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 ν_3 band.[9] The Q-branch central peaks appear at 946.7 ± 0.3 cm^{-1} for the $\nu_6 \rightarrow \nu_3 + \nu_6$ transition and at 947.6 ± 0.3 cm^{-1} for the fundamental ν_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.

We would like to thank J. P. Gordon, S. L. McCall, and C. H. Wang for helpful discussions, Mrs. D. F. Sturman for obtaining the SF_6 absorption spectra, Dr. J. E. Griffiths for interpretations of the SF_6 spectra and helpful discussions, and R. J. Kerl for technical assistance.

[1]N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Letters 13, 567 (1964).
[2]I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966).
[3]E. L. Hahn, Phys. Rev. 80, 580 (1950).
[4]S. L. McCall and E. L. Hahn, Phys. Rev. Letters 18, 908 (1967).
[5]C. K. N. Patel and R. E. Slusher, Phys. Rev. Letters 19, 1019 (1967).
[6]A. L. Bloom, Phys. Rev. 98, 1105 (1955).
[7]S. L. McCall and E. L. Hahn, to be published.
[8]See footnote 5 of Ref. 5.
[9]R. T. Lageman and E. A. Jones, J. Chem Phys. 19, 534 (1951).

PHOTON-ECHO DEPENDENCE ON INTENSITY*

A. Compaan, L. Q. Lambert, and I. D. Abella
Department of Physics, University of Chicago, Chicago, Illinois 60037
(Received 1 March 1968)

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-

tions $\vec{k}_1$ and $\vec{k}_2$, the echo radiates in direction $\vec{k} = 2\vec{k}_2 - \vec{k}_1$ with an intensity

$$I(\vec{k}) = \tfrac{1}{4}NI_0(\vec{k})\sin^2\theta_1 \sin^4\theta_2 \times\{N|[\exp i(\vec{k}+\vec{k}_1-2\vec{k}_2)\cdot\vec{r}]_{av}|^2-1\}.$$

The degree of excitation of the two-level system is determined by the integrated pulse amplitude

$$\theta = \gamma\int E(t)dt \propto \sqrt{I}_{\text{input}},$$

where γ is the "gyroelectric" ratio and E is the optical electric field amplitude. The exponential factor summed over the sample results in a directional pulse.

Figures 1(a) and 1(b) show, respectively, the result of varying the first and second pulse intensities plotted as a function of I for the former and I^2 for the latter. One observes a linear dependence in the region $\sin^2\theta_1 \approx \theta_1^2$, or $\sin^4\theta_2 \approx \theta_2^4$, respectively, in agreement with the above predictions, with departures at higher intensity, indicating that a significant superposition of excited and ground levels occurs in our sample. At higher intensities, the curves cannot be fitted well with the theoretical functions because of the multimode nature of the source and the nonuniformities in the laser-beam cross section. The echo signal is the sum of contributions from several modes, since the system does not itself select a favorable mode as in the induced transparency experiments.[5,6]

With these results as an average intensity calibration, we have tested the echo dependence on magnetic field orientation with respect to the optic axis, as a function of the incident pulse intensities. It had been shown[1,2] that a small external magnetic field parallel to the optical axis enhances the photon-echo signal. A tentative model[2,7] based on nonisotropic Cr^{3+} g factors, on Cr^{3+}-Al interactions, and on the relative superposition of ground and excited levels produced by the incident pulse was proposed to account for the dephasing observed as function of magnetic field strength, field orientation, and pulse separation. Although difficulties with this model have been pointed out,[7,8] the present experiment makes a direct experimental test of the dephasing dependence on the relative superposition of the $^4A_2$ and $^2E(\bar{E})$ levels in ruby. The dephasing was based on the forced precession of Al nuclei arising from the reorientation of the

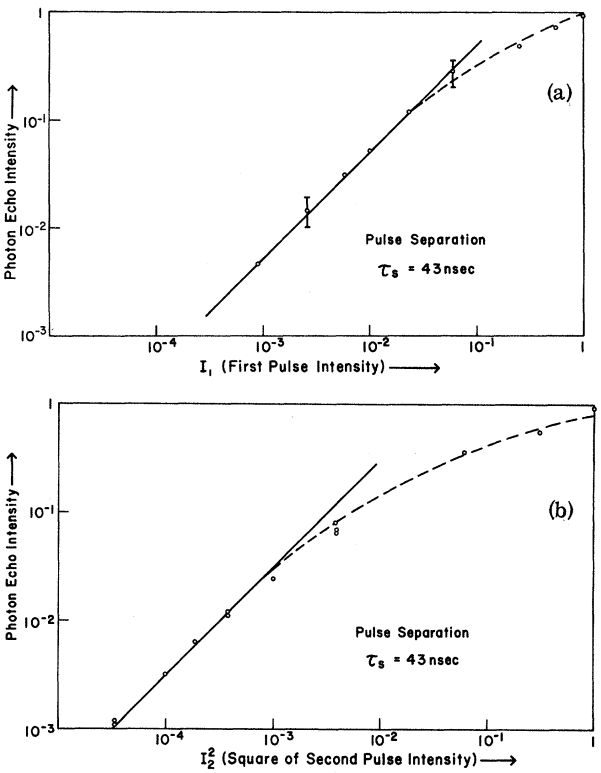

FIG. 1. (a) Echo intensity versus I_1, the first pulse intensity. (b) Echo intensity versus I_2^2, the square of the second pulse intensity. The external 200-G field was parallel to the sample optic axis.

