Electric-Dipole-Excited Electron Spin Resonance in InSb

B. D. McCombe and R. J. Wagner
Naval Research Laboratory, Washington, D. C. 20390
(Received 29 March 1971)

Electric-dipole-excited electron spin resonance (EDE-ESR) has been observed in several samples of InSb in the far infrared. A qualitative discussion of the linewidth is given in terms of conduction-band nonparabolicity. Electron g values for the lowest Landau level obtained from these measurements are in disagreement with previous calculations and indirect measurements. These accurate spin energies provide evidence against the proposed existence of a strong electron–TO-phonon interaction in InSb.

I. INTRODUCTION

Electric-dipole-excited electron spin resonance (EDE-ESR) was recently observed in $Hg_{1-x}Cd_xTe$ using a far-infrared pulsed gas laser system.[1] We report in this paper the observation of EDE-ESR in several samples of InSb using both the far-infrared laser system and a Fourier-transform interferometric spectrometer. EDE-ESR was previously observed in InSb in the microwave region at a single frequency.[2] From the present studies, accurate measurements of the spin energy and electron g value of the lowest Landau level are obtained as a function of magnetic field from 27 to 75 kG. These results are in disagreement with previous indirect measurements[3,4] and calculations[5,6] of the spin energy at high magnetic fields. These accurate determinations of the spin energy have important consequences concerning the interpretation of "polaron anomalies" in the impurity combined resonance spectra[4,7] as discussed below.

For InSb and other small-gap materials it has been shown that the electric-dipole-excited spin-flip transitions are primarily allowed by the so-called nonparabolicity mechanism. Theoretical calculations[8] for this mechanism, based on the Bowers-Yafet model, predict a transition at the spin-resonance energy $h\nu_s$ ($\Delta L = 0$; $\Delta s = -1$). Here L is the Landau quantum number, and s is the spin quantum number. This transition is allowed only for right circular polarization [the cyclotron-resonance-inactive (CRI) polarization] in InSb, since the g factor is negative. The matrix element for the EDE-ESR transition is given by[8]

$$\langle L, s=\downarrow | \vec{v} | L, s=\uparrow \rangle \approx \tfrac{2}{3}\pi\sqrt{2}\,|P|^2[1/E_g^2 - 1/(E_g+\Delta)^2]\,\nu_s k_z \qquad (1)$$

for $\vec{B} \parallel \vec{z}$, where $\vec{v}$ is the CRI velocity operator, E_g is the energy gap, Δ is the spin-orbit splitting energy, and P is the s-p momentum matrix element. Hence the transition probability is proportional to k_z^2, the square of the wave vector parallel to the magnetic field. This result affects the linewidth and the precise position of the transmission minimum.

II. EXPERIMENTAL

Transmission experiments were performed on several samples of n-type InSb having excess donor concentrations at liquid-nitrogen temperature between 2×10^{15} and 6×10^{15} cm^{-3} and mobilities of 2×10^5 and 3×10^5 cm^2/V sec. A dual-beam pulsed H_2O-D_2O-SO_2 laser and a vacuum interferometric spectrometer in conjunction with high-homogeneity superconducting solenoids were used to obtain the spectra. Details of these systems have been given elsewhere.[9,10]

III. RESULTS AND DISCUSSION

Results of transmission measurements at three laser wavelengths at 4.3 °K are shown in Fig. 1. These data were taken with unpolarized radiation. A check of the polarization properties of the line at 171.7 and 118.6 μm, using circular polarizers placed immediately before the samples, showed the line to be predominantly CRI, as expected. The narrow linewidth varies from approximately 80 G at 118.6 μm to 200 G at 73.4 μm. The magnetic field width at 118.6 μm corresponds to a frequency width of 0.16 cm^{-1}, which is at the limits of resolution of the best Fourier-transform spectroscopy. Hence a study of the line shape is only possible with a laser system.

The temperature dependence of the EDE-ESR line at 118.6 μm is shown in Fig. 2. The line broadens and shifts to higher magnetic field as the temperature is raised, the width varying from 79 G at 5.5 °K to 310 G at 26 °K. At 39 °K the line is so smeared out that it is not possible to make an accurate measurement of the width, which is estimated to be ≈ 700 G. The integrated absorption of the line remains roughly constant over this temperature range. This indicates that the observed line is due to free carriers and not electrons localized at impurities, since increasing temperature would thermally depopulate localized levels causing the line intensity to decrease drastically. A careful

