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Investigation of Pulse Delay in Self-Induced Transparency

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This paper gives the details of pulse delay in self-induced transparency in SF_6 with a CO_2 laser at 10.5915 μ. Results show that the propagation of intense CO_2 laser pulses in SF_6 can be described by self-induced transparency.

Recently, we have observed the phenomena of self-induced transparency (SIT) and photon echoes in a gaseous medium, using a gas laser as the source of coherent radiation.[1,2] The absorber was SF_6. Since then, a number of papers have been published[3-5] which suggest that the phenomena described in Ref. 1 were actually due to bleaching or saturation rather than SIT. The physical processes which lead to lossless propagation of coherent pulses in absorbing media have been described in great detail by McCall and Hahn in Refs. 6 and 7. The characteristic of self-induced transparency is an input intensity-dependent pulse delay which accompanies the lossless propagation and a non-Beer's-law behavior of the absorbing medium. According to Ref. 5, under certain conditions it is possible to observe a pulse delay arising from dispersion of the medium. However, intensity dependence of the pulse delay is a crucial test which determines whether the pulse propagation involves SIT or not. In this paper, we report the measurement of pulse delay as a function of input pulse intensity for propagation of CO_2 laser pulses in a gaseous cell containing SF_6. We show that the pulse delay at low intensities is negligibly small, indicating a Beer's-law propagation and negligible normal dispersion effects. The pulse delay behaves in a completely different manner as we increase the pulse intensity to reach the levels at which SIT is expected to set in. In addition, this paper also reports the maximum delay that we have seen as a function of pressure. The results of this paper strongly suggest that self-induced transparency exists in SF_6 when we propagate high-power CO_2 laser pulses in this medium.

The experimental setup used in this experiment was similar to that described in Ref. 1. The SF_6 absorption cell was 4.7 m long and was used only in single pass. Every attempt was made to ensure that the Q-switched CO_2 laser was operated in a single transverse and longitudinal mode of the optical cavity. The peak power obtained in pulses approximately 180 nsec long was ~ 2–3 kW. This gave intensities of ~ 2–3 kW/cm^2 in the absorbing medium. A weakly focusing input mirror gave the beam a uniform cross section of about 1 cm^2 throughout the absorption cell. The output pulses were detected with a fast Ge : Cu photodetector and were analyzed with a spectrometer to make sure that only the $P(20)$ transition was investigated. Under normal operation the CO_2 laser did not oscillate on any transition other than the $P(20)$ at 10.5915 μ. Other transitions had intensities less than 10^{-4} that of the $P(20)$ transition. The output from the detector was analyzed with a boxcar integrator with a gate width of ~ 20 nsec. Averaging of pulses was achieved for several seconds. The output from the boxcar integrator, in a swept gate mode, was recorded on an x-y recorder. The response time of the entire detector system was essentially limited by the 20-nsec gate of the boxcar.

