Using η_1 leads to $N_1 = 4\times10^5$ sec^{-1} for the photon arrival rate. The same quantity can be calculated theoretically as[5]

$$N_1 = 512\pi^5 hcd_{36}^2 \sin^2\theta P_p L \csc(\varphi_1+\varphi_2)\Delta\lambda_1\Delta\alpha_1 / n_p n_2 \lambda_1^5 \lambda_2 = 8.0\times10^5 \text{ sec}^{-1},$$

in reasonable agreement with experiment. Here d_{36} is a nonlinear coefficient of the crystal, θ and φ are internal angles, and $\Delta\alpha_1$ is the angle subtended at the crystal by detector 1.

This experiment has only set an upper bound on τ_C, the deviation from the coincidence condition, Eq. (1). Presumably, a photon can be no better localized in time than the inverse of its bandwidth. Thus the theoretical lower limit of τ_C is the inverse of the smaller bandwidth $\Delta\lambda_2 = 1.5$ nm, or $\tau_C = 2\times10^{-13}$ sec. One might possibly expect that τ_C is related to the coherence time of the pump laser. The He-Cd laser oscillates in about 15 longitudinal modes, with a total bandwidth about 10^9 Hz, corresponding to $\tau_C = 2\times10^{-10}$ sec, which is still too small to observe in the present experiment. With a single-mode laser, a coherence time >100 nsec is possible, so that one would expect a much smaller coincidence rate if τ_C were really determined by the pump coherence time.

The expected accidental coincidence rate is

$$\langle R_C(\text{acc})\rangle = \tau_G\langle R_1R_2\rangle = \tau_G\langle R_1\rangle\langle R_2\rangle + \tau_G\langle\Delta R_1\Delta R_2\rangle \quad (4)$$

(with backgrounds now included in the R's). Eq. (4) has been checked in the experiments of Figs. 2(b) and 4, where R_1 and R_2 are kept constant but Eqs. (1) and (2) are violated in order to eliminate real coincidences. In both cases R_C falls to a low level consistent with the first term on the right-hand side of Eq. (4), without any need to invoke the last term, which might arise from fluctuations of pump power. For large τ, $\langle R_C(\text{acc})\rangle$ was measured more accurately at higher R_1 and R_2, and agreed with $\tau_G\langle R_1\rangle\langle R_2\rangle$ within 10%. The same formula was also verified with a dc-powered incandescent light source. In summary, the maximum observed real coincidence rate was at least 100 times any unexplained residual coincidence rate.

The authors thank N. Knable for encouragement and cooperation, and G. Schappert, E. Sarachik, and B. Mollow for helpful theoretical discussions.

[1]Ya. B. Zel'dovich and D. N. Klyshko, Zh. Eksp. Teor. Fiz.–Pis'ma Red. 9, 69 (1969) [JETP Lett. 9, 40 (1969)].

[2]S. E. Harris, M. K. Oshman, and R. L. Byer, Phys. Rev. Lett. 18, 732 (1967); D. Magde and H. Mahr, Phys. Rev. Lett. 18, 905 (1967); and many later papers.

[3]D. Magde and H. Mahr, Phys. Rev. 171, 393 (1968).

[4]R. Foord, R. Jones, C. J. Oliver, and E. R. Pike, Appl. Opt. 8, 1975 (1969).

[5]D. A. Kleinman, Phys. Rev. 174, 1027 (1968); D. L. Weinberg, Appl. Phys. Lett. 14, 32 (1969).

FAR-INFRARED OBSERVATION OF ELECTRIC-DIPOLE–EXCITED ELECTRON-SPIN RESONANCE IN $Hg_{1-x}Cd_xTe$

B. D. McCombe, R. J. Wagner, and G. A. Prinz
Naval Research Laboratory, Washington, D. C. 20390
(Received 18 May 1970)

Electric-dipole–excited conduction-electron spin resonance has been observed for the first time in the far infrared in the II-VI semiconductor $Hg_{1-x}Cd_xTe$. Circular polarization and intensity studies as a function of magnetic field unambiguously identify this transition. Electron g values are obtained for the $L=0$ Landau level at several magnetic fields.

We report in this Letter what we believe to be the first observation of the electron-spin resonance transition excited by electric dipole radiation in a semiconductor in the far infrared.

A number of years ago "combined resonance" transitions, i.e., electric-dipole–excited transitions involving a change in the spin state of the carriers, were theoretically predicted for semiconductors with large spin-orbit interaction either having a small energy gap[1,2] (the nonparabolicity mechanism) or lacking a center-of-inversion symmetry[3,4] (the inversion-asymmetry mechanism).

