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Observation of the transparency of a resonant medium to zero-degree optical pulses*

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Experiments are described in which low-intensity laser pulses of zero area $[\int_{-\infty}^{\infty} \mathcal{E}(z,t)\,dt = 0]$ are propagated through a degenerate resonantly absorbing medium with greatly reduced absorption. These pulses are constructed either electro-optically or by allowing a non-zero-degree pulse to evolve toward zero area by means of a resonant absorption and reradiation process. We observe the transmission of as much as 65% of the energy of such pulses through a resonant absorber which attenuates the same cw laser by $e^{-\alpha L}$, with $\alpha L \approx 20$.

The transparency of a resonant absorber to pulses of large "area", $\theta = 2n\pi$ $(n = 1,2\ldots)$, has been a subject of active experimental[1] and theoretical[2] interest. In an early publication[3] discussing the inhibiting influence of level degeneracy on the transparency of large-area pulses, it was noted that zero-area pulses can propagate with low loss, irrespective of the degeneracy of the resonant transition. Such "zero-degree" pulses result if the pulse envelope undergoes a sign change in such a way that the tipping angle,

$$\theta_m(z,t) = (\mu_m/\hbar)\int_{-\infty}^{t} \mathcal{E}(z,t)\,dt,$$

goes to zero for $t \to \infty$. Here, $\mathcal{E}(z,t)$ is the envelope of the electric field, $\mathbf{E} = \hat{\epsilon}\mathcal{E}(z,t)\cos(\omega t - kz)$, and μ_m is the dipole matrix element for the transition involving the mth degenerate sublevel. The area $\theta_m(z,\infty) \equiv \theta_m(z)$ measures the degree of excitation of resonant molecules after passage of the pulse.[4] Recent articles have analyzed additional features of zero-degree pulse propagation.[5-7]

We describe here experiments in which zero-degree pulses have been constructed and propagated through a degenerate resonant absorber with dramatically reduced energy loss. We have also observed the evolution of small-area pulses into zero-degree pulses due to resonant absorption and reradiation process,[7,8] discussed in more detail below. The observations are made in the limit of low pulse intensities corresponding to small tipping angles. In this limit, where the atomic response can be treated in the harmonic oscillator approximation, the energy loss and pulse distortion can also be obtained by considering the absorption and dispersion of each Fourier component of the pulse separately, as discussed in Ref. 7.

A major requirement for low-loss propagation is that the pulse be able to coherently excite and deexcite a molecule in a time short compared to the homogeneous transverse relaxation time, T_2. For a $2n\pi$ pulse $(n \geq 1)$, the pulse energy must be sufficiently large to induce n complete transitions. Because of this, the required pulse energy (proportional to $1/\tau_{\text{pulse}}$) may be excessively large in cases where T_2 is very short. On the other hand, a zero-degree pulse need not induce a complete transition; hence, it can propagate with low loss even with small energy. If the absorption line is inhomogeneously broadened, small-energy zero-degree pulses will still propagate with low loss, provided the pulse envelope changes sign before the molecules can dephase due to the spread in their frequency distribution.[9]

The experimental arrangement is shown in Fig. 1(a). A cw laser with an internal mirror and diffraction grating line selector is operated on single P- or R-branch transitions of CO_2 or N_2O in the 10-μ region. The linearly polarized output of the laser is passed through a GaAs electro-optic modulator crystal with its **E** field oriented relative to the crystal axes[10] as shown in Fig. 1(b). The crystal is pulsed with a 5-kV/cm square pulse derived from a pressurized spark gap and an RG8/U-coaxial-cable pulse-forming network. The rise and fall time of the pulse is less than 0.3 nsec, and the duration can be varied by changing the length of the charging cable [C in Fig. 1(a)]. In the experiments reported here, pulses of 2- or 6-nsec duration were used. The pulse is propagated over the crystal, which is matched to 50 Ω by adjusting the capacitance of the crystal holder.