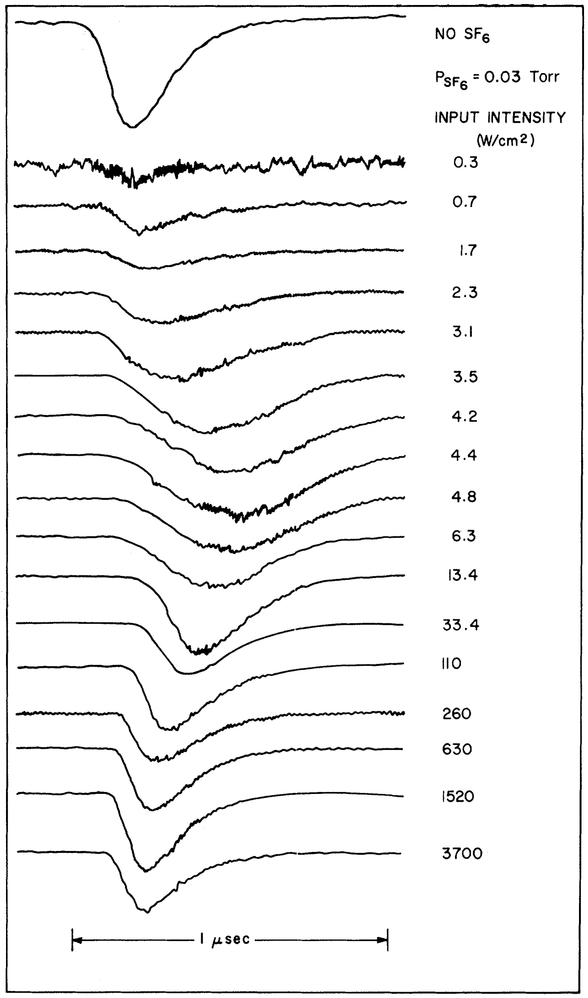

FIG. 1. Output pulse shape as a function of input intensities is shown by small numbers on the right-hand side. The topmost trace shows the output pulse without any SF_6 in the absorption cell. SF_6 pressure for the remaining pulses is 0.03 Torr. The intensity is arbitrary.

Figure 1 shows the shapes of some of the output pulses together with the shape of the input pulse into the 4.7-m-long absorption cell with SF_6 pressure of 0.03 Torr. The output pulses have been measured at a number of different input power levels to give intensity variation from about 0.15 W/cm^2 to about 4 kW/cm^2. At very low intensities, especially below about 0.5 W/cm^2, the output pulses are not delayed with respect to the output pulse in absence of SF_6 in the cell. At the same time, the intensity of the output pulse is very much less than that of the output pulse in the absence of the SF_6 cell, indicating a strong absorption in SF_6 as we expect. (The absorption constant in SF_6 is approximately 0.35 $cm^{-1} Torr^{-1}$ as measured by Patel and Slusher in Ref. 1. This gives an αl of about 4.8 at a SF_6 pressure of ~ 0.03 Torr.) For such intensities, the pulse is propagating in the Beer's-law regime through the absorbing cell. It was also found that the output intensity was linearly proportional to the input-pulse intensity in this range. As we increased the intensity the pulses were delayed, indicating a pulse propagation in the medium. At an intensity of about ~ 4 W/cm^2, we see that the delay suddenly increases to about 300 nsec. On further increase in input intensity, the pulse delay decreases as we go to the highest intensity of about 3 kW/cm^2. It is interesting to note that at and near the intensity where we see the maximum delay the output pulse shape is significantly different from the pulse shape in the absence of SF_6 in the absorption cell. This is in agreement with earlier results in Ref. 1. Also it is to be seen that the pulse width is different from the input pulse width, which is also understandable when we include a small amount of residual absorption in the medium. The delays from Fig. 1, and other similar runs for different SF_6 pressures are plotted on Fig. 2 as a function of input intensity. We see that for all pressures in the low-intensity regime there is no delay, indicating a Beer's-law propagation. As the intensity increases, the delay increases suddenly, and reaches a maximum at about 3–4 W/cm^2, beyond which it decreases as the input intensity is further increased. This result agrees with the theoretically expected behavior of lossless pulse propagation,[4] i.e., SIT in an absorbing medium. We have not shown the energy of the output pulse as a function of the input pulse energy because these results are similar to those in Ref. 1. Where the output pulse shows the maximum delay, the input pulse energy is approximately four to six times that of the propagating pulse, in agreement with results shown in Ref. 1. This result indicates that the absorption of the medium is reduced from about a factor of 100 to a factor of 4 when we increase intensity by about a factor of 2 from about 2 to about 4 W/cm^2. At this point, it should be mentioned that in this range of intensities, where delay is maximum, the intensity in the tail of the output pulse without any SF_6 is always smaller than that with SF_6 in the absorption cell. We have obtained output pulse shapes similar to the Fig. 2(b) of Ref. 1, where this argument is best seen, but for sake of space, we do not reproduce these here. The intensity dependence of the delay shown in Figs. 1 and 2, and the fact that the tail intensity of the propagating pulse with SF_6 in the cell is larger than without SF_6 conclusively rule out (a) saturation effects, and

