Measurement of the Transition Moment by the Optical Transient Nutation Effect*

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(Received 13 March 1969)

The observation of the optical transient nutation effect in SF_6 is found to be greatly facilitated by cooling the gas to dry-ice temperature; similar improvement is also expected in the observation of the self-induced transparency effect and the photon-echo effect in this gas. The improved experimental results demonstrate that it is possible to determine the transition moment of optical transitions directly on the basis of the optical nutation effect without the need of estimating any population densities which is required in the usual method based on the measurement of the absorption coefficient.

ALTHOUGH the optical nutation effect has recently been observed[1] on a rotational-vibrational transition of SF_6, the initial experimental results did not go beyond the mere demonstration of the existence of the effect; they did not provide much useful quantitative information on, for example, the frequency and damping of the nutations. In this paper, we report significantly improved results on the optical nutation effect that also allow one to determine the transition moment of the 10.57-μ transition of SF_6 directly from the observed nutation frequency. The improvement resulted primarily from cooling the gas to dry-ice temperature (SF_6 freezes at 140°K for 1-mm Hg pressure); cooling the gas reduces both the Doppler width of the line and the optimum operating pressure. We expect that cooling the gas should also greatly facilitate the observation of such related effects as the self-induced transparency effect[2,3] and, in particular, the photon-echo effect[4] in this gas.

The experimental setup used was similar to that in Ref. 1. The Q-switched CO_2 laser was tuned to the 10.57-μ line corresponding to the $P(18)$ line and to a single transverse and longitudinal mode by means of a diffraction grating and an iris aperture. The output was passed directly through a 57-cm-long SF_6 absorption cell equipped with NaCl Brewster windows and cooled to dry-ice temperature, 195°K. With this length cell, SF_6 pressures of 0.12 to 0.18 Torr resulted in nutations of large amplitude while net energy absorption in the cell was only $\lesssim 10\%$. The transmitted pulses were detected by a Ge:Au high-speed detector with response time of ≤ 2 nsec. This detector was calibrated for the 10.57-μ pulses using an Eppley thermopile.

Figure 1(a) shows scope traces of typical output pulses of the single-mode CO_2 laser, or the input to the cooled SF_6 cell. Figures 1(b) and 1(c) show examples of the pulse shape after passage through the SF_6 cell; the damped amplitude modulation near the front of the pulse is due to the optical nutation effect.[1,5–7] For reasons not yet completely clear to us, the shape of the pulse, though not so much the nutation frequency, was found to be more sensitive to the location of the detector in the beam cross section than expected[8]; Fig. 1(d) gives another trace of the output pulse when the detector position was slightly displaced from that for Fig. 1(c).

With these results on the optical nutation effect, which are greatly improved as compared with the initial results reported in Ref. 1, it is now possible to determine the transition moment of the corresponding SF_6 transition from the measured nutation frequency. In analogy with the nuclear magnetic transient nutation effect,[9] the measured nutation frequency in the optical case is[5]

$$\Omega=(\Omega_0^2+\Delta\omega^2)^{1/2}, \tag{1}$$

where

$$\Delta\omega=\omega_0-\omega_{mn} \tag{2}$$

is the difference between the frequency ω_0 of the incident radiation and the transition frequency ω_{mn}; all the frequencies here refer to the angular frequencies. In the case when the initial and final levels of the transition are both nondegenerate, Ω_0 is simply related to the transition moment $\langle m|P_z|n\rangle$ and the amplitude of the electric field $\bar{E}_z$ of a linearly polarized step-function-

* Work supported by Office of Naval Research, under Contract No. 00014-67-0077-0006. Use was also made of the facilities of the Cornell Materials Science Center.

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[8] We expect some variation of the nutation frequency across the beam due to the transverse variation of electric field strength of the incident beam. The large variation of the nutation amplitude may be related to the complicated diffraction effects commented upon by McCall and Hahn (Ref. 3).