The voltage pulse can be applied to the crystal a second time with the same or opposite polarity by allowing it to reflect from the end of an open or shorted 50-Ω cable [R in Fig. 1(a)] which terminates the crystal. Single pulses are obtained by terminating the cable with a 50-Ω resistor. For an incident field $E_0 \sin(\omega t - kz)$ polar-

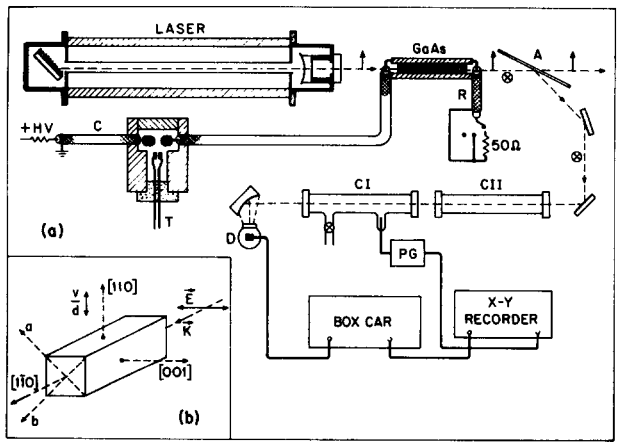

FIG. 1. (a) Schematic of experimental apparatus. Cell CII is in place only as described in text. The integrated output of the detector (D) is recorded as a function of pressure in cell CI. (b) Orientation of the GaAs modulator.

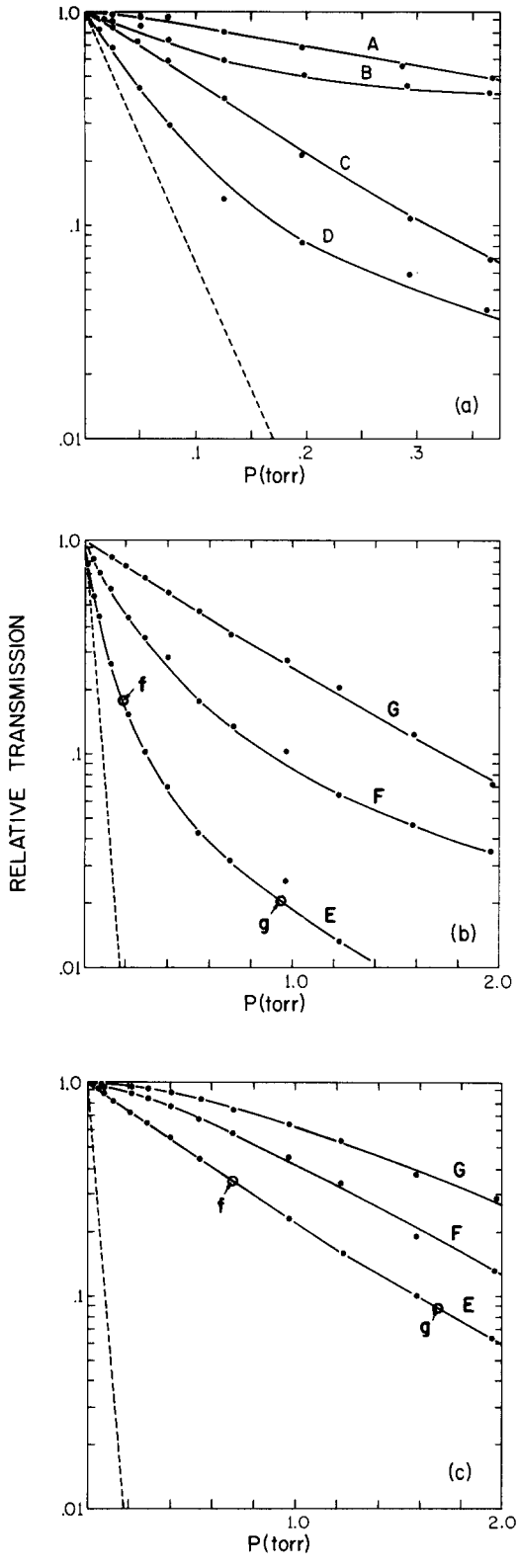

FIG. 2. Transmission of N_2O laser energy through the resonant NH_3 absorber as a function of pressure. Dashed lines give cw absorption. (a) Transmission through cell CI only for (A) 2-nsec zero-degree pulses, (B) 2-nsec in-phase pulses, (C) 6-nsec zero-degree pulses, and (D) 6-nsec in-phase pulses. (b) Transmission of single 6-nsec pulses through cell CI with cell CII filled at fixed pressures: Curve E is transmission with CII empty; curves F and G show transmission with CII filled to give absorption indicated by points f and g, respectively, on curve E. (c) Same as (b), for single 2-nsec pulses.

