

(3.18). It is clear that according to the argument advanced at the end of Sec. III, such a nonequilibrium  $D(\vec{P}_1, t)$  will decay just in the same manner as the  $D(\vec{P}_1, t)$  discussed in Sec. III.

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PHYSICAL REVIEW A

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## Pulse-Area-Pulse-Energy Description of a Traveling-Wave Laser Amplifier

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The effects of a transverse mode, degeneracy, overlapping transitions, background losses, and transverse population variations are included in an analysis of light-pulse amplification in a system with an inhomogeneously broadened gain profile. Over a wide range of resonant gain and background loss, the steady-state pulse energy is proportional to the inverse cube of the pulse width, in accord with the observations of Frova *et al.*

In previous papers,<sup>1,2</sup> which introduce the phenomenon of self-induced transparency, the authors presented a result which describes the evolution of the "area"  $A$  of a light pulse traveling through a resonant two-level medium. In the case of a plane-wave nondegenerate inhomogeneously broadened amplifying system at resonance, the area theorem states that

$$\frac{dA}{dz} = \frac{1}{2}\alpha \sin A, \quad (1)$$

where  $\alpha > 0$  is the linear "Beer's" gain of the resonant amplifying medium and  $A$  is equal to the final Bloch tipping angle  $\theta(z)$  of the fictitious electric polarization vector at exact resonance with the frequency  $\omega$  of a light pulse propagating in the  $z$  direction. The tipping angle is defined as

$$\theta(z) = (|\vec{p}|/\hbar) \int_{-\infty}^{+\infty} \mathcal{E}(z, t) dt, \quad (2)$$

where  $\vec{p}$  is the value of the dipole-moment matrix element and  $\mathcal{E}$  is the pulse envelope of the electric field parallel to  $\vec{p}$ . When properly generalized and related to energy gain estimates, the area theorem leads to a qualitative and quantitative description of light-pulse amplification in a traveling-wave laser with an inhomogeneously broadened gain profile. We will use much of the notation and some of the results of our previous papers.<sup>1,2</sup> The picture developed here may also be applied to the description of absorbing media, and relevant equations are easily obtained by changing the sign of  $\alpha$ . Mixed systems, in which some, but not all, state populations are inverted, may be considered in a similar way, although particular examples in the analysis to follow are not typical of mixed systems. A number of workers have investigated related problems, and extensive bibliographies have

been compiled.<sup>3</sup> The reader is referred to Ref. 2 for basic ideas concerning Eq. (1). Our purpose here is to expand and elaborate on that description as it applies to light-pulse amplification.

A light pulse may be described in a useful approximation in terms of two quantities, the pulse energy and pulse width. In the case of light pulses which rise and fall smoothly and have a constant instantaneous frequency, an equivalent description may be obtained in terms of the pulse energy and pulse area  $A$ , where  $A$  is proportional to the time integral of the electric field envelope  $\mathcal{E}$  of the light pulse. Such a description is necessarily incomplete, because the shape of a traveling-wave pulse is not calculated. Indeed, the procedure outlined here is by itself incapable of yielding complete and exact results. However, the simplicity of this approach makes it easy to include effects due to finite transverse-mode diameters, back-ground losses, and degenerate or overlapping two-level transitions.

A linear analysis leads to the conclusion that an unmodulated weak light pulse will often become modulated if the pulse width  $\tau$  is shorter than, or even comparable to, the inverse resonant amplifying bandwidth  $T_2^*$ . The same result is true in the nonlinear case,<sup>4</sup> at least for pulses of some shapes and widths shorter than about  $T_2^*$ . If  $\theta > \pi$  and  $\tau \gg T_2^*$ , pulse breakup is expected<sup>1,2,4</sup> to occur in a nondegenerate plane-wave amplifier because the electric polarization vector would display a time modulation of absorbed energy in a manner similar to that which would occur in the optical nutation effect.<sup>5</sup> Such breakup effects will be seen to be considerably less effective when transverse-mode and quantum-level degeneracy effects are taken into account. In the region of interest here, when  $\tau$  is greater than and nearly equal to  $T_2^*$  and  $\theta$  is less than  $\pi$ , but not small, the question of whether pulse frequency or amplitude modulation effects occur is quite important. At present, we have no complete answer to this question. The following analysis has a formal meaning but is of diminished relevance to theory and experiment if the light pulses are highly modulated. The discussion is, therefore, restricted to cases for which modulation effects are not important. We suspect, without real proof, that modulation effects are not important for pulses with temporal widths longer than some time of order  $T_2^*$ . That pulses can be shorter than  $T_2^*$ , and yet the following analysis apply, is supported by the work of Hopf and Scully<sup>4</sup> if one merely requires the qualitative prediction of pulse shortening. Interest has been focused on the pulse shape<sup>6</sup> formed by a long amplifying medium. However, without knowledge of limiting pulse shapes in the more general case considered here,

the importance of modulation effects is not easily estimated.

#### SINGLE-TRANSVERSE-MODE LIGHT PROPAGATION

The electric field  $\vec{E}$  of a light pulse traveling in the positive  $z$  direction may be expressed<sup>7</sup> as a sum over transverse modes:

$$\vec{E}(x, y, z, t) = \sum_m \mathcal{E}_m(z, t) \vec{\xi}_m(x, y, z) e^{+i(\omega t - kz)} + \text{c. c.}, \quad (3)$$

where  $\mathcal{E}_m$  is the pulse envelope for the  $m$ th mode described by the generally complex vector mode function  $\vec{\xi}_m(x, y, z)$ ,  $\omega$  and  $k$  denote, respectively, the central light frequency and wave vector, and c. c. is the complex conjugate term.

In many situations, a light pulse can be forced to exist in essentially one mode. For example, at microwave frequencies, a suitable choice of waveguide dimensions can result in a broad frequency range over which all transverse modes but one are highly attenuated. Selective diffraction losses can restrict otherwise free space-light propagation to single-transverse-mode propagation. It can be assumed that a structure exists which forces the electromagnetic field to exist in only one mode. For a laser operating in a single transverse mode, mode selection is done by the mechanical parts (e. g., finite aperture plasma tube) which comprise the laser. The light scattered into other transverse modes is attenuated in a short distance because such modes are highly damped or because the frequency  $\omega$  is below the higher-mode cutoff frequencies, and light of measureable intensity can only persist in the operating transverse mode.

