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The Decaying Second Order Isothermal Reaction

in a Weak Turbulence

by

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Abstract

A study has been made of the asymptotic behavior of a dynamically passive scalar when it is undergoing decay according to a second order reaction, and when it is being simultaneously convected by a weak, homogeneous turbulence. If $\bar{\Gamma}(t)$ is the mean concentration and $\bar{\Gamma}^2(t)$ the mean square fluctuation in concentration then the following asymptotic behavior is found:

$$\bar{\Gamma}(t) \sim t^{-1}, \quad \bar{\Gamma}^2(t) \sim t^{-\frac{1}{2}}$$

By an analysis of the non-diffusive limit it is shown that even the simplest closure scheme - the zero third moment approximation - may give useful decay estimates for relatively large initial scalar field fluctuations.

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Introduction

Study of the mixing of passive scalar fields by turbulence was initiated by Corrsin¹ and Obukhoff². Subsequently various facets of it have been pursued. In particular, an extension of considerable interest was introduced by Corrsin³ in his study of the statistics of an isothermal reacting mixture in isotropic turbulence and he was able to obtain estimates of the rate of decay of mean concentration and concentration fluctuations for certain limiting cases of both a first and a second order reaction.

This note is concerned wholly with a decaying, second-order, isothermal reaction in the limit of a weak, homogeneous turbulence. We first deduce explicit asymptotic decay rates for both the mean and the fluctuations and then show evidence that the zero third moment approximation, which is generally of very limited value in turbulence dynamics except asymptotically, is a useful closure technique even non-asymptotically when applied to the reaction field. Useful, that is, in predicting the decay of the mean and the fluctuations. It of course cannot represent spectral behavior adequately.

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1 A. N. Obukhoff, *Izvest, Akad. Nauk. S. S. R. Ser. Geograf. Geofiz.* 13, 58-69 (1949) (Translated by U. S. Army Chemical Corps, Camp Detrich).

2 S. Corrsin, *J. Aero. Sci.* 18, 417-423 (1951).

3 S. Corrsin, *Phys. Fluids* 1 42-47, (1958).

The Final Period of Decay

The material conservation equation for a reactant undergoing a second order isothermal reaction³ is

$$\frac{\partial \Gamma}{\partial t} + u_i \frac{\partial \Gamma}{\partial x_i} = D \nabla^2 \Gamma - C \Gamma^2 \quad (1)$$

where $\Gamma(x, t)$ is a random function of position and time, $u_i(x, t)$ is a turbulent velocity field whose behavior, by reason of the isothermal assumption, is uncoupled from that of the reactant, D is the diffusivity and C is the reaction rate.

Following Corrsin³ by defining $\bar{\Gamma}(t)$ as the mean and $\gamma(x, t)$ as the random fluctuation in concentration the following relationship holds;

$$\frac{d \bar{\Gamma}}{d t} = -C (\bar{\Gamma}^2 + \bar{\gamma}^2) \quad (2)$$

Also a correlation equation which makes explicit use of the homogeneity of the velocity and scalar fields is easily derived to be

$$\left(\frac{\partial}{\partial t} - 2D \frac{\partial^2}{\partial r_i \partial r_i} \right) \overline{\gamma \gamma'}(r, t) = -4C \bar{\Gamma}(t) \overline{\gamma \gamma'}(r, t) - 2C \overline{\gamma \gamma \gamma'}(r, t) - 2 \frac{\partial}{\partial r_j} \left[\overline{\gamma u_j \gamma'}(r, t) \right] \quad (3)$$

The isotropic form of (3) has been previously given by Corrsin³.

