# Longitudinal Multidomain Structures Formed in an Optically Bistable System under the Action of a Collimated Annular Beam 

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#### Abstract

Several longitudinal domains may form under the action of a collimated annular beam propagating in an optically bistable cavity-free system. The computer simulation of this phenomenon indicates two possible pathways for the domain formation process. © 2000 MAIK "Nauka/Interperiodica".


As is known, optically bistable cavity-free systems are studied in much detail within the framework of a wide-aperture approximation [1, 2], which ignores the effect of light diffraction. Taking this effect into account may significantly change the process of formation of the high-absorption regions [3-7] for both collimated and focused beams. It was previously demonstrated [3-7] that development of the switching waves for both initially Gaussian and annular beams proceeds, under certain conditions, virtually identically from the standpoint of forming several moving or immobile transverse domains. In particular, mobile kinks may develop under the action of a collimated annular structure [7].

Rozanov [2] explained the motion of high-absorption domains in terms of the focused Gaussian beams. In view of the fact that two kinks were formed under the action of collimated Gaussian beams [6], it was of interest to study whether this is possible under the action of an initially annular beam. This work showed that there are two possible pathways for the development of longitudinal two-domain structures. It should be emphasized that such structures can be observed not only in dynamic optical switching systems based on the absorption buildup, but in some other optical systems (potentially suited for a three-dimensional optical memory) as well.

The process in question is described by the following system of equations in dimensionless variables, determining the concentration $n$ of free electrons in the conduction band of a semiconductor (this concentration is measured in units of the maximum possible value and normalized to the input complex beam amplitude A):

$$
\begin{aligned}
& \frac{\partial A}{\partial z}+i D \Delta_{\perp} A+\frac{\delta_{0}}{2} \delta(n) A+i \alpha n A=0, \\
& 0<z<L_{z}, \\
& \frac{\partial n}{\partial t}+D_{\perp} \Delta_{\perp} n+D_{\|} \frac{\partial^{2} n}{\partial z^{2}}+q|A|^{2} \delta(n)-n
\end{aligned}
$$

with the initial and boundary conditions

$$
\begin{gather*}
\left.n\right|_{t=0}=\left.A\right|_{t=0}=\left.r \frac{\partial A}{\partial r}\right|_{r=0}=A(z, t, r=R) \\
\quad=\left.\frac{\partial n}{\partial z}\right|_{z=0, L_{z}}=\left.\frac{\partial n}{\partial r}\right|_{r=0}=\left.\frac{\partial n}{\partial r}\right|_{r=R}=0,  \tag{2}\\
A(z=0, t, r)=(r / a)^{2} \exp \left(-r^{2}\right)(1-\exp (-10 t)) . \tag{3}
\end{gather*}
$$

In Eqs. (1) and (2), the coordinate $z$ is measured in units of the diffraction length. Although the parameter $D$ in this case must be equal to unity, we retained this term in Eq. (1) for convenience of the numerical experiment. $L_{z}$ is the beam propagation pathlength. The transverse coordinate $r$ is normalized to a characteristic distribution radius of the initial beam profile, $R$ being the maximum value of this coordinate. The quantity $\delta_{0}$ characterizes the optical absorption at a diffraction wavelength, while $\delta(n)$ describes a nonlinear dependence of the absorption coefficient on the concentration of free electrons. The coefficient $\alpha$ equals the ratio of the initial beam power to a characteristic power of the self-action. This coefficient differs from zero when the radiation frequency deviates from the frequency of transition between selected levels; $\Delta_{\perp}=\frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial}{\partial r}\right)$ is the transverse Laplace operator written in the polar coordinates for an axisymmetric beam. The quantity $q$ characterizes the rate of the production of free electrons as a result of the light energy absorption in the semiconductor studied; $D_{\perp}$ and $D_{\|}$are dimensionless coefficients characterizing the transverse and longitudinal diffusion of charged particles; these coefficients are inversely proportional to, in particular, the square characteristic system dimensions along the longitudinal and transverse coordinates and to the pulse duration. The time $t$ is measured in units of the characteristic relaxation time. The coefficient $a$ determines the coordinate of the initial distribution of the light intensity. In this


Fig. 1. Longitudinal profile of the concentration of free electrons along the beam axis at various time instants $t=7.5$ (1), 11.25 (2), and 61.25 (3) calculated for the following parameters: $q=0.6084 ; D_{\|}=D_{\perp}=10^{-7}, \delta_{0}=0.625 ; \alpha=0 ; D=2.25$; and $a=0.57$.
paper, we assume that excitation of the donor levels is sufficiently small, so that the relaxation time is independent of the concentration of free electrons.

The absorption coefficient is approximated by the following function:

$$
\begin{equation*}
\delta(n)=(1-n) \exp [-\gamma(1-\beta n)], \tag{4}
\end{equation*}
$$

where $\gamma=2.553$ and $\gamma \beta=5$. This selection is explained by the fact that the condition $\gamma \beta>4$ corresponds to a bistable dependence of the concentration of free electrons in the input beam intensity within the framework of the point model.

Let the intensity of the beam entering into the medium be insufficient for switching the system into the upper excited state. As is known, a diffracted annular beam propagating in the medium transforms into a Gaussian beam. The transformed beam intensity at the axis may be considerably greater as compared to the initial radiation intensity. Therefore, it is possible to reach the level of intensity at which the system will be switched to the upper state in the bulk of the medium. In the high-concentration region thus formed, the light energy absorption sharply increases. After passage through this region, a drop in intensity is formed at the beam axis under certain conditions for charged particles. This accounts for the formation of an annular structure of the radiation intensity profile. As the radiation propagates further, the intensity at the beam axis increases again and the radiation intensity profile acquires the Gaussian shape.

As a result, the light beam propagating in the medium exhibits double self-focusing. This leads to the formation of two regions featuring a high concentration of charge carriers at the axis. The dynamics of formation of these regions is illustrated in Fig. 1. The first domain is formed close to the input cross section of the medium. The subsequent passage through the first domain by the annular beam leads to the formation of the second high-absorption domain. This is the first possible pathway of the process leading to the formation of two longitudinal domains. Note that the propagation of a short pulse may lead to situations in which the second domain is formed after the vanishing of the first domain. Such a phenomenon is observed when the relaxation time is shorter than the pulse duration, so that the pulse cannot completely "cover" the medium. This effect is very important for recording information by femtosecond pulses in a three-dimensional optical memory.

Another possible pathway for the longitudinal domain formation during the interaction of laser radiation with an optically bistable system is realized when the initial radiation intensity is sufficient to form a high-concentration domain at a certain distance from the beam axis in the vicinity of the input cross section. In this case, the pattern of the multidomain structure formation at the beam axis is significantly different from that described above.

Figure 2 shows the dynamics of formation of a multidomain structure by the second mechanism. According to this, two domains are formed in the first stageone at the front crystal face and another at the beam


Fig. 2. Longitudinal profile of the concentration of free electrons along the beam axis at various time instants $t=3.75$ (1), 7.5 (2), 13.75 (3), and 26.25 (4) calculated for the following parameters: $q=1.0 ; D_{\|}=D_{\perp}=5 \times 10^{-4}, \delta_{0}=5.0 ; \alpha=0 ; D=0.25$; and $a=0.5$.
axis in the bulk. In the example depicted in Fig. 1, the latter domain has a maximum intensity at $z=1.0$. This domain is formed by a mechanism of the annular beam focusing described above. The former longitudinal domain is due to the appearance of a high-absorption region at the input cross section of the medium. As a result, the radiation intensity profile exhibits, besides the main ring, an additional intensity minimum (replacing initial maximum). This additional ring structure provides for the formation of the third domain situated in front of the first longitudinal domain. The coordinate of the maximum intensity in the second (by the order of formation) domain is approximately 0.4 . This domain absorbs a part of the radiation energy, which results in that the first domain shifts away from the initial cross section. The domain structure described above is stationary.

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# Antireflecting Surface Coatings with Continuously Varying Complex Refractive Index 

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#### Abstract

Nonreflecting coatings for absorbing materials, based on thin films with an inhomogeneous complex refractive index, are considered. The reflectivity of these coatings is determined as a function of the film thickness (in units of the radiation wave length), optical constants, and the radiation incidence angle. It is shown that properly selected absorbing materials can be used to obtain surfaces with extremely low reflection coefficients. © 2000 MAIK "Nauka/Interperiodica".


High-performance antireflecting surface coatings are of great practical interest. In recent years, singlelayer coatings with inhomogeneous distribution of the refractive index (instead of multilayer structures composed of homogeneous films) have been a major focus of interest [1-3]. Total suppression of the electromagnetic radiation reflected from highly absorbing materials such as semiconductors (in solar cells) and metals (e.g., in "invisible" airplanes) is a very topical problem. However, well-known synthesis methods used to obtain antireflecting coatings can only be applied to fully transparent materials (i.e, the reflection coefficients of all media in the system can take only real values) $[1,4,5]$.

The purpose of this work was to investigate the properties of antireflecting coatings with continuously varying complex refractive indices.

Consider the reflection of a plane electromagnetic wave with a vacuum wavelength $\lambda$ incident at an angle $\varphi_{\mathrm{a}}$ on a thin film (subscript " f ") with the thickness $d_{\mathrm{f}}$. The film is inhomogeneous only in the direction perpendicular to the film surface (along the $z$-axis), has a complex refractive index $\hat{n}_{\mathrm{f}}(z)=n_{\mathrm{f}}(z)-i k_{\mathrm{f}}(z)$, and is supported on a massive homogeneous substrate (subscript " $s$ ") with a constant complex refractive index $\hat{n}_{\mathrm{s}}=n_{\mathrm{s}}-i k_{\mathrm{s}}$. The structure is surrounded by a transparent ambient medium (subscript "a") with a constant real refractive index $n_{\mathrm{a}}$. We assume that all the media possess magnetic permeabilities equal to unity. Since an analytical solution to the reflection problem for an inhomogeneous film is only known for several particular profiles of the refractive index [6,7] and expressions for the reflection coefficient are rather complicated even for real refractive indices, we have solved the reflection problem using a more universal approach: the direct numerical integration of the differential wave equation.

The numerical calculation of the reflectivity of a film with an inhomogeneous complex refractive index
is performed by the following method. The wave equation in the inhomogeneous medium (film) has the form

$$
\begin{equation*}
\frac{d^{2} \hat{U}_{\mathrm{f}}}{d z^{2}}-\alpha \frac{d}{d z}\left[\ln \hat{n}_{\mathrm{f}}^{2}(z)\right] \frac{d \hat{U}_{\mathrm{f}}}{d z}+\hat{\kappa}_{\mathrm{f}}^{2} \hat{U}_{\mathrm{f}}=0 \tag{1}
\end{equation*}
$$

where $\alpha=0$ and 1 for the $s$ and $p$ polarization, respectively; $\hat{U}_{\mathrm{f}}=U_{1 \mathrm{f}}+i U_{2 \mathrm{f}}$ is the unknown solution; $\hat{\mathrm{K}}_{\mathrm{f}}=$ $k_{0}\left(\hat{n}_{\mathrm{f}}^{2}(z)-n_{\mathrm{a}}^{2} \sin ^{2} \varphi_{\mathrm{a}}\right)^{1 / 2}$; and $k_{0}=2 \pi / \lambda$. For the numerical analysis of Eq. (1), we reduce it to a system of the first-order ordinary differential equations

$$
\left.\begin{array}{c}
U_{1 \mathrm{f}}^{\prime}=U_{3 \mathrm{f}}, \\
U_{2 \mathrm{f}}^{\prime}=U_{4 \mathrm{f}}, \\
U_{3 \mathrm{f}}^{\prime}=k_{0}^{2}\left[\left(n_{\mathrm{a}}^{2} \sin ^{2} \varphi_{\mathrm{a}}-\varepsilon_{\mathrm{f}}\right) U_{1 \mathrm{f}}+\varepsilon_{\mathrm{f}}^{(i)} U_{2 \mathrm{f}}\right] \\
+a\left[K_{1} U_{3 \mathrm{f}}+K_{2} U_{4 \mathrm{f}}\right] / M,  \tag{2}\\
U_{4 \mathrm{f}}^{\prime}= \\
k_{0}^{2}\left[\left(n_{\mathrm{a}}^{2} \sin ^{2} \varphi_{\mathrm{a}}-\varepsilon_{\mathrm{f}}\right) U_{2 \mathrm{f}}-\varepsilon_{\mathrm{f}}^{(i)} U_{\mathrm{ff}}\right] \\
+a\left[K_{1} U_{4 \mathrm{f}}-K_{2} U_{3 \mathrm{f}}\right] / M,
\end{array}\right)
$$

where

$$
\begin{gathered}
\varepsilon_{\mathrm{f}}-i \varepsilon_{\mathrm{f}}^{(i)}=\left(n_{\mathrm{f}}-i k_{\mathrm{f}}\right)^{2}, \quad K_{1}=2\left[n_{\mathrm{f}} n_{\mathrm{f}}^{\prime}+k_{\mathrm{f}} k_{\mathrm{f}}^{\prime}\right], \\
K_{2}=2\left[k_{\mathrm{f}} n_{\mathrm{f}}^{\prime}-n_{\mathrm{f}} k_{\mathrm{f}}^{\prime}\right], \text { and } M=n_{\mathrm{f}}^{2}+k_{\mathrm{f}}^{2} .
\end{gathered}
$$

Many different algorithms can be applied to solve system (2). We used the fourth- and fifth-order Runge-Kutta-Fehlberg scheme [8, 9], which is very reliable and convenient in practical applications. For the starting line, we have chosen the $z=d_{\mathrm{f}}$ plane. The uniform substrate features only an outgoing wave $\hat{U}_{\mathrm{s}}=$ $C \exp \left(i \hat{\kappa}_{\mathrm{s}} z\right)$, where $\hat{\kappa}_{\mathrm{s}}=\kappa_{\mathrm{s}}+i \kappa_{\mathrm{s}}^{(i)}=k_{0}\left(\hat{n}_{\mathrm{s}}^{2}-n_{\mathrm{a}}^{2} \sin ^{2} \varphi_{\mathrm{a}}\right)^{1 / 2}$ and $C$ is an arbitrary nonzero $(C \neq 0)$ constant deter-
mined by the wave amplitude. Therefore, we may set $C=1$. The continuity conditions $\hat{U}_{\mathrm{f}}\left(d_{\mathrm{f}}\right)=\hat{U}_{\mathrm{s}}\left(d_{\mathrm{f}}\right)$ and $\hat{U}_{\mathrm{f}}^{\prime}\left(d_{\mathrm{f}}\right)=\hat{U}_{\mathrm{s}}^{\prime}\left(d_{\mathrm{f}}\right)$ yield the following initial conditions for the $s$ polarization:

$$
\left.\begin{array}{c}
U_{1 \mathrm{f}}\left(d_{\mathrm{f}}\right) \equiv f_{1}=\left(\cos \kappa_{\mathrm{s}} d_{\mathrm{f}}\right) \exp \left(-\kappa_{\mathrm{s}}^{(i)} d_{\mathrm{f}}\right), \\
U_{2 \mathrm{f}}\left(d_{\mathrm{f}}\right) \equiv f_{2}=\left(\sin \kappa_{\mathrm{s}} d_{\mathrm{f}}\right) \exp \left(-\kappa_{\mathrm{s}}^{(i)} d_{\mathrm{f}}\right), \\
U_{3 \mathrm{f}}\left(d_{\mathrm{f}}\right)=-\left(\kappa_{\mathrm{s}} f_{2}+\kappa_{\mathrm{s}}^{(i)} f_{1}\right),  \tag{3}\\
U_{4 \mathrm{f}}\left(d_{\mathrm{f}}\right)=\kappa_{\mathrm{s}} f_{1}-\kappa_{\mathrm{s}}^{(i)} f_{2} .
\end{array}\right\}
$$

For the $p$ polarization, fields at the interfaces between two media must satisfy the following relationships: $\hat{U}_{\mathrm{f}}\left(d_{\mathrm{f}}\right)=\hat{U}_{\mathrm{s}}\left(d_{\mathrm{f}}\right)$ and $\left[\hat{n}_{\mathrm{f}}\left(d_{\mathrm{f}}\right)\right]^{-2} \hat{U}_{\mathrm{f}}^{\prime}\left(d_{\mathrm{f}}\right)=\hat{n}_{\mathrm{s}}^{-2} \hat{U}_{\mathrm{s}}^{\prime}\left(d_{\mathrm{f}}\right)$. Consequently, we obtain that $U_{\text {1f }}\left(d_{\mathrm{f}}\right)=f_{1}, U_{2 \mathrm{f}}\left(d_{\mathrm{f}}\right)=f_{2}$, and

$$
\left.\begin{array}{c}
U_{3 \mathrm{f}}\left(d_{\mathrm{f}}\right)=\left[\left(\varepsilon_{\mathrm{s}}^{(i)} \varepsilon_{\mathrm{d}}-\varepsilon_{\mathrm{s}} \varepsilon_{\mathrm{d}}^{(i)}\right)\left(\kappa_{\mathrm{s}} f_{1}-\kappa_{\mathrm{s}}^{(i)} f_{2}\right)\right. \\
\left.-\left(\varepsilon_{\mathrm{s}} \varepsilon_{\mathrm{d}}+\varepsilon_{\mathrm{s}}^{(i)} \varepsilon_{\mathrm{d}}^{(i)}\right)\left(\kappa_{\mathrm{s}} f_{2}+\kappa_{\mathrm{s}}^{(i)} f_{1}\right)\right] / L, \\
U_{4 \mathrm{f}}\left(d_{\mathrm{f}}\right)=\left[\left(\varepsilon_{\mathrm{s}}^{(i)} \varepsilon_{\mathrm{d}}^{(i)}+\varepsilon_{\mathrm{s}} \varepsilon_{\mathrm{d}}\right)\left(\kappa_{\mathrm{s}} f_{1}-\kappa_{\mathrm{s}}^{(i)} f_{2}\right)\right.  \tag{4}\\
\left.+\left(\varepsilon_{\mathrm{s}}^{(i)} \varepsilon_{\mathrm{d}}-\varepsilon_{\mathrm{s}} \varepsilon_{\mathrm{d}}^{(i)}\right)\left(\kappa_{\mathrm{s}} f_{2}+\kappa_{\mathrm{s}}^{(i)} f_{1}\right)\right] / L,
\end{array}\right\}
$$

where $\varepsilon_{\mathrm{s}}-i \varepsilon_{\mathrm{s}}^{(i)}=\left(n_{\mathrm{s}}-i k_{\mathrm{s}}\right)^{2}, \varepsilon_{\mathrm{d}} \equiv \varepsilon_{\mathrm{f}}\left(d_{\mathrm{f}}\right), \varepsilon_{\mathrm{d}}^{(i)} \equiv \varepsilon_{\mathrm{f}}^{(i)}\left(d_{\mathrm{f}}\right)$, and $L=\varepsilon_{\mathrm{s}}^{2}+\left(\varepsilon_{\mathrm{s}}^{(i)}\right)^{2}$. Calculating functions $U_{1}-U_{4}$ for $z=0$, taking into account that the solution in the ambient medium is given by the formula

$$
\begin{equation*}
\hat{U}_{\mathrm{a}}=\hat{A} \exp \left(i \kappa_{\mathrm{a}} z\right)+\hat{B} \exp \left(-i \kappa_{\mathrm{a}} z\right), \tag{5}
\end{equation*}
$$

and using the boundary conditions at $z=0$, we obtain the following expressions for complex amplitudes $\hat{A}$ and $\hat{B}$ of the incident and reflected waves,

$$
\begin{align*}
& \hat{A}=\left[\hat{U}_{\mathrm{f}}(0)+\left(\hat{U}_{\mathrm{f}}^{\prime}(0) /\left(i \kappa_{\mathrm{a}} \hat{p}\right)\right)\right] / 2,  \tag{6}\\
& \hat{B}=\left[\hat{U}_{\mathrm{f}}(0)-\left(\hat{U}_{\mathrm{f}}^{\prime}(0) /\left(i \kappa_{\mathrm{a}} \hat{p}\right)\right)\right] / 2, \tag{7}
\end{align*}
$$

where $\hat{U}_{\mathrm{f}}(0)=U_{1 \mathrm{f}}(0)+i U_{2 \mathrm{f}}(0) ; \quad \hat{U}_{\mathrm{f}}^{\prime}(0)=U_{3 \mathrm{f}}(0)+$ $i U_{4 \mathrm{f}}(0) ; \kappa_{\mathrm{a}}=k_{0} n_{\mathrm{a}} \cos \varphi_{\mathrm{a}}$; and $\hat{p}=1$ and $\hat{p}=\left(\hat{n}_{\mathrm{f}}(0) / n_{\mathrm{a}}\right)^{2}$ for the $s$ and $p$ polarization, respectively. The energy reflection coefficient for an inhomogeneous film is $R_{s, p}=(|\hat{B}| /|\hat{A}|)^{2}$.

In this study, we consider inhomogeneous coatings with the real part of the refractive index described by
either standard profiles,

$$
\begin{gather*}
n_{\mathrm{f}}(z)=n_{\mathrm{f} 0}+\left(n_{\mathrm{fd}}-n_{\mathrm{f} 0}\right)\left(z / d_{\mathrm{f}}\right)^{a},  \tag{8}\\
n_{\mathrm{f}}(z)=n_{\mathrm{f} 0} n_{\mathrm{fd}}\left[n_{\mathrm{fd}}-\left(n_{\mathrm{fd}}-n_{\mathrm{f} 0}\right)\left(z / d_{\mathrm{f}}\right)\right]^{-1},  \tag{9}\\
n_{\mathrm{f}}(z)=n_{\mathrm{f} 0}\left(n_{\mathrm{fd}} / n_{\mathrm{f} 0}\right)^{\left(z / d_{\mathrm{f}}\right)}, \tag{10}
\end{gather*}
$$

or a special profile proposed in [10],

$$
\begin{gather*}
n_{\mathrm{f}}(z)=n_{\mathrm{f} 0}+\left(n_{\mathrm{fd}}-n_{\mathrm{f} 0}\right) \\
\times\left[10\left(z / d_{\mathrm{f}}\right)^{3}-15\left(z / d_{\mathrm{f}}\right)^{4}+6\left(z / d_{\mathrm{f}}\right)^{5}\right] . \tag{11}
\end{gather*}
$$

The imaginary part $k(z)$ of the refractive index (the extinction coefficient) is only described by functions (8) and (11), in which parameters $n_{\mathrm{f} 0}$ and $n_{\mathrm{fd}}$ are replaced with $k_{\mathrm{f} 0}$ and $k_{\mathrm{fd}}$. Parameters $n_{\mathrm{f} 0}$ and $n_{\mathrm{fd}}$ are the real parts of the refractive index and $k_{\mathrm{f} 0}$ and $k_{\mathrm{fd}}$ are the extinction coefficients in the planes $z=0$ (the interface between the film and the ambient medium) and $z=d_{\mathrm{f}}$ (the interface between the film and the substrate), respectively.

Calculations (see Fig. 1a) clearly indicate that no effective decrease in reflectivity can be achieved with the aid of transparent inhomogeneous coatings $(k(z)=0)$ if the substrate material exhibits a nonzero absorption ( $k_{\mathrm{s}} \neq 0$ ). Nor will the antireflecting effect be observed when only the real part of the refractive index continuously varies up to $n_{\mathrm{a}}$ at the surface of the absorbing material for $k(z)=k_{\mathrm{s}}=$ const or, vice versa, when $k(z) \longrightarrow 0$ for $n_{\mathrm{f}}(z)=n_{\mathrm{s}}=$ const. The reflectivity of the absorbing substrate material can only be substantially lowered with a surface coating whose permittivity has both a real and imaginary part, which continuously varies from the values corresponding to the substrate parameters to the parameters of the ambient medium. In this case, the result strongly depends on the type of inhomogeneous profile for $n_{f}$ and $k_{\mathrm{f}}$ (Figs. 1b and 1d). For fully transparent materials, the best antireflecting coating was obtained for the refractive index profile (11) whereas, for absorbing materials, the lowest reflection coefficients were obtained for $n(z)$ and $k(z)$ both described by profile (8). In the latter case, the optimum parameters of expression (8) strongly depend on $d_{\mathrm{f}} / \lambda$ (Fig. 1e) and $k_{\mathrm{s}}$ (Fig. 1f) rather than on $\varphi_{\mathrm{a}}$ (Figs. 1c and 1e) and $n_{\mathrm{s}}$ (Fig. 1f). Note that the lowest reflectivities at the normal incidence (Fig. 1d) appear to be several orders of magnitude smaller than the corresponding values obtained for transparent materials with profile (11). At large slope angles, the reflectivity increases almost linearly with $\varphi_{\mathrm{a}}\left(R_{s, p} \longrightarrow 1\right.$ at $\left.\varphi_{\mathrm{a}} \longrightarrow 90^{\circ}\right)$.

Thus, absorbing materials enable the creation of more effective wideband antireflecting surfaces in comparison with transparent materials. This fact is clearly seen in Fig. 2a presenting the reflection coefficient $R_{0}$ as a function of the parameters $n_{\mathrm{s}}$ and $k_{\mathrm{s}}$ : the reflectivity abruptly lowers when $k_{\mathrm{s}}$ increases from 0 to $\sim 1$; in this


Fig. 1. Plots of (a, b, d) the reflectivity at normal incidence $R_{0}$ vs. $d_{\mathrm{f}} / \lambda$ and (c) the reflectivities at oblique incidence $R_{p}$ and $R_{s}$ vs. $\varphi_{\mathrm{a}}$ without steps in optical constants at interfaces (dot-and-dash and dashed curves for the $p$ and $s$ polarization, respectively). All curves were calculated for $n_{\mathrm{f} 0}=n_{\mathrm{a}}-1.0, k_{\mathrm{f} 0}=0, n_{\mathrm{fd}}=n_{\mathrm{s}}$, and $k_{\mathrm{fd}}=k_{\mathrm{s}}$, except for curves $l\left(k(z)=0, n_{\mathrm{f} 0}=n_{\mathrm{a}}, n_{\mathrm{fd}}=n_{\mathrm{s}}\right), 2\left(k(z)=k_{\mathrm{s}}\right.$, $n_{\mathrm{f} 0}=n_{\mathrm{a}}, n_{\mathrm{fd}}=n_{\mathrm{s}}$ ), and $3\left(n(z)=n_{\mathrm{s}}, k_{\mathrm{f} 0}=0, k_{\mathrm{fd}}=k_{\mathrm{s}}\right.$ ). The following profiles were studied. For $n(z)$ : profile ( 8 ) with $a=1.0$ (curves $1,2,4,9), 2.0(12), 3.0(14), 3.5(17), 4.3(19), 4.8(20), 5.0(5,15,21), 5.5(18,22)$, and 0.5 (7); profile (9) with $a=1.0$ (10, 11); profile (10) (8); and profile (11) ( $6,13,16$ ). For $k(z)$ : profile (8) with $a=1.0$ (3, 9, 10), 2.0 (12), 3.0 (14), 3.5 (17), 4.0 (11), 4.3 (19), $4.8(20), 5.0(15,21), 5.5(18,22)$, and $0.5(7,8)$ and profile $(11)(13,16)$. Plots of $(\mathrm{e}, \mathrm{f})$ the reflectivities $R_{0}$ and $R_{S, p}$ for $\varphi_{\mathrm{a}}=30^{\circ}$ vs. parameter $a$ (Figs. 1e and f) in the case when both $n(z)$ and $k(z)$ are described by the same profile (8) for $d_{\mathrm{f}} / \lambda=2.0$ (curve 23), $4.0(24), 6.0(25)$, and $3.0(26-32) ; n_{\mathrm{s}}=4.0(1-30), 2.0(32)$, and $1.3(31) ; k_{\mathrm{s}}=0(4-6), 1.0(2,7-25,27,31,32), 2.0(1,3,26)$, 3.0 (28), 4.0 (29), and 5.0 (30).
case, the particular value of the real part of the refractive index is of no significance. However, this is only so when equalities $n_{\mathrm{f} 0}=n_{\mathrm{a}}$ and $k_{\mathrm{f} 0}=0$ are satisfied with sufficiently high precision. Figure $2 b$ depicts the reflectivity as a function of the steps $\Delta n_{\mathrm{f} 0}=n_{\mathrm{f} 0}-n_{\mathrm{a}}$ and $k_{\mathrm{f} 0}$. From this figure we can see that, for extremely small values of these parameters ( $\Delta n_{\mathrm{f} 0}$ and $k_{\mathrm{f} 0} \leqslant 10^{-2}$ ) and transparent or weakly absorbing substrate materials, $R_{0}$ is almost independent of the steps in optical constants at $z=0$. For strongly absorbing materials, this dependence is linear (to the first approximation). If $k_{\mathrm{f} 0}<\Delta n_{\mathrm{f} 0}$, the effect of the step in $k_{\mathrm{f} 0}$ is barely perceptible.

The effect of the steps $\Delta n_{\mathrm{fd}}=n_{\mathrm{fd}}-n_{\mathrm{s}}$ or $\Delta k_{\mathrm{fd}}=k_{\mathrm{fd}}-$ $k_{\mathrm{s}}$ is not as pronounced, especially for $\Delta n_{\mathrm{f} 0} \neq 0$. Sometimes, it may be even advantageous if $n_{\mathrm{fd}} \neq n_{\mathrm{s}}$ and $k_{\mathrm{fd}} \neq$ $k_{\mathrm{s}}$ (Figs. 2c and 2d). Note also that, when $\Delta n_{\mathrm{fd}}=0$, the reflectivity as a function of $\Delta k_{\mathrm{fd}}$ may become strongly asymmetric (Fig. 2d, curves 23).

In conclusion, we formulate these main inferences. Antireflecting surfaces with lowest reflection coefficients can be obtained using absorbing (rather than transparent) materials containing a thin surface layer (in a surface coating) with both real and imaginary


Fig. 2. Plots of (a) the reflectivity $R_{0}$ vs. $n_{\mathrm{s}}$ for $k_{\mathrm{S}}=$ const (solid curves) and vs. $k_{\mathrm{s}}$ for $n_{\mathrm{s}}=$ const (dashed curves) with no steps in optical constants at interfaces ( $n_{\mathrm{f} 0}=n_{\mathrm{a}}, n_{\mathrm{fd}}=n_{\mathrm{s}}, k_{\mathrm{f} 0}=0$, and $k_{\mathrm{fd}}=k_{\mathrm{s}}$ ) and (b) $\mathrm{R}_{0}$ vs. $\Delta n_{\mathrm{f} 0}$ for $k_{\mathrm{f} 0}=0, n_{\mathrm{fd}}=n_{\mathrm{s}}$, and $k_{\mathrm{fd}}=k_{\mathrm{S}}$ (solid curves) and vs. $k_{\mathrm{f} 0}$ for $n_{\mathrm{f} 0}=n_{\mathrm{a}}, n_{\mathrm{fd}}=n_{\mathrm{s}}$, and $k_{\mathrm{fd}}=k_{\mathrm{s}}$ (dashed curves). Reflectivities $R_{p}$ (solid curves) and $R_{S}$ (dashed curves) at $\varphi_{\mathrm{a}}=45^{\circ}$ (c) vs. steps $\Delta n_{\mathrm{fd}}$ for $k_{\mathrm{fd}}=k_{\mathrm{s}}, n_{\mathrm{f} 0}=n_{\mathrm{a}}$, and $k_{\mathrm{f} 0}=0$ and (d) vs. $\Delta k_{\mathrm{fd}}$ for $n_{\mathrm{f} 0}=n_{\mathrm{a}}, k_{\mathrm{f} 0}=0$, and $n_{\mathrm{fd}}=n_{\mathrm{s}}$; both $n(z)$ and $k(z)$ are described by profile (8) for $a=4.8 ; d_{\mathrm{f}} / \lambda=3.0$ (a), 5.0 (b), and 2.0 (c and d); $n_{\mathrm{s}}=4.0$ [curves $4,5,7,9,12,13$ (solid), 14-18, 20, 22, 23], 2.5 (19), $1.5\left[6,8,10,11,13\right.$ (dashed), 21]; $k_{\mathrm{s}}=0(8), 0.1(9), 0.2(10), 0.5(11,12,17$, and 18$), 0.7(1), 1.0[2,13$ (dashed), 14 , $15,22,23], 2.0(19,21), 4.0(3), 5.0(20)$, and $6.0\left[13\right.$ (solid), 16); and $n_{\mathrm{a}}=1.0(2,4,9,12,13,16,17,20,22), 1.3(1,3,5,6,8,10$, $11,19,21), 2.0(7,14,18)$, and $3.0(15,23)$.
parts of the refractive index continuously varying from values corresponding to the substrate to those of the ambient medium. The governing factor for attaining an extremely low reflectivity is the accurate matching of the optical constants at the interface between the coating and the ambient medium, which is technologically a reasonably realizable task (even for $n_{\mathrm{a}}=1$ [11]).

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# Controlled Oscillations in an Autostochastic System 

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#### Abstract

Processes in a nonautonomous autostochastic system possessing an attractor of the Chen type were analyzed by numerical methods. The system was described by a set of equations for coupled oscillators with inertia. It is demonstrated that autooscillations can be controlled when a regular signal acts unidirectionally upon chaotic oscillations or, vice versa, when chaotic oscillations act upon the regular ones. © 2000 MAIK "Nauka/Interperiodica".


The possibility of controlling oscillations in autostochastic systems has been extensively studied in recent years [1-7]. Besides an important application to communication systems employing the principle of synchronized dynamic chaos, these investigations are of interest from the standpoint of the fundamental theory of nonlinear oscillations. Formulation of the concept of a chaotic autooscillating system has dramatically changed theoretical notions about many nonlinear phenomena studied and employed in various fields of science and technology.

Below we will consider the possibility of controlling oscillations in a system with chaotic dynamics possessing an attractor of the Chen type [8]. The study focuses on the derandomization of the chaotic oscillations as a result of the action of a regular external signal and on the chaotization of regular motions under the action of external chaotic oscillations. The controlled and controlling oscillators are described by the sets of equations for coupled oscillators with inertia.

The set of nonlinear differential equations for two unidirectionally coupled oscillators with inertia can be written, based on the general expression for a single oscillator with inertia [9], in the following form:

$$
\begin{gather*}
\ddot{x}_{i}+2 \delta_{i} x_{i}+\omega_{i}^{2} x_{i}=-\omega_{i} z_{i}+f_{i}\left(x_{i}, z_{i}\right)+m \dot{x}_{2}  \tag{1}\\
\dot{z}_{i}+b_{i} z_{i}=a_{i} x_{i}+\varphi_{i}\left(x_{i}\right),
\end{gather*}
$$

where $i=1,2 ; t$ is the time; $x_{i}$ and $z_{i}$ are the time- dependent variables; $\delta_{i}, \omega_{i}, a_{i}$, and $b_{i}$ are constant quantities; $m$ is a constant coupling coefficient; and $f_{i}\left(x_{i}, z_{i}\right)$ and $\varphi_{i}\left(x_{i}\right)$ are nonlinear functions.

Properties of the partial oscillators entering into system (1) are determined by expressions for the nonlinear functions. In particular [9], the classical Lorentz equations [10] can be transformed so as to acquire the gen-
eral form of Eqs. (1) for a single partial generator with $i=1$ and $m=0$. The well-known Chen equations [8]

$$
\begin{gathered}
\dot{x}=\sigma(y-x), \\
\dot{y}=\alpha y-r x-x z, \\
\dot{z}=x y-b z
\end{gathered}
$$

can also be converted so as to take the form of equations describing an oscillator with inertia. Based on the Chen equations, we may write a system of equations for two oscillators (1) by excluding the variable $y$, introducing the index $i$, and using the nonlinear functions

$$
\begin{gather*}
f_{i}\left(x_{i}, z_{i}\right)=-\left(1.5 \omega_{i} x_{i}+0.5 x_{i}^{2}+z_{i}\right) x_{i} \\
+\sigma_{i}\left(\alpha_{i}-2 r_{i}+1\right)\left(x_{i}+\omega_{i}\right),  \tag{2}\\
\varphi_{i}\left(x_{i}\right)=\left(\sigma_{i}-0.5 b_{i}\right) x_{i}^{2} .
\end{gather*}
$$

The coefficients $b_{i}, r_{i}, \sigma_{i}$, and $\alpha_{i}$ in Eqs. (1) and (2) coincide with the corresponding quantities in the original Chen equations modified by introducing the index $i$. The other quantities are defined as follows:

$$
\begin{gather*}
a=\left(2 \sigma_{i}-b_{i}\right) \omega_{i}, \quad 2 \delta_{i}=\sigma_{i}-\alpha_{i}, \\
\omega_{i}^{2}=b_{1}\left(r_{i}-1\right) . \tag{3}
\end{gather*}
$$

The Chen equations with $\sigma=35, \alpha=28, r=7$, and $b=3$ possess an unusual chaotic attractor (called the Chen attractor) in a projection onto the $\{x, z\}$ plane. Provided the same constants ( $\sigma_{i}=35, a_{i}=28, r_{i}=7$, and $b_{i}=3$ ), identical attractors in a projection onto the $\left\{x_{i}, z_{i}\right\}$ plane exist in the partial oscillators with inertia described by Eqs. (1)-(3) with $m=0$.

We have studied the task of controlling autooscillations in the system described by Eqs. (1)-(3). The regimes of induced derandomization and chaotization were analyzed for $\sigma_{1}=36, \sigma_{2}=30.5$ and $\sigma_{1}=30.5$,


Fig. 1. Bifurcation diagrams showing plots of the maximum values $\left[x_{1}\right]$ of the oscillating variable $x_{1}(t)$ versus the coupling parameter $m$ in the case of (a) derandomization of the initial chaotic autonomous autooscillations under the action of a regular external signal and (b) chaotization of the initial regular oscillations under the action of external chaotic oscillations.
$\sigma_{2}=36$. The other parameters of the partial oscillators were selected as follows:

$$
\alpha_{1}=\alpha_{2}=28, \quad r_{1}=r_{2}=7, \quad b_{1}=b_{2}=3
$$

Figures 1a and 1 b show the bifurcation diagrams illustrating the cases of derandomization of the chaotic oscillations and chaotization of the regular oscillations, respectively, in a nonautonomous regime. These data illustrate changes in the maximum values $\left[x_{1}\right]$ of the oscillating variable $x_{1}(t)$ observed in the system featuring chaotic autonomous autooscillations (Fig. 1a) and regular oscillations (Fig. 1b) plotted as functions of the coupling parameter $m$. The external oscillations $x_{2}(t)$ were regular (Fig. 1a) and chaotic (Fig. 1b).