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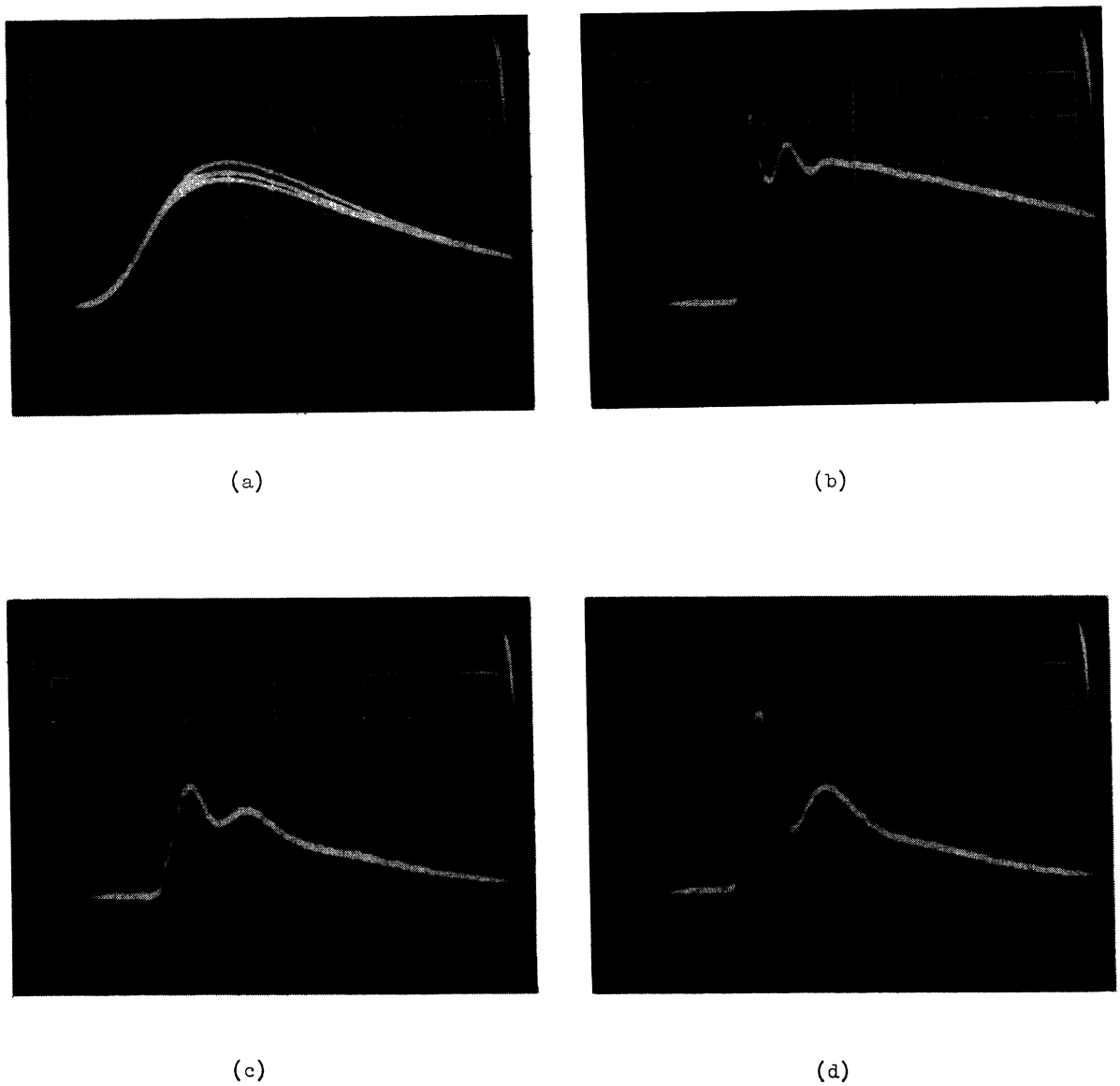