The classical arguments of final period turbulence, pertinent to asymptotic decay regimes, in which, effectively, for any given wave number the inertia contribution becomes negligible compared to diffusive effects can be used to omit legitimately the final term of (3)*. There remains a non-linearity due to the reaction which appears as the triple fluctuation term in (3) and precludes solving (2) and (3) as a closed set. However in the next

* Such has been done for the nonreacting scalar² and the scalar undergoing a first order reaction³.

section we show that even in the absence of molecular diffusion the exact statistical solution of a second order reaction in weak turbulence is always such that

$$\lim_{t \rightarrow \infty} \frac{\overline{f^3(t)}}{\overline{f(t)} \overline{f^2(t)}} \rightarrow 0$$

The existence of diffusion can be expected to strengthen this result.

Defining the Fourier transform of $\overline{f f^2}(\underline{k}, t)$ by $\phi(\underline{k}, t)$ and applying the zero third moment approximation to (3) we obtain the following restatements of (1) and (3).

$$\frac{d\overline{f(t)}}{dt} = -c \overline{f^2(t)} - c \int \phi(\underline{k}, t) d\underline{k}, \quad (4)$$

$$\left\{ \frac{\partial}{\partial t} + 2Dk^2 + 4c \overline{f(t)} \right\} \phi(\underline{k}, t) = 0. \quad (5)$$

Solving (5) to yield

$$\phi(\underline{k}, t) = \phi(\underline{k}, t_0) \exp\left\{-2Dk^2(t-t_0) - 4c \int_{t_0}^t \overline{f(t')} dt'\right\}, \quad (6)$$

where t_0 is a virtual time origin, and substituting (6) into (4) we obtain

$$\frac{d\overline{f(t)}}{dt} + c \overline{f^2(t)} = -c \left[\int \phi(\underline{k}, t_0) \exp\{-2Dk^2(t-t_0)\} d\underline{k} \right] \exp\left\{-4c \int_{t_0}^t \overline{f(t')} dt'\right\}. \quad (7)$$

It is easy to show that the asymptotic solution to (7) is, in agreement with Corrsin³,

$$\overline{f(t)} = \frac{1}{c(t-t_0)}, \quad t-t_0 \rightarrow \infty \quad (8)$$

The proof, whose detail we omit, consists of showing that the solution of (7) with $\phi(\underline{k}, t_0) = 0$, which we call $\overline{f_0(t)}$, satisfies $\overline{f_0(t)} \geq \overline{f^2(t)}$,

and $\bar{\Gamma}_0(t) = \frac{1}{C(t-t_0)}$, $t-t_0 \rightarrow \infty$. Also the solution of (7), with $D = 0$, which we call $\bar{\Gamma}_1(t)$, satisfies

$$\bar{\Gamma}_1(t) \leq \bar{\Gamma}(t) \leq \bar{\Gamma}_0(t)$$

and $\bar{\Gamma}_1(t) = \frac{1}{C(t-t_0)}$, $t-t_0 \rightarrow \infty$

Now the substitution of (8) into (5) yields

$$\phi(\underline{r}, t) = t^{-4} \cdot \phi(\underline{r}, t_0) \exp\{-2Dk^2(t-t_0)\}, \quad t-t_0 \rightarrow \infty$$

Thus

$$\bar{f}^2(t) = \int \phi(\underline{r}, t) d\underline{r} = t^{-4} \int \phi(\underline{r}, t_0) \exp\{-2Dk^2(t-t_0)\} d\underline{r}, \quad t-t_0 \rightarrow \infty. \quad (9)$$

The integral in (9) is identical with that which gives the asymptotic decay of a nonreacting scalar and the kinematic requirement of a quadratic behavior in k of the spherical shell mean of $\phi(\underline{r}, t)$ is also the same. Thus the integral in (9) behaves asymptotically as $t^{-3/2}$, and we have the result

$$\bar{f}^2(t) \sim t^{-1/2}, \quad t \rightarrow \infty$$

3 The Zero Third Moment Approximation

For weak turbulence in which the inertial transfer terms of (3) can be ignored, the role of diffusion in the decay of the mean and fluctuations is clear, and we argue below to the usefulness of a zero third moment

approximation applied to the scalar field in this case*. It is probable that similar arguments can be made in the more general case of strong turbulence, but they become quite intuitive and are not yet able to be verified by calculation. We therefore restrict ourselves to those cases in which the last term of (4) is negligible.