As seen from Fig. 1a, the transition from the chaotic to the regular regime under the action of an external regular signal takes place at $m=5.1$ and has a jumplike (rigid) character. However, a tendency towards the transition to a regular mode is also distinguished when the coupling parameter approaches the critical value. This trend is manifested by "concentration" of the points corresponding to maximum values of the oscillating variable.


Fig. 2. Induced oscillations in coupled systems: (a, c, e) power spectra and (b, d, f ) attractors for the oscillation process $x_{1}(t)$ observed ( $\mathrm{a}, \mathrm{b}$ ) in an autonomous chaotic regime with $m=0$, (c, d) upon derandomization induced by an external regular signal for $m=6$, and (e, f) upon chaotization of an initially regular process by an external chaotic signal for $m=3$.

The derandomization of chaotic oscillations, whereby a synchronism is established between the oscillations of two oscillators, is evidence of an internal resonance in the autostochastic system. The fact that the derandomization event has a jumplike character indicates that there exits a threshold for synchronization with respect to the external signal amplitude. This synchronization threshold can serve as a criterion of the system stochasticity [11].

For an initially regular oscillation process, the induced randomization (in contrast to the derandomization process considered above) develops in a "soft" manner. As seen in Fig. 1b, the maximum scatter of the oscillating variable $x_{1}(t)$ gradually increases with the coupling parameter $m$, the latter determining the intensity of action of the external random process $x_{2}(t)$. This pattern shows that there is no chaotization threshold. The controlled and controlling oscillations are not synchronized. The system features asynchronous interactions analogous to those observed in the asynchronous regime at a boundary of the trapping band in a nonautonomous system with regular dynamics [12].

Figure 2 presents the characteristic power spectra and the corresponding attractors illustrating the process
of induced synchronization (derandomization) and chaotization. Figures 2 a and 2 b show the power spectrum of an autonomous chaotic oscillation process and the corresponding attractor projected onto the $\left\{x_{1}, z_{1}\right\}$ plane. These data refer to $m=0$ in Fig. 1a. Figures 2c and 2 d show the power spectrum and the corresponding attractor in the case of synchronization $(m=6)$. Note that the same curves (Figs. 2c and 2d) virtually reflect the power spectrum and attractor of the controlling oscillation process. A trajectory of the imaging point projected onto the $\left\{x_{1}, x_{2}\right\}$ plane for $m=6$ appears as an ellipse extended along the point set $x_{1}=x_{2}$.

Figures 2 e and 2 f show the power spectrum and the corresponding attractor referring to the state $m=3$ in Fig. 1b. The power spectrum and attractor of the autonomous oscillations (for $m=0$ in Fig. 1b) are virtually the same as those in Figs. 2c and 2d, respectively, while the power spectrum and attractor of the controlling oscillations are similar to those depicted in Figs. 2a and $2 b$. Here, a trajectory of the imaging point projected onto the $\left\{x_{1}, x_{2}\right\}$ plane for $m=3$ reflects a chaotic behavior with a weakly pronounced orientation of motions in the direction of the point set $x_{1}=x_{2}$.

The results of the investigation of the effect of regular oscillations upon the chaotic process indicate that synchronization counteracts the chaotic motion and can be used as a stabilizing and controlling factor not only in systems with regular motions but in autostochastic oscillators as well. The induced synchronization allows a system operation mode to be switched from chaotic to regular.

The phenomenon of induced chaotization in a system with regular autonomous oscillations allows the same system to be used for obtaining induced chaotic
oscillations with a smoothly controlled width of the power spectrum.

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# A New Approach to the Development of Multifrequency Lasers 

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#### Abstract

A new approach to the creation of tunable lasers with synthesized spectra based on composite active media is proposed. This method eliminates irreproducibility of the energy distribution between components of the lasing spectrum, which commonly appears with an increase in their number. A four-frequency laser employing a single active medium is realized for the first time. © 2000 MAIK "Nauka/Interperiodica".


Creation of multifrequency lasers whose spectra can be synthesized and tuned rapidly is essential for the development of new effective methods in spectroscopy and data storage and transfer technology. Solving this task has become especially important in recent years with the advent of new active media and the development of electronic tuning techniques [1].

Progress in this field is hindered by the considerable instability and irreproducibility of the intensity distribution between components of the lasing spectrum, typical of multifrequency sources when the number of components exceeds two. When a common active medium is employed for the generation of all spectral components, this instability is caused by the strong coupling (competition) between them. In cases when the radiation of independent sources is effectively combined at the output and in equivalent cases (lasing from different regions of the active medium, etc.) the energies of different sources fluctuate independently and the corresponding "coupling coefficients" vary irreproducibly (e.g., due to temporal variations in the spatial structure of pumping).

In this report, we propose a new approach to this problem and demonstrate its effectiveness experimentally. This approach is based on the purposeful design of special composite active media for multifrequency lasers, containing several groups of active lasing centers in the same matrix. These centers are characterized by two features in the spectral properties (see Fig. 1a): (1) their absorption spectra considerably overlap and (2) their luminescence (or gain) spectra exhibit the Stokes shifts such that all of them contain a region $M_{j}=$ [ $\lambda_{1 j} ; \lambda_{2 j}$ ] overlapping neither with the absorption bands nor with other luminescence spectra (originating from the effective levels $s_{1}$ and $s_{2}$ ). In the process of multifrequency lasing, such a medium behaves like that with homogeneous broadening for the lasing spectrum components falling within the same $M_{j}$ region and with
inhomogeneous broadening for the components belonging to different $M_{j}$ regions.

The intensity distribution of the spectral components in such a laser can be modified by changing the relationship between losses common to the groups of lines originating from different $M_{j}$ regions, with subsequent fine tuning of the individual losses for the spectral lines of each particular region. Reproducibility of the relationship between the total energies in the groups of components from different regions (which is set at the first stage) is then secured automatically, because the influence of strong fluctuations in the energy and spatial structure of pumping of these groups is identical. To attain reproducibility in the intensity distribution between spectral components belonging to the same $M_{j}$ region and competing with each other, one can make use of the rules (known from the studies on intracavity laser spectroscopy) that determine the effect of frequency-selective losses on the laser spectrum and its evolution. Reproducibility can definitely be achieved for the case of two-component lasing in each $M_{j}$ region, which has been studied in [2].

Molecules, ions, complexes, etc., employed in tunable lasers can be listed among active laser centers appropriate for the proposed type of media. A variety of suitable matrices includes crystals, liquids, glasses, etc. Bearing in mind the special interest toward the systems that can be pumped, directly or indirectly, by powerful laser diode arrays, we chose dye molecules pumped by the radiation with a wavelength of $\Lambda=0.53 \mu \mathrm{~m}$ for the first experimental realization of the proposed laser. Examining laser dyes of this class, one can see that it is quite difficult to solve the problem by simply using dye combinations, because of the considerable overlap of their spectra. For this reason, cascade excitation of one dye by the other in combination with their simultaneous lasing has been used in the previous studies (see, e.g., $[3,4]$ ). Obviously, in this case it is difficult to con-


Fig. 1. (a) Schematic representation of the matching of component spectra in a composite active medium and (b) implementation of this scheme for the mixture of rhodamine 6G (shaded regions) with 6-aminophenolenone. Solid lines represent the absorption spectra and dashed lines represent the lasing spectra; $C$ is the acid concentration and $J$ is the normalized intensity.
trol the spectrum and to excite more than one component by an intermediate dye.

To implement the proposed approach, new dyes with the desired properties could be synthesized. Alternatively, one can rely on molecules whose properties vary under the influence of the matrix (or the solvent); if different types of molecules change their properties in a dissimilar way, then these can be used to compose the desired groups of lasing centers. In this study, we employed the second method using rhodamine 6 G and 6-aminophenolenone dyes. Under normal conditions, the spectra of these dyes do not comply with the requirements set above (see Fig. 1b). However, due to their chemical structure, the first molecule has no tendency to add on protons, while the second molecule readily acquires a hydrogen nucleus; thus, the spectral lines of the second substance shift with the pH value of the solution, while those of the first substance does not. When $\sim 0.8 \%$ of acid were added to the alcohol solvent (with a $2: 3$ ratio of the rhodamine 6G and 6-aminophe-
nolenon concentrations), both the above requirements were met (see Fig. 1a) and the laser efficiency was at maximum.

Using traditional techniques of introducing fre-quency-dependent losses [1], a four-frequency laser employing a common active medium (with two spectral components generated in each $M_{j}$ region) was created for the first time and investigated experimentally; effectiveness of the proposed approach was confirmed. Detailed comparison of the averaged spectrum (Fig. 2) with the spectra of separate pulses revealed that variations characterizing the scatter in their shape do not exceed $12 \%$ (without additional stabilization techniques and with the scatter in the energy and the spatial structure of pumping, as high as $\sim 100 \%$ ). Figure 2 demonstrates the possibility to control the spectrum, in particular, to influence the intensity distribution between spectral components by introducing extra losses $\Delta$ for one of the lines. As is seen, energy redistribution between the two components belonging to one


Fig. 2. Four-frequency laser spectrum and its variation with $\Delta$.
of the regions takes place, while neither relative, nor total energy, change for the two others. These results confirm our hypothesis concerning the properties of such an active medium and illustrate the simplicity of the tuning procedure. The effectiveness of this approach is further corroborated by the expansion of the multifrequency laser tuning range to $555-640 \mathrm{~nm}$.

Creation of the composite media containing three groups of active centers, combined with the method of single-stage spatial separation, will enable one to obtain lasing spectra consisting of 12 lines whose intensity and wavelength can be tuned simply and reproducibly, and to expand the wavelength range as well. There are no limitations related to the nature of the active centers or the matrix. Finally, we must note that development of such lasers is important for numerous applications, in particular, for selective photodynamic therapy and multifrequency spectroscopy (especially for
optochemical sensors), where the wavelength separation required exceeds the range of a single dye.

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# Translational Nonequilibrium of the Shock Wave Front in an Argon-Helium Gas Mixture 

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#### Abstract

It is experimentally established that the concentration of metastable atoms appearing in the front of a shock wave propagating in a gas phase is greater by one order of magnitude for an argon-helium 1:1 mixture than for pure argon with the same Mach number and initial pressure. The average "longitudinal" temperature of the shock wave front is determined. An expression for the concentration of excited particles is derived that takes into account the "longitudinal" character of the gas temperature in the shock wave front. © 2000 MAIK "Nauka/Interperiodica".


Previously [1], we experimentally studied the shock wave propagation in argon and demonstrated that there is a region of translational nonequilibrium in the wave front where the average temperature $\tilde{T}_{\mathrm{f}}$ is 1.7 times the equilibrium gas temperature behind the front. The degree of nonequilibrium in this region was characterized by the "temperature of collisions in the direction of the shock wave propagation" (or the "longitudinal" temperature) at the point $x=0$

$$
\begin{equation*}
T_{x=0}=\frac{m}{2 k}\left[\frac{1}{2} \sqrt{\frac{2 k T_{1}}{m}}+V_{2}+\frac{1}{2} \sqrt{\frac{2 k T_{2}}{m}}\right]^{2}, \tag{1}
\end{equation*}
$$

where $x$ is the distance from the wave front (expressed in units of the mean free path of a cold particle) measured in the shock wave propagation direction; $m$ is the mass of a particle; $k$ is the Boltzmann constant; $V_{2}$ is the hot gas flow velocity in the region behind the shock wave; $T_{1}$ and $T_{2}$ are the gas temperatures in the cold and hot regions, respectively. The origin of coordinates at the point $x=0$ is selected according to the Mott-Smith shock wave front model proposed in 1951 [2].

According to formula (1), a change in $V_{2}$ at the fixed $T_{1}$ and $T_{2}$ values leads to the corresponding change in the "longitudinal" temperature $T_{x=0}$, which must be experimentally manifested in variation of the front temperature $\tilde{T}_{\mathrm{f}}$. The purpose of our work was to check this hypothesis. We have experimentally studied the shock waves propagating in argon and in an argon-helium 1:1 mixture. For a shock wave with the Mach number $M=6$ ( $T_{1}=293 \mathrm{~K} ; T_{2}=3553 \mathrm{~K}$ ), formula (1) yields $T_{x=0}=11434 \mathrm{~K}$ for argon and $T_{x=0}=17086 \mathrm{~K}$ for the $\mathrm{Ar}-\mathrm{He}$ mixture. The effective particle mass $m$ for the mixture was calculated using the molecular weight of the gas mixture.

Velikodny̆̌ et al. [3] reported on the nonequilibrium emission from molecules $I_{2}$ behind the shock wave front in a strongly diluted two-component gas mixture (97-99.9\%). According to these experimental data, the emission intensity $I_{2}$ increased with the velocity of sound in the diluent gas, which was explained by a large partial Mach number in the heavy gas component. Since no information had been available on the crosssections and constants of elastic and inelastic processes (necessary for calculating the collision integral in the shock wave propagation direction), the consideration in [3] was restricted to the estimation of a maximum value of the collision integral. Even this approximation provided for a good agreement between calculated and experimental values. In our opinion, that approach was equivalent to a phenomenological determination of the average temperature $\tilde{T}_{\mathrm{f}}$ in the shock wave front [1]. This work expands the range of experimentally studied effects related to the translational nonequilibrium in a two-component gas mixture. The observed effects are described in terms of averaged phenomenological parameters.

The experiments were conducted in a stainless-steel single-diaphragm shock tube with a diameter of 50 mm . The stacked diaphragms were made of a $20-\mu \mathrm{m}$-thick Mylar film. The low-pressure chamber was initially evacuated to a pressure of $10^{-2}$ Torr, with a leak rate not exceeding $2 \times 10^{-4} \mathrm{Torr} / \mathrm{min}$. The lowpressure chamber was filled with an argon-hydrogen or a helium-argon-hydrogen gas mixture, hydrogen serving as the driving gas. The gas mixtures were prepared from high-purity argon and commercial-purity helium and hydrogen. The shock wave velocity was measured with the aid of piezoelectric transducers, which provided for a relative accuracy of not less than $1 \%$. The shock wave tube was provided with viewing ports made of $\mathrm{MgF}_{2}$ crystals. The spectral measurements were per-


Fig. 1. Experimental plots of the intensity of the $\mathrm{H}_{\alpha}$ Balmer line versus temperature $T_{2}$ for (1) $\mathrm{He}-\mathrm{Ar}-\mathrm{H}_{2}$ ( $50: 49.5: 0.5$, $P_{1}=20$ Torr) and (2) Ar- $\mathrm{H}_{2}$ (99: 1, $P_{1}=10$ Torr) mixtures.
formed using a BM-50 vacuum monochromator equipped with a diffraction grating ( $1200 \mathrm{~mm}^{-1}$ ) possessing a dispersion of $1.66 \mathrm{~nm} / \mathrm{mm}$. The spectral measurements were performed using a FEU-100 photomultiplier tube sensitive in a $170-800 \mathrm{~nm}$ spectral range, a DL-922 digital oscillograph, and an OSA-WP4 multichannel analyzer. The time resolution of the photomultiplier circuit was $1 \mu \mathrm{~s}$.

Previously [1], we described an empirical model for investigation of the translational nonequilibrium in a shock wave front in argon with a small admixture of hydrogen used as an indicator of the nonequilibrium state. According to this model, a certain amount of metastable argon atoms $\tilde{A} r \equiv \operatorname{Ar}\left\{4 s^{\prime}[1 / 2]_{0}\right\}$ with a concentration of $n^{0}(\tilde{A} r)$ appear in the shock wave front according to the reaction

$$
\begin{gather*}
\operatorname{Ar}\left\{{ }^{1} S_{0}\right\}+\operatorname{Ar}\left\{{ }^{1} S_{0}\right\}  \tag{2}\\
\longrightarrow \operatorname{Ar}\left\{4 s^{\prime}[1 / 2]_{0}\right\}+\operatorname{Ar}\left\{{ }^{1} S_{0}\right\}-11.72 \mathrm{eV}
\end{gather*}
$$

The experiments were performed with an $\mathrm{Ar}-\mathrm{H}_{2}$ 99: 1 mixture at an initial pressure of $P_{1}=10 \mathrm{Torr}$ and an $\mathrm{He}-\mathrm{Ar}-\mathrm{H}_{2} 50: 49.5: 0.5$ mixture at $P_{1}=20$ Torr. The main processes, occurring in these gas mixtures at temperatures $T_{2}<4000 \mathrm{~K}$ behind the shock wave front provided reaction (2) takes place in the front, are as follows:

$$
\begin{gather*}
\mathrm{H}_{2}+\mathrm{M} \longrightarrow 2 \mathrm{H}\{1 s\}+M,  \tag{3}\\
\tilde{\mathrm{~A} r}+\mathrm{H}\{1 s\} \longrightarrow \operatorname{Ar}\left\{{ }^{1} S_{0}\right\}+\mathrm{H}(3 s)-0.27 \mathrm{eV},  \tag{4}\\
\tilde{\mathrm{~A} r}+2 \mathrm{He} \longrightarrow \mathrm{Ar}\left\{4 s^{\prime}[1 / 2]_{1}\right\}+2 \mathrm{He}-0.1 \mathrm{eV},  \tag{5}\\
\tilde{\mathrm{~A}} \mathrm{H}+2 \mathrm{He} \longrightarrow \mathrm{Ar}\left\{4 s[3 / 2]_{1}\right\}+2 \mathrm{He}+0.1 \mathrm{eV},  \tag{6}\\
\mathrm{H}\{3 s\} \longrightarrow \mathrm{H}\{2 p\}+h v\left(\mathrm{H}_{\alpha}\right), \lambda=656.27 \mathrm{~nm}, \tag{7}
\end{gather*}
$$

$$
\begin{gather*}
\mathrm{H}\{2 p\} \longrightarrow \mathrm{H}\{1 s\}+h v\left(L_{\alpha}\right), \lambda=121.57 \mathrm{~nm},  \tag{8}\\
\operatorname{Ar}\left\{4 s^{\prime}[1 / 2]_{1}\right\} \longrightarrow \operatorname{Ar}\left\{{ }^{1} S_{0}\right\}+h v, \lambda=104.82 \mathrm{~nm},  \tag{9}\\
\operatorname{Ar}\left\{4 s[3 / 2]_{1}\right\} \longrightarrow \operatorname{Ar}\left\{{ }^{1} S_{0}\right\}+h v, \lambda=106.67 \mathrm{~nm} . \tag{10}
\end{gather*}
$$

In this work, we have followed process (7) by monitoring photons in the $\mathrm{H}_{\alpha}$ Balmer line emitted from the shock wave. This provided an estimate from below for the initial concentration of metastable argon atoms appearing in the shock wave front as a result of the translational nonequilibrium. Apparently, the number of photons emitted in the $\mathrm{H}_{\alpha}$ line cannot exceed the number of metastable argon atoms. The number of photons detected by the photomultiplier tube is $N_{h v}\left(\mathrm{H}_{\alpha}\right) \sim$
$\int_{0}^{\tau} U_{\mathrm{PMT}}\left(\mathrm{H}_{\alpha}\right) d t$, where $\tau$ is the light signal duration and $U_{\text {PMT }}$ is the photomultiplier output signal intensity at the $\mathrm{H}_{\alpha}$ line wavelength. When a multichannel spectral analyzer is employed, the number of photons registered at this wavelength is proportional to the analyzer signal amplitude $I\left(\mathrm{H}_{\alpha}\right)$.

Figure 1 shows the plots of the signal intensity $I\left(\mathrm{H}_{\alpha}\right)$ versus the temperature $\mathrm{T}_{2}$ measured in a series of experiments with the OSA-WP4 spectrum analyzer. In these experiments, the possible range of parameters was limited, on the one hand, by the sensitivity of the spectrum analyzer and, on the other hand, by the range of temperatures ( $T_{2} \sim 4000 \mathrm{~K}$ ) in which the model expressed by Eqs. (2)-(10) is valid. The intensity of the extreme left hand point for the curve representing the argon-hydrogen mixture was estimated from the results of measurements performed using the photomultiplier tube. Positions of the experimental points on the temperature scale were determined by the ability to vary the regime by changing the number of diaphragm films. As seen from Fig. 1, the curve for the $\mathrm{He}-\mathrm{Ar}-\mathrm{H}_{2}$ mixture lies by almost one order of magnitude above the curve for the $\mathrm{Ar}-\mathrm{H}_{2}$ mixture.

Previously [1], we used experimental data to determine the average shock wave front temperature $\tilde{T}_{\mathrm{f}}$ corresponding to the measured concentration of metastable argon atoms in terms of the Boltzmann formula $n^{0}(\tilde{A} r)=n \exp \left(-E(\tilde{A} r) / k \tilde{T}_{f}\right)$. This approach is based on the assumption that the population of a metastable level is established simultaneously with the equilibrium with respect to the translational degrees of freedom. This is possible provided that the probability of excitation of the metastable level upon a collision of two atoms is close to unity for the kinetic energies exceeding the energy of this level.

This formal definition does not provide for a clear physical meaning of the average temperature in the shock wave front where no equilibrium distribution of particles with respect to their velocities is established. However, it is known that the so-called "transverse" temperature $T_{\perp}$ in the front smoothly increases from $T_{1}$ to $T_{2}$ [4], while the "longitudinal" temperature $T_{x}$
exhibits, as seen from formula (1), a maximum in the vicinity of $x=0$. Let us calculate the average "longitudinal" temperature $\tilde{T}_{x}$. According to the phenomenological Mott-Smith model, $T_{x}$ tends to $T_{1}$ and $T_{2}$ at the edges of the temperature interval under consideration. Assuming the shock wave front width to be finite and the transitions from $T_{x}=T_{1}$ to $T_{x=0}$ and from $T_{x=0}$ to $T_{x}=T_{2}$ to be linear, we may introduce the average "longitudinal" temperature for the particles with opposite velocities directed along the $X$ axis:

$$
\begin{equation*}
\tilde{T}_{x}=\frac{1}{2 a} \int_{-a}^{a} T_{x} d x \tag{11}
\end{equation*}
$$

Here, $2 a$ is the conventional front width, $T_{x=-a}=T_{1}$, $T_{x=a}=T_{2}$, and $T_{x=0}$ is defined by formula (1). Thus, $T_{x}$ is determined for one-sixth of all particles. For the experiment described in [1], the concentration of metastable argon atoms appearing in the front is as follows:

$$
\begin{gather*}
n^{0}(\tilde{\mathrm{~A}} \mathrm{r}) \\
=\frac{1}{6} \frac{n_{1}+n_{2}}{2} \exp \left[-E(\tilde{\mathrm{~A}} \mathrm{r}) / k \tilde{T}_{x}\right]=2.1 \times 10^{8} \mathrm{~cm}^{-3} \tag{12}
\end{gather*}
$$

Here, the $n(x)$ value in the front is also assumed to vary linearly (from $n_{1}$ to $n_{2}$ ) with the coordinate; $E(\tilde{A} r$ ) being the energy of excitation of a given metastable level. The result calculated by formula (12) agrees well with the experimental value $n^{0}(\tilde{\mathrm{~A}} \mathrm{r})=2 \times 10^{8} \mathrm{~cm}^{-3}$. In this particular case, $\tilde{T}_{x}=6670 \mathrm{~K}$ and $T_{2}=3520 \mathrm{~K}$.

Figure 2 shows the plots of the concentration of metastable argon atoms $n^{0}(\tilde{A} r)$ versus $T_{2}$ calculated by formula (12). As seen, the plot for the $\mathrm{He}-\mathrm{Ar}-\mathrm{H}_{2}$ mixture lies by almost two order of magnitude above that for the $\mathrm{Ar}-\mathrm{H}_{2}$ mixture. A difference between the corresponding experimental plots is one order of magnitude lower. The discrepancy of calculation and experiment can be related to the fact that the calculation did not take into account the loss of metastable argon atoms via channels (5) and (6), which must decrease the $N_{h v}\left(\mathrm{H}_{\alpha}\right)$ value observed for the argon-helium mixture. Thus, experiments of this type allow the total rate constant for reactions (5) and (6) to be determined. The difference between calculation and experiment can be also related to the fact that a multicomponent mixture may


Fig. 2. Calculated plots of the concentration of metastable argon atoms $n^{0}(\tilde{A} r)$ versus temperature $T_{2}$ for (1) He-Ar$\mathrm{H}_{2}\left(50: 49.5: 0.5, P_{1}=20 \mathrm{Torr}\right)$ and (2) $\mathrm{Ar}-\mathrm{H}_{2}$ (99: 1, $P_{1}=10$ Torr) mixtures.
exhibit a partial front shape of the heavy component distorted [5] by interaction with the light component characterized by a very small partial Mach number (as a result, the wave front becomes more extended and loses a jumplike character). In other words, the light component occurring in the state of translational equilibrium produces effective "maxwellization" of the heavy component.

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# Asymmetry in the Nonlinear Resonance Interaction of the Capillary Oscillation Modes of a Charged Droplet 

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#### Abstract

An analytical expression is derived that describes the oscillations of a charged droplet with the initial deformation determined by a superposition of two oscillation modes. It is demonstrated that the fourth and sixth modes exhibit asymmetric energy exchange under the conditions of internal resonance in the second-order term with respect to a small deformation amplitude. © 2000 MAIK "Nauka/Interperiodica".


1. Investigation of the nonlinear interaction of oscillation modes in a charged droplet is an important task for both fundamental research and practical applications of this phenomenon. The original study of the nonlinear capillary oscillations of a charged droplet [1] showed that a system charged below a critical level (corresponding to stability with respect to the intrinsic charge) exhibits an internal nonlinear resonance between the fourth and sixth modes for $\omega_{6}^{2}=4 \omega_{4}^{2}$ in the second order of smallness with respect to the oscillation amplitudes. The presence of this resonance may change the critical conditions for realization of the system instability as compared to those predicted by a linear theory. Therefore, it was important to study this possibility in more detail.
2. Consider a droplet of the ideal perfectly conducting liquid with a density $\rho$ and a surface tension $\sigma$, bearing a charge $Q$. Let a deviation of the droplet shape from spherical (with a radius $R$ ) at the initial instant be determined by a superposition of two linear oscillation modes and characterized by an amplitude $\varepsilon$ and a zero initial velocity. In a spherical coordinate system with the origin at the droplet center, a mathematical description of the initial deformation using dimensionless variables where $\rho=\sigma=R=1$ for a quadratic approximation with respect to the small deformation $\varepsilon$ is provided by the following set of equations:

$$
\begin{gather*}
t=0: r=1+\xi_{0}+\varepsilon\left[h_{k} P_{k}(\mu)+h_{m} P_{m}(\mu)\right] ; \\
\xi_{0} \approx-\varepsilon^{2}\left[\frac{h_{k}^{2}}{(2 k+1)}+\frac{h_{m}^{2}}{(2 l+1)}\right]+O\left(\varepsilon^{3}\right) ;  \tag{1}\\
h_{k}+h_{m}=1 ; \quad \mu=\cos \theta .
\end{gather*}
$$

The volume of the initial deformed droplet is equal to that of the spherical droplet with the radius $R$. Below we consider the problem of nonlinear capillary oscillations of the charged droplet in a formulation analogous
to that used in [2]. However, the problem will be solved using a multiscale method with the initial condition (1), rather than by direct expansion. A solution determining the droplet shape at any instant to within the secondorder terms in the small parameter $\varepsilon$ is as follows:

$$
\begin{gathered}
r(\mu, t)=1+\varepsilon\left\{h_{k} \cos \left(\omega_{k} t\right) P_{k}(\mu)\right. \\
\left.+h_{m} \cos \left(\omega_{m} t\right) P_{m}(\mu)\right\} \\
-\varepsilon^{2}\left\{\frac { 1 } { 2 } \left[\frac{h_{k}^{2}}{(2 k+1)}\left(1+\cos \left(2 \omega_{k} t\right)\right)\right.\right. \\
\left.+\frac{h_{m}^{2}}{(2 m+1)}\left(1+\cos \left(2 \omega_{m} t\right)\right)\right] \\
\left.+\sum_{n=1}^{\infty}\left[N_{n}(t=0) \cos \left(\omega_{n} t\right)-N_{n}(t)\right] P_{n}(\mu)\right\}+O\left(\varepsilon^{3} t\right) ; \\
N_{n}(t)=\frac{1}{2}\left\{h_{k}^{2}\left[\lambda_{k k n}^{(-)}+\lambda_{k k n}^{(+)} \cos \left(2 \omega_{k} t\right)\right]\right. \\
+h_{k} h_{m}\left[\left[\lambda_{k m n}^{(+)}+\lambda_{m k n}^{(+)}\right] \cos \left(\left(\omega_{k}+\omega_{m}\right) t\right)\right. \\
+ \\
\left.\left[\lambda_{k m n}^{(-)}+\lambda_{m k n}^{(-)}\right] \cos \left(\left(\omega_{k}-\omega_{m}\right) t\right)\right] \\
\left.+h_{m}^{2}\left[\lambda_{m m n}^{(-)}+\lambda_{m m n}^{(+)} \cos \left(2 \omega_{m} t\right)\right]\right\} ; \\
\lambda_{m, j, n}^{( \pm)} \equiv \frac{\gamma_{m, j, n} \pm \omega_{m} \omega_{j} \eta_{m, j, n} ;}{\omega_{n}^{2}-\left(\omega_{m}^{ \pm} \omega_{j}\right)^{2}}
\end{gathered}
$$

$$
\begin{gathered}
\gamma_{m, j, n} \equiv k_{m, j, n}\left[\omega_{m}^{2}(n-m+1)+2 n[j(j+1)-1]\right. \\
+n[j(m+1)-m(2 m-2 n+7)+3] W] \\
+\alpha_{m, j, n}\left[\frac{1}{m} \omega_{m}^{2}+n W\right]
\end{gathered}
$$

$$
\begin{aligned}
\eta_{m, j, n} \equiv k_{m, j, n}\left(\frac{n}{2}-m+1\right)+\alpha_{m, j, n} \frac{1}{m}\left(1+\frac{n}{2 j}\right) \\
k_{m, j, n} \equiv\left[C_{m, j, n}^{0,0,0}\right]^{2} \\
\alpha_{m, j, n} \equiv-[m(m+1) j(j+1)]^{1 / 2} C_{m, j, n}^{0,0,0} C_{m, j, n}^{-1,1,0}
\end{aligned}
$$

$$
C_{m, j, n}^{0,0,0}=\left\{\begin{array}{l}
0, \quad \text { for }(m+j+n)=(2 g+1), g \text { is integer; } \\
\frac{(-1)^{g-n} g!(2 n+1)^{1 / 2}}{(g-m)!(g-j)!(g-n)!} \\
\times\left[\frac{(2 g-2 m)!(2 g-2 j)!(2 g-2 n)!}{(2 g+1)!}\right]^{1 / 2} \\
\text { for }(m+j+n)=2 g ; g \text { is integer; }
\end{array}\right.
$$

$$
\begin{gathered}
C_{m, j, n}^{-1,1,0} \equiv(2 n+1)^{1 / 2} n! \\
\times\left[\frac{(m+j-n)!m(m+1)}{(n+m-j)!(n-m+j)!(n+m+j+1)!j(j+1)}\right]^{1 / 2} \\
\times \sum_{p} \frac{(-1)^{m+1+p}(m+p-1)!(n+j-p+1)!}{p!(m-p+1)!(n-p)!(j-n+p-1)!} \\
W \equiv \frac{Q^{2}}{8 \pi} ; \quad \omega_{n}^{2} \equiv n(n-1)[(n+2)-W]
\end{gathered}
$$

In the last expression, the sum is taken over all integers $p$ for which all terms under the factorial sign are nonnegative. The quantities $C_{m, j, n}^{0,0,0}$ and $C_{m, j, n}^{-1,1,0}$ are the Clebsch-Gordon coefficients [3], which differ from zero provided that the bottom indexes satisfy the relationships $|m-j| \leq n \leq(m+j)$ and $(m+j+n)=2 g$, where $g$ is an integer.
3. The coefficients $\lambda_{k k n}^{( \pm)}$and $\lambda_{m m n}^{( \pm)}$differ from zero only for even $n$ belonging to the intervals $[0 ; 2 k]$ and $[0 ; 2 m]$, respectively, while $\lambda_{k m n}^{( \pm)}$and $\lambda_{m k n}^{( \pm)}$take nonzero vales only for $n$ within $[|k-m|,(k+m)]$ such that $n+k+m$ is even. Thus, simultaneous excitation of even and odd modes and their interaction in the second-order term would give rise to a perturbation in all even modes from the interval $[0 ; \max (2 k, 2 m)]$ and all odd modes from the interval $[|k-m|,(k+m)]$. Upon the initial excitation of two even or two odd modes, the second-order term will exhibit only the perturbation in even modes from the interval $[0 ; \max (2 k, 2 m)]$. In particular, for the initial excitation of the fourth and sixth modes (their
resonance is studied below), solution (2) takes the following form:

$$
\begin{aligned}
& r=1+ \varepsilon\left[h_{k} \cos \left(\omega_{4} t\right) P_{4}(\mu)+h_{m} \cos \left(\omega_{6} t\right) P_{6}(\mu)\right] \\
&+\varepsilon^{2}\left[A_{0}(t) P_{0}(\mu)+A_{2}(t) P_{2}(\mu)+A_{4}(t) P_{4}(\mu)\right. \\
&+A_{6}(t) P_{6}(\mu)+A_{8}(t) P_{8}(\mu) \\
&\left.+A_{10}(t) P_{10}(\mu)+A_{12}(t) P_{12}(\mu)\right]
\end{aligned}
$$

The coefficients $A_{j}(t)$ are rather cumbersome and we will write only the expressions for $A_{4}(t)$ and $A_{6}(t)$ that


Fig. 1. Time variation of the amplitudes of oscillation modes of a charged droplet excited in the second-order terms with respect to $\varepsilon=0.1$ in the vicinity of the internal resonance (for $\omega_{6}^{2}=4 \omega_{4}^{2}$ ): (1) zero mode; (2) second mode; (3) forth mode; (4) sixth mode; (5) eighth mode. The initial deformation in the form of a virtual perturbation. Only the sixth mode amplitude exhibits a resonance growth for the initial deformation $\varepsilon$.


Fig. 2. Time variation of the amplitudes of oscillation modes of a charged droplet showing resonance growth of the fourth mode amplitude with the initial deformation $\varepsilon$ : (6) tenth mode; (7) twelfth mode. Other notations as in Fig. 1.


Fig. 3. Time variation of the amplitudes of oscillation modes of a charged droplet showing resonance growth of the fourth and sixth modes with the initial deformations $0.5 \varepsilon$. Notations as in Figs. 1 and 2.
are important in the context of this paper. The other coefficients (readily obtained from Eq. (2)) are omitted for the sake of brevity.

$$
\begin{aligned}
A_{4}(t) & =\frac{108}{1001 \omega_{4}^{2}}\left(\frac{3}{4}(206+W)-(4+17 W) \cos \left(\omega_{4} t\right)\right. \\
- & \left.\frac{1}{4}(602-65 W) \cos \left(2 \omega_{4} t\right)\right) h_{4}^{2}+\frac{15}{286(8-3 W)} \\
& \times\left\{\omega_{6}^{-1}\left[4(4 W+19) \omega_{6}+(240-W) \omega_{4}\right]\right.
\end{aligned}
$$

$$
\begin{align*}
& \times \cos \left[\left(\omega_{4}-\omega_{6}\right) t\right]-8(4 W+19) \cos \left(\omega_{4} t\right) \\
& +\omega_{6}^{-1}\left[4(4 W+19) \omega_{6}-(240-W) \omega_{4}\right]  \tag{3}\\
& \left.\times \cos \left[\left(\omega_{4}+\omega_{6}\right) t\right]\right\} h_{4} h_{6}+\frac{7}{2431}\left\{12 \frac{23 W+572}{\omega_{4}^{2}}\right. \\
& +\frac{24\left(277 W^{2}-1084 W-10568\right)}{\omega_{4}^{2}(74-9 W)} \cos \left(\omega_{4} t\right) \\
& \left.-\frac{(3532-347 W)}{(74-9 W)} \cos \left(2 \omega_{6} t\right)\right\} h_{6}^{2} ; \\
& A_{6}(t)=\frac{15}{198 \omega_{6}^{2}}\{(610-11 \mathrm{~W}) \\
& +16 \frac{\left(1010-37 W-3 W^{2}\right)}{(8-3 W)} \cos \left(\omega_{6} t\right) \\
& \left.-\omega_{6}^{2} \frac{(526+3 W)}{6(8-3 W)} \cos \left(2 \omega_{4} t\right)\right\} h_{4}^{2}-\frac{28}{1683(74-9 W)} \\
& \times\left\{3(56 W-121) \cos \left(\omega_{6} t\right)-\omega_{4}^{-1}\left[1.5(56 W-121) \omega_{4}\right.\right. \\
& \left.+3(397-7 W) \omega_{6}\right] \cos \left[\left(\omega_{6}-\omega_{4}\right) t\right]  \tag{4}\\
& -\omega_{4}^{-1}\left[1.5(56 W-121) \omega_{4}-3(397-7 W) \omega_{6}\right] \\
& \left.\times \cos \left[\left(\omega_{6}+\omega_{4}\right) t\right]\right\} h_{4} h_{6} \\
& -\frac{100}{3553 \omega_{6}^{2}}\left\{-3(23 W+528)+16(11 W+1) \cos \left(\omega_{6} t\right)\right. \\
& \left.+(1568-107 W) \cos \left(2 \omega_{6} t\right)\right\} h_{6}^{2} .
\end{align*}
$$

The internal nonlinear resonance between the fourth and sixth modes is related to the appearance of summands containing terms proportional to $(8-3 W)^{-1}$ in expressions for the mode amplitudes. As seen from Eqs. (2)-(4), the amplitudes $A_{4}(t)$ and $A_{6}(t)$ are quadratic forms of the initial amplitudes $\varepsilon h_{4}$ and $\varepsilon h_{6}$. In the amplitude $A_{4}(t)$, the resonance term $(8-3 W)^{-1}$ appears in the summand proportional to $h_{4} h_{6}$, which implies that a resonance growth of $A_{4}(t)$ will take place only for the initial excitation of both fourth and sixth modes. In the amplitude $A_{6}(t)$, the resonance term enters in the summand proportional to $h_{4}^{2}$, so that a resonance growth of $A_{6}(t)$ is ensured by initial excitation of the fourth mode alone $\left(h_{6}=0\right)$. When only the sixth mode
is excited $\left(h_{4}=0\right)$, no resonance interaction between the fourth and sixth modes takes place. The above considerations are illustrated in Figs. 1-3 showing the plots of dimensionless amplitudes $A_{j}(t)$ versus dimensionless time $t$ for the modes excited in the vicinity of the resonance between fourth and sixth modes as a result of the interaction in the second-order term calculated by Eq. (2) (for $W=W_{*}=2.67$ ). As seen, the sixth mode amplitude grows only upon initial excitation of the fourth mode and the fourth mode amplitude grows only upon excitation of both fourth and sixth modes-in agreement with the above prediction.