ized as shown, the output in the perpendicular polarization, which is selected by analyzer A, is given by $E_0 \cos(\omega t - kz) \sin\Gamma$, where $\Gamma = \pi n_0^3 r_{41} Vl/\lambda d \approx \pi/8$ is the phase advance or retardation (determined by the sign of the electric field strength V/d) along axes a or b [Fig. 1(b)].[11]

The transparency effect was observed in CO_2, with the use of a 1.5-m heated gas cell, as well as in NH_3. The $\nu_2[asQ(8,7)]$ transition of $^{14}NH_3$ is coincident to within 10 MHz with the $P(13)$ 10.6-μ N_2O laser line and has a doppler half-width of 42 MHz.[12,13] The NH_3 absorption coefficient of 0.7 cm^{-1}/Torr is considerably larger than in CO_2 and, hence, allowed detailed studies for many absorption lengths. Figure 2 shows a logarithmic plot of experimental data for the transmission of 0.01-W/cm^2 $P(13)$ 10.6-μ N_2O laser pulses through a variable-pressure[14] 40-cm NH_3 absorption cell [CI in Fig. 1(a)]. Curve A in Fig. 2(a) shows the transmission of zero-degree pulses with two out-of-phase 2-nsec lobes, which are shorter than $T_2^* = 1/2\pi\Delta\nu \approx 4$ nsec (where $\Delta\nu$ is the Doppler half-width of the absorbing transition). The dashed line shows for comparison the transmission of low-intensity cw radiation as given by $e^{-\alpha L}$, where α is the linear absorption coefficient and L is the length of the sample cell. At low pressures, the number of absorption lengths, αL, is proportional to pressure since the pressure broadening is small compared to the Doppler width.[15] The effective absorption coefficient for the zero-degree pulses may be obtained from the logarithmic slope of curve A. By comparison with the cw absorption (dashed line), we note that at low pressures the absorption coefficient of the 2-nsec zero-degree pulses is smaller by a factor of more than 25 than the value of 0.7 cm^{-1}/Torr. (At pressures above 75 μ, corresponding to $\alpha L \approx 2$, the slope in curve A increases slightly, indicating an increased absorption; this is believed to be due to a small delay between the two pulses, which can result in an interference between the second pulse and the molecular reradiation after the first pulse).

For a short-duration pulse of nonzero area, the absorption coefficient is expected to be smaller than α when its duration is less than T_2^*. In this case, the energy absorbed becomes proportional to the pulse duration rather than the inverse linewidth. For a thin sample ($\alpha L < 1$), this reduction is not as great as for the zero-degree pulse of the same duration. For $\alpha L \gg 1$, propagation effects cause the pulse area to evolve toward zero (see below). Curve B in Fig. 2(a) shows the transmission of a pulse of nonzero area, consisting of two in-phase 2-nsec pulses. The initial absorption obtained from the slope of this curve is less than the cw absorption, but larger by a factor of 5 than that of the zero-degree pulse. At higher pressures, corresponding to several absorption lengths, this curve begins to flatten out and its slope becomes characteristic of that of a zero-degree pulse.

Similar behavior is seen for 6-nsec zero-degree pulses (curve C) and in-phase pulses (curve D), but these exhibit larger absorption since the pulse is now longer than T_2^*.

We now turn our attention to the evolution of a nonzero-area pulse toward zero. Saturation of the medium is negligible in our experiments since, for a pulse of duration $\tau_p = 12$ nsec (corresponding to two in-phase 6-nsec pulses) and $\mu = 0.2$ Debye,[13] the maximum pulse area is less than 2°. For these small angles, the above results can be quantitatively understood[16] in terms of a linearized theory[7] in which the thin-sample absorption is proportional to the overlap of the Fourier spectrum of the pulse with the resonance line. A zero-degree pulse has a Fourier spectrum which is zero on resonance. A similar spectrum, and hence absorption, will also result in this linear regime if a nonzero-area pulse whose spectral width is broader than the resonance line is allowed to propagate several absorption lengths. A consideration of linear dispersion shows that the phase relationships of the Fourier components are altered in such a way as to yield a zero-degree pulse.[17] For large tipping angles, the nonlinearity invalidates this analytic description, but we know from the area theorem[1-3] that the pulse area will decay toward zero if it is initially less than π.

