SELF-STEEPENING OF LIGHT PULSES

F. De Martini*† and C. H. Townes*

Department of Physics

T. K. Gustafson‡

Department of Electrical Engineering
and
Research Laboratory of Electronics
Massachusetts Institute of Technology
Cambridge, Massachusetts

P. L. Kelley

Lincoln Laboratory,** Massachusetts Institute of Technology
Lexington, Massachusetts

*Work supported by the National Aeronautics and Space Administration under Contract NsG-330 and the U. S. Air Force Cambridge Research Laboratories.

†Present address: Institut d'Optique, Faculte des Sciences, Orsay, France.

‡Work supported by the Joint Services Electronics Program (Contract DA28-043-AMC-02536 (E)).

**Operated with support from the U. S. Air Force.

ABSTRACT

The self-steepening, or change in shape, of light pulses due to propagation in a medium with an intensity-dependent index of refraction is investigated. The time required for the pulse to steepen into an optical shock is found, and the time development of the pulses studied for both zero and finite times of relaxation of the index of refraction. Analytic and numerical solutions are given for the pulse development in a number of cases. The frequency spectrum is obtained in the zero relaxation time limit and the largest peak intensities are found on the lower frequency side of the input spectrum. Although the rate of steepening is modified when the decay time of the pulse becomes as short as the relaxation time for the nonlinear part of the index of refraction, the time of decay can become arbitrarily short when there is no dispersion. Estimates are given of the thickness of the optical shock region and of the frequency spreading allowed by dispersion with the effect of relaxation included. The influence of self-steepening or pulse distortion in nonlinear optical experiments is discussed.

I. INTRODUCTION

An intensity-dependent index of refraction[1] will distort an optical pulse along its direction of propagation and can give rise to optical shocks (pulse self-steepening). If the index increases due to the nonlinearity (due, for example, to the Kerr effect) the trailing edge of the pulse steepens until its intensity falls as rapidly as the dispersion will allow, this steepening being analogous to the development of an acoustic shock on the leading edge of a sound wave. Steepening occurs on the trailing part of the pulse in materials where the velocity of the peak of the pulse is slower than that of the wings, because the trailing part of the pulse catches up with the peak.

In the absence of dispersion, a discontinuity is produced on the trailing edge which travels with a velocity appropriate for the maximum energy density in the pulse and the pulse tends asymptotically to a triangular shape. The front of the pulse travels faster than the rear and the pulse continues to broaden even after the discontinuity has occurred. As indicated above, dispersion prevents the development of an infinitely sharp jump and hence limits the Fourier spectrum of the pulse from spreading indefinitely.

In the situation where the pulse exhibits a periodic modulation (due, for example, to the beating of two nearby modes of the laser) the steepening occurs in each period of the beat and the pulse eventually appears as a series of sawteeth. The frequency spectrum spreads and has its most intense components on the lower frequency side. Such effects are observed in the spectrum of the output pulse from liquids such as CS_2 when two or more frequency components are initially present in the beam.[2] It should be noted that the asymmetry of the spectrum occurs even in the limit of zero relaxation time.

Electromagnetic shocks in the microwave frequency region and at lower frequencies have been discussed.[3] In these cases attention was concentrated on shocks which form on the leading edge of the pulse, such as may occur in ferrite materials, transition layers in semiconductors, and in transmission lines. The formation of electromagnetic shocks in the optical range has been discussed by Ikuta and Taniuti[4] and by Rosen.[5] However, these authors emphasize shocks which occur during an optical period due to the generation of optical harmonics, as may be seen from the time of shock formation they give, which is related to the period of the light wave. Although the response of nonlinearities in electronic polarization is very rapid (of order of the optical period); shocks during an optical period cannot be highly developed in real media because of color dispersion and absorption. Furthermore, the nonlinear polarization responsible for the harmonic response is usually an order of magnitude smaller than, for example, that due to molecular orientation. Optical shock formation has also been considered by Broer,[6] including the role of dispersion. A brief report on optical shocks by Joenk and Landauer[7] is rather closely related to the considerations presented here, and does apply to the case of molecular orientation.

Assume, now, an optical medium with a nonlinear dielectric response which is too slow for optical frequencies, but which decreases the wave velocity with increasing optical intensity. The characteristic distance for shock formation, assuming a very short relaxation time and neglecting dispersion, is given approximately by $z_s^{-1} = 3v_2 \ (d\rho/dt)_{Min}/v_o^2$, where $\rho = n_o^2 \ E^2/8\pi$ is approximately the energy density in the wave. The symbol $(d\rho/dt)_{Min}$ indicates the largest negative slope of the initial pulse in time, and n_o is the linear refractive index. Further, v_2 is a constant of the material given by $v_2 = 8\pi n_2 v_o/n_o^3$, where v_o is the linear velocity of propagation, and the index of refraction is given by $n = n_o + n_2 E^2$. Note that the period of the electromagnetic wave does not appear. For a Gaussian pulse, the shock distance is found to be $z_s = 0.19 \ n_o \ell/\delta n$, where $\ell = t_\ell v_o$, t_ℓ being the initial width of the Gaussian in time, and δn is the nonlinear index change at the peak of the pulse. In a "small-scale trapping" filament in CS_2, $\delta n/n_o$ of the order of 10^{-1} can be obtained. For Q-switched pulses normally available, t_ℓ is about 10 nsec, giving z_s about 5 meters. However, for a "mode-locked" laser t_ℓ can be less than 10^{-11} sec and hence pulse steepening can occur over propagation path lengths of less than a centimeter. Shocks may also develop in such short distances when the light intensity is rapidly modulated due to the mixture of Brillouin or Rayleigh-scattered waves with laser light.

In the case of an initially sinusoidal intensity variation, sidebands shifted by multiples of the modulation frequency are developed by the shock, with the most intense sidebands shifted downwards by about 15% at the shock distance. Further down shifting will occur beyond the shock distance.

In general, the intensity-dependent part of the refractive index can respond at the sum or difference of nearby optical frequencies in the laser pulse. In liquids, the most prominent non-linearity which responds to difference frequencies is normally due to the Kerr effect. A dominant part of the Kerr effect corresponds to molecular alignment by the optical fields, an alignment which will respond appreciably to frequency differences $\Delta\omega$ in the range $\Delta\omega\tau < 1$ where τ is the relaxation time for alignment and is normally in the range 10^{-9} - 10^{-13} sec. Molecular alignment is also at least partly responsible for self-trapping,[9] self-focusing,[10] stimulated Rayleigh-wing scattering,[11] and light-by-light scattering.[12]

A further important difference frequency nonlinearity, which can also produce self-steepening, is intensity-dependent anomalous dispersion due to saturation of an atomic or molecular transition. For a normal distribution of population, the self-steepening due to this process will occur on the leading edge of the pulse if the light frequency is below the atomic frequency and on the trailing edge of the pulse if the light frequency is above the atomic frequency. For an inverted population, the leading and trailing edges exchange roles. This effect is analogous to the induction of self-focusing and self-defocusing[13] due to intensity-dependent anomalous dispersion, and can produce steepening in laser pulses. Estimates of the maximum intensity-dependent index change give variations from 2×10^{-6} in the Xe gas laser to 5×10^{-3} in the GaAs semiconductor laser.

II. THE LIGHT-PULSE EQUATIONS WITHOUT RELAXATION

The light-pulse equation is readily obtained from the Poynting equation, which for a nonmagnetic material may be written as

$$\vec{E} \cdot \frac{\partial \vec{D}}{\partial t} + \vec{H} \cdot \frac{\partial \vec{H}}{\partial t} + c\vec{\nabla} \cdot (\vec{E} \times \vec{H}) = 0 \quad . \tag{1}$$

Assuming a linearly-polarized plane wave propagating in the z-direction, we may write (1) as

$$\overline{E\frac{\partial D}{\partial t}} + \frac{1}{2}\frac{\partial \overline{H^2}}{\partial t} + c\,\frac{\partial \overline{EH}}{\partial z} = 0 \qquad (2)$$

where the bar indicates averaging over optical periods. This averaging is carried out since we consider the case where the optical frequency components of the energy density do <u>not</u> contribute to the nonlinearity. In the spatial derivative term we may replace $\overline{EH}$, using Maxwell's equations, as follows

$$\overline{EH} = 8\pi\,\frac{v_p(\rho)}{c}\,\rho \qquad (3)$$

where ρ is given by

$$\rho = \frac{1}{16\pi}\,(\overline{ED} + \overline{H^2}) \qquad (4)$$

and $v_p(\rho)$ is the phase velocity, which is taken to be a nonlinear function of ρ. In writing (3), terms of order of the wave length over the characteristic pulse distance and the effects of color dispersion have been neglected. We may also rewrite the time-derivative term

$$\overline{E\frac{\partial D}{\partial t}} + \frac{1}{2}\frac{\partial \overline{H^2}}{\partial t} = \frac{1}{2}\,\frac{\partial\,(\overline{ED} + \overline{H^2})}{\partial t} + \frac{1}{2}\,\frac{\overline{ED}}{\epsilon}\,\frac{\partial\epsilon}{\partial t} = 8\pi\,\frac{\partial\rho}{\partial t} + \frac{4\pi\rho}{\epsilon}\,\frac{\partial\epsilon}{\partial t}\ , \qquad (5)$$

where $D = \epsilon E$ and where $\epsilon = c^2/v_p^2$. The light-pulse equation is then

$$\frac{\partial \rho}{\partial t} + \frac{\rho}{2\epsilon}\frac{\partial \epsilon}{\partial t} + \frac{\partial (v_p \rho)}{\partial z} = 0 \tag{6}$$

or, replacing ϵ by the phase velocity,

$$\frac{\partial \rho}{\partial t} - \frac{\rho}{v_p}\frac{\partial v_p}{\partial t} + \frac{\partial v_p \rho}{\partial z} = 0 \quad . \tag{7a}$$

In addition to the equation for ρ we also have the approximate equation of motion of the phase

$$\frac{\partial \Phi}{\partial t} + v_p \frac{\partial \Phi}{\partial z} = 0 \quad . \tag{8}$$

Assuming that the wave consists of a dominant initial Fourier component at or near frequency ω_o and propagation vector $k_o = \omega_o/v_o$ then we may write

$$\Phi = k_o z - \omega_o t + \phi \tag{9}$$

and we have for ϕ

$$\frac{\partial \phi}{\partial t} + v_p \frac{\partial \phi}{\partial z} = k_o (v_o - v_p) \quad . \tag{10}$$

In general, v_p is a function of other variables (in addition to ρ) such as temperature and pressure. We assume these do not change in time or space enough to significantly affect the nonlinear terms of importance. If relaxation is sufficiently rapid, we can take v_p to be a function of the instantaneous value of ρ and equation (7a) may be rewritten

$$\left[1 - \frac{\rho}{v_p}\frac{\partial v_p}{\partial \rho}\right]\frac{\partial \rho}{\partial t} + \left[v_p + \rho\frac{\partial v_p}{\partial \rho}\right]\frac{\partial \rho}{\partial z} = 0 \quad . \tag{7b}$$

Or, introducing the quantity

$$\sigma = \rho - \int \frac{\rho}{v_p}\frac{\partial v_p}{\partial \rho}\, d\rho + \sigma_o \quad , \tag{11}$$

where σ_o is independent of ρ and time, we may write the light-pulse equation as

$$\frac{\partial \sigma}{\partial t} + \frac{\partial (v_p \rho)}{\partial z} = 0 \quad . \tag{12}$$

If the material system has a high heat capacity and rapid exchange of energy between its modes, then molecular orientation does not appreciably change its temperature. In this case, the system is isothermal during passage of the light pulse and σ is the Gibbs free energy. The change in velocity due to the nonlinearity may be written

$$v_p = v_o - v_2\rho + v_4\rho^2 + \ldots \quad . \tag{13}$$

For most situations of interest $v_2 > 0$. If terms in ρ^2 and higher powers are neglected in (13), and assuming $v_2\rho \ll v_o$,[14] we may approximate (7b) by

$$-\frac{\partial \rho}{\partial t} = (v_o - 3v_2\rho)\frac{\partial \rho}{\partial z} = \frac{\partial (v_e \rho)}{\partial z} \quad . \tag{14}$$

Here,

$$v_e \equiv v_o - \frac{3}{2} v_2\rho = v_p - \frac{1}{2} v_2\rho \quad . \tag{15}$$

Also (10) becomes

$$\frac{\partial\phi}{\partial t} + v_o \frac{\partial\phi}{\partial z} = k_o \, v_2\rho \quad . \tag{16}$$

σ obeys the same equation as ρ provided $v_2\rho << v_o$. Equation (14) may be recognized as the continuity equation for energy density, with the effective velocity of the energy flow through a boundary given by v_e.

In the frame moving with linear velocity v_o along the z axis, (14) becomes

$$\frac{\partial\rho}{\partial t} = \frac{3}{2} v_2 \frac{\partial\rho^2}{\partial z} = 3v_2\rho \frac{\partial\rho}{\partial z} \quad , \tag{17}$$

where z is now the coordinate in the moving frame,[15] and (16) becomes

$$\frac{\partial\phi}{\partial t} = k_o \, v_2\rho \quad . \tag{18}$$

The solution of (11), the light pulse equation, can be obtained by the method of characteristics and is given in implicit form (for the boundary condition of an input signal varying in time at z = 0) by the set of equations

$$\rho(z,t) = \rho(0,t_o) \tag{19}$$

$$z = \{v_o - 3v_2\rho(0,t_o)\}(t-t_o) \quad . \tag{20}$$

Equation (19) indicates that along each characteristic curve leaving $z = 0$ at $t = t_o$, the density ρ remains constant. Equation (20) determines the family of characteristic curves, in this case straight lines. Taken together (19) and (20) describe the distortion of a pulse.

Another form of the solution of (14) is $\rho[z - (v_o - 3v_2\rho)(t-t_o)]$ where ρ is an arbitrary function. However, for physical reasons it must be single valued. It follows from this form, or from (19) and (20), that the peak intensity of the pulse is constant, and that the pulse may be constructed at any distance before formation of a shock by translating in time a given intensity from its initial time, an amount proportional to the product of $(v_o - 3v_2\rho)^{-1}$ and the distance.