The electric field may then be accurately expressed by

$$\vec{E}(x, y, z, t) = \mathcal{E}(z, t) \vec{\xi}(x, y, z) e^{+i(\omega t - kz)} + \text{c. c.}, \quad (4)$$

where  $\mathcal{E}$  and  $\vec{\xi}$  refer to the selected mode and  $\vec{\xi}$  varies only slightly for a change of  $z$  equal to a light wavelength  $\lambda$ . The operating mode function  $\vec{\xi}$  is orthogonal to other mode functions<sup>7</sup>:

$$\int dx dy \vec{\xi}(x, y, z) \cdot \vec{\xi}_m^*(x, y, z) = 0 \quad (5)$$

and satisfies the transverse equation

$$\left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \gamma^2 \right) \vec{\xi}(x, y, z) = 0, \quad (6)$$

so that  $k^2 = n^2 \omega^2 / c^2 - \gamma^2$ , where  $\epsilon$  is the host dielectric constant,  $n = +\sqrt{\epsilon}$ ,  $c$  is the vacuum value of light velocity, and  $\gamma$  is the mode eigenvalue. Boundary conditions and the value of  $\epsilon$ , which may depend on transverse position, are chosen so that  $\vec{\xi}$  is identical with the actual mode of operation. There

may be transverse or nonlinear effects which modify the above statements, but the fundamental assumption, upon which the following is based, is that Eq. (4) is accurate. It may be, because of unprescribed transverse effects, that  $\vec{\xi}$  does not have precisely some preconceived form (e. g., Gaussian). We assume that Eq. (4) is accurate, but insist that  $\vec{\xi}$  is to be determined by experimental conditions. The above is merely a mathematical device for properly choosing orthogonal transverse-mode functions.

Assuming that the background refractive index is dispersionless and that  $\alpha^{-1} \gg \lambda$ , Eq. (4) and Maxwell's equations prescribe an equation for the forward-traveling wave:

$$\begin{aligned} \frac{\partial \mathcal{E}}{\partial z} + \frac{n^2 \omega}{kc^2} \frac{\partial \mathcal{E}}{\partial t} \\ = i \frac{2\pi\omega}{\eta c} L^{-1} \int dx \int dy \vec{\mathcal{P}}(x, y, z) \cdot \vec{\xi}^*(x, y, z) \\ - \left( \frac{1}{2} \sigma' + L^{-1} \int dx \int dy \vec{\xi}^*(x, y, z) \cdot \frac{\partial \vec{\xi}(x, y, z)}{\partial z} \right) \mathcal{E}, \end{aligned} \quad (7)$$

where  $\eta = ck/\omega$  and  $L = \int dx dy |\vec{\xi}^*(x, y, z)|^2$ . The electric field  $\mathcal{E}(z, t)$  at the mode center is determined by the summation of source dipoles throughout the beam cross section because of diffraction. A loss term  $\frac{1}{2} \sigma' \mathcal{E}$  is introduced to include the effect of background broad-band linear loss (e. g., diffraction or conductivity losses). The polarization  $\vec{\mathcal{P}}$  in the laboratory reference frame is given by

$$\vec{\mathcal{P}}(x, y, z, t) = \vec{\mathcal{P}}(x, y, z, t) e^{+i(\omega t - kz)} + c. c. \quad (8)$$

Use of the assumption  $\alpha^{-1} \gg \lambda$  and the condition that the pulse spectral width is small compared to  $\omega$  allow the neglect of terms involving higher derivatives of  $\mathcal{E}$  and  $\vec{\mathcal{P}}$ .

In Eq. (7), the term involving  $\partial \vec{\xi} / \partial z$  refers to a transverse mode which changes with distance  $z$ . The term is included to allow a description of amplifiers in which the light is converging or diverging. The unimportant imaginary part of the term may be removed from the problem by redefining the phase  $kz$  in Eq. (4) to be  $kz + \Phi$ , where  $d\Phi/dz$  is equal to the imaginary part of the  $\partial \vec{\xi} / \partial z$  term. The remaining real part of the term may

be grouped with the background-loss parameter  $\sigma'$  to form an effective-loss parameter  $\sigma$ , which, in general, depends on distance  $z$ . It should be noted that  $\sigma$  may be positive, negative, or zero.

With Bloch's equations<sup>2,8</sup> describing the polarization  $\vec{\mathcal{P}}$ , the description of the system is formally complete. Derivations and results recorded in our earlier work will be recorded as needed.

#### GENERALIZED AREA THEOREMS

Results for a nondegenerate two-level system will be developed, and the effects of degeneracy or overlapping transitions will be included later. The gain profile is assumed to be symmetrical about  $\omega$ , so that the analysis is consistent with the assumption that the light pulse is not frequency modulated. All homogeneous relaxation times are assumed to be long compared to the light-pulse width.

At a particular point  $(x, y, z)$  in the light-pulse beam, the irradiating electric field  $\vec{\xi}(x, y, z)\mathcal{E}(z, t)$  causes the macroscopic Bloch vector of dipoles at exact resonance to tip to a final position at an angle

$$\theta(x, y, z) = A \frac{|\vec{\xi}^*(x, y, z) \cdot \hat{p}|}{|\vec{\xi}^*(0, 0, z) \cdot \hat{p}|} \quad (9)$$

away from their original direction.<sup>8</sup> The pulse area  $A$  is defined to be equal to the tipping angle in the beam center  $x=y=0$ , and  $\hat{p}$  is a unit vector parallel to the vector dipole-moment matrix element of the transition involved. The time integral  $\int_{-\infty}^{+\infty} \vec{\mathcal{P}}(x, y, z, t) dt$  of the envelope of the induced polarization  $\vec{\mathcal{P}}(x, y, z, t)$  at a point  $x, y, z$ , is shown in the proof<sup>2</sup> of Eq. (1) to be proportional to the sine of the tipping angle  $\theta(x, y, z)$ . It follows that the contribution to the pulse-area derivative  $dA/dz$  from the region around  $(x, y, z)$  is proportional to  $\sin\theta(x, y, z)$ . This term is multiplied by the initial local-population inversion density  $N_p(x, y, z)$  at frequency  $\omega$ , and by a factor  $|\vec{\xi}^*(x, y, z) \cdot \hat{p}|$ , which describes the projection of radiation from  $\vec{\mathcal{P}}(x, y, z, t)$  at a point  $(x, y, z)$  back onto the operating transverse mode. These comments follow directly from the application of Eq. (9) to the time-integrated Maxwell equation (7).

