# SELF-INDUCED TRANSPARENCY IN GASES

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(Received 13 September 1967)

Recently McCall and Hahn,[1] using a ruby laser system, have demonstrated their predictions[2] of self-induced transparency for propagation of intense coherent light pulses in absorbing media. We report further confirmation of this effect for propagation of $Q$-switched $CO_2$ laser radiation at 10.6 $\mu$ through gaseous $SF_6$. The gas system has the advantage that the relaxation times of the absorbing levels, the inhomogeneous Doppler linewidth, and the absorption coefficients can be easily controlled by variation of pressure, temperature, and buffer-gas additives. This permits more detailed study of the theoretical predictions and provides a new experimental technique for studying relaxation times of individual transitions in gases and gas mixtures. Since the high-repetition-rate $CO_2$ laser operates stably in a single longitudinal $TEM_{00}$ mode, the predicted pulse delays and shapes are readily studied. Over a range of $SF_6$ pressures (0.01 to 0.05 Torr) and buffer-gas–induced relaxation times, our results agree qualitatively with theoretical predictions for $T_2' > \tau_p$, and in addition demonstrate a narrowing of the transmitted pulse for homogeneous relaxation times shorter than the input pulse width ($T_2' < \tau_p$).

Recently it has been shown[3,4] that the $CO_2$ laser transition at 10.5915 $\mu$ lies very close to a rotational transition of the $\nu_3$ vibrational-rotational band of $SF_6$. As evidenced by the reported absorption coefficient, $\alpha \approx 0.3$ cm$^{-1}$ Torr$^{-1}$, the 10.5915-$\mu$ $CO_2$ laser line seems to lie within a 30-MHz Doppler width of one of the $SF_6$ transitions. From the measured $\alpha$ we estimate[5] a dipole moment $p \approx 3 \times 10^{-20}$ cm esu for $SF_6$.

The threshold for self-induced transparency can now be obtained from the condition[1,10]

$$\frac{2p}{\hbar}\int_{-\infty}^{+\infty} \mathcal{E}(t)dt = n(2\pi), \quad n = 1, 2, 3, \cdots, \qquad (1)$$

where $\mathcal{E}(t)$ is the electric field of input laser pulse. Equation (1), with $n = 1$, gives a threshold power $P_{th} \approx 10$ W cm$^{-2}$ for a $\tau_p$ of 200-300 nsec. The threshold for transparency, as described in Eq. (1), occurs when the electric field of the input light pulse becomes intense enough to drive coherently an absorbing molecule from its ground state to the excited state and then back to the ground state. Theory shows that such a pulse (hereafter called a $2\pi$ pulse) propagates without attenuation. Since the dephasing collisions (involving a change of rotational or vibrational level or a change in the translational velocity of the molecule without a change in the rotational level) would disrupt the coherency of this excitation, the threshold condition normally applies for a low pressure of the absorbing gas such that the dephasing relaxation time, $T_2'$, is longer than the pulse width $\tau_p$ (the case for $T_2' < \tau_p$ will be described later). However, the pressures at which our experiments were performed were high enough so that a large number of molecules were present in a wavelength. At low pressures, the random distribution of molecules over axial distances of the order of a wavelength is expected to change the character of self-induced transparency.

The $CO_2$ laser produced 200- to 300-nsec long pulses with peak power of ~20 kW at a repetition rate of 120 Hz. The 10.5915-$\mu$ transition was the strongest oscillating line with fluctuations of less than ~10% in the peak intensity as well as the pulse width. The laser output beam passed through the following components: an aperture, an ether (not aether[6]) absorption cell[7] for varying the intensity without deflecting the beam, a second aperture, a beam splitter, a focusing mirror, a two-pass 4.7-m $SF_6$ absorption cell (i.e., total absorption length $l \approx 9.4$ m) with a Brewster-angle window, a third aperture, collecting lenses, and a spectrometer. The split-out beam was monitored with another spectrometer. Both spectrometer resolutions were set to observe only the 10.5915-$\mu$ transition. Ge:Cu photoconductors were used as detectors. The detector response time, <10 nsec, was sufficient to resolve the details of the pulse shape. Gated integrators measured the total energy of the pulses at the input, $E_i$, and the output, $E_0$, of the $SF_6$ absorption tube.