Reported experimental observations of electric-dipole–excited transitions involving a spin flip thus far include the pure spin-flip transition (ΔL

$=0$, $\Delta S=-1$) for InSb in the microwave region,[5] and the spin-flip plus cyclotron transition ($\Delta L=1$, $\Delta S=-1$) for InSb[6,7] in the infrared. Here L is the Landau quantum number and S is the spin quantum number. The latter measurements demonstrated conclusively that the nonparabolicity mechanism is the dominant one for small-gap semiconductors. In addition there have been reports of spin-flip plus cyclotron combined resonance transitions in the stressed valence band of Ge.[8,9]

$Hg_{1-x}Cd_xTe$ is an excellent material for the study of combined resonance. For $x>0.16$ this alloy system has the InSb-type band structure with the conduction and valence bands of interest at the Γ point of the Brillouin-zone center. For $x<0.16$ the conduction and valence bands are inverted forming a zero thermal-energy-gap structure as in gray tin.[10] The energy gap for the alloy composition studied here ($x=0.193$) is of the order of 60 meV. A rough calculation indicates that combined resonance should be approximately 150 times stronger in this material than in InSb at the same magnetic field and carrier concentration.

The theory for the nonparabolicity mechanism predicts a transition at the spin resonance energy $h\nu_s$ ($\Delta L=0$, $\Delta S=-1$) which for materials having an InSb-type conduction band is allowed only for right-circular polarization,[3] i.e., the cyclotron-resonance inactive polarization. For a degenerate electron gas of low concentration (all electrons in the lowest Landau level with spin up), the integrated absorption for this transition is given by

$$(\alpha\Delta B)_{N-P}=\frac{32\pi^6|P|^4c\hbar}{27n}\left[\frac{1}{E_g^{\,2}}-\frac{1}{(E_g+\Delta)^2}\right]^2\frac{N^3}{B}, \quad (1)$$

where P is the conduction-valence-band momentum matrix element, E_g is the energy gap, Δ is the valence-band spin-orbit–splitting parameter, n is the index of refraction, N is the carrier concentration, and B is the magnetic field. The condition that all electrons be in the lowest energy level is approximately satisfied in our sample for the magnetic fields at which the transition was observed. The zero-field Fermi energy for this sample is estimated to be 10-11 meV at 4.2 K. At 7 kG (the lowest field at which the transition was observed) the energy of the $L=0$, spin-down Landau level is calculated to be approximately 10 meV. Hence there may be some population of the upper spin state at this field. This is, in fact, indicated by some of the measurements to be discussed.

The sample used in these experiments has an electron concentration of about 3.4×10^{15} cm^{-3} and a mobility of 2.7×10^5 $cm^2/V\ sec^{-1}$ at liquid-nitrogen temperature.[11] The sample thickness was 0.76 mm. A rather unique dual-beam, pulsed, water-vapor laser[12] was used as the source of far-infrared radiation. This system provides numerous relatively intense lines throughout the infrared. Further details of this system will be published elsewhere.[13] Light-pipe optics were used with the sample placed in the Faraday (longitudinal) configuration in helium exchange gas at the center of the bore of a Nb-Sn superconducting magnet capable of 95 kOe. The magnetic field was measured with a copper magnetoresistance probe calibrated by NMR.

The most crucial part of the experiment consisted of determining the polarization properties of the observed line. As indicated by the preceeding discussion, the electron-spin-resonance line is allowed only for right-circular polarization. In order to perform polarization studies, quartz plates were fabricated of the proper orientation and thickness to function as quarter-wave plates at 119 and 220 μm. Circular polarization was obtained by using these quarter-wave plates with a linear polarizer oriented at 45 deg with respect to the optic axis. These units were placed in the light pipe immediately before the sample at liquid-helium temperature. The sense of circular polarization was reversed by simply reversing the direction of the applied magnetic field. The degree of polarization and its sense were determined at both wavelengths by observing cyclotron resonance in InSb.