A generalization of Eq. (1) thus follows for the case of a nondegenerate two-level single-transverse-mode amplifier:

$$\frac{dA}{dz} = \frac{\alpha}{2} \frac{\int dx \int dy N_p(x, y, z) |\vec{\xi}^*(x, y, z) \cdot \hat{p}| |\vec{\xi}^*(0, 0, z) \cdot \hat{p}|}{\int dx \int dy |\vec{\xi}^*(x, y, z) \cdot \hat{p}|^2 N_p(x, y, z)} \sin \left( \frac{|\vec{\xi}^*(x, y, z) \cdot \hat{p}| A}{|\vec{\xi}^*(0, 0, z) \cdot \hat{p}|} \right) - \frac{1}{2} \sigma A, \quad (10)$$

where  $\alpha$  is a complicated factor involving  $\vec{\xi}$ ,  $N_p$ , etc. In the limit of small pulse areas, Eq. (10) reduces to

$$\frac{dA}{dz} = \frac{1}{2} (\alpha - \sigma) A, \quad (11)$$

and  $\alpha$  is therefore the resonant Beer's gain constant, which will everywhere in this paper be defined as the experimentally obtained resonant small signal gain/cm in the operating mode at frequency  $\omega$ . Equation (11), for a pulse of width  $\tau \gg T_2^*$ , is equivalent to

$$\frac{dU}{dz} = \alpha U - \sigma' U, \quad (12)$$

where  $U$  is the total pulse energy of a weak pulse of frequency  $\omega$  and bandwidth  $\Delta\nu_p \ll T_2^{*-1}$ .

In the case of a linearly polarized mode of constant transverse Gaussian<sup>7</sup> shape

$$|\vec{\xi}(x, y)|^2 = e^{-(x^2 + y^2)/r_0^2},$$

where  $r_0$  is the mode radius, with  $N_p$  constant and  $\hat{p}$  parallel to the electric field vector, Eq. (10) reduces to

$$\frac{dA}{dz} = \alpha \frac{1 - \cos A}{A} - \frac{1}{2} \sigma A. \quad (13)$$

If each or either level of a two-level system is degenerate, a suitable representation of the degenerate states can be chosen so that transitions occur only within pairs of states, each pair consisting of one ground and one excited state. Additionally, a two-level system may have overlapping transitions due to the presence of various species of amplifying atoms. It is assumed that the light pulse has a specified time-independent polarization, either through constraints such as Brewster windows or because the medium does not tend to change the light polarization. The spectral-gain profiles for the various transitions are each symmetrical and centered about  $\omega$ , so that it is consistent to ignore frequency modulation<sup>2</sup>; an area theorem for such systems will be derived shortly.

In systems for which the spectral-gain profiles are not all symmetrical about  $\omega$ , if the pulse width  $\tau$  is long compared with the inverse bandwidth of the most narrow spectral gain curve not symmetrical about  $\omega$ , the wings of the resonance only respond dispersively. There is then no resultant frequency modulation, and the area theorem to follow may be applied to such systems.

There are systems, e. g., free atoms with substantial hyperfine interactions, in which a number of transitions not independent of one another may occur. If the hyperfine splitting energy is  $\ll \hbar/\tau$ , the hyperfine levels may be considered degenerate. If the hyperfine splitting energy is  $\gg \hbar/\tau$ , then, for a given atom, only two of the hyperfine levels will be resonant, each atom may be considered as having two levels, and the amplifying system may

be regarded as being composed of overlapping independent transitions. If the hyperfine splitting energy is comparable to  $\hbar/\tau$ , however, there may develop a nonlinear polarization which frequency modulates the pulse. In this case, the pulse area is not defined, and there is no area theorem. It will therefore be assumed that any near degeneracies are characterized by an energy splitting either large or small compared with  $\hbar/\tau$ , or that the transitions are not coupled together and may be considered to be independent of each other. Then a quantum representation can be chosen so that transitions occur only between separate pairs of states, and the resonant amplifying system may be regarded as consisting of a number of independent amplifying systems, isolated from one another except for their interactions with the light pulse.

Defining dipole-moment matrix elements  $p_0 \hat{p}_0, p_1 \hat{p}_1, \dots$ , where  $\hat{p}_i p_i$  is the dipole-moment matrix element of the  $i$ th transition and  $\hat{p}_i = \hat{p}_i(x, y, z)$  is a unit vector, an area theorem including the effects of degeneracy can be formulated,<sup>2,9,10</sup>

$$\frac{dA}{dz} = \frac{1}{2} \alpha \left[ \sum_i \int dx \int dy N_i(x, y, z) D_i \sin(D_i A) \right] \times \left[ \sum_i \int dx \int dy N_i(x, y, z) D_i^2 \right]^{-1} - \frac{1}{2} \sigma A, \quad (14)$$

where

$$D_i = \frac{p_i}{p_0} \frac{|\hat{p}_i(x, y, z) \cdot \vec{\xi}^*(x, y, z)|}{|\hat{p}_0(0, 0, z) \cdot \vec{\xi}^*(0, 0, z)|}$$

and the pulse area  $A$  is chosen to be equal to the tipping angle at the beam center  $x = y = 0$  for atoms with dipole moment  $\vec{p}_0$ :

$$A = (p_0/\hbar) |\vec{\xi}^*(0, 0, z) \cdot \hat{p}_0| \int_{-\infty}^{+\infty} \mathcal{E}(z, t) dt. \quad (15)$$

The presence of the denominator ensures that  $\alpha$  remains the total resonant linear Beer's gain constant. The quantity  $N_i(x, y, z)$  is proportional to the local population inversion of the  $i$ th transition at frequency  $\omega$ . In the case of free particles in a gas, the prescribed representation results in each unit vector  $\hat{p}_i$  being parallel to the electric field polarization vector parallel to  $\vec{\xi}$ . For a  $J = 1$  to  $J' = 2$  transition, (e. g., the 6328-Å transition in Ne) with the initial-state populations differences  $N_i$  equal and independent of transverse position, Eq. (14), using Eq. (13), reduces to

$$\frac{dA}{dz} = \frac{2}{5} \alpha \left( \frac{1 - \cos A}{A} + 2 \frac{1 - \cos(\frac{1}{2}\sqrt{3}A)}{A} \right) - \frac{1}{2} \sigma A \quad (16)$$

in the linearly polarized Gaussian-mode case. The prescribed representation in this case is that described through a quantization axis parallel to  $\vec{\xi}$

so that the transitions obey the selection rule  $\Delta m = 0$ . The first term represents the  $M_{J'} = 0$  to  $M_J = 0$  transition with dipole moment  $p_0$ ; the second term represents the  $M_{J'} = \pm 1$  to  $M_J = \pm 1$  transitions with dipole moments  $\frac{1}{2}\sqrt{3} p_0$ . The factor  $\frac{2}{5}$  assures that  $\alpha$  remains the total resonant gain constant.