In terms of mean and root mean square concentrations the appropriate forms for (1) and (3) under weak turbulence are

$$\frac{d\bar{\Gamma}(t)}{dt} = -C\bar{\Gamma}(t) - C\bar{f}^2(t), \quad (10)$$

$$\frac{\partial \bar{f}^2(t)}{\partial t} = -2D \frac{\partial^2 \bar{f}}{\partial x_i^2} \frac{\partial \bar{f}}{\partial x_i} - 4C\bar{\Gamma}(t)\bar{f}^2(t) - 2C\bar{f}^3(t). \quad (11)$$

The diffusive term plays two roles. It contributes directly to the decay of the fluctuations and the mean, and it plays some role in determining the value of the triple moment $\bar{f}^3(t)$. If we consider the case $D = 0$, $\bar{f}^3(t)$, in the absence of a mechanism whose characteristic probability distribution is Gaussian, is likely to be larger. Thus we can expect that the limit of no diffusivity is almost certainly the one in which the role of $\bar{f}^3(t)$ is most crucially tested. In particular, if the zero third moment approximation is satisfactory for some range of initial values $\bar{\Gamma}(0)$, $\bar{f}^2(0)$ in the absence of diffusion, it is most likely to be so also for the same range of initial values when $D \neq 0$.

* By employing a zero fourth cumulant approximation, Corrsin³ obtained an equation for the mean concentration in situations in which the reactive terms dominate the decay. The point of emphasis here is that even the least subtle of the usual approximations is useful. This feature of second order reactions is analogous to Kraichnan's remark⁴ that closure schemes for turbulence are likely to be much more difficult for isotropic turbulence than for shear flows.

4 R. H. Kraichnan, in Proceedings of Symposium in Applied Mathematics (American Mathematical Society, Providence, Rhode Island, 1962) Vol. 13, p. 199.

The importance of the above remarks lies in the fact that the non-diffusive second order isothermal reaction in weak turbulence is able to be solved exactly, as we see below.

The system to be investigated is

$$\frac{d\Gamma}{dt} = -c\Gamma^2 \quad (12)$$

with initial statistics given by a probability density $P[\Gamma(0)]$.

It is convenient to consider $\Gamma^2(t)$ as a dimensionless concentration given by the ratio of the concentration field to its initial maximum value and to define a dimensionless time $\tau = ct$. Then it follows that the system to be solved is

$$\frac{d\Gamma^2}{d\tau} = -\Gamma^2, \quad 0 \leq \Gamma^2 \leq 1 \quad (13)$$

$$P[\Gamma^2(0)] \quad \text{prescribed.}$$

The solution of (13) for moments of any order is

$$\Gamma^{2n}(\tau) = \int \left(\frac{x}{1+x\tau} \right)^n P(x) dx, \quad (14)$$

from which $\overline{\Gamma^2}(\tau)$ and $\overline{\Gamma^{2n}}(\tau)$ can be obtained.

4 Asymptotic Results

The following limits can be deduced from (14) provided that $P[\Gamma(0)]$ is a reasonably well behaved function.

$$\begin{aligned} \lim_{\tau \rightarrow \infty} \overline{\Gamma^2}(\tau) &= \frac{1}{\tau}, \\ \lim_{\tau \rightarrow \infty} \overline{\Gamma^2}(\tau) &= O(\tau^{-4}), \\ \lim_{\tau \rightarrow \infty} \overline{\Gamma^3}(\tau) &= O(\tau^{-6}). \end{aligned}$$

The first two are in agreement with the final period results for $D = 0$, and the three combined together verify the statement made previously that

$$\lim_{t \rightarrow \infty} \frac{\bar{\gamma}^3(t)}{\bar{\Gamma}(t) \bar{\gamma}^2(t)} \rightarrow 0 .$$

5 The Multivariate Gaussian Initial Distribution

Solutions of (13) for arbitrary time are easily calculated once an explicit form for $\mathcal{P}[\Gamma(0)]$ is prescribed.