There are thus three distinct velocities of importance. One is the phase velocity of the optical wave, $v_p \equiv v_o - v_2\rho$. A second is the effective velocity of energy flow, $v_e \equiv v_o - 3/2\ v_2\rho$. The third is the envelope velocity, or the velocity of a given field intensity on the envelope of the optical wave, which is $v_{en} \equiv v_o - 3v_2\rho$.

At some distance a discontinuity or shock forms due to the crossing of two characteristics. This crossing distance is found from (20) as follows.

$$\frac{z}{[v_o - 3v_2\rho(0,t_o')]} + t_o' = t = \frac{z}{v_o - 3v_2\rho(0,t_o)} + t_o \tag{21}$$

that is,

$$\frac{1}{z} = \frac{\dfrac{1}{v_o - 3v_2\rho(0,t_o')} - \dfrac{1}{v_o - 3v_2\rho(0,t_o)}}{t_o - t_o'} \quad . \tag{22}$$

The characteristics that cross first must be initially adjacent so that we can take the limit $t_o' \to t_o$ to obtain

$$z^{-1} = -\frac{d\left(\dfrac{1}{v_{en}}\right)}{dt_o} = \frac{-3v_2 \dfrac{d\rho(t_o)}{dt_o}}{[v_o - 3v_2\rho(t_o)]^2} \quad . \tag{23}$$

The smallest distance occurs when the right-hand side of (23) is largest and positive (since the left-hand side of (23) is positive). This occurs for times greater than the time when the peak passes. In other words, the steepening occurs on the trailing edge of the pulse. The steepening distance z_s is then given by

$$z_s^{-1} = \left.\left(\frac{-3v_2 \dfrac{d\rho(t_o)}{dt_o}}{[v_o - 3v_2\rho(t_o)]^2}\right)\right|_{Max} \approx \left.\left(\frac{-3v_2}{v_o^2}\frac{d\rho}{dt_o}\right)\right|_{Max} \tag{24}$$

For a Gaussian initial pulse $\{\rho(0,t) = \rho_o \exp(-4t^2/t_\ell^2)\}$, (24) gives the steepening distance

$$z_s \approx \frac{1}{6}\sqrt{\frac{e}{2}}\ \frac{t_\ell\ v_o^2}{v_2\ \rho_o} = .194\ \frac{\ell n_o}{\delta n} \tag{25}$$

where δn is the nonlinear refractive index change at the pulse maximum and $\ell = v_o t_\ell$.

If the boundary conditions are those of a given variation in space at an initial time t = 0, the solution of (14) can be written in the form

$$\rho(z,t) = \rho(z_o,t_o) \tag{26}$$

$$z - z_o = \{v_o - 3v_2\rho(0,t_o)\}t \quad . \tag{27}$$

The characteristics will first cross at a time given by

$$t_s^{-1} = 3v_2 \left.\frac{d\rho}{dz_o}\right|_{Max} \approx v_o\ z_s^{-1} \tag{28}$$

To solve the phase equation in order to determine the Fourier spectrum of the pulse we introduce the coordinates

$$\zeta = \frac{1}{2}\ (z/v_o + t) \quad , \qquad \eta = \frac{1}{2}\ (z/v_o - t) \quad . \tag{29}$$

The phase equation (16) becomes

$$\frac{\partial\phi}{\partial\zeta} = \frac{\omega_o\ v_2}{v_o}\ \rho(\eta,\zeta) \tag{30}$$

which has the solution, assuming $\phi = 0$ at $z = 0$,

$$\phi = \frac{\omega_o v_2}{v_o} \int_{\frac{1}{2}(t - z/v_o)}^{\frac{1}{2}(t + z/v_o)} d\zeta \; \rho(\eta,\zeta) \quad . \tag{31}$$

We then use the characteristic equation (20) written in the form

$$\zeta(\eta,t_o') = \frac{1}{3v_2 \, \rho(t_o')} \left[\{3v_2\rho(t_o') - 2v_o\}\eta + \{3v_2\rho(t_o') - v_o\}t_o' \right] , \tag{32}$$

to change the variable of integration from ζ to t_o'

$$\phi = - \frac{\omega_o}{v_o} \int_{t_o}^{t-z/v_o} dt_o' \; \rho(t_o') \; \frac{\partial\zeta(\eta,t_o')}{\partial t_o'} \tag{33}$$

where $t_o' = t - z/[v_o - 3v_2\rho(t_o')]$.

Equation (33) may be integrated by parts to give

$$\phi = \omega_o \left\{ \frac{z v_2 \, \rho(t_o)}{v_o[v_o - 3v_2\rho(t_o)]} \left(1 - \ln\,[\rho(t_o)/\rho_o]\right) \right.$$

$$\left. - \, v_2 \int_{t_o}^{t-z/v_o} \rho(t_o')\,dt_o' + \frac{1}{3}\int_{t_o}^{t-z/v_o} \ln\,[\rho(t_o')/\rho_o] \; dt_o' \right\}. \tag{34}$$

The last two integrals can be easily carried out for specific input signals $\rho(t_o)$.

Appendix A gives some further properties of the light-pulse equation (14).

Figure 1 shows the development of a Gaussian pulse for the two types of boundary conditions discussed above. Note in Fig. 1b that the pulse moves backwards in the moving frame because of the intensity-dependent decrease in velocity.

Figures 2a-e show the evolution in shape and Fourier spectrum of a pulse which is initially sinusoidally modulated. This modulation could arise from the beating of two laser modes or from sidebands produced by a stimulated scattering process (e.g. Brillouin or Rayleigh scattering[16]). Bloembergen and Lallemand[2] have already discussed the beating of two frequencies together to produce, through Kerr effects, additional sidebands. Their perturbation approach is useful for the initial growth of additional frequency components. The present approach allows calculation of the frequency spectral distribution for cases where the additional components have become arbitrarily intense.

To examine the frequency spectrum of a pulse we take the time Fourier transform at a given point in space along the direction of propagation. To obtain the spectrum shown in Figs. 2d and 2e we have used a spatial boundary condition. The spectrum is given by

$$S(z,\omega_o+\Delta\omega) = \frac{c}{4\pi} \operatorname{Re} E(z,\omega_o+\Delta\omega)\; H^*(z,\omega_o+\Delta\omega)$$

$$\approx \frac{v_o}{T} \left| \int_{-T}^{T} dt\; \{\rho(z,t)\}^{1/2}\; e^{i\phi(z,t)}\; e^{i\Delta\omega t} \right|^2 , \qquad (35)$$

where $\Delta\omega$ is the frequency shift relative to the frequency ω_o. For a periodic pulse T is taken to be the modulation period.

The second line of (35), used to find the spectra in 2d and 2e, holds in the limit $v_o \gg v_2\rho$. This approximation tends to increase the high frequency components relative to the lower frequency components. The quantity $S(z,\omega_o + \Delta\omega)$ is the average power flowing per cm^2 per second in the spectral component at frequency $\omega_o + \Delta\omega$. Using the field equations this quantity can be shown in the lossless case to obey the generalized Manley-Rowe relations.[17] We note that the power spectrum develops one or more strong peaks on either side of ω_o; as z increases the shifts become more predominant. The prominent downward shifts come from the leading part of the pulse while the upward shifts come from the steepened region in the tail. The upward peaks shift in frequency faster because of the steepness of the tail. The ratio of peak spectral intensity on the upper side to the peak spectral intensity on the lower side becomes progressively smaller with distance. The amount of upward shifting will decrease when the relaxation time is taken to be non-zero. The same general features are observed in the power spectra of other pulses such as the Gaussian. Deviations in the spectra on the high frequency side occur when the steepness of the shock becomes comparable to the optical wavelength, because the approximations used to obtain the light pulse equations break down.

The downward frequency shift $\delta\omega$, estimated in Appendix B for the particular case of an N-wave or asymptotic pulse is

$$\delta\omega = - \omega_o \frac{z}{\ell} \frac{\delta n}{n_o} \quad . \tag{36}$$

A similar result has been obtained by Joenk and Landauer.[7] If expression (36) is used for the case of sinusoidal modulation, it predicts a 30 percent shift in frequency at z_s for the most prominent peak, which is to be compared with the 15 percent shift shown in Figs. 2d and 2e. As the pulse develops for distances larger than z_s, further downward shifting of the most intense spectral components will occur.

Even though present considerations are not strictly valid in the shock range ($t > t_s$) because of the finite relaxation time τ of the nonlinear dielectric response and because of dispersion, as well as the breakdown assumption that the characteristic pulse lengths are much greater than the optical wavelength, it is nevertheless valuable to examine the pulse behavior assuming the relaxation to be infinitely fast to gain insight into the behavior of solutions, particularly when $t_\ell \gg \tau$. After the discontinuity first occurs at t_s, the conservation equation (14) in differential form must be replaced by one in integral form. It is possible to show in the frame moving with velocity v_o that the velocity of the shock front is given by the Rankine-Hugoniot equation

$$v_s = \frac{3}{2} v_2 \, [\rho^2]/[\rho] = - \frac{3}{2} v_2 \, (\rho_+ + \rho_-) \tag{37}$$

where [ ] indicates the jump across the shock front of the quantity contained inside. ρ_+ and ρ_- are respectively energy densities immediately in front of and immediately behind the discontinuity. The negative sign shows that the shock moves backwards in the moving frame with a speed proportional to the average of the initial and final energy densities. For the parts of the pulse not on the discontinuity, the velocity is where $v_{\rho_\pm}$ are the velocities of the pulse just in front of and behind the shock. Equation (38) indicates the tail of the pulse is catching up with the shock, increasing the discontinuity with time, and thereby further increasing its backwards velocity (in the moving frame). This process continues until all or most of the tail catches up with the pulse discontinuity.

The entire pulse spreads in time because the front of the pulse travels faster than the shock region. Eventually the shock discontinuity begins to decrease in height because of this spreading. It can be shown [18] that the shape of the pulse tends, for $t \rightarrow \infty$, to a triangle (N-wave) for an initial function which is zero outside a finite interval. The height of the shock front in the N-wave decreases as $t^{-1/2}$ and the width increases as $t^{+1/2}$, the total energy being constant. If the energy density is periodic with period p (the case of a sinusoidal modulated pulse) the asymptotic solution is a series of sawtooth functions (i.e. a series of N-waves) with the discontinuity in each period proportional to p/t.

As the pulse steepens, more and more Fourier components appear in the pulse, the spread in Fourier components being given by $d\Delta k \approx 2\pi$, where d is the thickness of the shock. If the medium is dispersive, the spread in Δk gives a spread in linear velocity $\Delta v_o = |\frac{dv_o}{dk}|\Delta k \approx \frac{v_o^2}{n_o} |\frac{dn}{d\omega}|\Delta k$. This spread in velocities tends to dissipate the shock front. Equating this spreading velocity to the nonlinear velocity change which steepens the pulse, we obtain the following approximate result for the stable thickness

$$d_o \approx \frac{2\pi v_o^2}{3v_2\,\rho_o n_o} \left|\frac{dn}{d\omega}\right| = \frac{v_o}{3\delta n} \left|\frac{dn}{d\nu}\right| \tag{39}$$

where δn is the nonlinear index change. This distance is somewhat analogous to the Taylor thickness in acoustical shocks.[18] The frequency spread, as opposed to the downward shift given by (36), is

$$\Delta\nu_{d_o} \approx \frac{3v_2\rho_o n_o}{v_o \left|\frac{dn}{d\omega}\right|} = \frac{3\delta n}{\left|\frac{dn}{d\nu}\right|} \quad . \tag{40}$$

In CS_2, $\left|\frac{dn}{d\nu}\right| \approx 5 \times 10^{-6}$ cm and in a small scale trapped filament, the percentage changes in index range from about 0.1% up to a few tens of percent. Choosing $\delta n/n_o \approx .1$, we obtain $\Delta\nu_{d_o} \approx 10^5$ cm^{-1}, and $d_o \approx 10^{-5}$ cm, a distance smaller than a wavelength, which is small enough that some of the approximations used are no longer correct. We must include higher derivative terms which were dropped in obtaining the energy density and phase equationsbecause of the assumption that the distance (or time) over which the pulse changes is large compared to a wavelength (or period). The actual thickness is considerably increased and the frequency spread correspondingly reduced by relaxation of the nonlinear index as will be shown in the next section.

III. THE LIGHT PULSE EQUATIONS WITH RELAXATION

In Section II the pulse steepening equations are derived assuming an infinitely fast response of the nonlinear polarization. If the relaxation process is exponential, then the nonlinear index change $\delta\epsilon$ (where $\delta\epsilon = \epsilon - \epsilon_o$) can be written, to first order in ρ,[19]

$$\delta\epsilon(z,t) = \frac{2\epsilon_o v_2}{v_o} \int_{-\infty}^{t} \frac{dt'}{\tau} \rho(z,t') \; e^{-(t-t')/\tau} \quad . \tag{41}$$

In differential form, equation (41) is

$$\frac{\partial\delta\epsilon}{\partial t} = \frac{2\epsilon_o v_2 \rho}{v_o \tau} - \frac{\delta\epsilon}{\tau} \quad . \tag{42}$$

This may also be written as an equation for the phase velocity

$$\frac{\partial\delta v}{\partial t} = - \frac{v_2}{\tau} \rho - \delta v/\tau \tag{43}$$

where $\delta v = v_p - v_o$. In the moving frame (41) becomes

$$\delta\epsilon(z,t) = \frac{2\epsilon_o v_2}{v_o \tau} \int_{-\infty}^{t} \frac{dt'}{\tau} \; \rho(z + v_o[t-t'],t') e^{-(t-t')/\tau} \tag{44}$$

where z is in the moving frame. In differential form we have

$$\frac{\partial\delta\epsilon}{\partial t} - v_o \frac{\partial\delta\epsilon}{\partial z} = \frac{2\epsilon_o v_2 \rho}{v_o \tau} - \frac{\delta\epsilon}{\tau} \quad . \tag{45}$$

Equations (7a) and (8) together with (43) form a set of coupled equations describing the development of pulse steepening in the case of the nonzero relaxation time. In the limit when $\tau \to 0$ (7a) and (43) combine to give (15).