Cr electronic spin, with a resulting modulation of the photon-echo states by the time-dependent magnetic fields at the Cr site. This change in local field depends approximately on $\sin(\chi'-\chi)$, where χ and χ' are the spin orientations with respect to the optic axis before and after the first pulse, and the angular dependence of the echo on θ_1 is given to a first approximation by

$$\sin^2\tfrac{1}{2}\theta_1 \sin\chi.$$

Figure 2 shows the photon-echo signal as a function of field orientation with respect to the sample optic axis for various first-pulse intensities, $\propto\theta_1^2$, as calibrated by Fig. 1(a). The total field was maintained at a constant 200 G. We note that echoes are observed out to essentially 90° field angle (for fixed pulse separation, τ_s = 43 nsec). To within an experimental uncertainty of ±15%, the slopes of the curves for a number of different runs are independent of intensity at high intensity as well as in the region of $\sin\theta_1 \sim \theta_1$. Similar results have also been observed as a function of second-pulse intensity. This constitutes independent experimental evidence

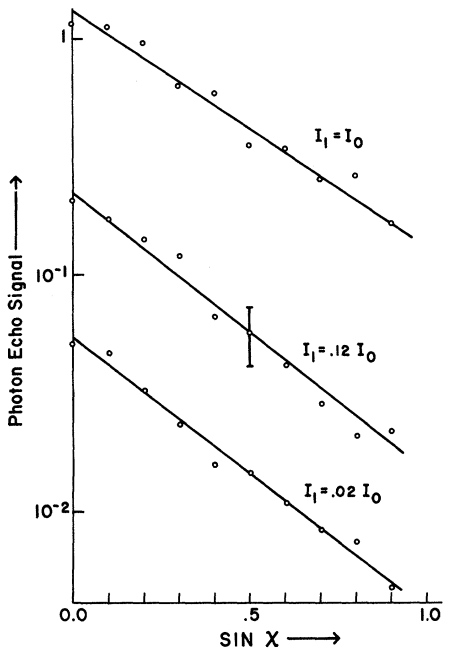

FIG. 2. Angular dependence results for several input intensities I_1, as calibrated by Fig. 1(a), where I_0 corresponds to an average θ_1 of the order of $\frac{1}{2}\pi$. Results from repeated runs show the slope to be independent of input intensity to within an uncertainty of ±15%.

that pulse-induced dephasing is not important here. The magnetic field effects are believed to be due to Cr^{3+}-Al interactions,[2,8] and the details remain to be calculated.

The technical assistance of Mr. Fred Rachford is gratefully acknowledged.

*Work supported by the Advanced Research Projects Agency (Project DEFENDER) and monitored by the U. S. Army Research Office-Durham, Contract No. DA-31-124-ARO-D-426.

[1]N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Phys. Rev. Letters 13, 567 (1964).

[2]I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966).

[3]I. D. Abella, A. Compaan, and L. Q. Lambert, Bull. Am. Phys. Soc. 13, 55 (1968).

[4]E. L. Hahn, Phys. Rev. 80, 580 (1950).

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

[6]C. K. N. Patel and R. E. Slusher, Phys. Rev. Letters 19, 1019 (1967).

[7]N. A. Kurnit and S. R. Hartmann, in Interaction of Radiation with Solids; Proceedings of the Cairo Solid State Conference at the American University at Cairo, Egypt, 1966, edited by A. Bishay (Plenum Press, Inc., New York, 1967), p. 693.

[8]D. Grischkowsky and S. R. Hartmann, Phys. Rev. Letters 20, 41 (1968).

NUCLEAR RELAXATION AND PAIR CORRELATION IN PARAMAGNETS

B. G. Silbernagel* and V. Jaccarino*
Department of Physics, University of California, Santa Barbara, California

and

P. Pincus†
Department of Physics, University of California, Los Angeles, California

and

J. H. Wernick
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 11 March 1968)

From NMR relaxation studies, experimental and theoretical evidence is obtained for electron-spin pair-correlation effects in paramagnets.

Nuclear spin-lattice relaxation caused by indirect hyperfine coupling between paramagnetic spins and the nuclei of nonmagnetic ions has been studied in the paramagnetic state of magnetically ordered systems. These measurements were made at sufficiently high temperatures for critical fluctuation phenomena to be unimportant. Direct measurements of the spin-lattice relaxation time (T_1) were made on the Al^{27} nuclei in the XAl_2 (X = rare earth) intermetallic compounds using transient techniques. An unexpected but substantial temperature variation of T_1 of Al^{27} is observed at high temperatures. It is also noted that a large, and as yet unexplained, temperature variation of the F^{19} linewidth ($\Delta H \sim 1/T_2 \sim 1/T_1$) exists well above T_N in MnF_2. We will demonstrate that both the sign and magnitude of the temperature dependences are associated with