By far the simplest initial distribution to work with is the multivariate Gaussian for which of course $\bar{\gamma}^3(0)$ is zero, and by which, therefore, we can examine the generation of triple moment due to the nonlinearity of the reaction. The Gaussian does not, however, lie between zero and unity and the possibility arises that even if the initial distribution is essentially located in that region subsequent distributions evolved by the decaying process may not be. The problem turns out to be not a significant one in practice since for all feasible initial Gaussians the fluctuations decay faster than the mean for all times.

Moreover the numerical results that will be presented in section 7 indicate that the decay rates of the mean and the fluctuations are insensitive even to the assumption of a zero triple moment as compared to that generated from an initial multivariate Gaussian, and thus the same decay can be expected to be insensitive to the precise form of the initial distribution if the initial triple moment be negligible.

Therefore for the remainder of the paper $\mathcal{P}[\Gamma(0)]$ is given the form

$$\mathcal{P}[\Gamma(0)] = \frac{1}{\sqrt{2\pi} [\bar{\gamma}^2(0)]^{1/2}} \exp \left\{ -\frac{1}{2} \left(\frac{\Gamma(0) - \bar{\Gamma}(0)}{[\bar{\gamma}^2(0)]^{1/2}} \right)^2 \right\} .$$

6

Small Time Results

Straight forward expansions of the exact solution with the above form of $P[\Gamma(t)]$ yield*

$$\begin{aligned}\bar{\Gamma}(z) = & \bar{\Gamma}(0) - [\bar{\Gamma}^2(0) + \bar{\gamma}^2(0)]z + [\bar{\Gamma}^3 + 3\bar{\Gamma}(0)\bar{\gamma}^2(0)]z^2 \\ & - [\bar{\Gamma}^4(0) + 6\bar{\Gamma}^2(0)\bar{\gamma}^2(0) + 3(\bar{\gamma}^2(0))^2]z^3 \\ & + [\bar{\Gamma}^5(0) + 10\bar{\Gamma}^3(0)\bar{\gamma}^2(0) + 15\bar{\Gamma}(0)(\bar{\gamma}^2(0))^2]z^4 - \dots,\end{aligned}$$

$$\begin{aligned}\bar{\gamma}^2(z) = & \bar{\gamma}^2(0) - [4\bar{\Gamma}(0)\bar{\gamma}^2(0)]z + [10\bar{\Gamma}^2(0)\bar{\gamma}^2(0) + 8(\bar{\gamma}^2(0))^2]z^2 \\ & - [20\bar{\Gamma}^3(0)\bar{\gamma}^2(0) + 48\bar{\Gamma}(0)(\bar{\gamma}^2(0))^2]z^3 + \dots.\end{aligned}$$

Similarly the zero third moment description of the same quantities can be shown to be

$$\begin{aligned}\bar{\Gamma}(z) = & \bar{\Gamma}(0) - [\bar{\Gamma}^2(0) + \bar{\gamma}^2(0)]z + [\bar{\Gamma}^3 + 3\bar{\Gamma}(0)\bar{\gamma}^2(0)]z^2 \\ & - [\bar{\Gamma}^4(0) + 6\bar{\Gamma}^2(0)\bar{\gamma}^2(0) + (\bar{\gamma}^2(0))^2]z^3 \\ & + [\bar{\Gamma}^5(0) + 10\bar{\Gamma}^3(0)\bar{\gamma}^2(0) + 5\bar{\Gamma}(0)(\bar{\gamma}^2(0))^2]z^4 - \dots,\end{aligned}$$

$$\begin{aligned}\bar{\gamma}^2(z) = & \bar{\gamma}^2(0) - [4\bar{\Gamma}(0)\bar{\gamma}^2(0)]z + [10\bar{\Gamma}^2(0)\bar{\gamma}^2(0) + 2(\bar{\gamma}^2(0))^2]z^2 \\ & - [20\bar{\Gamma}^3(0)\bar{\gamma}^2(0) + 10\bar{\Gamma}(0)(\bar{\gamma}^2(0))^2]z^3 + \dots.\end{aligned}$$

* It can be shown⁵ that Kraichnan's Direct Interaction Hypothesis applied to the same problem yields precisely the same results as the exact solution to the orders in z shown and, moreover, for longer times it is a closer approximation than the zero fourth cumulant applied to the same problem.