If we solve equation (43) for ρ and insert it into the second term on the left of (7a) and approximate v_p by v_o in the denominator of this term, the equation becomes

$$\frac{\partial \sigma}{\partial t} + \frac{\tau}{v_2 v_o}\left(\frac{\partial \delta v}{\partial t}\right)^2 + \frac{\partial (v_p \rho)}{\partial z} = 0 \qquad (46)$$

where

$$\sigma = \rho + \frac{(\delta v)^2}{2 v_2 v_o} + \sigma_o \qquad (47)$$

and σ_o is independent of time. The second term in (46) represents the irreversible part of the work done by the external sources on the volume element under consideration. Since this work must be positive unless the medium amplifies the wave, or there are other overriding losses such as those due to resistance, v_2 is normally positive. For an isothermal system, σ is just the Gibbs free energy made up of the energy stored in the field and dielectric and the remaining free energy. Furthermore, in the case of the alignment of polarizable molecules, $v_2 = K/T$, where K is a constant and T the temperature. It can then be shown that the part of the entropy density due to alignment is

$$s = - \frac{v_o (\delta \epsilon)^2}{8K\epsilon_o^2} = - \frac{(\delta v)^2}{2Kv_o} \quad . \qquad (48)$$

The electromagnetic field does not appear in this result since the entropy is an intrinsic property of the alignable molecules. The internal energy density, u, is found to be equal to ρ [defined by (4)] either from the relation $u = \sigma - Ts$ or from integrating EdD + HdH holding entropy (and hence $\delta\epsilon$) constant. In aligning the molecules under isothermal conditions, heat flows out of the alignment degree of freedom into the surroundings. If the relaxation time is nonzero, then part of this heat flow is irreversible, this part being related to the ratio of the relaxation time to the time over which the pulse is changing.

Equations (7a) and (43) still indicate the build up of a rarefaction shock. For pulse widths which are long compared to the relaxation time, the pulse builds up a sharp trailing edge of width τv_o in a distance given by (24). Further sharpening occurs with the thickness decaying to zero (neglecting loss and dispersion). The finite relaxation time of the nonlinearity does not intrinsically limit steepening and frequency broadening of the pulse. After steepening of the trailing edge to a width somewhat smaller than τv_o has occurred, the velocity change δv in the steep region and the tail should be determined primarily by the exponential decay of the peak velocity. Therefore

$$\delta v(z) = - \delta v_o \, e^{(z-z_o)/v_o \tau} , \quad \text{for } z < z_o \tag{49}$$

where z_o is the leading point or peak of the steep region and where δv_o is determined at this peak. In other words, we assume the velocity is insignificantly affected by the field in the tail region. The rate at which a point z is catching up with the front of the steep edge is given by

$$\Delta \dot{z} = \dot{z}_o - \dot{z} = \delta v_o - \delta v = - \delta v_o \, (1 - e^{-\Delta z / v_o \tau}) \tag{50}$$

which has the solution

$$\Delta z = v_o\tau \ \ln \left\{1 + e^{-\delta v_o \Delta t/v_o\tau} \left(e^{+\Delta z_o/v_o\tau} - 1\right)\right\} \quad , \tag{51}$$

where $\Delta z_o = z_o - z$ at $\Delta t = 0$. For values of $\Delta z_o/v_o\tau << 1$, the decay is always exponential; for $\Delta z_o/v_o\tau >> 1$, Δz decreases linearly with Δt for $\Delta z_o >> \delta v_o \Delta t$. More important, however; for all positive $\Delta z_o/v_o\tau$, z decays exponentially for large Δt's with a time constant $v_o\tau/\delta v_o$. If $t_\ell >> \tau$, the ratio $v_o/\delta v_o$ is roughly the ratio of duration of the pulse time t_ℓ to the time t_s for a shock to develop, so that the time constant is approximately $t_s\tau/t_\ell$. Thus, in the regime where $t_s > t_\ell > \tau$ the pulse sharpens exponentially with a time constant which is intermediate in value between t_s and τ. If the initial width of the pulse or length of the modulation cycle is less than $v_o\tau$ (i.e. $\tau >> t_\ell$) then the nonlinearity responds very little to the pulse during the intensity build up and steepening occurs with a time constant $v_o\tau^2/3v_2\rho_{Max}t_\ell$ or approximately $t_s\tau^2/t_\ell^2$. Neglecting dispersion, we see that exponential decay of the nonlinear index indicates an infinite time for infinite steepening.

We can include the combined effects of relaxation and dispersion in an estimation of the thickness of the steep region by noting that relaxation reduces the nonlinear velocity change through a shock front of thickness d by a factor $(1 - \exp\{-d/v_o\tau\})$. In the limit in which the distance d_o given by (23) is much larger than the relaxation length (i.e. $d_o >> v_o\tau$), the thickness reduces to (39) and the frequency spread is given by (40). However, when

$d_o \ll v_o\tau$ the thickness is $d = \sqrt{v_o \tau d_o}$. Relaxation can thus markedly increase the thickness. The frequency spread is also modified so that $\Delta\nu_d = \sqrt{\Delta\nu_{d_o}/\tau}$. For CS_2, $\tau = 3 \times 10^{-13}$ sec and since from the previous section $d_o \approx 10^{-5}$ cm, we have $v_o\tau \gg d_o$. The modified thickness is approximately 3μ and the frequency spread is 3000 cm^{-1}. This is the maximum frequency spread which can occur due to steepening. The degree of steepening and hence spreading depends strongly on experimental conditions.

The steepening of specific pulse profiles has been investigated by solving (43) and (7a) numerically using a modified Runge-Kutta technique. Results are shown in Figs. 3 through 6 for index changes expected to occur in small scale trapping $\left(\frac{v_2 \rho_o}{v_o} \simeq .1\right)$.

The evolution of a short pulse in a trapped beam can be approximated by the behavior of an intense pulse penetrating the nonlinear trapped region from a linear region. This is shown in Figs. 3 and 4 for two different ratios of the relaxation time to the shock time. In Fig. 3 this ratio is about .016 and in 2 shock times an amount of steepening occurs which is approximately equal to the steepening which occurs in 8 shock times for Fig. 4 where the ratio is about .25.

The shape taken by a periodically modulated optical pulse depends upon the ratio of the relaxation time of the dielectric to the modulation period. In Fig. 6 this ratio is relatively large, in which case the dielectric relaxation from one period of modulation can cause a continued dielectric constant fall off into the region occupied by the leading edge of the next cycle. This causes steepening on both the front and back edges of the modulation cycle. If, on the other hand, the above ratio is small, as is the case in Fig. 5, only the lagging edge of the periods steepen appreciably. In Figs. 5 and 6 the boundary conditions for the pulse were taken to be that of an initial sinusoidal spatial distribution at $t = 0$. With this condition the distortion of a single cycle after moving a distance z is nearly the same as that which occurs when the cycle moves a distance z from an input boundary if the steepening distance is much longer than the length of a modulation cycle.

As is seen from the results in Figs. 5 and 6, as well as equations (7a) and (43) and the discussion above, the distortion for a given z is strongly affected by the ratio of the relaxation time to the modulation frequency. The phase and frequency spectrum will also be affected; the spectrum spreads less as the relaxation time increases in agreement with the results of Lallemand.[2]

ACKNOWLEDGEMENTS

We would like to thank C. Cercignani, R. Y. Chiao, H. Cheng, and H. Schlossberg for useful discussions. We are particularly grateful to H. A. Haus for his helpful interest in this problem. One of us (F.D.M.) would like to thank the Consiglio Nazionale delle Ricerche, Rome, Italy, for partial support during the course of this work.

APPENDIX A

We examine in this appendix two of the overall features of the pulse described by (15), specifically the behavior of the center of energy and its spatial dispersion. For $t < t_s$ and for a pulse whose energy density tends to zero as $|z|^{-\epsilon}$ for $|z| \to \infty$, where $\epsilon > 1$, we shall show the following:

a) the center of energy $\bar{z} = \int_{-\infty}^{\infty} z\rho dz / \int_{-\infty}^{\infty} \rho dz$ travels at uniform speed.

b) The spatial dispersion of a symmetric pulse $\sigma^2 = \overline{(z - \bar{z})^2}$ increases quadratically with time.

Consider first the following lemma: the integral

$$I_n = \int_{-\infty}^{\infty} \rho^n \, dz \tag{A-1}$$

is constant in time if $n \geq 1$. To see this we differentiate (A-1) with respect to time

$$\frac{\partial I_n}{\partial t} = n \int_{-\infty}^{\infty} \rho^{n-1} \frac{\partial \rho}{\partial t} \, dz \tag{A-2}$$

and using (13) we have

$$\frac{\partial I_n}{\partial t} = 3v_2 n \int_{-\infty}^{\infty} \rho^n \frac{\partial \rho}{\partial z} \, dz \quad . \tag{A-3}$$

If time is held constant then we may rewrite this as

$$\frac{\partial I_n}{\partial t} = 3v_2 n \int_{\rho(z=-\infty)}^{\rho(z=+\infty)} \rho^n \, d\rho$$

$$= 3v_2 \frac{n}{n+1} \rho^{n+1} \Big|_{\rho(z=-\infty)}^{\rho(z=+\infty)}$$

$$= 0 \quad . \qquad \text{(A-4)}$$

Now consider the motion of the center of energy

$$\frac{d\bar{z}}{dt} = \frac{1}{I_1} \int_{-\infty}^{\infty} z \frac{\partial \rho}{\partial t} dz = \frac{3v_2}{I_1} \int z \rho \frac{\partial \rho}{\partial z} dz$$

$$= \frac{3v_2}{I_1} \left[\frac{1}{2} z \rho^2 \right]_{-\infty}^{\infty} - \frac{3v_2}{2I_1} \int_{-\infty}^{\infty} \rho^2 \, dz \quad . \qquad \text{(A-5)}$$

The first term is zero from the assumption above concerning the dependence of ρ for large $|z|$ while the second term is constant from the lemma. Therefore,

$$\frac{d\bar{z}}{dt} = -\frac{3v_2 I_2}{2I_1} \quad , \qquad \text{(A-5a)}$$

and

$$\bar{z} = -\frac{3v_2 I_2}{2I_1} t + \bar{z}_o \quad , \qquad \text{(A-6)}$$

the center of energy moves backwards in the moving frame linearly in time.

Next consider the function

$$\overline{z^2} = \frac{1}{I_1} \int z^2 \rho \, dz \tag{A-7}$$

differentiating with respect to time we obtain

$$\frac{d\overline{z^2}}{dt} = \frac{1}{I_1} \int z^2 \frac{\partial \rho}{\partial t} dz = - \frac{3v_2}{I_1} \int_{-\infty}^{\infty} z \rho^2 \, dz \tag{A-8}$$

and differentiating once more we have

$$\frac{d^2 \overline{z^2}}{dt^2} = \frac{6v_2^2}{I_1} \int_{-\infty}^{\infty} \rho^3 \, dz = \frac{6v_2^2}{I_1} I_3 \quad . \tag{A-9}$$

Using initial conditions, we then find for $\overline{z^2}$

$$\overline{z^2} = \frac{3v_2^2}{I_1} I_3 \, t^2 + At + \overline{z_o^2} \tag{A-10}$$

where

$$A = - \frac{3v_2}{I_1} \int_{-\infty}^{\infty} z \rho^2 \, dz \Big|_{t=o} \quad . \tag{A-11}$$

Therefore the dispersion is given by

$$\sigma^2 = \overline{(z - \bar{z})^2} = \frac{3v_2 I_3 t^2}{I_1} + At + \overline{z_o^2} - \left(\frac{3v_2 I_2}{2I_1}\right)^2 t^2$$

$$+ 3\bar{z}_o v_2 I_2 t/I_1 - \overline{z_o^2} \quad . \tag{A-12}$$

If we take the pulse symmetrical at $t = 0$ and further that $\bar{z}_o = 0$ then we have

$$\sigma^2(t) = \frac{9v_2^{\ 2}}{4I_1^{\ 2}} \left(\frac{4}{3} I_1 I_3 - I_2^2 \right) t^2 + \sigma^2(o) \quad . \tag{A-12a}$$

One can show using the Cauchy-Schwartz-Boniakowsky relation that $I_1 I_3 - I_2^2 > 0$, therefore

$$\sigma^2(t) \geqslant \sigma^2(o) \quad , \tag{A-13}$$

and the dispersion <u>increases</u> quadratically with time.

APPENDIX B

We examine here the development of an N-wave [the asymptotic solution to (15)]. We will assume at $z = 0$ an N-wave of the form

$$\rho = \frac{v_o t}{3v_2 \, (t+t_o)} \quad , \qquad 0 \leqslant t \leqslant t_1 \tag{B-1}$$

$$= 0 \quad , \quad \text{elsewhere}$$

and

$$\phi = 0 \quad , \tag{B-2}$$

where $t_1 = \ell/v_o$. Here ℓ is the initial pulse length in the medium at t_1, $t_o = \ell/\delta v_o - t_1$, and $\delta v_o = 3v_2\rho_{Max}(t_1)$.

The solution for ρ is

$$\rho = \frac{v_o t - z}{3v_2 \, (t+t_o)} \quad , \quad 0 \leqslant v_o t - z \leqslant z_o(t) \tag{B-3}$$

$$= 0 \quad , \quad \text{elsewhere}$$

where

$$z_o(t) = [\ell \delta v_o (t+t_o)]^{1/2} \quad . \tag{B-4}$$

From (16) and the boundary condition, the solution for the phase is

$$\phi = \frac{\omega_o}{3v_o} (z - v_o t) \ln\left\{1 - \frac{z}{v_o(t+t_o)}\right\} \quad , \; 0 \leq v_o t - z \leq z_o(t) \tag{B-5}$$

$$= 0 \qquad \text{elsewhere.}$$

The frequency shift at the peak ρ_{Max} is

$$\delta\omega(z) = -\frac{\partial\phi}{\partial t} = \frac{\omega_o}{3}\left[\ln\left\{1 - \frac{z}{v_o(t_M+t_o)}\right\} + \frac{z\, z_o(t_M)}{v_o(t_o+t_M)\,\{v_o t_o + z_o(t_M)\}}\right] \tag{B-6}$$

where t_M is the solution to

$$v_o t - z = z_o(t) \quad . \tag{B-7}$$

For $v_o t_o \gg z$

$$\delta\omega(z) \approx -\frac{\omega_o z \, v_2 \rho_{Max}}{v_o \ell} \tag{B-8}$$

which is in agreement with the result of reference 7.