(a) (b)

(c) (d)

FIG. 1. Pulses transmitted through SF_6 absorption cell. Horizontal scale is 50 nsec/division. (a) Multiple traces of pulses without SF_6 in cell. Peak intensity could be varied from $\sim 3\frac{1}{2}$ kW/cm² to ~600 W/cm² using attenuators between the laser and the SF_6 cell. (b) Output through the SF_6 cell for a high-intensity pulse. $P_{SF_6}=0.16$ Torr. Vertical scale is 665 W/cm² division. (c) Output through SF_6 cell for a low-intensity pulse. $P_{SF_6}=0.12$ Torr. Vertical scale is 333 W/cm² division. (d) Same as (c) except the detector was moved slightly across the beam.

type incident wave[1,5,6]:

$$\Omega_0^2=\frac{\bar{E}_z^2|\langle m|P_z|n\rangle|^2}{\hbar^2}, \tag{3}$$

provided the amplitude modulation due to the nutation effect is small compared to the incident intensity. Here $\hbar$ refers to Planck's constant and $\langle m|$ and $|n\rangle$ refer to the upper and lower states of the transition. On the other hand, if the upper and lower levels are both degenerate, as was pointed out in Ref. 6, there will not be a single nutation frequency unless thermal equilibrium among the degenerate states of each level is essentially maintained. In that case, Ω_0 becomes —for example, for the molecular rotational-vibrational transitions—simply related to the average value, $[\mathrm{Av}(|\langle m|P_z|n\rangle|^2)]^{1/2}$ of the transition moments between all the degenerate states of the upper and lower levels[6]:

$$\Omega_0^2=\frac{\bar{E}_z^2\mathrm{Av}(|\langle m|P_z|n\rangle|^2)}{\hbar^2}, \tag{4a}$$

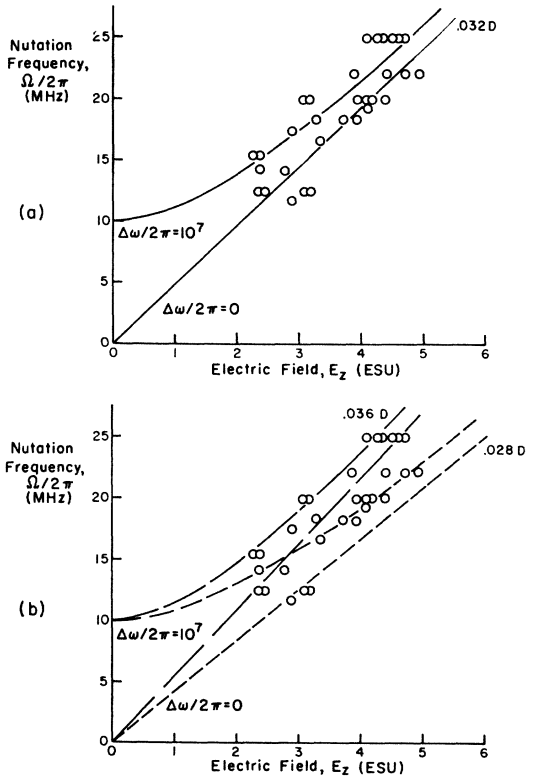

FIG. 2. Plots of observed nutation frequency Ω versus electric field amplitude. (a) Theoretical curves for $[\mathrm{Av}(|\langle m|P_z|n\rangle|^2)]^{1/2}=0.032$ D with $\Delta\omega/2\pi=0$ and 10 MHz. (b) Theoretical curves for $[\mathrm{Av}(|\langle m|P_z|n\rangle|^2)]^{1/2}=0.028$ and 0.036 D with $\Delta\omega/2\pi=0$ and 10 MHz. Each datum point represents an independent run, which includes refilling the absorption cell, recentering the detector, etc.