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Translated by P. Pozdeev

# Optical Low-Frequency Absorption of Small Metal Particles 

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#### Abstract

Absorption of the electromagnetic radiation by metal particles in a quasistatic region was estimated within the framework of a classical theory. The calculated values agree in the order of magnitude with experimental data on the frequency dependence of the effective absorption factor of silver, gold, and aluminum particles. The size effect in optical absorption in the near IR spectral range is discussed. © 2000 MAIK "Nauka/Interperiodica".


Optical measurements are traditionally used as a means of direct investigation of the properties of small particles. The so-called size effect-that is, dependence of the characteristics of small particles on their dimensions, determines the properties of dispersed media. Investigation of the characteristics of small particles is also of importance for the development of optically active coatings [1, 2].

Measurements of the optical absorption of small metal particles $(R=1-10 \mathrm{~nm})$ in the IR range $(\tilde{v}=$ $10-100 \mathrm{~cm}^{-1}$ ) revealed anomalous behavior, whereby the measured values exceeded by several orders of magnitude the estimates obtained for the electric dipole absorption [3]. Subsequent experiments [4-7] were devoted to the determination of the absorption coefficient $\gamma$ for specially prepared dielectric "pellets" containing a suspension of particles having nearly spherical shapes and approximately equal radii. In these samples, the quantity $\eta=\frac{4}{3} \pi R^{3} n_{p}$ characterizes the fraction of nanoparticles with the concentration $n_{p}$ in the dielectric matrix. For $\eta \ll 1$, the correlation between particles can be ignored and the absorption coefficient can be expressed as $\gamma(R, v)=n_{p} \sigma(R, v)$, where $\sigma$ is the cross section for the wave absorption by the nanoparticle.

Bondar' [8] used the experimental data obtained in [4-6] at $T=4.2 \mathrm{~K}$ and determined by a specially developed method the values of a dimensionless effective absorption factor $K_{\text {eff }}(v)=\sigma(v) / \pi R^{2}$ (see figure). The experimental $K_{\text {eff }}(v)$ curves were explained [8] by the "electrooptical" effect analogous to the FranzKeldysh effect observed in semiconductors exposed to a constant external electric field. It was suggested that the role of the external field in stimulating the absorption of electromagnetic waves is played by an intrinsic electric field of the surface layer of particles. However, according to the results of calculations performed by the density functional method $[9,10]$, this field is concentrated within a very thin surface layer (including
only the uppermost coordination shell of atoms) and decays exponentially on both sides of the surface. Therefore, this effect can be ignored for particles containing even a few atomic coordination shells.

The purpose of this paper is to demonstrate that there is essentially no anomaly in the absorption and the experimental curves constructed in [8] can be satisfactorily explained within the framework of classical electrodynamics taking into account the magnetic dipole absorption and the diffuse scattering of electrons from the particle surface.

Despite the fact that the absorption of electromagnetic waves by small particles was the subject for a large number of theoretical investigations [11-16], the size and frequency dependences of the absorption cross section $\sigma(R, v)$ predicted by these theories turned out to be considerably overstated [2]. This statement refers, for the most part, to attempts at establishing the quan-


Plots of the effective absorption factor $K_{\text {eff }}(\tilde{v})$ for small metal particles: ( 1 ) gold ( $R=70 \mathrm{~nm}$ ); (2) silver $(R=40 \mathrm{~nm}$ ); (3) aluminum ( $R=18.3 \mathrm{~nm}$ ). Solid curves show the results of calculations by formula (3), dashed curves (points) present the experimental data taken from [8].
tum effects in the dipole absorption. Indeed, in the region of cryogenic (helium) temperatures, the effect of the energy level quantization in particles can already be ignored at $R>1.5 \mathrm{~nm}$. The fraction of surface atoms, determined by the particle surface to volume ratio, is also an insignificant factor for particles with the radius $R>2 \mathrm{~nm}$. Therefore, the energetics of these particles can be described using a quasi-classical approach.

Let us consider the IR absorption of small particles with dimensions smaller than the mean free path of electrons in a massive metal (for Al at room temperature, this value is about 30 nm [17]) and much smaller than the electromagnetic wavelength. In this approximation, the field on the particle scale is homogeneous and the estimates can be obtained within the framework of the Mie theory. According to this, the particle absorption cross section is determined by the complex electric ( $\alpha_{\mathrm{e}}$ ) and magnetic ( $\alpha_{\mathrm{m}}$ ) polarizabilities:

$$
\begin{equation*}
\sigma=\frac{4 \pi \omega}{c} \operatorname{Im}\left(\alpha_{\mathrm{e}}+\alpha_{\mathrm{m}}\right) \tag{1}
\end{equation*}
$$

where $c$ is the speed of light in vacuum. For a system of close packed spherical particles, the particle surface is "rough", at least on the atomic scale, and the diffuse scattering of electrons from the particle surface is a physically reasonable factor [16]. The effective frequency of collisions for electrons in a particle is

$$
\begin{equation*}
\frac{1}{\tau_{\mathrm{eff}}}=\frac{1}{\tau_{0}}+\frac{V_{F}}{R}, \tag{2}
\end{equation*}
$$

where $\tau_{0}$ is the electron relaxation time in a massive metal and $v_{F}$ is the electron velocity on the Fermi surface. Estimates for the particles with dimensions in the range studied show that the effective frequency of collisions at $T=295 \mathrm{~K}$ is determined to an equal extent by the electron-phonon scattering in the bulk [17] and the electron scattering from the walls (surface), while at $T=4 \mathrm{~K}$ only the second factor in Eq. (2) is significant.

In the region far from the plasma resonance ( $\omega \ll \omega_{p}$ ), we may use Eq. (1) and definition of the dielectric permittivity $\varepsilon(\omega)=1-\omega_{p}^{2} /\left[\omega_{2}+i \omega / \tau_{\text {eff }}\right]$ to obtain a relationship

$$
\begin{equation*}
K_{\mathrm{eff}}=\frac{12 \pi \tilde{\mathrm{v}}^{2} c R}{\sigma_{D}}\left(1+\frac{16 \pi^{2} R^{2} \sigma_{D}^{2}}{90 A c^{2}}\right), \tag{3}
\end{equation*}
$$

where $\sigma_{D}=\omega_{p}^{2} \tau_{\text {eff }} / 4 \pi$ is the static conductivity, $\omega_{p}^{2}=$ $4 \pi n e^{2} / m, n=\left(4 \pi r_{s}^{3} / 3\right)^{-1}$ is the electron concentration, and $A=\left\{1\right.$ for $\omega \tau_{\text {eff }} \ll 1 ; \omega^{2} \tau_{\text {eff }}^{2}$ for $\left.\omega \tau_{\text {eff }} \gg 1\right\}$.

The estimate for $\omega \tau_{\text {eff }} \ll 1$ was obtained by Wood and Ashcroft in [15], where the absorption coefficient was calculated according to the Maxwell-Garnet theory assuming a small concentration of metal particles in a dielectric matrix and using a linear approximation
with respect to $\eta \ll 1$. The second term in (3), which corresponds to the magnetic dipole absorption (eddy currents), can be reduced to the form $\left(R / 22 r_{s}\right)^{4}$. For Al particles with $R>2 \mathrm{~nm}$, the magnetic component of absorption in (3) dominates over the electric component.

The results of calculations by formula (3) are depicted by solid curves in the figure, showing good quantitative agreement between theory and experiment. The frequency dependence of $K_{\text {eff }}(\tilde{v})$ is rather well reproduced. The values of $K_{\text {eff }}$ exhibit a weak temperature variation, in agreement with the results of measurements reported in [8]. For comparison, we have also performed calculations by the formula

$$
\begin{equation*}
K_{\mathrm{eff}}=\frac{12 B \pi^{2} \tilde{v}^{2} c R^{2}}{v_{F}} \tag{4}
\end{equation*}
$$

following from the calculations taking into account quantum effects in the dipole absorption cross section [13] with neglect of the correlation between one-electron energy levels ( $B \approx 3$; we also took into account the missing factor $2 \pi$ ). These estimates show a satisfactory agreement with the experimental values. However, formula (4) does not take into account the role of eddy currents and, hence, overstates the role of the electric dipole absorption.

In the near IR range ( $\lambda=1 \mu \mathrm{~m}$ ), Bondar' [8] calculated the dimensional dependence of the imaginary part of the specific dynamic polarizability $\alpha_{2}$ for small silver particles with $R=1.5-8 \mathrm{~nm}$ evaporated onto a quartz substrate. Calculations by the formula $K_{\text {eff }}=$ $8 \pi R \varepsilon_{m}^{1 / 2} \alpha_{2} / 3 \lambda$ yield the "experimental" values $K_{\text {eff }}(R) \approx$ 0.03 and 0.04 for $R=1.5$ and 8 nm , respectively. Here $\varepsilon_{m}=\left(1+\varepsilon_{0}\right) / 2$ and $\varepsilon_{0}=2.38$ is the dielectric constant for quartz.

Experimentally, there is virtually no size effect in $K_{\text {eff }}(R)$ in the near IR frequency range, while formula (4) leads to a relationship of the type $K_{\text {eff }}(R) \propto R^{2}$. At the same time, formulas (2) and (3) (for $\omega \tau_{\mathrm{eff}} \gg 1$ ) are indicative of a weak size effect in $K_{\text {eff }}(R)$ in the range of high frequencies and low temperatures, because the role of eddy currents at small $R$ is not as pronounced. Our estimates give $K_{\text {eff }}(R) \approx(1-1.5) \times 10^{3}$ for $R=$ $1.5-8 \mathrm{~nm}$. These estimates are about $1 / 30$ of the experimental values, but the law $K_{\text {eff }}(R) \approx$ const is qualitatively obeyed. The quantitative discrepancy can be related both to the nonspherical shape of particles [18] and to the presence of an oxide film on their surface [19].

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# Instability Development during Motion of a Metal-Insulator Phase Boundary 

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#### Abstract

Diffusion of a magnetic field into a conductor featuring a metal-insulator phase transition caused by Joule heating is theoretically studied. Development of a thermomagnetic instability at the metal-insulator interphase boundary moving in a fast wave regime is described in the linear stage depending on the wavenumber. Effect of the interphase boundary instability on the operation of a fast-speed electric current breaker based on $\mathrm{a}\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ solid solution is assessed. © 2000 MAIK "Nauka/Interperiodica".


Previously [1] we studied the process of magnetic field penetration into a substance featuring a metalinsulator phase transition of the first kind caused by Joule heating. Stationary subsonic solutions were obtained for a one-dimensional problem, corresponding to a system in which the magnetic field profile, interphase boundary, etc., propagate at a constant velocity $U_{s}$, and numerical calculations were performed for a particular case of the $\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ solid solution. This phenomenon can be used in high-power solid-state pulse current breakers of a new type [1, 2]. A most promising operation regime is offered by the fast wave mode [1], whereby $U_{s}$ is lower than the speed of sound in the metal or insulator phases but higher than the adiabatic sound velocity in a mixed (heterogeneous) phase. In this case, the heterogeneous region is missing and the system features a sharp metal-insulator interface [1].

From the standpoint of practical applications, it is important to study the stability of the metal-insulator interphase boundary moving under the conditions of Joule heating of the medium. No usual hydrodynamic instabilities can practically develop in the system since a difference of the phase densities is very small (on the order of $1 \%$ ) and the mass transfer rates in the medium are extremely small. For this reason, dominating instabilities are of the thermomagnetic type. A typical pattern of the inhomogeneous current distribution in the vicinity of the interphase boundary with an allowance for the boundary perturbations is depicted in the figure. As seen, the current lines concentrate at the dips, where the insulating phase goes into the metal, and diverge at the hills, where the metal phase protrudes. The convergence and divergence of the current lines leads to inhomogeneous heating of the $\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ phase with metallic conductivity and gives rise to the perturbation.

It was demonstrated [1] that diffusion of the magnetic field into $\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ is characterized by low velocities of the mass transfer and small values of the
magnetic Reynolds number, which implies that the motion of the medium can be ignored. We are interested only in the fast motion of the phase transition wave in a system featuring a sharp interface. Assuming that each phase possesses a constant conductivity, we may describe the magnetic field diffusion by the following equation:

$$
\begin{equation*}
\frac{\partial B}{\partial t}=\chi \Delta B, \tag{1}
\end{equation*}
$$

where $\chi=c^{2} /(4 \pi \mu \sigma)$ is the magnetic diffusion coefficient, $c$ is the speed of light in vacuum, $\mu$ is the magnetic permeability, $\sigma$ is the conductivity of the $\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ phase in the metallic state, and $\Delta=$ $\partial^{2} / \partial x^{2}+\partial^{2} / \partial z^{2}$. Since the phase transition is caused by the Joule heating of the medium, the velocity of the


Schematic diagram illustrating propagation of a perturbed metal-insulator interphase boundary and the current lines $\mathbf{J}$ corresponding to inhomogeneous heating of the metal phase.
interphase boundary motion is determined by the thermal balance equation:

$$
\begin{equation*}
\frac{\partial Q}{\partial t}=\sigma E^{2} \tag{2}
\end{equation*}
$$

where $Q$ is the specific amount of heat and $E$ is the electric field strength.

The figure schematically illustrates the motion of the metal-insulator interphase boundary at a velocity $U_{s}$ in the substance under the action of a magnetic field with the strength $B_{0}$. The magnetic field is oriented perpendicularly to the plane of the figure and has a constant magnitude in the insulating phase. The interphase boundary perturbations are assumed to be small. Under
the action of Joule heating, the interphase boundary slowly shifts toward the metal phase. It is suggested that the metal-insulator interphase boundary perturbation in the linear stage has the following form:

$$
\begin{equation*}
a=a_{0} \exp [\omega t] \cos k x \tag{3}
\end{equation*}
$$

where $a_{0}$ is the initial perturbation amplitude (small parameter), $k$ is the wavenumber, and $\omega$ is the perturbation amplitude growth increment. In the linear stage, the system parameters must obey the relationship $a \gg \lambda$, where $\lambda$ is the perturbation wavelength.

A solution to the diffusion equation in the metal phase can be found in the following form:

$$
\begin{equation*}
B(x, z, t)=B_{0}\left\{\exp \left[-\frac{z-U_{s} t}{\delta_{1}}\right]+\frac{a_{0} \exp [\omega t] \cos (k x)}{\delta_{1}} \exp \left[-\frac{z-U_{s} t}{\delta_{2}}\right]\right\} \tag{4}
\end{equation*}
$$

where $\delta_{1}$ is the skin layer depth at the unperturbed metal-insulator boundary, $\delta$ is the skin layer depth for the inhomogeneous (i.e., $x$-dependent) magnetic field component, and $B_{0}$ is the magnetic field strength in the insulator. The mobile boundary condition is formulated as $B(x, z, t)=B_{0}$. Upon expanding the left-hand part of the diffusion equation into a Taylor series in the vicinity of point $z=U_{s} t$ and retaining only terms that are linear
with respect to $a_{0}$, we obtain the mobile boundary condition in a simpler form of $z_{M I}-U_{s} t=a$. There are two additional boundary conditions: $B(x,-\infty, t)-=B_{0}$ and $B(x, \infty, t)=0$.

Substituting expression (4) into equation (1) and accomplishing transformations, we obtain an equation for the unknown coefficients:

$$
\begin{align*}
& \frac{U_{s}}{\delta_{1}} \exp \left[-\frac{z-U_{s} t}{\delta_{1}}\right]+\left(\omega+\frac{U_{s}}{\delta_{2}}\right) \frac{a_{0}}{\delta_{1}} \exp [\omega t] \cos (k x) \exp \left[-\frac{z-U_{s} t}{\delta_{2}}\right] \\
= & \frac{\chi}{\delta_{1}^{2}} \exp \left[-\frac{z-U_{s} t}{\delta_{1}}\right]+\left(\frac{\chi}{\delta_{2}^{2}}-k^{2} \chi\right) \frac{a_{0}}{\delta_{1}} \exp [\omega t] \cos (k x) \exp \left[-\frac{z-U_{s} t}{\delta_{2}}\right] . \tag{5}
\end{align*}
$$

Since $a_{0}$ is an arbitrary small number, equation (5) can be divided into two independent equations. The first of these yields an expression for the skin-layer thickness at the unperturbed boundary: $\delta_{1}=\chi / U_{s}$. The second is the quadratic equation $\chi / \delta_{2}^{2}-U_{s} / \delta_{2}-k^{2} \chi-\omega=0$ with the roots

$$
\delta_{2}=\frac{2}{\frac{U_{s}}{\chi} \pm \sqrt{\left(\frac{U_{s}}{\chi}\right)^{2}+4\left(k^{2}+\frac{\omega}{\chi}\right)}}
$$

The boundary conditions are obeyed only for the positive root $\left(\delta_{2}>0\right)$. A relationship between $k$ and $\omega$ can be derived from the thermal balance equation (2). For this purpose, we express $\mathbf{E}$ through $\mathbf{B}$ using the Maxwell equation $\operatorname{rot} \mathbf{B}=-\frac{4 \pi}{c} \sigma \mathbf{E}$ and substitute the solution (4) into Eq. (2). In calculating the square of vector $\mathbf{B}$, we restrict the expansion to terms of the first order with respect to the small perturbation amplitude. Integrating the resulting expression over time from $-\infty$ to $t$, we obtain the following expression for the specific heat:

$$
\begin{gather*}
Q(x, z, t)=\frac{c^{2} B_{0}^{2}}{16 \pi^{2} \sigma \rho}\left\{\frac{1}{2 U_{s} \delta_{1}} \exp \left[-\frac{2\left(z-U_{s} t\right)}{\delta_{1}}\right]\right. \\
\left.+\frac{2 a_{0} \cos (k x)}{\delta_{1}\left[\delta_{2} \delta_{1} \omega+U_{s}\left(\delta_{2}+\delta_{1}\right)\right]} \exp \left[-\frac{z\left(\delta_{2}+\delta_{1}\right)-\left(\delta_{1} \delta_{2} \omega+U_{s}\left(\delta_{2}+\delta_{1}\right)\right) t}{\delta_{2} \delta_{1}}\right]\right\} \tag{7}
\end{gather*}
$$

At the interphase boundary, the temperature must be equal to the phase transition temperature $T_{M I}$. As was demonstrated previously [1], the heat capacity can be considered as constant. Then, the boundary condition with an allowance for perturbations at the interphase
boundary can be written in the following form [1]:

$$
\begin{equation*}
Q(x, z, t)=Q_{M I}=c_{V} \Delta T \tag{8}
\end{equation*}
$$

which yields a relationship

$$
\begin{gather*}
1=\exp \left[-\frac{2\left(z-U_{s} t\right)}{\delta_{1}}\right] \\
+\frac{c^{2} a_{0} \cos (k x)}{\pi \sigma \delta_{1}\left(\delta_{2} \delta_{1} \omega+U_{s}\left(\delta_{2}+\delta_{1}\right)\right)} \exp [\omega t] \exp \left[-\frac{\left(z-U_{s} t\right)\left(\delta_{2}+\delta_{1}\right)}{\delta_{2} \delta_{1}}\right] \tag{9}
\end{gather*}
$$

Expanding the exponent terms in the right-hand part of Eq. (9) into series with respect to $z$, restricting the expansion to terms of the first order in $a_{0}$, and substituting $z=a_{0} \exp [\omega t] \cos k x$ and $\delta_{2}$ from (6), we arrive at a quadratic equation

$$
\begin{equation*}
\frac{4 \delta_{1}^{2} \omega^{2}}{\chi^{2}}-\frac{4 \omega}{\chi}+\left(\frac{1}{\delta_{1}}-\frac{1}{\delta_{1}^{2}}-4 k^{2}\right)=0 \tag{10}
\end{equation*}
$$

with the roots

$$
\begin{equation*}
\omega_{1,2}= \pm \frac{\chi k}{\delta_{1}} \tag{11}
\end{equation*}
$$

Since the initial perturbation was assumed to be small, we should reject the solution with negative increment that corresponds to infinite growth of the perturbation amplitude with time (for $t \longrightarrow-\infty$ ).

As demonstrated previously [1], the velocity of propagation of a stationary wave at $B_{0}=60 \mathrm{~T}$ is approximately $U_{s} \sim 3 \times 10^{5} \mathrm{~cm} / \mathrm{s}$. Assuming the conductivity of the $\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ phase in the metallic state to be $10^{3}(\Omega \mathrm{~cm})^{-1}$, we obtain $\delta_{1} \approx 0.27 \mathrm{~cm}$. This estimate indicates that instabilities with $k>1 \mathrm{~cm}^{-1}$ may significantly develop even for a process with a duration on the order of a few microseconds. The development of instabilities with greater wavenumbers $k$ can be limited by heat conductivity. In particular, the characteristic time of heat transfer between the interphase boundary
depressions and protrusions can be estimated as $(\pi / k)^{2} C_{p} \rho / \alpha$, where $C_{p} \rho$ is the isobaric heat capacity per unit volume and $\alpha$ is the thermal conductivity. The characteristic time of inhomogeneous heating is $\delta_{2} / U_{s}$. When the two characteristic times become comparable, the heat transfer begins to control the perturbation amplitude growth. However, the $\left(\mathrm{V}_{1-X} \mathrm{Cr}_{X}\right)_{2} \mathrm{O}_{3}$ solid solution is characterized by very large wavenumbers $k$ for which the amplitude growth virtually ceases $\left(\sim 10^{8} \mathrm{~cm}^{-1}\right)$. Thus, the heat transfer in this system does not practically limit the perturbation amplitude growth even for large $k$ values. These results indicate that thermomagnetic instability can be a factor significantly affecting operation of a current breaker of the type described in [2].

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# The Drift Resonance and Isolation of Charged Particles in a Toroidal Magnetic Trap Modeled Using the Newton-Lorentz Equation and a Set of the Drift Equations of Motion 

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#### Abstract

The motion of a charged particle in a toroidal-helicoidal magnetic field was modeled. The drift resonance of the particle with a magnetic perturbation and the phenomenon of particle trapping on an inhomogeneity of this field were studied using two approaches, based on solving a set of the drift equations or the exact Newton-Lorentz equation of motion. The two approaches exhibit a good quantitative coincidence in describing the drift resonance and show a qualitative agreement in describing the motion of a trapped particle. The approach based on the exact equations of motion reveals some new features in the phenomenon of particle trapping, which are of importance from the standpoint of determining the neoclassical transfer coefficients. © 2000 MAIK "Nauka/Interperiodica".


Introduction. The behavior of charged particles in variable spatially inhomogeneous magnetic and electric fields can be analyzed using both analytical and numerical methods. Both these approaches exhibit advantages and disadvantages of their own. The analytical approach, based on solving the equations of motion, provides for a general understanding of the process features. However, in the case of a complicated field configuration, this generality is reached at the expense of numerous simplifying assumptions that may be accompanied by the loss of important and interesting partial solutions. On the other hand, the numerical approach is helpful in the analysis of partial solutions. The obtained empirical features may stimulate the analytical investigation, which makes the two approaches-analytical and numerical-mutually complementary.

The numerical analysis is based on the combination of theoretical models of a given phenomenon with numerical methods. The models and methods are selected so as to provide for an optimum relationship between the computational time and accuracy. Analysis of the results obtained using various models and computation methods allows the reliability of these results to be assessed, which is an important part of each numerical model investigation.

The purpose of this study was to compare the results obtained by numerically solving two sets of the model equations of motion of a charged particle in a toroidalhelicoidal magnetic field for various approximations and particular cases. The two models represent the Newton-Lorentz equation and a set of the drift equations of motion [1]. The sets of differential equations were numerically solved by the Adams finite-difference method using the PECE scheme [2].

The Newton-Lorentz equation and a set of the drift equations. The motion of a charged particle in an electromagnetic fields obeys the Newton-Lorentz equation,

$$
\begin{equation*}
\frac{d \mathbf{p}}{d t}=e \mathbf{E}+\frac{e}{c}[\mathbf{V B}], \tag{1}
\end{equation*}
$$

which follows from the most general physical principles and offers exact description within the framework of the classical mechanics [3].

In the case of electric and magnetic fields representing arbitrary functions of space coordinates and time, this differential equation cannot be integrated in analytical functions. Therefore, practical results can be obtained using various approximations, one of these being called the drift approximation.

The drift approximation consists in averaging the exact equation of motion (1), as suggested by Bogolyubov and Krylov [4], under the assumption that (i) the Larmor radius is small compared to the field inhomogeneity scale, (ii) the field variation frequency is small compared to the Larmor frequency, and (iii) the electric field strength is sufficiently small [1]. The averaging eliminates fast Larmor oscillations of a particle and the final equations of motion acquire (for $[\nabla \mathbf{B}]=0$ ) the following form:

$$
\begin{gather*}
\frac{d \mathbf{r}}{d t}=V_{\|} \frac{\mathbf{B}}{B}+\frac{c}{B^{2}}[\mathbf{E} \mathbf{B}]+\frac{m_{j} c\left(2 V_{\|}^{2}+V_{\perp}^{2}\right)}{2 e B^{3}}[\mathbf{B} \nabla B], \\
\frac{d W}{d t}=e \mathbf{E} \frac{d \mathbf{r}}{d t}+\frac{m_{j} V_{\perp}^{2}}{2 B} \frac{\partial B}{\partial t},  \tag{2}\\
\frac{d \mu}{d t}=0,
\end{gather*}
$$



Fig. 1. The projection of a trajectory of a charged particle, moving in a toroidal-helicoidal field, onto the meridional cross section of the tore. The numerical calculations were performed using (a) a set of the drift equation and (b) the Newton-Lorentz equation.
where $\mathbf{r}$ is the radius-vector of the particle center; $W$ is the kinetic energy; $V_{\|}$and $V_{\perp}$ are the particle velocity components parallel and perpendicular to the magnetic field; $m_{j}$ and $e$ are the particle mass and charge, respectively; and $\mu$ is the transverse adiabatic invariant.

Although realization of a numerical solution of the set (2) presents a more complicated task as compared to the case of Eq. (1), the drift approximation provides for a considerable reduction in the overall computation time. This economy is related to the fact that the drift approximation neglects the Larmor oscillations of the particle, thus making the computation step markedly greater as compared to that necessary for a numerical solution of the exact equation (Fig. 1). In our case, the particle moves in a helicoidal magnetic field of a toroidal torsatron trap with $L=3$ [5]. Small turns depicted with magnification in the bottom panel of Fig. 1 correspond to the Larmor motions. The top panel shows a part of the particle trajectory (frequently called the
hairpin bend). In what follows, bottom panels in the figures show magnified portions of the trajectories indicated by frames in the top panels.

Below, we will demonstrate reliability of the results obtained by numerically solving the set of drift equations (2), as confirmed by comparison with a numerical solution of the exact equation (1). Both models-exact equation and drift approximation-of the particle motion in the electromagnetic field refer to the electromagnetic fields of thermonuclear toroidal traps (torsatrons). We will consider two important physical phenomena for particles moving in such a toroidal-helicoidal field: the drift resonance of a charged particle with a magnetic perturbation [5] and the effect of particle trapping on an inhomogeneity of the field.

Drift resonance. An analysis of the motion of a charged particle in a toroidal-helicoidal magnetic field shows that the equations of motion in the drift approximation are essentially the equations for a nonlinear


Fig. 2. "Drift islands" (with a drift rotation angle $l^{*}=1 / 3$ ) representing the traces of the charged particle trajectory in the meridional cross section of the tore. The numerical calculations were performed using (a) a set of the drift equation and (b) the Newton-Lorentz equation.
oscillator, the drift angle of the particle rotation over the tore playing the role of the intrinsic frequency. The particle trajectory trace covers a certain toroidal surface.

In the presence of a helicoidal magnetic perturbation, with the spatial frequency being a multiple of the intrinsic frequency of the effective nonlinear oscillator, the system exhibits a resonance. The resonance is manifested in the appearance of a helicoidal tube, formed by the particle trajectory, lying on the toroidal surface. In the meridional cross section of the tore, the tube appears as a set of islands (called the drift islands).

A question naturally arises as to whether the resonance is manifested in solving the exact equation of motion (1) as well, or whether this is only a consequence of the drift approximation. The results of numerical modeling of the particle motion in a toroidalhelicoidal field using the Newton-Lorentz equation show that a resonance of the charged particle with helicoidal perturbation really does takes place (Fig. 2).


Fig. 3. Motion of the "drift islands" in a toroidal-helicoidal field with the amplitude varying at a frequency of 950 Hz . The numerical calculations were performed using (a) a set of the drift equation and (b) the Newton-Lorentz equation.

Using the numerical solution to Eq. (1), it is possible to study the fine effect of the "drift island motion" caused by a change in the amplitude of the helicoidal magnetic field [5]. Moreover, this can be done with an allowance for the Coulomb interaction of particles in the thermonuclear plasma (Fig. 3) [6]. The Coulomb interaction of a probing particle with plasma species takes place at a plasma density of $10^{14} \mathrm{~cm}^{-3}$ and a temperature of 10 keV at the center of the magnetic configuration.

The numerical investigation was performed for various magnitudes and shapes of the plasma density profile and various frequencies of the helicoidal magnetic field. The results of the model calculations show a good agreement between the numerical solutions of Eq. (1) and set (2).

Trapped particles. Moving along the tore, a particle with a small ratio of the longitudinal velocity component (i.e., that parallel to the magnetic field) to the


Fig. 4. The projection of a trajectory of a charged particle trapped on the toroidal inhomogeneity of the toroidal-helicoidal magnetic field onto the meridional cross section of the tore. The numerical calculations were performed using (a) a set of the drift equation and (b) the Newton-Lorentz equation. Symbol I indicates the particle reflection points.
total velocity $\left(V_{\|} / V\right)$ exhibits refections from the regions of strong magnetic field. This is the so-called trapped particle. According to the shape of the drift trajectory, we may distinguish the particles trapped on a toroidal inhomogeneity and those trapped on a helicoidal inhomogeneity of the toroidal-helicoidal magnetic field.

It would by interesting to compare the trajectories of a trapped particle obtained by solving the exact Eq. (1) and the approximated set (2) (Fig. 4a). Figure 4b (bottom) shows a magnified fragment of the corresponding hairpin bend trajectory, with indicated points where the particle is reflected from the regions of strong magnetic field. Note that the trajectories obtained using Eqs. (1) and (2) exhibit only a qualitative similarity. This (i.e., the lack of quantitative coincidence) is explained, first, by uncertainty the drift approximation and, second, by the impossibility of exactly matching the initial velocities in the problems described by Eqs. (1) and (2) ( $V_{\|}^{0}$, $V_{\perp}^{0} \Rightarrow V_{r}^{0}, V_{\vartheta}^{0}, V_{\phi}^{0}$. The latter implies that the drift approximation represents a generalization of the class of partial solutions with various directions of the transverse component of the initial velocity $\left(\mathbf{V}_{\perp}^{0}\right)$, whereas the exact partial solutions differs for the trapped particles.

Conclusions. An analysis of the results of our investigation leads to the following conclusions:
(1) For a charged particle moving in a toroidal-helicoidal field, the drift resonance with a helicoidal magnetic perturbation takes place even when the particle motion is modeled using the exact Newton-Lorentz equation. Therefore, this phenomenon is not a consequence of the drift approximation.
(2) The drift approximation provides for a sufficiently good description of the fine effect of the "drift island motion" in a variable helicoidal magnetic field with an allowance for the Coulomb interaction of particles in a thermonuclear plasma trap.
(3) The results of numerically solving the NewtonLorentz equation and the set of the drift equations show only a qualitative agreement in the case of trapped particles. Therefore, analysis of a solution to the exact Newton-Lorentz equation, ensuring more precise quantitative results, is preferred for a numerical investigation of the motion of trapped particles. Thus, a methodological value of using the Newton-Lorentz equation in modeling the particle motion consists in that this approach (in contrast to that based on the drift approximation) does not lose important partial solutions. The results of our study are indicative of the need to refine the neoclassical transfer coefficients that should be calculated with an allowance for the deviation of orbits of the trapped particles.

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# Electric Charge Affects the Formation of a Wavy Microrelief on the Surface of a Viscoelastic Medium 

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#### Abstract

The electric charge transferred with a heavy-current ion beam obliquely incident on the surface of a viscoelastic medium influences the formation of a wavy microrelief on the irradiated surface. © 2000 MAIK "Nauka/Interperiodica".


1. Development of nanoelectronic technologies poses many theoretical problems, including the task of establishing the laws of formation of a wavy relief on the initially flat charged surface of a viscoelastic material under the action of external factors giving rise to the normal and tangential surface stresses. For example, the sputtering of a semiconductor ( $\mathrm{Si}, \mathrm{Ge}, \mathrm{GaAs}$ ) target by a medium-energy $(\sim 1-10 \mathrm{keV})$ ion beam with a current density of $j \sim 1-10 \mathrm{~mA} / \mathrm{cm}^{2}$ incident at an angle deviating from normal to the surface leads to the formation of an ordered wavy relief with a characteristic scale on the order of $1 \mu \mathrm{~m}$ (see, e.g., experimental results reported in [1-4]).

The existing theoretical models (see $[2,5-8]$ and references therein) are not always adequate to the real experimental conditions studied. In particular, the role of an important factor such as the electric charge accumulated on the irradiated surface was never taken into account. However, the ion bombardment may lead to development of the instability of a subsurface semiconductor layer (amorphized under the ion beam action) with respect to the accumulated charge. This problem is of interest from the standpoint of assessing prospects for using the beam-induced ordered wave microrelief on the semiconductor surfaces in solving the practical tasks of nanoelectronics [1-4].

Let us estimate the strength of the electric field at the semiconductor surface bombarded by an ion beam. For a beam current density of $j \sim 10 \mathrm{~mA} / \mathrm{cm}^{2}$, the rate of charge delivery per unit surface area $\left(1 \mathrm{~cm}^{2}\right)$ is $\sim 10^{-2} \mathrm{C} / \mathrm{s}$. A characteristic time of the Maxwell charge relaxation is determined by the well-known expression $\tau \sim \varepsilon_{0} \varepsilon \eta^{-1}$, where $\varepsilon_{0}$ is the dielectric constant, $\varepsilon$ is the relative permittivity of the substance studied (in most semiconductors, $\varepsilon \sim 10$ ), and $\eta$ is the specific conductivity of this substance. The specific conductivity of a semiconductor amorphized by an intense ion bombardment is determined by the formula [9]

$$
\eta=\eta_{*} \exp (-E / k T)
$$

where $E$ is the activation energy, $k$ is the Boltzmann constant, $T$ is the absolute temperature, and $\eta_{*}$ has the meaning of conductivity at the infinite temperature.

For silicon-a semiconductor in which most of the experiments with the wave relief formation were con-ducted-the activation energy in the amorphous state is rather small $(E=0.75 \mathrm{eV})$ and the conductivity at $T \approx$ 300 K is on the order of $\eta \sim 10^{-7} \mathrm{~S} / \mathrm{m}$ (extrapolation of the data reported in [9]). Then, the characteristic time of the Maxwell charge relaxation on the surface of silicon amorphized by the intense ion bombardment can be estimated at $\tau \sim 10^{-3} \mathrm{~s}$. This time is sufficient for complete relaxation of the electric charge transferred by the ion beam, so that the semiconductor surface in the dynamic equilibrium will acquire a certain surface charge density $\kappa$. The corresponding electric field strength $E$ at the ion-irradiated surface of the amorphized silicon target for the given physical parameters is on the order of $10^{6} \mathrm{~V} / \mathrm{m}$. This value is sufficient for the development of instability of a viscoelastic amorphized surface with respect to its own charge.
2. For the electrohydrodynamic analysis below, we will consider a flat layer of a viscoelastic conducting fluid (amorphous semiconductor) with density $\rho$, surface tension $\gamma$, kinematic viscosity $\nu_{0}$, viscosity relaxation time $\tau$, and thickness $d$ on a solid substrate occurring in the gravitational force field $\mathbf{g}$. We assume that the free surface is subjected to a continuous action of an oblique (deviating from the surface normal) momentum flux transmitted with the ion beam incident on the surface. The task is to determine the spectrum of wave motions appearing on the free surface and analyze these motions with respect to their temporal stability on the surface homogeneously charged at the surface density $\kappa$.

Let $\Pi_{j k}=\delta V_{j} V_{k}$ be the tensor of the momentum flux density of the external action in the region over the amorphized layer surface. Here, $V_{j}$ are the velocity components of the beam incident at an angle $\beta$ relative
to the surface normal and $\delta$ is the volume density of the layer. We will solve a two-dimensional problem to calculate the spectrum of motions in the medium relative to a Cartesian system $X O Z$ with the $O Z$ axis oriented upward and parallel to the normal ( $\mathbf{n}_{z} \|-\mathbf{g}$ ), with the solid bottom of the layer situated at $z=-d$. The equation describing the shape of the perturbed free surface of the layer is $z=\xi(x, t),|\xi| \ll d$. We will assume that phenomena related to the supply of substance to the system can be ignored. The perturbation $\xi(x, t)$ of the initially flat surface will be considered in the form of a traveling wave with a wavenumber $k$ and a complex frequency $s: \xi \sim \exp (s t-i k x)$. The viscous and elastic properties of the medium are described using the complex viscosity $v$ defined by the Maxwell formula [10]: $v=v(1+s \tau)^{-1}$.

A solution to the problem outlined above can be found by a method analogous to that used in similar electrohydrodynamic situations [7, 8, 11]. In this way, we readily obtain the following dispersion equation (written in the dimensionless coordinates $\rho=\gamma=g=1$ ):

$$
\begin{gathered}
k^{2} q\left(4 s\left(k^{2}+q^{2}\right)-(3 k+q) \frac{W}{v}\right) \\
+\frac{s \omega_{0}^{2}}{v^{2}}(k \cosh (k d) \sinh (q d)-q \sinh (k d) \cosh (q d)) \\
-2 k^{3} q\left(2 s-\frac{W}{v}\right)(k \cosh (k d) \cosh (q d) \\
-q \sinh (k d) \sinh (q d))+\left(k^{2}+q^{2}\right)\left(s\left(k^{2}+q^{2}\right)-k^{2} \frac{W}{v}\right) \\
\times(k \sinh (k d) \sinh (q d)-q \cosh (k d) \cosh (q d))=0 \\
q^{2} \equiv k^{2}+s / v ; \quad w \equiv 4 \pi \kappa^{2} \\
W \equiv-(\delta / \rho) \sqrt{\rho /(g \gamma)} V^{2} \cos (2 \beta) \\
\omega_{0}^{2} \equiv k\left(1+k^{2}\right)-w k^{2}
\end{gathered}
$$

This dispersion equation is rather cumbersome and the behavior of branches corresponding to various sets of parameters should be analyzed by numerical methods. In the case of $\tau=0$, we deal with a usual strongly viscous fluid. Here, the dispersion equation possesses a complex solution describing the capillary motions capable both of the Tonks-Frenkel instability $\left(\omega_{0}<0\right)$ [11] and of a shear instability caused by the force action of the obliquely incident ion beam [7]. In addition, this equation has an infinite number of real roots describing the decrements of aperiodic motions arising when the capillary motions are reflected from the layer bottom [11]. If the fluid possesses elastic properties $(\tau \neq 0)$, another infinite family of decaying aperiodic solutions appears that describes the elastic waves reflected from the bot-


Fig. 1. Plots of the real $\operatorname{Re} s(W)$ and imaginary $\operatorname{Im} s(W)$ components of the dimensionless complex frequency $s$ versus the dimensionless parameter $W$ calculated for $\tau=0, k=1$, $k d=1, v=0.5$ and various values of the parameter $w$. The first index at a curve corresponds to the branch number (two branches are depicted) and the second index refers to the value of the parameter $w=0(1), 1(2), 2(3)$, and $3(4)$ used in the calculation.
tom. The solutions of these types were analyzed previously $[8,11]$ and are not considered in this paper, nor will the corresponding branches be depicted (so as not to overload figures) in the plots illustrating the results of numerical analysis of the system studied.

