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Some Results on Coherent Radiative Phenomena with 0π Pulses*

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The propagation behavior of nontrivial 0π pulses $[\int_{-\infty}^{\infty} \mathcal{E}(t, z)\, dt = 0,\ \int_{-\infty}^{\infty} \mathcal{E}^2(t, z)\, dt > 0]$ in attenuating media is examined. It is found that under appropriate conditions 0π pulses exhibit anomalously low absorption which is a direct consequency of atomic coherence. Specific analytical results valid for an unbroadened medium are presented, while the corresponding solutions for an inhomogeneously broadened resonance are calculated numerically. These two results are consistent in the limit of $T_2^* \to \infty$. Furthermore, the influences of T_2, T_2^*, and level degeneracy on the characteristics of propagation are determined. Finally, the behavior of more general 0π pulses is studied. The criteria for the production of 0π pulses are qualitatively established by numerical methods, and the influence of the relative phase of pulse pairs on propagation is examined. The latter indicates a method for the measurement of relative pulse phase.

I. INTRODUCTION

Recent developments in the theory of propagation of electromagnetic radiation in attenuating media have focused attention on self-induced transparency[1,2] (SIT) and other coherent radiative phenomena.[3] In their initial work on SIT, McCall and Hahn[1] showed that an electromagnetic pulse of the form

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

interacting with an attenuator consisting of a resonant inhomogeneously broadened ensemble of two-level atoms, evolved in a manner that could be determined by knowledge of the pulse area $\theta(z)$ as defined by

$$\theta(z) = \frac{\wp^2}{2\hbar^2}\int_{-\infty}^{\infty} dt\, \mathcal{E}(t, z). \qquad (2)$$

The symbol $\hat{\epsilon}$ represents a unit polarization vector, $\mathcal{E}(t, z)$ is the slowly varying pulse envelope,[4] and $\wp$ corresponds to the dipole moment matrix element connecting the two levels of the atomic radiators. In such cases, the area develops according to the equation

$$\frac{d\theta(z)}{dz} = -\tfrac{1}{2}\alpha \sin\theta(z), \qquad (3)$$

where α is the weak-field linear attenuation constant.[5] Under these circumstances, pulses with $\theta(0) = 2n\pi$ have a constant stable area and pass through the attenuator with little or no energy loss. As it propagates, the pulse separates into a series of n pulses, each of which possesses an area of 2π with an envelope that is essentially in the shape of a hyperbolic secant. Pulses with $\theta(0) = 2n\pi \pm \epsilon$ ($\epsilon < \pi$, $n = 1, 2, \ldots$) evolve to the closest stable value [see Eq. (3)], so that, for example, if $\theta(0) = 2\pi - \epsilon$, then $\theta(z)$ tends to 2π, or if $\theta(0) < \pi$, then $\theta(z)$ tends to zero. Consequently, it is held that pulses with $\theta(0) > \pi$ exhibit SIT, while those pulses with $\theta(0) < \pi$ do not. This argument depends critically on the absence of any phase shifts, i.e., that the envelope function $\mathcal{E}(t, z)$ is always nonnegative. If $\mathcal{E}(t, z)$ is allowed to possess both positive and negative values, then the possibility arises that pulses with $\theta(0) < \pi$ might also exhibit SIT. The most obvious case consists of two mutually inverted 2π hyperbolic secant pulses of widths $\hat{t}_1$ and $\hat{t}_2$ well separated by a time T. This paper considers the nature of 0π pulses [i.e., $\theta(z) = 0$] and the manner in which they evolve. It is observed that phase shifts are customarily produced in homogeneously broadened amplifiers[6] and to a lesser extent in multiple-pass attenuators and single-pass attenuators with multiple pulses. Thus, 0π pulse configurations must be taken into account to properly describe the behavior of such systems followed by a single-pass attenuator.

Section II presents the equations governing the behavior of inhomogeneously broadened and unbroadened[7] attenuators. The latter, and considerably simpler model, provides a class of exact solutions for the 0π [$\theta(z)=0$] condition, and it is shown that the pulse evolution obtained from that model very nearly approximates the behavior in the substantially more complicated inhomogeneously broadened case. Section III investigates the dependence of the pulse evolution pattern on dephasing processes, level degeneracy, and the inhomogeneous linewidth of the resonance. Section IV establishes criteria for the production of nontrivial (finite, nonvanishing energy) 0π pulses in attenuating media, along with a description of the circumstances for which pulses with $\theta(0) < \pi$ will exhibit SIT.

II. 0π PULSE PROPAGATION

Since the formulas describing pulse propagation in an inhomogeneously broadened medium have been presented in detail elsewhere,[6] only an elementary outline of the relevant results is given here. The medium of two-level atoms is analyzed with density-matrix methods and is characterized by an absorption constant α, relaxation parameters T_1 and T_2, and inhomogeneous frequency distribution $\sigma(\omega)$.[8] The response of the medium is summarized in terms of a susceptibility χ which is the Fourier transform of the population inversion:

$$\chi(T, t, z) = [2\pi\sigma(\nu)]^{-1} \int_{-\infty}^{\infty} d\omega\, \sigma(\omega) \cos[(\omega-\nu)T] \times [\rho_{aa}(\omega, t, z) - \rho_{bb}(\omega, t, z)]. \quad (4)$$