where

$$\mathrm{Av}(|\langle m|P_z|n\rangle|^2)=\frac{1}{2}\left(\frac{1}{2J_m+1}+\frac{1}{2J_n+1}\right)\times\sum_{M_m,M_n}|\langle v_m J_m l_m M_m|P_z|v_n J_n l_n M_n\rangle|^2. \quad (4b)$$

Here the quantum numbers J, l, M, and v refer, respectively, to the total angular momentum, the projection of J along a symmetry axis of the molecule, the projection of J along the space-fixed z axis, and the vibrational state of the molecular level. P_z in Eqs. (3) and (4) is the z component of the electric dipole operator of the molecule defined relative to the space-fixed axis.

In the case of SF_6, considerable uncertainties still exist as to the actual rotational levels involved in the 10.57-μ transition. Since our experimental results show clearly the presence of a single nutation frequency, and since the $J=0\rightarrow 0$ transition is forbidden, we can assume that Eq. (4) must apply, i.e., thermal equilibrium among the degenerate states is essentially maintained during the passage of the pulse. With the measured value of the nutation frequency Ω as a function of the applied $\bar{E}_z$ field, we can now determine the value of $[\mathrm{Av}(|\langle m|P_z|n\rangle|^2)]^{1/2}$ on the basis of Eqs. (1) and (4).

Figure 2 gives the measured nutation frequencies for various values of $\bar{E}_z$ obtained on a large number of *independent* experimental runs. The scatter in the data points shown gives, therefore, a good indication of the general reproducibility of the results obtained with our simple setup. Part of the difficulty was simply due to the relatively poor readability of the photographs of the type shown in Fig. 1; with still better shaped input pulse into the SF_6 cell, this type of uncertainty can be minimized. Another very important factor was the variation in the value of $\Delta\omega$ from run to run, since the CO_2 laser we used was not frequency-stabilized and the value of $\Delta\omega$ for each run could be anywhere up to the half-width of the 10.57-μ line of SF_6, which is about 10 MHz at dry-ice temperature and at the pressure used. With a frequency-stabilized laser, the scatter in the data points due to this difficulty could be eliminated, and one should be able to determine Ω_0, and hence the transition moment, from the measured nutation frequency Ω with greatly improved accuracy. Even with the data points shown in Fig. 2, one can already determine the value of the transition moment with reasonable accuracy. To fit the data points shown in Fig. 2, the transition moment for the 10.57-μ line of SF_6 is estimated to be

$$[\mathrm{Av}(|\langle m|P_z|n\rangle|^2)]^{1/2}=0.032\pm0.003\ \mathrm{D}. \quad (5)$$

We plot in Fig. 2(a) the calculated nutation frequency as a function of the applied $\bar{E}_z$ field using this value, Eq. (5), for the transition moment and assuming $\Delta\omega=0$ in Eq. (1). To account for the fluctuations in the value of $\Delta\omega$, we also show the case for $\Delta\omega=2\pi\times10^7$ sec^{-1}, or approximately the half-width of the SF_6 line. It can be seen that most of the data points fall within the two calculated curves. In order to give a rough indication of the accuracy of this result, Eq. (5), we also show in Fig. 2(b) similar calculated curves for two other values of the transition moment: 0.036 and 0.028 D. These two values evidently do not fit the data; thus, the value for the transition moment assumed in Eq. (5) is probably quite good.[10]

[10] We note that Patel and Slusher (Ref. 2) also estimate this transition moment to be ~0.03 D. This agreement may, however, be fortuitous for two reasons: (1) In their estimate based upon the measured absorption coefficient, it was assumed that 1/300 of the SF_6 molecules are in the lower level of this transition. Since the rotational levels involved in the transition are still not certain to date, it is not possible to estimate this population density with confidence. (2) Their Eq. (1) refers to a $\theta=2\pi$ pulse, which is not the "threshold" for the onset of self-induced transparency. According to McCall and Hahn (Refs. 2 and 3), the threshold occurs at θ slightly greater than π. Using the threshold intensity of 10 W/cm² for a linearly polarized wave of pulse duration ~250 nsec as given by Patel and Slusher, we obtain a value of 0.046 D for the transition moment on the basis of the prescription given by McCall and Hahn. On the other hand, since it is extremely difficult to pinpoint this threshold from the self-induced transparency data because of the very slow variation of the absorbed energy with the incident intensity of the pulse, the threshold value might actually be greater than the value used. There is, therefore, no apparent discrepancy between the self-induced transparency data of Patel and Slusher and the optical nutation results given here.

