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Influence of Degeneracy on Coherent Pulse Propagation in an Inhomogeneously Broadened Attenuator*

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The influence of degeneracy on coherent pulse propagation in an inhomogeneously broadened attenuator is examined by computer calculations, and the connection with self-induced transparency in a nondegenerate system is described. Specific modifications that arise in a degenerate system are the development of pulse shapes characteristic of the degeneracy, the association of a finite loss with propagation, a reduction of pulse delay times, and the suppression of pulse separation of multi-2π pulses. Additionally, results of calculations pertaining to experiments on pulse propagation in SF_6 are presented which indicate two basically different phenomena. One corresponds to delay phenomena qualitatively similar to nondegenerate self-induced transparency, while the other is a coherent saturation effect generally characteristic of degenerate media. The computed results fully substantiate our earlier experimental findings.

I. INTRODUCTION

THE properties of coherent pulse propagation in attenuating media have recently received intensive study.[1] Considerable activity has centered on the nonlinear features associated with the transmission of coherent radiation. A number of significant and surprising results have emerged. One such result is the phenomenon of self-induced transparency (SIT). Aside from practical considerations, this effect is of interest because it involves the coupling of Schrödinger's and Maxwell's equations through a nonlinear dynamical process and is completely solvable under certain simple conditions.

It has been shown that the effect of level degeneracy has a strong influence on the propagation behavior of coherent pulses, and hence, on SIT. Given the one-to-one correspondence between the rotating frame well known in NMR[2] and the rotating frame appropriate for discussion of electric-dipole transitions as described by Hartmann,[3] it is pertinent to inquire why degeneracy effects frequently arise in the latter but not in the former. *Spatial* degeneracy does not occur in NMR because the static magnetic field destroys the isotropy of space by establishing a preferred direction. More formally, the

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[1] S. L. McCall and E. L. Hahn, Phys. Rev. Letters **18**, 908 (1967); Frederic A. Hopf, C. K. Rhodes, G. L. Lamb, Jr., and Marlan O. Scully, Phys. Rev. (to be published); F. T. Arecchi, V. DeGiorgio, and S. G. Someda, Phys. Letters **27A**, 588 (1968); S. L. McCall and E. L. Hahn, Phys. Rev. **183**, 457 (1969) (this article briefly notes degeneracy effects); C. K. N. Patel and R. E. Slusher, Phys. Rev. Letters **19**, 1019 (1967); F. T. Arecchi, G. L. Masserini, and P. Schwendimann, Riv. Nuovo Cimento **1**, 181 (1969); F. A. Hopf and M. O. Scully, Phys. Rev. (to be published).

[2] A principal reference and guide to the NMR literature is A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961).

[3] I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. **141**, 391 (1966).

unperturbed part of the Hamiltonian in NMR does not commute with the group of rotations. It follows that systems possessing different magnetic moments are not degenerate.[4] This failure in the analog between the two rotating frames is also manifested in the fact that in NMR the rotating frame is in R^3 coordinate space (x,y,z) while the electric dipole analog is in pseudospace.

An earlier publication[5] presented calculations, valid for an attenuating medium, of propagating pulse shapes with the use of an unbroadened model.[6] This article is an extension of that previous work and incorporates the effect of an inhomogeneously broadened resonance. We begin with a brief theoretical review and describe the extension to degenerate systems. Specific results are calculated for $Q(j)$ (i.e., $j\rightarrow j$) transitions,[7] since spatial degeneracy is commonly present[8] in a wide variety of materials. We point out that the origin of the degeneracy in a particular situation may be spatial or may arise from the presence of overlapping transitions. As it turns out, the well-known problems associated with SF_6 involve both of these considerations.

Finally, we present the details of calculations which are pertinent to some experimental work[9] using SF_6. These computations, which agree very well with the experimental findings, describe two essentially different phenomena. The first is the significant reradiation and pulse delay which qualitatively resemble SIT in a nondegenerate system. This occurs for input pulses whose angle $\theta(0)$ is in the vicinity of π. The second phenomenon involves input pulses for which the inequality $\theta(0)\gg\pi$ is valid. A pulse-steepening effect appears for pulse input angles in that region. This is *not* a saturation process in the language of rate equations. On the contrary, it is shown to arise as a result of a completely coherent excitation in the presence of sufficient level degeneracy.

II. THEORETICAL OUTLINE

The theoretical analysis of a degenerate inhomogeneously broadened ensemble of two-level radiators interacting with a classical electromagnetic field involves a straightforward generalization of the nondegenerate theory. Since a previous publication[10] has examined that theory in considerable detail, only an outline of the extension to the degenerate case will be presented here.

Consistent with customary notation, we take the electromagnetic field as a linearly polarized plane running wave of the form

$$\mathbf{E}(t,z)=\hat{\epsilon}\,\mathcal{E}(t,z)\cos(kz-\nu t)\,, \tag{1}$$

where $\hat{\epsilon}$ designates a unit polarization vector. The electric field envelope function $\mathcal{E}(t,z)$ is assumed to be slowly varying[11]; more precisely, this assumption is stated as

$$\partial\mathcal{E}(t,z)/\partial z\ll k\,\mathcal{E}(t,z) \tag{2}$$

and

$$\partial\mathcal{E}(t,z)/\partial t\ll \nu\,\mathcal{E}(t,z) \tag{3}$$

for all z and t. The field interacts with a set of ensembles, indexed by $\{A\}$, of two-level systems described by a corresponding set of 2×2 density matrices $\rho_i=\rho(\omega,t,z,\wp_i)$, $i\epsilon\{A\}$.[12] Each density matrix ρ_i has elements $(\rho_i)_{aa}$, $(\rho_i)_{bb}$, and $(\rho_i)_{ab}=(\rho_i)_{ba}{}^*$ where the subscripts a and b index the upper and lower states, respectively. The radiators, in the laboratory frame, have resonant frequencies ω; the ith ensemble possesses a dipole matrix element $\wp_i$; the relaxation processes for all the ensembles are characterized by the relaxation parameters[13] T_1 and T_2. Since the medium is attenuating, the equations of motion are supplemented by the boundary condition[14]

$$\lim_{t\to-\infty}(\rho_i)_{\alpha\beta}=\delta_{\alpha b}\delta_{\beta b}$$

for all $i\epsilon\{A\}$, and for all $z\epsilon[0,L]$. Furthermore, the ith ensemble has an inhomogeneous frequency distribution $\sigma_i(\omega)$ which is a normalized Gaussian centered at $\omega=\omega_0$

[4] This ignores the special case of an accidental degeneracy. Throughout this paper degeneracy is regarded in the "normal" sense as described by Wigner in E. P. Wigner, *Group Theory and Its Application to the Quantum Mechanics of Atomic Spectra* (Academic Press Inc., New York, 1959), expanded edition, p. 120. The standard counterexample of accidental degeneracy is the Coulomb problem as discussed in V. Fock, Z. Physik **98**, 145 (1935).