A net area theorem always is derived from a superposition of terms  $\sin\theta$ . Consequently, an area theorem for a given arbitrary system always takes the form

$$\frac{dA}{dz} = \frac{1}{2} \alpha S(A) - \frac{1}{2} \sigma A, \quad (17)$$

where  $S(A) = A - B_3 A^3 + B_5 A^5 - \dots$  is an odd function of  $A$ . The coefficients  $B_3, B_5$ , etc., are all positive in unmixed amplifying systems, and dependent on distance  $z$  if the form of the transverse-mode function varies.

The pulse area evolves according to Eq. (17) and eventually approaches the value  $A_0$  determined by the smallest nonzero solution of the equation  $S(A) = \sigma A / \alpha$ , as illustrated in Fig. 1. If the mode-function form or diameter varies with distance, then the pulse area may be imagined to track the intercept  $A_0(z)$ , as  $A_0(z)$  changes with variations of  $S(A)$  or the effective-loss parameter  $\sigma$ .

#### ENERGY GAINS IN INHOMOGENEOUSLY BROADENED SYSTEMS

The second function of interest here is the mode center pulse energy/cm<sup>2</sup>  $\mathcal{T}$  defined by

$$\mathcal{T}(z) = \frac{\eta c}{2\pi} \int |\mathcal{E}(z, t)|^2 |\vec{\xi}(0, 0, z)|^2 dt. \quad (18)$$

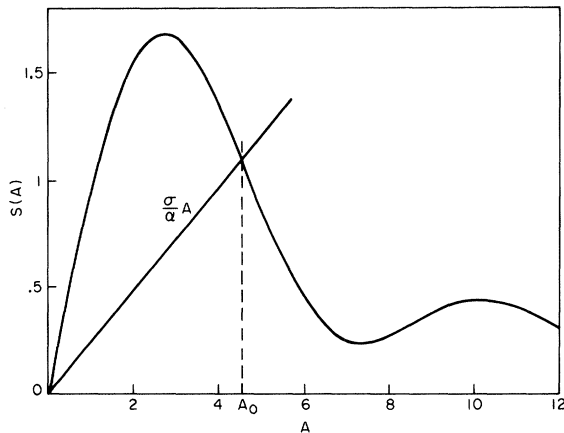


FIG. 1. The curve is  $S(A)$  for a  $J=1$  to  $J=2$  transition with a linearly polarized Gaussian transverse-mode profile. The densities  $N_i$  vary radially as  $1 - \frac{1}{2} I(r)/I(0)$ , where  $I(r) = I(0) e^{-r^2/r_0^2}$  is the mode intensity at radial distance  $r$  and  $r_0$  is a mode radius. The straight line has slope  $\sigma/\alpha$ , and the intercept  $A_0 \approx 4\frac{1}{2}$  rad is the steady-state pulse area.

For finite transverse-mode diameters, the total pulse energy is

$$U(z) = \int dx \int dy |\vec{\xi}(x, y, z)|^2 / |\vec{\xi}(0, 0, z)|^2 \mathcal{T}(z). \quad (19)$$

Weak pulses of small bandwidth and small area are uniformly and linearly amplified according to the equation

$$d\mathcal{T}(z)/dz = \alpha \mathcal{T}(z) - \sigma \mathcal{T}(z).$$

If the pulse bandwidth or area is not small, the energy gain due to resonant interaction will in general change, and a factor  $F$  is included to take this into account:

$$\begin{aligned} \frac{d\mathcal{T}(z)}{dz} &= \alpha F(A, \mathcal{T}, \text{"pulse shape," "mode profile"}) \mathcal{T}(z) \\ &\quad - \sigma \mathcal{T}(z). \end{aligned} \quad (20)$$

By definition of  $\alpha$ ,  $F=1$  when  $A$  and  $\mathcal{T}$  are small. The "pulse shape" variable describes time aspects of the pulse  $\mathcal{E}(z, t)$  except for the pulse area and energy. A pulse might have, for example, a "Gaussian" shape. The "mode profile" variable includes effects due to both a transverse intensity variation of the single-mode light pulse and overlapping or degenerate resonant transitions.

In the case where the light pulse is a plane wave and there is neither degeneracy nor overlapping transitions, the corresponding quantity  $F$  displays characteristic oscillations, with nearly zero values at  $2\pi, 4\pi$ , etc. Furthermore, the value of  $F$  for given  $A$  or  $\mathcal{T}$  is reasonably independent<sup>2</sup> of the pulse shape. This independence carries over to the more general case above, and the dependence of  $F$  on the pulse shape will be neglected. Let  $F$  in the plane-wave nondegenerate case be denoted by  $F_p(\theta, \mathcal{T})$ , where  $\theta$  and  $\mathcal{T}$  refer to values at  $x=y=0$  for the transition with dipole moment  $\vec{p}_0$ .

Use of Eq. (19) allows (20) to be rewritten in terms of the total pulse energy  $U(z)$ :

$$\frac{dU(z)}{dz} = \alpha F(A, \mathcal{T}, \text{"mode profile"}) U(z) - \sigma' U(z), \quad (21)$$

where  $\sigma'$  refers to real losses and not those changes in  $U(z)$  due to a distance  $z$ , dependent on mode profile or diameter. In this equation, the term involving  $F$  is the total change/cm of energy in the resonant atoms. The energy emitted by an atom involved in the  $i$ th transition at position  $(x, y, z)$  will be calculated first and then a subsequent summation over transitions and positions  $x, y$  will produce a formula for the total resonant energy change/cm. Identification of this result

with the term  $\alpha F U(z)$  above results in an expression for  $F$ .