The function $D(T)$ is defined as the normalized Fourier transform of the frequency distribution. The medium responds to the electric field as

$$\frac{\partial\chi(T, t, z)}{\partial t} = \frac{\wp^2}{2\hbar^2}\, \mathcal{E}(t, z) \int_{t_0}^{t} dt'\, \mathcal{E}(t', z) \times e^{-(t-t')/T_2} \times [\chi(T+t-t', t', z) + \chi(T-t+t', t', z)], \quad (5)$$

where $\wp$ is the dipole matrix element and we have taken the limit $T_1 \to \infty$. Thus, we have

$$\chi(T, t_0, z) = D(T), \quad (6)$$

where the quantity t_0 represents some time before the arrival of the pulse. The field responds to the medium according to the formula

$$\frac{\partial\mathcal{E}(t, z)}{\partial z} + \frac{(1/c)\partial\mathcal{E}(t, z)}{\partial t} = \alpha \int_{t_0}^{t} dt'\, \mathcal{E}(t', z)\, e^{-(t-t')/T_2}\, \chi(t-t', t', z), \quad (7)$$

where

$$\alpha = \wp^2 \nu N \pi \sigma(\nu)/c\epsilon\hbar. \quad (8)$$

N is the density of atomic systems, c is the velocity of light, and ϵ is the dielectric constant of the inert background. The energy of the pulse is directly proportional to the quality $\tau(z)$ which is given by

$$\tau(z) = (\wp/\hbar)^2 \int_{-\infty}^{\infty} dt\, \mathcal{E}^2(t, z). \quad (9)$$

With this normalization, $\theta^2(z)/\tau(z)$ is the pulse length. For the case in which the input energy is sufficiently small, the system behaves linearly and the energy $\tau(z)$ decreases exponentially in z. When $T_2 \gg T_2^*$ and the medium is nondegenerate, $\tau(z)$ is proportional to $e^{-\alpha z}$. The small signal attenuation factor for an absorbing medium of length L cm is defined as $\tau(L)/\tau(0)$, provided that $\tau(0)$ is sufficiently small. The modifications that are associated with degeneracy are examined in Ref. 9, while those arising in the case of $T_2 \approx T_2^*$ are analyzed in Ref. 6.

In the fully coherent limit of $\lim T_1 \to \infty$ and $\lim T_2 \to \infty$, expression (3) can be derived directly from formulas (5) and (7). Furthermore, in the limit $T_2^* \to \infty$, the following simple equation governing the dynamical evolution of the pulse envelope $\mathcal{E}(t, z)$ obtains:

$$\frac{\partial\mathcal{E}(t, z)}{\partial z} + \frac{1}{c}\, \frac{\partial\mathcal{E}(t, z)}{\partial t} = \alpha' \sin\!\left(\frac{\wp}{\hbar} \int_{-\infty}^{t} dt'\, \mathcal{E}(t', z)\right), \quad (10)$$

where

$$\alpha' = \wp^2 \nu N / z c \epsilon \hbar.$$

If the inhomogeneous distribution $\sigma(\omega)$ is assumed to be Gaussian with a standard deviation $2/T_2^*$, then the connection between α and α' is the following:

$$\alpha = \wp^2 \nu N (\pi)^{1/2}\, T_2^*/zc\epsilon\hbar = \alpha' (\pi)^{1/2}\, T_2^*. \quad (11)$$

For the solution of Eq. (10), the electric field envelope $\mathcal{E}(t, z)$ is expressed in terms of a function $\phi(t, z)$ such that $\partial\phi(t, z)/\partial t = (\wp/\hbar)\,\mathcal{E}(t, z)$; then $\phi(t, z)$ satisfies the nonlinear partial differential equation[10]

$$\frac{\partial^2\phi(t', z)}{\partial z\, \partial t'} = -\alpha' \sin\phi(t', z), \quad (12)$$

in which $t' = t - z/c$. Although the general solution of Eq. (11) is unknown, a class of particular solutions may be generated by means of a Bäcklund transformation.[11,12] Two simple types of 0π pulses have been found among these solutions. The first is given by

$$\frac{\wp\mathcal{E}(t,z)}{\hbar}=\frac{A[(2/\hat{t}_1)\,\mathrm{sech}\,X-(2/\hat{t}_2)\,\mathrm{sech}\,Y]}{1-B(\tanh X\tanh Y+\mathrm{sech}\,X\,\mathrm{sech}\,Y)}, \quad (13)$$

where

$$\begin{aligned} A&=(\hat{t}_2^2-\hat{t}_1^2)/(\hat{t}_2^2+\hat{t}_1^2),\\ B&=2\hat{t}_1\hat{t}_2/(\hat{t}_2^2+\hat{t}_1^2),\\ X&=(t-z/v_1)/\hat{t}_1,\\ Y&=(t-z/v_2)/\hat{t}_2,\\ 1/v_1&=(1/c)+\alpha'\hat{t}_2^2,\\ 1/v_2&=(1/c)+\alpha'\hat{t}_2^2. \end{aligned} \quad (14)$$

The pulse profile given by Eq. (13) eventually evolves into two relatively inverted 2π hyperbolic secant pulses whose widths are determined by $\hat{t}_1$ and $\hat{t}_2$ and which have velocities[13] v_1 and v_2. A pulse that behaves in this fashion will be referred to as a "separating" 0π solution. However, if $\hat{t}_2<0$, then expression (13) represents a 4π pulse whose properties have been previously examined.[11] Another solution which corresponds to a 0π pulse is given by

$$\wp\mathcal{E}(t,z)/\hbar=(4/\hat{t}_e)\,\mathrm{sech}X\{[\cos Y-(\hat{t}_f/\hat{t}_e)\,\sin Y\tanh X]\times[1+(\hat{t}_f/\hat{t}_e)^2\,\sin^2 Y\,\mathrm{sech}^2 X]^{-1}\}, \quad (15)$$

where

$$\begin{aligned} X&=(t-z/v_e)/\hat{t}_e,\\ Y&=(t-z/v_e)/\hat{t}_f,\\ 1/v_e&=(1/c)+[\alpha'/(\hat{t}_e^{-2}+\hat{t}_f^{-2})],\\ 1/v_f&=(1/c)-[\alpha'/(\hat{t}_e^{-2}+\hat{t}_f^{-2})],\\ 1/\hat{t}_1&=(1/\hat{t}_e)+i(1/\hat{t}_f),\\ 1/\hat{t}_2&=(1/\hat{t}_e)-i(1/\hat{t}_f). \end{aligned}$$

In contrast to the previously described "separating" solution, this latter solution remains compact, and hence, will be described as "nonseparating." The computer calculations performed for an inhomogeneously broadened medium show examples of both types of propogation.