The present experiment has, therefore, demonstrated for the first time the possibility of determining the transition moment of optical transitions on the basis of the optical nutation effect. The method avoids the need for determining the population densities of the levels involved, as is required in the usual method based upon the measurement of absorption coefficients. The estimate of the population densities can often be uncertain and impossible if the assignment of the transition is not known, as is the case in the 10.57-μ transition of SF_6.

No attempt has yet been made to relate directly the observed damping of the amplitude modulation due to the nutation effect to the relevant relaxation rates of the optical transition, since the observed damping may also be seriously affected by the presence of Doppler broadening of the line and by certain complicated diffraction effects[3] as the self-modulating pulse of inhomogeneous wave propagates through the resonance medium.

PHYSICAL REVIEW VOLUME 184, NUMBER 2 10 AUGUST 1969

Effect of Radio-Frequency Fields on Mössbauer Spectra*

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The effect of a radio-frequency field on Mössbauer spectra is reconsidered. Both the ground and the excited state of the source nuclei are simultaneously exposed to the rf magnetic field. As an example, the effect of the rf field on ^{57}Fe nuclei in a source with magnetic splitting (detected by a single-line absorber) is discussed in detail for two experimental geometries. Mössbauer spectra taken at a constant velocity, but with varying radio frequency, show that the main features can be understood in terms of two-quantum processes (rf-induced transitions between the magnetic sublevels preceding or subsequent to the emission of a γ quantum), as long as the rf amplitude is small compared to the static magnetic field at the nucleus.

I. INTRODUCTION

THE first theory of the influence of a radio-frequency (rf) field on the nuclear Zeeman lines has been given by Hack and Hammermesh,[1] whose treatment is an extension of the Weisskopf-Wigner theory of spontaneous emission. They give an expression for the emission-line shape for the case in which the rf field acts only on the excited state of the nuclei. Recently, Mitin[2] investigated the problem of "γ magnetic resonance" in a ferromagnet and calculated, using second-order perturbation theory, the effective absorption cross section for two-quantum transitions (simultaneous absorption of a Mössbauer quantum and a quantum of the external rf field). No external constant magnetic field was assumed, and his final expression is for the Bloch-wall activity in a fine powder of particles with several domains.

At the present time, it is by no means clear whether the *pure* rf resonance effect on the Mössbauer line shape has been observed at all. Heiman *et al.*[3] showed experimentally that in thin iron foils rf fields generate acoustical sidebands in the Mössbauer spectra and suggested that the resonance effect reported by Matthias[4] is due to sideband overlap with the normal Mössbauer lines. Perlow,[5] on the other hand, demonstrates the destruction of the Mössbauer hyperfine pattern by a nonresonant rf field and attributes the effect to rf-induced domain-wall motion causing alterations in the directions of hf fields. Both phenomena make it difficult to observe the rf resonance effect on the Mössbauer line shape in ferromagnetics. Working with single-domain samples still requires suppression of the disturbing influence of magnetostriction. Besides solving the problem for Mössbauer isotopes in ferromagnets by appropriate experimental means, it is worthwhile to investigate the possibilities of the rf method for isotopes with long-lived excited states in samples without magnetostriction. Matthias[4] has already mentioned that Mössbauer NMR might become an experimental technique of high sensitivity. Particularly, a small linewidth makes magnetic hf splitting observable in relatively small fields; therefore, a large ratio of the rf field amplitude to the dc magnetic field can be obtained without relying on the hyperfine enhancement which gives ferromagnets their favored position among host materials.