The quantity  $F_p$  involves an average over the inhomogeneously broadened resonance at  $x = y = 0$  for the transition with dipole moment  $\vec{p}_0$ . The dynamics of nondegenerate atoms may be completely described by a torque equation<sup>2,8</sup> which is derived from an interaction

$$-\vec{p}_i(x, y, z, t) \cdot \vec{E}(x, y, z, t),$$

where  $\vec{p}_i(x, y, z, t)$  is the dipole-moment operator for the  $i$ th atom at  $x, y, z$  at time  $t$ . In the case here, therefore, the torque equation involves the driving electric field in the form

$$p_i | \hat{p}_i \cdot \vec{\xi}^*(x, y, z) | \mathcal{E}(z, t),$$

which is proportional to  $D_i \mathcal{E}(z, t)$ , apart from the phase of  $\hat{p}_i \cdot \vec{\xi}(x, y, z)$ , which cannot affect a result for energy change. Consequently, since the energy change of atoms of dipole-moment matrix element  $\vec{p}_0$  at  $x = y = 0$  is proportional to  $\mathcal{T}(z) F_p(A, \mathcal{T})$ , it follows that the energy change of atoms with dipole-moment matrix element  $\vec{p}_i$  at position  $x, y, z$  is proportional to  $D_i^2 F_p(D_i A, D_i^2 \mathcal{T})$ , since, for a given pulse width,  $\mathcal{T}$  is proportional to  $\mathcal{E}^2$ , and only  $D_i \mathcal{E}$  can be involved in the energy-change expression.

The net energy absorbed is the resultant sum over transitions and positions so that, upon identifying the total resonant energy change with the term  $\alpha F U(z)$ , we have

$$\alpha F(A, \mathcal{T}) U(z) = \Delta \int dx \int dy \sum_i N_i(x, y, z) D_i^2 \mathcal{T}(z) \times F_p(D_i A, D_i^2 \mathcal{T}), \quad (22)$$

where  $\Delta$  is a multiplicative factor.

In the limit of small  $A(z)$  and  $\mathcal{T}(z)$ ,  $F_p = F = 1$ , and use of Eq. (19) allows  $\Delta$  to be determined so that

$$F = \left[ \sum_i \int dx \int dy D_i^2 N_i(x, y, z) F_p(D_i A, D_i^2 \mathcal{T}) \right] \times \left[ \sum_i \int dx \int dy D_i^2 N_i(x, y, z) \right]^{-1}, \quad (23)$$

where  $F_p$  is evaluated according to the indicated functional dependence and the denominator ensures that  $F = 1$  in the limit of small pulse area and pulse bandwidth.

For  $\tau \gg T_2^*$ , there is no dependence<sup>2</sup> of  $F_0$  and therefore  $F_p$  on  $\mathcal{T}$ , because a change in  $\mathcal{T}$  for constant pulse area  $A$  only results in a larger excitation bandwidth, which is reflected in the factor  $\mathcal{T}$  in Eq. (20). As  $\tau$  becomes comparable with  $T_2^*$ , for constant  $A$ , the larger bandwidth of excitation begins to include the wings of the resonance, and  $F$  decreases with increasing  $\mathcal{T}$ . If the resonant spectral-gain profiles for various positions and transitions vary in a way not simply proportional to  $N_i(x, y, z)$ , Eq. (23) must further be generalized

to take into account this fact through a variation in the dependence of the various  $F_p$  on the effective interacting pulse energy  $D_i^2 \mathcal{T}$ .

If  $\tau \gg T_2^*$  and the amplifying transition is bell shaped,  $F$  is an even function of  $A$ , never exceeds unity, and approaches the value 1 as  $A$  tends to 0 (except for pulse shapes which undergo 180° phase reversals and have zero net area). For  $\tau \gg T_2^*$ , it can be shown that  $F(A)$  decreases towards zero for increasing  $A$ , eventually becoming proportional to  $1/A$ , possibly with some oscillations. This limiting case will not be of primary interest in the analysis to follow.

It is convenient to introduce the pulse-energy dependence parametrically through the defined pulse width  $\tau$ , where

$$\tau(z) = \beta \left[ \int_{-\infty}^{+\infty} \mathcal{E}(t, z) dt \right]^2 / \int_{-\infty}^{+\infty} \mathcal{E}(t, z)^2 dt, \quad (24)$$

and  $\beta$  is of order unity. The value of  $\beta$  is formally arbitrary but may be chosen for convenience, perhaps so that  $\tau$  is the time between half-maximum intensity times for a particular pulse shape. The nonlinear gain function  $F(A, \mathcal{T})$  appearing in Eq. (20) may now be regarded as a function of  $A$  and  $\tau$  instead. The numerator and denominator of Eq. (24) are, respectively, proportional to the pulse area squared and the pulse energy, so that from Eqs. (17) and (20) we have

$$\frac{d\tau}{dz} = \alpha \tau [S(A)/A - F(A, \tau)]. \quad (25)$$

In the plane-wave nondegenerate case, the energy change of exactly resonant atoms is proportional<sup>8</sup> to  $1 - \cos\theta = 1 - \cos A$ . The total energy change of all atoms is proportional to the product of some average energy change and an average bandwidth of excitation. Choosing  $1 - \cos\theta$  as a typical change of energy content and  $\tau^{-1} \propto \mathcal{T}/\theta^2$  as excitation bandwidth in the limit  $\tau \gg T_2^*$ , an expression  $2(1 - \cos\theta)/\theta^2$  follows for an estimate of  $F_p(\theta, 0)$ . This expression does not, however, include the effects of power broadening, important for  $\theta > 2\pi$ , and consequently does not behave as  $1/\theta$  for large  $\theta$ . For small  $A$ , the effects of bandwidth can be formulated through a linear calculation. A Gaussian pulse, interacting with a system with a Gaussian spectral-gain profile, is associated with

$$F(0, \tau) = \tau / (\tau^2 + 2T_2^{*2})^{1/2},$$

where  $\tau$  is the time between half-maximum intensity times, and  $T_2^* = \sqrt{2} (\ln 2) / \tau \Delta\nu$ , where  $\Delta\nu$  is the frequency width (Hz) between half-maximum gain frequencies.

A surprisingly good fit to  $F_p(\theta, \mathcal{T})$ , especially useful for estimates in the region  $0-2\pi$ , is

$$F_p(\theta, \tau) = \frac{2(1 - \cos\theta)}{\theta^2} \left( \frac{\tau^2}{\tau^2 + 2T_2^{*2}} \right)^{1/2}, \quad (26)$$

where the dependence of  $F_p$  on  $\tau$  is introduced through the appropriately defined pulse width  $\tau$ .

In the region  $0-2\pi$ , this formula is an excellent fit to values calculated for a pulse of Gaussian shape of width  $\tau \gg T_2^*$ . The bandwidth factor has the proper limits for  $\tau \gg T_2^*$  and  $\tau \ll T_2^*$  and should describe bandwidth effects to a reasonable accuracy. Near  $2\frac{1}{2}\pi$ , with  $\tau \gg T_2^*$ , this formula yields a value of  $F_p$  only about 35% smaller than the value numerically calculated<sup>2</sup> for a pulse of Gaussian shape.