Figure 1 shows the plots of the real and imaginary components of the complex frequency $s$ versus the dimensionless parameter $W$ characterizing stability of the free surface of a viscous $(\tau=0)$ fluid with respect to shear stresses [7, 8]. The plots refer to various values of the parameter $w$ describing stability of the surface with respect to the intrinsic charge [11]. In this figure, the branches 1-3 (first index) correspond to capillary motions of the fluid (the second figure in the branch index refer to various values of the parameter $w$ used in the calculation).

As the surface charge density increases, the critical value of the parameter $W$ (corresponding to the onset of the instability development) shifts toward negative region (Fig. 2). This corresponds (in complete agreement with the experimental data $[1-4]$ ) to the extension toward the region of smaller (compared to the interval


Fig. 2. Relationship between the critical values of the parameters $W$ and $w$. This plot divides the plane of parameters ( $W ; w$ ) into the regions of stable (to the left) and unstable (to the right) states.
$\beta \in(\pi / 4 ; \pi / 2)$ obtained in the approximation neglecting the surface charging [7]) values of the ion beam incidence angle at which an ordered wavy relief appears on the irradiated surface. The limitation with respect to the maximum beam incidence angle is related to the parameter $W$ (or the ion beam energy). The upper limit for the incidence angle ( $\beta=\pi / 2$ ) was obtained in the approximation neglecting the ion beam refraction; a real limiting value has to be smaller, as was noted in [1-4]. According to the experimental data reported in [4] for an ion beam energy of 9 keV , the ordered wave microre-
lief was formed in the range of incidence angles from $39^{\circ}$ to $70^{\circ}$. This range tended to narrow with an increasing ion beam energy, in agreement with predictions of the model proposed above.

As for the oscillatory instability that can be developed if the elastic properties of the medium are taken into account [8], the results of our numerical calculations (Fig. 3) show that the presence of the surface charge leads to more rigid critical conditions concerning the instability development. This is manifested by an increase in the $W$ values for which the instability can develop (whereby branches 2 and 4 in Fig. 3 emerge to the region Res >0). The increment of this instability increases with the surface charge density (i.e., with the $w$ value). This is illustrated in Fig. 3, where branches 1-4 correspond to the viscoelastic wave motions; branches 6 and 7 , to the aperiodic decaying motions; and branch 5 describes the aforementioned aperiodic instability with respect to shear.

Conclusion. Taking into account the effect of the semiconductor target surface charging by the ion beam sputtering the material ensures the model predictions are consistent with the experimental results. Moreover, experimental data on the range of the ion beam incidence angles for which the development of a wave microrelief is observed can be used to estimate the conductivity of the semiconductor amorphized by the ion beam bombardment, which may significantly differ


Fig. 3. Plots of the real $\operatorname{Re} s(W)$ and imaginary $\operatorname{Im} s(W)$ components of the dimensionless complex frequency $s$ versus the dimensionless parameter $W$ calculated for $\tau=0.4, k=1, k d=1, v=0.5$ and two different values of the parameter $w=0$ (a); 50 (b). See the text for explanation of the curve numbers.
from the conductivity of the same semiconductor in the crystalline state.

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# Photoluminescence of Porous Silicon Layers Formed in Ion-Implanted Silicon Wafers 

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#### Abstract

Implantation of the $\mathrm{B}^{+}$and $\mathrm{N}^{+}$ions or a $\mathrm{B}^{+}+\mathrm{N}^{+}$combination into silicon substrates affects the photoluminescence properties of porous silicon (por-Si) layers prepared on the ion-modified wafers. The postimplantation anneals lead to significant changes in the por-Si emission bands. Models explaining the observed phenomena are suggested. © 2000 MAIK "Nauka/Interperiodica".


Beginning with the discovery of intense luminescence from porous silicon (por-Si), much effort has been spent to obtain stable por-Si-based light-emitting structures with controlled spectroscopic characteristics [1]. The main tasks of these investigations were to increase the stability of por-Si and to find a means of controlling the luminescent properties of this material, that is, the position and intensity of the emission band. Attempts of reaching these goals are most frequently based on using various post-anodizing treatments of the por-Si surface [1].

We believe that a promising way for solving the above tasks is offered by the methods of ion implantation into the por-Si layers and/or into the initial silicon wafers. This is justified, on the one hand, by the requirement of decreasing the resistance of a por- Si layer by introducing doping impurities [2], which is necessary in order to obtain effective electroluminescent structures based on por-Si. On the other hand, attempts at reaching the same purpose by using a traditional diffusion process involving a high-temperature impurity activation often irreversibly decrease the photoluminescence (PL) intensity and change the spectral position of the PL band [2]. The main advantage of ion implantation is apparently the possibility of controlled impurity introduction virtually at room temperature. A disadvantage is the production of defects in the course of ion bombardment, which implies the need for the postimplantation annealing. As was demonstrated [2], the intensity of PL from por-Si usually decreases both during the dopant implantation and in the course of subsequent annealing. The implantation of inert gas ions into por-Si always decreases the intensity of emission from this material [2, 3]. These experiments showed that the radiation stability of por-Si is markedly higher as compared to that of a single-crystalline silicon target [4].

An alternative method of por-Si doping by applying an organic solution containing the dopant, followed by rapid thermal annealing, was suggested by Pastukhov [5]. Such a treatment, involving a combined doping with boron and phosphorus, led to a considerable increase in the PL intensity without significantly changing the spectral position of the emission band.
Apparently, another effective means of controlling the luminescent properties of por-Si consists in the modification of the initial silicon surface (prior to the anodic treatment), in particular, by ion implantation [6-8]. The intense blue PL from por-Si prepared by anodizing silicon wafers implanted with carbon ions was explained by the formation of $\beta$-SiC precipitates (leading to a decrease in the length of silicon fibers) [6] and even by the emission from por-SiC [7]. Pavesi et al. [8] suggested that the porosity of silicon can be controlled by implanting Si ions into the initial wafer. In this context, the purpose of our work was to study the effect of preanodizing implantation of $\mathrm{B}^{+}$and $\mathrm{N}^{+}$ions on the PL properties of por-Si.

The initial (100)-oriented p-Si wafers (KDB-10 grade) with a resistivity of $10 \Omega \mathrm{~cm}$ were modified by implanting $\mathrm{B}^{+}$or $\mathrm{N}^{+}$ions or by a combined implantation of the ions of both types into the front side of a wafer. The ions were implanted at an energy of $E=$ 175 keV to a fluence of $2 \times 10^{15} \mathrm{~cm}^{-2}$ (for the combination, the total fluence was $4 \times 10^{15} \mathrm{~cm}^{-2}$ ). The ion beam current density did not exceed $0.1 \mu \mathrm{~A} / \mathrm{cm}^{2}$. The por-Si samples were prepared using the conventional method of electrochemical etching. Prior to this, an aluminum contact film was applied onto the rear side of the wafer. Then, por-Si layers were formed by anodic etching of the wafer in a teflon (PTFE) cell with a platinum electrode. The process was conducted in the dark using a $48 \%$ aqueous solution of the $\mathrm{HF}-\mathrm{C}_{2} \mathrm{H}_{5} \mathrm{OH}(1: 1)$ mixture. The etching was performed for 5 min at a current


Fig. 1. The PL spectra of por-Si formed on (1) the initial (unimplanted) silicon wafer and on the silicon implanted with (2) $\mathrm{B}^{+}$or (3) $\mathrm{N}^{+}$ions and (4) with the combination $\mathrm{B}^{+}+\mathrm{N}^{+}$.
density of $30 \mathrm{~mA} / \mathrm{cm}^{2}$. Then, the samples were placed into a special setup and subjected to a pulsed thermal annealing in an argon atmosphere for 30 s at 800 K . The PL spectra were excited by a nitrogen laser $(\lambda=$ 337.1 nm ) producing pulses with a duration of 10 ns and a power of 5 kW .

Figure 1 shows the typical spectra of PL from porSi formed on the initial (unimplanted) silicon wafer and on the samples implanted with $\mathrm{B}^{+}$or $\mathrm{N}^{+}$ions and with their combination (curves 1-4, respectively). The PL spectrum of the unimplanted por-Si has a characteristic shape with a maximum at $660-670 \mathrm{~nm}$, which is related to the quantum-confinement effect manifestation. The spectra of por-Si samples prepared using the ionimplanted wafers are very slightly shifted toward lower energies.

The samples ion-implanted with $\mathrm{B}^{+}$ions (Fig. 1, curve 2 ) and $\mathrm{N}^{+}$(curve 3) show a considerable decrease in the total PL intensity. This effect, analogous to that observed in a-Si:H films obtained in the glow discharge [9] can be due to the internal electric fields (created by charged donors and acceptors) decreasing the probability of recombination in the couples of charge carriers responsible for the PL. A decrease in the PL intensity may be also related to the ion-implantationinduced defects providing for an additional channel of the nonradiative recombination.

This last assumption is consistent with the shortwave shift observed in the PL spectra of por-Si formed on the material ion-doped with $\mathrm{B}^{+}$or $\mathrm{N}^{+}$ions. It was demonstrated [10-12] that the quantum confinement effect is manifested in the optical characteristics of nanoparticles by a shortwave shift of their optical spectrum upon the adsorption of charged particles. There are different explanations of this effect. The shift can be attributed to (i) an increase in the exciton energy


Fig. 2. The effect of rapid thermal annealing ( 30 s at 800 K ) on the PL spectra of por-Si formed on (1) the initial (unimplanted) silicon wafer and on the silicon implanted with (2) $\mathrm{B}^{+}$or (3) $\mathrm{N}^{+}$ions and (4) with the combination $\mathrm{B}^{+}+\mathrm{N}^{+}$. The intensity is multiplied by the factor $(1,3,4) 10$ and (2) 2 .
under the action of an internal electric field of excess charges [10]; (ii) a decrease in the oscillator strength for the exciton transition under the action of trapped electrons and holes [11]; or (iii) the excess electrons occupying lower vacant levels of the nanoparticle, whereby a greater energy is required for exciting the electron to a higher valence level [12].

Another evidence in favor of using ion implantation is that the PL spectrum of por-Si formed on the silicon wafer upon the combined $\mathrm{B}^{+}+\mathrm{N}^{+}$implantation (Fig. 1, curve 4) exhibits a growth in the emission intensity. This fact is most probably related to the compensation effects (that is, to the formation of donor-acceptor complexes involving doping impurities) and, hence, to the formation of an additional recombination-active channel.

In the following stage of investigation, we performed rapid thermal annealing (RTA) by heating the samples for 30 s to 800 K in an argon atmosphere. As is seen from Fig. 2, this treatment results in the lowenergy shift of the long-wavelength emission line in the spectra of both the unimplanted sample and that doped with $\mathrm{B}^{+}+\mathrm{N}^{+}$, which can be explained by the annealinginduced coarsening of nanostructures. As is known [13], por-Si possesses a laminar structure manifested in a distribution of the silicon filament diameter from small (at the surface) to large (in the bulk). Upon thermal annealing, the thin surface filaments lose radiative ability and their emission occurs for the most part from deeper layers containing thick filaments. The drop in the PL intensity in the long-wavelength range is typical of anneals in the temperature range employed [14].

In addition to the usual long-wavelength emission at $650-700 \mathrm{~nm}$, the RTA results in activation of the PL band at 450 nm . The high-energy emission can be explained within the framework of the model proposed
by Kanemitsu [14]. According to this, the RTA at 800 K leads to the rupture of hydroxyl $(\mathrm{OH})$ groups. As a result, a structure of the silicon filament $-\mathrm{SiO}_{x}$ layer is formed in the inner por-Si layers. This gives rise to a local distortion of the electron states of the quantum filaments, which accounts for the high-energy PL from the annealed por-Si samples. The samples implanted with $\mathrm{B}^{+}$ions exhibit a very sharp increase in the PL intensity at 450 nm (Fig. 2, curve 2). Apparently, the RTA process electrically activated boron atoms in the subsurface layer of por-Si. The segregation of boron atoms at the silicon fiber- $\mathrm{SiO}_{x}$ interface may lead to the appearance of additional energy levels in this region, which would result in the additional high- energy PL. At the same time, this sensitization effect is not as pronounced in the samples implanted with a $\mathrm{B}^{+}+\mathrm{N}^{+}$combination (Fig. 2, curve 4), which is probably explained by a greater total implantation dose leading to a greater amount of unannealed defects.

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# The Barkhausen Effect in a Hydrogen-Saturated $\mathrm{Fe}-\mathrm{B}-\mathrm{Si}-\mathrm{Ni}$ Amorphous Alloy 

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#### Abstract

Is is established that saturation of a $\mathrm{Fe}_{78} \mathrm{~B}_{12} \mathrm{Si}_{9} \mathrm{Ni}_{1}$ amorphous alloy ribbon with hydrogen leads to a several-fold increase in the emf observed under the Barkhausen effect conditions. The magnitude of the observed effect is related to the hydrogen content in the alloy. © 2000 MAIK "Nauka/Interperiodica".


Despite a considerable number of publications concerning the interaction of hydrogen with amorphous metal alloys, only a few of these are devoted to studying the properties of samples in the course of or immediately upon the hydrogenation process.

The very first investigations in this direction revealed a number of effects previously unknown in the physics of hydrogen interaction with the amorphous metal alloys. These include a several-fold increase in the creep rate in a material loaded to a level considerably below the yield point [1], the loss of mechanical strength of the amorphous metal alloys oversaturated with hydrogen, the so-called Pen-X effect [2], etc.

Evidently, these changes in the deformation properties of amorphous metal alloys must be related to considerable changes in their structure. However, a very fast rate of the transition of an amorphous metal alloy from the state of hydrogen oversaturation to that where the sample is virtually free of hydrogen hinders the study of changes occurring in the metal. This refers both to the conventional methods of structural investigations and to the study of physical properties. Indeed, the duration of measurements must be sufficiently small to introduce no significant errors related to the time instability of the state of a hydrogen-saturated amorphous metal alloy.

It was found that a convenient parameter is offered by the emf $\left(\varepsilon_{\mathrm{B}}\right)$ accompanying the Barkhausen effect measurements, which has proved to be a reliable characteristic of various alloys, especially, crystalline ones [3, 4].

The experimental procedure involved the two-sided hydrogen saturation of a $30-\mu \mathrm{m}$-thick amorphous metal ribbon made of a $\mathrm{Fe}_{78} \mathrm{~B}_{12} \mathrm{Si}_{9} \mathrm{Ni}_{1}$ (2HCP grade) alloy. The hydrogenation was effected in an acid electrolyte medium ( $1 \mathrm{~N} \mathrm{H}_{2} \mathrm{SO}_{4}+100 \mathrm{mg} / \mathrm{l} \mathrm{As}_{2} \mathrm{O}_{3}$ ) at a cathode current density of $50 \mathrm{~A} / \mathrm{m}^{2}$. The $\varepsilon_{\mathrm{B}}$ measurements were performed on a SKIF-1 setup [5]. The Barkhausen transducer represented a coil measuring the magnetic
flux related to the Barkhausen oscillations and converting these oscillations into emf (the Barkhausen emf). A magnetic field in the sample was generated by an external electric magnet switched at a frequency of 50 Hz , which provided for a homogeneous magnetization reversal in the whole sample. A control sample represented the 2HCP alloy in the as-received state. Thermal annealing of the amorphous metal alloy samples was performed by heating them in vacuum or in a liquid neutral medium.

The results of our investigations revealed several previously unreported effects.

The introduction of hydrogen into metal leads to several-fold increase in the $\varepsilon_{\mathrm{B}}$ value (Fig. 1). As the hydrogenation time increases, the $\varepsilon_{\mathrm{B}}(t)$ curve exhibits saturation. Upon termination of the saturation process, the $\varepsilon_{\mathrm{B}}$ value gradually (over a period of several days) returns to a value close to the initial level (Fig. 2). Repeated saturation of the same sample with hydrogen (Fig. 2) leads to the corresponding increase in $\varepsilon_{\mathrm{B}}$. A sample of the alloy studied could withstand several hydrogenation-outgassing cycles before being destroyed. An increase in the number of cycles is accompanied by the accumulation of residual changes, whereby the $\varepsilon_{\mathrm{B}}$ amplitude in each cycle gradually decreases. It was established that the residual changes in $\varepsilon_{\mathrm{B}}$ could be eliminated by heating the sample to $100^{\circ} \mathrm{C}$, after which the sample showed the $\varepsilon_{\mathrm{B}}$ value characteristic of the initial state.

We have studied the effect of the thermal treatment temperature on the rate of $\varepsilon_{\mathrm{B}}$ recovery to the initial level (i.e., that observed before hydrogenation). It was found that increasing the temperature above $60^{\circ} \mathrm{C}$ sharply accelerates the recovery, which takes a few minutes at $120^{\circ} \mathrm{C}$ instead of three days at $20^{\circ} \mathrm{C}$. Since no structural changes in this temperature interval were reported for alloys of the type studied, we may suggest that the only reason for the $\varepsilon_{\mathrm{B}}$ recovery is the hydrogen escape from


Fig. 1. Effects of (1) the duration of hydrogenation treatment and (2) the annealing temperature on the Barkhausen emf in the 2 HCP alloy ( $\varepsilon_{\mathrm{B}}^{*}$ is the ratio of emf value in the hydrogen-saturated alloy to that in the control sample).


Fig. 2. Dynamics of the Barkhausen emf variation in the 2HCP alloy subjected to hydrogenation-annealing cycles. Solid portions of the curve correspond to a 10-min hydrogenation; dashed portions, to a 3-day storage at room temperature $\left(\varepsilon_{\mathrm{B}}^{*}\right.$ is the ratio of emf value in the hydrogen-saturated alloy to that in the control sample).
the metal. This assumption was confirmed by the results of special electrochemical investigations.

Thus, we have discovered the effect of hydrogen on the $\varepsilon_{\mathrm{B}}$ value in amorphous metal alloys of the 2 HCP type and showed reversibility of the process related to the presence of hydrogen in the metal bulk.

A change in the $\varepsilon_{\mathrm{B}}$ value in the alloy studied can be achieved by annealing the sample at $300-650^{\circ} \mathrm{C}$
(Fig. 1). However, even heating the sample to temperatures above the crystallization temperature does not provide for the $\varepsilon_{\mathrm{B}}$ variation such as that observed during hydrogen saturation of the same sample. Naturally, heating the alloy to these temperatures affects the metal structure and leads to irreversible changes in the sample, which results in that the $\varepsilon_{\mathrm{B}}$ value reaches a constant level and remains unchanged upon the thermal treatment.

Thus, there are grounds to believe that the amorphous metal alloy oversaturated with hydrogen features a special structural state characterized by a reduced potential barrier for the magnetic domain $180^{\circ}$-boundary passing from one equilibrium state to another. It is commonly accepted that the magnetic properties of ferromagnetic amorphous alloys are determined by the transition metal atoms (iron, cobalt, nickel), while the atoms of elements (metalloids) stabilizing the amorphous state are believed to be nonmagnetic [6]. According to these notions, the average magnetic moment $\tilde{\mu}$ is determined essentially by the magnetic moment of metal atoms $\tilde{\mu}_{f}$ and their concentration $c$ :

$$
\tilde{\mu}=c \tilde{\mu}_{f}+(1-c) \tilde{\mu}_{n}=c \tilde{\mu}_{f}
$$

where $\tilde{\mu}_{n}$ is the magnetic moment of a metalloid atom, which is assumed to be insignificant and taken equal to zero. As the hydrogen content in the alloy increases, the total contribution due to the magnetic moments of metal atoms must, at first glance, decrease. However, hydrogen is not a typical metalloid and hydrogen atoms present in the metal may dramatically change the situation; this is reflected by the results of our measurements.

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# Ferromagnetic Resonance Affected by a Superconductor 

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#### Abstract

The effect of a high- $T_{\mathrm{c}}$ superconductor on the uniform ferromagnetic resonance was experimentally studied using an $\mathrm{YBa}_{2} \mathrm{Cu}_{3} \mathrm{O}_{7-\delta}$ film placed onto an yttrium-iron-garnet disk. A peak in the resonance-bandwidth-temperature characteristic is revealed. The peak value is attained at a temperature slightly above $T_{\mathrm{c}}$. The maximum bandwidth exceeds that for liquid-nitrogen temperature by a factor of 6. © 2000 MAIK "Nauka/Interperiodica".


In recent years, the effect of a high $-T_{c}$ superconductor on the propagation of magnetostatic waves (i.e., long spin waves) in an adjacent ferrite film has been the subject of extensive experimental and theoretical study (see, e.g., [1-5]). In particular, it was found that a superconductor film may affect the surface wave attenuation, which increases if the temperature is above $T_{\mathrm{c}}$ and decreases otherwise [3]. This property can, in principle, be used for the contactless measurement of $T_{\mathrm{c}}$. Unfortunately, the existing techniques used to measure the magnetostatic wave attenuation exhibit insufficient accuracy and, most importantly, it is still unclear under what conditions the effect takes place. The above considerations inspired our investigation into the effect of a superconductor on the uniform ferromagnetic resonance. To the best of our knowledge, this aspect of the problem is explored for the first time. The line of research we pursue may reveal new phenomena related to microwave interactions in ferrite/superconductor structures.

To set the stage for optimum interaction of processes in a superconductor and a ferrite film, the former was placed onto an yttrium-iron-garnet (YIG) disk with a diameter of $\sim 1 \mathrm{~mm}$ and a thickness of $20 \mu \mathrm{~m}$. The disks were fabricated by depositing YIG films onto gadolinium-gallium-garnet substrates and etching away the excess part of the film. To achieve a sufficiently large filling factor and, hence, appreciable magnitude of the ferromagnetic resonance, a microstrip resonator was employed (Fig. 1a), the disk being situated at the antinode of the magnetic field. The resonator comprises two microstrip waveguide sections with a total length of $\lambda / 2$ (such resonators were used as elements of microstrip filters [6]). The loaded Q-factor of the resonator was $\sim 20$.

The $\mathrm{YBa}_{2} \mathrm{Cu}_{3} \mathrm{O}_{7-\delta}$ superconductor film with a thickness of $0.1 \mu \mathrm{~m}$ was deposited onto a $\mathrm{SrTiO}_{3}$ substrate by laser sputtering. The working area of the film $(\sim 1.5 \times 1.5 \mathrm{~mm})$ was small enough to avoid serious decrease in the resonator $Q$, which is lowered predominantly by the $\mathrm{SrTiO}_{3}$ substrate (Fig. 1b).

The microstrip resonator was placed into a cryostat, whose temperature $T$ was adjusted by blowing nitrogen vapor. Microwave power was coupled in and out via thin coaxial waveguides. Being normal to the plane of the disk, a magnetostatic field was varied within the range $2.8-3.3 \mathrm{kOe}$ so that the ferromagnetic resonance occurred at a constant frequency of 3.36 GHz .

The curves of the uniform ferromagnetic resonance (Fig. 1c) were displayed by an R2-57 panoramic meter, being superimposed onto the resonance curves of the microstrip resonator (also note weak resonances caused by nonuniform types of precession). The bandwidth of the ferromagnetic resonance $\Delta f$ was measured at a level of $N_{\text {res }}-3 \mathrm{~dB}$, where $N_{\text {res }}$ is the attenuation at the peak of the resonance curve (Fig. 1c). Indeed, the measured values of $\Delta f$ were not equal to $\gamma \Delta H$, where $\Delta H$ is the dissipation parameter. However, the discrepancy is unimportant in terms of this study.

Figure 2 shows the temperature dependences of $\Delta f$ measured with and without the superconductor film. Also displayed is the temperature dependence of the superconductor film resistance, the magnetic field being the same as that for the resonance measurements. The resistance was measured between two contacts deposited onto the film from which the sample for the resonance measurements was cut out. Figure 2 indicates that the loss introduced by the superconductor sharply rises near $T_{\mathrm{c}}$, namely, at a temperature $\sim 10 \mathrm{~K}$ higher than $T_{\mathrm{c}}$ ( $T_{\mathrm{c}}$ was conventionally determined at the


Fig. 1. (a, b) Schematic representation of the microstrip resonator and (c) a plot of transmitted power vs. frequency, displayed by an R2-57 meter. Panel (a) is the top view. Panel (b) is the view in section AB. Panels (a) and (b) show (1) a glass-ceramic substrate, (2) a copper strip, (3) a gadolin-ium-gallium-garnet substrate, (4) a YIG disk, (5) a superconductor film, and (6) a $\mathrm{SrTiO}_{3}$ substrate.


Fig. 2. Temperature dependence of $(1,2)$ the ferromagnetic resonance bandwidth (1) in the absence and (2) in the presence of a superconductor film and (3) of the resistance of the film. Curve 3 is obtained with the same magnetic field as that used in the resonance measurements.
middle point of the steeply descending segment of the resistance characteristic). Note that another sample showed a peak value of $\Delta f$ lower by about $10 \%$, the corresponding temperature being almost the same.

The reasons for the peak in the $\Delta f$ versus $T$ curve may be as follows:
(1) The losses due to eddy currents in the superconductor film. It was experimentally demonstrated that, if a ferrite film is in contact with a normal metal film, the contribution of eddy currents to the magnetostatic wave attenuation in the ferrite film exhibits a maximum at a certain surface resistance of the metallic film [7]. If this mechanism also dominates in the structure under study, then the fact that $\Delta f$ peaks at a temperature close to $T_{\mathrm{c}}$ is simply due to the abrupt change in the superconductor resistance near $T_{\mathrm{c}}$.
(2) The nonuniformity of the dc or the microwave magnetic field in the disk related to the structural inhomogeneity of the superconductor film or to the presence of vortices.

The observed behavior of $\Delta f$ may also be caused by other factors specific to superconductor films. If so, the existence of a maximum $\Delta f$ near $T_{\mathrm{c}}$ is physically significant, as in the case of the vortex-induced field nonuniformity.

To assess the effect of the surface resistance of the superconductor film, we replaced it with a metallic one and measured $\Delta f$ at a constant (room) temperature. Several metals and thicknesses were tried so that the surface resistance $\rho_{s}$ was changed within the same range as that of the superconductor film. The result was somewhat surprising: the points fitted to a common curve in the $\rho_{s}$ versus $\Delta f$ plane. The curve exhibited a maximum at a value of $\rho_{s}$ close to the surface resistance of the superconductor film at the temperature where $\Delta f$ is maximum, but the maximum value of $\Delta f$ for the metallic films was a few times larger than that for the superconductor film. One may interpret this by the fact that a superconductor film exerts a considerably different influence than a metallic film does. Anyhow, the question as to the role of the surface resistance remains unanswered. In fact, one has yet to find out which mechanism underlies the action of a superconductor film on the ferromagnetic resonance in an adjacent ferrite film.

To elucidate this mechanism and, in particular, to ascertain the factors that determine the position of the peak in the $\Delta f$ versus $T$ curve, it seems wise, as a first step, to carry out similar measurements for films of high $-T_{\mathrm{c}}$ superconductors differing in composition and thickness.

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# Polymer Films Studied by Scanning Tunneling Microscopy 

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#### Abstract

Polymer films with thicknesses up to 300 nm were investigated by scanning tunneling microscopy. It was demonstrated that the films contain areas whose images change depending on the scan parameters, which can be explained by the emission processes. © 2000 MAIK "Nauka/Interperiodica".


The discovery of high electric conduction in thin polymer films at the end of the 1980s [1, 2] and the development of methods to achieve this state [3] suggest good prospects for the use of these materials as elements of molecular electronics. First of all, this is related to the fact that the polymer conductivity can be reversibly changed in a wide dynamic range under the influence of diverse physical parameters, the required magnitudes of a driving action being rather low.

At the same time, given the number of reports on the unaccountably high electric conductivity of the polymer films ( $\left.\sigma>10^{6}(\Omega \mathrm{~cm})^{-1}[4]\right)$ and the anomalously high current densities ( $j>10^{5} \mathrm{~A} / \mathrm{cm}^{2}$ ) that these films can withstand with no apparent damage, one is prompted to seek for trivial explanations of the effect such as a short circuit or intergrowth of a metal dendrite between electrodes [5]; moreover, doubts in the existence of the effect as such may arise.

Answers to some of the critical questions could be provided by the studies of the structure and morphology of a polymer film subjected to various factors causing the film transition into the high-conductivity state. By now, only the results of indirect studies of the structural parameters of conducting regions in polymers are available [6, 7]. A single experiment in which the conducting domains were observed immediately in the conduction regime was performed with liquid crystals [8]. However, this technique only provided observation of the sites where the conducting domains emerged on the surface, while it was impossible to make conclusions on their structure and conduction properties.

The purpose of our study was to investigate the structure of conducting domains in thin polymer films by scanning tunneling microscopy (STM), which enables one to examine the structure and electrical properties of nanometer-sized objects.

The study was carried out in air using an SMM2000T scanning multimicroscope providing a lateral resolution of 0.3 nm and an order of magnitude better resolution in the vertical direction. In the operation regimes used, the tunnel current was in the $0.01-1 \mathrm{nA}$
range, and the voltage across the tip-sample gap was varied from 5 to 1000 mV . The image was recorded in the constant tunnel current mode.

Below, we report the results obtained for a polymer from the polyheteroarylene series. The substrates were made of the optically polished [100]-oriented silicon with a natural oxide layer, over which a gold layer was deposited. The films were obtained by spin-coating from a cyclohexanone solution in a centrifuge; the samples were first dried for 30 min at room temperature and then baked for another 30 min in a drying chamber at $100-150^{\circ} \mathrm{C}$ to remove the residual solvent. The film thickness was about 100 nm .

One of the first and basic results obtained, which enabled us to perform the study of polyheteroarylene films by STM, is that the tunnel current flowing in the circuit consisting of the STM tip, the tunneling gap, the polymer film, and the metallic substrate, can be reliably "locked." This is not a trivial fact, since the film thickness ranges from 50 to 100 nm . Apparently, the possibility of observing the polymer film surface is favored by the phenomenon of the so-called spontaneous conduction in these materials reported previously [9].

The STM image of the polymer film surface represents a pattern of fibrillar or granular structure typical of this kind of object. At a relatively flat portion of the surface, we managed to observe the structure corresponding to the regions of the polymer crystallinity with a parallel arrangement of molecules (see Fig. 1).

Studying the influence of the measurement conditions (current and voltage) on the quality of recorded images, we established that the STM images from certain regions of the polymer film depend on the voltage across the tunnel gap. Figure 2 shows images of the same region of the film recorded at different voltages increasing from 130 to 830 mV . One can clearly see that spots of enhanced brightness, whose area and contrast increase with the voltage, appear on the background of the existing relief pattern.

To correctly interpret this effect, let us analyze the measurement procedure in more detail. The image is


Fig. 1. An STM image of a 100 -nm-thick polymer film showing a region with ordered arrangement of the polymer chains (scan area, $90 \times 90 \AA$ ). A three-dimensional representation with virtual illumination source is shown to the right.


Fig. 2. Variation of the emission center distribution over the polymer film surface with the voltage $V$ applied across the tunneling gap: (a) $V=130 \mathrm{mV}, h=10 \mathrm{~nm}$; (b) $V=230 \mathrm{mV}, h=14 \mathrm{~nm}$; (c) $V=430 \mathrm{mV}, h=19 \mathrm{~nm}$; (d) $V=630 \mathrm{mV}, h=28 \mathrm{~nm}$; (e) $V=730 \mathrm{mV}$, $h=38 \mathrm{~nm}$; (f) $V=830 \mathrm{mV}, h=46 \mathrm{~nm}$ ( $h$ is the height of a typical image element). The scan area is $0.6 \times 0.6 \mu \mathrm{~m}$, and the tunnel current is 0.1 nA ; three-dimensional representation is shown.
obtained in the constant current mode. The feedback loop keeps the tunnel current constant by varying the distance between the tip and the surface under study, which is equivalent to recording the surface relief pattern. In this regime, the average tip-surface distance is
maintained on a level close to the "tunneling gap" value of about $10 \AA$. Other reasons can cause local current variations, e.g., changes in the effective work function (EWF) of electrons at a given region of the surface. However, the contribution from the EWF variations is usually
considerably smaller that from the relief variations, which follows from the well-known relationship [10]

$$
I \propto\left(\frac{V}{d}\right) \exp \left(-A \bar{\varphi}^{1 / 2} d\right)
$$

where $A=1.025 \mathrm{eV}^{-1 / 2} \AA^{-1}, \bar{\varphi}$ is the average height of the potential barrier between electrodes, $V$ is the potential difference, and $d$ is the width of the tunneling gap.

From the series of images presented in Fig. 2, one can see that new relief features are formed in addition to the previously existing ones. This can be related either to the mass transfer caused by some lithographic process or to the emission phenomena.

As a rule, lithography takes place under fairly large applied voltages (usually, $V \geq 10 \mathrm{~V}$ ) on the surface of metals, where atoms can migrate easily. In the experiment under discussion, $V<1 \mathrm{~V}$, which is well below the level necessary to transfer the metal atoms and polymer molecules. In addition, restoration of the initial relief pattern changed by the mass transfer must be difficult (even impossible under the conditions of decreasing voltage); meanwhile, the recorded surface image depends only on the magnitude of the applied voltage, irrespective of the direction of its variation. When the tunnel current was increased, no changes in the initial relief pattern were recorded. This indicates that deformations induced by the Joule heat were absent.

Thus, one can suppose with confidence that the observed changes in the surface image presented in Fig. 2 are related to the emission processes in certain regions of the polymer film under study. This can be called a "pseudorelief," originating from relatively large local variations of the EWF. Then, the pseudorelief height is to be proportional to the emission current density.

Based on the available data on the structure of polymer films, it is reasonable to assume that the observed regions of reduced EWF correspond to the sites of emergence of the conduction channels responsible for the high conductivity of these polymers. This conclusion is corroborated by the finding that the emitting area of a channel increases with the voltage applied, which can occur due to the well-known effect of the current pinching [11, 12]. In addition, the measured dimensions of these regions are in good agreement with the estimates obtained by other methods.

By now, it has been known that the modified STM technique enables one to investigate emission properties of the diamond-like films [13, 14] and graphite heterostructures [15]. Interest in these objects is related to the negative electron affinity and, consequently, good
emission properties of these materials. The emission centers in those cases are predominantly located at the nanocrystallite interfaces. A special feature of the system under consideration is that emission occurs both at the grain boundaries and on the grains themselves. This means that, in principle, it is possible to induce emission over the whole film area. Taking into account the simplicity of their fabrication and the rather wide choice of physical parameters that control their local electrical properties, polyheteroarylene films can provide promising objects to create the emission electronic devices.

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# A Special Feature in the Magnetic Moment Orientation of Highly Magnetic Phase Particles in Magnetically Hard Fe-Co-Ni-Al Alloys 

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#### Abstract

Orientation of the magnetic moments of the particles of a highly magnetic phase in a magnetically hard $\mathrm{Fe}-\mathrm{Co}-\mathrm{Ni}-\mathrm{Al}$ alloy (YuN15DK25BA grade) was studied by Mössbauer spectroscopy. The results indicate that the magnetic moments, instead of being oriented along the axes of anisotropy of the particles, fluctuate with an average amplitude of $\theta=45^{\circ}$ and a characteristic time $\tau_{\mathrm{s}} \approx 6 \times 10^{-9} \mathrm{~s}$ relative to these axes. © 2000 MAIK "Nauka/Interperiodica".


Metal alloys of the $\mathrm{Fe}-\mathrm{Co}-\mathrm{Ni}-\mathrm{Al}$ system are widely used in permanent magnets. In the highly coercive state, the structure of these alloys comprises singledomain anisotropic particles of a highly magnetic $\alpha^{\prime}$ phase separated by interlayers of a weakly magnetic $\alpha$ phase [1]. During the thermomagnetic processing, the acicular $\alpha$ ' phase particles are oriented so that their long axes are aligned in the magnetic field, which gives rise to the induced uniaxial magnetic anisotropy with a constant exceeding the crystallographic anisotropy constant by more than one order in magnitude [2]. The presence of magnetically uniaxial particles and the parallel orientation of their long axes upon thermomagnetic treatment lead to the formation of interacting domains (IDs) representing macroscopic regions containing large numbers of single-domain particles magnetized in the same direction [3-5].

The appearance and behavior of IDs in the system studied exhibit some features that make these regions different from the usual domains observed in uniaxial multidomain ferroelectrics. In particular, it was established that there are no interlayers between IDs, and their boundaries are not parallel to the easy magnetization axis (EA) [3, 4] determined by the direction of predominant orientation of the long axes of particles in the domains. As a result, magnetic charges appear on the domain walls [5] and the question as to how the magnetic moments of single-domain particles are oriented inside IDs in the absence of external magnetic field is still open.

In order to elucidate this problem, we have studied a magnetically hard alloy of the YuN15DK25BA grade containing (wt \%) $48 \mathrm{Fe}, 25 \mathrm{Co}, 14 \mathrm{Ni}, 8.5 \mathrm{Al}, 3.5 \mathrm{Cu}$, $0.2 \mathrm{Ti}, 0.6 \mathrm{Nb}$, and 0.2 S by Mössbauer spectroscopy. The magnetically hard alloy was characterized by a residual induction $B_{r}=12500 \mathrm{Gs}$ and a coercive force
of $H_{c}=780 \mathrm{Oe}$. The IDs, visualized with the aid of a magnetic suspension, had a width of $30 \mu \mathrm{~m}$. In a demagnetized sample, the boundaries of IDs made an angle not exceeding $7^{\circ}$ with the EA. A sample for the Mössbauer measurements had the shape of a thin disk with a diameter of 12 mm cut from a massive alloy along the plane perpendicular to the EA. The sample surface was ground and polished, after which the sample thickness was $50 \mu \mathrm{~m}$. The Mössbauer spectra were excited by $\gamma$-radiation from a ${ }^{57}$ Co source in a chromium matrix. The beam divergence was $5^{\circ}$. Prior to measurements, the sample was demagnetized so that the intrinsic demagnetizing field would not influence the orientation of the magnetic moments of particles in the domains. Demagnetization was achieved by exposure of the sample to an alternating magnetic field parallel to the EA, with the field amplitude having been decreased to zero.