[5] C. K. Rhodes, A. Szöke, and A. Javan, Phys. Rev. Letters **21**, 1151 (1968).

[6] In this context, the unbroadened attenuator is a model consisting of an ensemble of noninteracting radiators each of which is on exact resonance. The radiative lifetime is assumed as infinitely long.

[7] In the case of spatial degeneracy we are concerned with the transformation properties of the states under rotations. They are specified by the angular momentum quantum number j so that these considerations are not necessarily restricted to molecular rotational-vibrational states. Our interest is centered on $Q(j)$ transitions because they most nearly satisfy the conditions for SIT. See Ref. 5 in this regard.

[8] In nearly all molecular gas systems presently under study, for example, CO_2, HCN, N_2, SF_6, N_2O, and H_2O, one is dealing uniformly with levels of relatively high angular momentum. Indeed, this situation is not unknown in solids as the high spin levels of the 1.06-μ transition of Nd lasers illustrate.

[9] C. K. Rhodes and A. Szöke, Phys. Rev. **184**, 25 (1969).

[10] Frederic A. Hopf and Marlan O. Scully, Phys. Rev. **179**, 399 (1969).

[11] This is known in the literature as the slowly varying envelope assumption (SVEA). It enables one to neglect certain derivative terms in the basic equation of motion for the envelope function $\mathcal{E}(t,z)$.

[12] The expression $i\epsilon\{A\}$ states that the element i is a member of the set $\{A\}$.

[13] The relaxation parameters T_1 and T_2 correspond to the energy transfer and dephasing times, respectively.

[14] By an attenuating medium, we mean one in which all the atomic systems are initially in the ground state. The $\delta_{\alpha b}$ and $\delta_{\beta b}$ are Kronecker deltas with $\alpha,\beta\epsilon\{a,b\}$. The medium is treated as finite and one dimensional so that the variable z is constrained to lie in the closed interval $[0,L]$ for some length $L<\infty$.

with standard deviation[15] $2/T_2^*$; for a unidirectional running wave field in the medium this treatment is also appropriate for Doppler broadening. The density of the ith ensemble is given by N_i. The polarization of the medium $\mathbf{P}(t,z)$ is the ensemble average and the sum over the i ensembles of all the microscopic dipoles induced by the field and is expressed in the form

$$P(t,z)=\hat{\epsilon}\sum_{i\epsilon\{A\}}N_i\wp_i\int d\omega\{\sigma_i(\omega)[(\rho_i)_{ab}+(\rho_i)_{ba}]\}. \quad (4)$$

The response of the atoms with the coupling constant $\wp_i$ is summarized in a susceptibility function $\chi_i(T,t,z)$ which is the cosine Fourier transform of the population difference.

$$\chi_i(T,t,z)=[2\pi\sigma_i(\nu)]^{-1}\int_{-\infty}^{\infty}d\omega\sigma_i(\omega)\times\cos\{(\omega-\nu)T\}[(\rho_i)_{bb}-(\rho_i)_{aa}]. \quad (5)$$

The electric field envelope $\mathcal{E}(t,z)$ then obeys the equation of motion[15]

$$\partial\mathcal{E}(t,z)/\partial z+(1/c)[\partial\mathcal{E}(t,z)/\partial t]=-\sum_i\alpha_i\int_{-\infty}^{t}dt'\mathcal{E}(t',z)\times\exp\{-(t-t')/T_2\}\chi_i(t-t',t',z), \quad (6)$$

where c is the velocity of light in the inert background and

$$\alpha_i=\wp_i^2\omega N_i\pi\sigma_i(\nu)/2\hbar c. \quad (7)$$

The dynamical evolution of the susceptibility χ_i is described by Eq. (8):

$$\partial\chi_i(T,t,z)/\partial t=[D_i(T)-\chi_i(T,t,z)]/T_1-(\wp_i^2/2\hbar^2)\mathcal{E}(t,z)\times\int_{-\infty}^{t}dt'\mathcal{E}(t',z)\exp[-(t-t')/T_2]\times\{\chi_i(T+t-t',t',z)+\chi_i(T-t+t',t'z)\}. \quad (8)$$

$D_i(T)$ is the normalized Fourier transform of the frequency distribution function $\sigma_i(\omega)$. The initial condition associated with Eq. (8) for χ_i is[15]

$$\lim_{t\to-\infty}\chi_i(T,t,z)=D_i(T)=(T_2^*\sqrt{\pi})^{-1}\exp[-T^2/(T_2^*)^2], \quad \text{for all } i\epsilon\{A\}. \quad (9)$$

The set of dipole matrix elements, $\{\wp_i|i\epsilon\{1,2,\ldots\}\}$ is dealt with conveniently if they are indexed with the following convention. The matrix elements are rewritten in the form

$$\wp_i=r_i\wp, \quad (10)$$

with the assumption that $\wp_1=\wp$ is the maximum element and $\wp_k\leq\wp_l\to k\geq l$. Thus, $r_1=1$ and $r_k\leq r_l\to k\geq l$. Similarly, the densities N_i are defined as

$$N_i=n_iN, \quad (11)$$

with the convention $n_1=1$; this normalizes all the remaining densities to N_1. The energy of the pulse, $S(z)=(c\epsilon/8\pi)(\hbar^2/\wp^2)\tau(z)$, is proportional to the quantity $\tau(z)$, where

$$\tau(z)=(\wp^2/\hbar^2)\int_{-\infty}^{\infty}\mathcal{E}^2(t,z)dt. \quad (12)$$

