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OBSERVATION OF THE OPTICAL TRANSIENT NUTATION EFFECT*

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The optical nutation effect has been observed on a rotational-vibrational transition in gaseous SF_6 at 10.6 μ.

In this Letter, we report the observation of the optical analog of the nuclear magnetic transient nutation effect.[1] The optical effect was observed on a rotational-vibrational transition of the ν_3 absorption band[2] of gaseous SF_6 using the 1.6-μ output of a Q-switched CO_2 laser. This effect is closely related to the recently observed self-induced transparency effect[3,4] and the photon echo effect.[5,6] It can be observed under suitable conditions[7,8] when a step-function–type, monochromatic, coherent light pulse passes through a resonant absorption medium or a maser medium.[9] The effect would show up in the form of a damped amplitude modulation near the leading edge of the light pulse.

Physically, the optical nutation effect occurs because the intense coherent light pulse can drive the molecules successively from the lower state in the case of a resonant absorption medium, to a coherent superposition state where the induced dipole moments of the molecules oscillate in phase resulting in a large macroscopic electric dipole moment, to the upper state, again to the coherent superposition state, and finally back to the lower state; the cycle repeats but is subject to damping by the relaxation processes in the medium. The oscillation in the population difference reacts back on the field and amplitude modulates the light wave. The phenomenon is analogous to the nuclear magnetic transient nutation effect.

In the absence of any inhomogeneous broadening of the transition, the rate of damping of the "optical nutations" is equal to the inverse of the homogeneous relaxation time[3] T_2'. For molecular vibration-rotational transitions, the angular frequency of the amplitude modulation, or the nutation frequency Ω, is proportional to the applied constant electric field amplitude, $\bar{E}$, of the light wave[8]:

$$\Omega^2 = \frac{|\bar{E}|^2}{6\hbar^2}\left[\frac{1}{2J_m+1}+\frac{1}{2J_n+1}\right]S_{mn} \tag{1}$$

when the amplitude of modulation is small compared with $\bar{E}$, where S_{mn} is the so-called line strength of the transition and is given by

$$S_{mn} = \sum_{M_m M_n} |\langle v_m J_m l_m M_m | \times \vec{P} | v_n J_n l_n M_n \rangle|^2 \tag{2}$$

in terms of the appropriate matrix elements of the electric dipole operator $\vec{P}$ of the molecule defined relative to the space-fixed axes. Here $\hbar$ is Planck's constant. The quantum numbers J, l, M, and v refer, respectively, to the total angular momentum, the projection of $\vec{J}$ along a symmetry axis of the molecule, the projection of $\vec{J}$ along the space-fixed z axis, and the vibrational

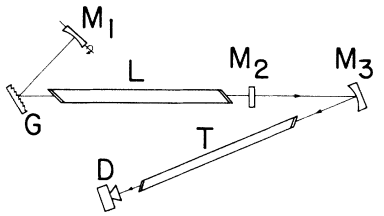

FIG. 1. Experimental setup for the observation of the optical transient nutation effect. M_1: rotating mirror Q-switch, gold coated, 10-m radius of curvature. G: Bausch and Lomb grating, 75 lines/mm, 12-μ blaze. L: 2-m laser tube with NaCl Brewster windows. M_2: flat, dielectric-coated germanium mirror, 15% transmitting. M_3: gold-coated mirror, 2 m radius of curvature. T: 1.8-m-long SF_6 absorption tube with NaCl Brewster windows. D: Ge:Au detector.

state of the molecular level; the subscripts m and n refer, respectively, to the upper and lower levels in the transition. It is then clear that the basic requirement for the observation of the optical transient nutation effect is that

$$\Omega > 1/T_2'; \tag{3}$$

to see a few nutations, the pulse duration must obviously be several times $2\pi/\Omega$. In the presence of inhomogeneous broadening, the behavior of the optical nutations would of course be modified; however, there will be significant modifications only[7] when the inhomogeneous broadening is much larger than Ω.