The  $2\pi$  hyperbolic secant "self-induced transparency" pulse<sup>1,2</sup> has the plane-wave values  $F_p(2\pi) = 0$ ,  $S(2\pi) = 0$ , and  $\sigma = 0$  and applies stably for  $\alpha < 0$ . A  $\pi$ -pulse solution<sup>2</sup> has the plane-wave values  $F_p(\pi) = \frac{1}{2}$ ,  $S(\pi) = 0$ , and  $\sigma = 0$  and applies in the region  $\alpha > 0$ ,  $\tau \gg T_2^*$ .

The posed analysis can be usefully performed graphically. In Fig. 2, a trajectory is drawn on a graph of curves of  $F(A, \tau)$  plotted against  $A$  for various pulse widths  $\tau$ . The propagating pulse area, for a given input pulse area, is determined as a function of distance  $z$  alone, through Eq. (17). The solution to the equation for the pulse energy, Eq. (20), is then specified through the knowledge of  $A(z)$ . The final pulse area stabilizes at the value  $A_0$ , where  $dA/dz = 0$ , so that  $S(A_0) = \sigma A_0/\alpha$ . Finally, with pulse area so stabilized, the pulse energy continues to change until the gain function saturates when  $\tau$  shortens to value  $\tau_0$ , where  $d\tau/dz = 0$ , so that  $F(A_0, \tau_0) = \sigma/\alpha$ .

In Fig. 2, two possible trajectories are shown: (I) The input pulse is such that  $\tau$  shortens to a time about equal to  $T_2^*$  while  $A$  is still changing towards  $A_0$ ; (II)  $A$  becomes close to  $A_0$  before  $\tau$  becomes comparable with  $T_2^*$ . A more complicated system would result in a figure of similar appearance. The curves of  $F(A, \tau)$  and  $S(A)$  may be imagined to change with distance  $z$  if some parameter [e. g., the mode function  $\bar{\xi}(x, y, z)$  or the total inversion] depends on distance  $z$ . The actual pulse area and pulse width track the operating point that would be obtained through setting  $dA/dz = d\tau/dz = 0$ . If  $\tau < T_2^*$ , the description above may not apply if pulse-amplitude modulation is not taken into account in this analysis.

#### SMALL-SIGNAL THEORY

If both  $\theta$  and  $T_2^*/\tau$  are small, but nonlinear effects are nevertheless operative in both Eqs. (17) and (20), and if furthermore the finite bandwidth of the amplifying transition is operative in Eq. (20), we may expand the functions  $S(A)$  and  $F(A, \tau)$  in a Taylor series, keeping Eq. (24) in mind, and re-

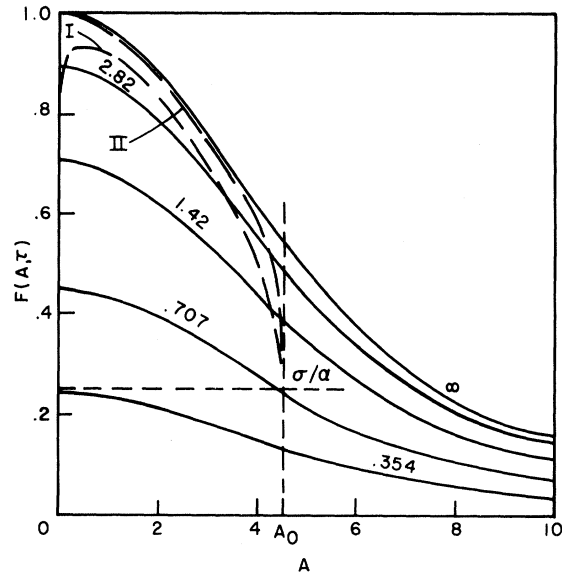


FIG. 2. Sketched trajectories in the  $F$ - $A$  plane of evolving pulse areas and energies, the pulse energy defined parametrically through the pulse area  $A$  [Eq. (15)] and pulse width  $\tau$  [Eq. (24)]. The time  $T_2^*$  is such that Eq. (28) applies in the limit  $A \rightarrow 0$ , but with  $\tau/T_2^*$  merely small. The solid curves represent  $F(A, \tau)$ , with the adjoining numbers being values of  $\tau/T_2^*$ . The system is that described in the caption of Fig. 1. The approximate formula for  $F_p$  given by Eq. (26) was used to obtain numerical values of  $F(A, \tau)$ . Curves I and II are trajectories of evolving pulse areas and energies, discussed in more detail in the text. The steady-state pulse area  $A_0$  is determined in Fig. 1, and the steady-state pulse width is about  $0.7 T_2^*$ .

tain only the leading terms. In this approximation

$$\frac{dA}{dz} = \frac{1}{2}\alpha (A - B_3 A^3) - \frac{1}{2}\sigma A, \quad (27)$$

$$\frac{d\tau}{dz} = \alpha \tau \left( 1 - \delta A^2 - \frac{T_2^{*2}}{\tau^2} \right) - \sigma \tau. \quad (28)$$

The value<sup>2</sup> of  $B_3$  in the case of no degeneracy has been determined for the plane-wave case to be  $\frac{1}{8}$ , and in the Gaussian mode case to be  $\frac{1}{12}$ . The parameter  $T_2^*$ , about equal to the inverse amplifying bandwidth, is now precisely defined by Eqs. (19) and (16), once  $\beta$  is chosen, by the condition of linear ( $\theta \ll \pi$ ) gain saturation in the limit of large  $\tau/T_2^*$ .

The value of  $\delta$  may be determined by expanding  $F_p(A, \tau)$  to order  $A^2$  and substituting the result into Eq. (23). A comparison of Eqs. (14) and (23) reveals that the ratio  $\delta/B_3$  is independent of the mode profile and only depends on the pulse shape. Consequently,  $\delta/B_3$  may be calculated in the plane-wave nondegenerate case and  $\delta$  then determined

from the particular value of  $B_3$  for the system of interest. The square pulse case<sup>2</sup> prescribes the analytical value  $\delta/B_3 = \frac{1}{2}$ , while numerical calculations<sup>2</sup> for a Gaussian pulse shape specify a slightly higher value of  $\delta/B_3$ . The functional  $F$  must be an even function of  $A$ , so that there can be no terms linear in  $A$  in the expansion Eq. (28) of  $F$ . A linear term  $T_2^*/\tau$  will not be present if the spectral distribution function derivative has no discontinuity. The bandwidth reduction factor  $T_2^{*2}/\tau^2$  is equivalent to a factor  $(\Delta\nu_p/\Delta\nu)^2$ , where  $\Delta\nu_p$  is the light-pulse bandwidth and  $\Delta\nu$  the amplifying bandwidth.