Assuming that $T_2^*\ll T_2$ [16] and $\mathcal{E}(t,z)$ very small, the usual Beer's-law attenuation

$$d\tau(z)/dz=-\sum_i r_i^2n_i\alpha\tau(z) \quad (13)$$

obtains where α [assuming $\sigma_i(\omega)=\sigma(\omega)$, for all $i\epsilon\{A\}$] is given by

$$\alpha=\wp^2\omega N\pi\sigma(\omega)/2\hbar\epsilon. \quad (14)$$

Typical circumstances[17] are $n_i=1$ for all i. In such cases, the $i=1$ radiators will tend to dominate the linear absorption and the nonlinear behavior as well. Furthermore, if $\theta(z)$ is defined in terms of the maximum dipole moment as

$$\theta(z)=(\wp/\hbar)\int_{-\infty}^{\infty}\mathcal{E}(t,z)dt, \quad (15)$$

then $d\theta(z)/dz$ is found in a straightforward manner[18] as

$$d\theta(z)/dz=-(\alpha/2)\sum_i n_ir_i\sin[r_i\theta(z)]. \quad (16)$$

Naturally, the stable areas associated with Eq. (15) $[d\theta(z)/dz=0,\ d/d\theta(d\theta(z)/dz)<0]$ depend on the sets $\{r_i\}$ and $\{n_i\}$. However, certain solutions for a $Q(j)$ transition are easily determined. In that case, $n_i=1$ for all i and $r_i=(j-i+1)/j$, $i=1, 2, 3, \ldots$, so that $\theta(z)=2\pi j$ and, if j is even, $\theta(z)=\pi j$ represent stable pulse areas. We emphasize that a stable pulse area does *not* imply SIT. As an example, consider $Q(2)$. Then $\theta(z)=2\pi$ is stable, although it is impossible that this value corresponds to a transparent[19] pulse, since for the ensemble indexed by $i=2$ ($r_2=\frac{1}{2}$, equivalently $m_j=1$), the atoms near the resonant frequency are turned through an angle of π producing a partial inversion. This process represents a considerable absorption of energy. This energy loss at a fixed $\theta(z)$ is accomplished

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[15] The dephasing time T_2^* is chosen to give a simple form for $D(T)$ [see Eq. (19)]. The full width at half-height of $\sigma(\omega)$ is $\Delta\omega_{\text{inhom}}=3.3/T_2^*$. The choice of identical, resonant, degenerate Doppler lines will adequately approximate the actual physical situation of overlapping degenerate Doppler lines so long as T_2^* is chosen to be much smaller than the pulse width. The definitions of $\chi(T,t,z)$, α, and $D(T)$ differ from the convention of Ref. 10 by a minus sign. The convention used in this paper makes α and $D(T)$ positive numbers.

[16] When $T_2\approx T_2^*$, the gain is depressed as the line is broadened. For the quantitative details, the reader may consult Ref. 10.

[17] Spatial degeneracy is a common example, since a condition for thermal equilibrium is equal population of all m sublevels.

[18] Naturally, we have assumed $T_2\gg\tau_{\text{pulse}}$ throughout this discussion.

[19] In this context, we use the term "transparent" to designate lossless propagation regardless of the pulse distortion.

by an increase in the pulse width[20] $\hat{t}(z)$ and a simultaneous decrease in the average electric field amplitude[20] $\mathcal{E}_0(z)$ such that $\mathcal{E}_0(z)\hat{t}(z)$ is constant while $\mathcal{E}_0^2(z)\hat{t}(z)$ decreases monotonically, tending eventually to zero. We refer to this process, in which a pulse loses energy at constant $\theta(z)$ resulting in pulse broadening, as fixed-area attenuation. By its basic nature, this effect is accompanied by considerable pulse delay and reradiation[1] and differs substantially from the small signal or Beer's-law behavior with which no delay phenomena are associated.

A simple argument suggests that both $\tau(z)$ and $\hat{t}(z)$ behave roughly exponentially in z under these conditions. As a definite example, consider a $Q(2)$ attenuator and a pulse for which $\theta(z)=2\pi$. Recalling the discussion above, this will invert the $m_j=1$ radiators over a width in the inhomogeneous distribution given approximately by $\pi/\hat{t}(z)$. Assuming that this is the dominant loss mechanism,[21] the rate of energy loss goes as $-1/\hat{t}(z)$. Since the energy $S(z)$ is proportional to[20] $\theta^2(z)/\hat{t}(z)$, and since θ is constant, then the loss per unit length, $dS(z)/dz$, is proportional to $S(z)$, leading to an exponential law. This produces an exponential increase in $\hat{t}(z)$ and a corresponding decrease for the pulse energy.

For specific cases, the stable and unstable areas associated with expression (16) can be easily computed. For the purpose of illustration, we quote some of the results for the cases $Q(2)$, $Q(10)$, and the continuum model.[9] A $Q(2)$ transition will have stable values of pulse angle at $\theta(z)=2\pi$ and $\theta(z)=4\pi$ with the first unstable solution appearing at $\theta(z)=1.06\pi$. The lowest solutions for $Q(10)$ are 2.36π, 4.27π (stable), and 1.37π (unstable). These values, of course, do not exhaust all of the solutions. At sufficiently high spin, the systems approximate classical behavior which in the limit corresponds to a continuum model. In this case the summation appearing on the right-hand side of Eq. (16) becomes an integral of the form $\int n(r)r\sin(r\theta)dr$. Assuming that $n(r)$ is independent of r, $n(r)=n_0$ [this is the appropriate density for correspondence with the limit of a $Q(j)$ transition], then $d\theta(z)/dz$ is proportional to $-j_1(\theta)$, where $j_1(\theta)$ is the spherical Bessel function of the first kind of order 1.[9] Therefore, the first stable value occurs at $\theta(z)\approx 1.43\pi$. For $P(j)$ and $R(j)$, the values for the ratio of the dipole moments is $r_i=[j^2-(j-i+1)^2/j]^{1/2}$. Because r_i takes on irrational values, there are no straightforward solutions of Eq. (16) for this case, even for $j=2$. One notes that for $J\gg 1$, the function r_j is peaked strongly near $r=1$, so that many of the levels, and especially the dominant ones, well tend to stay in phase with one another. For a continuum model, the sum in Eq. (16) is replaced by $\int drr^2(\sin r\theta)/(1-r^2)^{1/2}$. This integral does not reduce to a simple closed form, but its general characteristics will be similar to an ordinary Bessel function. The solutions of Eq. (16) in this case are at 1.2π (unstable) and 2.2π (stable). The clustering of the matrix elements near the maximum means that a $\theta\approx\pi$ to 2π pules will behave more like one propagating in a nondegenerate medium if it is in a $P(j)$ rather than a $Q(j)$ attenuator.