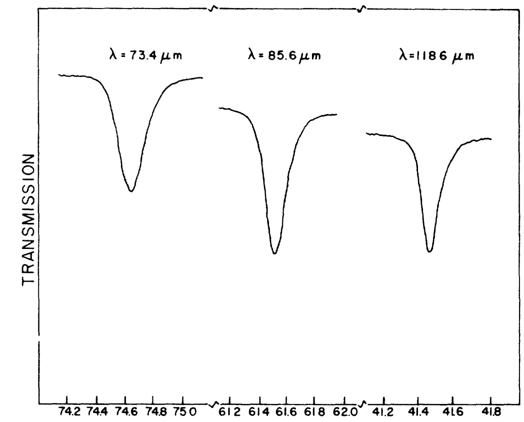

FIG. 1. Transmission vs magnetic field for three different H_2O laser wavelengths as indicated at 4.3 °K. The InSb sample had a carrier concentration of 6×10^{15} cm^{-3} and a mobility of 2×10^{5} cm^2/V sec.

search revealed no additional absorption structure in any of the samples which might be attributable to donor-impurity electron spin resonance.

Although very narrow by semiconductor standards, the EDE-ESR lines are much broader than the microwave ESR in InSb.[11] The width of the lines can be approximately accounted for by energy broadening due to the k_z^2 dependence of the transition probability [Eq. (1)] and the large nonparabolicity of the conduction band. For $H\neq0$ the energy difference between the spin states of the lowest Landau level at $k_z=0$ is larger than the corresponding energy difference at $k_z\neq0$. Since the transition probability is proportional to k_z^2 the peak absorption must occur for some value of k_z between $k=0$ and $k_z=k_F$ (the Fermi wave vector) for degenerate statistics. The singularity in the density of states at $k_z=0$ will favor small values of k_z but the peak will certainly occur for $|k_z|>0$. Hence the observed absorption peak occurs at lower energy (higher field) than is calculated for the energy levels at $k_z=0$. In addition, this mechanism should broaden the line by an amount somewhat less than, but of the order of, the energy difference

$$h[\nu_s(k_z=0)-\nu_s(k_z=k_F)]$$

for $kT<\epsilon_F'$. Here ϵ_F' is the Fermi energy at $T=0$ measured with respect to the bottom of the lowest spin-up Landau level. For $kT\gg\epsilon_F'$ the largest value of k_z which is significantly populated is of the order of $[2m^*kT/\hbar^2]^{1/2}$; and hence the line broadens and shifts to lower energies as the temperature is increased.

We may *estimate* the width and magnitude of the shift of the peak position from the approximate ($\Delta\gg E_g$) Bowers-Yafet expression for the magnetic energy levels,[12]

$$\left\{\begin{matrix} E_{L\uparrow} \\ E_{L\downarrow}\end{matrix}\right\} = E_g \times\frac{1}{2}\left\{1+\left[1+\frac{8P^2}{3E_g^2}\left(k_z^2+\frac{eB}{c\hbar}\,(2L+1\mp\tfrac{1}{2})\right)\right]^{1/2}\right\}. \quad (2)$$

The maximum spin-energy variation is given by

$$\Delta E_s=(E_{0\downarrow}-E_{0\uparrow})_{k_z=0}-(E_{0\downarrow}-E_{0\uparrow})_{k_z=k_{\max}}\,, \quad (3)$$

where $k_{\max}\approx k_F$ for $kT<\epsilon_F'$ and $k_{\max}\approx(2m^*kT/\hbar^2)^{1/2}$ for $kT\gg\epsilon_F'$. At 41.5 kG (the line position at 118.6 μm) $\epsilon_F'\approx0.75$ meV—as calculated for the sample of Fig. 1, ($N=6\times10^{15}$ cm^{-3}). From Eq. (3), ΔE_s is 0.096 meV or ΔB_s is 370 G, which is somewhat larger than, but comparable to, the observed linewidth. This agreement does not preclude other broadening mechanisms in addition to spin lattice relaxation. For example, due to the spin-orbit interaction, there can be an orbital contribution to the relaxation time.

The maximum shift in the position of the peak due to nonparabolicity must be of the order of the observed width, or 80 G at 118.6 μm. This corresponds to a maximum shift in the line position from the $k_z=0$ value of ≈ 0.2%. Our estimated experimental error in magnetic field determination is approximately 0.2–0.3%, hence the observed peak positions at low temperature are an excellent

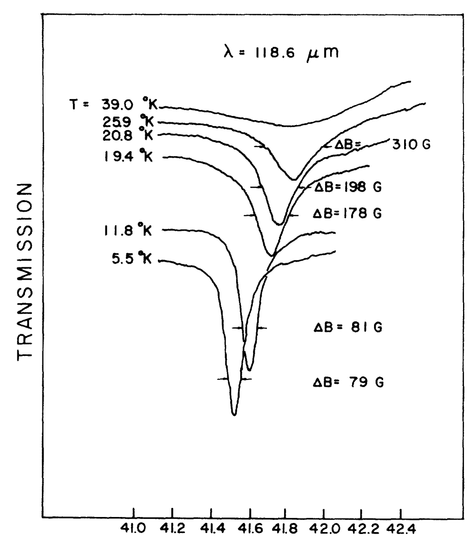

FIG. 2. Transmission vs magnetic field at 118.6 μm for several sample temperatures as indicated. The full widths in magnetic field at half-peak absorption constant are also shown.