Figure 1 illustrates a comparison which reveals the influence of inhomogeneous broadening on propagation behavior. As these results show, particular solutions of Eq. (12), valid in the absence of inhomogeneous broadening, can be found analytical-

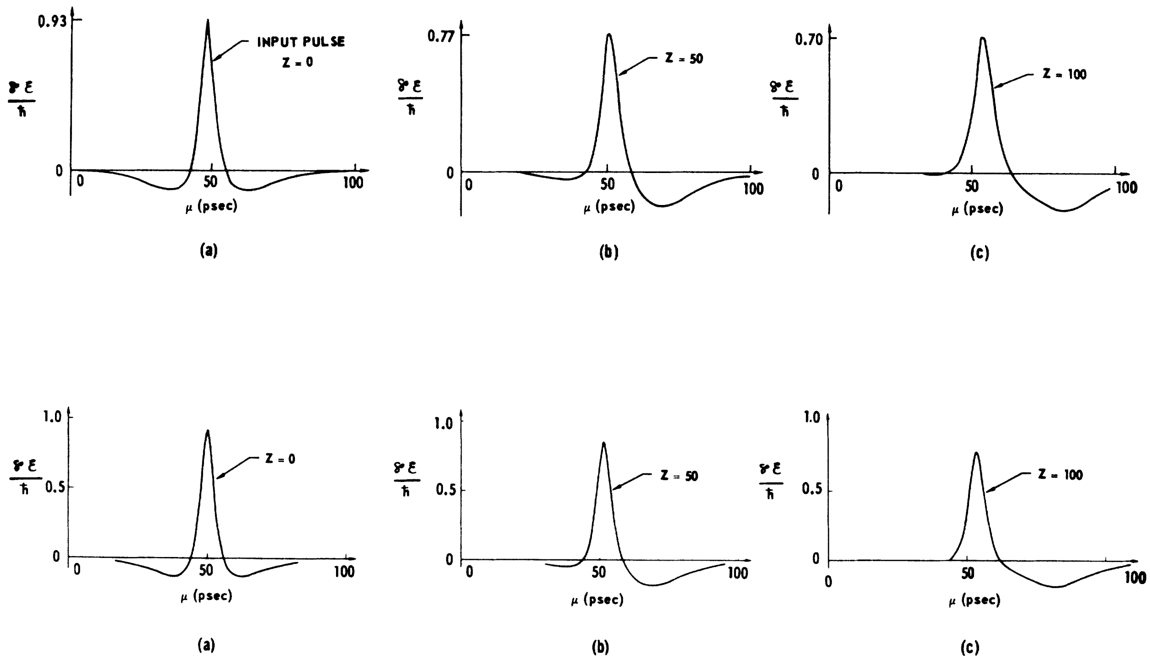

FIG. 1. Electric field envelope propagation behavior for an inhomogeneously broadened attenuator (upper sequence) and for the unbroadened model (lower sequence). The parameters for the former are $T_2^*=1.0$ psec, $T_2=10^6$ psec, and $\alpha=0.0563\ \mathrm{cm}^{-1}$ while those for the latter are $\hat{t}_1=2.7$ psec, $\hat{t}_2=10.75$ psec, and $\alpha'=0.002\ \mathrm{cm}^{-1}$. (a) represents identical input pulses for both cases, (b) $z=50$ cm, and (c) $z=100$ cm. The ordinates are in units of $\wp\mathcal{E}(\mu,L)/\hbar$; the abscissas denote retarded time $\mu=(t-z/c)$ in psec.

ly. Equations (13) and (15) represent two explicit examples. The evolution of expression (13) is shown in the lower sequence ($z = 0, 50, 100$) of Fig. 1. The parameters associated with those curves are $\hat{t}_1 = 2.7$ psec, $\hat{t}_2 = 10.75$ psec, and $\alpha' = 0.002$ cm^{-1}. For the case in which inhomogeneous broadening is present, it is necessary to solve Eqs. (5) and (7) numerically. This solution, for the same input envelope $\mathcal{E}(t, 0)$ as the former, is represented by the upper sequence ($z = 0, 50, 100$) of Fig. 1. The relevant parameters in this calculation are $T_2^* = 1.0$ psec, $T_2 = 10^6$ psec, and $\alpha = 0.0563\ \text{cm}^{-1}$. It is apparent, for these choices of α and α', that the propagation behaviors differ negligibly. The appropriate value of α' was determined by matching curves of the two solutions.

The solution given by Eq. (13) propagates with rigorously conserved energy. Although a small loss is actually present for the inhomogeneously broadened medium, this loss is sufficiently small so that the inclusion of inhomogeneous broadening modifies the propagation behavior in a minimal way. A minor loss term is introduced, but otherwise the pulse-propagation behavior is essentially unaffected (cf. Fig. 1).

Physically, a very simple process is occurring. Absorption takes place at the leading edge of the pulse while this same energy is returned to the pulse tail by stimulated emission. The phase reversals indicated by the nodes of $\mathcal{E}(t, z)$ are associated with the transitions between absorption and emission. The radiators that are on exact resonance begin and terminate their interaction precisely in the ground state. This is not exactly true for radiators that are off resonance, and hence, a slight loss results due to a small net absorption caused by the wings of the inhomogeneous line.