Figure 1 shows a typical Mössbauer spectrum displaying a Zeeman sextet, related to Fe atoms in the $\alpha$ ' phase, and the central peak due to the paramagnetic $\alpha$ phase iron. The effective magnetic field acting upon the iron nuclei in the $\alpha^{\prime}$ phase has a strength of $H_{\mathrm{e}}=$ 345 kOe . The isomer shifts for the $\alpha$ and $\alpha^{\prime}$ phase iron nuclei are 0.14 and $0.03 \mathrm{~mm} / \mathrm{s}$, respectively. There are two circumstances that are worth noting.

1. The second and fifth lines of the spectrum are characterized by definite intensities. Should the magnetic moments of particles in the $\alpha$ ' phase be parallel to the EA, these lines would be absent as a result of the polarization effect [6].
2. The edge lines in the Mössbauer spectrum are much wider than the middle ones ( $\Gamma_{1.6}=0.98 \mathrm{~mm} / \mathrm{s}$ against $\Gamma_{3.4}=0.40 \mathrm{~mm} / \mathrm{s}$ ), which is typical of the relaxation spectra $[7,8]$. However, this broadening can also be due to a statistical distribution of the effective mag-


Fig. 1. A typical Mössbauer spectrum of the YuN15DK25BA alloy. The sample was cut perpendicularly to the easy magnetization axis.


Fig. 2. A graphical solution to Eq. (1).
netic fields on the ${ }^{57} \mathrm{Fe}$ nuclei, which may take place in alloys. As is known, the $\alpha^{\prime}$ phase particles represent a solid solution based on the Fe -Co system [1]. It was demonstrated [9] that a statistical distribution of Fe and Co atoms in the nearest vicinity of Fe nuclei in the $\mathrm{Fe}-$ Co alloys produces no significant broadening of the Mössbauer spectrum. For example, a $25-75 \%$ variation in the Co content in these alloys increases the spectral lines only by $0.04 \mathrm{~mm} / \mathrm{s}$, the width of the edge lines not exceeding $0.30 \mathrm{~mm} / \mathrm{s}$ [9]. Therefore, we may suggest that the broadening of lines belonging to the $\alpha^{\prime}$ phase is due to fluctuations of the effective magnetic field on the iron nuclei in this phase. We believe that this process is related to fluctuations in the directions of magnetic moments of the single-domain particles in IDs. Indeed, the $\mathrm{Fe}-\mathrm{Cr}-\mathrm{Co}$ alloy not subjected to a thermomagnetic treatment (i.e., containing no IDs) the lines in the Mössbauer spectrum corresponding to the $\alpha^{\prime}$ phase exhibit no broadening [10]; otherwise, the spectra are very much alike in both shape and parameters.

A factor responsible for fluctuations of the directions of magnetic moments of the single-domain particles in IDs is probably the presence of magnetic charges in the vicinity of these domains [3,5]. The tendency to decrease the magnetostatic energy of the surface and bulk magnetic charges leads to chaotic fluctuations in the directions of magnetic moments from one domain to another, which results in the closure of the magnetic flux between the IDs. In other words, the fluctuations of magnetic moments play the role of the closing domains in the usual domain structure.

We may estimate the average amplitude of fluctuations of the magnetic moment orientation in the IDs. For this purpose, we will use an equation obtained previously [11],

$$
\begin{equation*}
\cos ^{3} \theta+\frac{9 k-12}{3 k+4} \cos \theta+\frac{8-12 k}{3 k+4}=0 \tag{1}
\end{equation*}
$$

which allows the angle of scatter $\theta$ in the magnetic texture of a permanent magnet to be determined using the parameter $k$ representing the ratio of areas under the second and first or fifths and sixth lines in the Mössbauer spectrum ( $k=S_{2} / S_{1}$ or $S_{5} / S_{6}$ ). Here, the $\theta$ angle can be considered as the average amplitude of fluctuations of the magnetic moments of particles in the $\alpha^{\prime}$ phase relative to their long axes. Figure 2 shows a graphical solution to Eq. (1), according to which the alloy studied in this work is characterized by $\theta=45^{\circ}$.

A condition for manifestation of the fluctuational phenomena in the Mössbauer spectra is $[7,8]$

$$
\begin{equation*}
v_{L} \tau_{\mathrm{s}} \approx 1 \tag{2}
\end{equation*}
$$

where $v_{\mathrm{L}}$ is the frequency of the Larmor precession of nuclear spins in the effective magnetic field $H_{\mathrm{e}}$, and $\tau_{\mathrm{s}}$ is the characteristic time of fluctuation of the effective magnetic field at the nucleus. We may use Eq. (2) to estimate a characteristic time for the magnetic moments of particles fluctuating relative to their axes of anisotropy in the $\alpha$ ' phase, which yields $\tau_{\mathrm{s}} \approx 6 \times 10^{-9} \mathrm{~s}$.

It should be noted that magnetic characteristics achieved in practice for the $\mathrm{Fe}-\mathrm{Co}-\mathrm{Ni}-\mathrm{Al}$ alloys are lower than the values calculated by a model assuming homogeneous rotation of the magnetization vectors of single-domain particles [1]. The discrepancy is usually explained in terms of the incoherent rotation of the magnetization vectors of single-domain particles and the magnetic interactions between these particles. Chaotic fluctuations of the magnetic moments of particles imply incoherence of their rotation in the course of the magnetization reversal. Thus, the observed phenomenon may offer a link explaining the discrepancy between theory and experiment.

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# Phase Diffraction Gratings Formed Based on the Phenomenon of Single Crystal Twinning 

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#### Abstract

Phase diffraction gratings can be created using the phenomenon of crystal twinning. Conditions of the grating formation and possibilities for increasing the quality of gratings are considered. © 2000 MAIK "Nauka/Interperiodica".


In recent years, we have extensively studied the effect of twinning in crystals [1-4]. It would be also very important to find practical applications for this phenomenon in modern technologies. In this context, we have investigated the possibility of using crystal twinning to obtain phase gratings now widely employed in optical devices.

The principal feature of a phase diffraction grating is a surface relief, obtained predominantly by mechanical methods, in the form of linear grooves with triangular cross section on the smooth surface of a material reflecting light waves. Below, we demonstrate that a similar relief on a crystal surface can be obtained by partly converting a near-surface region of the crystal into a twinned orientation. Indeed, every twinned ridge creates a defect (step) with a triangular cross section on the crystal surface. The step height $H$ can be controlled to vary from hundreds of Angströms to tens of microns.

Figure 1 shows a scheme that can be used to obtain a system of parallel twinned inclusions (Fig. 2) forming a phase diffraction grating. In order to induce the formation of twinning dislocations, it is necessary to apply localized loads that would lead to the concentration of sufficiently high internal stresses determined by the Schmidt orientational factor. In our case, the loads can be developed by spherical indentors 2 in Fig. 1 spaced by the distances $L$ and $D$ along the $O X$ and $O Y$ axes, respectively. Localized deformation of a crystal surface with such an indentor gives rise to several wedgeshaped twins around the indentor, which propagate in several crystallographic directions rather than along a single axis. For example, six favorable twinning directions were observed in bismuth single crystals deformed on the (111) cleavage plane [1-4]. In order to separate two mutually perpendicular directions necessary for the grating formation, we may use, for example, additional applied stresses acting along preselected axes so as to facilitate twinning in these directions (making them preferred to the other possible directions).

Another means of separating preferred twinning directions is provided by the anisotropic electroplastic effect observed for twinning in single crystals [6]. In this case, the dislocation processes accompanying twinning in the directions of electric current passage are predominantly stimulated.

In order to reduce the number of twins originating from a concentrators of applied stresses, it is recommended to apply local stresses using the indentors of spherical shape. The number of twins can be controlled, for example, by changing the temperature of a deformed crystal. Increasing the temperature (within certain interval) usually decreases the contribution of twinning to the plastic deformation and results in a lower number of twins originating from the stress con-


Fig. 1. A scheme of obtaining phase diffraction gratings using the phenomenon of single crystal twinning: (1) sample surface; (2) stress concentrators; (3) twinned inclusions.


Fig. 2. A system of parallel twinning inclusions forming a phase diffraction grating on a single crystal surface.
centrator [6]. A decrease in the number of unfavorable twins in a crystal can be also provided by ion irradiation of the crystal [1-3,5]. In the case of deformed bismuth crystals, an especially pronounced effect of decreasing the number of twins accompanied by stimulating the mobility of twinning dislocations was observed in the samples irradiated by zirconium ions [5].

When a single crystal surface is deformed by localized indentors spaced by a distance $D$, the boundaries of twinned ridges with the width $h$ are separated by a layer of untwinned material with a thickness $d$. A light wave normally incident onto such a grating will give
rise to beams reflected in the reverse direction, which would negatively affect the diffraction pattern. In order to reduce the $d$ value to zero, it is necessary to increase either the external control stresses $\sigma$ (Fig. 1) or the load applied to the spherical indentors (2 in Fig. 1).

Thus, we have demonstrated the possibility to obtain phase diffraction gratings by means of controlled twinning in single crystals.

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# Preparation and Investigation of the Structure and Properties of $\mathrm{Al}_{2} \mathrm{O}_{3}$ Plasma-Detonation Coatings 

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#### Abstract

Using a specially designed and constructed plasmatron, combining the plasma and detonation technologies, aluminum oxide coatings with high adhesion and strength ( 19.8 GPa ) were obtained for the first time. The samples were studied by the transmission electron microscopy and X-ray diffraction methods and characterized for the first time by the Rutherford backscattering and proton elastic resonance measurements. It was found that the aluminum oxide coatings are composed of $0.20-0.25 \mu \mathrm{~m}$ crystallites, belonging to the $\alpha-, \beta-, \gamma-$, and $\delta-\mathrm{Al}_{2} \mathrm{O}_{3}$ crystal modifications, and have a transition region containing AlFe nanocrystals with a size of $20-40 \mathrm{~nm}$ and a considerable dislocation density. The aluminum oxide coatings exhibit significant carbonization, with a maximum carbon concentration on the surface reaching $20 \mathrm{at} . \%$. The relative phase composition calculated using the X-ray diffraction data is as follows: $\gamma$ - $\mathrm{Al}_{2} \mathrm{O}_{3}, 60 \% ; \alpha-\mathrm{Al}_{2} \mathrm{O}_{3}, 30 \% ; \beta$ - and $\delta-\mathrm{Al}_{2} \mathrm{O}_{3}$, balance to $100 \%$. © 2000 MAIK "Nauka/Interperiodica".


At present, the arsenal of effective means of increasing the reliability and durability of structural materials used for the fabrication of machine parts, equipment, technological fittings, and tools includes various surface treatment methods. In particular, the most widely employed methods of surface hardening are based on the formation of various coatings. A new technology capable of modifying the surface of structural materials and forming coatings possessing protective and other useful properties is the plasma-detonation spraying (PDS) [1]. The PDS technology is based on the use of pulsed plasma streams transferred by a detonation wave in the form of a plasma impulse. As a result, the plasma stream velocity increases from $100-300$ to 5000$8000 \mathrm{~m} / \mathrm{s}$ and the power flux density grows up to $10^{7}-$ $10^{8} \mathrm{~W} / \mathrm{cm}^{2}$ [1]. These factors provide for the possibility of obtaining high-performance ceramic and metalceramic coatings on metal substrates by the PDS method.

The phase composition of an aluminum oxide coating depends on the method and technological parameters of deposition, substrate temperature, size of deposited particles, and some other factors. These coatings are conventionally obtained by gas-plasma, plasma, or detonation spraying methods [2-4]. As is known, coatings obtained by the gas-plasma spraying of corundum $\left(\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}\right)$ are composed almost completely of $\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}$, while the plasma and detonation spraying processes result in two-phase coatings composed of $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ ( $5-10 \%$ ) and $\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}(90-95 \%)$. However, data on the sequence of phase transitions converting a low-temperature modification $\left(\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}\right)$ into the high-temperature
one ( $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ ), as well as on the quantitative characteristics of this process, are rather contradictory. For example, Kozlova [6] showed that $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ coatings is composed of the primary powder particles, while Bartenev et al. [7] ascertained that the final coating is formed as a result of the sequence of phase transitions involved in the detonation spraying process rather than the initial corundum. Therefore, we could not predict a priori the phase composition of a plasma-detonation aluminum oxide coating deposited onto a steel substrate in our experiments. The purpose of this study was to determine the elemental and phase composition of an aluminum oxide layer obtained for the first time by the PDS method.

A setup for the plasma-detonation spraying employs a gas mixture burning in the detonation regime as a factor initiating the pulsed plasma. As a result, a pulsed plasma stream with a pulse duration of $1-3 \mathrm{~ms}$ and a repetition frequency of $2-20 \mathrm{~Hz}$, a power flux density of $10^{5}-10^{6} \mathrm{~W} / \mathrm{cm}^{2}$, a velocity of $5000-8000 \mathrm{~m} / \mathrm{s}$, and a temperature of $25000-30000 \mathrm{~K}$ is emitted from the plasma-detonation generator. The stream carries a current of $1-3 \mathrm{kA}$. The rate of heating and cooling of the surface layer of a substrate reaches $10^{5}-10^{6} \mathrm{~K} / \mathrm{s}$.

The oxide coatings were deposited with the aid of a special plasma-detonation burner (Fig. 1). The main parts of this device are a replaceable nozzle 1 , a gasodynamic combustion chamber 2 where the gas components are mixed and burned to detonate, and a spark plug 3 providing for the gas mixture ignition at a frequency of 20 Hz . The gas mixture components and cooling water are supplied via pipelines 4 . In order to
increase the pulse power in the PDS deposition setup [1], the gas mixture was burned under the conditions of a nonstationary detonation process. The efficiency of the PDS method was increased by initiating the gas mixture detonation in an electromagnetic field created between two coaxial electrodes in the reaction chamber. A metal electrode mounted at the axis of the reaction chamber exhibits erosion in an overheated state, which provides for the introduction of metal vapors into the plasma stream. Detonation of the gas mixture filling the reaction chamber is initiated by a detonation gun. The gas mixture components are introduced into the reaction chamber via special channels in the detonation gun.

In the presence of the electric field $E$ created by the electric energy converter, the gas layer behind the denotation wave carries the electric current. This provides for an additional energy supply to the gas. When the detonation wave leaves the nozzle of the combustion chamber, the electric current flows from the electrode (anode) to the surface of coated article (cathode) via the plasma stream and the sprayed material.

The material to be deposited is supplied with a transport gas via a channel in the side wall of the reaction chamber. The plasma is confined to the chamber axis under the action of ponderomotive forces, which markedly increases its energy density. The high-energy gas stream strikes the article surface to form an impact compression layer. The current passing in the plasma stream leads to an additional heating of the material and the energy deposition in the impact compression layer.

In our experiments, an aluminum oxide coating was deposited by PDS onto a 3-mm-thick steel plate. The coating material was an electrocorundum powder (20-45 $\mu \mathrm{m}$ fraction) representing the $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ crystal modification. The experiments were performed using an Impuls-2 plasmatron operated in the regime with $W=10 \mathrm{~kW}$ using a propane-butane mixture as the plasma-forming gas; the corundum powder was supplied at a rate of $m=3 \mathrm{~kg} / \mathrm{h}$. The pulsed plasma stream had a power flux density of $5 \times 10^{6}-10^{7} \mathrm{~W} / \mathrm{cm}^{2}$, a temperature of $(2.5-3) \times 10^{4} \mathrm{~K}$, and a velocity of up to $7 \mathrm{~km} / \mathrm{s}$ at a plasmatron operation frequency of 20 Hz . A distance from the plasmatron nozzle edge to the substrate surface was $20-40 \mathrm{~mm}$. The coatings were deposited at a rate of $1 \mathrm{~m}^{2} / \mathrm{h}$ and had a total thickness of $0.3-0.8 \mathrm{~mm}$.

To our knowledge, we have performed for the first time the elemental analysis of the aluminum oxide coatings obtained by PDS. Figures $2 a$ and $2 b$ show the spectra obtained by Rutherford backscattering (RBS) and proton elastic resonance (PER) spectra of an $\mathrm{Al}_{2} \mathrm{O}_{3}$ coating in steel. The RBS and PER spectra exhibit the peaks of oxygen, aluminum, iron, and a considerable amount of carbon in the uppermost surface layer of the coating. The empirical formula of a film formed by the


Fig. 1. Schematic diagram of a microwave detonation plasma-gun (plasmatron): (1) replaceable nozzle; (2) gasodynamic combustion chamber; (3) spark plug; (4) pipelines supplying gases, air, oxygen, and cooling water; (5) powdered coating material.
plasma-detonation spraying process can be expressed as $\mathrm{Al}_{38} \mathrm{O}_{59} \mathrm{Fe}_{3}$. The concentration of carbon in the surface layers of the coating reached $20 \mathrm{at} . \%$. The appearance of iron in the oxide coating is explained by erosion of a steel electrode in the course of the plasma stream formation in the combustion chamber of the plasmatron [8-11].

Data of the local phase and structure analysis performed by the electron microdiffraction in a transmission electron microscope showed that the coating possesses a multiphase polycrystalline structure comprising a mixture of $\alpha-, \beta-, \gamma-$, and $\delta-\mathrm{Al}_{2} \mathrm{O}_{3}$ modifications and AlFe. The $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ crystallites with a size of about $0.25 \mu \mathrm{~m}$ exhibit a regular faceting and contain no structural defects. The other phases ( $\beta-, \gamma-$, and $\delta-\mathrm{Al}_{2} \mathrm{O}_{3}$ ) appear as crystalline grains with the dimensions ranging from 0.1 to $0.3 \mu \mathrm{~m}$. These crystallites exhibit a substructure of defects representing isolated dislocations. The coating regions adjacent to the substrate contain a transition layer representing an AlFe intermetallic compound possessing a polycrystalline structure with a grain size of $20-40 \mathrm{~nm}$. This layer is characterized by a high dislocation density, which is related to the impact action of the plasma-detonation wave.

The X-ray diffraction analysis, which provides integral information about the surface layer several


Fig. 2. Data on the elemental and phase composition of an $\mathrm{Al}_{2} \mathrm{O}_{3}$ coating obtained by PDS on a substrate spaced by 40 mm from the plasmatron nozzle: (a) $\mathrm{He}^{+}$RBS spectrum; (b) PER spectrum; (c) X-ray diffractogram.
microns thick, also showed the presence of $\alpha$ - and $\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}$ phases (Fig. 2b). The phase ratio calculated from these data was as follows: $\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}, 60 \%$; $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}, 30 \% ; \beta$ - and $\delta-\mathrm{Al}_{2} \mathrm{O}_{3}$, balance to $100 \%$.

An analysis of the relationship between the phase composition and physicochemical and mechanical properties of the aluminum oxide coatings obtained by PDS showed that an increase in the content of a stable $\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}$ phase, as well as the other (metastable) phases, leads to a decreases in the hardness, wear resistance, and corrosion resistance of the coating. On the other hand, these components improve the coating adhesion to substrate and increase the coating material density. As is known [5, 6], the polymorphous $\gamma \longleftrightarrow \alpha$ transition occurring in $\mathrm{Al}_{2} \mathrm{O}_{3}$ on heating to $1050-1300^{\circ} \mathrm{C}$ involves considerable volumetric changes. This may result in cracking of the coating and its separation from the substrate, provided that a two-phase coating contains more than $80 \%$ of the $\gamma-\mathrm{Al}_{2} \mathrm{O}_{3}$ phase or a thickness exceeding 0.3 mm .

In our coatings, the presence of several phases led to a gradient of relaxation stresses that inhibited the formation of cracks. The average density of PDS coatings was $3.90 \mathrm{~g} / \mathrm{cm}^{3}$, which is somewhat lower than the den-
sity of $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}\left(3.98 \mathrm{~g} / \mathrm{cm}^{3}[4,11]\right)$. At the same time, the maximum hardness of the PDS coating (1.98 GPa) and the thermal conductivity coefficient ( $38 \mathrm{~W} / \mathrm{m} \mathrm{K}$ ) were close to the corresponding parameters of $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ [4, 11].

Thus, plasma-detonation spraying of $\mathrm{Al}_{2} \mathrm{O}_{3}$ allows compact multiphase coatings to be obtained. The coatings have a good adhesion to substrate (which is related to the presence of a stable low-temperature phase and metastable phases) and high mechanical properties (owing to a high content of the $\alpha-\mathrm{Al}_{2} \mathrm{O}_{3}$ phase).

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# Energy Production in Ambipolar Reactors with D-T, D- ${ }^{3} \mathrm{He}$, and D-D Fuel Cycles 

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#### Abstract

Kinetics of the D-T, D- ${ }^{3} \mathrm{He}$, and D-D (catalyzed) fuel cycles in an ambipolar reactor were analyzed. The plasma characteristics and the plasma confinement system parameters necessary for the effective energy production in the ambipolar reactor operated with these fuels were calculated. © 2000 MAIK "Nauka/Interperiodica".


This paper reports on a comparative analysis of the characteristics of a thermonuclear plasma and the parameters of a magnetic confinement system for D-T and the alternative ( $\mathrm{D}-{ }^{3} \mathrm{He}, \mathrm{D}-\mathrm{D}$ ) fuels. By thermonuclear plasma we imply the plasma featuring intensive thermonuclear reactions with a power gain coefficient of $Q \geq 10$. An ambipolar system was selected because the condition $Q \geq 10$ for the alternative fuel cycles is fulfilled with $\beta>0.4[1,2]$ ( $\beta$ is the ratio of the plasma pressure to that produced by the external magnetic field). The MHD stability can be provided in an axially symmetric ambipolar system [3], which excludes the neoclassical transfer processes. It was experimentally demonstrated that the transverse confinement time in such a system can be controlled provided that the anomalous transfer by the radial electrostatic field is suppressed [4].

The main advantage of the $\mathrm{D}-{ }^{3} \mathrm{He}$ reactor over the $\mathrm{D}-\mathrm{T}$ system is a very low level of neutron fluxes originating from the plasma, which increases the working life of the first wall up to 30-40 years. The D-D reactor requires no gas blanket, which implies no need in providing for the tritium or helium-3 supply. We will analyze the energy balance and the whole plasma confinement system of an ambipolar reactor. The main task of this analysis is to assess the characteristics of a thermonuclear plasma and the related magnetic confinement system for the effective energy production in the ambipolar reactor using the $\mathrm{D}-\mathrm{T}, \mathrm{D}-{ }^{3} \mathrm{He}$, and $\mathrm{D}-\mathrm{D}$ fuels. The D-D fuel cycle is considered in two variants: (i) fully catalyzed, whereby both ${ }^{3} \mathrm{He}$ and tritium produced in the reaction are completely used as a secondary fuel; (ii) partly catalyzed, whereby only ${ }^{3} \mathrm{He}$ is completely used while the unburned tritium fraction is not
returned to the reactor (in order to reduce the neutron flux). The power balance equation for an ambipolar reactor plasma is described by the following equation:
$P_{f}+P_{c}+P_{p}=P_{\|}+P_{\perp}+P_{n}+P_{r}+\sum_{a}\left(P_{0 a}+P_{a}\right),(1)$
where $P_{f}$ is the power of the thermonuclear reaction; $P_{c}$ and $P_{p}$ are the powers of the external heating of plasma in the central section and in the protective ion barrier, respectively; $P_{\|}$are the longitudinal power losses [5]; $P_{\perp}$ are the transverse losses characterized by the transverse confinement time $\tau_{\perp}$ assumed to be the same for all types of charged particles; $P_{n}$ is the power of neutrons; $P_{r}$ are the radiative losses (bremsstrahlung and cyclotron effects); $P_{0 a}$ are the losses related to the production of charged particles in the region of losses; and $P_{a}$ is the power carried by charged products scattered in the region of losses (subscript "a" indicates the ion products of various types). The cyclotron losses for $T_{e}=5-100 \mathrm{keV}$ are determined by the Trubnikov formula [6] and for $T_{e}=100-1000 \mathrm{keV}$, by a modified expression [7].

The losses for bremsstrahlung on ions ( $P^{e-i}$ ) and electrons ( $P^{e-e}$ ) are calculated by integrating the electron emission cross sections for ions and electrons with respect to the electron flux in the space of velocities with a relativistic equilibrium electron distribution function [8]. In the region of $T_{e}>10 \mathrm{keV}$, where the Born approximation is valid [9], the results of numerical calculations performed for the emission cross sections for ions [10] and electrons [9] can be approximated (to within a $2 \%$ error) by the following

Parameters of the ambipolar $\mathrm{D}-\mathrm{T}, \mathrm{D}-{ }^{3} \mathrm{H}$, and $\mathrm{D}-\mathrm{D}$

| Parameter** | D-T | D- ${ }^{3} \mathrm{He}$ |  | D-D |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
| $x_{3 \mathrm{He}}=n_{3 \mathrm{He}} / n_{\mathrm{D}}$ | - | 1 | 0.5 | 0.09 | 0.09 |
| $x_{\mathrm{T}}=n_{\mathrm{T}} / n_{\mathrm{D}}$ | 1 | 0.00079* | 0.0012* | 0.009 | 0.0028* |
| $n_{\mathrm{D} c}, 10^{20} \mathrm{~m}^{-3}$ | 0.82 | 1.35 | 1.87 | 3.13 | 3.63 |
| $n_{e c}, 10^{20} \mathrm{~m}^{-3}$ | 1.67 | 4.12 | 3.86 | 4.07 | 4.59 |
| $n_{e p}, 10^{20} \mathrm{~m}^{-3}$ | 1.29 | 1.08 | 1.00 | 0.91 | 1.02 |
| $T_{e c}=T_{i}, \mathrm{keV}$ | 15 | 65 | 65 | 65 | 65 |
| $T_{e p}, \mathrm{keV}$ | 25 | 130 | 130 | 130 | 130 |
| $\varphi_{i}, \mathrm{keV}$ | 60 | 260 | 260 | 300 | 300 |
| $\varphi_{e}, \mathrm{keV}$ | 114 | 488 | 473 | 520 | 504 |
| $\varphi_{b}, \mathrm{keV}$ | 90 | 390 | 390 | 450 | 450 |
| $B_{0 c}, \mathrm{~T}$ | 3.3 | 5.4 | 5.4 | 5.7 | 6.1 |
| $B_{c}, \mathrm{~T}$ | 3.0 | 3.0 | 3.0 | 3.1 | 3.4 |
| $B_{0 p}, \mathrm{~T}$ | 5.1 | 5.6 | 5.6 | 5.8 | 6.2 |
| $B_{p}, \mathrm{~T}$ | 4.9 | 4.9 | 4.9 | 5.2 | 5.6 |
| $B_{m}, \mathrm{~T}$ | 14.8 | 14.8 | 14.8 | 15.6 | 16.8 |
| $\beta_{c}$ | 0.2 | 0.7 | 0.7 | 0.7 | 0.7 |
| $\beta_{p}$ | 0.077 | 0.23 | 0.22 | 0.19 | 0.19 |
| $\tau_{\perp}, \mathrm{s}$ | 1.3 | 7.1 | 6.3 | 14 | 14 |
| $L_{c}, \mathrm{~m}$ | 10 | 44 | 42 | 44 | 56 |
| $P_{f}$, MW | 157 | 685 | 654 | 685 | 885 |
| $P_{c}$, MW | 6.25 | 33.3 | 31.9 | 33.7 | 43.5 |
| $P_{p}, \mathrm{MW}$ | 9.44 | 35.2 | 33.5 | 34.8 | 44.9 |
| $P_{p-c}$, MW | 1.59 | 0.93 | 0.77 | 0.53 | 0.70 |
| $\xi_{r}=P_{r c} / P_{f}, \%$ | 1.3 | 36 | 29 | 27 | 36 |
| $\xi_{n}=P_{n c} / P_{f}, \%$ | 80 | 1.6 | 3.6 | 36 | 20 |
| $q_{n}, \mathrm{MW} / \mathrm{m}^{3}$ | 2.0 | 0.04 | 0.09 | 0.90 | 0.50 |
| $q_{w}, \mathrm{MW} / \mathrm{m}^{3}$ | 2.03 | 0.94 | 0.82 | 1.58 | 1.40 |

* D-D reaction products retained in the plasma;
** subscripts " $c$ " and " $p$ " refer to the central section and the protective ion barrier, respectively; $B_{m}$ is the field strength in the blocks; $P_{r c}$ are the radiative losses from the central section.
formulas:

$$
\begin{gather*}
P^{e-i}=\frac{32}{2} \sqrt{\frac{2}{\pi}} \alpha r_{e}^{2} m_{e} c^{3} n_{e}^{2} Z_{e f f}^{2} V \sqrt{y} \\
\times\left\{\frac{9}{8} \sqrt{\frac{\pi}{2}} \sqrt{y}\left[\ln \left(2 y+\frac{1}{2}\right)+\frac{3}{2}-C\right]+\exp (-2 y)\right\},  \tag{2}\\
p^{e-e}=\frac{4 C_{F}}{\sqrt{\pi}} \alpha r_{e}^{2} m_{e} c^{3} n_{e}^{2} V y^{3 / 2}  \tag{3}\\
\times\left(1+1.17 y+0.28 y^{2}-0.6 y^{3}\right), \quad y<1, \\
p^{e-e}=24 \alpha r_{e}^{2} m_{e} c^{3} n_{e}^{2} V y\left[\ln (2 y)+\frac{5}{4}-C\right], \quad y \geq 1, \tag{4}
\end{gather*}
$$

where $\alpha$ is the constant of the fine structure; $r_{e}$ is the
classical electron radius; $m_{e}$ is the electron mass; $c$ is the speed of light; $C=0.5772 \ldots$ is the Euler constant; $C_{F}=(5 / 9)\left(44-3 \pi^{2}\right) \approx 8 ; Z_{\text {eff }}$ is the effective ion charge in the plasma; $n_{e}$ is the electron concentration; $V$ is the volume of plasma; and $y$ is the electron temperature expressed in the rest energy units $m_{e} c^{2}$. In the nonrelativistic and ultrarelativistic cases, formulas (2)-(4) transform into the corresponding limiting expressions.

The product energy losses $P_{a}$, the energy transferred from the products to the background plasma, and the pressure of the products can be calculated using the distribution functions $f_{a}$ determined by solving the Fok-ker-Planck equation with the corresponding boundary conditions [2]. In the coordinates of $v$ (velocity) versus $\theta$ (pitch angle), the stationary Fokker- Planck equation for products of the "a" type ( $a=p, \alpha,{ }^{3} \mathrm{He}, \mathrm{T}$ ) is as
follows:

$$
\begin{align*}
& -\frac{1}{v^{2}} \frac{\partial}{\partial v} v^{2}\left[D_{v v}^{c} \frac{\partial f_{a}}{\partial v}-\left(A_{v}^{c}+A_{v}^{n}\right) f_{a}\right] \\
& -\frac{1}{v^{2} \sin \theta} \frac{\partial}{\partial \theta} \sin \theta D_{\theta \theta}^{c} \frac{\partial f_{a}}{\partial \theta}=S_{a}-\frac{f_{a}}{\tau_{\perp}}-\frac{f_{a}}{\tau_{n a}} \tag{5}
\end{align*}
$$

Here, $D_{V v}^{c}, D_{\theta \theta}^{c}$, and $A_{v}^{c}$ are the coefficients of the Coulomb diffusion and the Coulomb dynamic friction, respectively [11]; $A_{v}^{n}$ is the coefficient of friction due to the elastic nuclear scattering, calculated using the process cross sections [12]; $S_{a}$ is the particle source; and $\tau_{n a}$ is the time of product escape into the region of losses as a result of scattering on the nuclei. A characteristic feature of the product distribution functions obtained as described above is that their shapes in the region of thermal energies are close to the Maxwell distribution, while in the region of higher (epithermal) energies, the distributions are close to solutions obtained in the high-energy approximation [13].

The results of calculations of the parameters of ambipolar reactors working on the $\mathrm{D}-\mathrm{T}, \mathrm{D}-{ }^{3} \mathrm{He}$, and D-D fuels are presented in the table. For all types of fuel, the specific power of synthesis is taken equal to $5 \mathrm{MW} / \mathrm{m}^{3}$, the cyclotron radiation reflection coefficient is 0.8 , the plasma radius in the central section is 1 m , and the protecting ion barrier length is $L_{p}=3 \mathrm{~m}$. The electron temperature in the central section $\left(T_{e c}\right)$ and the fuel ion temperature ( $T_{i}$ ) are also assumed to be equal. For a given value of the electrostatic protecting ion barrier $\varphi_{i}$, the electron-confining potential $\varphi_{e}$ was determined according to the Pastukhov model [5], and the thermal barrier height $\varphi_{b}$ was calculated by the formula taken from [14]. The electron temperature in the ion barrier ( $T_{e p}$ ) corresponds to a minimum energy spent for the external heating of plasma in the reactor. The length $L_{c}$ of the central section is determined by a condition for the power gain coefficient in the reactor plasma $Q=$ $P_{f} /\left(P_{c}+P_{p}\right)=10$. The necessary value of $\tau_{\perp}$ corresponds to the power gain coefficient in the plasma of the central section $Q_{c}=P_{f} /\left(P_{c}+P_{p-c}\right)=10$, where $P_{p-c}$ is the power transmitted from hot electrons of the ion barrier to the central section. For estimating the neutron flux and the total thermal flux to the wall, the wall radius was taken equal to the plasma radius. For comparison, the table also presents data on the concentra-
tions of deuterium $n_{\mathrm{D}}$ and electrons $n_{e}$, the magnetic induction in vacuum $B_{0}$ and in the plasma $B$, and the $\beta$ values of the plasma.

An analysis of the results obtained within the framework of the kinetic model described above shows that the ambipolar plasma confinement systems offers good reactor prospects for both the $\mathrm{D}-\mathrm{T}$ fuel cycle and the alternative $\mathrm{D}-{ }^{3} \mathrm{He}$ and $\mathrm{D}-\mathrm{D}$ cycles. However, there are certain principal difficulties encountered in providing the required values of parameters of the ambipolar confinement system, including sufficiently high temperatures and potential barriers and relatively large (on the level of $\sim 10 \mathrm{~s}$ ) radial transfer time.

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# Cantilever Tip-Surface Contact as a Catastrophe Machine 

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#### Abstract

The contact of a cantilever tip with a sample surface in a scanning-probe microscopy is considered for the first time as a catastrophe machine. Using an approach of the catastrophe theory, behavior of the tipsurface system is analyzed depending on the internal parameters (stiffness and coordinates of the cantilever holder). The tip jumps and the adhesion force variations in the course of the vertical and lateral cantilever motions are calculated. Projections of the equilibrium surface are constructed and the regimes are established that eliminate the "jumps to contact" and "microslips." A new method for the surface vacancy diagnostics and a new algorithm for the controlled tip approach to the surface are proposed. © 2000 MAIK "Nauka/Interperiodica".


Sliding friction was originally studied within the framework of simple theoretical models of independent [1] and coupled [2] oscillators. In both theories, the interatomic forces were modeled by springs arranged perpendicularly or parallel to the sliding direction [3]. The discovery of atomic-force [4], friction [5], and adhesion [6] microscopes, with a cantilever probe being in common for all these techniques, gives a new life to the old models. Indeed, using springs one cannot only model the atomic bonds but simulate the cantilever motions as well [7-11]. Interaction of the cantilever tip with a sample surface can be described by a potential, the parameters of which have to be determined, in the general case, taking into account both the contact geometry and the types of chemical bonds in the system studied.

Devices that can feature the events called "catastrophes," whereby the system exhibits a jumplike transition from one state to another, are called catastrophe machines [12]. In this paper, the mechanical part of scanning-probe microscopes [4-6] is considered as adhesional or frictional catastrophe machines schematically depicted in Figs. 1a and 2a, respectively. According to this approach, a spring of variable stiffness $k$ with a tip (representing a tungsten atom) fixed on one end modeling the cantilever of a scanning-probe microscope. Another end of the spring is fixed at the holder. The spring axis can be perpendicular (Fig. 1a) or parallel (Fig. 2a) to the sample surface. The spring can both expand and contract, on the one hand, due to a change in coordinates of the holder $\left(Z_{h}\right.$ or $\left.X_{h}\right)$ and the tip $\left(Z_{t}\right.$ or $X_{t}$ ) and, on the other hand, as a result of variation of the stiffness $k$. The tip interacts with the surface modeled by a single tungsten atom (Fig. 1a) or by a chain of such atoms (Fig. 2a). The potential of interactions of the sur-
face atoms with each other and with the tip atom was represented, without loss of generality, by the same Morse potential for tungsten [13].

The state of an adhesional or frictional catastrophe machine is described by three quantities: tip coordinate ( $Z_{t}$ or $X_{t}$ ); cantilever holder coordinate $\left(Z_{h}\right.$ or $\left.X_{h}\right)$; and variable stiffness $k$. Once all the three parameters are given, the potential energy of the entire system is uniquely determined. The spring will stretch so as to minimize the energy (at least on the local level). Let us consider the holder coordinate and the spring stiffness as the control parameters and the tip coordinate, as the internal parameter of the system. This choice is justified by the fact that the design of scannning-probe microscopes provides for the principal possibility of setting and controlling only the holder coordinate and the cantilever stiffness, while the tip coordinate can be only monitored by measuring the cantilever deviations.

In the three-dimensional space of states of the catastrophe machine, the states in which the tip occurs in equilibrium form smooth surfaces called the equilibrium surfaces $Z_{t}\left(k, Z_{h}\right)$ and $X_{t}\left(k, X_{h}\right)$. In each equilibrium state of the tip, represented by a point on this surface, the force of the adhesion interaction between tip and surface is compensated by the force of spring stretching. The total potential energy of the tip-surface system represents a sum of the term describing the tip-surface interaction, $U_{a}\left(Z_{t}-Z_{s}\right)$ or $U_{a}\left(X_{t}-X_{s}\right)$, and the term describing the energy stored in the spring, $k\left(Z_{t}-Z_{h}\right)^{2} / 2$ or $k\left(X_{t}-X_{h}\right)^{2} / 2$. In order to derive an equation describing the equilibrium surface, we have to minimize the total potential energy of the system with respect to the internal parameter, taking into account that $Z_{s}=X_{s}=0$.