In the case of both degenerate and nondegenerate attenuators, the pulse behavior is determined to a large extent by Eq. (16). If one considers only attenuators of few absorption lengths and pulses whose area is not much greater than 2π, the right-hand side of Eq. (16) is qualitatively similar for $P(j)$, $Q(j)$ and nondegenerate media, so that, qualitatively speaking, one expects considerable similarities in pulse development among the various cases. The major differences arise from the shifting of the values of θ that represent the stable and unstable solutions of Eq. (16). However, in the absence of an independent means of accurately determining the area of the pulse (or equally, the maximum dipole moment), one cannot tell from the experiments which energies correspond to which θ. Indeed one "measures" θ by observing, for instance, the value of $\theta(0)$ for the maximum delay time [approximately at the lowest, unstable solution of Eq. (16)]. Thus the determination of θ will be model-dependent. The single feature of the experiment that is independent of the model is that the "knee," or transition from small-signal attenuation to large-signal "transparency," should occur over no more than an order of magnitude or so of variation of input energy, and should coincide with the maximum delay time. At high θ, however, the right-hand side of Eq. (16) goes to zero for the degenerate case, but not for the nondegenerate case. Thus one expects to see the greatest differences for $\theta\gg 2\pi$. These similarities and dissimilarities are discussed further in the section dealing with the experimental results.

III. RESULTS OF CALCULATIONS FOR $Q(j)$ ATTENUATORS

The propagation behavior of pulses in $Q(j)$ attenuators is presented in this section. Since sufficiently

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[20] We define the quantities $\hat{t}(z)$ and $\mathcal{E}_0(z)$ in the following sense:

$$S(z)=(c\epsilon/8\pi)\int_{-\infty}^{\infty}\mathcal{E}(t,z)dt\equiv(c\epsilon/8\pi)\,\mathcal{E}_0^2(z)\hat{t}(z)$$

and

$$\theta(z)=(\wp/\hbar)\int_{-\infty}^{\infty}\mathcal{E}(t,z)dt\equiv(\wp/\hbar)\,\mathcal{E}_0(z)\hat{t}(z)$$

are two expressions which determine $\hat{t}(z)$ and $\mathcal{E}_0(z)$ unambiguously in the case where no phase reversals in $\mathcal{E}(t,z)$ are present. For the details of this singular case where $\theta(z)=0$ is a possibility, the reader should refer to Frederic A. Hopf, C. K. Rhodes, G. L. Lamb, Jr., and Marlan O. Scully (unpublished). Similar arguments were used by E. Courtens in Chania, Crete, 1969 (unpublished) and by S. L. McCall in Flagstaff, Ariz., 1969 (unpublished).

[21] The actual loss is pulse-shape dependent on account of the inhomogeneous broadening. For $Q(2)$, the $m_j=2$ atoms will generally also produce a loss directly related to the wings of the inhomogeneous distribution even when $\theta(z)=2\pi$. The reader should examine the off-resonance behavior of the state amplitudes for a square-wave pulse at $\theta(z)=2\pi$ and $\theta(z)=\pi$ as an explicit example. The 2π hyperbolic secant pulse is the only known case where the upper state amplitude vanishes independent of the detuning from exact resonance. Thus, the assumption we have made ignores the influence of these effects.

general results are not available in closed analytic form, the following calculations were performed numerically using the equations presented in Sec. II.

The most elementary nontrivial case which exhibits the features of the general result is a $Q(2)$ $(j=2\rightarrow j=2)$ attenuator. Recall that a definite propagating pulse shape can be calculated for $Q(j)$ in the absence of inhomogeneous broadening.[5] The general conclusion of these calculations was "that for a $j\rightarrow j\pm1$ transition (j arbitrary) an undistorted pulse propagating without attenuation cannot exist except for the special cases $0\rightarrow 1$ and $\frac{1}{2}\rightarrow\frac{3}{2}$. However, $j\rightarrow j$ transitions (j arbitrary) admit, in principle, solutions corresponding to lossless, undistorted, finite-energy, propagating pulses. The steady-state pulse shape for $j\rightarrow j$ is multiply peaked with j maxima for integral j and $j+\frac{1}{2}$ maxima for half-integral j."[5] This result, based on the unbroadened[6] model, is unfortunately unable to furnish information concerning the evolution of an arbitrary input pulse.[22] Hence, two points are of direct interest:

(1) Under which conditions, if any, does an arbitrary input pulse develop into the multiply peaked configuration characteristic of the degeneracy?

(2) In the degenerate case, how severe are the losses[5,23] associated with the presence of the inhomogeneous distribution? This will affect the pulse stability.

Calculations, which were performed for $Q(2)$, $Q(3)$, and $Q(4)$ attenuators, produced the following results. It was found that the multiply peaked pulse shape was characteristic of the pulse evolution provided that the input pulse had sufficient area $\theta(0)$. This sufficient area is determined by the condition $\theta(0)\approx 2\pi$ relative to the minimum matrix element. For $Q(j)$ transition, recall that the dipole matrix elements are proportional[24] to the magnetic quantum number m. Thus the input pulse corresponds generally to $\approx 2\pi m$ for the mth sublevel. A precise examination of the $Q(2)$ case revealed a slight deviation from the $2\pi m$ condition. Actually, it was found that the tendency to develop into the multiply peaked configuration was maximized when $\theta(0)=2\pi j+\epsilon$, where the $\epsilon\ll 1$ is associated with a small modification due to the presence of inhomogeneous broadening. Figures 1(b)–1(d) illustrate the results for $Q(2)$, $Q(3)$, and $Q(4)$ attenuators, respectively. In all three cases, the input pulse shape, labeled $z=0$, does not represent a special or preferred $\mathcal{E}(0,t)$; it was taken proportional to $(t/C_0)\exp\{-(t/C_0)^2\}$, where C_0 defines the pulse width, for computational convenience only. Additionally, the time $T_2{}^*$ is taken as unity and the small signal attenuation constant α is identical in all three cases. The attenuation constant α is such that a weak pulse $[\theta(0)\ll 1]$ would sustain approximately a factor of 10^3 attenuation in energy after propagating a distance of 120 cm.

The output pulses clearly exhibit a modulation that is directly associated with the degeneracy. The pulses for $Q(j)$ develop j maxima. Although the angle $\theta(z)$, as expected, is very nearly a constant, these pulses do not propagate without distortion. Furthermore, a finite, albeit very small, loss is associated with the propagation due to the presence of inhomogeneous broadening. This small parasitic loss will eventually cause the pulse to be completely damped, thus destroying SIT in its strictest sense. For the case of $Q(2)$, it is worth noting that the input pulse possesses $\theta(0)=4\pi$ relative to the largest matrix element associated with that transition and that a nondegenerate system with a 4π input pulse would have evolved into two very well separated 2π pulses under the same conditions.[25] With this comparison in mind, one influence of degeneracy is such as to suppress pulse separation.