On the basis of Eqs. (1)-(3) and recent estimates of various SF_6 constants,[4,6,10] it is clear that the optical transient nutation effect should be observable in this gas using the CO_2 laser as the source of coherent radiation. Our experimental setup is shown in Fig. 1. The Q-switched laser produces pulses of up to 5-kW/cm^2 peak power in a single cavity mode and on a single vibrational-rotational transition. Any one of about 14 P branches or 11 R branches of the $00^0 1$-$10^0 0$ transitions can be chosen by rotating the grating. For this experiment we operated on the $P(20)$ transition at 10.6 μ. The rotating-mirror Q switch was operated at about 150 revolutions per second. The detector is a high-speed gold-doped germanium unit made by Santa Barbara Research, with a response time of ≤ 2 nsec.

Figure 2(a) shows multiple scope traces of typical output pulses of the single-mode CO_2 laser,

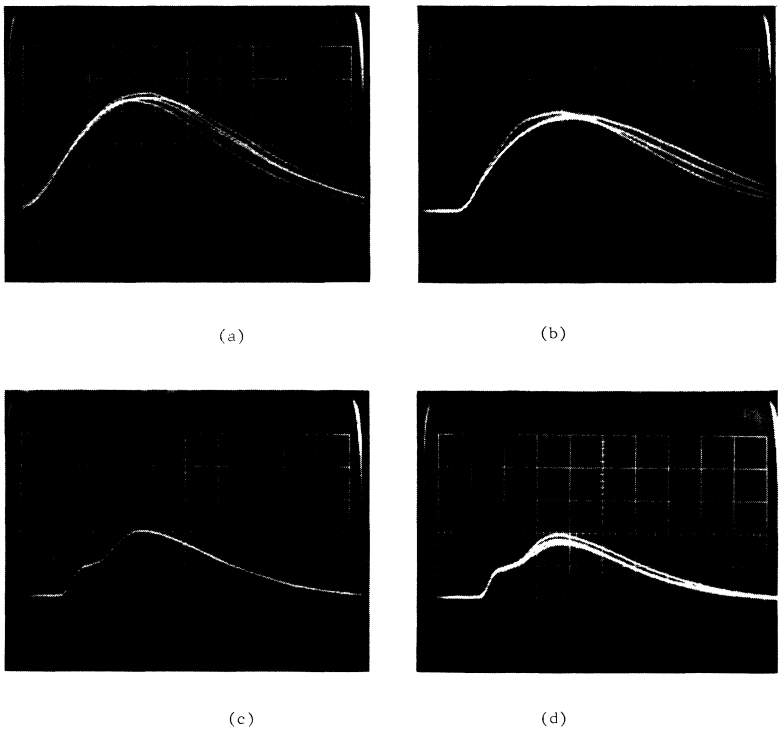

FIG. 2. Input and output pulses from SF_6 absorption tube. Horizontal scale is 50 nsec/division. (a) Multiple traces of input pulses. Peak intensity shown $\approx$3.6 kW/cm^2. (b) Output through SF_6 tube with $P_{SF_6} \approx 0.05$ Torr. Peak intensity $\approx$3 kW/cm^2. (c) Output pulse from SF_6 tube with $P_{SF_6} \approx 0.2$ Torr. Peak intensity $\approx$500 W/cm^2. (d) Multiple traces under same conditions as (c).