Fig. 1. (a) A schematic diagram of the adhesional catastrophe machine: (S) sample surface; (T) probing tip; (C) cantilever spring; (H) cantilever holder. (b) Plots of the tip coordinate $Z_{t}$ versus the spring holder coordinate $Z_{h}$ for various stiffness values (indicated at the curves). (c) Plots of the adhesion force variation for a spring holder with $k=$ $0.25 \mathrm{~N} / \mathrm{m}$ moved to and from the surface; $\Delta F_{a}$ are the force jumps at the instants of "sticking" and "desticking." Thin and thick arrows in (a-c) indicate directions of the holder motion and the tip jumps. Thick and thin solid curves in (b, c) correspond to stable and unstable branches.

For an adhesional catastrophe machine, the resulting equation is as follows [8, 9]:

$$
\begin{gather*}
\partial U_{\mathrm{tot}}\left(Z_{t}, Z_{h}, k\right) / \partial Z_{t} \\
=\partial U_{a}\left(Z_{t}\right) / \partial Z_{t}+k\left(Z_{t}-Z_{h}\right)=0 . \tag{1}
\end{gather*}
$$

By the same token, for a frictional catastrophe machine, we obtain [9-11]

$$
\begin{gather*}
\partial U_{\text {tot }}\left(X_{t}, X_{h}, k\right) / \partial X_{t}  \tag{2}\\
=\partial U_{a}\left(X_{t}\right) / \partial X_{t}+k\left(X_{t}-X_{h}\right)=0 .
\end{gather*}
$$

The solutions to these equations are conveniently represented in the form of the curves $X_{t}\left(X_{h}\right)$ and $Z_{t}\left(Z_{h}\right)$ representing the cross sections of the corresponding equilibrium surfaces by the planes of constant stiffness $k=$ const. Figures 1 b and 2 b show examples of such curves for some values of the stiffness frequently encountered in experiments.

Now, we will show how a catastrophe can develop in the adhesional catastrophe machine with a fixed stiff-


Fig. 2. (a) A schematic diagram of the frictional catastrophe machine (notations the same as in Fig. 1). (b) Plots of the tip coordinate $X_{t}$ versus the spring holder coordinate $X_{h}$ for various stiffness values (indicated at the curves). (c) Plots of the friction force variation for a spring holder with $k=0.25 \mathrm{~N} / \mathrm{m}$ moved along the surface; $\Delta F_{a}$ are the force jumps at the instants of single "microslips" and "desticking". Thick arrows in $(\mathrm{a}-\mathrm{c})$ indicate directions of the holder motion, while single and double thin arrows indicate the single and double tip jumps. Thick and thin solid curves in (b, c) correspond to stable and unstable branches.
ness of $k=0.25 \mathrm{~N} / \mathrm{m}$. Let the system to occur at point 4 (Fig. 1b) and the holder to approach the surface. The catastrophe ("sticking") arises when the system reaches point 1 . Then, the equilibrium breaks and a "jump to contact" takes place whereby the tip shifts from point 1 to 2 , displacing by $\Delta Z \approx 0.2 \mathrm{~nm}$. Now let us move the holder away from the surface. In this case, the catastrophe ("desticking") takes pace when the system reaches point 3 . Here, the equilibrium breaks again and the tip "jumps out of contact" to point 4 , displacing by $\Delta Z \approx$ 0.44 nm . Thus, the system turns back to the initial state. For "stiff" springs, both the "jumps to and out of contact" and the area of the resulting hysteresis cycle is smaller than those for "soft" springs (cf. cycles 8-5-6-$7-8$ and $4-1-2-3-4$ in Fig. 1b). When the stiffness exceeds a critical level of $0.8 \mathrm{~N} / \mathrm{m}(k=2$ or $1000 \mathrm{~N} / \mathrm{m})$, the catastrophes become impossible since there is a single equilibrium tip coordinate for any position of the holder. For large stiffness values (exceeding $1000 \mathrm{~N} / \mathrm{m}$ ), the $Z_{t}\left(Z_{h}\right)$ curve degenerates into a straight line $Z_{t}=Z_{h}$.

Now, let us consider the catastrophe development in the frictional catastrophe machine with a fixed stiffness of $k=0.25 \mathrm{~N} / \mathrm{m}$. Let the system to occur at point 4 (Fig. 2b) and the holder to move on the left along the $X$ axis. The catastrophe ("sticking") arises when the system reaches point 1 . Then, the equilibrium breaks and a "microslip" takes place whereby the tip jumps from point 1 to a new equilibrium at point 2 , displacing by $\Delta X \approx 0.17 \mathrm{~nm}$, or at point 10 with $\Delta X \approx 0.39 \mathrm{~nm}$ (the latter case corresponds to a double "microslip" resulting in the transition to another branch). As the holder shifts further to point 5 , the equilibrium is broken again and another "microslip" takes place, with the tip jumping to point 6 . Once a "double" microslip had occurred at point 1 , the system would move from point 10 to 6 without jumps. Now let us reverse the direction of the holder motion. In this case, the equilibrium is broken when the system passes from point 6 to 7 . By analogy with the above, either single (to point 8) or double (to point 9) "microslip" can take place. As the holder shifts further in the same direction, the equilibrium is broken again at point 3 , with the tip jumping to point 4 . Once a "double" microslip had occurred at point 7, the system moves from point 9 to 4 without jumps. Note that only single "microslips" are possible for a spring stiffness of $k=0.5 \mathrm{~N} / \mathrm{m}$ and no hysteresis at all is observed for large stiffness values (exceeding $30 \mathrm{~N} / \mathrm{m}$ ), whereby the $X_{t}\left(X_{h}\right)$ curve degenerates into a straight line $X_{t}=X_{h}$ (Fig. 2b). Thus, the cycles of holder scanning may give rise to the hysteresis loops corresponding to single "microslips" (e.g., 1-2-3-4-1 or 5-6-7-8-5) and to more complicated paths involving double "microslips" ( $1-10-7-9-1$ ) or combinations of single and double "microslips" (1-2-5-6-7-9-1), which were experimentally observed in [5]. It is interesting to note that the tip may, in principle, fall onto an unstable branch as well. In this case, the tip is situated strictly above an atom of the chain (see black points in Fig. 2b).

At the instants of "jumps to contact" and "microslips," the total potential energy of the system drops, the difference $\Delta U_{\text {tot }}$ being spent both for the excitation of elastic oscillations and for the deformation of contacting surfaces. The jumps of the force $\Delta F_{a}$ at the instants of "microslips" $[5,11,14]$ and "desticking" $[6,14]$ measured experimentally (e.g., by scan-ning-probe microscopes of the NT-MDT Solver type) can be calculated within the framework of the proposed model using the $F_{a}\left(X_{h}\right)$ or $F_{a}\left(Z_{h}\right)$ curves. Figures 1c and 2c show the plots of $F_{a}\left(X_{h}\right)$ and $F_{a}\left(Z_{h}\right)$ for $k=0.25 \mathrm{~N} / \mathrm{m}$. As seen, the force jump modulus $\Delta F_{a}$ at the instant of "sticking" (transition from point 1 to 2 ) amounts to 0.53 nN , while the value corresponding to "desticking" (transition from point 3 to 4 ) is almost twice as great, $\Delta F_{a}=1.1 \mathrm{nN}$ (Fig. 1c). The force jump modulus $\Delta F_{a}$ for a single "microslip" (transition from point 1 to 2 ) is 0.43 nN , while a double "microslip" (transition from point 1 to 10 ) is characterized by a more than twice


Fig. 3. Projections of the equilibrium surfaces onto the planes of control parameters for (a) adhesional catastrophe machine $Z_{t}\left(k, Z_{h}\right)$ and (b) frictional catastrophe machine $X_{t}\left(k, Z_{h}\right)$, showing the folder lines and junction points; $\psi$ is the gradient of the tip coordinate as function of the holder coordinate. Points in the cross-hatched region ( $\psi<0$ ) correspond to the projections of three points of the equilibrium surface, while points in the open region ( $\psi>0$ ) correspond to a single equilibrium point.
greater value of $\Delta F_{a}=0.98 \mathrm{nN}$ (Fig. 2b). The area of the hysteresis cycles in the $X_{t}\left(X_{h}\right)$ or $Z_{t}\left(Z_{h}\right)$ curves, as well as that in the $F_{a}\left(X_{h}\right)$ or $F_{a}\left(Z_{h}\right)$ curves, increases with decreasing stiffness of the springs. This is related to the fact that both ends of the stable branches of these curves "extend" in the opposite directions along the $X_{h}$ or $Z_{h}$ axes.

Proceeding from the above model considerations, we may suggest a new method for diagnostics of the surface point defects (vacancies) based on the detection of tip jumps that are more pronounced over these sites. Indeed, the region of a single vacancy corresponds to the "matching" of two stable branches, a bivacancy corresponds to the conjugation of three branches, and so on. Therefore, the tip will exhibit double jumps in the vicinity of a singe vacancy, tripe jumps at bivacancies, etc. By assessing multiplicity of the tip jumps, it is possible to determine the dimensions of vacancy clusters. We may suggest that the double "microslips" originally observed in [5] were related to the tip jumping over vacancies. However, it must be taken into account that the tip jumping over vacancies may fall on the second stable branch rather than on the adjacent one to provide for the release of a greater kinetic energy. The proposed method is markedly different from the conventional techniques based on the constant-force line bending, which is used for the point defect observation in the atomic force microscope [15, 16].

For the investigation of bifurcations in the equilibrium tip position, it is convenient to use a theory describing the features of smooth mapping in the catastrophe theory [12]. Consider the planes of control parameters $\left(k, Z_{h}\right)$ and $\left(k, X_{h}\right)$ for the catastrophe machines considered above. Let us construct projections of the solutions to Eqs. (1) and (2) (that is, of the smooth equilibrium surfaces) onto these planes. These
projections involve the folder lines and the junction points. The folder point projections represent the catastrophe lines, or the "sticking" curves, dividing the plane of the control parameters into two parts (Fig. 3). Points in the cross-hatched region correspond to projections of three (Fig. 3a) or more (Fig. 3b) points of the equilibrium surface, while points in the open part correspond to a single equilibrium point. The points on the curve correspond to either two (Fig. 3a) or three (Fig. 3b) equilibrium points. When the catastrophe curve is crossed on the passage from the cross-hatched to open region, two inverse images (of the three) merge together and then disappear (exhibiting a feature called the folder line). On approaching the sharp vertices, the three inverse images merge to form another feature, the junction point. The jumps can be avoided provided that it is known how to pass from one to another point on the equilibrium surface. For example, it is seen from Fig. 3a that crossing of the catastrophe curve on the passage by the segment AF from point A to F leads to the "jump to contact" (transition from point 1 to 2 in Fig. 1b), while moving from F to A by the same segment corresponds to the "jump out of contact" (transition from point 3 to 4 in Fig. 1b). However, any catastrophes are excluded on traveling along the broken line ABCDEF by-passing the junction point. For this purpose, it is sufficient to increase the stiffness above the critical level ( $0.8 \mathrm{~N} / \mathrm{m}$ ) on approaching the catastrophe line (points B and E ) and by-pass the junction point on the right. By the same token, it is possible to avoid "microslips" in the cycles of lateral sliding (Fig. 3b) by increasing the spring stiffness (above $30 \mathrm{~N} / \mathrm{m}$ ) on approaching the catastrophe curve at the points B and E .

Based on the above analysis of states of the catastrophe machine, we may suggest a new algorithm for the controlled tip approach to the sample surface. According to this, the approach is conducted so as to control the parameter $\psi\left(Z_{h}, k\right)=\operatorname{grad}\left[Z_{h}\left(Z_{t}\right)\right]$. The positions $Z_{h}$ at which the parameter $\psi$ turns zero correspond to crossing the catastrophe curve on the plane of the control parameters (Fig. 3a). The positions where $\psi<0$ correspond to the cross-hatched region and those where $\psi>0$, to the open region. Thus, by controlling the approach so that $\psi$ is always positive, we may avoid a catastrophe. The essence of the algorithm is that, when the $\psi$ value approaches zero (but not when it turns zero!), the feedback must operate so as to increase the cantilever stiffness. As soon as the $\psi$ value tends to increase, the feedback circuit may be switched off.

The laws of the catastrophe development described above must be taken into account in designing atomic force microscopes (in particular, in calculating the friction forces [17] and the phase shifts in the dynamic mode [18]) and in developing methods for the calculation of interatomic potentials using the tip loading
curves [19]. The proposed approach can also be applied to more realistic models, using a greater number of atoms at the contact zone and taking into account the relaxation effects. In addition, this approach can be applied to the analysis of capillary, electrostatic, magnetic, and other interactions between a probing tip and the surface.

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# Rapid Monitoring of the Charging Properties of Ion-Doped MOS Structures 

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#### Abstract

A method for the analytical modeling of the high-frequency capacitance-voltage ( $C-V$ ) characteristics of ion-doped MOS structures is proposed. The approach is based on the Gaussian approximation of the implanted dopant concentration profile and the exhaustion layer approximation (in the region of capacitance modulation). The theoretical $C-V$ curves calculated by the proposed method were applied to monitoring the charging parameters of MOS structures. The new method is recommended for the rapid evaluation of the charging characteristics of a $\mathrm{SiO}_{2}$ /Si interface. © 2000 MAIK "Nauka/Interperiodica".


Methods based on the comparative analysis of experimental and theoretical $C-V$ curves, which are very informative and can be readily automated, are still widely used for evaluation of the charging properties of silicon-oxide interfaces in semiconductor devices [1,2]. The validity of this approach depends both on the accuracy of experimental measurements and on the correctness of constructing theoretical characteristics determined by a semiconductor doping profile, which can be significantly inhomogeneous in the ion-doped MOS structures.

Most adequate results are provided by numerical calculations of the theoretical $C-V$ curves based on the real depth profiles of a dopant and the mobile charge concentration in a semiconductor, but this method is rather cumbersome and time-consuming because of an unstable character of the boundary problem solution for a Poisson equation with the right-hand part containing a term corresponding to a complicated dopant depthconcentration profile. This approach is not always justified when it is only necessary to rapidly obtain estimates of the charging parameters of a $\mathrm{SiO}_{2} / \mathrm{Si}$ interface. On the other hand, estimates based on the theoretical $C-V$ curves analytically constructed using a concept of the space-charge region in the semiconductor and simple approximations of the dopant profile (represented, e.g., by the effective concentration [3], $\delta$-function, or displaced rectangle [4]) frequently lead to considerable errors in the charging parameters, in particular, in the energy spectrum of surface states [5].

The depth-concentration profile of an implanted impurity in the $\mathrm{SiO}_{2} / \mathrm{Si}$ system [6] can be described by a Gaussian distribution function. The postimplantation heat treatments lead to a change in the initial profile, which can be calculated using Fick's equations. It was established that the diffusion length of a dopant in most of the practically important cases is either markedly greater (for prolonged anneals) or significantly smaller
(for a short-time low-temperature treatment) as compared to the standard displacement of ions implanted into silicon [7]. The resulting dopant depth-concentration profile can be described with an allowance of the dopant segregation at the $\mathrm{SiO}_{2} / \mathrm{Si}$ interface by the following modified Gaussian distribution function [7]:

$$
\begin{gather*}
N(x)=N_{B} \\
+\frac{D_{I}}{\sqrt{2 \pi} \sigma}\left(\exp \left[-\frac{\left(x_{c}-x\right)^{2}}{2 \sigma^{2}}\right]+\gamma \exp \left[-\frac{\left(x_{c}+x\right)^{2}}{2 \sigma^{2}}\right]\right), \tag{1}
\end{gather*}
$$

where $N_{B}$ is the base impurity concentration in silicon, $D_{I}$ is the dose of implanted ions, $x_{c}$ is the centroid of the implanted impurity, and $\gamma$ is a coefficient taking into account the boundary profile. For phosphorus, the latter coefficient is $\gamma=1$, while for boron (upon a typical heat treatment at $\left.900-1100^{\circ} \mathrm{C}\right)|\gamma|<0.1$ [7]. The effective standard deviation of the dopant impurity is $\sigma=\sqrt{\Delta R_{\mathrm{PSi}}^{2}+2 \sum_{j} D_{j} t_{j}}$, where $D_{j}$ is the dopant diffusion coefficient at a temperature of the $j$ th anneal, $t_{j}$ is the duration of the $j$ th anneal, and $\Delta R_{\mathrm{PSi}}$ is the meansquare displacement of the implanted ions in silicon.

In the case of modulated (differential) capacitance measurements [8], the most informative region of the capacitance variation is that where the surface potential $\psi_{s}$ varies from 0 (flat bands) to $2 \varphi_{0}$ corresponding the
onset of strong inversion. Here, $\varphi_{0}=\frac{k T}{q} \ln \left(\frac{N(x=0)}{n_{i}}\right)$
is the Fermi potential on the semiconductor surface, where $k$ is the Boltzmann constant, $T$ is the absolute temperature, $q$ is the electron charge, $n_{i}$ is the intrinsic charge carrier concentration, and $N(x=0)$ is the dopant concentration at the semiconductor surface. Under these conditions we may use the approximation of an


Fig. 1. The $C-V$ curves obtained by $(1,3)$ experimental methods and $(2,4,5)$ theoretical calculations for a MOS structure on a silicon substrate $(1,2)$ before and $(3,4,5)$ after implantation of phosphorus ions $\left(E=100 \mathrm{keV} ; D_{I}=\right.$ $0.06 \mu \mathrm{C} / \mathrm{cm}^{2}$ ). The $C-V$ curves of ion-doped samples were numerically calculated using the approximations of (4) an effective constant dopant concentration and (5) a Gaussian profile of the implanted dopant. The base dopant (phosphorus) concentration is $N_{B}=3 \times 10^{14} \mathrm{~cm}^{-3}$ and the silicon oxide layer thickness is $d_{i}=24 \mathrm{~nm}$.
exhaustion layer in the semiconductor, in which case the charge $Q_{B}$ in the space-charge region is determined by the first integral of the Poisson equation with the right-hand part in the form of (1) and can be expressed as follows [9]:

$$
\begin{align*}
Q_{B}= & q\left[N_{B} x_{0}+\frac{D_{I}}{2}\left(\operatorname{erf} \frac{x_{c}}{\sigma \sqrt{2}}+\operatorname{erf} \frac{x_{0}-x_{c}}{\sigma \sqrt{2}}\right)\right.  \tag{2}\\
& \left.+\gamma\left(\operatorname{erf} \frac{x_{0}+x_{c}}{\sigma \sqrt{2}}-\operatorname{erf} \frac{x_{c}}{\sigma \sqrt{2}}\right)\right],
\end{align*}
$$

where $x_{0}$ is the depth of the depletion region and $\operatorname{erf}(x)=\frac{2}{\sqrt{\pi}} \int_{0}^{x} \exp \left(-t^{2}\right) d t$ is the error function (probability integral). The capacitance $C_{s c}$ of the space-charge region is defined as the derivative of the charge of this region $Q_{B}$ with respect to the surface potential $\psi_{s}$. The latter potential can be determined analytically as the second integral of the Poisson equation at $x=0$ [9]:

$$
\begin{gather*}
\psi_{s}=-\frac{q}{2 \varepsilon_{s}}\left\{N_{B} x_{0}^{2}\right. \\
+D_{I}\left[\sigma \sqrt{\frac{2}{\pi}}\left(\exp \left(-\frac{x_{c}^{2}}{2 \sigma^{2}}\right)-\exp \left(-\frac{\left(x_{0}-x_{c}\right)^{2}}{2 \sigma^{2}}\right)\right)\right. \\
\left.+x_{c}\left(\operatorname{erf} \frac{x_{c}}{\sigma \sqrt{2}}+\operatorname{erf} \frac{x_{0}-x_{c}}{\sigma \sqrt{2}}\right)\right] \tag{3}
\end{gather*}
$$



Fig. 2. Theoretical energy spectra of the density of surface states for a MOS structure on a silicon substrate (1) before and $(2,3)$ after implantation of phosphorus ions $(E=100 \mathrm{keV}$; $D_{I}=0.06 \mu \mathrm{C} / \mathrm{cm}^{2}$ ). The spectra of ion-doped samples were calculated using (2) the approximations of an effective constant dopant concentration and (3) an analytical $C-V$ curve for a Gaussian profile of the implanted dopant. The base dopant (phosphorus) concentration is $N_{B}=3 \times 10^{14} \mathrm{~cm}^{-3}$ and the silicon oxide layer thickness is $d_{i}=24 \mathrm{~nm}$.

$$
\begin{gathered}
+\gamma\left[x_{c}\left[\operatorname{erf} \frac{x_{c}}{\sigma \sqrt{2}}-\operatorname{erf} \frac{x_{0}+x_{c}}{\sigma \sqrt{2}}\right]\right. \\
\left.\left.+\sigma \sqrt{\frac{2}{\pi}}\left[\exp \left(-\frac{x_{c}^{2}}{2 \sigma^{2}}\right)-\exp \left(-\frac{\left(x_{0}+x_{c}\right)^{2}}{2 \sigma^{2}}\right)\right]\right]\right\}
\end{gathered}
$$

Both the charge and the band bending in the spacecharge region depend on the parameter $x_{0}$. Taking the derivative $d Q_{B} / d \psi_{s}$ of the parametric function, we obtain the following expression for the capacitance of the space-charge region:

$$
\begin{equation*}
C_{s c}=\frac{d Q_{B}}{d \psi_{s}}=\frac{d Q_{B} / d x_{0}}{d \psi_{s} / d x_{0}}=\frac{\varepsilon_{s}}{x_{0}} . \tag{4}
\end{equation*}
$$

The total capacitance $C$ of the MOS structure is calculated by the formula

$$
\begin{equation*}
C=\frac{C_{i} C_{s c}}{C_{i}+C_{s c}}=\frac{C_{i} \varepsilon_{s}}{C_{i} x_{0}+\varepsilon_{s}}, \tag{5}
\end{equation*}
$$

where $C_{i}$ is the intrinsic (geometric) capacitance of the oxide.

The gate voltage $V_{\mathrm{g}}$ corresponding to the band bending $\psi_{s}$ is given by the well-known formula [1]

$$
\begin{equation*}
V_{\mathrm{g}}=-\frac{Q_{B}\left(x_{0}\right)}{C_{i}}+\psi_{s}\left(x_{0}\right) . \tag{6}
\end{equation*}
$$

Equations (5) and (6) represent a parametric relationship $C\left(V_{\mathrm{g}}\right)$ for $C$ and $V_{\mathrm{g}}$ depending on the parameter
$x_{0}$-the depth of the depletion region. By varying the parameter $x_{0}$ from the effective Debye length $L_{\mathrm{D}(\mathrm{eff})}=$ $\sqrt{\varepsilon_{s} k T /\left[q^{2} N(x=0)\right]}$ to a maximum value $x_{0(\max )}$ attained in the case of strong inversion, we may calculate the $C-V$ curve in the region of capacitance modulation. Matching this curve to the values of capacitance in the regions of enrichment $\left(C=C_{i}\right)$ and strong inversion $\left(C=C_{\text {min }}\right)$, which coincide with the experimental values, we obtain the theoretical high-frequency $C-V$ curve of an ion-doped MOS structure. Attaining the region of strong inversion is indicated by the surface potential $\psi_{s}$ approaching its maximum value $2 \varphi_{0}$.

Thus, we have analytically constructed the total high-frequency $C-V$ curve of an ion-doped MOS structure. Note that this analytical $C-V$ curve virtually coincides with the characteristics calculated by numerically integrating the Poisson equation with a Gaussian profile of the dopant concentration (Fig. 1). As an example, Fig. 2 shows the energy spectrum of surface states in a MOS structure calculated using the proposed method. As seen, the ion implantation does not significantly distort the PS spectrum and only slightly increases the spectral density of surface states rather than giving rise to a feature that can be misinterpreted as a monolevel (as does the effective concentration approximation presented for the comparison).

Thus, using a Gaussian distribution function to approximate the implanted dopant profile, we may
avoid the need in numerically solving the Poisson equation and analytically construct with high precision the $C-V$ curves of an ion-doped MOS structure to be used for comparison with the results of high-frequency $C-V$ measurements.

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# The Dislocation Anharmonicity in Silicon 

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#### Abstract

The effect of the dislocation anharmonicity on the Young modulus in bending silicon single crystals was studied. It is shown that changes in the Young modulus are caused by the contribution of dislocations to the fourth-order elasticity moduli. It is revealed experimentally that the bending deformation in both donor and acceptor silicon crystals favors the appearance mainly of the edge dislocations with the axes forming an angle $\theta \sim 90^{\circ}$ with the Burgers vector. © 2000 MAIK "Nauka/Interperiodica".


As is well known, study of the propagation of elastic waves in a condensed medium provides information on their velocity and energy losses, the defect structure of this medium, etc. The knowledge of the wave velocity and the specific density allows one to determine the elastic constants of this medium, which, in turn, are related to the interatomic forces [1-3]. Variation of the elastic moduli of solids can be caused by various technological operations giving rise to defect formation. For example, the presence of dislocations in a medium results in considerable changes in the nonlinear elastic moduli, which depend on the type and specific features of these dislocations. This statement was repeatedly confirmed by experiments on various metals [3, 4]. However, the dislocation anharmonicity in semiconductors is almost unstudied, although the linear defects in semiconductors are formed both during crystal growth and the operation of semiconductor-based devices and integrated microcircuits. Therefore, we undertook this study to establish a relationship between the dislocation structure and the dislocation anharmonicity in silicon crystals during their deformation.

The experiments were made on dislocation-free $n$ and $p$-type silicon wafers with a fixed resistivity $\rho$ ranging from 10.00 to $0.01 \Omega \mathrm{~cm}$ in different specimens. Edge dislocations were introduced by $30-\mathrm{min}$ annealing of the wafers under loading along the [111] direction at a temperature of $1000^{\circ} \mathrm{C}$ [5]. The fixed dislocation density in different wafers ranged from $10^{4}$ to $10^{6} \mathrm{~cm}^{-2}$.

The effect of the dislocation anharmonicity on the Young modulus $E_{0}$ was recorded during the bending of silicon single crystals. Then the data obtained were analyzed using the nonlinear Hooke's law for solids,

$$
\begin{equation*}
\sigma=E_{0} \varepsilon+\alpha \varepsilon^{2}+\beta \varepsilon^{3} \tag{1}
\end{equation*}
$$

and an expression for the elastic energy per unit volume
of the crystal (which follows from Hooke's law)

$$
\begin{equation*}
W=\frac{1}{2} E_{0} \varepsilon^{2}+\frac{1}{3} \alpha \varepsilon^{3}+\frac{1}{4} \beta \varepsilon^{4} . \tag{2}
\end{equation*}
$$

Here, $\alpha$ and $\beta$ are linear combinations of the third- and fourth-order elasticity moduli (in Pa), respectively, and $\varepsilon$ is the elastic deformation.

The modulus $\alpha$ in Eq. (2) is the coefficient at the term containing elastic deformation in an odd power and, therefore, the alternating-sign contribution of this term into the elastic energy depends on the sign of $\varepsilon$. Thus, in the case of bending down, the upper part of the wafer is compressed with respect to the neutral layer ( $\alpha \varepsilon^{3}<0$ ), whereas the lower part is extended ( $\alpha \varepsilon^{3}>0$ ), so that averaging over the cross section yields the zero value [6].

Unlike the above case, any alternating-sign deformation results in the same change in $W$ in the term containing $\beta$. While calculating the dislocation contribution to the nonlinear moduli, one has to take into account the lattice $\left(\beta_{\mathrm{l}}\right)$, dislocation $\left(\beta_{\mathrm{d}}\right)$, and concentration $\left(\beta_{\mathrm{c}}\right)$ anharmonicities: ${ }^{1}$

$$
\begin{equation*}
\beta=\beta_{1}+\beta_{d}+\beta_{c} . \tag{3}
\end{equation*}
$$

In the presence of a dislocation structure, we have [4]

$$
\begin{equation*}
\beta_{\mathrm{d}}=\frac{\left(1+3 v-7 v \sin ^{2} \theta\right) N_{\mathrm{d}}^{-} L^{4} \Omega^{4} E_{0}^{4}}{160\left(1+v-3 v \sin ^{2} \theta\right)^{4} b^{2} g^{3} \mu^{3}} \tag{4}
\end{equation*}
$$

and $\beta_{1} \ll \beta_{d}$. Here, $\Omega$ is the orientation factor; $\mu$ is the shear modulus; $g$ is the dimensionless coefficient characterizing the extension of the elastic field of dislocations; $N_{\mathrm{d}}$ and $L$ are the density and the length of dislocations, respectively; $v=0.27$ is the Poisson ratio for

[^0]silicon [3]; and $\theta$ is the angle between the Burgers vector $b$ and the dislocation axis.

For an edge dislocation, the sign of the modulus $\beta_{1} \approx \beta_{d}$ is positive, whereas, for the prevailing screw component, the sign is negative. Therefore, measuring $\beta$ of the dislocation-containing specimen with a low charge carrier concentration, one can determine the nature of the linear defects in a semiconductor.

For the experimental determination of the sign of $\beta$, we employed the method of a composite vibrator using piezoeceramic transducers with two metallized faces glued to the wafer ends. This provided excitation of the zero modes of the Lamb waves along the [110] direction. The width of the 6 - to $8-\mathrm{mm}$-long transducers was chosen to be as close as possible to the wafer thickness (400-500 $\mu \mathrm{m}$ ).

Then, the wafers were bent along the [111] direction on a specially designed device with the spherical supports 0.5 mm in diameter. The wafer bending was measured with a micrometer to within an accuracy of $\pm 2.5 \mu \mathrm{~m}$. The maximum bending at the wafer center was $800 \mu \mathrm{~m}$ (the total wafer length was 60 mm ). More pronounced deformations led to complete failure of the specimen.

The wafer studied was connected in the feedback circuit of a high-frequency resonance amplifier. The amplitude of the resonance frequency was monitored with the aid of an S1-83 oscillograph and an RCh3-070002 frequency meter. The effect of deformation on the elastic wave velocity in a semiconductor was measured within a frequency range from 3 to 7 MHz . First, the vibrational contour was tuned to the resonance frequency and maintained at this frequency for three minutes to attain the thermal equilibrium and diminish the drift of the resonance frequency. Then, the frequency meter readings were recorded under load and upon unloading the crystal. The relative changes in the frequency caused by a drift of the ambient medium parameters did not exceed $10^{-5}$.

When longitudinal vibrations are excited in a semiconductor silicon wafer of length $L_{\mathrm{PL}}$ and thickness $h_{\mathrm{PL}}$ subjected to the three-point bending, the total deformation is a sum of the vibrational deformation $\varepsilon_{v}$ and the static deformation $\varepsilon_{0}$. In this case, the vibration parameters are characterized by the dynamic modulus

$$
\begin{equation*}
E_{\mathrm{d}}=E_{0}\left(1+\left(\frac{3 \beta}{2 E_{0}}-\frac{\alpha^{2}}{E_{0}^{2}} \frac{h_{\mathrm{PL}}^{2}}{6 r_{\mathrm{PL}}^{2}}\right) .\right. \tag{5}
\end{equation*}
$$

Here, the curvature radius $r_{\mathrm{PL}}^{2}$ at small bending is determined by the second derivative of the wafer profile $y(x)$. For the loading scheme used with the fixed dis-


Influence of the bending deflection $d$ on the change of the vibrational frequency in (a) phosphorus-doped specimens $(\rho=10 \Omega \mathrm{~cm})$ with the dislocation densities $N_{\mathrm{d}}=8 \times 10^{4}(1)$, $7 \times 10^{5}(2)$, and $2 \times 10^{6} \mathrm{~cm}^{-2}(3)$ and (b) boron-doped specimens $(\rho=20 \Omega \mathrm{~cm})$ with $N_{\mathrm{d}} 1 \times 10^{5}(1), 5 \times 10^{5}(2)$, and $1 \times 10^{6} \mathrm{~cm}^{-2}(3)$.
tance $P$ between the supports, the wafer profile is described by the known function [4]

$$
\begin{equation*}
y(x)=d\left(-\frac{4 x^{3}}{p^{3}}+\frac{6 x^{2}}{p^{2}}-1\right), \tag{6}
\end{equation*}
$$

where $d$ is the bending deflection (sag).
In order to determine the elastic moduli, we used the measuring scheme consisting of a self-excited genera-
tor with the resonance condition being determined by the total phase

$$
\begin{gather*}
\varphi=2 \pi f \int_{-L / 2}^{L / 2} \frac{d x}{v(x)} \\
=\frac{2 \pi f}{\mathrm{v}_{0}} \int_{-L / 2}^{L / 2} \frac{d x}{\sqrt{\left(1+\left(\frac{3 \beta}{2 E_{0}}-\frac{\alpha^{2}}{E_{0}^{2}}\right) \frac{h_{\mathrm{PL}}^{2}}{6 r_{\mathrm{PL}}^{2}}\right)}}=2 \pi N  \tag{7}\\
N=1,2,3 \ldots
\end{gather*}
$$

Here, $v=\sqrt{\frac{E}{d_{\mathrm{Si}}}}$ is the sound velocity in the crystal and $d_{\mathrm{Si}}$ is the silicon density.

Integration of this equation with due regard for Eqs. (5) and (6) yields an expression for the relative change in the generation frequency $\Delta f=f-f_{0}$ of the ultrasonic signals of the vibrational system with the deformed wafer:

$$
\begin{equation*}
\frac{\Delta f}{f} \approx \frac{d f}{f}=4 \frac{h_{\mathrm{PL}}^{2}}{p^{3} L_{\mathrm{PL}}}(\Delta d)^{2}\left(\frac{3 \beta}{2 E_{0}}-\frac{\alpha^{2}}{E_{0}^{2}}\right) \tag{8}
\end{equation*}
$$

Here, $f_{0}$ and $d_{0}$ are the frequency of self-excited vibra and the bending deflection in the absence of deformation, $f$ and $d$ are the same quantities under deformation, and $\Delta d=d-d_{0}$.

For a more convenient analysis, let us represent Eq. (8), with due regard for Eq. (3) and for the absence of the term containing $\alpha$ during specimen bending, by the following equations:

$$
\begin{align*}
& \left\{\frac{\Delta f}{(\Delta d)^{2}}\right\}_{\mathrm{S}}=4\left(\frac{h_{\mathrm{PL}}^{2}}{\rho_{\mathrm{PL}}^{3} L_{\mathrm{PL}}}\right)_{\mathrm{S}} f_{\mathrm{S}}\left[\frac{3 \beta_{\mathrm{S}}}{2 E_{0}}\right]  \tag{9}\\
& \left\{\frac{\Delta f}{(\Delta d)^{2}}\right\}_{\mathrm{d}}=4\left(\frac{h_{P L}^{2}}{\rho_{\mathrm{PL}}^{3} L_{\mathrm{PL}}}\right)_{\mathrm{d}} f_{\mathrm{d}}\left[\frac{3 \beta_{\mathrm{d}}}{2 E_{0}}\right] \tag{10}
\end{align*}
$$

where the dislocation-free silicon is indicated by the subscript S and the silicon with dislocations, by the subscript d.

One can readily see that the right-hand sides of Eqs. (9) and (10) determine the slope of the experimental curves constructed in the $\Delta f$ versus $(\Delta d)^{2}$ coordinates, whereas their difference yields the value of the
dislocation contribution to the modulus measured:

$$
\begin{align*}
\Delta \beta= & \beta_{\mathrm{d}}-\beta_{\mathrm{S}}=\frac{E_{0}}{6 f_{\mathrm{d}}}\left(\frac{p_{\mathrm{PL}}^{3} L_{\mathrm{PL}}}{h_{\mathrm{PL}}^{2}}\right)_{\mathrm{d}}\left(\frac{\Delta f}{\Delta d^{2}}\right)_{\mathrm{d}}  \tag{11}\\
& -\frac{E_{0}}{6 f_{\mathrm{S}}}\left(\frac{p_{\mathrm{PL}}^{3} L_{\mathrm{PL}}}{h_{\mathrm{PL}}^{2}}\right)_{\mathrm{S}}\left(\frac{\Delta f}{\Delta d^{2}}\right)_{\mathrm{S}}
\end{align*}
$$

The contribution of dislocations to the corresponding modulus can be determined by comparing the $\Delta \beta$ values for the specimens with different dislocation densities $N_{\mathrm{d}}$. In this case, the effects associated with the free carrier contribution to the nonlinear modulus $\beta$ are excluded.

Typical results obtained in our study are shown in the figure. They indicate that, for the dislocation structure of $n$ - and $p$-type silicon, the anharmonicity is positive $(\beta>0)$. Therefore, the key factor here is the edge nature of the dislocations.

Thus, it has been established that the deformation modes used for both donor and acceptor silicon specimens provide the appearance mainly of the edge dislocations with the axes forming an angle $\theta \sim 90^{\circ}$ with the Burgers vectors.

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Translated by L. Man

# Electrostatic Mechanism of a Wavy Microrelief Formation on a Semiconductor Surface Sputtered by an Ion Beam 

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#### Abstract

It is shown that modeling of a physical mechanism of the formation of an ordered wavy microrelief on the semiconductor surface under the action of a high-energy ion beam can be considered as an electrostatic problem. © 2000 MAIK "Nauka/Interperiodica".


The development of various methods for creating ordered spatially periodic wavy structures with the characteristic wave length ranging within $\sim 0.1-1.0 \mu$ on the semiconductor surface is of a considerable interest for solving various problems of modern nanotechnology. These structures can be formed under the action of a powerful laser radiation [1-3] or due to sputtering of the semiconductor surface by ion beams with energies exceeding 1 eV [4-6].

Despite the visual similarity of the microreliefs obtained under the action of various factors on a material, the physical mechanisms of the microrelief formation under the action of a laser radiation and an ion beam are essentially different. In the first case, the relief is formed due to the surface melting of a solid, with a characteristic time of the microrelief formation typical of the liquid surface (ranging within $\sim 10^{-9}-10^{-11} \mathrm{~s}$ ). In the second case, a considerably lower power of the ion beam does not change the solid state of the semiconductor but makes it amorphous; therefore, the microrelief is gradually formed in the course of sputtering of the semiconductor surface over a longer time $\left(\sim 10^{2}-10^{9} \mathrm{~s}\right)$.

The effect of an electric field, which is induced by surface charging, was ignored in the first theoretical models of the formation of an ordered microrelief on the semiconductor surface irradiated with an ion beam [4-6], although it was well known that this effect played the decisive role in the microrelief formation under laser irradiation [1-3]. This field should also play an important role in the process of ion beam sputtering, if only because of the fact that the near-surface electric field decelerates the incident ions.