5. E. E. O'Brien, To be published.

The implication of the above is that, at least for small times, the error in computing decay of the mean using the zero third moment approximation is no greater than $O\left[\left(\frac{\bar{f}^2(0)}{\bar{\Gamma}^2(0)}\right)^2\right]$ and similarly the error in computing $\bar{f}^2(t)$ is no greater than $O\left[\left(\frac{\bar{f}^2(0)}{\bar{\Gamma}^2(0)}\right)\right]$.

7 Numerical Calculations

Since the asymptotic behavior of the approximation is identical with that of the exact solution, it is plausible that the error introduced by it will be satisfactorily small for all time, for some range of values of $\bar{f}^2(0)/\bar{\Gamma}^2(0)$. To test this notion numerical computations were carried out for both solutions using the following sets of initial conditions

$$\begin{aligned}\bar{\Gamma}^2(0) &= 0.5, \\ (\bar{f}^2(0))^{1/2} &= .05, 0.1 \text{ and } 0.2.\end{aligned}$$

The last corresponds approximately to a Gaussian for which 98 percent of its area is between zero and one, and this is taken to be the largest physically sensible intensity that can be examined with a multivariate Gaussian initial condition.

Figures 1, 2 and 3 show decay of the mean and the r. m. s. fluctuations from the exact solution and as predicted by the zero third moment approximation. Even for an initial concentration field relative intensity, defined by $(\bar{f}^2)^{1/2}/\bar{\Gamma}$, of .4 (Figure 3) the mean, as represented by the approximation, is within 2 percent of the exact. Decay of r. m. s. fluctuations is predicted to within 10 percent for an initial relative intensity of .2 (Figure 2).

8 Conclusion

We can conclude that the second order reaction problem will be more tolerant of statistical approximations on the concentration fluctuations than is the case for the nonreacting scalar or for the reactant undergoing a first order reaction. It seems possible that the argument may be able to be extended to strong turbulence by noting that Mills and Corrsin⁶ obtained the empirical result that a skew-isotropic field of temperature fluctuations behind a heated grid tended rapidly toward non-skewness. The implication is that the inertia terms which we have suppressed by the assumption of weak turbulence do not generate skewness of the scalar field even when the turbulence is intense.

9 Acknowledgement

The author is indebted to Professor Michael Bentwich for extended discussions of the matter in this paper and for some of the calculations of section 5, and to Mr. Richard Reis who carried out the computations presented as Figures 1, ~~and 2~~ and 3.

The study was supported by a National Science Foundation Grant, No. 31-82.

⁶ R. R. Mills, Jr. and S. Corrsin. N.A.S.A. Memo 5-5-59 W, May, 1959.

List of Figures

(1). Decay of mean concentration and r. m. s. fluctuations.

$$\bar{C}(t) = 0.5, \sqrt{C^2(t)} = 0.05.$$

(2). Decay of mean concentration and r. m. s. fluctuations.

$$\bar{C}(t) = 0.5, \sqrt{C^2(t)} = 0.1.$$

(3). Decay of mean concentration and r. m. s. fluctuations.