Figure 1 shows the measured $E_0$ vs $E_i$ for a number of $SF_6$ pressures. The maximum $E_i$ and $E_0$, $10^4$ on the arbitrary scale, corre-

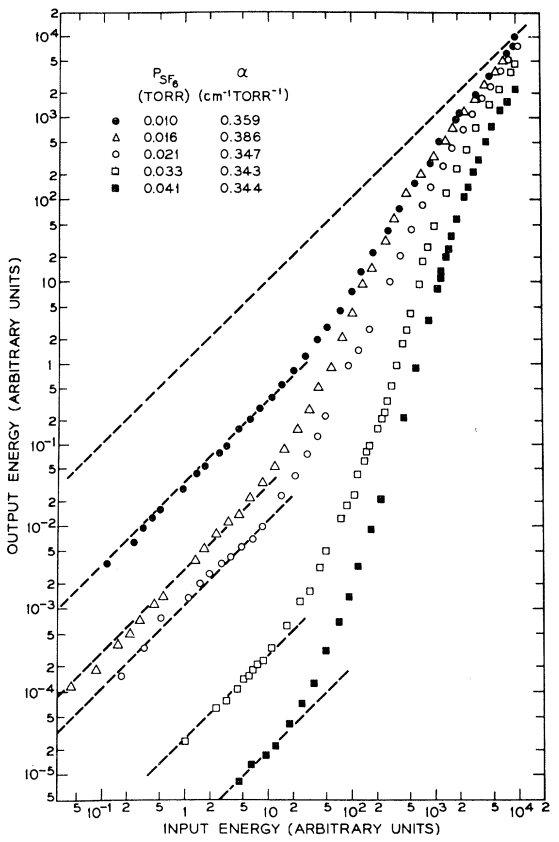


FIG. 1. $E_0$, as a function of $E_i$, for transmission through 9.4 m length of various $SF_6$ pressures. The maximum $E_i$ and $E_0$ (without $SF_6$), $10^4$ on the arbitrary scale, corresponds to a peak input intensity of $\sim 10^4$ W cm$^{-2}$. The absorption coefficients, $\alpha$, are obtained from the linear portions of the curves at the low end of the input energy scale. (The maximum $E_0$ corresponds to no $SF_6$ in the absorption cell.)

sponds to an input peak intensity of $\sim 10^4$ W cm$^{-2}$. At the low end of the $E_i$ scale we see classical absorption in $SF_6$. $E_0$ increases linearly with $E_i$, and $\alpha$ varies linearly with pressure, i.e., Beer's law is obeyed. From this classical region we obtain $\alpha \approx 0.344$ cm$^{-1}$ Torr$^{-1}$, in reasonable agreement with other measurements.[4,8] With increasing $E_i$, the $E_0$ curves for all $SF_6$ pressures depart from linearity at approximately the same input level, and the output increases faster than linearly with the input, signifying the onset of self-induced transparency in $SF_6$. At the highest $SF_6$ pressure, the output increases by $\sim 10^4$ for a factor of 10 increase in $E_i$ above threshold. For the highest $E_i$, $SF_6$ is nearly transparent and $E_0$ again begins to vary linearly with $E_i$. The threshold intensity for the transparency effect was ~10 W cm$^{-2}$ in agreement with calculations using Eq. (1). Note that this threshold is independent of $SF_6$ pressure over the range shown in Fig. 1, indicating that $T_2' > \tau_p$, since dephasing collisions should cause an apparent increase in $P_{th}$.