It is conceivable that different input pulse shapes would follow very different evolutions and arrive at substantially dissimilar output pulse shapes. In order to check this possibility, the propagating pulse shape for $Q(2)$ calculated without considering inhomogeneous broadening was used as the input field. Figure 1(a) shows the results. All the conditions of the attenuating medium were identical to those corresponding to Fig. 1(b). A small loss is again associated with the propagation. Most importantly, however, the output pulse shapes of Figs. 1(a) and 1(b) strongly resemble one another. The general features of the pulse development could depend so severely on the input pulse form that observation of the degeneracy modulated pulse shapes would be impossible. The opposite tendency is actually present.

The result shown in Fig. 1(a) demonstrates that the influence of inhomogeneous broadening on a properly modulated pulse is minimal. The principal reason for this is that these pulses are chosen in such a way that the medium is left with a negligible macroscopic polar-

[22] See footnote 3 of Ref. 5 and the text referred to therein. We point out a critical misprint occurring in that footnote; "$\tau_{\text{pulse}}\ll T_2$" should read "$\tau_{\text{pulse}}\ll T_2$ invalid." Physically, this problem arises since no process is available for *reversible* dephasing in the unbroadened model. In the absence of such dephasing, an arbitrary input pulse will broaden so that $\tau_{\text{pulse}}\approx T_2$. However, the presence of an inhomogeneous width allows a reversible dephasing process to occur which limits the reradiation and enables the inequality $\tau_{\text{pulse}}\ll T_2$ to be preserved for an arbitrary input pulse. Hence, an inhomogeneous width is essential in any discussion concerning pulse evolution.

[23] The 2π hyperbolic secant pulse has the remarkable property that it is effectively 2π independent of the radiator's position in the inhomogeneous distribution; the atomic systems begin in the ground state and end up in the ground state independent of the detuning parameter $\Delta\omega=\omega-\nu$. This is not a general property valid for an arbitrary $\mathcal{E}(t,z)$. In such cases where this property is absent propagation is necessarily accompanied by loss.

[24] For $Q(j)$ transitions, using the conventions of Ref. (5), the matrix elements are $\wp_m=\wp m$, where $\wp$ is a constant and $m=0$, ±1, ±2, ..., $\pm j$. For a symmetric top molecule, the additional κ degeneracy may be included although we have assumed that the κ degeneracy is broken. See C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill Book Co., New York, 1955), p. 96, for the details of these matrix elements.

[25] S. L. McCall and E. L. Hahn, Phys. Rev. Letters **18**, 908 (1968); G. L. Lamb, Jr., Phys. Letters **25A**, 181 (1967).

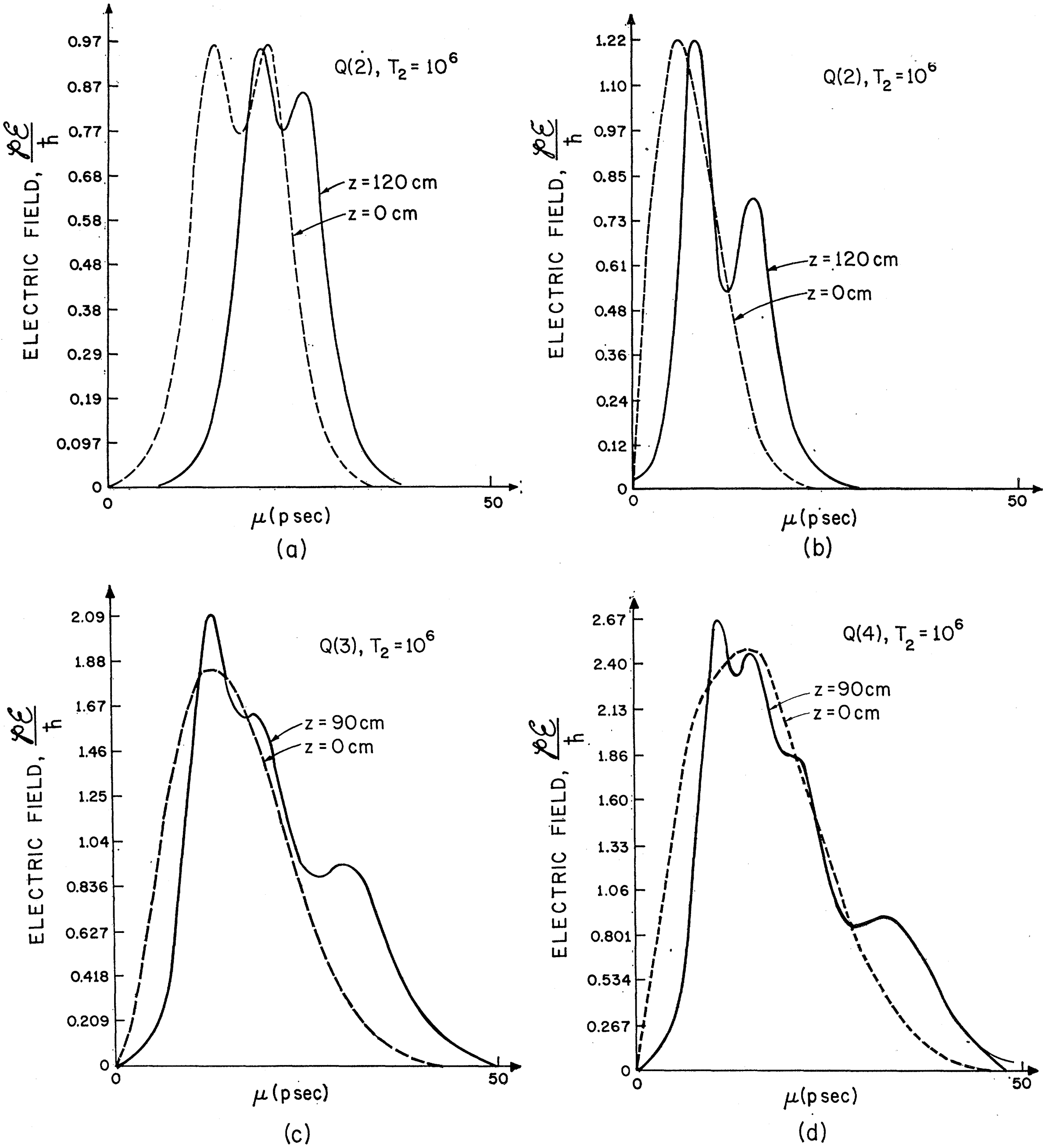

FIG. 1. (a) Time dependence of the electric field envelopes of the input and output pulses for an inhomogeneously broadened $Q(2)$ attenuator using as input data the propagating pulse shape calculated in the absence of inhomogeneous broadening. (b)–(d) Time dependence of the electric field envelopes of the input and output pulses for an inhomogeneously broadened $Q(2)$, $Q(3)$, and $Q(4)$ attenuators. The input pulse is taken in the functional form $(t/C_0)\exp[-(t/C_0)^2]$. The abscissa represents retarded time in units of the inverse inhomogeneous width T_2^*. For the ordinate, $\wp\mathcal{E}(t,\mu)/\hbar$, $\wp$ is the maximum dipole matrix element associated with the transition; it was taken as unity.

ization density. In this situation, one does not have to rely on the inhomogeneous broadening to destroy the macroscopic polarization. The success of the unbroadened model depends on this feature. Thus, the difference of the two pulse envelopes illustrated in Fig. 1(a) is a measure of the contribution of the wings of the inhomogeneous distribution and is readily seen to be small.