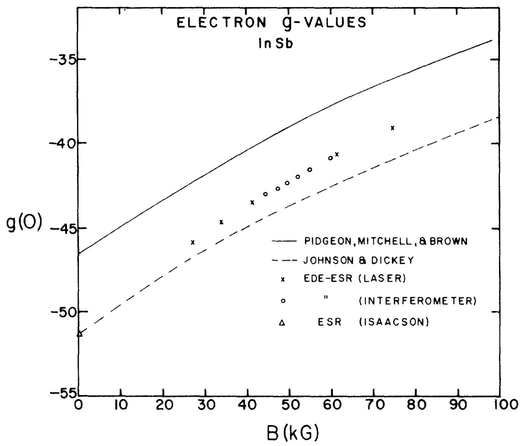

FIG. 3. Electron g values vs magnetic field. The legend indicates the origin of the lines and points. Experimental points were obtained at 4.3 °K for $\vec{B} \parallel [111]$ from the expression $g_{expt} = h\nu_s/\beta B$, where β is the Bohr magneton. The Pidgeon-Mitchell-Brown calculation is shown for $\vec{B} \parallel [111]$, and the Johnson-Dickey calculation is for $\vec{B} \parallel [110]$.

approximation to the spin energy at $k_z = 0$ at low temperature for this sample. This is not true of $Hg_{1-x}Cd_xTe$ ($x = 0.193$) where the correction is very large due to the extremely large nonparabolicty.[1]

As the temperature is increased, kT becomes appreciably larger than ϵ_F' and k_{max} becomes greater than k_F. Under these conditions we expect the line to shift to higher fields and broaden. For $T \approx 40$ °K, we calculate $\Delta E_s = 0.33$ meV or $\Delta B_s = 1500$ G in reasonable agreement with the observed width of 700 G and shift of 450 G. Part of the calculated shift with temperature may be offset by the opposite shift resulting from the decrease of the energy gap.

One puzzling feature of the data which is not accounted for by the above picture is the broadening of the line as the magnetic field is increased. Since ϵ_F' decreases as H is increased, it appears that the line should narrow as the magnetic field increases. The broadening may be related to the fact that as ϵ_F' decreases the sample changes from a region of degenerate statistics to nondegenerate statistics. To clear up this point and also to ascertain the exact position of the observed peak, a numerical calculation of the line profile is in progress and will be reported in a subsequent more detailed publication.

At 140.85 μm an asymmetric broadening of the line to lower field was observed which appears to be due to an unresolved splitting. This is the field region where the spin-down cyclotron resonance energy is approximately equal to the LO-phonon energy, and a resonant electron-phonon interaction may be responsible for the splitting.

The results of the g-value measurement for the $L = 0$ Landau level are shown in Fig. 3. Our experimental points are compared with the values calculated from fits to both interband data[5] (solid line) and intraband data[6] (dashed line). The present experimental values fall *between* the two calculations. They approach the dashed line at lower fields, as expected, since this calculation was adjusted to fit Isaacson's[11] low-field microwave ESR data (triangle).

A discrepancy between the interband and intraband determination of both the cyclotron mass and the g value in InSb and α-Sn has been previously pointed out.[6,13] Hence it is not surprising that our values disagree with the interband calculations (solid line); however, it is surprising that the present results deviate appreciably from the intraband data (at the highest experimental field the deviation is $\approx 4.5\%$). The present results are also in disagreement with our own previous measurements of the spin energy obtained from the difference between the combined resonance and spin-down cyclotron resonance data.[3] This is not well understood and is being investigated further. Differences between our [111] measurements and the [110] calculation of Johnson and Dickey cannot be accounted for by g-value anisotropy. This anisotropy gives a shift of only about 1% at 75 kG and is a linear function of magnetic field.[14]

An important consequence of this discrepancy in the spin-energies vs magnetic field concerns the controversy over the existence of an impurity electron–TO-phonon interaction in InSb.[4,7,15] Two separate interpretations have been given of very sim-