$$\bar{C}(t) = 0.5, \sqrt{C^2(t)} = 0.2.$$

--- ZERO THIRD MOMENT APPROXIMATION
 ——— EXACT SOLUTION

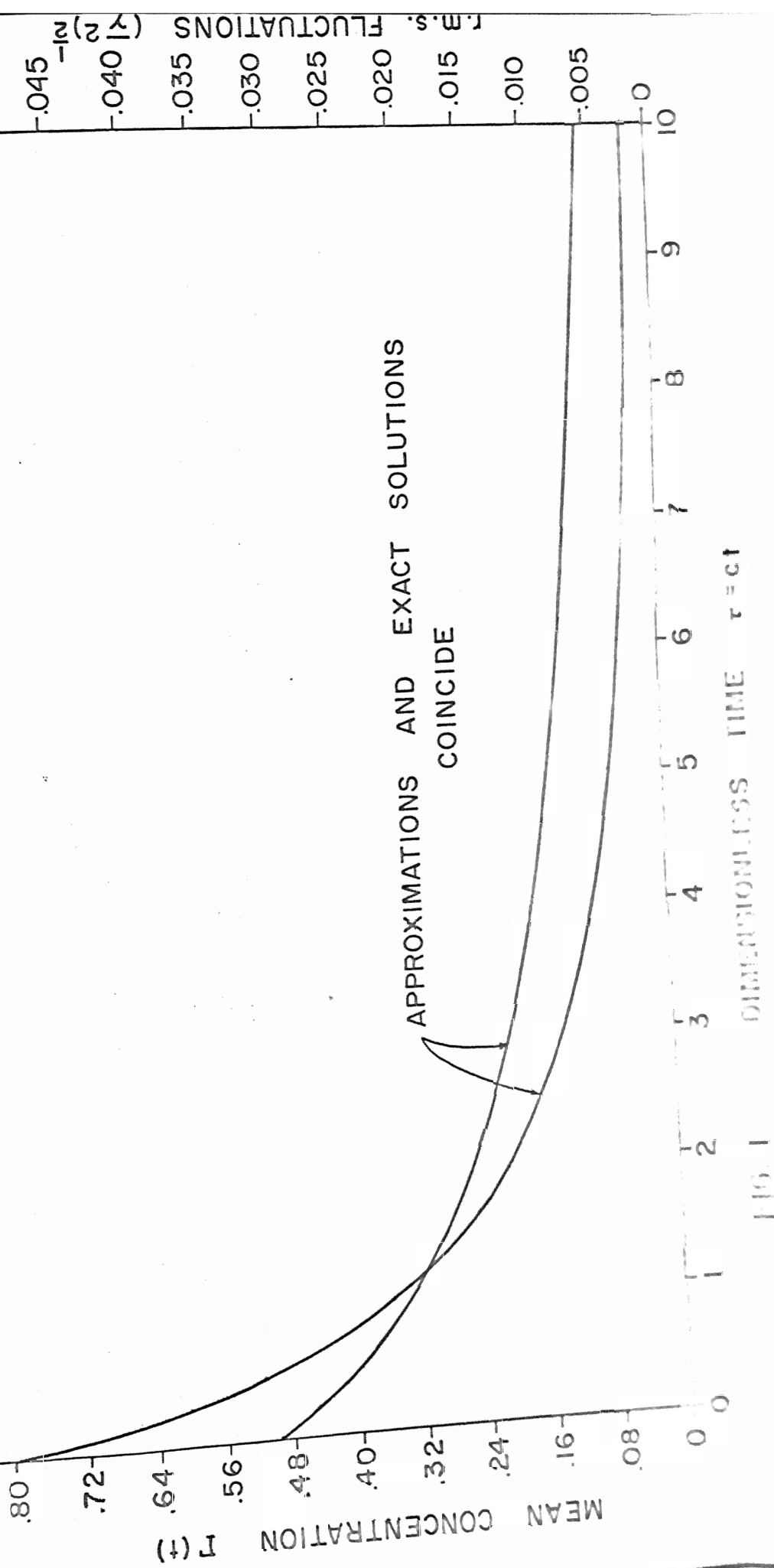