or the input into the SF_6 cell. As can be seen, the pulse shape, especially near the front end, is quite stable and can always be repeated. Figure 2(b) shows the pulse shape after passage through the SF_6 cell at a low pressure of 0.05 Torr; the pulse is simply attenuated, with the leading part attenuated more heavily giving the appearance of a slight steepening of the front edge. As the SF_6 pressure was increased to 0.2 Torr, an amplitude modulation on the front part of the pulse unmistakably appeared[11]; Fig. 2(c) shows a scope trace of such a pulse. Figure 2(d) shows multiple scope traces of such pulses. As can be seen, again the pulse shape with the amplitude modulation is highly stable and can be easily repeated. Similar amplitude modulation can be seen under the same conditions when the detector samples different points of the cross section of the output beam or a longer radius of curvature mirror was used for M_3 shown in Fig. 1. It should be pointed out that the detector was located near the center of the output beam of the SF_6 cell for Fig. 2(b) but displaced somewhat from the center for Figs. 2(c) and 2(d). The exact shape of the amplitude modulation would vary depending upon the local beam intensity.

This amplitude modulation is believed to be due to the optical transient nutation effect. Numerically, this interpretation is quite consistent with previous experimental results on SF_6. The observed period is of the order of 50 nsec. On the basis of the data given in Ref. 4, the light intensity required to give a nutation frequency $\Omega/2\pi$ of 20 MHz should be approximately[12] 160 W/cm^2. The average intensity of the output light pulse where the modulation appeared as shown in Figs. 2(c) and 2(d) was approximately 200 W/cm^2. Furthermore, according to Ref. 6, T_2' in pure SF_6 is of the order of 22 nsec at 1 Torr; thus, the nutations should be damped out at our pressure of 0.2 Torr in about 100 nsec, which is consistent with our observations. To make the results more meaningful quantitatively, one would obviously have to modify the input pulse shape to resemble more a step-function pulse so that the average intensity would remain nearly constant where the nutations occur; then the nutation frequency would be directly proportional to the constant part of the E-field strength according to Eq. (1) and the damping rate would give direct information on the linewidth of the transition. For our particular experimental arrangement, there was only a fairly narrow pressure range where the optical transient nutation effect could be seen. Below about 0.1 Torr, the amplitude of the modulation was apparently too small to be seen; above about 0.4 Torr, the attenuation of the entire 10.6-μ pulse through the SF_6 cell became too large and our detector was not sensitive enough to see the residual output pulse. Within this intermediate pressure range, however, the effect was always repeatable. To extend the range where the effect can be seen, one should obviously use a longer SF_6 cell thus allowing a reduction of pressure.

While our results are only qualitative and a more quantitative study is difficult at this time, the effect is there and clearly seen. Further improvement can undoubtedly be made, although it is probably not particularly important in the case of SF_6. The extension of such studies to the 00^01-10^00 vibrational-rotational transitions in the CO_2 laser medium itself would be highly valuable, since the nutation frequencies would give directly the line-strength factors for these transitions. These transitions are forbidden in the harmonic and linear approximations and become allowed only when the mechanical and electrical anharmonicities in the molecule are taken into account; it will be of interest to check the calculated line-strength factors. Furthermore, from the damping of the optical nutations, one could also learn more about the realxation processes of these CO_2 laser levels. Hopefully, the demonstration of the effect in SF_6 will lead to more activities in these directions.

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[11]The authors of Ref. 4 also mention that "at intermediate intensities above the threshold some pulses have two or more maxima." It is possible that the effect they observed was the optical nutation effect; comparison with our results is, however, difficult to make due to the lack of explicit data on the effect they observed.

[12]The experimental results on self-induced transparency of Ref. 4 show that for SF_6 a 2π pulse at an intensity level of approximately 10 W/cm^2 is about 200 nsec long. For a 2π pulse, $\Omega/2\pi$ times the pulse length is equal to 1; thus, 160 W/cm^2 is required to yield $\Omega/2\pi = 20$ MHz.

ISOTOPIC PHASE SEPARATION IN SOLID He^3-He^4 MIXTURES*

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The excess pressure due to phase separation of solid He^3-He^4 mixtures held at constant volume has been measured. From this we obtain the phase-separation temperature as a function of the mole fraction, the excess volume, and the pressure dependence of the critical temperature.