17 May 1967

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14. This corresponds to assuming $n = n_o + n_2\overline{E^2}$ for $n_2\overline{E^2} << n_o$ in which case $v_2 = 8\pi n_2/n_o^4$.

15. The fields are still referred back to the frame fixed with the medium, in other words they are the fields measured in the fixed frame.

16. Exactly in the forward direction, the Brillouin shift is zero in liquids and also in the exact forward direction the Rayleigh-wing effect gives a mode with zero gain and one with a small loss constant of the order Ω/ω_o smaller than the corresponding stimulated Rayleigh gain for large angles, where Ω is the beat frequency. However, these components are present at small angles as well as in the backward direction where they could be reflected into the forward direction.

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FIGURE CAPTIONS

Fig. 1a. Development in distance of a Gaussian input pulse in time, $z_o = 0$, $z_1 = z_s/2$, $z_2 = z_s$. (The time axes for each curve are translated by $- z_i/v_o$).

Fig. 1b. Development in time of a Gaussian initial pulse in space. $t_o = 0$, $t_1 = t_s/2$, $t_2 = t_s$. (The pulse is shown in the frame moving with velocity v_o).

Fig. 2a. Development in distance of intensity of a sinusoidal input in time of the form $\rho = \rho_o \sin^2 \pi t/T$, where T is the modulation period. $z_o = 0$, $z_1 = z_s/2$, $z_2 = z_s$. (The time axis for each curve is translated by $- z_i/v_o$).

Here $v_2\rho_o/v_o = .1$.

Fig. 2b. Development in time of a sinusoidal initial pulse in space $t_o = 0$, $t_1 = t_s/2$, $t_2 = t_s$. (The pulse is shown in the frame moving with velocity v_o).

Fig. 2c. Development in distance of the phase for a sinusoidal input field. $z_1 = z_s/2$, $z_2 = z_s$. At $z = 0$, $\phi = 0$. (The time axis for each curve is translated by $- z_i/v_o$). ω_o is the optical frequency, T is the modulation period.

Fig. 2d. Spectrum of the sinusoidal pulse at z_1 and z_2 for $\omega_o T = 100$. The frequency is in units of half the initial modulation frequency, the even multiples being absent. At z_s the most intense peak is about 2000 wave numbers below ω_o for the case of a ruby laser.

Fig. 2e. Spectrum of the sinusoidal pulse at z_1 and z_2 for $\omega_o T = 500$. The frequency is in units of half the initial modulation frequency, the even multiples being absent. At z_s, the most intense peak is about 2000 wave numbers below ω_o for the case of a ruby laser.

Fig. 3a. Propagation of a pulse in the stationary frame. The initial pulse just before entering the medium is Gaussian with the height and width normalized to unity. $\frac{v_2\rho_o}{v_o} = .2, \left(\frac{t_s v_o}{\ell} = .291\right)$. $\tau = .0017\ t_s$, $t_1 = .89\ t_s$, $t_2 = 2.61\ t_s$, $t_3 = 6.97\ t_s$.

Fig. 3b. The integrated energy density $U = \int_{-\infty}^{\infty} \rho\ dz$ as a function of time (normalized to the shock time).

Fig. 4a. Propagation of a pulse in the stationary frame. The input pulse just before entering the medium is Gaussian with the height and width normalized to unity. $\frac{v_2\rho_o}{v_2} = .2, \left(\frac{t_s v_o}{\ell} = .291\right)$. $\tau = .229\ t_s$. $t_1 = .89\ t_s$, $t_2 = 1.77\ t_s$, $t_3 = 5.22\ t_s$, $t_4 = 8.72\ t_s$, $t_5 = 12.29\ t_s$.

Fig. 4b. The integrated energy density as a function of time (normalized to the shock time).

Fig. 5a. Propagation of a pulse in the stationary frame. The initial pulse is sinusoidal of the form $\rho_o \sin^2\left(\frac{\pi z}{v_o T}\right)$. $\frac{v_2 \rho_o}{v_o} = .1 \left(\frac{t_s v_o}{\ell} = .531\right)$. $\tau = .0094\ t_s$, $t_1 = 0$, $t_2 = 1.66\ t_s$, $t_3 = 3.13\ t_s$.

Fig. 5b. The integrated energy density as a function of time (normalized to the shock time).

Fig. 6a. Propagation of a pulse in the stationary frame. The initial pulse is sinusoidal of the form $\rho_o \sin^2\left(\frac{\pi z}{v_o T}\right)$. Distance is normalized to modulation wavelengths. $\frac{v_2 \rho_o}{v_o} = .075 \left(\frac{t_s v_o}{\ell} = .707\right)$. $\tau = .354\ t_s$, $t_1 = 0$, $t_2 = 1.74\ t_s$, $t_3 = 4.56\ t_s$.

Fig. 6b. The integrated energy density as a function of time (normalized to the shock time).

