# PHOTON ECHOES IN GASES

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Photon echoes following two pulses of $CO_2$ laser radiation incident on $SF_6$ gas have been observed. Homogeneous relaxation times for $SF_6$ colliding with $SF_6$, He, Ne, and $H_2$ are shown to be easily measurable from the decay of photon-echo amplitude.

Photon echoes have previously been observed for two ruby laser pulses incident on an absorbing ruby sample.[1,2] These echoes result from the coherent excitation of the electric dipoles of a two-level absorbing medium and are analogous to the nuclear spin echoes.[3] We report the observation of photon echoes in gaseous $SF_6$ using two $CO_2$ lasers as sources of coherent radiation at 10.6 $\mu$. These echoes are closely related to the self-induced transparency phenomenon which has been observed in both of the above systems.[4,5] The advantages of mode control, single-frequency operation, and the high repetition rate of the $CO_2$ laser allows us to study the echo characteristics with relative ease and accuracy. From the time decay of the echo amplitude, we have obtained the collision-induced homogeneous relaxation time $T_2'$ as a function of $SF_6$ and buffer-gas (helium, neon, and hydrogen) pressures. From these we obtain the pressure-independent collision cross sections for homogeneous dephasing of the levels responsible for the absorption of the 10.6-$\mu$ radiation in $SF_6$. For a given relative orientation of the electric field polarization of the first two excitation laser pulses, the echo electric field polarization was found to be parallel to that of the second excitation pulse in contrast to the earlier results in the ruby system.[1,2]

Two separate $CO_2$ lasers were used to generate the two linearly polarized optical pulses required for forming the echo in $SF_6$. The pulse width $\tau$ of 200 nsec was the minimum obtainable for the rotating-mirror $Q$ switch. The first optical pulse applied at $t=0$ excites the molecules to a super-radiant state which dephases because of the Doppler broadening (inhomogeneous relaxation time, $T_2^* \approx 6$ nsec) of the absorbing transition. The second pulse applied at $t=\tau_S$ results in an effective time reversal which rephases the super-radiant state to produce a photon echo at $t=2\tau_S+\frac{1}{2}\tau$.[1,2,6] The pulse separation could be continuously varied from 0 to 10 $\mu$sec by adjusting the reflection angle from a common rotating-mirror $Q$ switch (repetition rate = 120 Hz). However, the minimum pulse width of ~200 nsec limited $\tau_S$ to greater than 0.5 $\mu$sec for echo observation. The two light beams were made collinear with a germanium beam splitter and were directed through a 4.7-m $SF_6$ absorption cell. The output beams from the $SF_6$ cell were collected by a lens and detected through a spectrometer to isolate the 10.5915-$\mu$ $CO_2$ laser transition. A Ge:Cu photoconductor (4.2°K) was used as the fast detector with a response time of shorter than 20 nsec. The two beams were about 1 cm diam at the input of the absorption cell, expanded to about 2 cm diam at the output of the $SF_6$ cell, and were collinear to better than $10^{-3}$ rad. The intensities of both the pulses could be varied continuously by two ether absorption cells.[5]

Figure 1 shows a typical sequence of the output pulses. As the separation $\tau_S$ between the first two incident pulses was varied from ~0.5 to ~5 $\mu$sec, the third output pulse appeared at $2\tau_S+\frac{1}{2}\tau$ identifying it as the photon echo. The intensities of the two exciting pulses were ~1 and ~4 W cm$^{-2}$, respectively. From our earlier results[5] on threshold intensity required for self-induced transparency (~10 W cm$^{-2}$) in $SF_6$, it follows that incident pulse intensities in Fig. 1 correspond to ~90°-180° rotations of the pseudodipole moment.[1,2] As the incident pulse intensities were decreased or increased, the echo amplitude gradually decreased and vanished when the

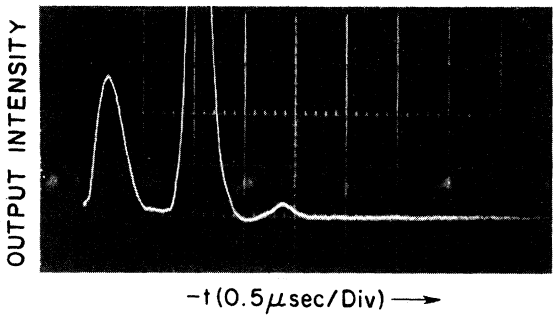


FIG. 1. Typical oscilloscope trace of output pulses from $SF_6$ cell at $P_{SF_6} \approx 0.015$ Torr. The first two pulses are transmitted $CO_2$ laser pulses, and the third is the photon echo. The second laser pulse is about a factor of 4 off scale.

