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Self-Induced Transparency of Degenerate Transitions with Thermally Equilibrated Levels

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Pulse propagation in SF_6 has been investigated using 200-nsec pulses of a CO_2 laser. Operating at $\lambda = 10.53$ μm ($P14$ transition) we have observed a breakup of the pulse envelope, an oscillatory energy transmission, and an energy-dependent pulse delay. We interpret our data in terms of the existing theory of self-induced transparency; the corresponding Bloch-type equations were derived assuming thermal equilibrium between rotational M sublevels.

Recently several authors[1-3] have investigated the propagation of short coherent optical pulses in SF_6 at the frequency of the CO_2 laser lines $P16$ to $P20$ of the 10.6-μm band. Patel and Slusher[1] interpreted their results with $P20$ pulses in terms of the theory of self-induced transparency (SIT) as first derived by McCall and Hahn[4] on the basis of a nondegenerate transition. From measurements of the photon echo they claimed the J-transition values involved to be 0-1 or 1-1.[5] Contrary to these authors, Rhodes and Szöke[2] believed that the SF_6 transitions interacting with $P16$ to $P20$ pulses were highly degenerate and gave strong evidence of the degeneracy at the frequency $P16$ by investigations of pulse transmission. We have measured the transmission of SF_6 with CO_2 laser pulses of the line $P14$ (10.53 μm). We wish to show that the propagation behavior at this frequency differs drastically from the transmission characteristics of both $P16$ and $P20$ pulses. Our results are believed to be consistent with a properly modified theory of SIT where the degeneracy of the upper and lower states is effectively removed by the assumption of a strong relaxation coupling.

According to the basic theory of SIT,[4] nondegenerate two-level absorbers are expected to interact with resonant coherent light pulses in such a way as to establish a regular alternation between absorption of light and subsequent coherent re-emission. This exchange of energy yields a modulation of the time envelope of the light pulse and an oscillatory behavior of the energy transmission. It also gives rise to an energy-dependent pulse delay. Experimental observation or pure SIT has been reported only on one atomic line of Rb vapor.[6]

In the limit of constant light frequency the pulse envelope has been shown to remain smooth if the resonant transitions are degenerate.[7] The degeneracy of the SF_6 transitions resonant with CO_2 laser radiation is expected to arise for two reasons: (1) The density of absorption lines is quite large in the 10-μm range to make an overlap of several $(v, J, K) \rightarrow (v', J', K')$ transitions rather probable. In fact high-resolution Lamb-dip spectroscopy[8] has shown high multiplicity of lines within the range of the CO_2 lines $P16$ (10.55 μm) and $P18$ (10.57 μm). (2) Each vibrational rotational level (v, J, K) consists of $2J+1$ sublevels which correspond to the projections M of the rotational angular momentum J on a space-fixed axis (the additional K degeneracy is assumed to be removed). In view of this spectral complexity Rhodes and Szöke[2] have interpreted their results with $P16$ pulses in terms of a theory derived for degenerate transitions. Contrary to these findings, our experiments of $P14$ pulse

propagation in SF_6 are indeed compatible with the theory of SIT for one nondegenerate transition, though the SF_6 transition coupling to $P14$ pulses is surely degenerate. Following Tang and Statz[9] we are led to the following interpretation. The population density within each set of degenerate M sublevels is always in thermal equilibrium; this assumption is equivalent to a random orientation of the molecules in space for all times.

We adopt the notation $|1m\rangle$ for the upper states, where 1 denotes the set of quantum numbers (v, J, K) and m the projection quantum number of spatial orientation. Correspondingly, $|2n\rangle$ is used for the lower states. The electric field of the optical pulses, given as

$$\vec{E} = \hat{x}\epsilon(t, z)\cos(\omega t - kz),$$

is introduced into the equation of motion of the density matrix, $i\hbar\dot{\rho} = [H, \rho]$, through a Hamiltonian $H = H_0 - \vec{p}\cdot\vec{E}$, where H_0 refers to the unperturbed system with equilibrated populations among the upper and lower degenerate states.