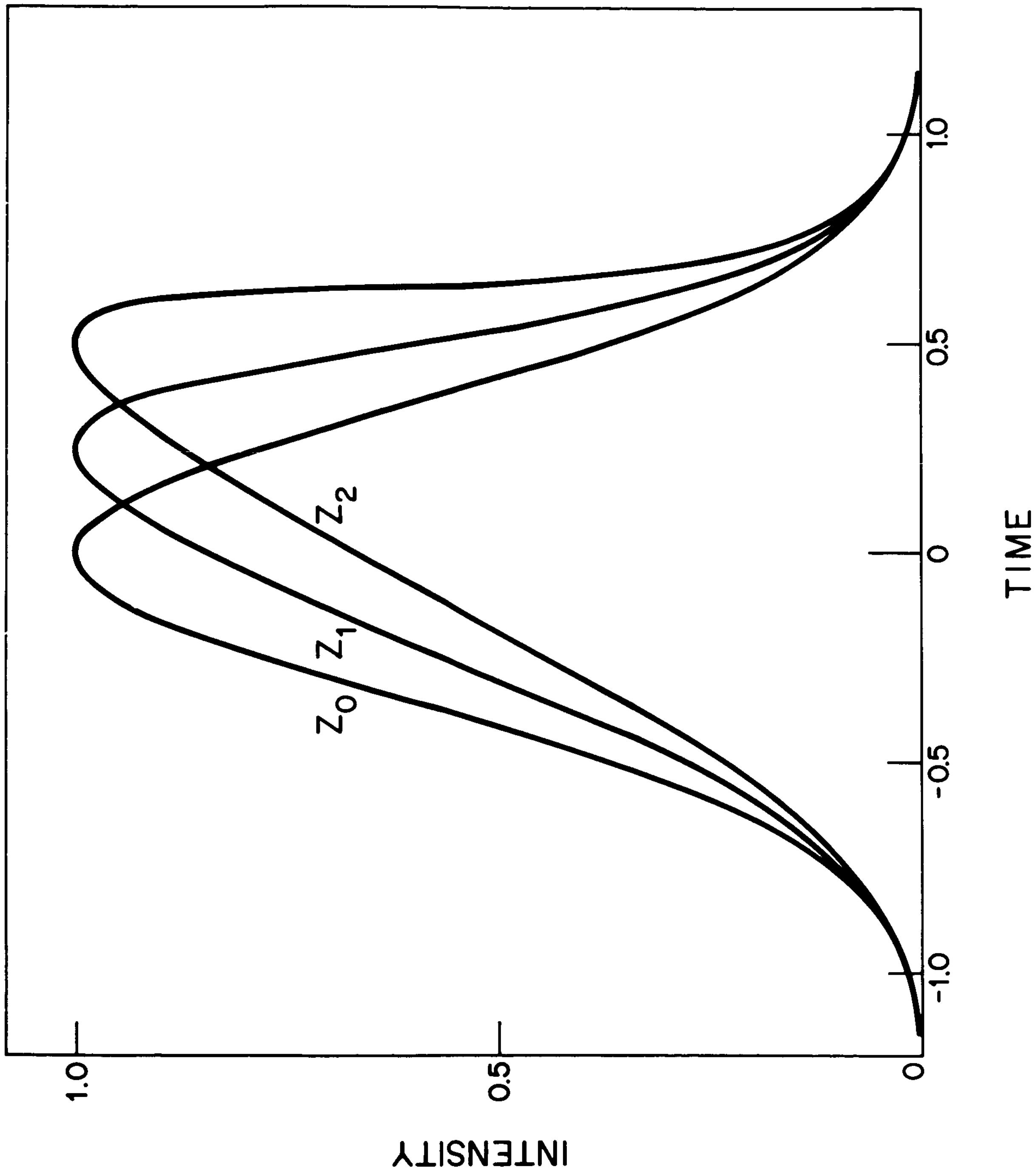


Fig. 1a

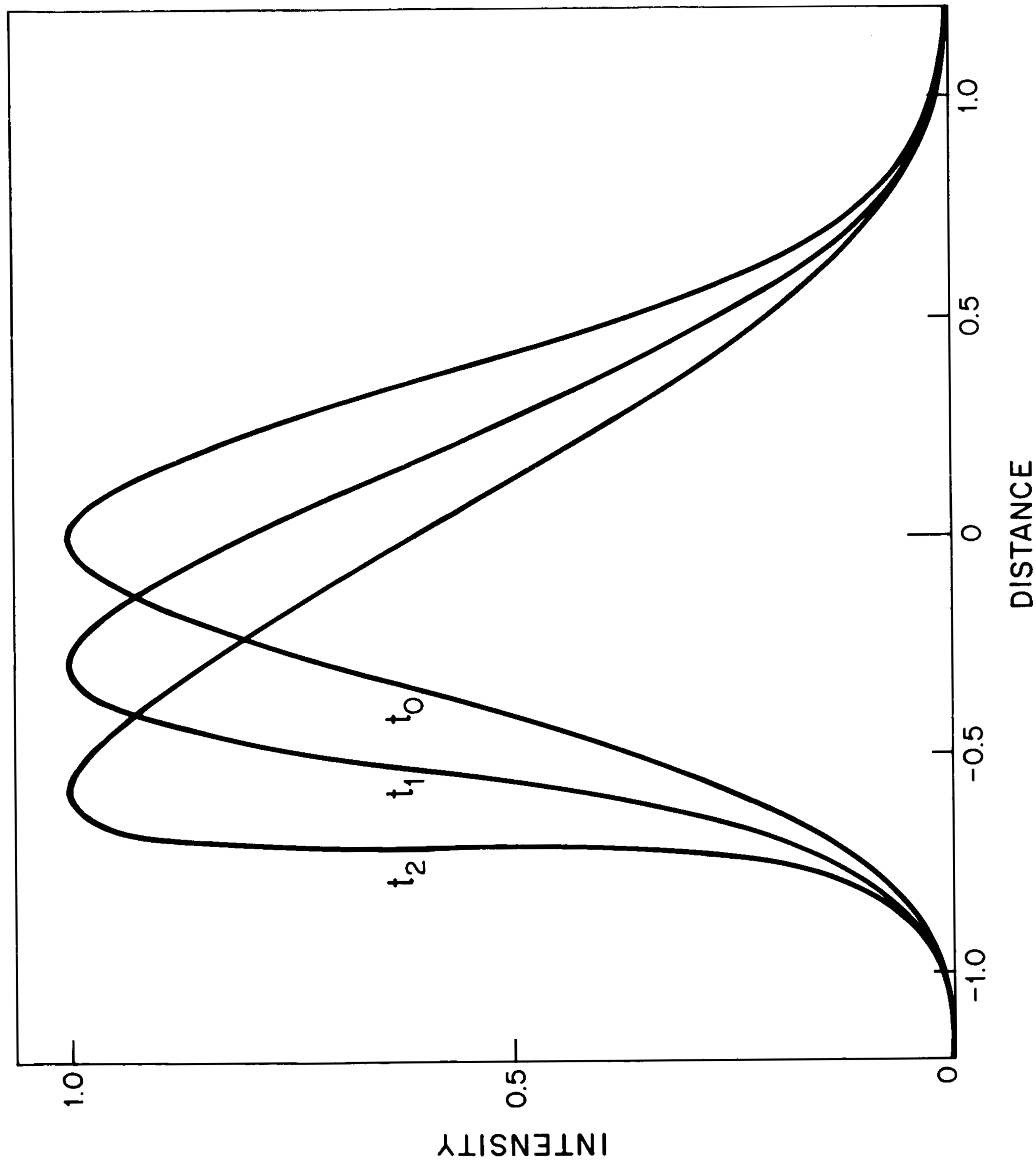


Fig. 1b

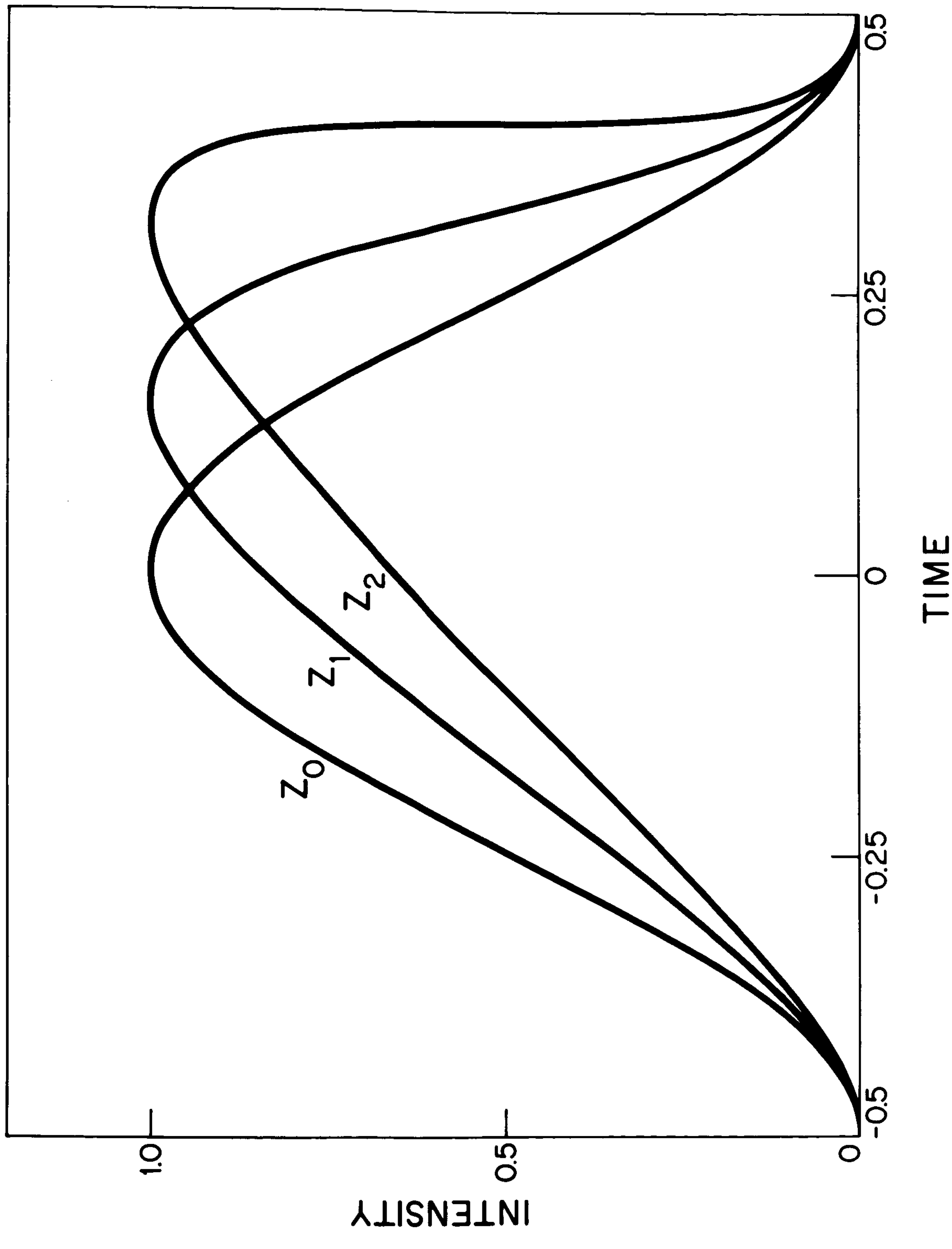


Fig. 2a

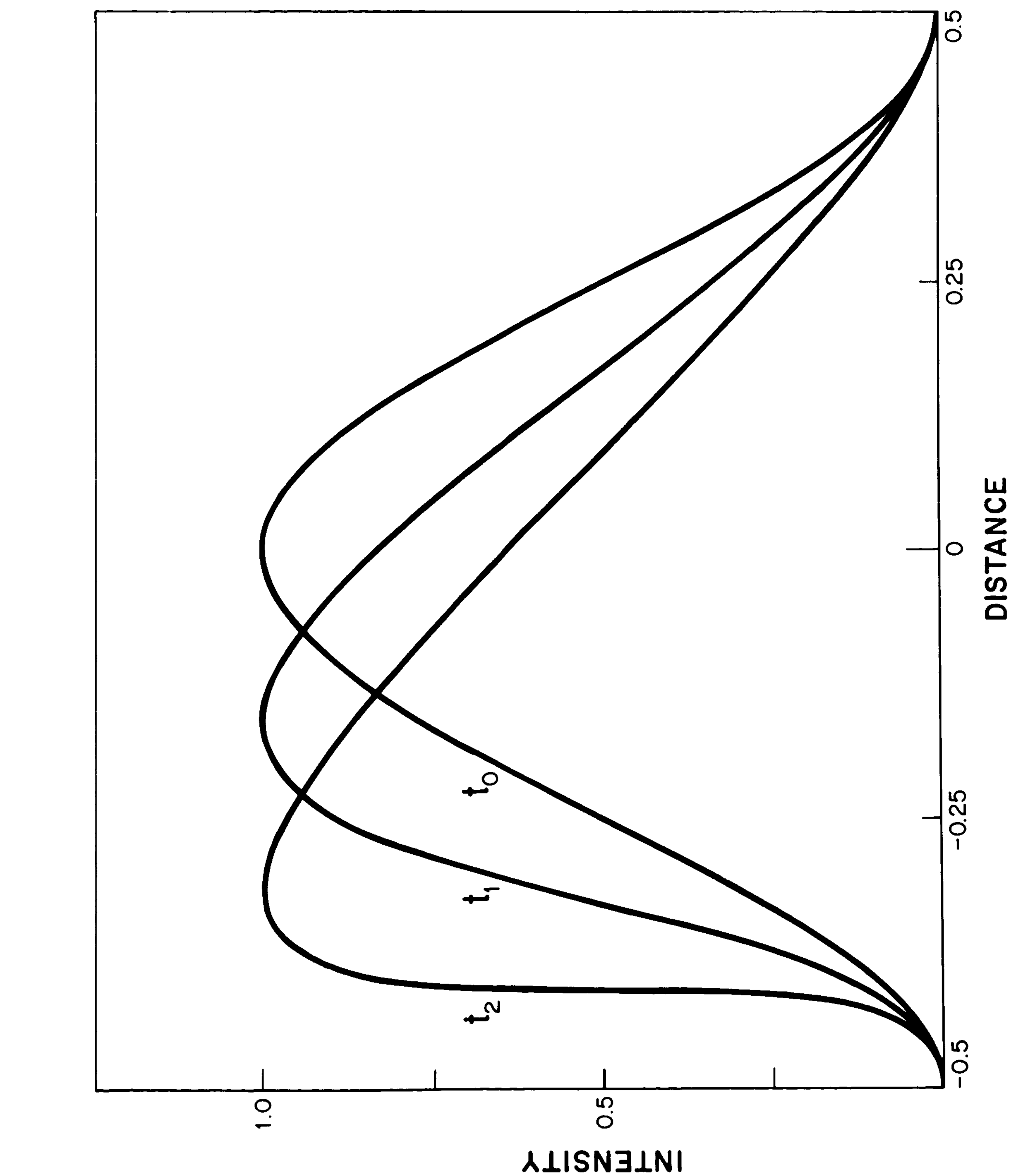
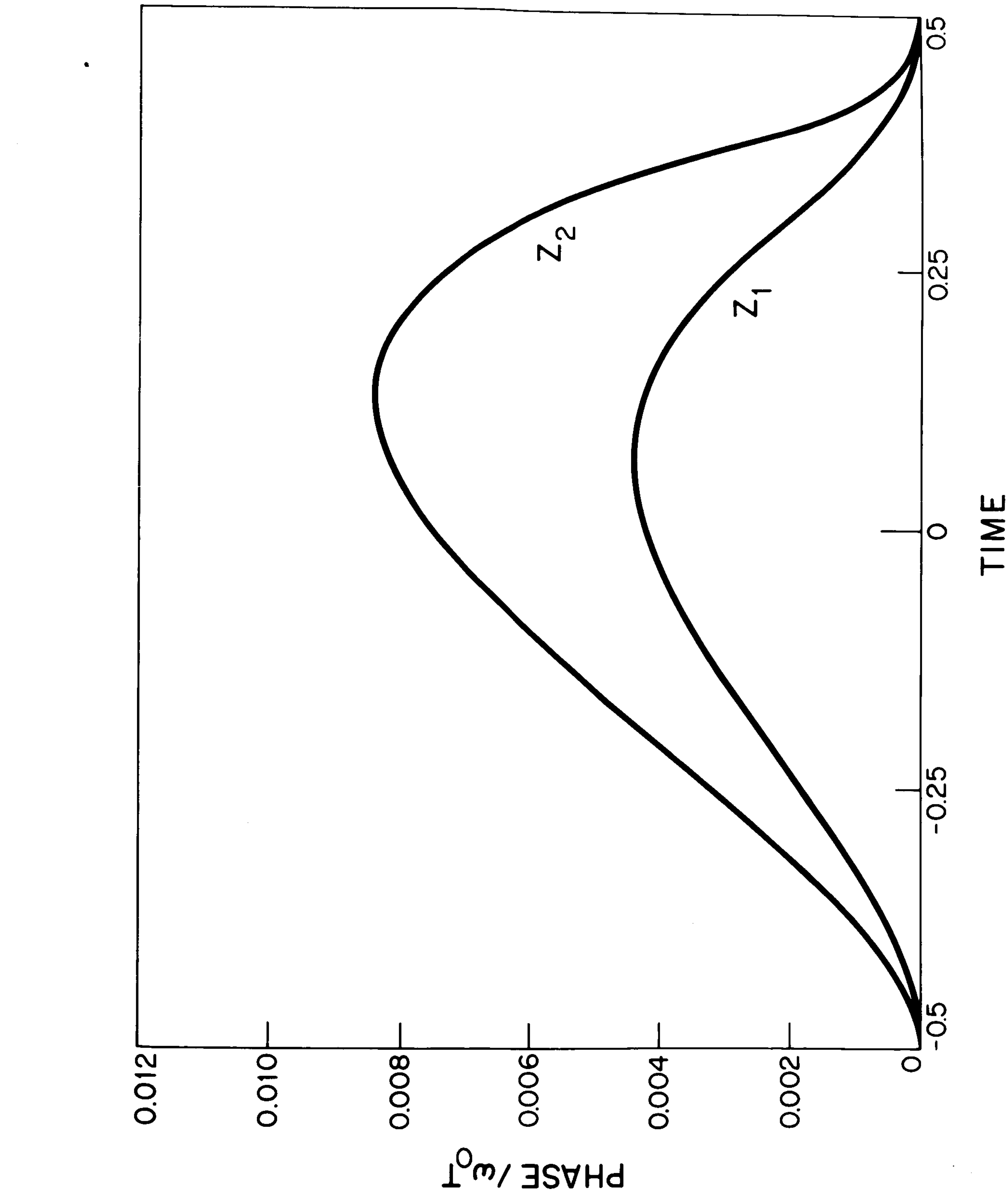


Fig. 2b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0.010
0.008
0.006
0.004
0.002
0
-0.5
-0.25
0
0.25
0.5
TIME
Z2
Z1