By observing the specific heat of a mixture of solid He^3 and He^4, Edwards, McWilliams, and Daunt[1] (EMD) discovered, in 1962, that these mixtures undergo phase separation below a certain temperature. Recently a microscopic theory of the phase separation has been presented by Mullin.[2] In this paper we report measurements of the excess pressure P^E due to phase separation of He^3-He^4 mixtures held at constant volume. In addition to the phase-separation temperature as a function of the mole fraction of He^3, x, the measurements give the excess volume v^E and the pressure dependence of the critical temperature, which are quantities predicted by the Mullin theory.

EMD found that the specific heat due to mixing could be represented quite well by the regular solution model. In this model[3] one employs various excess quantities which are generally defined as the increase in that quantity upon mixing. The excess Gibbs function is given by

$$g^E = x(1-x)\Delta E(P), \tag{1}$$

where ΔE is the energy of mixing. This leads to a critical temperature for phase separation of

$$T_c = \Delta E/2R. \tag{2}$$

The specific heat in the phase-separation region is expressed as

$$C = 2RT_c(1-2x_{ps})(dx/dT)_{ps}, \tag{3}$$

where the subscript "ps" means along the phase-separation curve. For the excess volume we have

$$v^E = (\partial g^E/\partial P)_T = x(1-x)2R(\partial T_c/\partial P). \tag{4}$$

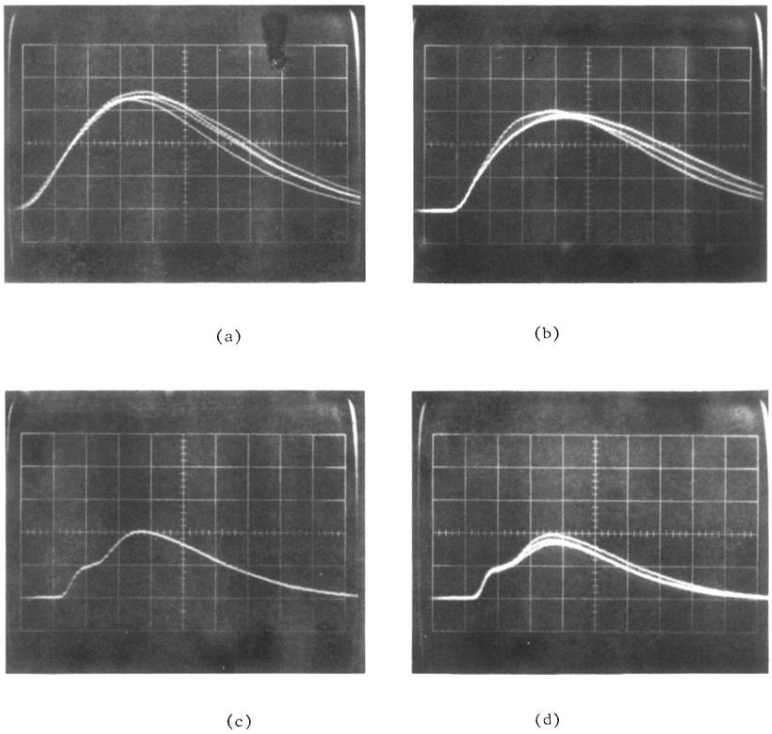
Mullin finds that the solution is almost regular, but the essential feature is the difference in molar volumes of the pure He^3 and He^4, $v_3 - v_4$, which would be zero in the classical theory. He obtains for the excess Gibbs function

$$g^E = x(1-x)[a(P)-b(P)x], \tag{5}$$

where the small term in x in the square bracket causes a slight departure from the regular solution. The main effect of this extra term is to cause asymmetry in the phase-separation curve. For the excess volume Mullin obtains

$$v^E \simeq -\sigma x(1-x)/P, \tag{6}$$

with $\sigma/P \simeq 0.4$ cm^3/mole. He computes the T-x



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