The solution of Eq. (27) is given through the exponential decay of the squared-reciprocal pulse area.

$$[A(z)]^{-2} = \frac{B_3}{\alpha - \sigma} + \left( [A(0)]^{-2} - \frac{\alpha B_3}{\alpha - \sigma} \right) e^{-(\alpha - \sigma)z}, \quad (29)$$

where  $A(0)$  is the input pulse area at  $z = 0$ . It is assumed that  $\alpha$  and  $\sigma$  do not depend on distance  $z$ . The final stable operating area at very large  $z$  is

$$A_0 = [(\alpha - \sigma)/\alpha B_3]^{1/2}. \quad (30)$$

Substituting Eq. (24) into Eq. (28) and using Eq. (27) leads to the equation, for the changing pulse width,

$$\frac{d\tau}{dz} = \alpha\tau \left( \frac{T_2^{*2}}{\tau^2} - (B_3 - \delta)A^2 \right). \quad (31)$$

If  $T_2^{*2}/\tau^2 > (B_3 - \delta)A^2$ , the pulse width lengthens, but if  $(B_3 - \delta)A^2 > T_2^{*2}/\tau^2$ , the pulse width shortens. If a short weak pulse of width  $\tau(0)$  is injected at  $z = 0$ , the nonlinear term may be neglected, and  $\tau$  behaves for small  $z$  as

$$\tau(z) = [\tau(0)^2 + 2\alpha T_2^{*2}z]^{1/2}, \quad (32)$$

but if the medium is predominantly nonlinear for small  $z$ , the bandwidth term  $T_2^{*2}/\tau^2$  may be neglected, and for some distance  $z \leq z'$ ,

$$\tau(z) = \tau(0)[A(z)/A(0)]^{2(1 - \delta/B_3)} \times e^{-(\alpha - \sigma)(1 - \delta/B_3)z},$$

where  $A(z)$  is given by Eq. (29). If  $A(z) \ll 1$  and  $T_2^*/\tau \ll 1$ ,  $\tau(z) = \tau(0)$  as expected. If we assume that  $A(z') = A_0$  to good accuracy, then for  $z > z'$ ,  $\tau(z)$  is given by

$$\tau^2(z) = \frac{T_2^{*2}}{(1 - \sigma/\alpha)(1 - \delta/B_3)} + \left( \tau^2(z') - \frac{T_2^{*2}}{(1 - \sigma/\alpha)(1 - \delta/B_3)} \right) \times e^{-2(\alpha - \sigma)(1 - \delta/B_3)(z - z')}, \quad (33)$$

and approaches the final operating pulse width  $\tau_0$

found by setting  $dA/dz = d\tau/dz = 0$  in (27) and (31),

$$\tau_0 = \frac{T_2^*}{(1 - \sigma/\alpha)^{1/2}(1 - \delta/B_3)^{1/2}}. \quad (34)$$

According to Eqs. (15), (18), and (24),

$$\mathcal{I}(z) = \frac{\beta \hbar^2 \eta c}{2\pi \rho_0^2} \frac{|\tilde{\xi}(0, 0, z)|^2}{|\tilde{\xi}(0, 0, z) \cdot \hat{p}_0|^2} \frac{A^2}{\tau}, \quad (35)$$

so that

$$\mathcal{I}_0 = \frac{\beta \hbar^2 \eta c}{2\pi \rho_0^2} \frac{|\tilde{\xi}(0, 0, z)|^2}{|\tilde{\xi}(0, 0, z) \cdot \hat{p}_0|^2} \frac{(1 - \sigma/\alpha)^{3/2}(1 - \delta/B_3)^{1/2}}{B_3 T_2^*}. \quad (36)$$

It should be noted that

$$\tau_0 \tau_0^3 = \frac{\beta \hbar^2 \eta c T_2^{*2} |\tilde{\xi}(0, 0, z)|^2}{2\pi \rho_0^2 B_3 (1 - \delta/B_3) |\tilde{\xi}(0, 0, z) \cdot \hat{p}_0|^2} \quad (37)$$

is independent of the resonant gain  $\alpha$  and the background loss  $\sigma$ . A closely related quantity

$$A_0 \tau_0 = \frac{T_2^*}{[B_3(1 - \delta/B_3)]^{1/2}} \quad (38)$$

is likewise independent of  $\alpha$  and  $\sigma$ . Frova *et al.*<sup>11</sup> pointed out that their observations of the output of a He-Ne laser mode locked<sup>12</sup> by a Ne absorption cell are in good agreement with Eq. (37); reasonable quantitative agreement is reached in their case if one averages over the discharge length as well as the transverse distance in the evaluation of  $B_3$ . The data of Fox and Smith<sup>13</sup> are also in accord with Eq. (37). Both experiments involved the  $J = 1$  to  $J' = 2$ , 6328-Å neon transition. To a degree of approximation largely determined by operating conditions, a mode-locked laser may be thought of as an unidirectional traveling-wave laser amplifier with a steady-state pulse. However, lack of precise knowledge of the longitudinal and radial dependence of the density of excited atoms causes an uncertainty in the relevant value of  $B_3$ . Furthermore, overlap of the mode-locked pulse with itself upon reflection from a cavity mirror, atomic memory times comparable to the transit time of a pulse between mirrors, frequency modulation of the mode-locked pulses, and other effects are not here taken into account, so that only qualitative agreement with Eq. (37) should be expected. The agreement between experiment and Eq. (37) should be regarded as incompletely explained. If such mode-locked laser outputs conform with Eq. (37) or an analogous equation taking into account possible frequency modulation, then nontrivial but direct-measurement mode-locked pulse parameters yield a value for the dipole moment of the transition.