incident intensities changed by a factor of 10. The peak intensity of the echo seen in Fig. 1 for $\tau_S \approx 1$ $\mu$sec is ~10% of the first transmitted pulse. For the $SF_6$ density of $5\times10^{14}$ cm$^{-3}$ in Fig. 1, the fraction of the incident pulse absorbed in exciting the super-radiant state is ~90% and the homogeneous relaxation time $T_2'$ is ~1.7 $\mu$sec. For these parameters the echo amplitude extrapolated to $\tau_S \approx 0$ is of the same order of magnitude as the absorbed fraction of the first pulse, and this is to be expected when the two beams are well aligned through the $SF_6$ absorption cell. Under certain conditions, however, it can be shown[7] from the theory of self-induced transparency effect that it is possible to obtain an echo whose amplitude is greater than that of the transmitted first pulse. The observed echo pulse width of $\sim\tau$ is expected because $\tau \approx 30T_2^*$, and only ~1/30 of the Doppler-broadened $SF_6$ absorption line is affected by the exciting pulses. Since an estimated 1/300 of the total number of $SF_6$ molecules are in the lower level responsible for the absorption of the 10.5915-$\mu$ $CO_2$ laser radiation,[8] and since $\tau \approx 30T_2^*$, only ~$10^{10}$-$10^{11}$ molecules cm$^{-3}$ participate in the echo formation. Thus it is interesting to note that the echo is formed under the condition of only a few $SF_6$ molecules per wavelength of the $CO_2$ radiation.

The decay of the echo intensity is a measure of $T_2'$ for the particular vibrational-rotational $SF_6$ levels excited by the 10.5915-$\mu$ $CO_2$ laser transition. This relaxation is caused largely by collision-induced change of rotational state or translational velocity of the molecules. The echo electric field is expected to vary as $\exp(-2\tau_S/T_2')$ or the echo intensity as $\exp(-4\tau_S/T_2')$. Figure 2 shows the exponential dependence of the echo amplitude as a function of the separation ($2\tau_S$) of the echo from the peak of the first pulse for pure $SF_6$ and mixtures of $SF_6$ with He, Ne, and $H_2$. From these data the following dephasing collision cross sections $\langle\sigma\rangle$ and homogeneous relaxation times $T_2'$ can be deduced. For $SF_6$-$SF_6$ collisions, $T_2'(SF_6\text{-}SF_6) = 22$ nsec Torr, $\langle\sigma\rangle_{SF_6\text{-}SF_6} = 1.05\times10^{-14}$ cm$^2$; for $SF_6$-He collisions, $T_2'(SF_6\text{-He}) = 33$ nsec Torr, $\langle\sigma\rangle_{SF_6\text{-He}} = 1.2\times10^{-15}$ cm$^2$; for $SF_6$-Ne collisions, $T_2'(SF_6\text{-Ne}) = 46.4$ nsec Torr, $\langle\sigma\rangle_{SF_6\text{-Ne}} = 1.9\times10^{-15}$ cm$^2$; and for $SF_6$-$H_2$ collisions, $T_2'(SF_6\text{-}H_2) = 16.4$ nsec Torr, $\langle\sigma\rangle_{SF_6\text{-}H_2} = 1.7\times10^{-15}$ cm$^2$. These cross sections are nearly pressure independent over a range of pressures from 0.002 to 0.015 Torr of $SF_6$ and from 0.001 to 0.02 Torr of buffer gases. The $SF_6$-$SF_6$ cross sections agree very well with the previous results from self-induced transparency effects.[5] The buffer-gas collision cross sections (above) are more accurate and larger by about a factor of 5 than those reported in Ref. 5 because the self-induced transparency results were based on lower limit estimates obtained from buffer-gas effects on self-induced transparency threshold.

The echo polarization was found to be parallel to the polarization of the second exciting pulse when the electric fields, $\vec{E}_1$ and $\vec{E}_2$, of the exciting pulses were polarized at an angle of $\psi$. The amplitude varied as approximately $\cos^2\psi$, i.e., decreased from a maximum to zero (with the observable signal-to-noise ratio, $\geq$200, for the echo when $\vec{E}_1$ and $\vec{E}_2$ are parallel) as $\psi$ increased from 0° to 90°. This behavior contrasts to the results for the ruby system,[2] where the echo polarization was found at an angle of $2\psi$ with respect to $\vec{E}_1$ and the echo amplitude was expected to be independent of $\psi$. Our results can be explained if the right- and the left-circularly polarized components of the linearly polarized incident pulses do not act independently on the $SF_6$ transition. Consider for example a $J = 0 \leftrightarrow 1$ transition with the selection rule $\Delta M = 0$. ($J$ is the total-angular-momentum quantum number and $M$ is its projection on a space-fixed axis de-