The evolution of a small-area pulse into a low-loss zero-degree pulse can alternately be described in the time domain in the following way[7,8]: As the pulse enters the medium, it excites an oscillating macroscopic polarization, which for a resonant absorber is phased so as to radiate a field which adds destructively to the incident field. The polarization continues to radiate after the pulse passes and produces a negatively phased lobe on the trailing edge of $\mathcal{E}(z,t)$. For small absorption ($\alpha z \ll 1$, where z is the penetration depth into the sample) and in the inhomogeneously broadened limit, the coherent "ringing"[8] of the medium lasts for a time comparable to the inverse spectral width of excited atoms, which is given by the longer of the pulse width τ_p or the inverse inhomogeneous width T_2^*. For a homogeneously broadened line, the coherent ringing time will be given by the transverse relaxation time T_2. In either case, the polarization decays before all of the absorbed energy is coherently reradiated. If $\alpha z > 1$, on the other hand, a sufficiently large fraction of the incident pulse has been absorbed and reradiated into the negative lobe to enable this lobe to extract an appreciable fraction of the absorbed energy. This lobe is further amplified as the pulse propagates so that the pulse area approaches zero with little further loss in pulse energy. Significant pulse reshaping, resulting in the development of a pulse with many lobes, is obtained for $\alpha z \gg 1$.[18]

In order to further verify this evolution of small-area pulses into zero-degree pulses, transmission measurements were made on the pulse obtained by first passing a single small-area pulse through an additional 40-cm absorption cell (CII in Fig. 1) placed in front of the first cell. The subsequent absorption in cell CI for pulses which had passed through many absorption lengths in CII was seen to be characteristic of that obtained for zero-degree pulses. Transmission measurements as a function of pressure in cell CI are shown for single 6- and 2-nsec pulses in Figs. 2(b) and 2(c), respectively. Curve E in each of these figures gives the transmission with cell CII empty. With cell CII filled at a fixed pressure to give absorption as indicated by the point labelled f on curve E, the fraction of energy transmitted by cell CI was again measured as a function of pressure and is plotted as curve F in each figure. As is seen from these curves, we note that the transmission characteristics of the pulse after passage through cell CII exhibit reduced loss, as compared with the characteristics of the original input pulse. With cell CII filled at a higher pressure corresponding to a larger number of absorption lengths, we obtain an output pulse with an area more nearly equal to zero. This is seen from curve G, which shows the transmission vs pressure in cell CI with cell CII filled to give absorption as indicated by point g on curve E. The 6-nsec pulse evolves into one which has an absorption coefficient of only $\frac{1}{20}$ that of the cw; the 2-nsec pulse, into one which has only $\frac{1}{100}$. At a pressure of 1 Torr, where the cw beam is attenuated by $e^{-\alpha L}$ with $\alpha L \approx 20$,[15] 65% of the energy of the 2-nsec pulse which emerges from cell CII is transmitted by cell CI.

It should be noted that, at pressures sufficiently high that collision broadening becomes larger than the Doppler width, the dashed curve, which represents the cw absorption, becomes parallel to the abscissa. In the pressure region where T_2 becomes shorter than the pulse duration, the absorption for all pulses asymptotically approaches the high-pressure limit of the dashed curve. At a fixed pressure, the cw absorption maintains a constant logarithmic slope as a function of sample length; the zero-area pulse would also exhibit a constant but greatly reduced slope.

The use of short pulses from mode-locked lasers should allow the effects described here to be useful for long-distance propagation through absorbers with considerably shorter relaxation times. In the case of atmospheric propagation, for example, typical values for pressure broadening range between 1 and 20 MHz/Torr. Pulses 10—100 times shorter than those utilized here can be expected to result in reduced loss due to resonant absorption.

Other aspects of this work which are still the subject of experimental investigation include the study of these effects in the high-intensity limit where the nonlinear effects become important, as well as the off-resonance case, and the observation of the actual pulse-envelope distortion.

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[14]The curves in Fig. 2 were measured as a continuous function of pressure. The points correspond to pressure at which the Pirani pressure gauge was calibrated by means of a McCleod gauge at pressures above 150 mTorr and by means of the cw absorption at lower pressures.
[15]The pressure broadening of ~ 25 MHz/Torr (see Ref. 12) does not become significant until pressures exceed several hundred mTorr.
[16]cf., Fig. 5 of Ref. 7. A more detailed analysis of these results will be presented elsewhere.
[17]It should be noted that regardless of the continuous changes in pulse envelope, the carrier frequency remains unshifted if the laser frequency coincides with the center of a symmetrically shaped absorption line (see Refs. 6 and 7).
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