A typical output pulse near the threshold is shown in Fig. 2(b). Notice that the output pulse is delayed by ~0.5 $\mu$sec, its width $\tau$ is greater, and its shape is more symmetric than the input pulse. More important is the fact that its intensity is larger than the input pulse tail seen on this double-exposure photograph. Multiple double exposures similar to Fig. 2(b) showed that the output pulse intensity always exceeded the corresponding maximum input-pulse tail intensity over a major fraction of its duration. This observation eliminates the possibility that the transparency and the delayed output pulse are caused by "hole burning," i.e., the saturation of the absorbing transition.[4]

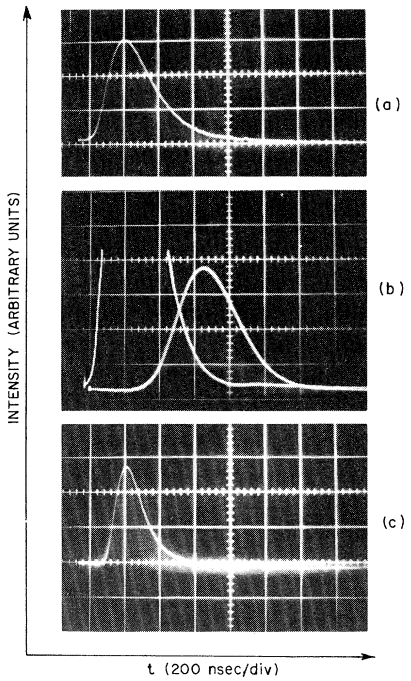


FIG. 2. Input and output pulse shapes just above $P_{th}$. (a) Typical input pulse as measured through the 9.4-m absorption cell but without any $SF_6$. (b) Double exposure of input and output. Input same as in (a) but with the vertical scale expanded by 4. The delayed output pulse with 0.04 Torr $SF_6$. (c) Output pulse with 0.04 Torr $SF_6$ and 2 Torr helium. Input intensity is adjusted to be just above threshold for this mixture. Notice the near total absence of a delay and the narrowing of the output pulse as compared with the input.

Pulse delays up to a maximim of 0.7 $\mu$sec were observed near the threshold ($l = 9.4$ m), and for higher input pulse intensities the output pulse delay steadily decreased, until at the maximum input intensity, the output pulse shape was a slightly delayed (~50 nsec) replica of the input pulse. At intermediate input intensities above the threshold some output pulses had two or more maxima; however, these shapes depended critically on the input pulse shape and focusing of the beam through the $SF_6$ cell. Halving the absorption length $l$ from 9.4 to 4.7 m resulted in a decrease in the output pulse delay from 0.7 to 0.4 $\mu$sec.

The above results are predicted by the theory of McCall and Hahn[1] in the limit $T_2' > \tau_p$. The output pulse delay at threshold is

$$\tau_D \approx \pi\hbar\alpha l/2p\mathcal{E}_{\mathrm{th}}, \qquad (2)$$

where $\mathcal{E}_{\mathrm{th}}$ is the threshold electric field. At 0.04 Torr of $SF_6$, and $\tau_p = 200$ nsec for $l \approx 9.4$ m, Eq. (2) predicts a delay of 1.3 $\mu$sec, which is about a factor of 2 larger than the observed delay at this pressure. The electric field of the delayed output pulses has the expected[1] sech form on the trailing edges, but the leading edges appear more abruptly as could be expected since the delayed output pulse must still start with the input pulse. The delayed output pulses are expected to be wider than the incident pulses since the electric field of the propagating pulse decreases by a factor of 2 to 3 because of the beam divergence and losses[1] associated with finite $T_2'$. In order to maintain the threshold condition with decreasing $\mathcal{E}$, $\tau$ must increase. As $\tau$ approaches $T_2'$ one first expects a finite but non-Beer's-law loss in the pulse electric field; however, $\tau$ cannot be greater than $T_2'$ because beyond this point a $2\pi$ pulse cannot propagate and the pulse will decay according to Beer's law. Thus $T_2'$ sets an upper limit on the maximum output pulse width.

As $\mathcal{E}$ increases above threshold, the molecules can undergo a series of $2\pi$ excitations corresponding to $n > 1$ in Eq. (1). The observed multiple maxima may be associated with the breakup of the propagating pulse into this series of $2\pi$ pulses. At maximum applied $\mathcal{E}$ the series of delayed pulses seem to merge into a single pulse closely resembling the input pulse but slightly delayed. This decreased delay is given by Eq. (2) but the $\tau$ is that given for one $2\pi$ cycle predicted by Eq. (1) with the maximum applied $\mathcal{E}$. The observed increase in $\tau_D$ (at threshold) with increasing $SF_6$ path length is also predicted by Eq. (2). The observed increase in delay for an increase of $l$ from 4.7 to 9.4 m cannot occur for a transparency caused by "hole burning."