The pseudoelectric polarization $\vec{P}$ is defined as

$$\vec{p} = \hat{x}_1 u + \hat{x}_2 v - \hat{x}_3 k W/\omega, \tag{1}$$

with the following components:

$$u = \tfrac{1}{2}N\sum_{m,n} p_{1m,2n}(\rho_{1m,2n} + \rho_{2n,1m}),$$
$$v = \tfrac{1}{2}iN\sum_{m,n} p_{1m,2n}(\rho_{1m,2n} - \rho_{2n,1m}), \tag{2}$$
$$W = N\frac{\hbar\omega_0}{2}\frac{2}{(2J_1+1)^{-1} + (2J_2+1)^{-1}} \times\left(\sum_{m'}\frac{\rho_{1m',1m'}}{2J_1+1} - \sum_{n'}\frac{\rho_{2n',2n'}}{2J_2+1}\right),$$

where

$$k = \frac{1}{\hbar}\left[\frac{1}{6}\left(\frac{1}{2J_1+1} + \frac{1}{2J_2+1}\right)\sum_{m,n}|\vec{P}_{1m,2n}|^2\right]^{1/2}. \tag{3}$$

N is the number of particles involved and ω_0 the transition frequency. In a rotating frame one finds an equation of motion of the Bloch type,

$$d\vec{P}'/dt = \vec{P}' \times (\hat{x}_1' k\epsilon + \hat{x}_2' \Delta\omega), \tag{4}$$

with $\Delta\omega = \omega_0 - \omega$. The quantity $\theta = k\int_{-\infty}^{+\infty}\epsilon dt$ is called the pulse area.

It is interesting to see that Eq. (4) is of the same form as derived for a nondegenerate two-level transition.[10] In other words, the existing theory of SIT[4] can be applied to our system when the single dipole moment is replaced by the average dipole moment $P_{av} = k\hbar$. We refer to this situation as "effectively nondegenerate".

We have used a CO_2 laser with three prisms, a rotating mirror, and a piezoelectric tuner. The emission occurred in a TEM_{q00} mode. The beam passed a NaCl lens of 10 m focal length, a diaphragm, and an attenuation cell before entering the SF_6 cell of 525 cm length. Two detectors (Ge:Au and Ge:Hg) of 15 nsec risetime observed the incoming and transmitted pulses. The transmitted pulses were measured about 10 cm behind the exit window at the center of the beam where the transverse intensity variation was negligible. The longitudinal intensity distribution was found to be most uniform when the diaphragm was limited approximately to the first Fresnel zone.

We measured the time dependence of the transmitted pulses, the transmitted energy, and the delay curve versus the incident pulse energy at a pressure of 30 mTorr. In Fig. 1(a)–1(c) are presented oscilloscope traces of incident and transmitted pulses. The transmitted pulses of

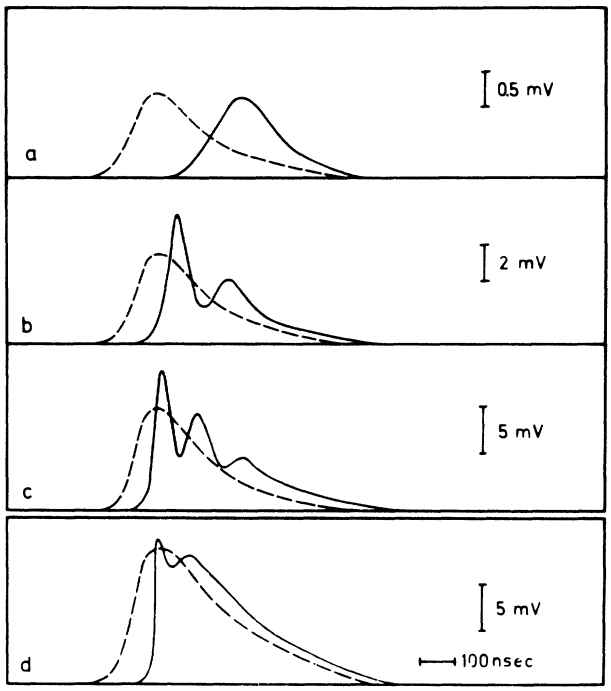

FIG. 1. Oscilloscope traces of input (broken curve) and output (solid curve) pulses transmitted through 525 cm of SF_6 at 30 mTorr. (a)–(c) show pulses of frequency $P14$ at different intensity levels and (d) shows typical $P16$ pulses. The intensity levels are given in terms of voltage sensitivity. The time scale in (a)–(c) is the same as plotted in (d).