This considerable similarity among the solutions of the two models has been previously noted[11] in regard to 4π and 2π pulses. Both models are soluble for the 2π pulse and lead to the identical analytic result (i.e., an envelope waveform described by a hyperbolic secant). These results strongly suggest that the uncountably infinite class of solutions generated by the Bäcklund transformation technique will closely approximate the corresponding solutions for propagation in an inhomogeneously broadened medium; of course, with the proviso that the range of validity be constrained to sufficiently short distances so that the loss can be properly neglected. This restriction is also necessary because the velocity of propagation of an isolated 2π pulse behaves like $\hat{t}^{-2}$ [for pulse widths $\hat{t}$ short compared to $1/(\alpha' c)^{1/2}$] in the unbroadened model, whereas in the inhomogeneously broadened case this velocity goes as $\hat{t}^{-1}$ (for $T_2^* \ll \hat{t}$).[14] Thus, once the $2n\pi$ or 0π pulse completely separates, the individual pulses will recede from one another at different rates in the two models.

The 0π pulse shapes obtained from the unbroadened model rigorously represent propagation with conserved energy and area and thereby induce a strict transparency in the medium. On this basis it is expected that the "separating" solutions will exhibit SIT in an inhomogeneously broadened medium as well. Indeed, the pulse shown in Fig. 1 has constant energy within the limits of accuracy of the computer program (2–3%). Section III will deal with the nature of propagation of this pulse form in the presence of T_2 and level degeneracies, and under the influence of different values of T_2^*.

III. VARIATION OF MEDIUM PARAMETERS

In this section, various numerical solutions of the nonlinear equations (5) and (7) are considered in relation to the influence of different medium conditions on the development of 0π pulses. The properties of the attenuating medium considered in this section are T_2^*, T_2, and level degeneracy.[9] In each case, the input electric field envelope $\mathcal{E}(t, 0)$ is identical to that shown in Fig. 1(a)(i.e., $z = 0$).

The existence of 0π pulses in an inhomogeneously broadened medium was demonstrated in Secs. I and II. The two solutions shown in Fig. 1 represent a comparison between inhomogeneous by broadened and unbroadened media, in which it was necessary to determine the appropriate value of α' by graphically matching curves. It is natural to examine the limit $T_2^* \to \infty$ using the value of α' implied by Eq. (11). The parameter α varies with T_2^* such that $\lim \alpha = 0$ ($T_2^* \to 0$). Thus since the small-signal absorption grows as T_2^* increases, then for a fixed attenuator length L the product αL is similarly enlarged.[15] This implies that the greater variations in the pulse envelope $\mathcal{E}(t, L)$ are associated with the larger values of α, hence T_2^*. The particular parameters used in this study were $\alpha = 0.0563\ \text{cm}^{-1}$, $T_2^* = 1.0$ psec; $\alpha = 5.85\ \text{cm}^{-1}$, $T_2^* = 104.0$ psec; and in the limit $T_2^* \to \infty$, $\alpha' = 0.0318\ \text{psec}^{-1}\ \text{cm}^{-1}$. In these cases the effect of T_2 was reduced to a negligible level by the selection of a value significantly greater than any of the other characteristic times of the system. The value $T_2 = 10^6$ psec well satisfied this condition. Figure 2 illustrates the output electric field envelope waveforms $\mathcal{E}(t, L)$ corresponding to the two numerical solutions [Fig. 2(b) and 2(c)] and that given by Eq. (13) [Fig. 2(d)]. As expected, it is readily observed that the greater waveform changes are associated with the larger value of T_2^* and that the unbroadened medium solution provides the proper limiting pulse shape as $T_2^* \to \infty$.[16]

The dependence of the output-pulse configuration (given identical input data) on the dephasing parameter T_2 as obtained by numerical solution of formulas (5) and (7) is shown in Fig. 3. In these calculations, the small-signal absorption is held constant

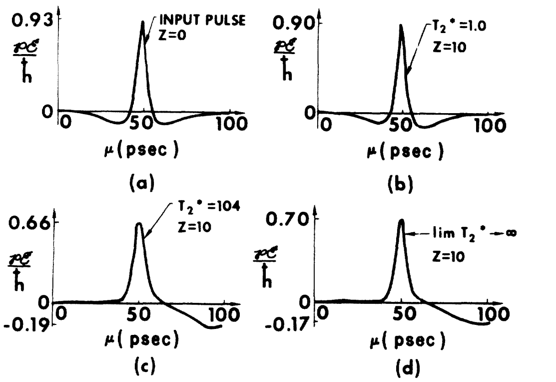

FIG. 2. Electric field envelope vs retarded time for four cases. The ordinates are in units of $\wp\mathcal{E}(\mu, L)/\hbar$; the abscissas are labeled by retarded time $\mu=(t-z/c)$ in psec. (a) Input-pulse waveform ($z=0.0$ cm) used in all cases as given by Eq. (13). (b) Output-pulse waveform ($z=L=10.0$ cm) obtained by numerical integration with $T_2^*=1.0$ psec, $\alpha=0.0563$ cm^{-1}, and $T_2=10^6$ psec. (c) Output-pulse waveform ($z=L=10.0$ cm) obtained by numerical integration with $T_2^*=104.0$ psec, $\alpha=5.85$ cm^{-1}, and $T_2=10^6$ psec. (d) Output-pulse waveform ($z=L=10.0$ cm) given by formula (13) with $\alpha'=0.0318$ cm^{-1}, $\hat{t}=2.7$ psec, and $\hat{t}_2=10.75$ psec.

as T_2 is varied. This involves increasing the value of the density of radiators N as T_2 decreases in order to compensate for the greater width of the atomic resonance. The appropriate value of α corresponding to a given T_2 are determined by methods developed in Ref. 6. The particular values are $\alpha=0.0563\text{ cm}^{-1}$ for $T_2=10^6$ psec; $\alpha=0.0573\text{ cm}^{-1}$ for $T_2=52$ psec; and $\alpha=0.0583\text{ cm}^{-1}$ for $T_2=13$ psec. It is clear that both the positive and negative lobes of the pulse are attenuated and the tendency for pulse separation is suppressed when T_2 is comparable to the pulse width.[17] It is also interesting to note that the delay of the main central peak of the pulse exhibits an independence of T_2, at least over the range of T_2 explored here.