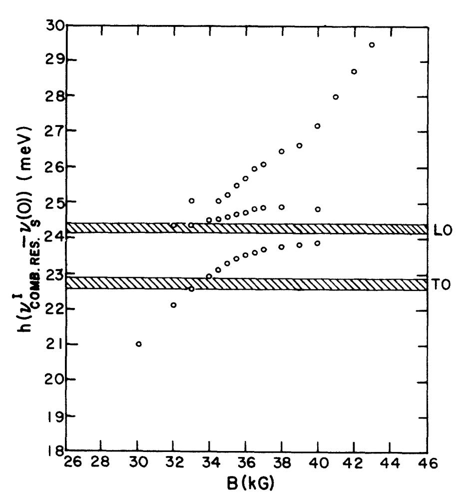

FIG. 4. A plot of the impurity combined resonance energy $h\nu^I_{comb. res.}$ from Ref. 6 minus the free carrier spin resonance energy $h\nu_s(0)$ vs magnetic field. The origin of the shaded bands indicated by LO and TO are discussed in the text. All data were taken with $\vec{B} \parallel [111]$.

ilar impurity combined resonance data in InSb. The first (Ref. 4) invoked an electron–TO-phonon interaction in addition to the usual electron–LO-phonon interaction to explain the *two* "polaron anomalies" observed in the combined resonance spectra; the second (Ref. 7) asserted that only the electron–LO-phonon interaction was important, and the upper "polaron anomaly" was due to coupling to an impurity excited state. Of crucial importance in the interpretation of this data is a knowledge of the impurity spin energy of the lowest Landau level. Figure 4 shows the combined resonance data of Ref. 7 with the $L=0$ spin energies obtained from the EDE-ESR measurements subtracted. The shaded bands labeled LO and TO cover the ranges of the values of these phonon energies quoted in the literature (see Ref. 7). The impurity spin energy will be smaller than the measured free carrier spin energy due to the effects of nonparabolicity on the impurity binding energies. This will raise the experimental points even higher. It is obvious that the lowest "anomaly" occurs at ω_{LO} and *not* ω_{TO}. Hence the interpretation in terms of LO-phonon coupling alone is apparently correct, and the impurity electron–TO-phonon coupling must be much weaker.

ACKNOWLEDGMENTS

The authors are indebted to Dr. E. J. Johnson for communication of his calculated spin energies. Thanks are also due Dr. G. A. Prinz for experimental assistance and discussions.

[1]B. D. McCombe, R. J. Wagner, and G. A. Prinz, Phys. Rev. Letters 25, 87 (1970).

[2]R. L. Bell, Phys. Rev. Letters 9, 52 (1962).

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

[4]D. H. Dickey and D. M. Larsen, Phys. Rev. Letters 20, 65 (1968).

[5]C. R. Pidgeon, D. L. Mitchell, and R. N. Brown, Phys. Rev. 154, 737 (1967).

[6]E. J. Johnson and D. H. Dickey, Phys. Rev. B 1, 2676 (1970).

[7]B. D. McCombe and R. Kaplan, Phys. Rev. Letters 21, 756 (1968).

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

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

[10]R. Kaplan, Appl. Opt. 6, 685 (1967).

[11]R. A. Isaacson, Phys. Rev. 169, 312 (1968).

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

[13]S. H. Groves, C. R. Pidgeon, A. W. Ewald, and R. J. Wagner, J. Phys. Chem. Solids 31, 2031 (1970).

[14]B. D. McCombe, Solid State Commun. 6, 533 (1968).

[15]B. D. McCombe, R. J. Wagner, and G. A. Prinz, Solid State Commun. 7, 1381 (1969).

PHYSICAL REVIEW B VOLUME 4, NUMBER 4 15 AUGUST 1971

Low-Temperature Grüneisen Parameters for Silicon and Aluminum†

W. B. Gauster
Sandia Laboratories, Albuquerque, New Mexico 87115
(Received 29 March 1971)

From measurements of the inertial thermoelastic stress produced by pulse heating a portion of a sample, the Grüneisen parameters of silicon and aluminum as a function of temperature were obtained directly between 5 and 290 °K. Pulses of 1.5-MeV average-energy electrons, lasting approximately 40 nsec, were used as the heating agent, and the stresses were recorded with a quartz gauge bonded to the back face of the sample. For silicon, the low-temperature stress measurements indicated a limit of the Grüneisen parameter of about 0.2, which is lower than the value calculated from thermal-expansion data but in agreement with that calculated from pressure derivatives of elastic constants. For aluminum (6061 alloy-97% Al), the low-temperature limit of the lattice Grüneisen parameter was found to be 1.7 and the electronic contribution was estimated to be 1.7 as well, agreeing within experimental uncertainty (approximately 25%) with published values based on thermal-expansion measurements on pure aluminum.

I. INTRODUCTION

Recently, precise measurements of elastic-stress pulses resulting from the heating of solid samples by brief bursts of MeV electrons have been related to the Grüneisen parameters of the absorbing materials.[1–4] Absolute determinations of room-temperature Grüneisen parameters were accomplished by combining the elastic displacement measurements with electron-beam dosimetry.[3,4] Related free-carrier effects in semiconductors have been analyzed.[5]

The purpose of this paper is to report the measurement of thermoelastic stresses to determine