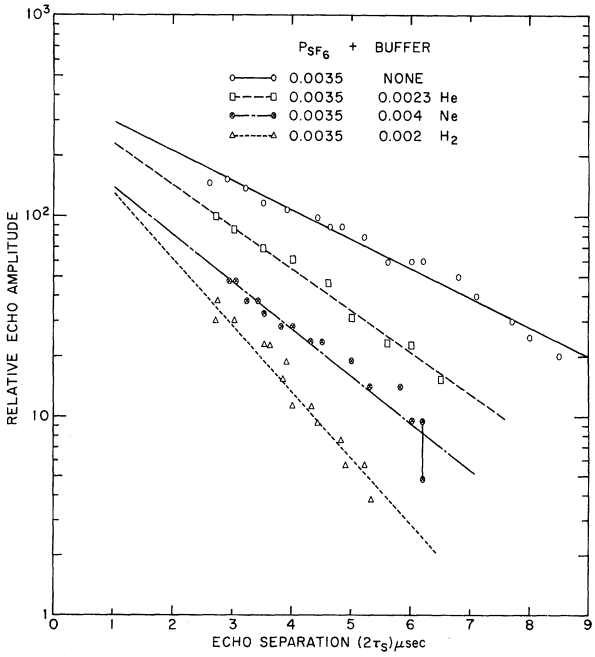


FIG. 2. Dependence of echo amplitude on $2\tau_S$ showing an exponential decay for pure $SF_6$ and $SF_6$ with He, Ne, and $H_2$. [Echo amplitude varies as $\exp(-4\tau_S/T_2')$.]

fined by $\vec{E}_1$.) The echo formed in this case can be shown to have polarization properties observed here. However, for $SF_6$ high-resolution (~0.3 $cm^{-1}$) absorption spectroscopy shows that the $CO_2$ laser transition at 944.15 $cm^{-1}$ falls 2-3 $cm^{-1}$ away from the line center toward the low frequency or $P$ branch ($\Delta J = -1$) components of the vibrational band. The absorption line appears to consist of two superimposed vibrational bands. The temperature dependence of one of the overlapping absorption peaks points towards its arising from a "hot" band $\nu_6 \rightarrow \nu_3 + \nu_6$. The temperature-independent band is identified as the fundamental $\nu_3$ band.[9] The $Q$-branch central peaks appear at $946.7 \pm 0.3$ $cm^{-1}$ for the $\nu_6 \rightarrow \nu_3 + \nu_6$ transition and at $947.6 \pm 0.3$ $cm^{-1}$ for the fundamental $\nu_3$ transition. Thus the laser at 944.15 $cm^{-1}$ probably coincides with a $P$-branch transition with a $J$ quantum number between 5 and 50. For large $J$, the rotational motion can be considered classically. The vibrational motion for a $P$-branch transition can be considered as a two-level quantum system and would result in an induced dipole perpendicular to the classical angular momentum $\vec{J}$. This semiclassical model is an interesting way to study the echo formation, but probably does not accurately describe the lower $J$ values of interest for $SF_6$. Details of these models and their relation to the echo formation and self-induced transparency[5] in $SF_6$ will be published elsewhere.

In conclusion we have shown that photon echoes can be observed in gases for vibrational-rotational transitions. The echo amplitude decay gives an accurate measure of $T_2'$ and dephasing collision cross sections for the particular levels excited. By using shorter pulses, it should be possible to investigate relaxation phenomena in smaller volumes of samples at higher gas pressures. This may be a versatile means of studying dynamic processes in nonequilibrium gases and gas mixtures. Although the photon-echo technique is limited, at present, to a few coincidences of absorbing transitions and laser frequencies, tunable lasers may, in the future, make photon echoes a valuable experimental technique.

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## PHOTON-ECHO DEPENDENCE ON INTENSITY*

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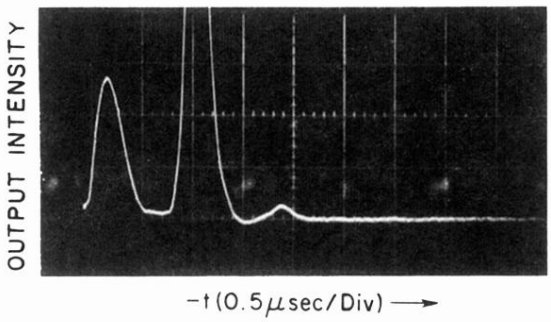
We report here on observations of photon echoes[1,2] in ruby as a function of input laser-pulse intensity.[3] These results are used to estimate the average degree of excitation of the ruby sample. The echo intensity is then studied as a function of external magnetic field orientation for various degrees of excitation. It is therefore possible to make an independent test of the validity of a tentative[2] model for echo dephasing.

Photon echoes, the optical analog of spin echoes,[4] are observed from a 0.03%-$Cr^{3+}$ sample at 2.2°K after excitation by two ruby-laser pulses, originating from a liquid-nitrogen cooled source. The apparatus used has been previously described,[1,2] with the important addition of two optical attenuators (train of polarizers) which are inserted into each pulse path. These are critically selected and adjusted so that reproducible changes of intensity are possible without impairing the image overlap of the two pulses on the ruby sample.

Previous calculations[2] have predicted the functional dependence of the echo intensity on the input intensity. For two pulses, incident at direc-

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