We conclude this section with some remarks concerning the origin of the wave form modulation produced by the degeneracy. The effect can be understood in the following sense. Consider a $Q(2)$ attenuator, without inhomogeneous broadening, as an example. In the rotating frame[3] the vectors associated with the two m sublevels precess at rates related by a factor of 2. Thus, at any fixed point z during the entire pulse, one vector precesses a total angle of 4π while the other precesses exactly 2π. We denote the respective precession angles of these two vectors by the ordered pair (θ_1,θ_2) where the subscripts refer to the magnetic quantum number m. Since the medium is initially in phase during the first portion of the pulse $(\approx\delta, \approx 2\delta)$, $\delta\ll 1$, precisely *out* of phase at $(\pi,2\pi)$ midway through the pulse, and very nearly in phase again $(\approx 2\pi-\delta, \approx 4\pi-2\delta)$, $\delta\ll 1$ during the latter portion of the pulse. The modulation minimum occurring on the input pulse $(z=0)$ shown in Fig. 1(a) is naturally associated with the out-of-phase condition. Conversely, the maxima are directly related to the in-phase condition. The presence of inhomogeneous broadening does not alter the basic ingredients of this argument.

IV. EFFECTS OF EXTREME DEGENERACY AND COMPARISON OF CALCULATIONS AND EXPERIMENTAL RESULTS WITH SF_6

In this section, we examine the effects of extreme level degeneracy (the number of participating levels $\gg 1$ with a wide distribution of matrix elements) and compare the results of specific calculations with experimental work[9] on SF_6. Two basic phenomena were studied. One corresponds to very strong excitation $[\theta(0)\gg 1]$ and ≈ 20 absorption lengths in the medium; pulses sharpen at their leading edge, while the front part gets absorbed. The other pertains to much lower excitation and only two absorption lengths; considerable reradiation can be distinguished and long delays result which qualitatively resemble the behavior of a nondegenerate two-level system in SIT.

It is appropriate to emphasize that the region of the SF_6 spectrum which absorbs radiation originating from the $P(16)$ transition[26] of CO_2 is spatially degenerate and most probably consists of many overlapping transitions.[27] The relevant spectral properties of SF_6 have been discussed elsewhere.[9] Since the conclusion of that work, several explicit confirmations of those findings have been disclosed.[28] In the presence of the overwhelming influence of these data, it is difficult to understand arguments[29] that conclude that the SF_6 transition which absorbs the CO_2 $P(20)$ line is a well-resolved[30] resonance of low spin ($j_{\text{upper}}\leq 1$, $j_{\text{lower}}\leq 1$).

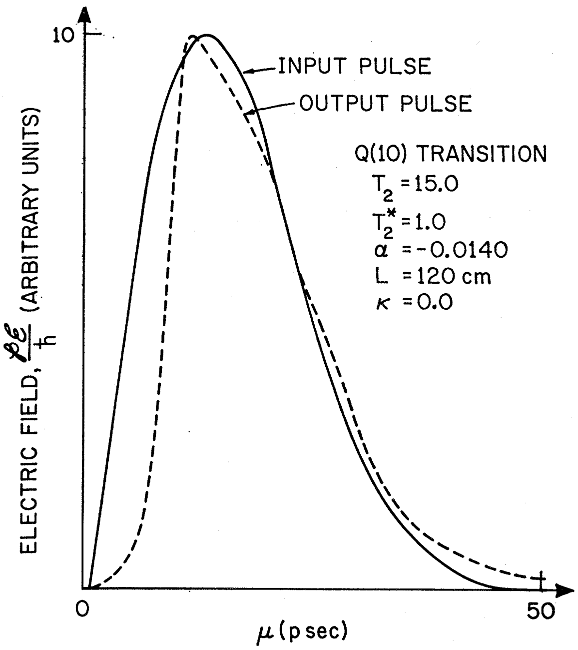

FIG. 2. Time dependence of the electric field envelopes (proportional to the square root of intensity) of the input and output pulses for an inhomogeneously broadened $Q(10)$ attenuator. The input pulse is taken in the functional form $(t/\tau_0)\exp[-(t/\tau_0)^2]$. The abscissa represents retarded time and the ordinate is proportional to the electric field envelope $\mathcal{E}(t,z)$. The parameters α and L refer to a *linear* loss per unit length and the attenuator length, respectively. The pulse sharpening occurs at the leading edge while the remainder of the pulse is essentially unaffected. The corresponding experimental data are shown in Fig. 8 of Ref. 9.

The pulse-sharpening effect arises as a direct consequence of the level degeneracy. As described in an earlier publication (Ref. 9, Sec. IV B), for high intensity input pulses $[\theta(0)\gg\pi]$ the medium is *coherently* saturated in the sense that an additional increment of excitation does not change its macroscopic state. The macroscopic polarization tends to a small quantity limiting both absorption and reradiation; the macroscopic energy content of the medium approaches a constant value which represents a loss from the pulse. The computational results reported here pertain directly to the experimental findings illustrated in Fig. 8 of Ref. 9. These calculations, with the parameters T_2, T_2^*, and α (or equivalently the small signal attenuation) appropriately adjusted to correspond to the experimental situation, showed a pulse-steepening effect remarkably similar to the one observed as Fig. 2 illustrates. Note that the leading edge is depleted while the trailing edge is essentially unchanged. For the purposes of the calculation, the transition was assumed to be $Q(10)$. For this part of the problem the choice of

[26] The $P(16)$ transition to which we refer is the common 10.6-μ laser transition that occurs on the $10°0 \rightarrow 00°1$ rotation-vibration band in CO_2.

[27] Because of the high density of states in the 10.6-μ band of SF_6, the conclusions concerning the $P(16)$ transitions are valid for some other transitions as well [e.g., the $P(18)$ and $P(20)$ lines].

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[30] "Well resolved," in this context, means resolution with respect to the inhomogeneous width; in this case, the Doppler width.