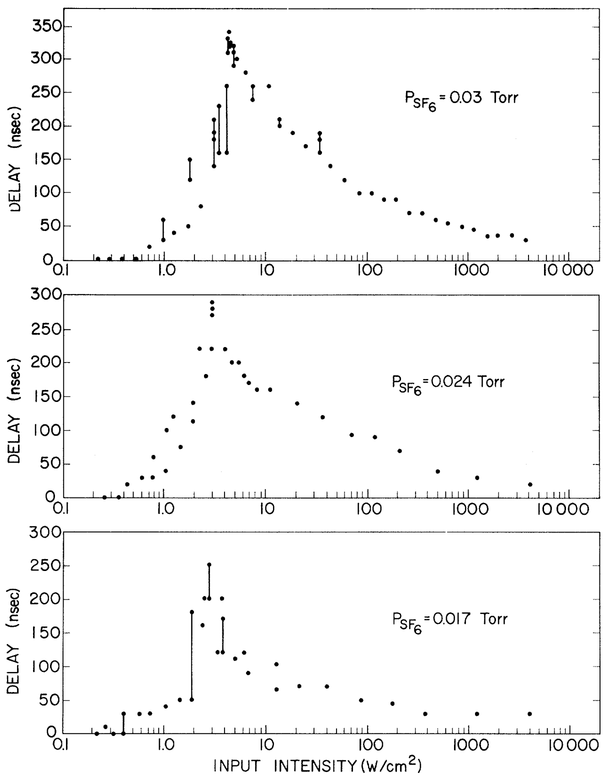

FIG. 2. Pulse delay as a function of input pulse intensity plotted for a number of different SF_6 pressures, showing the peak in pulse delay when the input pulse angle is approximately π. This indicates that for a π pulse angle the input intensity is approximately 3–4 W/cm^2 with an input pulse width of approximately 180 nsec.

(b) linear dispersion effects, as explanations for the pulse-propagation effects in SF_6. In fact, the above results, which indicate a sort of nonlinear behavior, can be explained only by invoking the self-induced transparency described by McCall and Hahn.[6, 7] It should be pointed out that the input pulse width is approximately 180 nsec even though the pulse width shown at the top of Fig. 1 looks somewhat wider. This is so because we are using a finite gate of about 20 nsec to sweep through the pulse for sampling at different times. The homogeneous relaxation time T_2' for the different SF_6 pressures shown in Fig. 2 varies from about 2 to about 0.8 μsec, and thus the ratio of pulse width to the homogeneous relaxation time is greater than approximately 4.

The results shown above in Fig. 2 for delay versus intensity are in rough agreement with what one expects from the SIT calculations given by Hopf and Scully in Ref. 4. Our experimental results show that the interaction between SF_6 and the CO_2 laser pulses is a coherent one and the phenomenon is described by the self-induced transparency theory.

Figure 3 shows maximum pulse delay as a function of SF_6 pressure for the 4.7-m-long absorption cell. It is seen that the maximum delay is linear with pressure, for pressures up to about 0.025 Torr. At higher pressures the delay reaches a maximum and then decreases. This is easily understood when we remember that the delay is proportional to the absorption coefficient only when the pulse width is much shorter than T_2'. At $P_{SF_6} \approx 0.025$ Torr, T_2'/τ_p is only 5, and thus a departure from the theory is not unexpected. On Fig. 3, we show the input intensity I_{crit} at which the maximum delay occurs as a function of SF_6 pressure. I_{crit} is independent of P_{SF_6} only for low SF_6 pressures where $T_2' \gg \tau_p$ as expected. I_{crit} increases as P_{SF_6} is increased at the same SF_6 pressures where the maximum delay-versus-pressure curve departs from linearity. Both of these are results of the breakdown of the requirement $T_2' \gg \tau_p$ for SIT. It is interesting to note that the pulse delay shows a