1. Let a homogeneous monoenergetic ion beam with the current density $\mathbf{j}_{0}$ be incident for a certain period of time onto a semiconductor plate at an angle $\alpha$ to the surface normal. The bottom surface of this plate is maintained at the zero potential (see figure). The incident high-energy ions penetrate the semiconductor to a depth $h$ and, thus, destroy the ordered crystal structure
and give rise to the formation of a large number of defects. Because of a high density of the ion flow, the subsurface layer of the thickness $h$ passes into a disordered amorphous state with the dielectric constant $\varepsilon_{1}$ and electric conductivity $\sigma_{1}$ lower than the corresponding characteristics $\varepsilon_{2}$ and $\sigma_{2}$ of the semiconductor in the initial state. The constant ion flow penetrating to the indicated depth (where the origin of the Cartesian system is placed, with the $z$-axis being normal to the free surface) charges both the free surface and the interface between the amorphous and crystalline layers. In the steady-state mode, the energy of ions penetrating the semiconductor is determined by their initial energy in the source (at the accelerator cut output) and the electric field intensity at the free surface.

Let us calculate the electric field strengths in the system considered. Above the free surface, we have $z=h+\xi(x, t)$ (the medium with subscript 0 ), inside the amorphized layer $0 \leq z \leq h+\xi$ (the medium with subscript 1), whereas inside the crystalline layer $-d \leq z \leq 0$ (the medium with subscript 2). A perturbation of the free surface $\xi=\xi(x, t)$ is associated with the formation of an amorphized viscoelastic layer on the free surface under the effect of a momentum flux (caused by the beam ions) and a wavy relief with the amplitude $\sim(k T / \gamma)^{1 / 2}$, where $k$ is the Boltzmann constant, $T$ is the absolute temperature, and $\gamma$ is the tension coefficient at the free surface [7-9].

In order to determine the electric field potentials and the currents flowing in the system, we have to solve the following problem:

$$
\begin{gather*}
\operatorname{div} \mathbf{E}_{m}=0, \quad \mathbf{E}_{m}=-\nabla \Phi_{m} ; \quad \operatorname{div} \mathbf{j}_{m}=0 ;  \tag{1}\\
\operatorname{rot} \mathbf{E}_{m}=0 ; \quad(m=0,1,2) ; \\
z=h+\xi: E_{0 n}-\varepsilon_{1} E_{1 n}=4 \pi \kappa_{h} ; \quad \Phi_{0}=\Phi_{1} ; \\
\frac{\partial \kappa_{h}}{\partial t}+\sigma_{1}\left(\mathbf{n} \cdot \mathbf{E}_{1}\right)+\operatorname{div}_{\Sigma}\left(\kappa_{h} \mathbf{u}_{\tau}+\kappa_{h} b_{h} E_{1 \tau} \mathbf{n}_{\tau}\right)=0 ; \tag{2}
\end{gather*}
$$

$$
\begin{gather*}
z=0: \varepsilon_{1} E_{1 n}-\varepsilon_{2} E_{2 n}=4 \pi \kappa_{0} \\
\Phi_{1}=\Phi_{2} ; \quad j_{0 n}=j_{1 n}-j_{2 n} \\
\frac{\partial \kappa_{0}}{\partial t}+\sigma_{1}\left(\mathbf{n} \cdot \mathbf{E}_{1}\right)+\sigma_{2}\left(\mathbf{n} \cdot \mathbf{E}_{2}\right)-j_{0 n} \mathbf{n}  \tag{3}\\
+\operatorname{div}_{\Sigma}\left(\kappa_{0} \mathbf{u}_{\tau}+\kappa_{0} b_{0} E_{2 \tau} \mathbf{n}_{\tau}\right)=0 \\
z=-d: \Phi_{2}=0  \tag{4}\\
\nabla_{\Sigma} \equiv \frac{\partial}{\partial x} \mathbf{n}_{x}+\frac{\partial}{\partial y} \mathbf{n}_{y} ; \quad \mathbf{j}_{m}=\sigma_{m}\left(\mathbf{n} \cdot \mathbf{E}_{m}\right)
\end{gather*}
$$

Here, $\Phi_{m}$ is the electric potential; $\kappa_{h}(x, t)$ and $\kappa_{0}(x, t)$ are the charge density at the free surface and at the interface, respectively; and $\mathbf{u}_{\tau}$ is the component of the velocity field of the medium motion tangential to the surface. Because of a considerable viscosity of the amorphized layer and in accordance with the experimental data [4-5], the latter factor is assumed to be insignificant and is neglected in our considerations.
2. Since the perturbation amplitude $\xi=\xi(x, t)$, of the free surface of the amorphized layer is rather small, deviations caused by this perturbation from the equilibrium values of the potentials, surface charge density, and current density in the incident beam would also be small. For definiteness, we set the perturbation $\xi$ in the form $\xi=\xi_{0} \exp (i k x)$, where $k$ is the wavenumber. Then, it is natural to search for the solution of the problem (1)-(4) in the form

$$
\begin{gather*}
\Phi_{0}=\Phi_{0}^{0}+C_{1} \exp (-k z) \exp (s t-i k x) \\
\Phi_{1}=\Phi_{1}^{0}+\left[C_{2} \exp (k z)+C_{3} \exp (-k z)\right] \exp (s t-i k x) \\
\Phi_{2}=\Phi_{2}^{0}+C_{4} \sinh [k(z+d)] \exp (s t-i k x) \\
\kappa_{h}=\kappa_{h}^{0}+C_{5} \exp (s t-i k x)  \tag{5}\\
\kappa_{0}=\kappa_{0}^{0}+C_{6} \exp (s t-i k x) \\
j_{0}=j_{0}^{0}+C_{7} \exp (s t-i k x)
\end{gather*}
$$

Here, the superscript 0 indicates the equilibrium values for the unperturbed free surface $z=h$. In writing a pos-


Schematic representation of the physical model of the interaction between an ion beam and a semiconductor surface.
sible solution for $\Phi_{2}$, we took into account the boundary condition (4).

Expanding the boundary conditions (2) in the vicinity of $z=h$ in the approximation linear with respect to $\xi$ and substituting the possible solutions (5) into the obtained relationships, we can determine the unknown constants $C_{m}$. In the zero approximation, we arrive at a self-consisted problem for determining the stationary fields and currents, the solution to which has the form

$$
\begin{gathered}
\kappa_{h}^{0}=\kappa_{0}^{0} ; \quad E_{0}^{0}=8 \pi \kappa_{h}^{0} ; \quad E_{1}^{0}=0 ; \quad E_{2}^{0}=8 \pi \varepsilon_{2}^{-1} \kappa_{0}^{0} ; \\
\kappa_{0}^{0}=\frac{j_{0} \varepsilon_{2} \cos \alpha}{8 \pi \sigma_{2}} ; \\
E_{0}^{0}=\frac{\varepsilon_{2}^{2} p^{2} q^{2} l \cos ^{2} \alpha}{\sigma_{2}^{2} m} \\
\times\left\{1-\left[1+\frac{4 \Delta \phi \sigma_{2}^{2} m}{\varepsilon_{2}^{2} p^{2} q^{2} l^{2} \cos ^{2} \alpha}\right]^{1 / 2}\right\}
\end{gathered}
$$

where $p, q$, and $m$ are the concentration, the charge, and the ion mass in the beam, respectively; $q \Delta \phi$ is the initial ion energy; and $l$ is the path traveled by an ion along the normal at the free surface in the field $E_{0}^{0}$.

In order to determine corrections of the first-order smallness, we have to solve an inhomogeneous system of seven algebraic equations with respect to the unknown coefficients $C_{m}$. This can readily be done on a computer with the use of one of the well known packages of programs for analytical computationsMATHCAD or MATHEMATICA. However, the solutions thus obtained are rather cumbersome. For this reason, we restrict the consideration to a brief analysis of the solution for $C_{7}$, the most important coefficient in the context of this article.
3. The expression for $C_{7}$ is complex. Being represented in the exponential form, it determines the amplitude of the correction to $j_{0}^{0}$ and the addition to the correction phase. Equating the imaginary part of $C_{7}$ to zero, we arrive at an algebraic equation with respect to $s$, the exponent of the rate of the exponential change in the correction amplitude with time. The solution of the latter equation is rather simple (provided that the mobilities of the charge carriers at the free surface and at the interface are equal, $b_{h}=b_{0}=b$ ):

$$
\begin{align*}
& s^{2}\left\{2 \varepsilon_{2}\left(\varepsilon_{2} \sigma_{1}-\varepsilon_{1} \sigma_{2}\right)[1+\exp (2 k h)] \cosh ^{2}(d k)\right. \\
& -2 \varepsilon_{1} \sigma_{1}\left[1+\exp (2 k h)-\varepsilon_{1}+\varepsilon_{1} \exp (2 k h)\right] \sinh ^{2}(d k) \\
& -\varepsilon_{1}\left[-\varepsilon_{2} \sigma_{1}(1+\exp (2 k h))+\sigma_{2}(-1+\exp (2 k h)\right. \\
& \left.\left.\left.\quad+2 \varepsilon_{1}+2 \varepsilon_{1} \exp (2 k h)\right) \sinh (2 d k)\right]\right\} \tag{6}
\end{align*}
$$

$$
\begin{aligned}
& +s\left\{-8 \pi \varepsilon_{1} \sigma_{1} \sinh (d k)\left[\sigma_{2} \cosh (d k)(1+\exp (2 k h))\right.\right. \\
& \left.\left.\quad+\sigma_{1} \sinh (d k)(-1+\exp (2 k h))\right]\right\} \\
& +32 \pi^{2} b^{2} \kappa^{2} \sigma_{1}(-1+\exp (2 k h)) \sinh ^{2}(d k)=0
\end{aligned}
$$

The above equation has two real roots-positive and negative. The negative root corresponds to the temporal exponential attenuation of the correction of the first order of smallness. The positive root corresponds to exponential increase of the correction, that is, of the contribution of the term $\sim \exp [s t-i(k x+\beta)]$ (where $\beta$ is the phase) to the incident beam current density with time. In the close vicinity of the positive root of (6), we have $\beta \approx \pm \pi$. This implies that the spatial variable of the component of the ion beam incident onto the semiconductor surface is in counterphase with the initial perturbation of the free surface $\xi=\xi_{0} \exp (i k x)$, so that the dips of the wavy surface relief correspond to the maxima of the ion concentration in the beam. As a result, the amplitude of the wavy relief on the semiconductor surface increases with time because of a more intense sputtering at the dips [4-5].
4. Conclusion. With neglect of the hydrodynamic motion, the possible mechanism of the formation of an ordered wavy relief on the semiconductor surface dur-
ing its sputtering by a high-energy ion beam can be considered as a purely electrostatic process.

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Translated by L. Man

# Dielectric Spectra of Sodium Niobate-Based Binary Systems 

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#### Abstract

Dielectric properties of the $\mathrm{Na}_{1-x} \mathrm{Li}_{x} \mathrm{NbO}_{3}, \mathrm{Na}_{1-x} \mathrm{~K}_{x} \mathrm{NbO}_{3}$, and $\mathrm{Na}_{1-x} \mathrm{~Pb}_{x} \mathrm{Nb}_{1-x} \mathrm{Ti}_{x} \mathrm{O}_{3}$ systems have been studied in the frequency range from 120 Hz to 20 kHz . A considerable dispersion of the dielectric constant in the antiferroelectric solid solutions is established, which indicates the possible formation of ferroelectric clusters in the antiferroelectric matrix. When the probing field frequency decreases in the range of low $x$ values, the temperature curves of the dielectric constant acquire some relaxation extrema. It is suggested that the observed effects are caused by oxygen vacancies. © 2000 MAIK "Nauka/Interperiodica".


The compositional disorder and various types of point and extended defects inherent in polycrystalline specimens result in a certain diffusion of the phase transitions [1]. In some cases, these spatially inhomogeneous media can exhibit states characteristic of glasslike systems (phases of the dipole glass type) [2]. The dipole glass states are often observed in the systems of solid solutions containing regions where the antiferro-electric-ferroelectric phase transitions take place. One of the criteria of the transition of a system into the dipole glass state is the dispersion in the dielectric constant in the vicinity of the phase transition temperature, whereby the maxima of the temperature curves of the real ( $\varepsilon^{\prime}$ ) and imaginary ( $\varepsilon^{\prime \prime}$ ) parts of the dielectric constant shift toward higher temperatures with an increase of the probing field frequency.

The present study was undertaken in order to reveal the possible dipole glass properties in the vicinity of the antiferroelectric-ferroelectric phase transitions in the $\mathrm{NaNbO}_{3}$-based binary systems (at low concentrations of the second components) by methods used in the physics of dielectrics.

The ceramic specimens of the $\mathrm{Na}_{x} \mathrm{Li}_{1-x} \mathrm{NbO}_{3}(0 \leq$ $x \leq 0.145), \mathrm{Na}_{x} \mathrm{~K}_{1-x} \mathrm{NbO}_{3}(0 \leq x \leq 0.1)$, and $\mathrm{Na}_{x} \mathrm{~Pb}_{1-x} \mathrm{Nb}_{x} \mathrm{Ti}_{1-x} \mathrm{O}_{3}(0 \leq x \leq 0.2)$ systems were obtained by solid-phase synthesis with subsequent hot pressing. The measurements were made on $1 \times 10 \mathrm{~mm}$ disks with fused silver electrodes. Using an E8-2 alter-nating-voltage bridge in the frequency range from 120 Hz to 20 kHz , we measured the temperature dependences of $\varepsilon^{\prime}$ and the dielectric loss tangent $\tan \delta$.

It was revealed experimentally for the $\mathrm{Na}_{x} \mathrm{Li}_{1-x} \mathrm{NbO}_{3}$ system in the composition range $0<x \leq 0.0075$, for the $\mathrm{Na}_{1-x} \mathrm{~K}_{x} \mathrm{NbO}_{3}$ system in the range $0<x \leq 0.0125$, and for the $\mathrm{Na}_{x} \mathrm{~Pb}_{1-x} \mathrm{Nb}_{x} \mathrm{Ti}_{1-x} \mathrm{O}_{3}$ system in the range $0<x \leq$ 0.005 that, the temperature corresponding to the maxi-
mum dielectric constant depends on the probing field frequency. In pure $\mathrm{NaNbO}_{3}$ and in compounds of all the other compositions, no maxima of the dielectric constant were observed on the $\varepsilon(T)$ curves at any frequencies.

The $\varepsilon^{\prime}(T)$ curves obtained for some compositions lying within the above concentration ranges of the binary systems are shown in Fig. 1. Note that a pronounced dispersion in $\varepsilon$ takes place for the above antiferroelectric compositions, which is not typical of the antiferroelectric compounds [3]. This fact can be explained by the formation of ferroelectric clusters in the antiferroelectric matrix of the specimens studied. This assumption is confirmed by the existence of the ferroelectric hysteresis loop $[4,5]$ similar to that observed earlier in pure sodium niobate.

With a decrease in the probing field frequency, some relaxation extrema appear on the $\varepsilon^{\prime}(T)$ curves. The $\tan \delta(T)$ dependences at the frequencies within 120130 Hz are slightly nonmonotonic at the temperatures corresponding to the main phase transition (i.e., the transition observed at a frequency of 20 kHz in the temperature range from 300 to 800 K ). The additional extrema on the $\varepsilon^{\prime}(T)$ curves for the $(\mathrm{Na}, \mathrm{Pb})(\mathrm{Nb}, \mathrm{Ti}) \mathrm{O}_{3}$ system appear at $f=600 \mathrm{~Hz}$; in the $\left(\mathrm{Na}, \mathrm{Li}^{2}\right) \mathrm{NbO}_{3}$ and $(\mathrm{Na}, \mathrm{K}) \mathrm{NbO}_{3}$ systems, such extrema are observed at $f=$ 400 Hz .

It should be emphasized that no variation of the temperature corresponding to the maximum dielectric constant with frequency is observed for the first system in the stable antiferroelectric region, whereas in two other systems this takes place in the vicinity of the antiferro-electric-ferroelectric phase transition.

Previously, the appearance of additional maxima with a decrease of the probing field frequency was reported in sodium bismuth titanate and sodium niobate single crystals [6]. This phenomenon was explained by


Fig. 1. Temperature dependences of the dielectric constant of sodium niobate-based binary systems measured at various frequencies: (a) $\mathrm{Na}_{0.9975} \mathrm{Li}_{0.0025}$; (b) $\mathrm{Na}_{0.9975} \mathrm{~K}_{0.0025} \mathrm{NbO}_{3}$; (c) $\mathrm{Na}_{0.998} \mathrm{~Pb}_{0.002} \mathrm{Nb}_{0.998} \mathrm{Ti}_{0.002} \mathrm{O}_{3}$. Dashed lines show the data for annealed specimens.
the existence of oxygen vacancies. In order to verify this result, we annealed our ceramic specimens for $10-15 \mathrm{~h}$ in an oxidizing atmosphere at $T=900 \mathrm{~K}$. Upon annealing, only one $\varepsilon$ maximum remained, which corresponded to the main phase transition. The specimen conductivity considerably decreased, whereas the $\tan \delta(T)$ curve became monotonic. Therefore, the additional extrema on the $\varepsilon^{\prime}(T)$ curves in our experiments can also be caused by oxygen vacancies.

Thus, the sodium niobate-based ceramic systems studied in our experiments showed no dipole glass properties. Our data may support the opinion that the $x-T$ phase diagrams of $(\mathrm{Na}, \mathrm{Pb})(\mathrm{Nb}, \mathrm{Ti}) \mathrm{O}_{3},(\mathrm{Na}, \mathrm{Li}) \mathrm{NbO}_{3}$, and $(\mathrm{Na}, \mathrm{K}) \mathrm{NbO}_{3}$ solid solutions have regions of antiferroelectric phases, which can initially contain ferroelectric clusters. This is indicated both by a strong dispersion of $\varepsilon$ in the antiferroelectric compounds and by the manifestations of irreversibility in the form of the ferroelectric hysteresis loops. In turn, the strong mesoscopic inhomogeneity (structure clusterization) enhances the influence of the defect state (oxygen vacancies) on the dielectric properties of these objects, which is seen from the appearance of additional extrema on the $\varepsilon(T)$ curves.

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# Probability Symmetry Broken upon Rapid Period-Doubling Bifurcation 

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#### Abstract

A quadratic one-dimensional mapping with noise is used to model the phenomenon of violation of the equal probability of variants in the postbifurcation system evolution upon a fast change of the control parameter. The case of a period-doubling bifurcation, after which only one of the two types of oscillations with different phases may occur in the system, is analyzed. The laws of the phenomenon are considered and its possible mechanism is discussed. © 2000 MAIK "Nauka/Interperiodica".


1. Dealing with the nonlinear world, one frequently encounters bifurcation situations, whereby the initial state (motion) of a system loses stability in response to a change in the control parameter, replaced by two equiprobable variants of the subsequent existence and development pathways. In particular, a period-doubling bifurcation leads to the appearance of a cycle with a double period (in the vicinity of the cycle in which the system has lost stability), with two equiprobable variants of the subsequent motion differing only in phase (i.e., in the timeshift per period of the unstable motion). Then, the question naturally arises as to which of the two variants of the postbifurcation development will be realized in the system provided that the control parameter $r$ varies at a certain finite rate $S$ in the presence of a noise with the variance $\sigma^{2}$. The interest in answering this question is related both to studying fundamental problems in physics and biology (see, e.g., reviews [1, 2]) and to solving numerous practical tasks.

In the case where the control parameter varies very slowly (adiabatically) after the period-doubling bifurcation, the system should occur in the vicinity of the cycle where stability was lost and where selection of the subsequent state is determined by the noise. If the noise probability distribution is symmetric, the final states of the system are equiprobable as well: $p_{1}=p_{2}=1 / 2$. In the opposite limit of infinitesimal noise ( $\sigma^{2} \longrightarrow 0$ ), either the first $\left(p_{1}=1, p_{2}=0\right)$ or the second $\left(p_{1}=0, p_{2}=1\right)$ final state is realized with certainty, depending on the initial conditions and irrespective of the rate $S$, which corresponds to completely predictable behavior. These opposite limiting cases of the bifurcation transition are called the "stochastic" and "dynamic" variants, respectively [3, 4].

Should the control parameter vary at a sufficiently high rate and the noise be present, an intermediate case is realized, whereby the final states can only be predicted at a probability lower than unity and the proba-
bility distribution is no longer symmetric $\left(p_{1} \neq p_{2}\right)$. In this case, it is a common practice to establish a conditional boundary between the stochastic and dynamic variants of the bifurcation transition, based on the criterion of attaining a certain final state at a preset probability (e.g., $p_{1}=0.75 ; p_{2}=0.25$ ). Butkovskiĭ et al. [3] studied, using numerical methods, the properties of a logistic mapping of several types [see Eqs. (2) below] with additive noise. It was established that the noise level and the rate of the control parameter variation corresponding to this boundary are related by a universal power law:

$$
\begin{equation*}
\sigma_{\mathrm{c}}^{2}=C S^{\alpha} \tag{1}
\end{equation*}
$$

Here, the exponent $\alpha$ and the coefficient $C$ depend on the initial conditions, $\alpha$ being maximum when the initial coordinate $x_{0}$ is set at the boundaries of the "attraction domain" of the final state [4]. ${ }^{1}$

The aim of this study was to elucidate a mechanism responsible for the violation of the final state probability symmetry in systems subject to a period-doubling bifurcation upon rapid change in the control parameter, to determine the role of the universal relationship (1), and to refine the concept of a fast transition. For this purpose, we have considered a greater number of models as compared to that used in the previous studies and gained a markedly greater volume of statistical data.
2. A classical model system with period-doubling bifurcations is offered by a one-dimensional mapping of the type $x_{n+1}=f\left(x_{n}\right)$, where $f\left(x_{n}\right)$ is a function with quadratic maximum, $x$ is a dynamic variable, and $n=0$,

[^1]

Fig. 1. The shapes of various mapping functions for different values of the control parameters: (a) mapping (2); (b) mapping (4); (c) mapping (6); (d) the structure of the attraction domains of states 1 (thin portions of the abscissa axis) and 2 (thick segments) for mapping (2) and its second iteration $x_{n+2}=f\left(f\left(x_{n}\right)\right)$.
$1,2, \ldots$ is the discrete time. We have studied five mapping functions of this type:

$$
\begin{gather*}
x_{n+1}=r x_{n}\left(1-x_{n}\right),  \tag{2}\\
x_{n+1}=r-x_{n}^{2},  \tag{3}\\
x_{n+1}=-\left(x_{n} / r\right)\left(r+x_{n}\right),  \tag{4}\\
x_{n+1}=r\left(1-r^{3} x_{n}^{2}\right),  \tag{5}\\
x_{n+1}=1-r x_{n}^{2} \tag{6}
\end{gather*}
$$

These functions, which can be converted one into another by the corresponding substitution of variables, differ by the character of dependence of their stationary solutions $x^{*}$ (equilibrium point, cycle of period 1) on the parameter $r$. In Fig. 1, these solutions are represented by intersections with the diagonal $x_{n}=x_{n+1} .{ }^{2}$ The mapping function (4) has a zero derivative ( $\gamma=$ $d x^{*} / d r=0$ ), which implies that point $x^{*}$ does not change its position in the one-dimensional phase space (Fig. 1b). For the other functions, the derivatives are nonzero $(\gamma \neq 0)$ and may differ in both magnitude and sign. In this case, the points $x^{*}$ change their positions depending on the parameter (Figs 1a and 1c).

[^2]The control parameter was varied as described by a piecewise linear function:

$$
r(n)=\left\{\begin{array}{l}
r_{1}+S n, \quad n \leq N,  \tag{7}\\
r_{1}+S N=r_{2}, \quad n>N^{\prime},
\end{array}\right.
$$

where $r_{1}$ and $r_{2}$ are the initial and final values of the control parameter and $S$ is the parameter variation rate (determined as $S=\Delta r / N$, where $\Delta r=r_{2}-r_{1}$ is the change in the control parameter and $N$ is the number of iterations corresponding to this change). The bifurcation (critical) value of the parameter $r_{\mathrm{c}}$, for which the multiplier is $\left|\left(d x_{n+1} / d x_{n}\right)\right|_{x^{*}}=1$, falls within the interval $r_{1}<r_{\mathrm{c}}<r_{2}$. At $r=r_{\mathrm{c}}$, the point (state) $x^{*}$ loses stability and a cycle of period 2 appears in the vicinity of this point. In Fig. 1d showing mapping (2) and the corresponding second iteration $x_{n+2}=f\left(f\left(x_{n}\right)\right)$, the doubleperiod cycle corresponds to the equilibrium points 1 and 2. For a dynamic system with $x_{n+2}=f\left(f\left(x_{n}\right)\right)$, these points appear as attractors with the attraction basins indicated by thick solid segments on the abscissa axis. For system (2), the different parts of the abscissa axis represent the "attraction domains" of the final states.

The noise was introduced into the mapping functions in the form of a small additive $x_{n} \longrightarrow x_{n}+\xi_{n}$, where $\xi_{n}$ is a random sequence with a distribution law close to the normal (Gaussian) with a zero mean value. In the calculations, the initial condition was taken in the


Fig. 2. Behavior of the system modeled by mapping (2): (a) a logarithmic plot of the critical noise variance versus the control parameter variation rate for a large $\Delta r$; (b) logarithmic plots of the critical noise variance versus the control parameter variation rate for an odd $N$ and various $\Delta r$; dashed line shows analytical estimates calculated by formula (8); (c) the plots of ( $x_{N}+\xi_{N}$ ) and $x^{*}$ for $r=r_{2}$ versus the number of iteration steps at a fixed $\Delta r$ value.
form $x_{0}=x^{*}\left(r_{1}\right)$ (Figs. 1a-1c) and determined analytically using Eqs. (2)-(6). Then the iteration procedure was conducted including a number of steps $(n \gg N)$ that were sufficiently large for the transient processes to decay, after which the form of the final state was determined. Using the results of 250 numerical experiments, the probabilities $p_{1,2}$ for a given double-period state to be attained were calculated as a function of the noise variance $\sigma^{2}$ for various values of $S$. The smoothened $p_{1,2}(\ln S)$ plots were used to determine the critical noise level $\sigma_{\mathrm{c}}^{2}$ for which the probabilities $p_{1,2}$ were equal to the preset values 0.75 and 0.25 . By these estimates, the plots of $\ln \left(\sigma_{\mathrm{c}}^{2}\right)$ versus $\ln S$ were constructed.
3. An analysis of the results of our numerical experiments leads to the following conclusions:
(a) In the general case (arbitrary $\Delta r$ ), dependence of the critical noise dispersion $\sigma_{\mathrm{c}}^{2}$ on the parameter variation rate $S$ differs from the power law and is determined by the type of mapping. This is illustrated by Fig. 2a showing that the plot of $\ln \left(\sigma_{c}^{2}\right)$ versus $\ln S$ exhibits two
branches corresponding to odd and even $N$, none of which can be approximated by a linear function. Moreover, this plots depends on the $\Delta r$ value as is seen from Fig. 2b (presenting the plots for odd $N$ ), where various $\Delta r$ at the same $S$ correspond to different $\sigma_{\mathrm{c}}^{2}$.
(b) The power relationship (1) is valid only for small changes of the control parameter $\Delta r$, which is seen in Fig. 2b showing that portions of the plots with large $k$ values can be approximated by straight lines. A large $k$ corresponds to small $\Delta r_{k}=2^{-3(k-1)} \Delta r_{1}\left(\Delta r_{1}\right.$ was taken to be equal to a difference of the control parameter values for the first and second period-doubling bifurcations). Note that the points corresponding to small $N(N=1$ on the right-hand ends of the plots and $N=200$ on their left-hand ends) also fit virtually to the same straight line.
(c) The above features and the very fact of violation of the final state probability symmetry in the systems studied is related to a shift of the "center of gravity" of the distribution function at the end of the fast transition (for $\left\langle x_{N}+\xi_{N}\right\rangle$, where angular brackets denote the ensemble-average) with respect to the boundary
between the attraction domains of different final states. These boundaries are formed by unstable equilibrium points $x^{*}$ and their images as illustrated in Fig. 1d (for details, see [5]). Over the interval $n=(0, N)$ (corresponding to $r$ changing from $r_{1}$ to $r_{2}$ ), the $x^{*}$ value varies monotonically with time, while $x_{n}$ exhibits oscillations with a double quasi-period. Fluctuations, which are suppressed in the first stage of the transition (for $r<r_{c}$ ), grow in the second stage. Accordingly, the dispersion $\left\langle\left(\left(x_{N}+\xi_{N}\right)-\left\langle x_{N}+\xi_{N}\right\rangle\right)^{2}\right\rangle$, which is not large for small $N$ values, increases with $N$ and the fluctuation component eventually becomes dominating. As is seen in Fig. 2c, there is a certain $N_{1}$ below which the quantity ( $x_{N}+\xi_{N}$ ) on each subsequent iteration step falls on different sides of the boundary $x^{*}$. This makes the different final states alternatively more probable (which results in separation of the points corresponding to odd and even $N$ values in the right-hand part of Fig. 2a). For $N>N_{1}$, where the oscillating $\left(x_{N}+\xi_{N}\right)$ values fall on one side of the boundary, the parity of $N$ no longer affects the $\sigma_{\mathrm{c}}^{2}$ versus $S$ curve. As the $N$ value increases further, the development of fluctuations in the second stage of the transition increases the random character of the process, as reflected by the leveling of the probability of different final states.

The shift of $\left\langle x_{N}+\xi_{N}\right\rangle$ from boundary toward the domain of attraction of one of the possible final states (making this state more probable) depends both on the type of mapping and on the fast transition parameters. In particular, for mapping (4) with the derivative $\gamma=0$, the position of the boundary $x^{*}$ between the attraction domains of the final states is independent of $r$; for $x_{0}=$ $x^{*}$, the value of $\left\langle x_{N}+\xi_{N}\right\rangle=x^{*}$ also remains constant. Here, the symmetrically distributed noise equiprobably "drives" the system toward both possible evolution pathways for any $N$. In the general case of a mapping with $\gamma \neq 0$, including mappings of the types (2), (3), (5), and (6), the shift $\left\langle x_{N}+\xi_{N}\right\rangle$ is nonzero and the final state probability symmetry is broken.
(d) The dashed line in Fig. 2b shows estimates of the $\sigma_{\mathrm{c}}^{2}$ values for various $S$ obtained in the region of small $\Delta r$ assuming that $x^{*}(r)$ is a linear function of $r$ (i.e., $\gamma=$ const). For $N=1$, the shift is $\left\langle x_{N}+\xi_{N}\right\rangle-x^{*}=\Delta x=S \gamma$.

Then the probability of a given final state for a normal (Gaussian) noise with a zero mean is

$$
\begin{equation*}
p_{1}=\int_{-\infty}^{\Delta x} \rho(\xi) d \xi=\frac{1}{2}\left[1+\operatorname{erf}\left(\frac{S \gamma}{\sigma \sqrt{2}}\right)\right], \tag{8}
\end{equation*}
$$

where $\rho(\xi)$ is the noise probability density function. The error function for the preset critical value $p_{1}=0.75$ (used to determine the final variant) is $\operatorname{erf}\left(S \gamma /\left(\sigma_{\mathrm{c}} 2^{1 / 2}\right)\right)=$ $1 / 2$, which corresponds to $S \gamma /\left(\sigma_{c} 2^{1 / 2}\right) \approx 0.477$. As seen in Fig. 2b, the slopes of the calculated plots of $\ln \left(\sigma_{\mathrm{c}}^{2}\right)$ versus $\ln S$ corresponding to small $\Delta r$ values, as well as the variation of the positions of points corresponding to $N=1$, agree with the line of estimates.
4. The phenomenon of violation of the final state probability symmetry upon the fast period-doubling bifurcation in the discrete models studied is determined by the boundaries of the attraction domains of different final states, which move in the phase space in response to variation of the control parameter. The levels of noise corresponding to the conditional boundary separating the "stochastic" and "dynamic" variants of the bifurcation transition are determined by properties of the model mapping. In the general case, the power relationship between the noise level and the control parameter variation rate predicted in [3] is valid only in a small vicinity of the critical (bifurcation) value.

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# Influence of the Surface on the Impurity Solubility in Small-Size Crystals 

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#### Abstract

The surface effects on the process of impurity dissolution in semiconductor crystals has been analyzed. Computations performed for the suggested model showed that the surface effects are essential for spherical crystals with a radius smaller than thirteen lattice periods (i.e., for nanocrystals). It has been established that the melting points and the limiting impurity solubilities of impurities decrease with the crystal size. © 2000 MAIK "Nauka/Interperiodica".


Recently, particular attention has been given to the study of various nanostructures for which the influence of the surface on the impurity solubility is especially important.

Under equilibrium conditions, the impurity concentration in a crystal corresponds to the minimum free energy of the system. A system consisting of a semiconductor crystal of finite dimensions and an external phase (ambient medium) is considered here by the method of the minimum free energy, which is applicable to the systems that exchange neither particles nor energy with the ambient medium.

Free energy of a crystal with due regard for the surface energy. We assume that the system under consideration consists of atoms of only two kinds, $A$ and $B$, with the $A$ atoms being dissolved in a crystal consisting of $B$ atoms. For definiteness, we also assume that the $A$ atoms can be located only in the lattice sites (if necessary, this model can be extended to include the impurity atoms located in interstitials). We use the following notation: $N^{B}$ is the number of lattice sites in the crystal, $N_{A}^{B}$ is the number of $A$ atoms in the crystal, $N_{B}^{B}$ is the number of $B$ atoms in the crystal, and $N_{A}^{E}, N_{B}^{E}$ is the number of $A$ and $B$ atoms in the external phase. The free energy of this system can be written as

$$
\begin{gather*}
G=G^{E}\left(N_{A}^{E}, N_{A}^{E}\right)+g_{A}^{B} N_{A}^{B} \\
+\left(g_{A}^{B} N_{A}^{B}-G^{S}\left(N_{B}^{B}\right)\right)-k T \ln \frac{N^{B}!}{N_{A}^{B}!N_{B}^{B}!}, \tag{1}
\end{gather*}
$$

where $g_{A}^{B}$ and $g_{B}^{B}$ are the free energies of the $A$ and $B$ atoms in the crystal lattice, $G^{E}\left(N_{A}^{E}, N_{A}^{E}\right)$ is the free energy of the external phase, and $G^{S}\left(N_{B}^{B}\right)$ is the surface energy.

When minimizing the free energy, one has to take into account the conservation laws for particles of each kind and the coupling equations for the number of sites in the crystal

$$
\begin{align*}
& \varphi_{A} \equiv N_{A}-N_{A}^{B}-N_{A}^{E}=0, \\
& \varphi_{B} \equiv N_{B}-N_{B}^{B}-N_{B}^{E}=0,  \tag{2}\\
& \varphi^{B} \equiv N^{B}-N_{A}^{B}-N_{B}^{B}=0 .
\end{align*}
$$

The first two equations in set (2) express the law of conservation of the particle number in the closed system. The third expression follows from the law of conservation of the number of lattice sites.

The minimum of the free energy given by Eq. (1) with due regard for conditions (2) can be determined by the method of Lagrange multipliers. The thermodynamic equilibrium of the system corresponds to the minimum of the following functional:

$$
\begin{equation*}
\Psi=G+\lambda_{A} \varphi_{A}+\lambda_{B} \varphi_{B}+\lambda^{B} \varphi^{B} \tag{3}
\end{equation*}
$$

where $\lambda_{A}, \lambda_{B}$, and $\lambda^{B}$ are the Lagrange multipliers. Minimizing functional (3) with respect to the variables $N_{A}^{E}, N_{B}^{E}, N_{A}^{B}, N_{B}^{B}$, and $N^{B}$, we arrive at the system of equations

$$
\left\{\begin{array}{l}
\mu_{A}^{E}-\lambda_{A}=0, \\
\mu_{B}^{E}-\lambda_{B}=0, \\
g_{A}^{B}+k T \ln N_{A}^{B}-\lambda^{A}-\lambda_{B}=0,  \tag{4}\\
g_{B}^{B}+k T \ln N_{B}^{B}-\lambda^{B}-\lambda_{B}-\frac{\partial G^{S}\left(N_{B}^{B}\right)}{\partial N_{B}^{B}}=0, \\
\lambda^{B}-k T \ln N^{B}=0 .
\end{array}\right.
$$

Here, we used the following definition of the chemical potential:

$$
\begin{equation*}
\mu_{A}^{E}=\frac{\partial G^{E}}{\partial N_{A}^{E}}, \quad \mu_{B}^{E}=\frac{\partial G^{E}}{\partial N_{B}^{E}} . \tag{5}
\end{equation*}
$$

The chemical potential of atoms in the external phase can be represented by a standard expression

$$
\begin{align*}
\mu_{A}^{E} & =\mu_{0 A}^{E}+k T \ln a_{A}^{E}, \\
\mu_{B}^{E} & =\mu_{0 B}^{E}+k T \ln a_{B}^{E}, \tag{6}
\end{align*}
$$

where $a_{A}^{E}$ and $a_{B}^{E}$ are the activities of the $A$ and $B$ atoms in the external phase. Such a representation of the chemical potential leads to the following solution of system (4):

$$
\begin{align*}
N_{A}^{B} & =N^{B} a_{A} \exp \left\{-\frac{g_{A}^{B}-\mu_{0 A}^{E}}{k T}\right\}=N^{B} a_{A} \exp \left\{-\frac{\Delta g_{A}^{B}}{k T}\right\}, \\
N_{B}^{B} & =N^{B} a_{B} \exp \left\{-\frac{g_{B}^{B}-\mu_{0 B}^{E}}{k T}\right\} \exp \left\{\frac{1}{k T} \frac{\partial G^{S}\left(N_{B}^{B}\right)}{\partial N_{B}^{B}}\right\}(7  \tag{7}\\
& =N^{B} a_{B} \exp \left\{-\frac{\Delta G_{B}}{k T}\right\} \exp \left\{\frac{1}{k T} \frac{\partial G^{S}\left(N_{B}^{B}\right)}{\partial N_{B}^{B}}\right\},
\end{align*}
$$

where $\Delta g_{A}^{B}=g_{A}^{B}-\mu_{0 A}^{E}$ is the change in the free energy of an $A$ atom from the external phase during its incorporation into the crystal and $\Delta G_{B}^{\text {melt }}=g_{B}^{B}-\mu_{0 B}^{E}$ is the free energy of melting of the crystal consisting of $B$ atoms.

In the approximation of $N_{B}^{B} \approx N^{B}$, the second expression in (7) yields the activity of $B$ atoms with due regard for the effect of the surface energy:

$$
\begin{equation*}
a_{B}=\exp \left\{\frac{\Delta G_{\text {melt }}^{B}}{k T}\right\} \exp \left\{-\frac{1}{k T} \frac{\partial G^{S}\left(N_{B}^{B}\right)}{\partial N_{B}^{B}}\right\} . \tag{8}
\end{equation*}
$$

Since the activities of the solution components are interrelated, the surface effects would influence the impurity activity, and, thus, the impurity solubility. As will be shown below, the smaller the crystal size, the more pronounced the influence of the surface effects. Consider the influence of the surface on the solubility of an impurity for a spherical crystal.