On addition of a buffer gas to $SF_6$, we expect to reduce $T_2'$. As long as $T_2' \gg \tau_p$, $P_{\mathrm{th}}$ is not expected to change; however, the shape, width, and the maximum delay of the propagating $2\pi$ pulse at threshold may change with $T_2'$. We mentioned earlier that a propagating $2\pi$ pulse expands in order to make up for any depletion of $\mathcal{E}$; however, the output pulse width is limited to ~$T_2'$ for non-Beer's-law propagation. Hence, a change in the output pulse width, $\tau$, at threshold on addition of a buffer gas gives a good indication of $T_2'$, and we expect a narrowing of output pulse in the limit of $T_2' < \tau_p$. Accompanying the narrowing, an increase in $P_{\mathrm{th}}$ is to be expected. The output pulse for 2 Torr of He added to 0.04 Torr $SF_6$ is seen in Fig. 2(c) for input powers slightly above threshold, and shows a narrowing and a reduction in $\tau_D$ as compared with the delayed pulse seen in Fig. 2(b) without the buffer gas. For 0.04 Torr $SF_6$, the pulse width decreased progressively from ~520 nsec for 0 Torr He to about 140 nsec for 10 Torr He, and $\tau_D$ decreased from a typical value of ~500 nsec to ~0 nsec. Both of these effects are qualitatively understood in terms of the self-induced transparency in the limit $T_2' < \tau_p$. Figure 3 shows $E_i$ vs $E_0$ characteristics for 0.02 Torr $SF_6$ when He was added as a buffer. An increase in $P_{\mathrm{th}}$ is observed as He pressure is increased, and at 10 Torr He, $E_0$ varies linearly with $E_i$ up to very nearly the maximum available power from the laser. Below 0.5 Torr He there was no significant change in curves of $E_0$ vs $E_i$ indicating that $T_2' \gtrsim \tau_p$ at 0.5 Torr He. Both of these observations give a collisional dephasing frequency $\gamma_{SF_6\text{-He}} \approx 10^6$ sec$^{-1}$ Torr$^{-1}$, corresponding to a dephasing collision cross section $\langle\sigma\rangle_{SF_6\text{-He}} \approx 2\times10^{-16}$ cm$^2$ for the $SF_6$ levels responsible for the absorption of the 10.5915-$\mu$ radiation.

Addition of 1 Torr of $H_2$ was more effective than 1 Torr of He, and $\langle\sigma\rangle_{SF_6\text{-}H_2}$ was ~50% larger than $\langle\sigma\rangle_{SF_6\text{-He}}$. Argon, on the other hand, was a factor of 2 less effective than He at the same pressure. Thus $\langle\sigma\rangle_{SF_6\text{-Ar}}$ is larger than $\langle\sigma\rangle_{SF_6\text{-He}}$ by ~50%. Addition of ethylene, whose $\nu_7$ frequency is close to the $\nu_3$ frequency of $SF_6$ responsible for the absorption

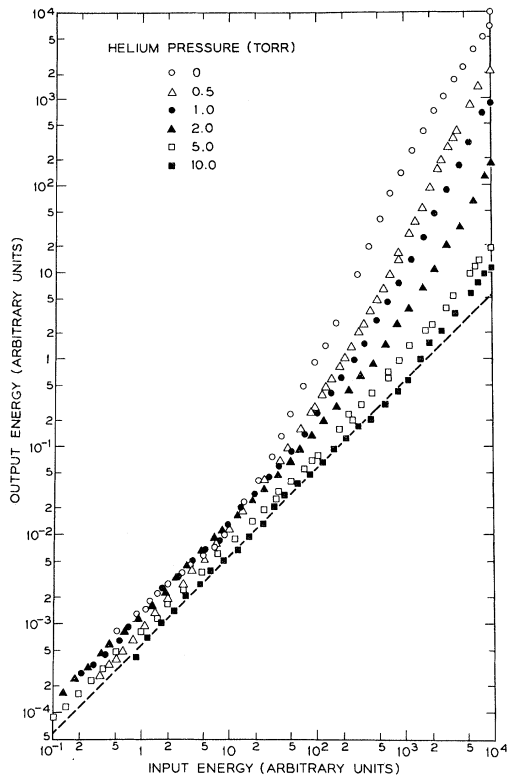


FIG. 3. $E_0$, as a function of $E_i$, for 0.02 Torr $SF_6$ showing the effect of helium buffer gas at various pressures. Notice the increase in $P_{th}$ as helium pressure increased. (The maximum output point corresponds to no $SF_6$ in the absorption cell.)