FIG. 1

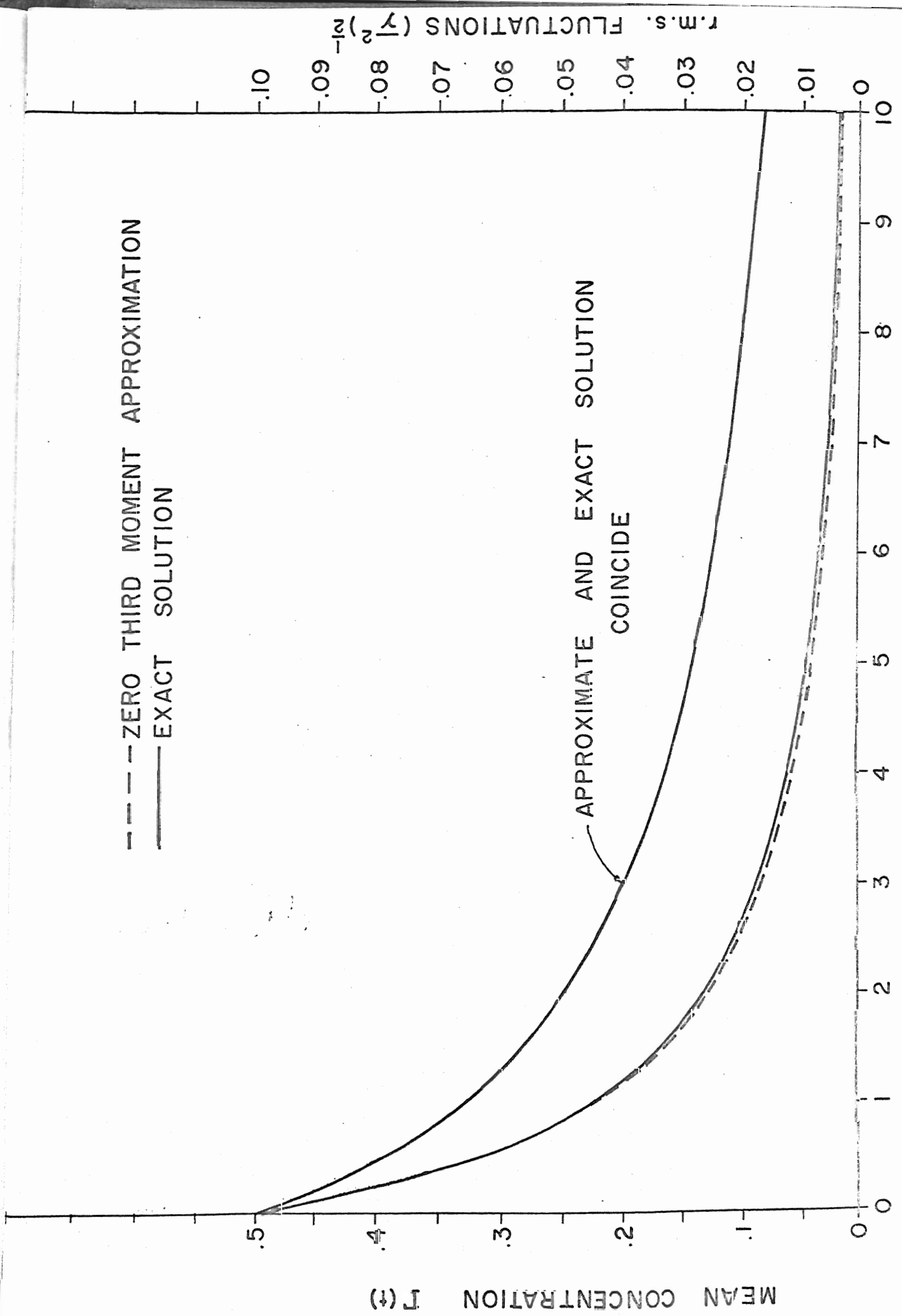


FIG. 2 DIMENSIONLESS TIME $\tau = ct$