In this paper, the effects of the rf field on both the ground and excited states are treated on an equal

* Work performed under the auspices of the U. S. Atomic Energy Commission.
† On leave from Institut für Theoretische Physik A, Technische Universität Braunschweig, Braunschweig, Germany.
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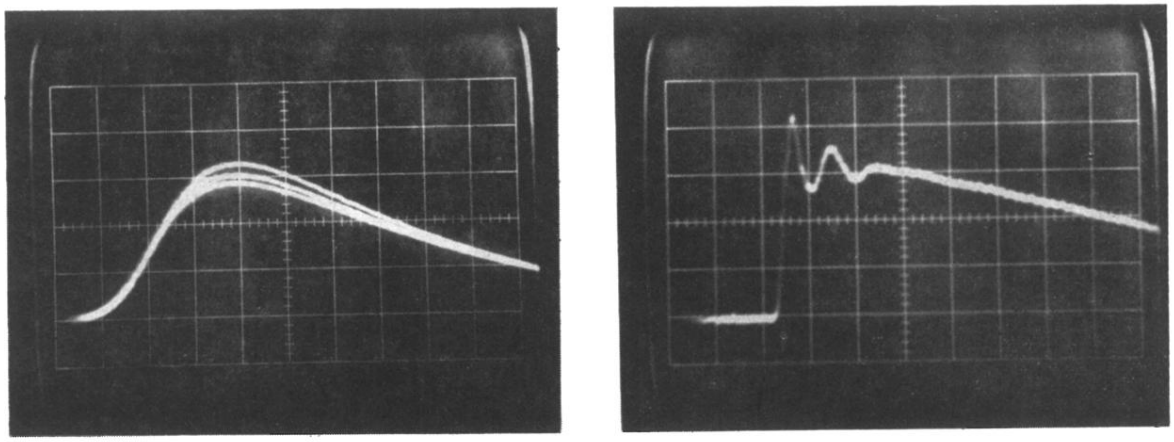
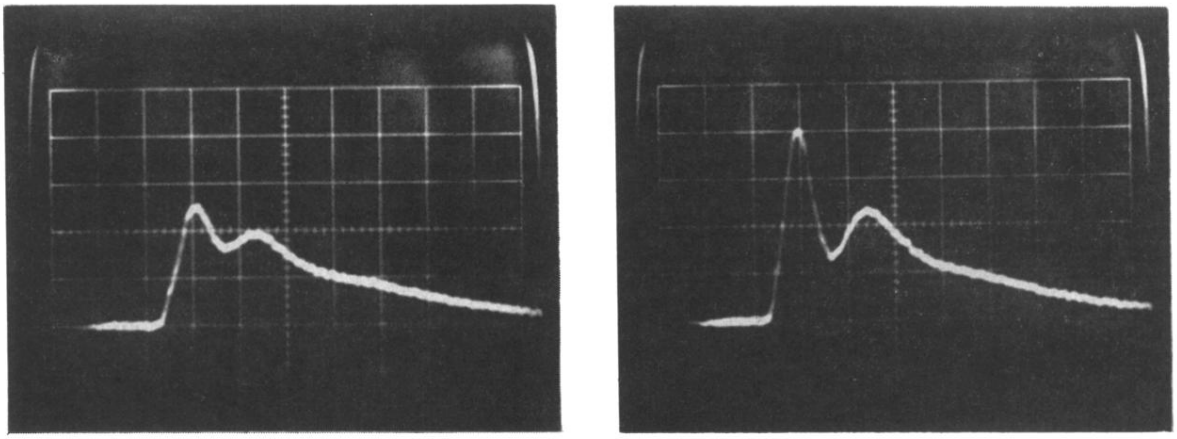


(a) (b)



(c) (d)

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