1(b) and 1(c) break up into separated parts of increasing individual width and decreasing height. It is interesting to note that the peak intensities exceed those of the input pulses. In addition the transmitted pulses are more and more delayed with decreasing input energy. This behavior of pulse breakup with a corresponding energy-dependent delay of the peak pulse is in agreement with calculations for a nondegenerate absorber.[4] A similar envelope modulation has been observed using $P12$ (10.51 μm) light pulses. It is instructive to notice the contrast between the modulation of $P14$ pulses [Fig. 1(a)–1(c)] and $P16$ pulses. In Fig. 1(d) we present the incident and transmitted pulses for the $P16$ transition. In this case the envelope modulation is very small, with a distinct steepening of the leading edge. This modulation roughly agrees with observations of Rhodes and Szöke[2] who considered the smooth pulse envelope as an indication of high degeneracy. Our observations with $P20$ pulses are equally well consistent with pulse shapes as reported by Patel.[1] We have found a pronounced frequency dependence of $P14$ and $P12$ pulse splitting which was consistent with the local absorption profile of SF_6.[10] The breakup was most evident at the frequencies $P14_{\text{center}} + 15$ MHz and $P12_{\text{center}} - 20$ MHz.

In Fig. 2 the energy transmission is plotted versus the input energy. The curves fitted to the experimental points are presented for the two lines $P14$ and $P16$. The $P14$ transmission rises rapidly for small input energies and exhibits three well-resolved peaks. They should be characterized by pulse areas $\theta = 2\pi n$, i.e., the input pulse energies of the transmission

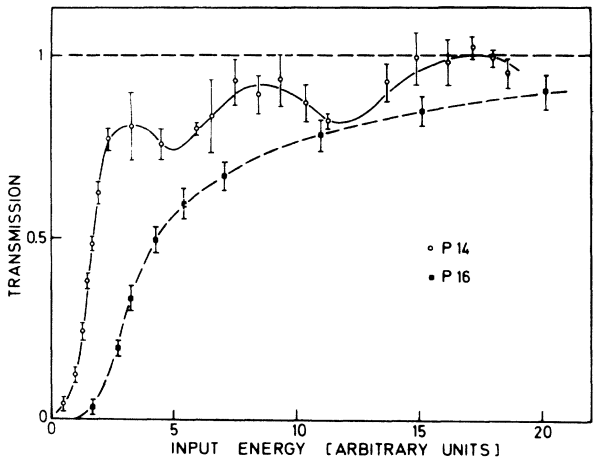

FIG. 2. Energy transmission ratio versus input energy of $P14$ and $P16$ pulses.

maxima are predicted to be proportional to n^2. This relation holds quite well in our experiments if one takes into account the influence of the dephasing time T_2, which tends to shift the input pulse area related to the first transmission maximum to values somewhat larger than 2π. In our case the homogeneous dephasing time T_2 is approximately[11] 770 nsec for a pressure of 30 mTorr. This value is roughly four times the pulse width of about 200 nsec.

From the condition $\theta = 2\pi n$ we estimate the numerical value of the average dipole moment of the transition to be $P_{av} \simeq 6 \times 10^{-19}$ esu cm. For a pulse duration of approximately 200 nsec the threshold intensity for the rapid increase of transmission is of the order of 0.1 W/cm^2. From Fig. 2 it is readily seen that the $P16$ transmission curve approaches total transparency monotonically which is considered to be typical for a degenerate transition and differs drastically from the $P14$ transmission curve.