The theory of pulse propagation in a degenerate inhomogeneously broadened attenuating medium is developed in Ref. 9. It is shown there that pulses with stable areas $\theta(z)>0$ do not induce strict SIT in the medium. However, for $Q(j)$ transitions, there exist particular pulse shapes having $\theta(z)=2j\pi$ which essentially drive each sublevel through a multiple of 2π. This greatly minimizes the attenuation and leads to propagation with small (~ 5–10%) loss through many absorption lenghts. However, the 0π pulses represent a constant area condition for all degenerate media [not just $Q(j)$] and leave the radiators, for all sublevels sufficiently near resonance,[18] back in their respective initial states. Thus it might be expected that degenerate media would be transparent to such pulses. However, it is impossible for the separating solutions to exhibit transparency in this manner, since 2π pulses are normally absorbed in degenerate media.

The results of some numerical calculations for certain types of degenerate media are illustrated in Fig. 4. The parameters of the media are adjusted so that the *small-signal* attenuations are identical in all cases. The graph of normalized pulse energy $\tau(z)/\tau(0)$ vs propagation distance z indicates that the attenuation increases with the number of degenerate levels.[19] It is apparent from Fig. 4(e) that SIT, even in approximate form, does not exist for 0π pulses in degenerate media. Furthermore, the pulses do not separate in the fashion suggested by the nondegenerate case. Indeed, the development of the pulses in the $Q(4)$ and $Q(8)$ bears a strong resemblance to the behavior of a "nonseparating" 0π pulse [cf. Eq. (15)]. This result is a consequence of both the nonlinear[20] nature of the interaction and the pulse-shape–dependent response of radiators in the wings of the inhomogeneous distribution.

IV. GENERAL 0π PULSES

This section summarizes the results of several numerical calculations which utilize more general input-pulse waveforms than the particular one used in the previous computations [see Fig. 2(a)]. For the present calculations, the electric field envelope is constructed by a superposition (relatively inverted) of two hyperbolic secant pulses which are 4 psec in width and are separated by 10 psec. The initial area (at $z=0$ cm) of these two constituent pulses are designated by θ_1 and θ_2. For appropriate values of θ_1 and θ_2, these pulses approximate the type of waveforms which are produced in an amplifier and exhibit a phase reversal.[6] Since our interest is primarily directed at 0π pulse configurations, we restrict our attention to inputs for which

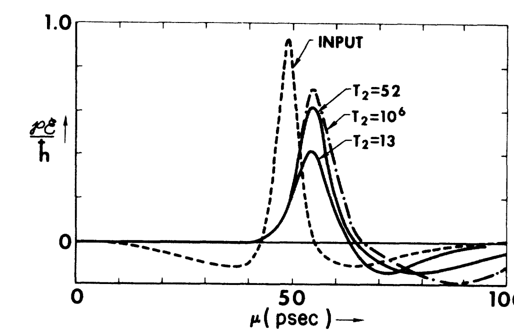

FIG. 3. Dependence of the output-pulse waveforms on T_2 for fixed small-signal attenuation. The ordinate is in units of $\wp\mathcal{E}(\mu, z)/\hbar$; the abscissa is labeled by retarded time $\mu=(t-z/c)$ in psec. The waveforms shown correspond to the input pulse $\mathcal{E}(t, 0)$ and to the output pulses for $T_2=10^6$, $T_2=52$, and $T_2=13$ psec.

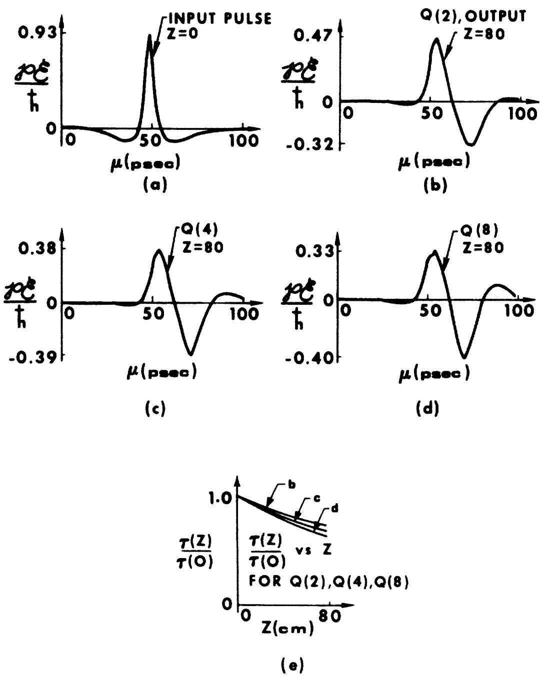

FIG. 4. Dependence of propagation on level degeneracy for $Q(2)$, $Q(4)$, and $Q(8)$ transitions (Ref. 19). For insets (a)–(d), the ordinates are in units of $\wp\mathcal{E}(\mu, z)/\hbar$; the abscissas are labeled by retarded time $\mu = (t - z/c)$ in psec. For (e), the ordinate denotes normalized pulse energy $\tau(z)/\tau(0)$, while the abscissa is in units of propagation length z cm. (a) Input pulse used in all calculations. (b), (c), and (d) Output pulses at $z = 80$ cm for $Q(2)$, $Q(4)$, $Q(8)$ attenuators, respectively. (e) Normalized pulses energy $\tau(z)/\tau(0)$ vs z.

$0 \leq \theta_1 + \theta_2 < \pi$ is valid. The media considered in this section are identical to that described in Sec. II.