Fig. 2c

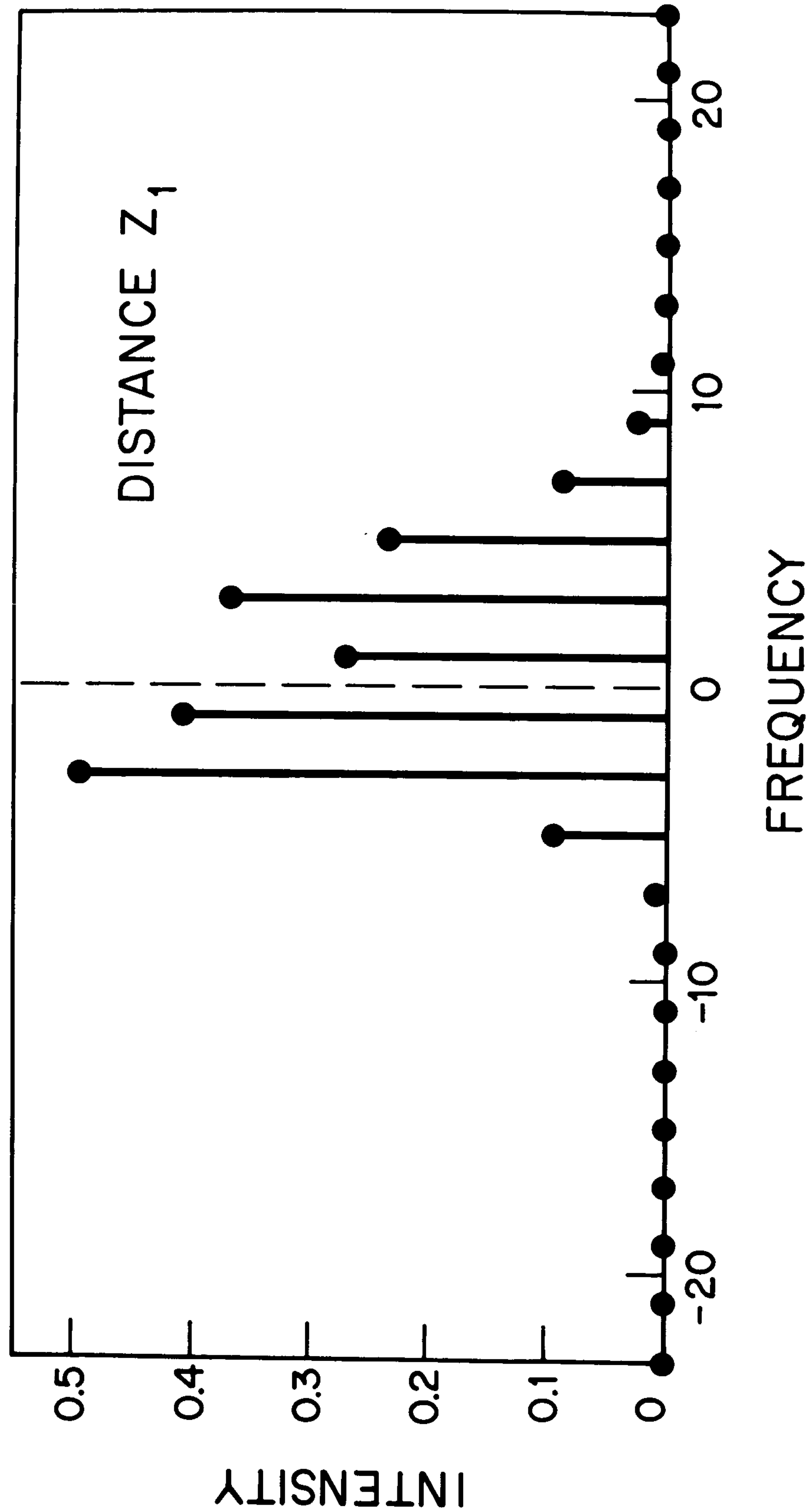


Fig. 2d

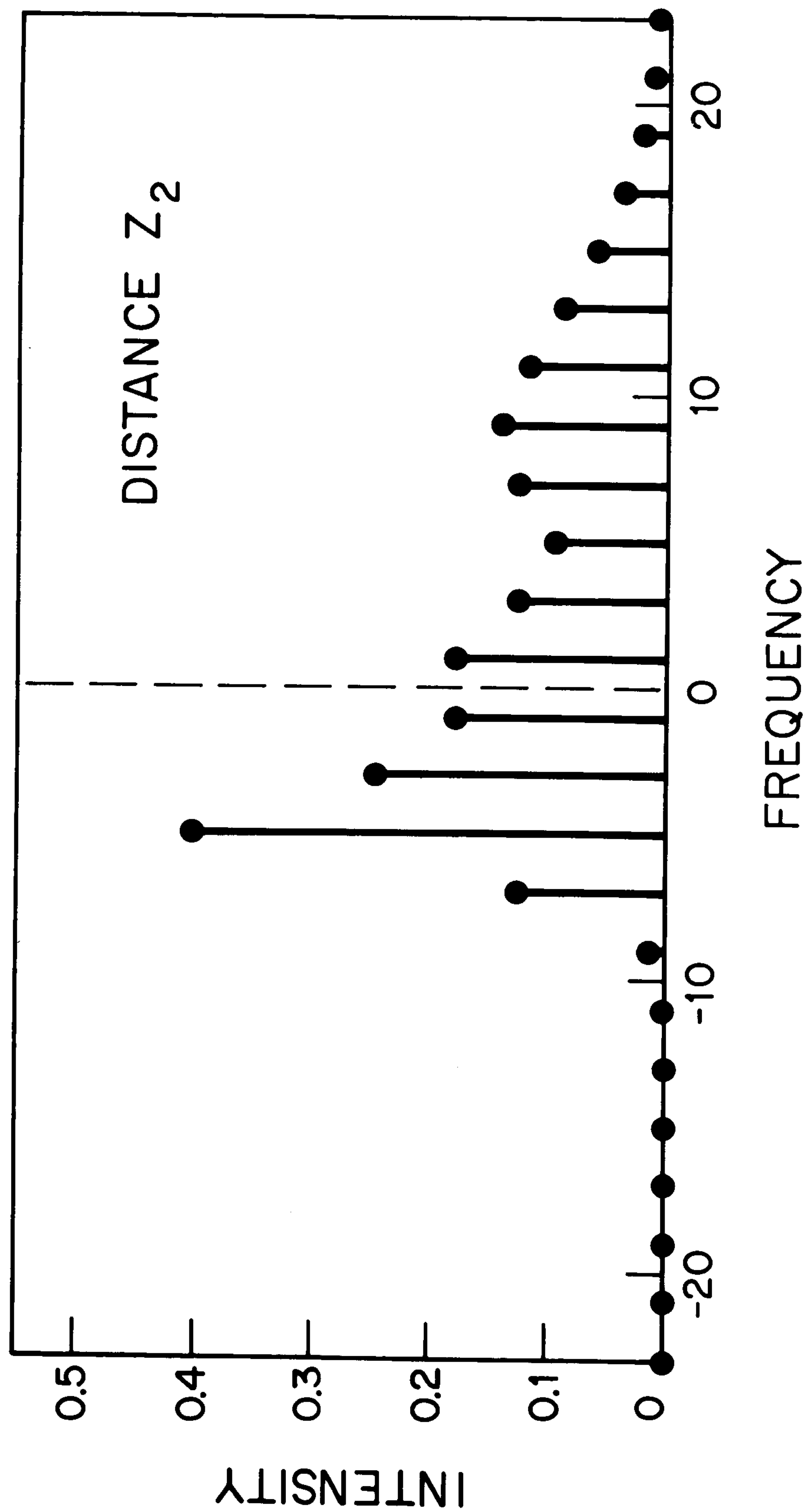
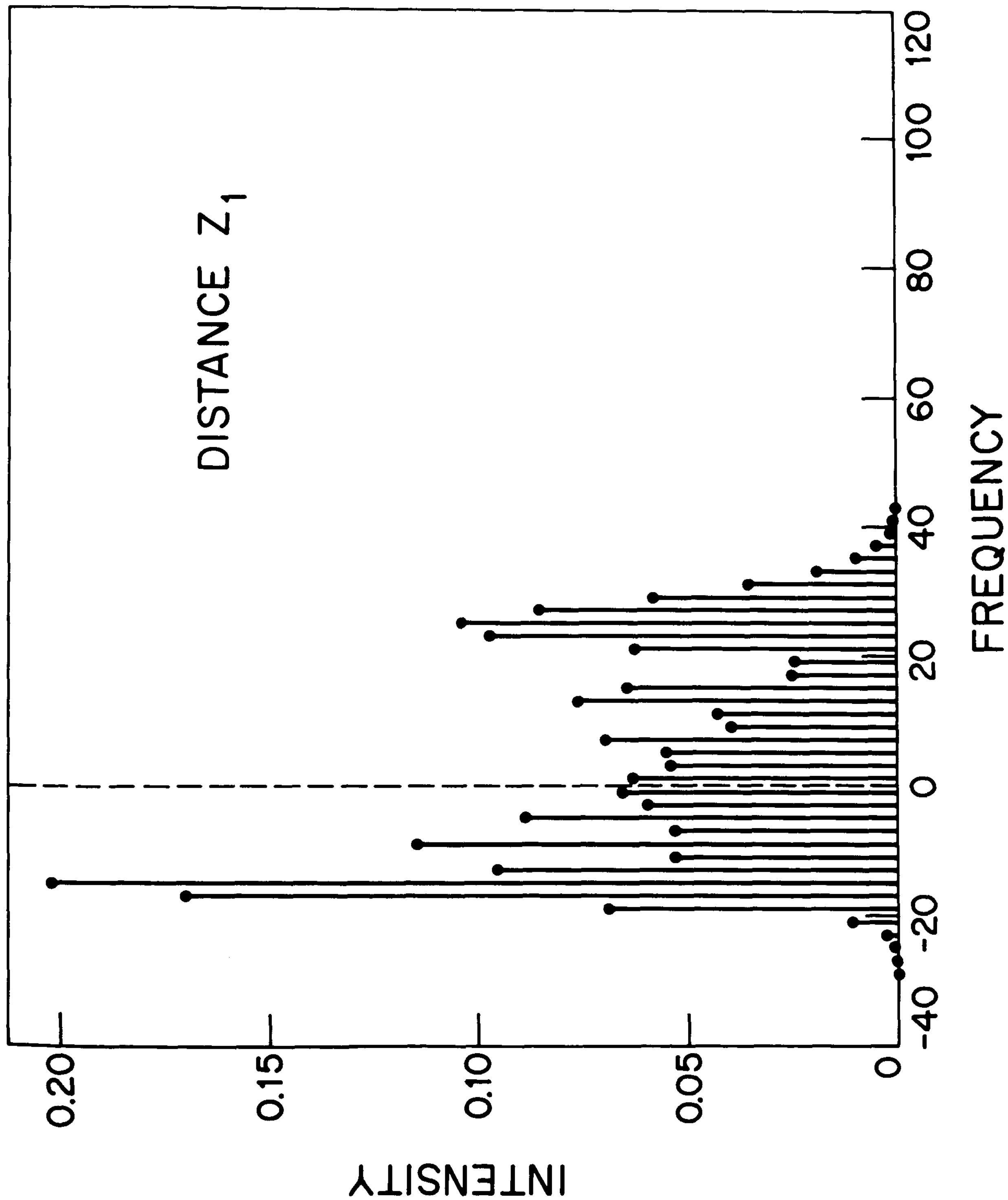


Fig. 2d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FREQUENCY
INTENSITY
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0
20
40
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80
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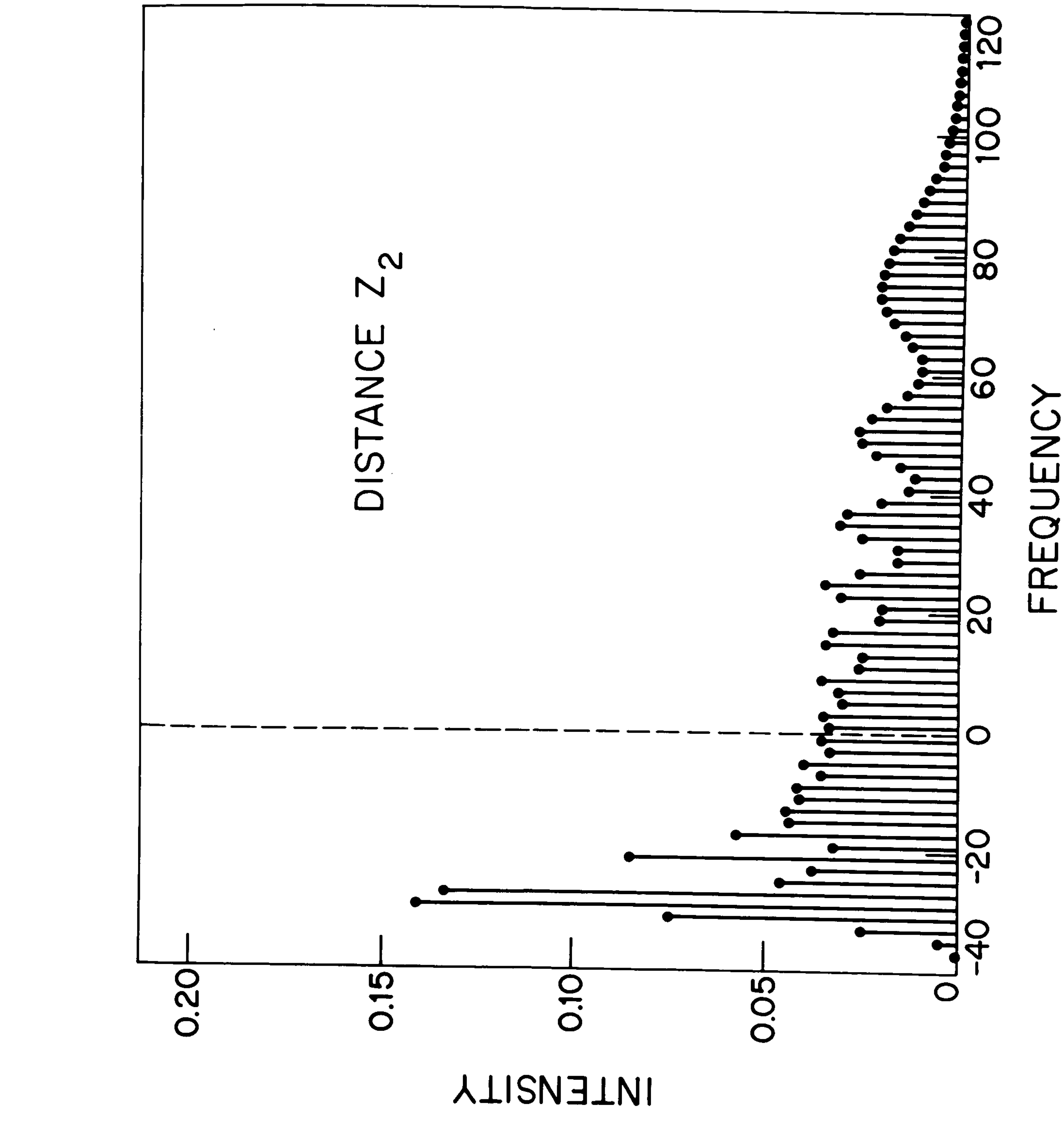
0
0.05
0.10
0.15
0.20

Fig. 2e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INTENSITY
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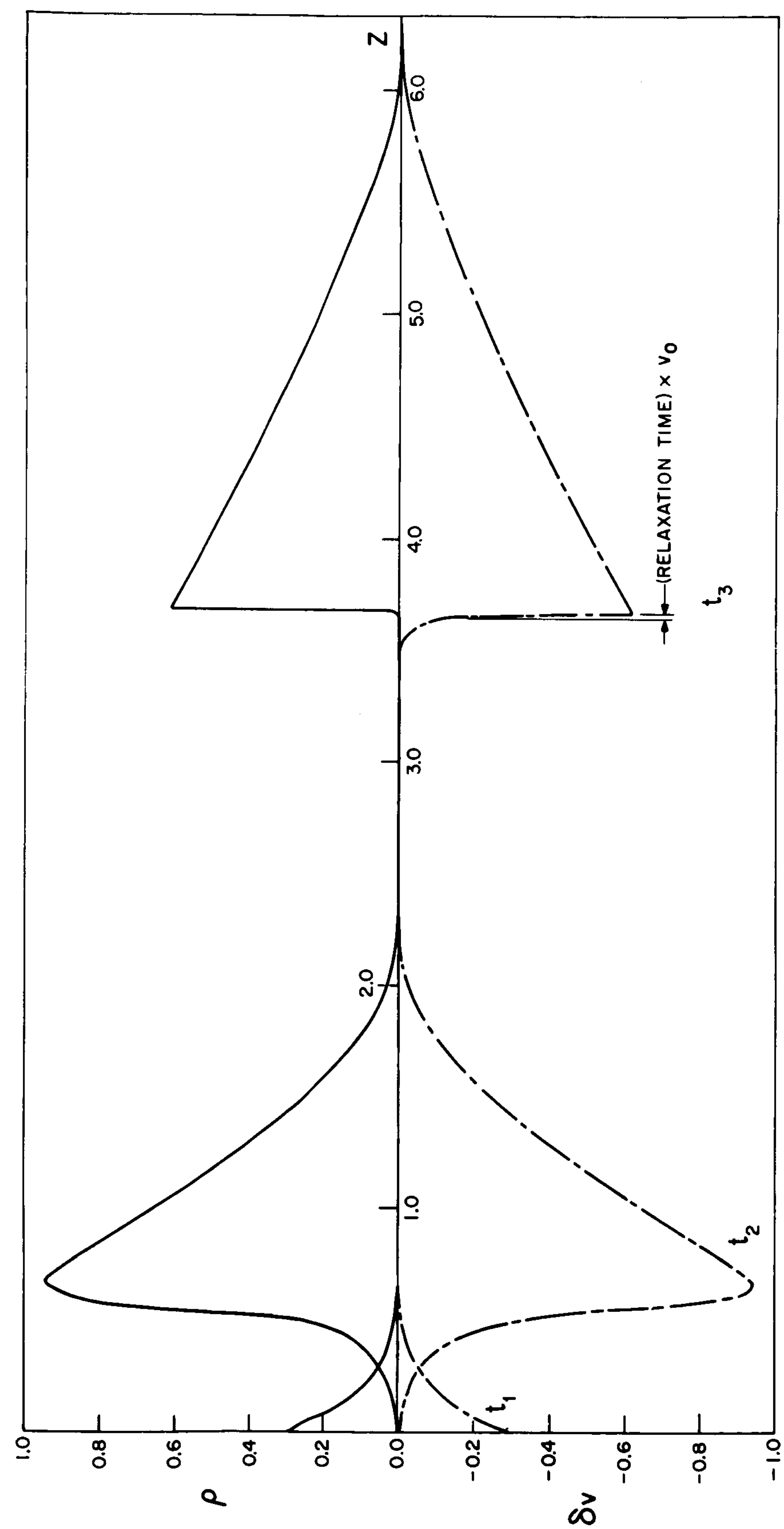
80
100
120