The region of applicability of the small signal calculation may be assessed through the alternating power series expansion following Eq. (16). Keeping one term  $B_3 A^5$  more than in the small



signal calculation, if the extra term  $B_5 A^5$  is small, a corrected solution  $A_0$  to the equation  $\alpha(A - B_3 A^3 + B_5 A^5) = \sigma A$  is given by

$$A_0^2 = \frac{\alpha - \sigma}{\alpha B_3} + \frac{B_5}{B_3^3} \left( \frac{\alpha - \sigma}{\alpha} \right)^2, \quad (39)$$

and the fractional error in calculating  $A_0$  by Eq. (30) is therefore about  $B_5 A_0^2 / 2B_3$ . In the nondegenerate plane-wave, Gaussian-, and Lorentzian-mode cases, with  $A$  equal to the beam center tipping angle, respectively,<sup>2</sup>

$$B_3 = \frac{1}{6}, \frac{1}{12}, \frac{1}{18},$$

$$B_5 = \frac{1}{120}, \frac{1}{360}, \frac{1}{600},$$

$$B_3/B_5 = 20, 30, 33\frac{1}{3}.$$

If  $B_5 A_0^2 / 2B_3$  is required to be less than 10%, then  $A < 0.64\pi$ ,  $0.78\pi$ , and  $0.81\pi$ , respectively. The important parameter  $B_3 A_0^2$ , which indicates the amount of nonlinearity, is, for 10% error,  $< \frac{2}{3}$ ,  $\frac{1}{2}$ ,  $0.370$ , respectively. A similar analysis indicates that the errors in Eqs. (34) and (37) should be about twice the error in Eq. (30). As the transverse mode becomes less "square," the importance of higher nonlinearities increases. Degeneracy effects will further increase the importance of higher nonlinearities. But for  $B_3 A_0^2 < 0.4$  (corresponding to  $\sigma/\alpha > 0.6$  and  $\tau > 2.3 T_2^*$ ), the small signal calculation should be quite accurate, and small signal theory applies over a large range of pulse areas.

### CONCLUSIONS

The pulse-area-pulse-energy approach to light-pulse propagation problems allows a quick and quantitative estimate of the changing and steady-state pulse energy, width, and area. The small signal calculations are reasonably accurate over a sizable range of pulse areas and pulse widths, and result in a steady-state  $\tau_0 \tau_0^3$  law, which agrees with measurements,<sup>11</sup> in spite of the fact that the pulses are frequency modulated.<sup>14</sup>

The point at which a pulse breaks up<sup>2,4,5,15</sup> into two smaller pulses may be taken as an upper limit for the region of relevance of the pulse-energy-pulse-area calculations. However, it is not presently clear that such breakup effects necessarily occur in systems for which transverse-mode and degeneracy effects are large. Indeed, for the subject system (envisioned as approximating conditions in a He-Ne plasma tube) depicted by Figs. 1 and 2, the condition  $S(A) > 0$  applies for all  $A$  [in contrast to the nondegenerate plane-wave case for which  $S(A) = \sin A$ ]. We may conclude that the net emitted energy by atoms with resonant frequency  $\omega$  is a monotonically increasing function of time, for a given pulse of any area, because the electric field is sufficiently inhomogeneous over the beam diameter to average out oscillations. In any event, if the breakup of a pulse in such systems should occur, it will require a much larger distance to set in than might be estimated from plane-wave nondegenerate results.

The region of applicability of the pulse-area-pulse-energy formalism in this paper is purposely limited to the region  $\tau > T_2^*$  in order to avoid the possibility of severe amplitude modulation effects<sup>4</sup> for shorter  $\tau_0$ . Investigations<sup>6</sup> of the homogeneously broadened amplifier indicate that the pulse formed by a sufficiently long amplifying medium may be free of severe modulation, even for  $\tau \ll T_2$ . If a similar situation holds in the inhomogeneously broadened line case, the estimates given here will be reasonably accurate. At this time, however, we must regard the large- $z$  description of modulation effects stimulated by short pulses in highly amplifying media as an open question.

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PHYSICAL REVIEW A

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## Nuclear Magnetic Resonance in Solid Helium-3–Helium-4 Mixtures between 0.3 and 2.0 °K \*

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Nuclear-magnetic-resonance techniques have been used to study vacancy diffusion and the exchange interaction of He<sup>3</sup> in solid He<sup>3</sup>–He<sup>4</sup> mixtures between 0.3 and 2.0 °K for molar volumes between 20 and 22 cc/mole. The concentrations studied are 32.1, 7.78, and 1.94% He<sup>3</sup> in He<sup>4</sup>. There is good agreement between the diffusion activation energies determined from the  $T_2$  measurements and those obtained by measuring the diffusion constant directly using the field-gradient technique. These activation energies are consistently lower for mixtures than for pure He<sup>3</sup>. The  $T_1$  data in the Zeeman-exchange plateau region indicate that the exchange interaction is independent of concentration. The  $T_2$  data in the exchange-narrowed region are not in agreement with the theoretical result obtained by allowing the moments of the line shape to become concentration dependent. The experimental values of  $T_2$  are much lower than the predicted values. This deviation is qualitatively explained by postulating the existence of two spin species: those that strongly experience the effects of exchange and those that do not. A small fraction of the isolated spins can then dominate the  $T_2$  relaxation process.

### I. INTRODUCTION

The magnetic properties of solid He<sup>3</sup> have been the subject of intense experimental and theoretical investigation for the past ten years.<sup>1–7</sup> The most interesting feature of this solid is that the van der Waals binding energy is not much larger than the kinetic zero-point energy, so that the atoms undergo large-amplitude zero-point vibrations, and there is an unusually large amount of overlap between the wave functions of atoms occupying adjacent lattice sites. As a result of the atomic overlap and the symmetry requirements for pairs of fermions, there is a probability that a pair of atoms will mutually tunnel between adjacent lattice sites and interchange their positions. This process leads to an exchange energy in the Hamiltonian specifying the system.<sup>8</sup> The magnitude of the exchange interaction is essentially proportional to the electrostatic energy developed by the overlapping atoms and the scalar product of the nuclear-

spin orientations. The exchange process appears to be most probable when the pair of atoms are in the singlet state.<sup>6,7</sup>

The effects of He<sup>4</sup> impurities in the He<sup>3</sup> lattice have also received experimental<sup>9–14</sup> and theoretical<sup>15</sup> attention. Recent nuclear resonance experiments seem to indicate that the excess volume around a He<sup>4</sup> impurity permits the neighboring pairs of He<sup>3</sup> atoms to have a greatly “enhanced” exchange interaction.<sup>13,14</sup> In this work, we have investigated the possibility of such effects from a different point of view. By studying the nuclear resonance properties of solid mixtures of He<sup>3</sup> and He<sup>4</sup> with He<sup>3</sup> concentrations between 2 and 30%, we hoped to understand what effects the He<sup>4</sup> would produce on neighboring He<sup>3</sup> atoms undergoing the exchange tunneling, and what effects the dilution process has on the spectral density function responsible for the relaxation processes.

In Sec. II, the theory of how diffusion and exchange affect  $T_1$  and  $T_2$  is reviewed and extended