In the absence of analytic techniques, there does not appear to be any way of precisely determining the conditions for the development of SIT for general values of θ_1 and θ_2. Therefore, the values of θ_1 and θ_2 used as limiting values must be regarded as highly approximate. Nevertheless, three clearly distinct pulse behavior patterns were found. The first is represented by a pulse that is absorbed in a straightforward manner. In that case, θ_1 and θ_2 are small (i.e., $\ll \pi$) and the energy-vs-z curve $[\tau(z)]$ conforms to Beer's Law.[21] For sufficiently small θ_1 and θ_2, this is expected on the basis of an argument which considers the two lobes as interacting independently with the medium. This approach is equivalent to a superposition principle and explicitly ignores the nonlinear properties of the interaction. Hence, its validity is restricted to a certain domain of sufficiently small angles θ_1 and θ_2. The computer results indicate that this domain is given by[22] $|\theta_1| < \frac{1}{2}\pi$. This is consistent with the results for a single pulse,[23] since $\max(\theta_1 + \theta_2) < \pi$.

The opposite extreme occurs if both lobes possess angles in the vicinity of 2π(i.e., $\theta_1 \cong 2\pi$, $\theta_2 \cong -2\pi$). "Separating" 0π pulses result in this case. The computer estimated limits for this situation are $\theta_1 \geq \frac{3}{2}\pi$ and $\theta_2 \leq -\pi$. Two examples are presented: Figure 5 illustrates the energy-vs-distance $[\tau(z)]$ curves, while Fig. 6 shows the time dependence and evolution of the pulse envelope. The second row (b) in Fig. 6 is an exemplification of direct separation of the two lobes. The areas of both lobes tend to 2π pulses. The corresponding curve (b) of Fig. 5 indicates that very little loss is associated with this propagation. In the third row of Fig. 6, the pulse behavior for the reversal of the lobe order (smaller first) is shown.[24] If θ_1 is small enough, it forms into the wider, hence slower, of the resulting 2π pulses and "passes through" the other lobe. Surprisingly, the nonlinear features of the interaction occurring when the two lobes are superimposed are not conspicuous. Curve (c) in Fig. 5 presents the corresponding $\tau(z)$ curve.

Another possibility involves pulses which have envelopes resembling the form expressed by Eq. (15) (i.e., a modulated hyperbolic secant). One example is illustrated in row (a) of Fig. 6; the corresponding energy curve $[\tau(z)]$ is curve (a) of Fig. 5. These pulses give no indication of any separation into individual components. However, they appear to go asymptotically to a nonvanishing energy.[25] This behavior is similar to simple π or 2π in the case of ordinary SIT. They exhibit large "delays" at various input energies down to some critical value. This value is not easily determined because of the

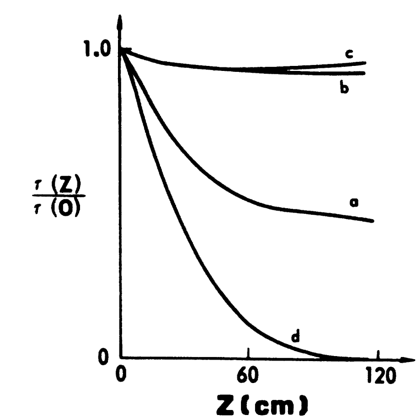

FIG. 5. Normalized pulse energy $\tau(z)/\tau(0)$ vs propagation distance z in cm. (a) For the pulse sequence (a) of Fig. 6; (b) for the pulse sequence (b) of Fig. 6; (c) for the pulse sequence (c) of Fig. 6; (d) the result corresponding to a pulse with $\theta(0) = 0.9\pi$ (no phase reversals) propagating in an attenuator identical to that for (a)–(c) above.

limitations of the computer program. At that point, no "delays" will occur. An example of a simple (no phase reversal) 0.9π pulse is offered in curve (d) of Fig. 5 as a contrast to the other cases. That pulse while developing nonlinearly is, nevertheless, very strongly absorbed.

The presence of a phase shift can influence the gross properties of the pulse evolution pattern. In order to isolate the phase-shift effect, further computations on the propagation of pulse pairs were performed. The shapes of the two pulses were assumed to be hyperbolic secants with $|\theta_1| = |\theta_2|$.[26] The widths of these pulses were 4 psec, while the relative delay (peak separation) was 10 psec. The evolution behavior of the pulse pairs, with and without phase reversal, was examined as a function of θ.[26] For these calculations,[27] the small-signal attenuation constant α was $0.0563\ \text{cm}^{-1}$ and the attenuator length L corresponded to 120 cm. T_2^* was 1 psec, and T_2 was 10^6 psec. The relevant comparison examined the intensity $I(t, L)$ of the output pulses for the phase reversed and nonphase reversed inputs. It was found that for $\theta = 2\pi$, the output intensity showed a negligible dependence on the phase reversal. This is expected, since the first 2π hyperbolic secant pulse leaves the atomic systems back in their ground states throughout the entire inhomogeneous distribution. Since no phase information is contained in this state, the propagation of the second pulse is unaffected by the relative phase of the first. However, for $\theta \neq 2\pi$, the atomic radiators do possess phase information significantly altering the behavior of the phase reversed and nonphase reversed pulse pairs. The general tendency of this difference is indicated by consideration of the case $\theta = 1.25\pi$. The in-phase pulses evolved into a single 2π pulse, while the phase reversed case maintained two clearly distinct pulses. The difference in the output waveforms, between one pulse and two, was obvious. In fact, if $\pi < \theta < \frac{3}{2}\pi$, there is a general inclination for the in-phase pairs to merge into a single pulse while the phase reversed pairs favor a multipulse configuration. Also, the loss associated with propagation is significantly less for the phase reversed situation. This behavior is preserved, with slight modification, when degenerate levels are considered as explicit calculations for a $P(7)$ transition indicate. This effect enables the determination of relative optical phase and is a nonlinear analog of ordinary interference.