The results of the circular polarization study at 118.6 μm are shown in Fig. 1. The upper trace of this figure shows the relative transmission as a function of magnetic field for the cyclotron-resonance inactive polarization (CRI) (right-circular polarization in the convention of Refs. 2 and 7). The lower trace of the figure shows the transmission for the cyclotron-resonance active polarization (CRA) (left-circular polarization). The monotonically increasing background in these figures is due to a combination of plasma and cyclotron-resonance effects. There is a strong absorption line appearing in the CRI polarization but essentially nothing in the CRA polarization. Similar results were obtained at 220 μm. This is strong evidence that the observed line is indeed electric-dipole–excited electron spin resonance. The fact that the line occurs only for the

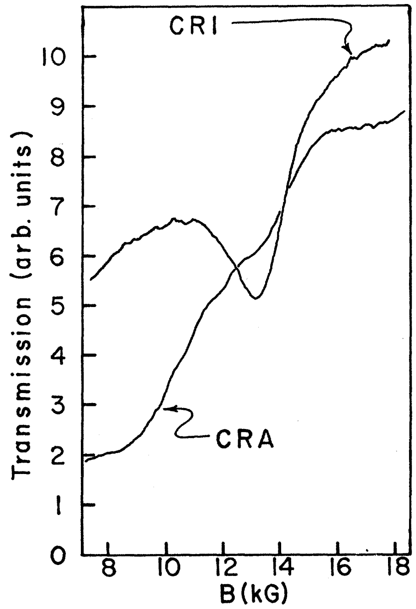

FIG. 1. A plot of relative transmission at 4.3°K for the observed line for two opposite circular polarizations, CRI and CRA. The laser wavelength is 118.6 μm.

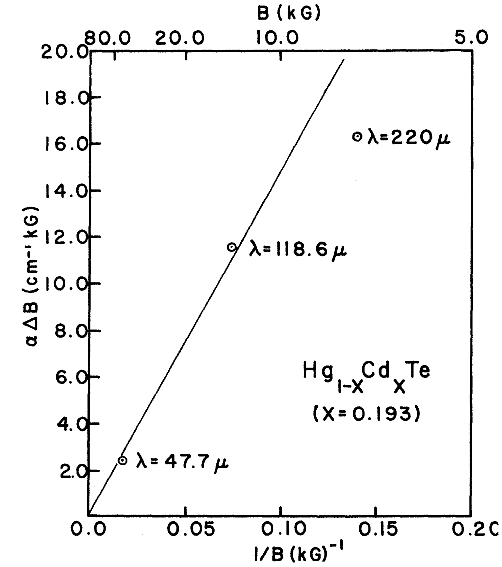

FIG. 2. Integrated absorption ($\alpha\Delta B$) obtained at three laser wavelengths versus $1/B$. The solid straight line is drawn for reference purposes.

CRI polarization shows conclusively that it cannot be due to spin-down cyclotron resonance in this extremely nonparabolic conduction band.[14]

Further evidence that the observed transition is electron spin resonance excited by electric dipole radiation is provided by a study of the magnetic field dependence of the intensity of this line. The product of the peak absorption constant α times the full width at $\frac{1}{2}$ peak absorption ΔB is taken as a reasonable measure of the integrated absorption which can be compared with the theoretical prediction of Eq. (1). A plot of $\alpha\Delta B$ is shown in Fig. 2 for data taken at three different laser wavelengths. Certainly to well within experimental error the two lowest points fall on a straight line drawn through the origin as indicated in the figure. This is in agreement with the theoretical predictions of Eq. (1). There appears to be a "tailing off" of the integrated absorption at 220 μm. This is consistent with a partial population of the upper spin state which reduces the intensity.

It is of interest to compare quantitatively the calculated and experimental integrated absorption. At $\lambda = 118.6$ μm the line occurs at a magnetic field of 13.5 kG. Assuming a helium-temperature carrier concentration of 2.3×10^{15} cm^{-3}, the calculated value of $\alpha\Delta B$ at this field from Eq. (1) using $\Delta = 0.96$ eV, $E_g = 0.056$ eV, and $E_P = \hbar^2P^2/2m = 18.5$ eV is given by $(\alpha\Delta B)_{NP} = 38$ cm^{-1} kG; the measured value is about 12 cm^{-1} kG, a reasonable agreement. The parameters for the calculation were obtained as indicated below.

From the peak positions of the electron-spin-resonance line we can obtain the effective g value of the lowest Landau level as a function of magnetic field from the usual expression $h\nu_s(0) = g(0)\beta H$, where β is the Bohr magneton. The g-value results are plotted versus magnetic field in Fig. 3. In obtaining these points no correction was made to the line positions to take account of any possible plasma effects. It does not appear

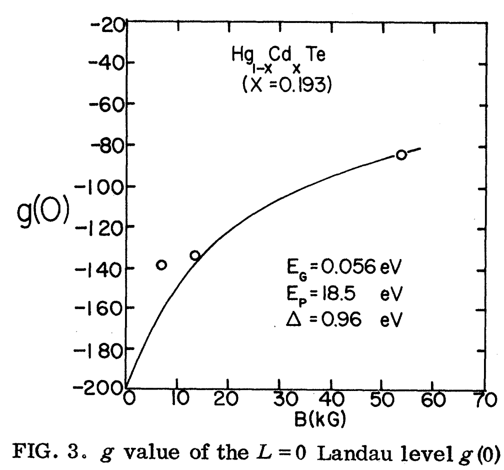

FIG. 3. g value of the $L=0$ Landau level $g(0)$, plotted increasing negatively downward, versus magnetic field. Open circles: experimental points; solid line: fit to Bowers and Yafet theory with the parameters indicated.