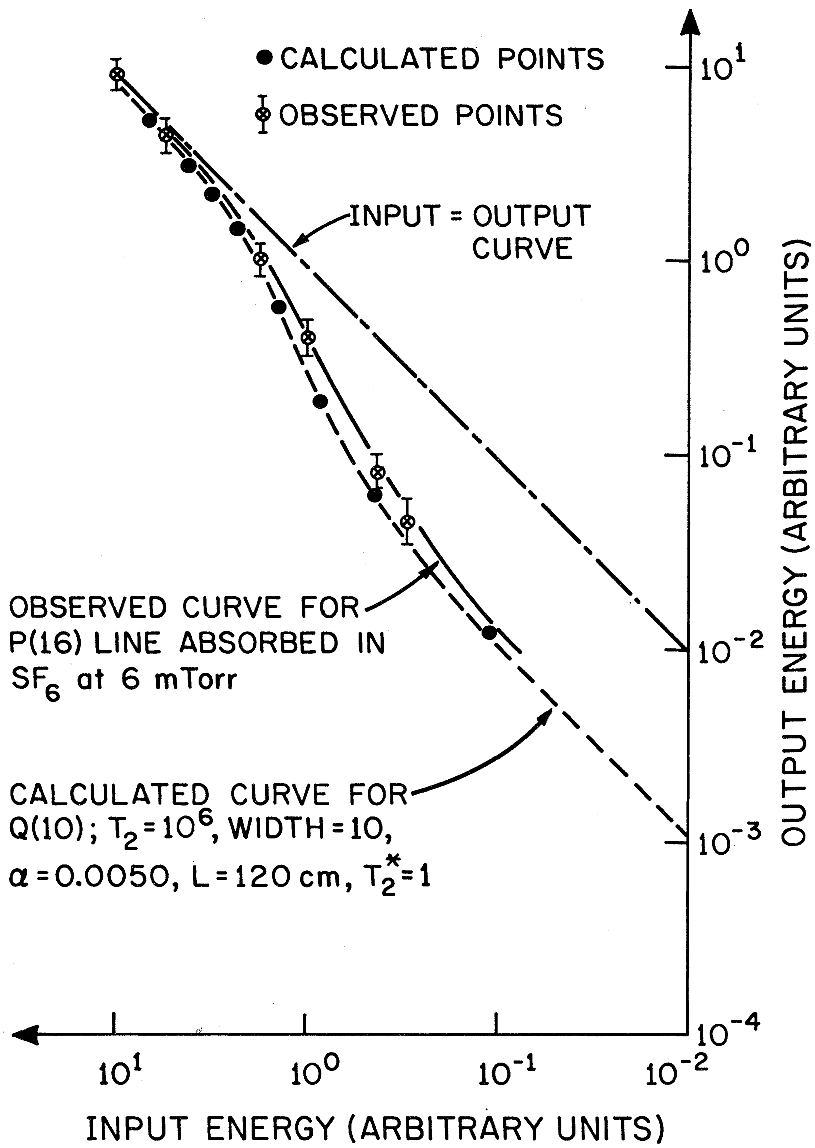

FIG. 3. Calculated and experimental saturation curves. The ordinate is pulse output energy while the abscissa corresponds to the input pulse energy.

a $Q(10)$ transition is merely representative of a situation involving a large range of dipole matrix elements. The results are not influenced appreciable by the choice of another comparable transition.[31] Basically, the pulse-sharpening effect occurs because, for large $\theta(z)$, the distribution of dipoles dephases leaving the system in a saturated[32] state. Hence, the effect emerges as a general phenomenon associated with the response of highly degenerate media to very intense, coherent pulses.

Delay and reradiation phenomena were observed[9] with input pulses whose angles were in the vicinity of π. Since the degeneracy produces a parasitic loss, these phenomena were most prominent in a sample of around two absorption lengths. Again assuming a $Q(10)$ transition, calculations were performed with the parameters T_2, T_2^*, and α appropriately adjusted in correspondence with the experiment. The computed quantities were the output pulse energy versus the input pulse energy (saturation curve) and the output pulse delay versus the input pulse energy (delay curve). The results of these calculations, together with the experimental points, are illustrated in Figs. 3 (saturation) and 4 (delay). We found that the computed saturation curve,

[31] As described in Ref. 9, the identical computations were performed for a P-branch transition with angular momentum quantum number $j=21$, $P(21)$. The conclusion was identical.

[32] This is not a saturation process in the language of rate equations. For high excitation, the individual dipoles tend to cancel thus leading to an effectively saturated state in which the macroscopic properties of the medium (polarization and energy content) are unchanged by an additional increment of excitation. We refer the reader to Secs. IV A and IV B of Ref. 9 for a further discussion of these considerations.

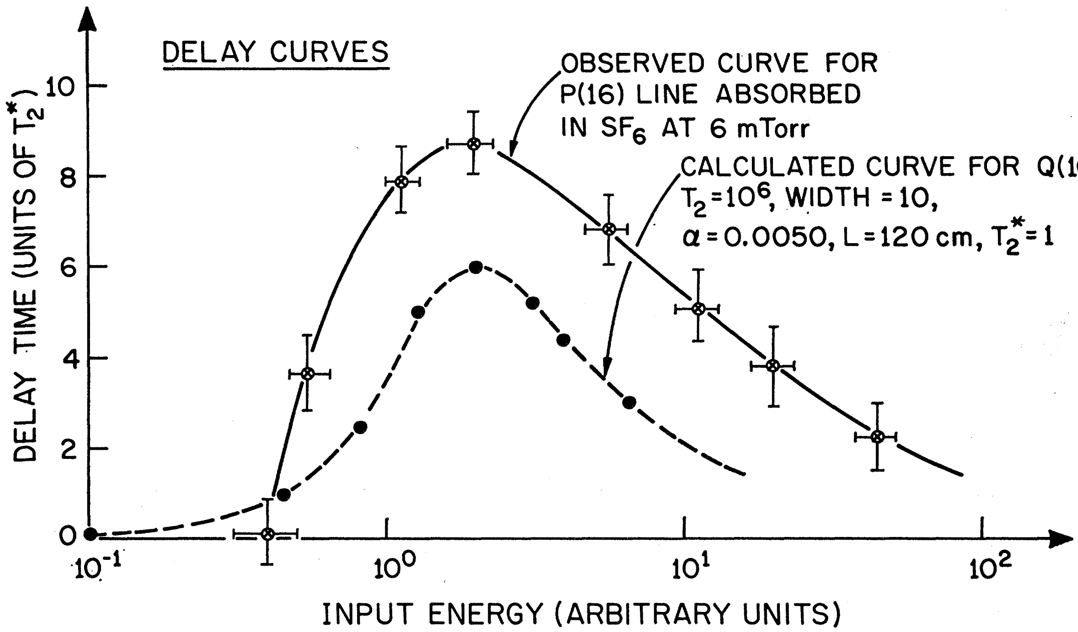

FIG. 4. Calculated and experimental delay curves. The ordinate represents pulse delay (position of the pulse peak) in units of T_2^* while the abscissa of Fig. 3 is in the *same* units as in this figure, indicating pulse input energy.