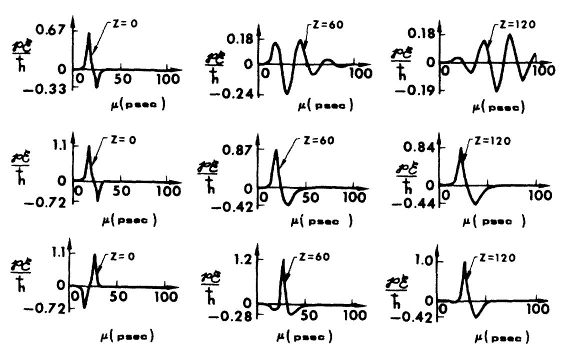

FIG. 6. Three propagating pulse sequences (a)–(c). (a) is the top line (i), (ii), (iii); (b) is the middle line (iv), (v), (vi); (c) is the bottom line (vii), (viii), (ix). The ordinates are in units of $\wp\mathcal{E}(\mu, z)/\hbar$; the abscissas are in units of retarded time $\mu = (t - z/c)$ in psec. The propagation distance z in cm is noted in each frame.

V. CONCLUSIONS

The nontrivial 0π pulse has been shown to exist, and by virtue of an energy loss associated with short T_2, is clearly a direct consequence of atomic coherence. The waveforms produced in an inhomogeneously broadened medium are similar (but not identical) to those associated with the unbroadened model.[7] It is noted that two different forms of 0π pulses are expected to occur. The "separating" variety, which is the analog of the well-known 4π pulse, evolves into two distinct (relatively inverted) 2π pulses. The second type or "nonseparating" pulse, which is the analog of the ordinary 2π pulse, remains intact. It evolves into the form of a rapidly modulated hyperbolic secant with large delay times determined by the input-pulse widths and energies, and whose modulation is specified by the initial peak separation. The former results are expected whenever the positive and negative lobes are near 2π, and the latter when both lobes are near π. These two species of 0π pulses exhibit SIT. However, if both lobes are small ($\ll \pi$), the pulse is absorbed in the normal fashion according to Beer's Law. Furthermore, it is possible to produce curves of output energy vs input energy and delay time vs input energy, using only 0π pulses as inputs, which closely approximate those obtained from the ordinary SIT effect without phase reversals.

Therefore, it is clear that considerable care must be exercised in predicating predictions on Eq. (3) if pulses possessing pulse shifts are involved. These shifts can occur in amplifiers[6] and multiple-pass attenuators.[28] They can also originate in an attenuator in which T_2^* and T_2 are as long as or comparable to the pulse width. These conclusions can be inferred from the following: (i) the fact that the waveforms calculated for the inhomogeneously broadened medium go smoothly over into those associated with the unbroadened model for the limit $T_2^* \to \infty$[29]; and (ii) that input pulses which have areas differing from $2n\pi$ develop phase reversals in the unbroadened attenuator. Thus, for example, a pulse with $\theta(0) > \pi$ could form phase shifts and separate into a sequence of alternately inverted 2π pulses.

It should be pointed out the $\theta(z)=0$ condition does not necessarily imply nearly lossless propagation. The nonlinearity of the interaction manifests itself in such a way that sufficiently weak zero-area pulses are damped in the normal Beer's law fashion. More precisely, the pulse-evolution patterns shown in Fig. 1 do not follow for a similarly shaped input pulse whose amplitude is reduced by an order of magnitude. Strong damping results. Thus, under comparable input conditions, this feature causes degenerate attenuators to have more losses than nondegenerate ones, since the ensembles with the relatively small dipole matrix elements experience a correspondingly weaker interaction. This produces a residual attenuation which is roughly proportional to the degeneracy.

These calculations also reveal the sensitivity of the propagation behavior on the relative phase of pulse pairs. This optical phase can be determined by its influence on the propagation. Less loss also generally results for the phase reversed pairs *vis-à-vis* the in-phase pair.

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[4]The electric field envelope $\mathcal{E}(t,z)$ is slowly varying in the following sense: $|\partial\mathcal{E}(t,z)|/\partial z \ll |\kappa\mathcal{E}(t,z)|$ and $|\partial\mathcal{E}(t,z)|/\partial t \ll |\omega\mathcal{E}(t,z)|$ for all t and z. Hence, the left-hand sides of the inequalities can always be neglected in comparison with the right-hand sides. This considerably simplifies the equation of motion for $\mathcal{E}(t,z)$ by the elimination of those derivative terms.

[5]See expression (8) and the associated text for a precise definition of the linear attenuation constant α.

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

[7]The concept of an unbroadened attenuator sometimes causes confusion. It is a model of noninteracting radiators each of which is on exact resonance. The approximations implied in this model are valid in the limit of sufficiently small inhomogeneous broadening and sufficiently long relaxation times T_1 and T_2.

[8]α is the small-signal attenuation constant, the relaxation parameters T_1 and T_2 correspond to the energy transfer and dephasing times, respectively, and the inhomogeneous distribution function $\sigma(\omega)$ which has a width in frequency space of $\Delta\omega_{\text{inhom}}$. The full width at half-height of $\sigma(\omega)$ is $\Delta\omega_{\text{inhom}}=3.3/T_2^*$.

[9]Frederic A. Hopf, Charles K. Rhodes, and Abraham Szöke, Phys. Rev. B 1, 2833 (1970).

[10]The electric field envelope $\mathcal{E}(t,z)$ is always assumed to vanish in the limit $t\to-\infty$. Hence, $\lim \mathcal{E}(t,z)=0$ as $t\to-\infty$ for all $z\epsilon[0,L]$.

[11]G. L. Lamb, Jr., Phys. Letters 25A, 181 (1967).

[12]A. Seeger, H. Donth, and A. Kochendorfer, Z. Physik 134, 173 (1953).

[13]The velocities v_1 and v_2 correspond to the velocities of the two maxima of $|\mathcal{E}(t,z)|$.

[14]For "nonseparating" solutions the loss consideration will dominate, whereas for "separating" solutions, the velocity effect will prevail.

[15]Recall that physically the pulse shape evolves through a continuous process of absorption and emission. However, since the energy must be absorbed before it can be reemitted, and since the number of absorption lengths αL is a measure of the number of times this absorption-emission cycle is repeated, the product αL is an index of the pulse evolution.