of 10.5915-$\mu$ radiation, gives results qualitatively similar to those for He.

In conclusion, we have shown that self-induced transparency predicted by McCall and Hahn[2] can be observed in absorbing gases for input pulse intensities as low as 10 W cm$^{-2}$. Although qualitative aspects of the theory have been verified, the experimental results shown here include ranges of relaxation phenomena which remain to be analyzed in quantitative detail. Since $CO_2$ lasers with cw powers well above the threshold requirement are available, it is of interest to study propagation of pulses long compared with the various relaxation times. The low threshold intensities should also make it possible to study relaxation times by the photon echo technique.[9] In addition it should be possible to study the limit of low gas pressures where molecules are sparsely distributed within distances of the order of a wavelength, which is of interest for propagation of coherent light through the outer atmosphere of the earth and the gaseous clouds surrounding the stars and the nebulae.

We thank Professor E. L. Hahn and S. L. McCall for a number of enlightening discussions, and Professor Hahn for encouragement during early phase of these experiments. We also thank J. P. Gordon and P. A. Wolff for critical comments on the manuscript, and R. J. Kerl for technical assistance.

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[1]S. L. McCall and E. L. Hahn, Phys. Rev. Letters 18, 908 (1967).

[2]S. L. McCall and E. L. Hahn, Bull. Am. Phys. Soc. 10, 1189 (1965).

[3]H. Brunet, Compt. Rend. 264, 1721 (1967).

[4]O. R. Wood and S. E. Schwartz, Appl. Phys. Letters 11, 88 (1967).

[5]We have assumed that because of the Boltzmann distribution of molecules among the various rotational levels, only about 1/300 of the total number of molecules lie in the ground-state vibrational-rotational level of $SF_6$ responsible for absorption of the 10.5915-$\mu$ radiation.

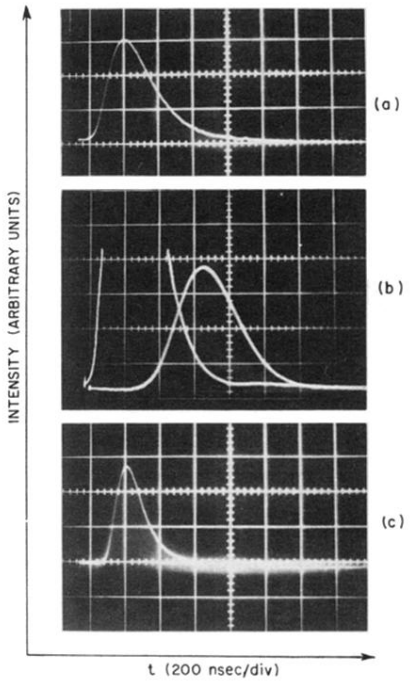
[6]For propagation of electromagnetic radiation through aether, see, for example, M. Born and E. Wolf, Principles of Optics (Pergamon Press, New York, 1964), pp. xxi-xxvii.

[7]The absorption coefficient of ether was measured to be $\sim 8\times 10^{-4}$ cm$^{-1}$ Torr$^{-1}$ at 10.6 $\mu$ and was linear up to ether pressures as high as 500 Torr.

[8]For the pulses used here $\tau_p \gg \frac{1}{2}\pi\Delta\nu_D$ so that cw and pulse measurements should agree.

[9]N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Letters 13, 567 (1964).

[10]The random orientations of the internuclear axes of the gas molecules require an average over the effective electric field-dipole interaction which may modify the nature of the threshold; however, this averaging is unnecessary for the vibrational transition of interest in $SF_6$ because of its high molecular symmetry ($O_h$).

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