It is well known that the delay of the transmitted pulses is a function of θ. The detailed nature of the delay curve strongly depends upon the degree of degeneracy. In Fig. 3(a) our observations of the pulse delays (first maximum) are plotted versus the input energy for the $P14$ frequency (same units as in Fig. 2). For comparison two theoretical curves calculated for a nondegenerate[3] and a degenerate transition[7] are given in Fig. 3(b). The calculated curves are normalized to the same peak value. A comparison of our experimental delay curve with the theoretical curves strongly suggests that the transition interacting with the $P14$ emission is of a nondegenerate type.

Our data are believed to be incompatible with a theory based on isolated degenerate transitions; they lead us to the assumption of an "effectively nondegenerate" transition. The difference between our results at the line $P14$ and those at $P16$ and $P20$ may be explained as follows: In the range of the lines $P16$ and $P20$ several SF_6 transitions $(v, J, K) \rightarrow (v', J', K')$ overlap within their Doppler widths, thus giving rise to a degeneracy in addition to that of spatial orientation. Consequently, laser pulses couple to several averaged dipole moments differing in magnitude; a distinct breakup of the incident pulse is not expected in this case. It is interesting to note that Hocker and Tang[12] have observed an appreciable break up of $P18$ pulses in SF_6 at lower temperature (dry ice) where the Doppler widths and, correspondingly, the overlap of absorption

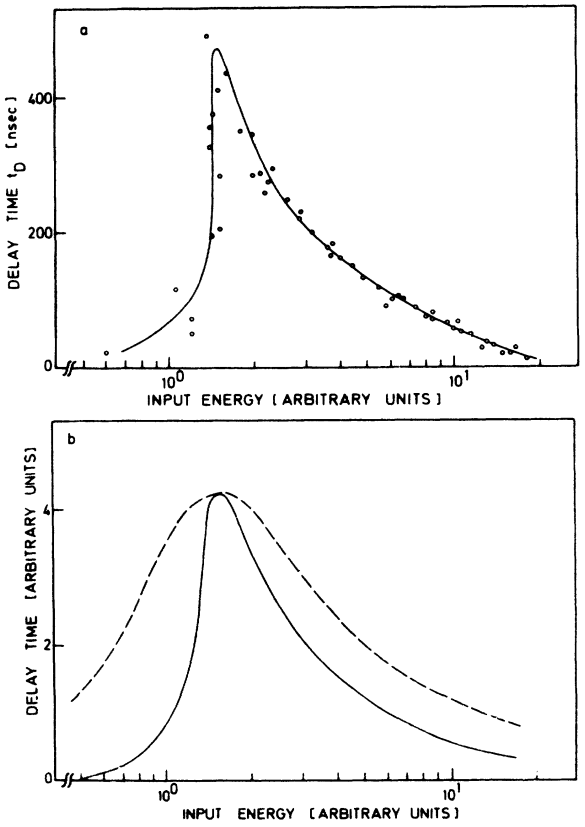

FIG. 3. Delay of the peak pulse versus input energy (a) obtained with $P14$ pulses, and (b) calculated for a nondegenerate (solid line) and a highly degenerate (broken line) transition (Q10).

lines are reduced. We feel that in our case ($P12$ and $P14$ frequencies) no overlap of the SF_6 transitions occurs even at 300°K.

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Differential Cross Sections for K-Shell Ionization of Surface Atoms by Electron Impact*

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We report first differential cross-section measurements for inner-shell ionization of surface atoms by electron impact. Comparison is made with calculations derived from a simple extension of the Burhop theory for ionization of isolated atoms. The trends of the predicted values are in qualitative agreement with experiment.

Excitation or ionization of the inner shells of atoms in molecular gases and in solids by electron impact has received considerable attention.[1-3] A related phenomenon is the ionization of atoms adsorbed on solid surfaces. Measurements of total inner-shell ionization cross sections as a function of primary energy have been made for gas, solid, and surface systems.[1-4] Until now, however, differential cross sections, which provide a better test of inelastic scattering theory, have been lacking. This has been due partially to sensitivity and absorption problems involved in the measurements. Detection of the ionization of chemisorbed atoms on surfaces has the distinct advantage that large numbers of atoms ($\sim 10^{13}$) are involved in the scattering, resulting