the computed delay curve, and even the details of the pulse reshaping were all very similar to the experimental data. The agreement of the saturation curves, although manifest, is by itself not sufficient evidence of a genuine coherent effect. Similar behavior is expected on the basis of a simple rate equations model, as indicated by Hopf and Scully.[1] However, the occurrence of the pulse delay in the nonlinear region of the saturation curve constitutes unequivocal evidence of a coherent radiative effect. Theory predicts[33] that the pulse delay should be small at low intensities (i.e., in the linear region), small at high intensities, and reach a maximum at an intermediate pulse input intensity. In Fig. 4, both the calculated and experimental curves exhibit this behavior. Qualitatively, the agreement of these curves is very good.

The discrepancy in the magnitudes of the delay curves is due almost entirely to the shape of the input pulse, although errors are expected to arise due to the inadequacies of the model, the model dependence of certain details of pulse behavior, and the influence of the nonuniformity of the beam. In the theoretical calculations for the delay curves, the input pulse has a very sharp drop at the trailing edge, so that the reradiation occurred in a region in time that was well separated from the initial radiation, i.e., so that if t_p is the time at which the output peak occurred, then $\mathcal{E}(t_p-L/c, L) \gg \mathcal{E}(t_p,0)$.[34] In the experiment, the input pulse had a rapid rise followed by a slow dropoff at the trailing edge, so that it was difficult to determine how much of the radiated peak came from the input pulse and how much from the reradiation. Several computer calculations were done with $Q(6)$ using a triangular input that resembled the experimental input pulse. The result was that the output pulses closely resembled the experimental outputs in all details. The reradiated peaks corresponded to the position of the observed peaks within 10%, and the first peak in the output pulse (coming from the peak of the input), was seen to have a power which varied in direct proportion to the input energy exactly as observed. The situation with $\theta(0) = 1.5\pi$, for $Q(6)$, near the maximum delay was compared with $P(6)$ and the nondegenerate case, giving similar outputs, with the maxima occurring at the same times, but with considerably larger reradiated peaks in the cases of nondegeneracy and $P(6)$. The same run for $Q(6)$ was then checked against the results of $\theta(0)=1.3\pi$ in $P(6)$ and $\theta(0)=1.1\pi$ in a nondegenerate attenuator, so that the comparison was between the outputs near the expected maximum delay.[35] This comparison showed that the reradiated peaks had similar

[33] Frederic A. Hopf and Marlan O. Scully, Phys. Rev. (to be published). Among other things, this work examines the dependence of the pulse delay on the input pulse energy and angle. The calculations performed in connection with this article show that the behavior of a degenerate system is qualitatively similar.

[34] Although the reradiated peak is not much larger than the original radiation, it nonetheless is larger and cannot be due to rate-equation effects. Furthermore, numerical calculations have been made which indicate that no large frequency sweeps or other such phenomena are expected to occur in a nonresonant $Q(j)$ attenuator of a few absorption lengths which might possibly give rise to similar experimental results.

[35] The maximum delay in a nondegenerate system, assuming $\tau_{\text{pulse}} \ll T_2$, occurs at the input angle $\theta(0) \approx \pi$. Refer to Hopf and Scully (Ref. 1) for details.

amplitudes, and there were somewhat larger delays for $P(6)$ and the nondegenerate cases, respectively. The expected similarities between the models was explained earlier, and these calculations indeed show that in the absence of an independent way of determining θ, there is no way to distinguish easily which case one is dealing with if observations are restricted to the region around the maximum delay.

V. CONCLUSIONS

Some modifications of coherent pulse propagation in attenuating media which originate from degeneracy have been examined. It is found that the effect of SIT, first described by Hahn and McCall, is modified in an essential way. Among these modifications are the development of pulse shapes for large θ which are characteristic of the degeneracy, the association of a finite loss with propagation, a reduction of delay times, and the suppression of pulse separation. Unfortunately, the latter three qualitatively tend to modify the propagation behavior in a manner similar to the introduction of a finite T_2. It also appears that, in the presence of attenuation, propagation can occur at constant angle (fixed area attenuation). This implies considerable pulse broadening and resembles nondegenerate SIT, in accord with experimental findings. The resemblance is so close, that on the basis of transmission curves, delay curves, and pulse shapes alone is hard to distinguish between the two.

At high excitations $[\theta(0)\gg\pi]$ sufficiently degenerate media depart from the similarity to the nondegenerate case. A coherent saturation occurs which induces pulse sharpening. This type of behavior provides an essential distinction between degenerate and nondegenerate media. It can be understood without the additional complication of inhomogeneous broadening. Indeed, the influence of inhomogeneous broadening on the high-intensity behavior of a degenerate system can be regarded as arbitrarily small if the saturation width is sufficiently large.

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Analysis of Periodic Schottky Deviations

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The theory of periodic Schottky deviations is studied numerically. Corrections to experimental surface reflection coefficients for several refractory metals are given. The corresponding corrected estimates of the surface inner potentials are in fair agreement with bulk band-theory calculations.

I. INTRODUCTION

An estimate of the electron inner potential of metals in the surface region and the confirmation of proposed shapes of the one-dimensional surface-potential barrier can be made by measuring the complex surface reflection coefficient μ ($\mu=|\mu|e^{i\arg\mu}$).[1] Reflection coefficients that have been obtained from an analysis of periodic deviations in the Schottky effect in thermionic emission from polycrystalline wires have been reported for several metals.[2–4] There are several approximate analytic expressions that describe the periodic deviations.[5,6] However, the expression given by Miller and Good[5] has been favored more recently by experimentalists because the generalized WKB wave functions used by Miller and Good are more accurate for realistic one-dimensional barriers than the usual WKB wave functions.[1,5,7] Recent experimental results, though, have pointed out two discrepancies. First, if the Sommerfeld box model[8] of the surface potential is used to calculate reflection coefficients, it is necessary

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[8] The Sommerfeld box model of a metal is a one-dimensional surface potential model in which the metal's inner potential is a constant $-W_a$. This constant value joins the classical image motive just outside the metal surface. See Fig. 1 and Eq. (1) of Ref. 5, and footnote 9 of Ref. 2.