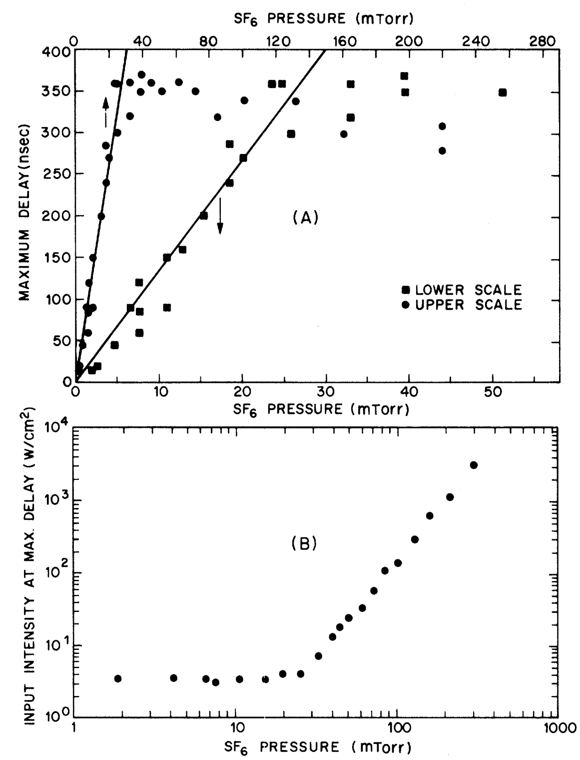

FIG. 3. (a) Maximum pulse delay as a function of SF_6 pressure showing a linear dependence expected from the theory, at low pressures where $T_2' \gg \tau_p$. At higher pressure the curve departs from linearity. (b) Input intensity for maximum pulse delay as a function of SF_6 pressure (see text).

distinct maximum as input intensity is changed, even for SF_6 pressures as high as 300 mTorr, where $T_2'/\tau_p \approx \frac{1}{2}$. This indicates that SIT exists even when $T_2' < \tau_p$, but the results are no longer clean and easy to interpret.

From the theory of Hopf and Scully,[4] we should expect a maximum delay of the propagating pulse when the input pulse intensity is such that the pulse angle θ, given in Eq. (1), is approximately π:

$$\theta = (2p/\hbar) \int_{-\infty}^{\infty} E\, dt \quad . \tag{1}$$

Thus, the intensity required for a π pulse seems to be about 3–4 W/cm^2, which (using a measured pulse width of approximately 180 nsec) gives us an electric dipole moment for the absorbing transition of about $(2.7\text{–}3.3)\times 10^{-20}$ esu.[8] This is to be compared with the earlier estimate of the dipole moment from the strength of the absorption, which depended on an estimate of the number of absorbing molecules. The dipole moment obtained from Eq. (1) involves no approximations other than the size of the propagating beam, which is known to within about a factor of 2. Thus, the dipole of the SF_6 absorbing transition is also known to within a factor of 2 and is given by $(2.7\text{–}3.3)\times 10^{-20}$ esu.

At the present time, we are attempting to observe the pulse delay versus input intensity at different SF_6 temperatures to elucidate the levels which cause absorption of the 10.6-μ radiation. The photon-echo results of Patel and Slusher[2] suggest that the absorption of the 10.6-μ radiation was caused by a $J=1 \rightarrow J=0$ or a $J=1 \rightarrow J=1$ transition. The self-induced transparency only exists in a clean form for low J-value transitions as indicated in Refs. 9 and 10. Thus, the results shown in Fig. 1, as well as those of Refs. 2 and 11, indicate that the absorbing transition which gives rise to the coherent interaction between the SF_6 and the CO_2 laser pulses must be a low-J transition. If the photon-echo results are accurate enough, it should be a 0-1 or 1-1 transition.

In this paper, we have shown that the propagation of CO_2 laser pulses investigated in SF_6 is indeed the phenomenon known as the self-induced transparency, and the pulse-delay versus input-intensity results prove beyond any doubt that the phenomenon is not that of bleaching. We have also obtained a good estimate of the dipole moment for the transition which causes the absorption of 10.6-μ radiation in SF_6. In spite of the fact that the experimental arrangement uses a Gaussian beam profile and the theory uses a plane-wave approximation, there is a reasonable agreement between theory and the experiment.

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

[7] S. L. McCall and E. L. Hahn, Phys. Rev. (to be published).

[8] $\int_{-\infty}^{\infty} E\, dt$ for the input pulse is calculated by numerical integration from observed intensity versus t pulse shape.

[9] The problems associated with SIT and other coherent effects with high J-value transitions were first mentioned in Ref. 1 (see Ref. 10). S. L. McCall and E. L. Hahn (Ref. 10) have given a detailed description of this effect in SIT. See Ref. 11 for photon-echo details with high-J transitions. The SIT problem was later also dealt with in Ref. 3.

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