Fig. 2e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δv
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0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
-1.0
1.0
2.0
3.0
4.0
5.0
6.0
Z
t_1
t_2
t_3
(RELAXATION TIME) × v_0

Fig. 3a

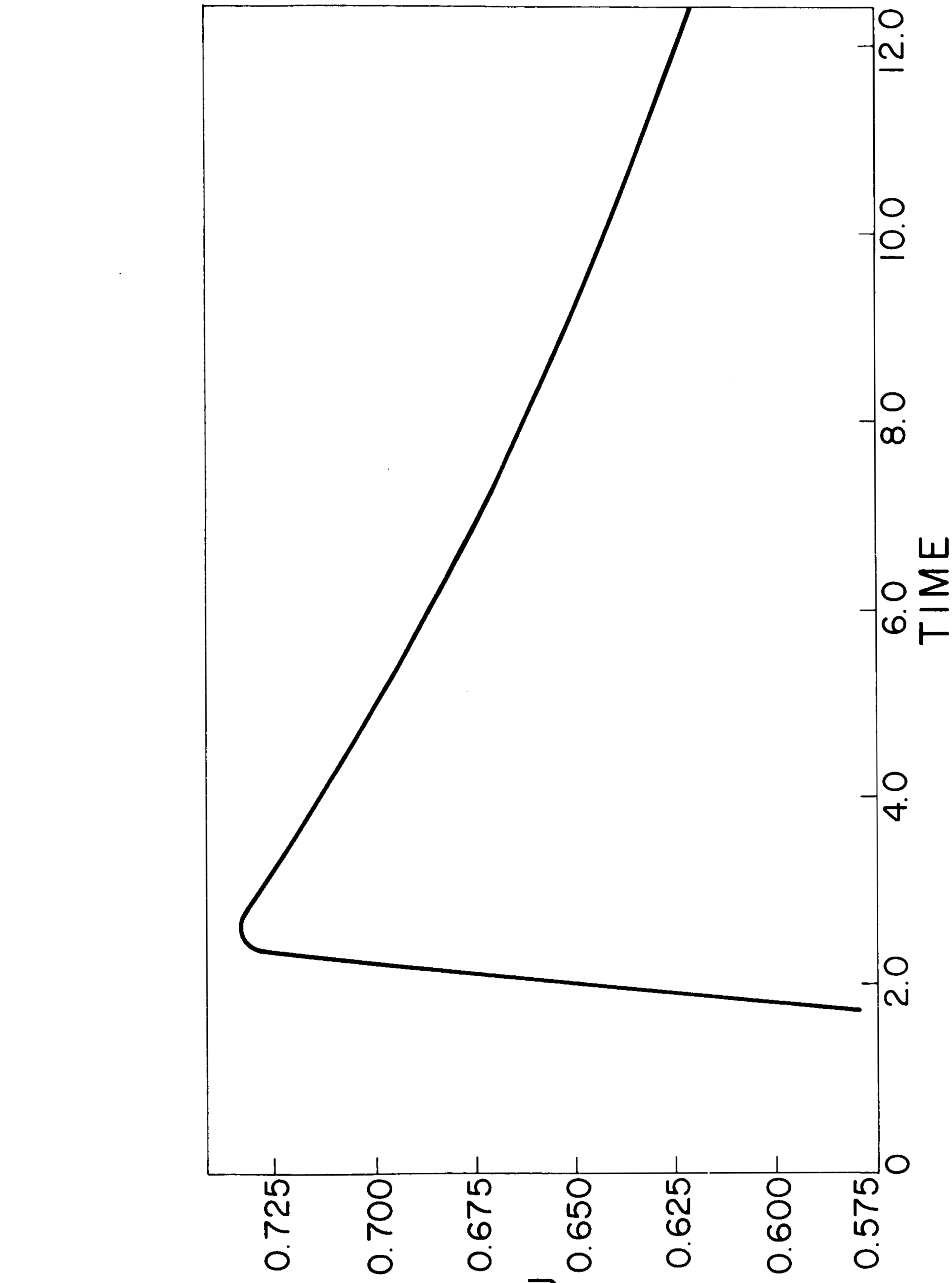
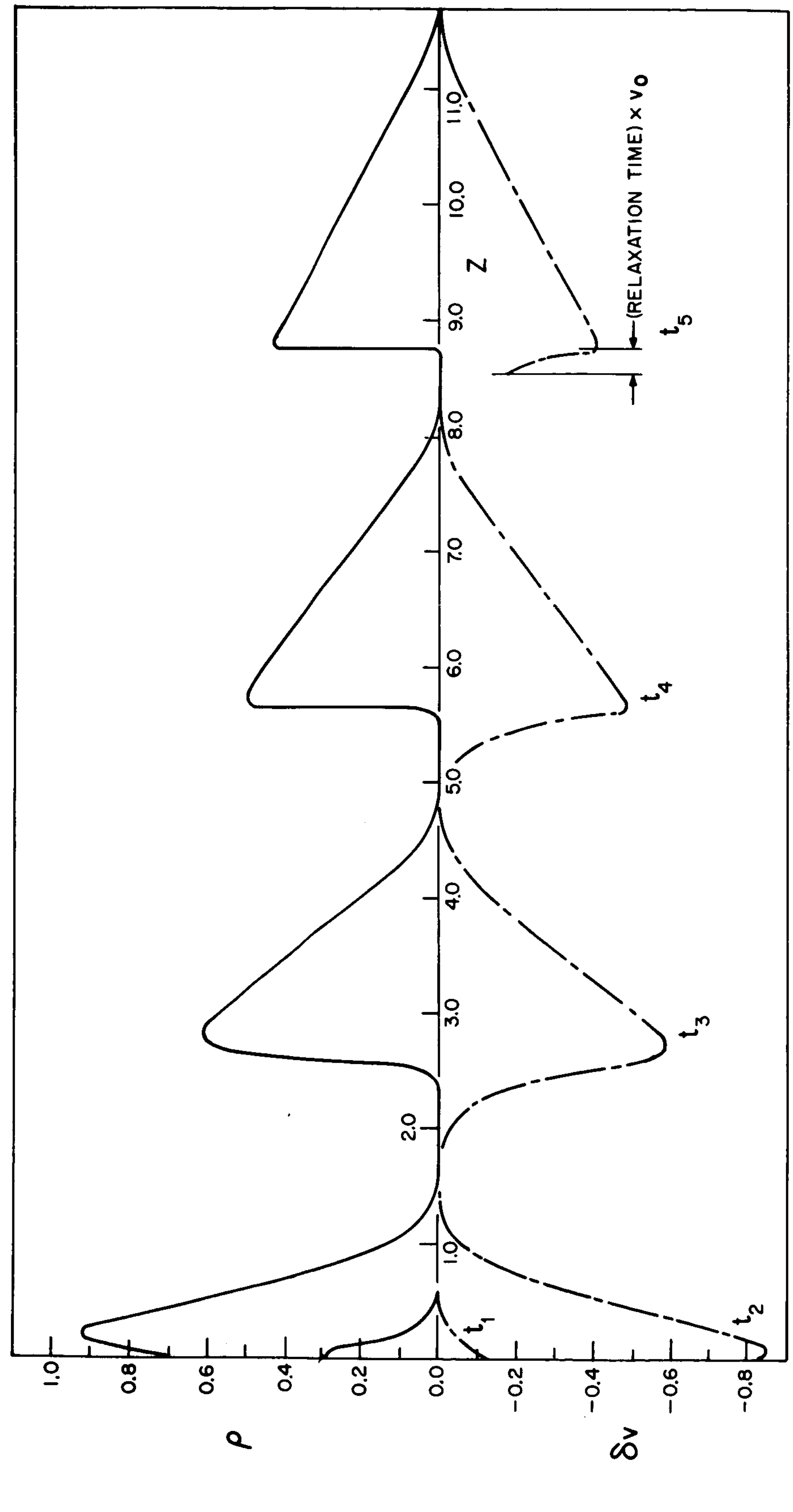
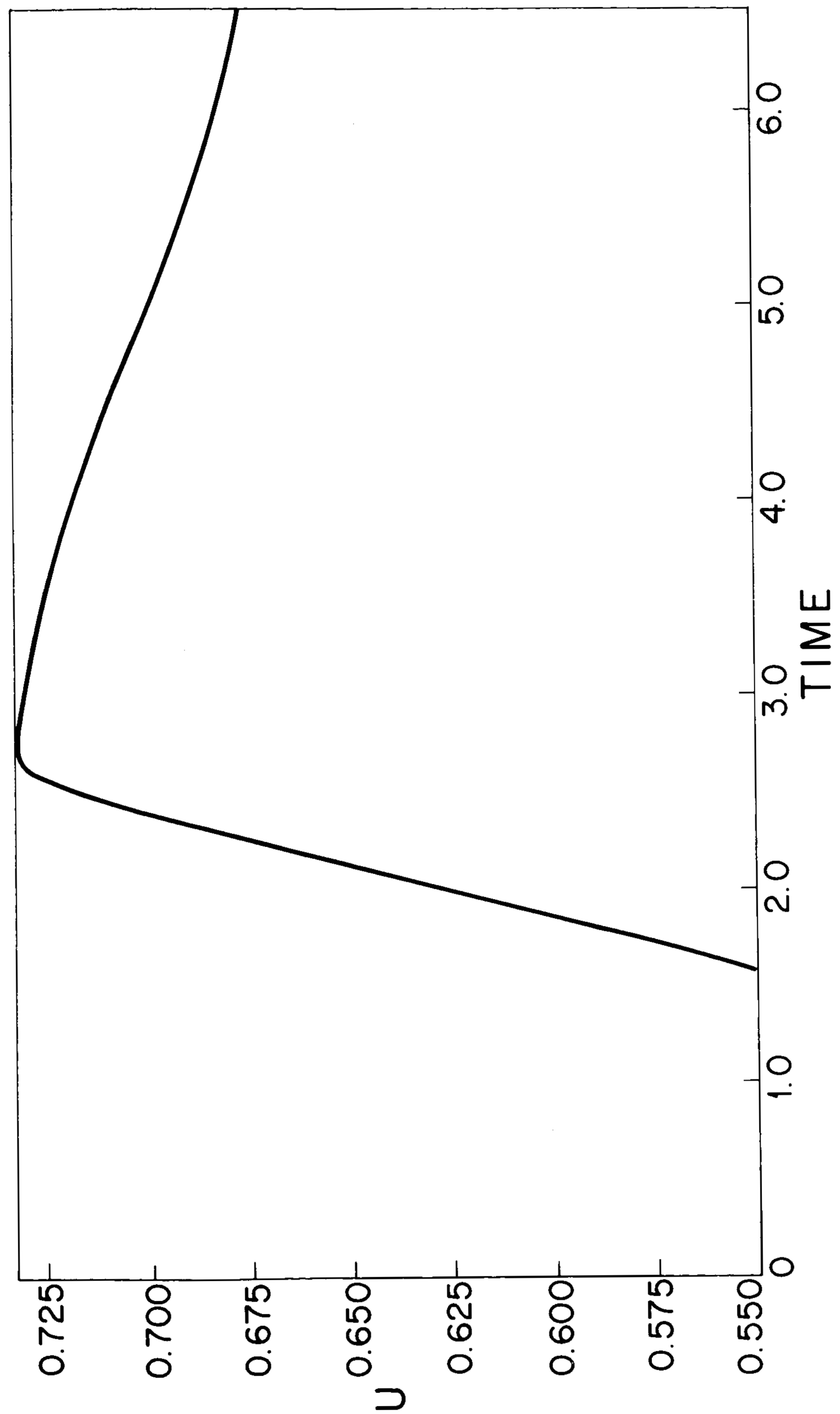


Fig. 3b



ρ
δv
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
9.0
10.0
11.0
Z
t1
t2
t3
t4
t5
(RELAXATION TIME) × v0