- - - ZERO THIRD MOMENT APPROXIMATION
 ——— EXACT SOLUTION

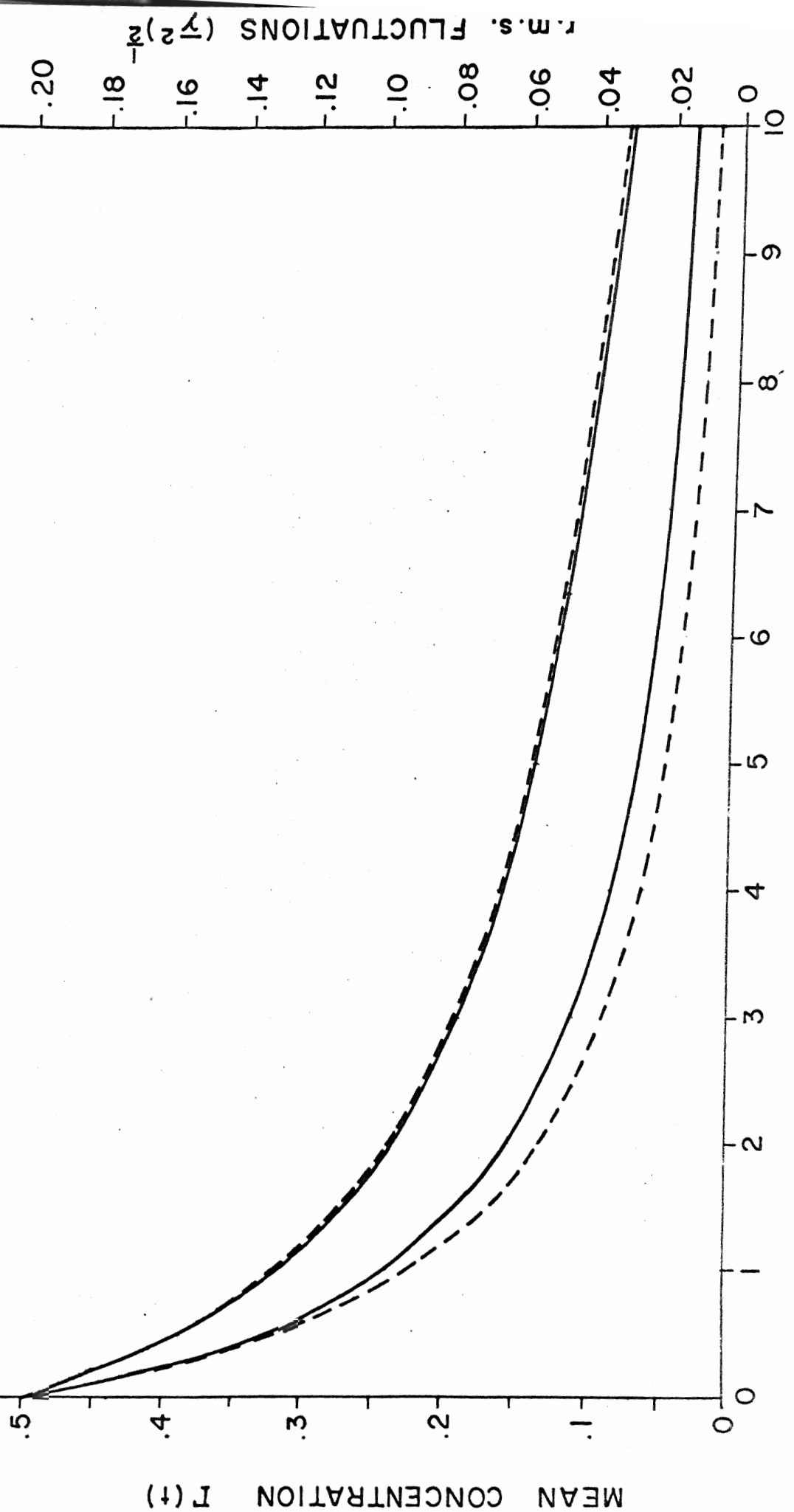


FIG. 3 DIMENSIONLESS TIME $\tau = ct$