that plasma effects are important for spin transitions since the only possible coupling is via the spin-orbit interaction which is relatively weak. This assertion is substantiated by the microwave measurements of electron spin resonance in InSb.[5, 15] Both these measurements were performed in the presence of strong plasma-shifted cyclotron resonance, and no effects of the plasma on the spin lines were observed. Figure 3 also shows, as the solid line, the g value calculated from the theory of Bowers and Yafet.[16] The parameters in this theory, E_g, Δ, and E_P, were obtained as follows: Δ and E_P were obtained from a fit to cyclotron-resonance and combined-resonance ($\Delta L=1$, $\Delta S=-1$) data taken on a sample having $x=0.203$[17]; the energy gap was then adjusted to provide a fit to the transition at 53.6 kG. The resulting calculated curve shows a significant deviation from the experimental point at 7 kG, the calculated g values being larger in absolute magnitude. This discrepancy can be attributed to partial population of the upper spin state of the lowest Landau level as indicated by the intensity studies and the estimate of the Fermi level. Calculations show that partial population of this level, occupying states away from $k_z=0$, can account for a shift to lower energy in the observed transition energy due to the large nonparabolicity. This shift, which results from blocking of transitions at and near $k_z=0$, is of the correct order of magnitude to account for the apparent discrepancy in the g value. In the limit of very low magnetic fields the measured g value is that at the Fermi energy. This is also consistent with observations of the linewidth which showed a decrease of more than a factor of 1.5 for this lowest field line compared with the line at 13.5 kG.

The authors would like to express their gratitude to Professor R. N. Dexter of the University of Wisconsin for making these experiments possible through the loan of the excellent $Hg_{1-x}Cd_xTe$ samples which were originally grown by Honeywell, Inc. We would also like to thank Dr. M. A. Kinch of Texas Instruments, Inc., for suggesting the possibility that the observed line was a spin transition.

[1]Y. Yafet, in *Solid State Physics,* edited by F. Seitz and D. Turnbull (Academic, New York, 1963), Vol. 14, p. 1.

[2]V. I. Sheka, Fiz. Tverd. Tela 6, 3099 (1964) [Sov. Phys. Solid State 6, 2470 (1965)].

[3]E. I. Rashba and V. I. Sheka, Fiz. Tverd. Tela 3, 1735 (1961) [Sov. Phys. Solid State 3, 1257 (1961)].

[4]E. I. Rashba and V. I. Sheka, Fiz. Tverd. Tela 3, 1863 (1961) [Sov. Phys. Solid State 3, 1357 (1961)].

[5]R. L. Bell, Phys. Rev. Lett. 9, 52 (1962).

[6]B. D. McCombe, S. G. Bishop, and R. Kaplan, Phys. Rev. Lett. 18, 748 (1967).

[7]B. D. McCombe, Phys. Rev. 181, 1206 (1969).

[8]J. C. Hensel, Phys. Rev. Lett. 21, 983 (1968).

[9]J. C. Hensel and K. Suzuki, Phys. Rev. Lett. 22, 838 (1969).

[10]T. C. Harman, in *Proceedings of the International Conference on II-VI Semiconducting Compounds, Providence, R. I., 1967,* edited by D. G. Thomas (Benjamin, New York, 1968), p. 982.

[11]This is sample "*J*" of J. D. Wiley and R. N. Dexter, Phys. Rev. 181, 1181 (1969).

[12]G. A. Prinz and R. J. Wagner, Phys. Lett. 30A, 520 (1969).

[13]R. J. Wagner and G. A. Prinz, to be published.

[14]A similar line was reported at 195 μm in $Hg_{0.8}Cd_{0.8}Te$ which was attributed to spin-down cyclotron resonance. R. A. Lancaster, K. J. Button, B. Lax, T. L. Cronburg, and D. Soderman, Bull. Amer. Phys. Soc. 15, 365 (1970).

[15]G. Bemski, Phys. Rev. Lett. 4, 62 (1960).

[16]R. Bowers and Y. Yafet, Phys. Rev. 115, 1165 (1959).

[17]B. D. McCombe, R. J. Wagner, and G. A. Prinz, to be published.