[16]From the point of view of these calculations, this limit is essentially achieved when $T_2^* \gg$ than the pulse width $\hat{t}$. In this case, the relevant pulse width $\hat{t}$ is ~ 10 psec, so that $T_2^*=104.0$ psec can be regarded as a valid approximation of infinity.

[17]The relevant pulse width is determined by the time scale of significant variations in the pulse envelope, and thus, can conceivably depend upon z. A good estimate of this time is given by the distance between the nodes of pulse envelopes ~ 20 psec.

[18]In the frequency domain, "sufficiently near resonance" denotes a strip in the inhomogeneous distribution $\sigma(\omega)$ of width 2π times the inverse pulse width $\hat{t}$ which is centered at the pulse carrier frequency ν. Hence, describing the frequency interval $\nu-\pi/\hat{t}\le\omega\le\nu+\pi/\hat{t}$.

[19]Since the transition examined was $Q(j)$, the number of participating degenerate levels is $\frac{1}{2}(2j+1-1)=j$. Hence the degeneracy of the medium is directly proportional to the angular momentum quantum number j.

[20]In this context, the term "nonlinear" refers to the dependence on the magnitude of the optical electric field $|\mathcal{E}(t,z)|$.

[21]Beer's law predicts an exponential decay by the small-

signal attenuation constat α in cm^{-1}. More precisely, $\tau(z)/\tau(0)=e^{-\alpha z}$.

[22]This domain is then equivalent to the interior of a square of side π, centered at the origin, in the θ_1-θ_2 plane.

[23]It is well known that single pulses with an area $\theta(0)<\pi$ do not exhibit SIT; see Ref. 1.

[24]Thus, the limits on θ_1 and θ_2 given above can be equivalently expressed as $\theta_1 \leq -\pi$ and $\theta_2 \geq 1.5$.

[25]The $\tau(z)/\tau(0)$ curve (a) in Fig. 5 is artificially depressed for $z \geq 100$ cm, since the calculation of $\tau(z)$ is truncated at $\mu=100$, and part of the pulse waveform obviously extended beyond that point. See Fig. 6(iii).

[26]Since $|\theta_1| = |\theta_2|$ in the following discussion, we define $|\theta_1| = |\theta_2| \equiv \theta$.

[27]The medium is assumed to be nondegenerate.

[28]Frederic A. Hopf (unpublished).

[29]This has been observed for π pulses in amplifiers and for 0π and 2π pulses in attenuators.

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Light-Scattering Measurement of Concentration Fluctuations in Phenol-Water near its Critical Point*

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The intensity and spectral width of light scattered by a critical mixture of phenol and water have been measured as a function of temperature both above and below the critical temperature T_c. The temperature dependence of the scattered intensity was fitted to $I \propto (T-T_c)^{-\gamma}$ for $T>T_c$ and $I \propto (T_c-T)^{-\gamma'}$ for $T<T_c$. Also measured was the ratio $R_I \equiv I(\Delta T)/I(-\Delta T)$ of intensities scattered at a given temperature interval ΔT above and below T_c. The measured values of γ, γ', and R_I were quite close to the predictions of the 3-*D* lattice gas model. The spectral width Γ was measured using a photon correlation method, and the data were fitted to $\Gamma = DK^2\,(1+K^2\xi_\Gamma^2)$, with the diffusion constant $D=D_0|T-T_c|^{\gamma^*}$ and $\xi_\Gamma=\xi_{0\Gamma}|T-T_c|^{\nu_\Gamma}$ The Fixman term $K^2\xi_\Gamma^2$ (K being the photon momentum transfer) was observed only above the critical temperature. The value of ν and the values of γ^* both above and below T_c were in fairly good agreement with the theory of Kadanoff and Swift. The spectral width measurements also provided the ratio $R_D \equiv D(-\Delta T)/D(\Delta T)$, a quantity for which no theoretical prediction exists. Comparison of this work on phenol-water with that of Swinney and Cummins and others on CO_2 near its gas-liquid critical point reveals remarkable similarities between the two systems.

I. INTRODUCTION

The last few years have witnessed a rapid growth of interest in the critical behavior of systems undergoing a second-order phase transition. Experiments in a wide variety of systems including ferromagnets, simple fluids, and binary liquid mixtures, reveal a striking similarity in their behavior near the critical point. This similarity is a reflection of the fact that fluctuations which develop near the critical point are of sufficiently long range as to be remarkably insensitive to the detailed form of the atomic interactions in the system.[1] As a result, the critical behavior of many of these systems can be characterized by a small number of dimensionless parameters.

The work described here is a study of the temperature dependence of the magnitude and lifetime Γ^{-1} of concentration fluctuations in a critical mixture of phenol and water. Measurements were made both above and below the critical temperature T_c. The lifetime measurements below T_c appear to be the first which have been reported for a binary mixture. A preliminary account of this work has already appeared.[2,3]

The temperature dependence of the magnitude of the fluctuations, which can be characterized by the exponents γ and γ', was measured by following changes in the average intensity I of light scattered by the system. The lifetime of the concentration fluctuations was determined by using a photon correlation method to analyze the fluctuating light intensity. The lifetime is inversely proportional to the mutual diffusion coefficient D whose temperature dependence is described by the exponent γ^*.

In addition to determining γ and γ^*, measurements were made of the following dimensionless intensity and diffusion coefficient ratios:

$$R_I = I(\Delta T)/I(-\Delta T);\; R_D = D(-\Delta T)/D(\Delta T)\,. \quad (1)$$

Here the argument $\Delta T(-\Delta T)$ refers to measurements made at equal temperature intervals above (below) T_c. The experimentally determined values of γ and R_I can be compared with predictions of the three-dimensional (3-D) lattice gas model.[4,5] Recent theoretical work of Kadanoff and Swift[6,7]