Fig. 4a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0.650
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0.600
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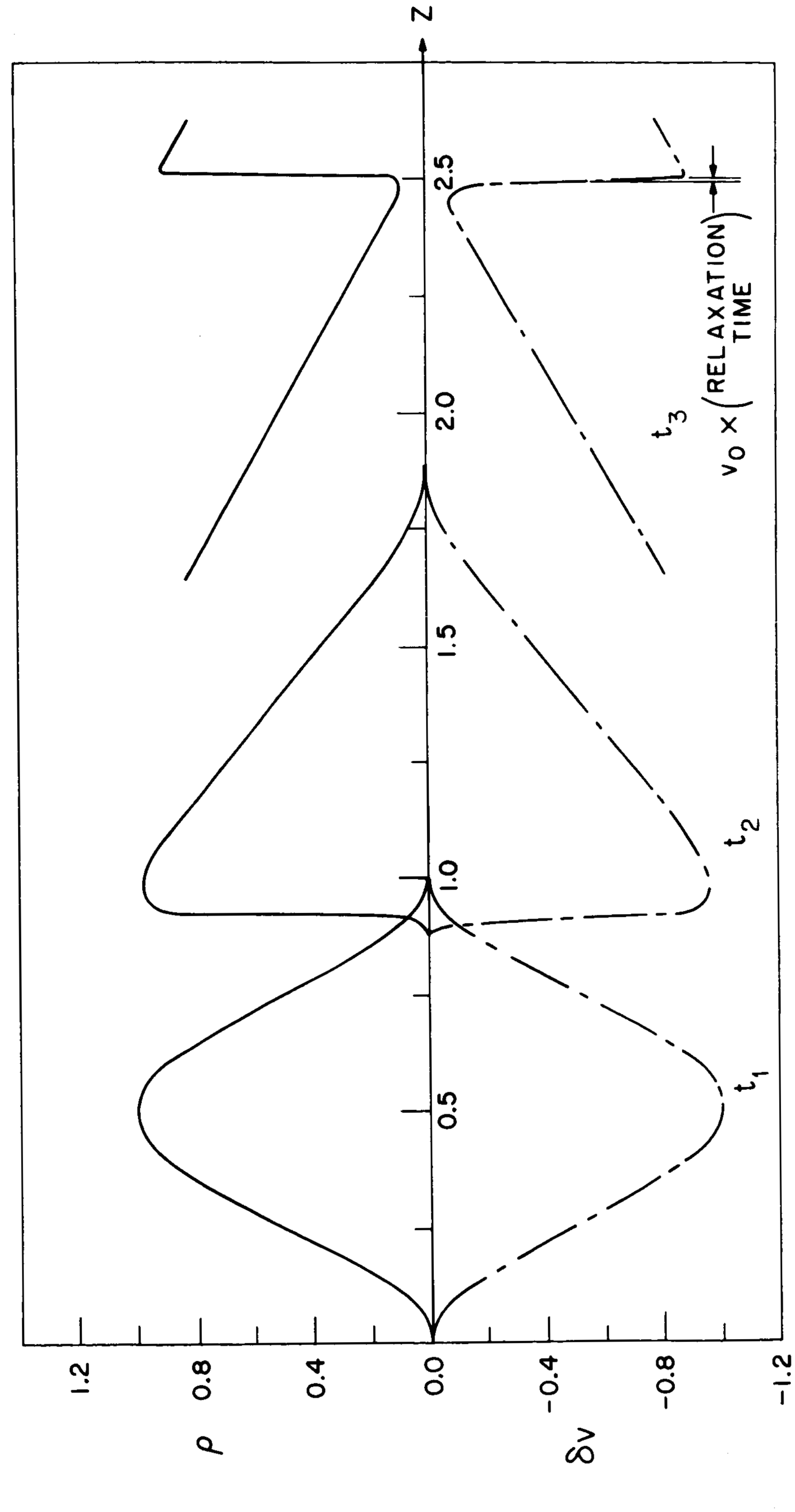
0.550
0
1.0
2.0
3.0
4.0
5.0
6.0
TIME

Fig. 4b



ρ
δv
1.2
0.8
0.4
0.0
-0.4
-0.8
-1.2
0.5
1.0
1.5
2.0
2.5
Z
t_1
t_2
t_3
$v_0 \times$ (RELAXATION TIME)

Fig. 5a

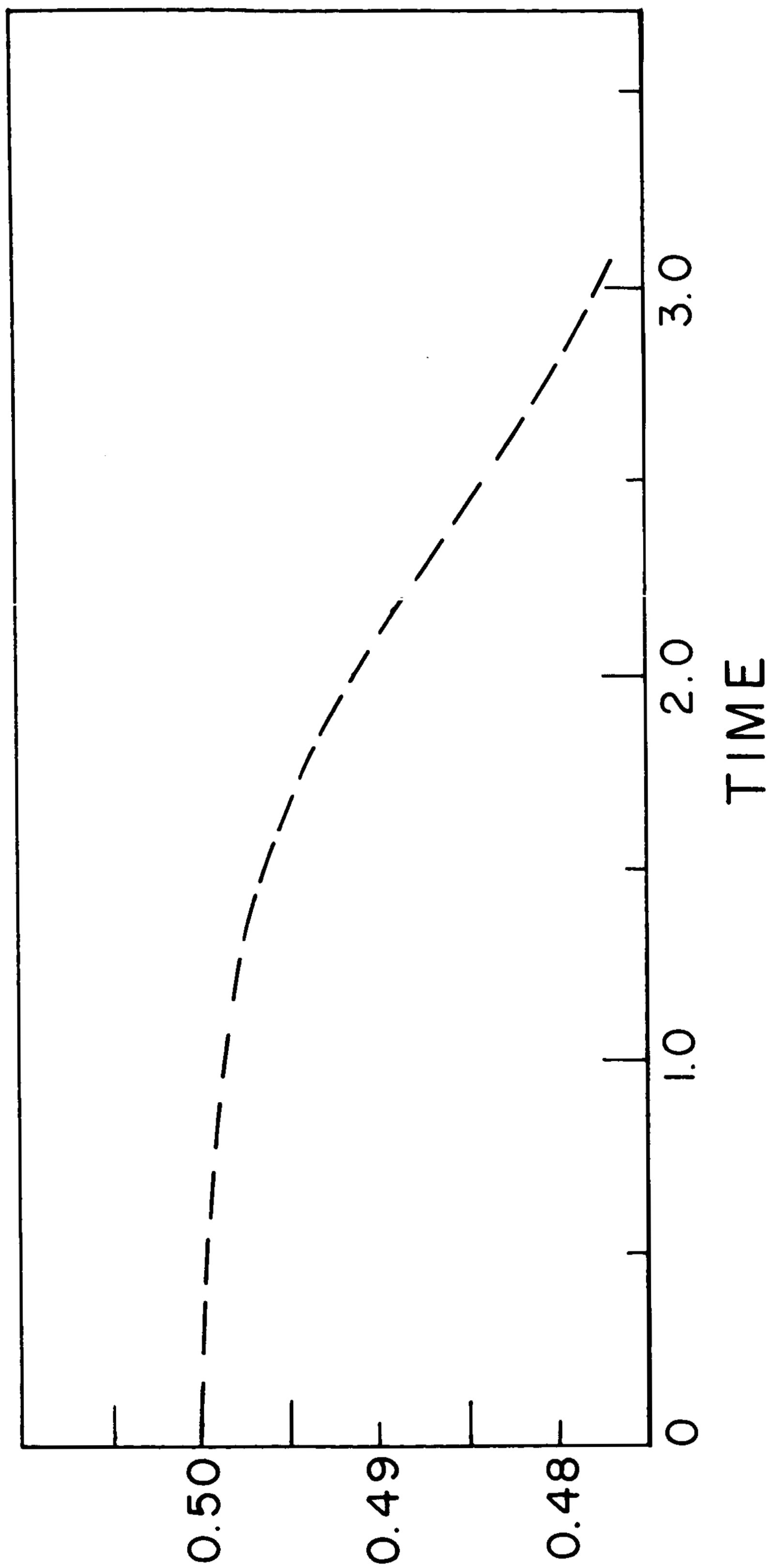
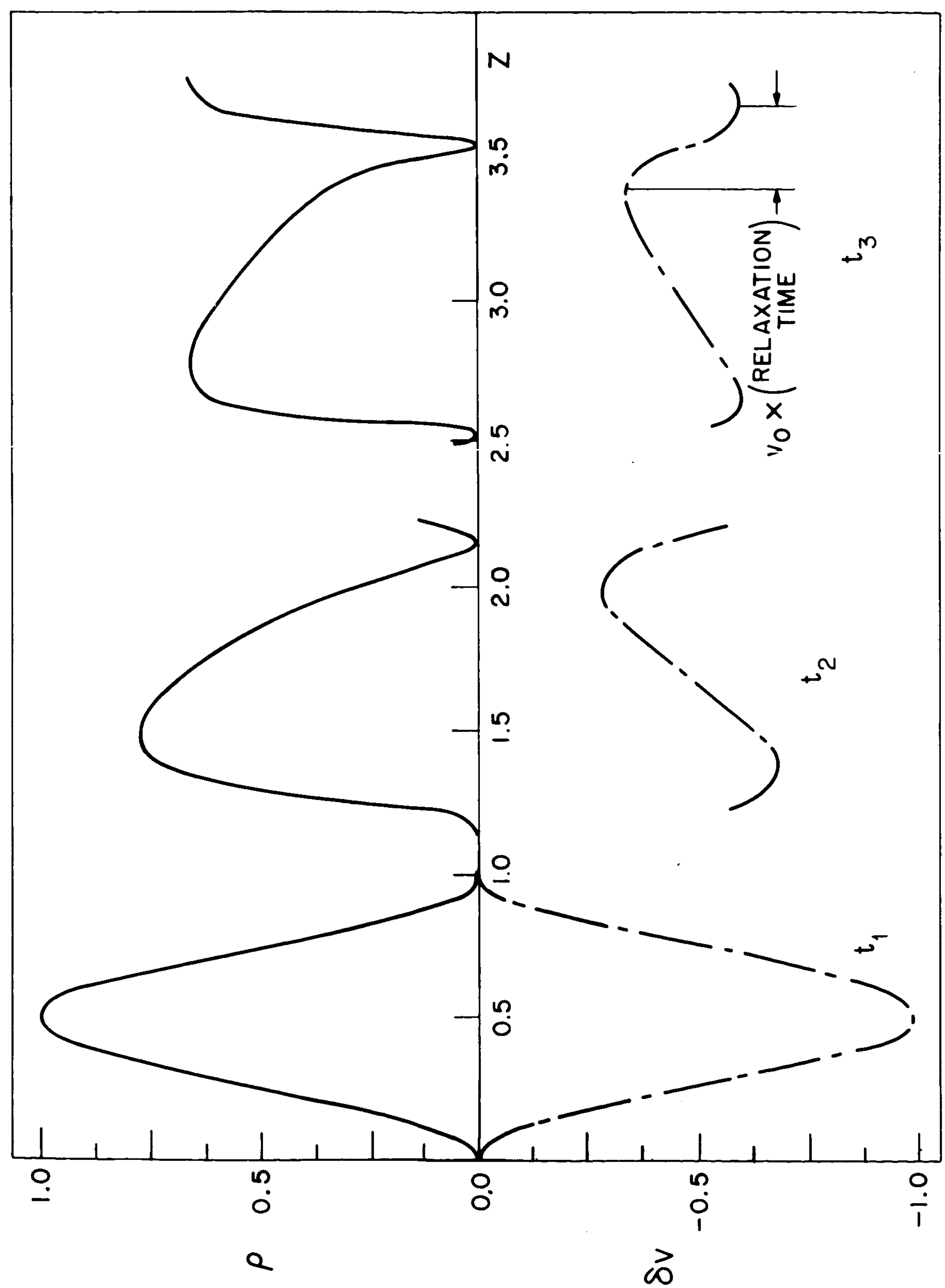


Fig. 5b



ρ
δv
1.0
0.5
0.0
-0.5
-1.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
Z
t1
t2
t3
v0 × (RELAXATION TIME)

Fig. 6a

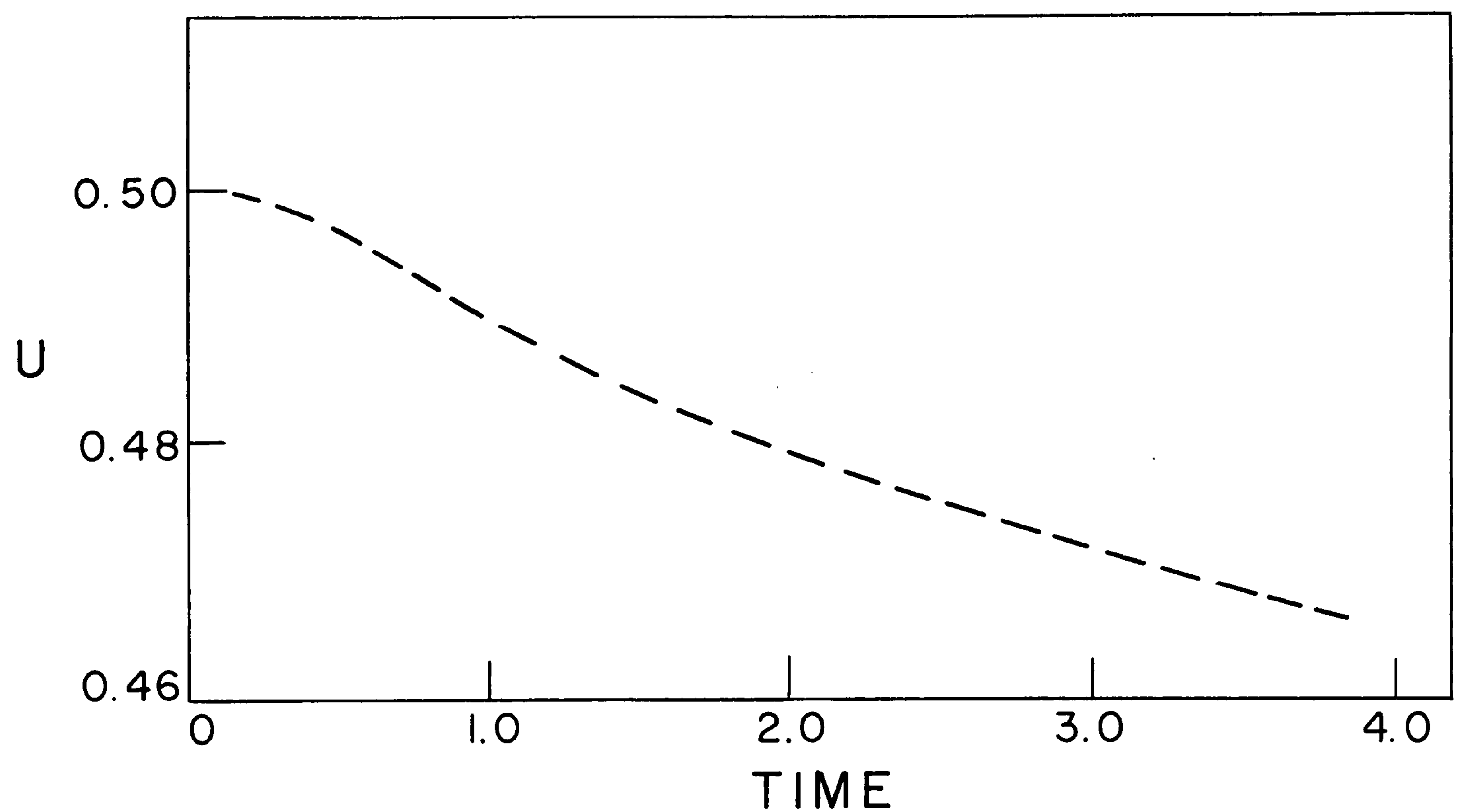


Fig. 6b