Impurity solubility in spherical crystals (quantum dots). We assume that the surface effects are caused by the presence of dangling bonds at the interface with the crystal matrix. Then the surface energy can be written as

$$
\begin{equation*}
G^{S}=N_{B}^{S} \Delta H_{B}^{S}, \tag{9}
\end{equation*}
$$

where $N_{B}^{S}$ is the number of the dangling bonds on the surface and $\Delta H_{B}^{S}=\frac{1}{4} \Delta H_{B}^{\text {melt }}$ is the enthalpy of dangling bonds [1].

Using simple geometric considerations, we arrive at the expression

$$
\begin{equation*}
S=4 \pi R^{2}=a^{2} N_{B}^{2}, \quad V=\frac{4 \pi}{3} R^{3}=N_{B}^{B} a^{3}, \tag{10}
\end{equation*}
$$

where $S$ is the surface area, $V$ the volume, $a$, the lattice parameter, and $R$, the radius of the crystal. It follows from Eq. (10) that $N_{B}^{S}=4 \pi\left(N_{B}^{B}\right)^{2 / 3}$ and, therefore, the surface energy can be written as

$$
\begin{equation*}
G^{S}=\sigma\left(N_{B}^{B}\right)^{2 / 3} \tag{11}
\end{equation*}
$$

where $\sigma=\pi \Delta H_{B}^{\text {melt }}$.
Substituting formula (8) for the activity into Eq. (11), one can obtain the following analytical expressions for the liquidus and solidus curves:

$$
\begin{gather*}
X_{B}=\frac{N_{B}^{E}}{N_{B}^{E}+N_{A}^{E}}=\frac{a_{B}}{\gamma_{B}} \\
=\frac{1}{\gamma_{B}} \exp \left\{\frac{\Delta G_{B}^{\mathrm{melt}}}{k T}\right\} \exp \left\{-\frac{2 \sigma\left(N_{B}^{B}\right)^{-1 / 3}}{3 k T}\right\}, \\
X_{A}=\frac{N_{A}^{E}}{N_{B}^{E}+N_{A}^{E}}=\frac{a_{A}}{\gamma_{A}}  \tag{12}\\
=\frac{1}{\gamma_{A}}\left(1-\frac{1}{\gamma_{B}} \exp \left\{\frac{\Delta G_{B}^{\text {melt }}}{k T}\right\} \exp \left\{-\frac{2 \sigma\left(N_{B}^{B}-\right)^{-1 / 3}}{3 k T}\right\}\right), \\
N_{A}^{B}=N^{B} \gamma_{A}\left(1-\frac{1}{\gamma_{B}} \exp \left\{\frac{\Delta G_{B}^{\mathrm{melt}}}{k T}\right\}\right. \\
\left.\times \exp \left\{-\frac{2 \sigma\left(N_{B}^{B}\right)^{-1 / 3}}{3 k T}\right\}\right) \exp \left\{-\frac{\Delta g_{A}^{B}}{k T}\right\},
\end{gather*}
$$

where $\gamma_{A}$ and $\gamma_{B}$ are the activity coefficients describing the interactions of particles in the external phase. Following the model of a quasi-chemical interaction, the activity coefficients can be expressed as [2]

$$
\begin{align*}
\ln \gamma_{B} & =\Omega \frac{\left(1-X_{B}\right)^{2}}{k T}  \tag{13}\\
\ln \gamma_{A} & =\Omega \frac{\left(X_{B}\right)^{2}}{k T} .
\end{align*}
$$

Using Eq. (12), we modeled the liquidus curve in the Si-Sn system well described with $\Omega=0.254 \mathrm{eV}$. With due regard for this representation of the activity


Solidus curves calculated for the $\mathrm{Si}-\mathrm{Sn}$ system featuring crystals of different size $R=10 a$ (1), 20a (2), (3) $\infty$. Symbols indicate the experimental data [2].
coefficient, we managed to determine the solubility curve for tin in silicon and the parameters of tin defect formation:

$$
\begin{gather*}
\Delta g_{\mathrm{Sn}}^{\mathrm{Si}}=\Delta h_{\mathrm{Sn}}^{\mathrm{Si}}-T \Delta s_{\mathrm{Sn}}^{\mathrm{Si}}, \\
\Delta h_{\mathrm{Sn}}^{\mathrm{Si}}=0.25 \mathrm{eV}, \quad \Delta s_{\mathrm{Sn}}^{\mathrm{Si}}=-4.6 k . \tag{14}
\end{gather*}
$$

Using the parameters (14), we calculated the process of impurity dissolution in small spherical crystals (see figure). It is seen that the impurity solubility
decreases with the crystal radius and, moreover, the melting point of the crystal decreases as well.

The computations show that the surface produces a strong effect if the surface energy exceeds one tenth of the enthalpy of melting:

$$
\begin{equation*}
0.1 \Delta H_{B}^{\mathrm{melt}} \leq \frac{2 \sigma}{2\left(N_{B}^{B}\right)^{1 / 3}} . \tag{15}
\end{equation*}
$$

Using expressions (10) and (15), one can determine the characteristic dimension above which the surface effects would disappear:

$$
\begin{equation*}
R_{c}=a\left(\frac{3}{4 \pi}\right)^{1 / 3} \frac{20 \sigma}{3 H_{B}^{\text {melt }}}=4.14 a \frac{\sigma}{H_{B}^{\text {melt }}} . \tag{16}
\end{equation*}
$$

Proceeding from the assumptions used in the calculation of the surface energy of a crystal (11), we obtain that the characteristic size below which the surface effects become essential equals $R_{c} \approx 13 a\left(N_{B}^{B} \approx 9000\right.$ ), this value being independent of the material of the crystal.

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Translated by L. Man

# The Effect of Condensation on the Parameters of Pulsed Supersonic Streams 

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#### Abstract

We report on the results of mass-spectrometric measurements conducted in the pulsed supersonic streams of argon, nitrogen, and helium. Evolution of the time parameters of a stream determined as functions of the retarding pressure $P_{0}$ is compared to the pattern of variation of the mass peak intensities corresponding to monomers and clusters. The transition of a gas flow into the condensation regime is accompanied by changes in the flight time and the halfwidth of the monomer signal measured in the molecular beam formed from the supersonic stream. It is demonstrated that the monitoring of the time parameters of a pulsed stream allows different condensation stages to be distinguished. © 2000 MAIK "Nauka/Interperiodica".


The efflux of a gas flow into vacuum may be accompanied by the formation of clusters, the properties of which depend on the flow retardation parameters, gas composition, and processes occurring in the stream. Actively participating in the energy exchange, clusters may also affect the gas-phase, chemical, and plasmachemical processes in the streams. Therefore, investigation of the phenomenon of condensation in gas streams and the effect of this process upon the gas dynamics is an important task.

The purpose of this work was to study the effect of condensation on the time characteristics of the pulsed streams of argon, nitrogen, and helium. The experiments were performed using a set of gasodynamic setups of the LEMPUS type designed and constructed at Novosibirsk State University [1]. The pulsed gas streams in a wide range of retarding pressures were produced using a device combining an electromagnetic valve and a supersonic nozzle with an edge diameter of $d_{*}=1 \mathrm{~mm}$. An $0.53-\mathrm{mm}$-diam skimmer was used to cut a molecular beam that was fed into a detector of an MS-7303 quadrupole mass spectrometer modified so as to perform measurements in a pulsed mode. The mass spectrometer instrumentation allowed changes in the shape of each pulse to be monitored for any mass peak within the mass range studied. The time was measured from the instant when an $0.5-\mathrm{ms}$-long starting pulse came to the electromagnetic valve. The pulse repetition rate was 5 Hz , which provided optimum conditions for properly evacuating the vacuum system. The experimental data were obtained for a nozzle-skimmer spacing of $x / d_{*}=175$ and 80 at a retarding temperature of $T_{0}=295 \mathrm{~K}$.

Previously [2], we demonstrated that evolution of the mass peak intensities of Ar monomers and clusters measured in the pulsed gas stream agrees with the
behavior observed in the stationary streams. By monitoring the time sweep of the mass spectrometer signal, we determined the time of arrival $T$ of the leading front of the pulse and the pulse halfwidth $D$. Figure 1 shows the plots of $T$ and $D$ versus retarding pressure $P_{0}$ for monomers and small clusters in argon and nitrogen streams. At small $P_{0}$ values, a difference in the pulse halfwidth and shape for monomers are insignificant, while a shift in the time of arrival of the leading pulse


Fig. 1.


Fig. 2.
front is caused only by the difference in the Ar and $\mathrm{N}_{2}$ efflux velocities. When small clusters appeared in the stream (for $P_{0}>40$ and 200 kPa for argon and nitrogen, respectively), the halfwidth of the monomer signal sharply increased. At the same time, no changes were observed in the time of arrival of the leading pulse front due to additional stream acceleration caused by the condensation heat evolution [3].

At pressures corresponding to the gas flow transition into a regime featuring the formation of large clusters (i.e., at $P_{0}>90$ and 500 kPa for argon and nitrogen, respectively) [3], we observed a retardation of the leading pulse front for monomers, while no significant changes took place for the pulses of cluster components. For each particular gas, these effects depend of the retarding pressure and are virtually independent of the Knudsen number of the skimmer [4] at the point of molecular beam cutting. This is confirmed by comparative data presented in Fig. 1 for Ar beams measured in systems with different nozzle-skimmer spacings ( $x / d=80$ and 175 for black and gray circles, respectively). No sharp changes in the $T$ and $D$ values were observed for helium (open circles) in the entire range of
$P_{0}$, since helium exhibits no condensation in this range of parameters. Factors responsible for a small synchronous decrease in $T$ (accompanied by a smooth increase in $D$ ) with growing $P_{0}$ prior to the onset of condensation in $\mathrm{Ar}, \mathrm{N}_{2}$, and He are still unclear. These factors are probably related to features of the gasodynamic process involved in the formation of pulsed flows. A weak increase in the $T$ value in helium is apparently due to the stream retardation at high $P_{0}$ in a gas pillow formed in front of the skimmer.

Evolution of the time parameters of the gas pulses, especially of their halfwidths, is correlated with the change in intensity of the peaks of monomers and small clusters measured at the molecular beam axis, as illustrated by the data for Ar and $\mathrm{N}_{2}$ in Fig. 2. A sharp growth in the monomer pulse halfwidth (circles) begins at the same values of $P_{0}$ for which the drop in the peak intensity is observed that is attributed to the onset of cluster formation in the stream. A significant delay in the time of arrival of the leading front of the monomer pulse (triangles) is observed for the $P_{0}$ values corresponding to the secondary increase in intensity attributed to the formation of large clusters in the stream and their fragmentation caused by the impact of probing electrons in the mass spectrometer detector.

Thus, evolution of the time parameters, pulse halfwidth, and time of the leading front arrival measured by pulsed molecular-beam mass spectrometry may provide additional information on the process of cluster formation in pulsed streams of gases and gas mixtures.

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# Multichannel Signal Detection in a Multimode Optical-Fiber Interferometer: Reducing the Effect of Spurious Signals 

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#### Abstract

Detection of a useful signal against spurious ones in a multimode optical-fiber interferometer is studied theoretically and experimentally. The theoretical analysis is based on a probabilistic approach. The computation and the experiment demonstrate that the multichannel technique can significantly reduce the probability of wrong signal detection as compared to the case of a single-channel photodiode scheme. © 2000 MAIK "Nauka/Interperiodica".


This paper continues the analysis of the operation of a multimode optical-fiber interferometer (MFI), presented in [1,2]. We here demonstrate some advantages of multichannel detection as a practical means to counter spurious signals.

MFIs usually serve as spatially-distributed measuring systems affected by various external factors, which usually include both useful and spurious (noninformative) signals. We think that the latter factors produce the most harmful noise (or, more properly, interference) in real MFIs operating as sensors of physical quantities. Experiments show that other types of noise in the instrument, namely, those from a laser source or a photodetector, are weaker. We therefore ignore them here.

On the whole, behavior of the MFI signals, both useful and spurious ones, is studied in sufficient detail as evidenced by literature on the so-called modal noise in the optical-fiber transmission lines [3, 4] and in the data transfer systems using intermodal interference (i.e., differential phase modulation of modes) [5, 6]. In addition, our work [1] looked more closely at the formation and behavior of different components of the MFI signal from the standpoint of sensor systems.

Let a multimode fiber of length $L$ be illuminated by a coherent light source at a frequency of $\omega$. Then the interference of various modes gives rise to a speckle pattern at the output of the fiber. Let $S$ denote the speckle area that is covered by the optical detector. According to [2], the corresponding light intensity can be expressed as

$$
\begin{equation*}
I(L, S)=I_{0}\left(1+\rho_{0}+\rho_{1}\right) \tag{1}
\end{equation*}
$$

where

$$
I_{0}=\frac{1}{2} \sum_{i=1}^{N} A_{i}^{2} \int_{S} E_{i}^{2} d s
$$

is a constant (zero-frequency) component,

$$
\rho_{0}=\frac{1}{I_{0}} \sum_{\substack{i=1 \\ i \neq k}}^{N} \sum_{k=1}^{N} A_{i} A_{k}^{*}\left\{\int_{S} E_{i} E_{k}^{*} d s\right\} \cos \psi_{i k}^{0}
$$

is a slowly varying component, and

$$
\rho_{1}=\frac{1}{I_{0}} \sum_{\substack{i=1 \\ i \neq k}}^{N} \sum_{\substack{ \\i \neq 1}} A_{i} A_{k}^{*}\left\{\int_{S} E_{i} E_{k}^{*} d s\right\} \sin \psi_{i k}^{0} \delta \Psi_{i k}
$$

is a useful signal component.
The intermodal phaseshift angles $\psi_{i k}$, which are implicitly involved in expression (1), can be expanded as $\psi_{i k}=\psi_{i k}^{0}+\delta \psi_{i k}$. The term $\psi_{i k}^{0}$ is a quasi-stationary portion of the phaseshift related to the modal parameters and the ambient conditions such as temperature, pressure, etc. The term $\delta \psi_{i k}$ refers to rapidly varying actions on the fiber, which are usually small ( $\delta \psi_{i k} \ll 1$ ).

Let us focus on the term $\rho_{1}$, since it provides information on the external factors and can easily be extracted from signal (1) by filtering in the operating frequency band.

We assume that $\delta \psi_{i k}$ is related to (i) a useful signal that is to be measured, and (ii) spurious perturbations acting in the operating frequency band. Thus, $\delta \psi_{i k}=$ $\delta \psi_{i k}^{S}+\delta \psi_{i k}^{n}$.

Let $S$ denote the magnitude of the useful signal and $n$ denote the total magnitude of the spurious action. Then the $\rho_{1}$ component of the MFI signal can be written as a sum of two terms:

$$
\begin{equation*}
\rho_{1}=K_{S} S+K_{n} n=\rho_{S}+\rho_{n}, \tag{2}
\end{equation*}
$$

where the coefficients $K_{S}$ and $K_{n}$ are expressed as

$$
\begin{align*}
& K_{S}=\frac{1}{I_{0}} \sum_{\substack{i=1 \\
i \neq k}}^{N} \sum_{\substack{ }}^{N}\left(A_{i} A_{k}^{*}\left\{\int_{S} E_{i} E_{k}^{*} d s\right\} \sin \psi_{i k}^{0} K_{i k}^{S}\right), \\
& K_{n}=\frac{1}{I_{0}} \sum_{\substack{i=1 \\
i \neq k}}^{N} \sum_{k}^{N}\left(A_{i} A_{k}^{*}\left\{\int_{S} E_{i} E_{k}^{*} d s\right\} \sin \psi_{i k}^{0} K_{i k}^{n}\right) . \tag{3}
\end{align*}
$$

Note that formulas (3) include the phaseshift sensitivity coefficients with respect to the useful signal and spurious factors: $K_{i k}^{S}=\left(\delta \psi_{i k}^{S} / S\right)$ and $K_{i k}^{n}=\left(\delta \psi_{i k}^{n} / n\right)$. Our previous study [1] of a multimode fiber exposed to mechanical disturbances that, with due regard for mode coupling, $K_{i k}^{S}$ and $K_{i k}^{n}$ depend on the mode numbers $i$ and $k$ so that $K_{S}$ and $K_{n}$ must vary with external conditions in an uncorrelated fashion. Consequently, both $\rho_{S}$ and $\rho_{n}$ components in the MFI output signal experience independent fading effects. This circumstance seriously hinders the detection of a useful signal against spurious ones. To solve the problem, we suggest using multichannel detection, whereby the radiation from the fiber is divided between $M$ photodiodes and the magnitudes of their output signals are summed up [2].

To assess potential advantages of multichannel detection, we introduce the following detection criterion: if the magnitude of the output signal (in the operating frequency band) is above a given threshold $X_{0}$, then the action on the fiber is considered as relevant.

Let us treat the problem in probabilistic terms [2]. Time variations in $\Psi_{i k}^{0}$ subject to external conditions depend on changes in the fiber parameters and also relate to the mode coupling and birefringence processes. In practice, these variations are unpredictable at any instant and therefore can be treated as random variables. According to the central limiting theorem, with a sufficiently large number of the summands in formulas (3), the values of $K_{S}$ and $K_{n}$ at a given instant are independent Gaussian random variables with a zero mean and a certain variance $\sigma_{K}^{2}$. We assume that $S$ is a deterministic (regular) variable and the value of $n$ at the instant of measurement is a zero-mean Gaussian random variable with the variance $\sigma_{n}^{2}$. Then $\rho_{S}$ in Eq. (2) is also a Gaussian random variable with the rms deviation $\sigma=S \sigma_{K}$.

To make the considerations simple and illustrative, let us assume that $S$ and $n$ have the same dimension.

This allows us to introduce the parameter $A^{2}=S^{2} / \sigma_{n}^{2}$, which has a meaning of the MFI signal-to-interference ratio. Otherwise, the relative levels of the useful signal and the interference would be represented by a complicated parameter, but the results would be generally similar. Thus, $\rho_{n}$ is the product of two independent Gaussian random variables ( $n$ and $K_{n}$ ), while $\sigma=S \sigma_{K}=$ $A \sigma_{n} \sigma_{K}$.

The above implies that, if $S=0$ (only a spurious signal is present), the magnitude of the output MFI signal $x^{(0)}$ equals the modulus of the spurious component $x^{(0)}=\left|\rho_{n}\right|$. If $x^{(0)}$ becomes larger than $X_{0}$, a false operation of the system takes place: a spike in the spurious signal is confused with an informative signal. With a nonzero $S$, we have $x^{(S)}=\left|\rho_{S}+\rho_{n}\right|$. Here, a wrong decision that results in missing a useful signal can be made if $x^{(S)}$ is smaller than $X_{0}$. The four possible outcomes are summarized as follows:

$$
\left\{\begin{array}{l}
S=0: x^{(0)}=\left|\rho_{n}\right|=\left|K_{n} n\right|, \\
x^{(0)}<X_{0}, \text { correct decision; } \\
x^{(0)}>X_{0}, \text { wrong decision; }  \tag{4}\\
S \neq 0: x^{(S)}=\left|\rho_{S}+\rho_{n}\right|=\left|\rho_{S}+K_{n} n\right|, \\
x^{(S)}>X_{0}, \text { correct decision; } \\
x^{(S)}<X_{0}, \text { wrong decision. }
\end{array}\right.
$$

For the multichannel detection with summing signals from $M$ photodiodes, the situation is much the same except that the MFI signals are more complicated combinations of random variables:

$$
\left\{\begin{array}{l}
S=0: x^{(0)}=\sum_{i=1}^{M}\left|K_{n}^{i} n\right|,  \tag{5}\\
x^{(0)}<X_{0}, \text { correct decision; } \\
x^{(0)}>X_{0}, \text { wrong decision; } \\
S \neq 0: x^{(S)}=\sum_{i=1}^{M}\left|\rho_{S}^{i}+K_{n}^{i} n\right|, \\
x^{(S)}>X_{0}, \text { correct decision; } \\
x^{(S)}<X_{0}, \text { wrong decision }
\end{array}\right.
$$

Here, $\rho_{S}^{i}, K_{n}^{i}$, and $n$ are independent zero-mean Gaussian random variables with rms deviations $\sigma, \sigma_{K}$, and $\sigma_{n}=\sigma /\left(A \sigma_{K}\right)$, respectively. (We assume that the photodiodes are equivalent in terms of the aperture conditions and the magnitude of intensity oscillation.)

The paragraphs below deal with estimation of the probability of wrong detection (PWD) and with the ability of reducing this error by the multichannel tech-



Fig. 1. Normalized histograms of the MFI signal computed for $A=5$ ( $M$ denotes the number of the photodetector channels).
nique. Solving the former problem reduces to finding the probability density functions (PDFs) of "signals" (4) and (5), selecting an optimum threshold $X_{0}$, and evaluating the corresponding integrals over the regions $x^{(S)}<X_{0}$ and $x^{(0)}>X_{0}$. The PDFs were found by computer-aided simulation, since it would be laborious to derive them from analytical expressions for $M \sim 10$. (The analytical expressions involve convolutions and are similar to those used in [2].) A special application program was employed to generate independent zero-mean Gaussian random variables, including ten variables $\rho_{S}^{i}$ with the variance $\sigma^{2}=1$, ten variables $K_{n}^{i}$ with the variance $\sigma_{K}^{2}=1$, and the variable $n$ with the variance $\sigma_{n}^{2}=1 / A^{2}$ (i.e., we assumed $S=1$ ). The computations were performed for $10^{5}$ values of each variable. For every set of variables, the $x^{(0)}$ and $x^{(S)}$ values were computed using expressions (4) for $M=1,2,4,6,8$, or 10 , and their histograms were constructed, the range of values being divided into 500 intervals. Upon normalization, these histograms represented the desired PDFs. Figure 1 gives an example of the normalized histograms of $x^{(0)}$ and $x^{(S)}$ for $M=1$ or 6 and the signal-to-interference ratio $A=5$.

It is expedient to select the operation threshold $X_{0}$ in a way similar to the criterion of ideal observer in a multichannel discrete communication system with parallel channels [7, 8]. Specifically, we set $X_{0}$ according to the intersection point of the respective PDFs for $x^{(0)}$ and $x^{(S)}$ (obtained from histograms). In the case under consideration, this value minimizes the total PWD determined as the sum of the probability of missing a useful signal $\left(x^{(S)}<X_{0}\right)$ and the probability of false operation $\left(x^{(0)}>X_{0}\right)$. For each $M$ and $A$, we evaluated $X_{0}$ from histograms and then computed the areas under the normalized histograms so as to estimate the PWD. Figure 2 shows the PWD as a function of the number of channels
$M$ for different values of the signal-to-interference ratio $A$.

The simulation results were verified in an experiment using a setup including a $\mathrm{He}-\mathrm{Ne}$ laser $(\lambda=0.63 \mu \mathrm{~m}$, $P=2 \mu \mathrm{~W}$ ) and a multimode graded-index fiber with a length of 480 m . The output radiation from the fiber was fed to a ten-channel optical detector designed as a linear photodiode array. The output signals of the detector circuit were digitized and processed by a computer. A reference signal was also transmitted to the computer so as to specify instants when a useful signal was applied. A $5-\mathrm{m}$ portion of the fiber was heated in the $20-40^{\circ} \mathrm{C}$ range, and another small segment ( $\sim 10 \mathrm{~m}$ ) was periodically subjected to weak mechanical disturbances, simulating a useful signal action, with the help of an electromagnetic device. No special sources of noise were used; produced solely by the natural laboratory environment, spurious signals were easy to detect if channel gains were adjusted appropriately.

The MFI signals were detected in the presence of slow heating and short-term mechanical disturbances


Fig. 2. The graphs of PWD vs. the number of channels obtained from the results of (1) simulation and (2) measurements. (The scatter is due to taking into account all possible combinations of $M$ photodiodes for $M \neq 10$.)

## (a)



Fig. 3. Fragments of time records showing (a) four photodiode outputs and (b) a result of the sum-of-magnitudes processing with $M=10$. The broken lines depict an optimum detection threshold.
with a pulse length of about 0.5 s and a pulse repetition period of 10 s . Figure 3a shows fragments of the output time records for four photodiodes. One can easily see the characteristic features of the useful and spurious signals, namely, slow amplitude fluctuations (fading) in both of them and the absence of correlation between the two random processes, whether they are in the same channel or not. The computer performed the sum-ofmagnitudes processing for two, four, six, eight, or ten channels. A fragment of a resultant signal for ten channels is shown in Fig. 3b.

Based on the time records, we determined the sample sets of MFI signal values $x^{(S)}$ and $x^{(0)}$ obtained with and without the useful signal action, respectively. [A rough estimate of $x^{(0)}$ was obtained from the peak value of the MFI signal for a 0.5 -s interval immediately before the useful signal action.] From these data, a value of $X_{0}$ was selected for each combination of $M$ channels. (In Fig. 3, the threshold is indicated by the horizontal lines.) Finally, the PWD was estimated from the corresponding numbers of the $x^{(0)}$ and $x^{(S)}$ values that are above or below $X_{0}$.

Figure 2 shows the PWD values estimated from the experimental data. Note that the signal-to-interference ratio determined from the photodetector output signals was between 4 and 5, the operating frequency range
being $1-100 \mathrm{~Hz}$. The ratio was evaluated from opticalreceiver outputs.

Figure 2 clearly demonstrates that single-channel MFI signal detection is characterized by a very high PWD even at an appreciable signal-to-interference ratio. This is hardly surprising, since the useful and spurious signals at the MFI output depend on the coefficients subject to independent and uncorrelated fading [see expression (2)]. The effect of MFI fading can be reduced by multichannel detection with the sum-ofmagnitudes processing [2]. This technique essentially increases a "distance" between the spurious component in the absence of the informative action and the MFI signal in the presence of such action, thus decreasing the PWD. For example, the PWD is lowered by more than one order in magnitude for $M=10$. It is seen that the experimental results agree well with the theoretical estimates, showing a pronounced advantage of the multichannel detection.

In conclusion, we have demonstrated some advantages of the multichannel detection in an MFI sensor in the case of the simplest (threshold) detection criterion. We believe that this approach furnished with a more efficient criterion could be instrumental in solving some serious problems with MFI and increasing performance of the fiber-based sensor devices, thus making them much more practical.

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# Electric Current and Heat Induced Acoustic Emission in Cadmium Sulfide 

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#### Abstract

Acoustic emission (AE) induced by the electric current passing through a cadmium sulfide (CdS) single crystal was studied. In the temperature range $T=300-450 \mathrm{~K}$, the intensity of the AE signals excited in the CdS crystal exposed to a constant electric field increases with the current density. It is suggested that AE in CdS is due to the dislocations breaking off and moving under the action of direct electric current and thermoelastic stresses. The activation energy for this process ( $E_{\mathrm{a}}=0.35 \pm 0.5 \mathrm{eV}$ ) was estimated for a current density in the range of $j=(1-7) \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}$. © 2000 MAIK "Nauka/Interperiodica".


As is well known, the external mechanical, electrical, or thermal factors acting upon dislocated crystals favor an increase in the mobility of linear defects [1-4]. This behavior is also typical of semiconductor structures operating in strong electric and thermal fields. Acting on the crystals containing linear defects, the above factors cause the generation of sound waves and pulses (acoustic emission) related to the motion of dislocations, crack formation, and fracture of the material $[5,6]$.

Despite this phenomenon being widespread, the acoustic emission (AE) in semiconductors under the action of permanent external factors is still insufficiently studied. In particular, virtually no data were reported on the AE in dislocated semiconductors exposed to stationary thermal and electric fields. Therefore, the purpose of our work was to study the AE induced by electric current and heat in dislocated crystals of cadmium sulfide (CdS).

The initial material was obtained by growing CdS single crystals using the Davydov-Markov technique [7] on a [0001]-oriented sapphire substrate. The singlecrystal CdS ingots were cut into $50-\mathrm{mm}$-diam disks with a resistivity of $20 \Omega \mathrm{~cm}$ and an average growth dislocation density of $10^{5} \mathrm{~cm}^{-2}$. The disks were polished and subjected to a conventional physicochemical treatment [7]. Finally, the dislocated samples in the form of $25 \times 10 \times 2 \mathrm{~mm}$ plates for the AE experiments were prepared from these disks by the scribing technique.

The AE signals generated in a CdS sample were detected by a piezoelectric transducer lying on the sample plate surface. The electric response $U(t)$ of the transducer (see the inset in Fig. 1) was displayed on an S9-8 digital storage oscillograph and transmitted via a common bus channel to a computer for the determination of the spectral composition $U(\omega)$ of the AE signal.

This analysis was performed using a fast Fourier transform (FFT) algorithm (Fig. 1, curve 1 ) implementing the Welch periodogram technique [8].

The FFT curves were smoothened using the energy minimization method [9, 10]. For this purpose, an experimental data array $\tilde{X}$ was represented as a sum of the true spectrum $\bar{X}$ and the noise component $Y$, after which a regularization process was conducted based on the functional $F(\bar{X})[9,10]$

$$
\begin{equation*}
F(\bar{X})=A(\bar{X}, \tilde{X})+\lambda B(\bar{X}), \tag{1}
\end{equation*}
$$

where

$$
A(\bar{X}, \tilde{X})=\frac{1}{2} \sum_{i=1}^{n-1}\left[\bar{X}_{i+1}-\bar{X}_{i}\right]^{2}
$$

is the difference functional and

$$
B(\bar{X})=\chi^{2}=\sum_{i=1}^{n} \frac{\left(\tilde{X}_{i}-\bar{X}_{i}\right)^{2}}{\sigma_{i}^{2}}
$$

is the smoothening functional characterizing deviation of the true spectrum from the experimental $\lambda$-parametric regularization. In the latter term, $\sigma_{i}$ is the meansquare deviation of $\tilde{X}$ and $n$ is the number of experimental points. In the first step, the numerical solution $\bar{X}$ was taken equal to $\tilde{X}$ at a certain value of the parameter $\lambda$. Then the functional (1) was minimized and the corresponding $\bar{X}$ and $\chi^{2}$ distributions were calculated. The calculation procedure was terminated when the $\chi^{2}$ value obtained in the sequential calculation steps decreased below $10^{-4}$. Should this level not be attained, the regularization parameter was taken equal


Fig. 1. Spectral density of the AE signal power measured in the course of annealing of dislocated $\left(10^{5} \mathrm{~cm}^{-2}\right) \mathrm{CdS}$ crystal plates at a current density of $j=5 \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}$ and $T=303 \mathrm{~K}$ : (1) Fourier transform; (2) smoothened curve. The inset shows a typical oscillogram of the $U(t)$ signal measured during the annealing.
to $\lambda=\lambda \frac{\chi^{2}}{n}$. Figures 1 (curve 2 ) and 2 (curves $1-4$ ) show the AE spectra (smoothened as described above) measured in the course of the electrothermal annealing of dislocated CdS crystals.

The AE processes were studied on CdS samples placed into an electric furnace and heated to reach a certain stationary temperature (the temperature was established within 3.5-4 h upon switching on the heaters). The AE spectra measured (at fixed temperatures $T$ ) in the course of heating were used to evaluate the AE signal energy $W \sim \sum_{n} U_{n}^{2}$ ( $U_{n}$ are the amplitudes of harmonics in the experimental spectrum). It was found that (similar to results reported for silicon [4]), the process of establishing a stationary temperature in the absence of a direct electric current $(j=0)$ in the sample was accompanied by a monotonic decrease in the AE response intensity. The stationary $U(t)$ value was attained $1-1.5 \mathrm{~h}$ after attaining the state of thermal equilibrium (Fig. 2, curve 5).

A direct electric current with a density of $j=(1-7) \times$ $10^{5} \mathrm{~A} / \mathrm{m}^{2}$ passing through a sample led to a sharp increase in the AE response amplitude, which remained constant even after keeping the sample at a fixed temperature for 36 h . Figure 2 shows a typical AE response from a semiconductor crystal with dislocations, with a clearly pronounced maximum at $f \approx 0.2 \mathrm{~Hz}$. This pattern may be related only to dislocations breaking off
and moving under the action of a constant electric field, in agreement with the observations reported in [3].

According to [3, 11], the average frequency of the dislocation transitions

$$
\begin{equation*}
f=f_{0} \exp \left(-\frac{W_{p}-F(j) b}{k T}\right) \tag{2}
\end{equation*}
$$

is determined by the temperature and by the electrostimulated force $F(j)$ acting upon dislocations; $f_{0}$ is the


Fig. 2. AE spectra of dislocated CdS crystal plates annealed under various conditions: ( 1 ) $j=5 \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}, T=303 \mathrm{~K}$; (2) $j=5 \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}, T=333 \mathrm{~K}$; (3) $j=5 \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}, T=$ $363 \mathrm{~K} ;(4) j=7 \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}, T=343 \mathrm{~K} ;(5) j=0, T=363 \mathrm{~K}$.


Fig. 3. (1-3) Temperature dependence of the AE signal intensity and (4) AE activation energy spectrum for a dislocated CdS crystal (dislocation density, $10^{5} \mathrm{~cm}^{-2}$ ), measured at a current density of $j=5 \times 10^{5} \mathrm{~A} / \mathrm{m}^{2}$ : (1) $f=2.2 \mathrm{~Hz}, E_{\mathrm{a}}=$ 0.1 eV ; (2) $f=1.8 \mathrm{~Hz}, E_{\mathrm{a}}=0.19 \mathrm{eV}$; (3) $f=1.5 \mathrm{~Hz}, E_{\mathrm{a}}=$ 0.28 eV ; (4) $E_{\mathrm{a}}=0.35 \mathrm{eV}$.
preexponential factor (determining the frequency of dislocation transitions at $T \longrightarrow \infty), W_{p}$ is the Peierls barrier, and $b$ is the Burgers vector.

In the absence of mechanical stresses, the dislocations are subject to pinning on impurities and other defects. Under the action of a constant electric field, the segments of dislocations exhibit bending depending on the magnitude and sign of the effective charge. The extent of bending increases until the break-off stress is reached:

$$
\begin{equation*}
\tau_{k}=\frac{2 E_{0}}{\pi b^{2} L_{\mathrm{s}}} \tag{3}
\end{equation*}
$$

which depends on the energy $E_{0}$ of dislocation binding to the defects. Therefore, dislocations or their segments can travel by-passing defects only provided that there are fluctuations assisted by the electric field. Upon changing position, dislocations are pinned again in the new quasiequilibrium state and occur there waiting for the following thermal fluctuations and mechanical actions stimulated by the electric field. According to this model, the dislocation motion proceeds in the field of Peierls forces determined by the corresponding potential barrier:

$$
\begin{equation*}
W_{p}=\frac{G b^{2}}{\pi(1-v)} \exp \left(-\frac{4 \pi d}{2 b(1-v)}\right) \tag{4}
\end{equation*}
$$

where $G=1.5 \times 10^{11} \mathrm{~Pa}$ is the shear modulus, $v=0.3$ is the Poisson coefficient, $b=1.1 \mathrm{~nm}$ is the Burgers vector, and $d=0.3 \mathrm{~nm}$ is the interplanar spacing in the crystal lattice. Substituting these numerical values into
expression (4), we may estimate the Peierls potential barrier for cadmium sulfide: $W_{p} \sim 0.5 \mathrm{eV}$.

Increasing current discharge is accompanied by the growing rate of the dislocation transitions and, hence, leads to a shortwave shift of the spectrum along the frequency axis (Fig. 2, curve 4). Increasing the intensity of thermal action leads to a growth in the number of dislocations surmounting the potential barrier.

By measuring the temperature dependence of the AE signal intensity for crystal samples containing growth dislocations, we may estimate the activation energy for the process of current-assisted dislocation transition over the barrier. It is expected that dislocation segments of various lengths have to overcome different barriers: for the same nature of dislocation segment pinning centers, the break-off probability must increase with the length of bent segments.

As is known [11], the dislocation segments of various length $L$ obey an exponential probability distribution

$$
\begin{equation*}
P=1-\left[1-\left(1+\frac{L-L_{\mathrm{s}}}{L_{n}}\right) \exp \left(-\frac{L-L_{\mathrm{s}}}{L_{n}}\right)\right]^{n} \tag{5}
\end{equation*}
$$

In this relationship, describing the spectral composition of the AE signal, $L_{\mathrm{s}}$ is the dislocation segment length determined by pinning on the neighboring defect and impurity centers, $L_{n}$ is the dislocation loop length, and $n=\left(L_{n} / L_{\mathrm{s}}\right)-1$. According to Eq. (5), a maximum probability corresponds to the break-off of the longest segments of a linear defect; this results in further elongation of the defect and leads to the gradual break-off of the whole dislocation [11]. Therefore, the activation barrier must decrease with increasing length of dislocation segments. By processing the experimental temperature dependences (Fig. 2, curves 1-3), it is possible to determine the activation energy values for various segments from their frequency characteristics. The experimental points determined for different frequencies $f$ (Fig. 2) fit well to the same straight line in the coordinates of $\ln U$ versus $1 / T$ (Fig. 3, curves 1-3). Curve 4 in Fig. 3 shows the results of the numerical calculation by Eq. (4). As seen, $E_{\text {a }}$ increases with the dislocation segment length $L$ to reach 0.5 eV for the extremum of $U(f)$ (Fig. 2), thus showing good agreement between experiment and calculations according to Eq. (4).

As for a decrease in $E_{\text {a }}$ predicted in the range of $0-0.5 \mathrm{~Hz}$, this feature may be related to a difference in the nature of pinning centers for various dislocation segments [11].

Thus, the acoustic emission observed in dislocated crystal plates of cadmium sulfide is related to the phenomenon of dislocations breaking off and surmounting energy barriers under the action of stationary thermal fields $(T \approx 400 \mathrm{~K})$ and electric currents $(j=1-7 \times$ $\left.10^{5} \mathrm{~A} / \mathrm{m}^{2}\right)$. Switching off the electric current $(F(j) \longrightarrow 0)$ decreases the probability of surmounting the barrier by means of thermal fluctuations, which results in the
complete vanishing of the AE signal upon attaining a stationary state.

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[^0]:    ${ }^{1}$ Here, the concentration anharmonicity is understood as the contribution of free charge carriers to the nonlinear elasticity modulus $\beta$.

[^1]:    ${ }^{1}$ By the "attraction domain" we imply a part of the attraction basin of the double-period cycle, from which the system falls into one of the final states with different phases.

[^2]:    ${ }^{2}$ The other point of intersection with the diagonal corresponds to an unstable solution $x^{* *}$; this point separates the domains of finite and infinite solutions.

