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EFFECTS OF ELECTRON-OPTICAL-PHONON INTERACTION IN THE COMBINED RESONANCE SPECTRA OF InSb

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The effects of electron–optical-phonon coupling on the combined resonance of both free and localized electrons have been studied in InSb. The results do not confirm the existence of a strong electron–TO-phonon interaction. Additional magneto-optical structure in the case of localized electrons appears to be due to discrete impurity excitations. The influence of conduction-band nonparabolicity on the impurity state binding energies has been observed.

Anomalies due to electron–optical-phonon coupling have recently been observed[1] in the combined resonance of electrons localized at impurities in InSb. The anomalies were interpreted as evidencing an electron–TO-phonon interaction comparable in strength to the electron–LO-phonon interaction which is responsible for the usual polaron effects. This result was somewhat surprising, since the analysis[2-6] of earlier observations of interband and electron cyclotron resonance absorption in InSb had excluded electron–TO-phonon coupling. Furthermore, there does not appear to be a satisfactory theoretical explanation for the coupling involving TO phonons.

The present Letter describes an experimental study of the effects of electron–optical-phonon coupling on the combined resonance of both free and localized electrons in InSb. Since the inclusion of the discrete "impurity" states of the localized electrons complicates the energy level structure, this structure is described initially in some detail. The methods used in obtaining and analyzing the data are then outlined. Results for the free-electron and localized-electron cases are presented separately, and are compared with those of earlier investigations.

The energy levels of interest in the present work are shown schematically in Fig. 1 for a magnetic field at which the cyclotron resonance energy is substantially larger than the impurity binding energy. For InSb, this situation obtains for fields greater than about 5 kG. The $N=0$ and $N=1$ free-carrier Landau levels are indicated by parabolas for effective spin vectors parallel ($\uparrow$) and antiparallel ($\downarrow$) to the magnetic field. Discrete impurity states of the electrons are identified by quantum-number sets ($lm\lambda$) in the manner of Wallis and Bowlden[7]; the Landau quantum number is given in terms of this set by $N=l+\frac{1}{2}(m+|m|)$. The solid arrows show the combined resonance transitions for free electrons $(N=0;\uparrow)\rightarrow(N=1;\downarrow)$, and localized electrons $(000;\uparrow)\rightarrow(010;\downarrow)$. Strong anomalies in combined resonance are expected for magnetic fields at which the optical phonon energy is comparable

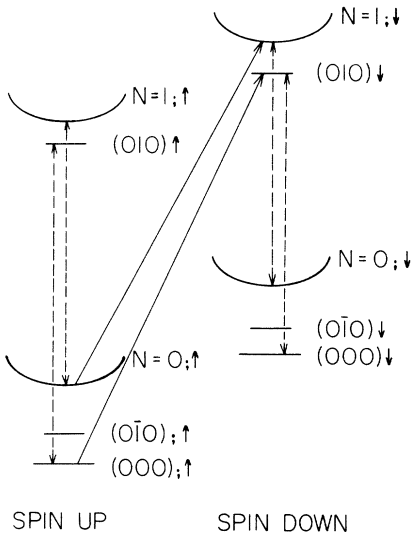

FIG. 1. Schematic representation of the energy levels of free and localized electrons in InSb in the high-field case. The lowest two Landau levels for both spin orientations are indicated by parabolas. Impurity states of importance in the present work have been included and are identified using the notation of Ref. 7. Combined resonance transitions are shown by solid arrows, while dashed arrows indicate spin-up and spin-down cyclotron resonance energies. The energy levels are not drawn to scale.

with the energy difference $E(N=1;\downarrow)-E(N=0;\downarrow)$ for free electrons, and $E(010;\downarrow)-E(000;\downarrow)$ for localized electrons. These energy differences, which correspond to the spin-down cyclotron eneriges of free and localized electrons, respectively, are indicates by dashed arrows in Fig. 1. The corresponding spin-up cyclotron energies are similarly indicated.

The transmission measurements were performed on two single-crystal samples of Se-doped InSb with excess donor concentrations of 8×10^{14} and 1.5×10^{15} cm^{-3}. The sample thicknesses were 2.5 and 1.7 mm, respectively. An interferometric spectrometer was used at photon energies between 10 and 60 meV, in conjunction with a superconducting solenoid capable of producing 58 kG. The solenoid field-current relation was obtained to 0.1% accuracy using NMR techniques. Samples were mounted in the Voigt geometry using wire-grid polarizers to obtain radiation whose electric field vector was parallel to the applied magnetic field. Liquid-helium-cooled Ge photoconductive detectors doped with Ga, Be, or Zn were used to cover the desired spectral region with optimum sensitivity. Sample temperatures in the range between 4.2 and 50°K could be maintained by appropriate choice of current in a sample heater coil and pressure in the helium-exchange gas cooling system. Temperature was monitored with a Ge thermometer in thermal contact with the sample holder. By this means the degree of carrier freezeout could be controlled, and magneto-optical transitions due to free and localized electrons could be observed separately or simultaneously.

It is of primary importance to obtain from the data the energy (or energies) at which strong interaction with the optical phonons occurs. The experimental measurements yield directly the magnetic field dependence of the combined resonance transition energies. If the $N=0$ spin-flip contributions to the latter are subtracted, the resulting spin-down cyclotron energies may be plotted versus magnetic field. When the cyclotron energy is nearly equal to the energy of an interacting optical phonon, two branches are expected in such a plot, one approaching the optical-phonon energy asymptotically from below and the other from above, with a discontinuity at the optical-phonon energy. More than one discontinuity may be observed[8] if additional phonons or discrete electronic states are involved in the coupling. In order for this procedure to give reliable results, the field-dependent spin-flip energy for the $N=0$ Landau level must be known accurately. Previous measurements[9] have provided this information for free carriers, including the field dependence of the spin-flip energy due to the conduction-band nonparabolicity. Corresponding information for localized electrons is not available, and must be deduced from the free electron results and knowledge of the $(100;\uparrow)$ and $(000;\downarrow)$ impurity-state binding energies.

Free-Electron Combined Resonance.—Free-electron combined resonance spectra were obtained for sample temperatures of about 30°K, which virtually eliminated the localized-electron transitions. In the spectral region of interest for these measurements, magnetic-field-independent absorption peaks at 34.2, 37.4, and 38.0 meV were observed. These correspond to multiphonon absorption peaks reported earlier.[10] The presence of these absorption lines increases the difficulty of obtaining accurate data in the thick samples necessary to observe the combined resonance transitions.

The free-electron combined resonance spectra showed two absorption peaks at fields for which the spin-down free-carrier cyclotron energy was comparable with the optical-phonon energy. As the field was increased, a gradual transfer of intensity occurred from the lower to the higher energy peak. The latter was considerably broadened in the strong-coupling region, but approached the low-field width at sufficiently high fields. The data analysis method described above has been followed in obtaining the plot of spin-down cyclotron energy versus magnetic field for free electrons, shown in Fig. 2. Both of the transition energy branches are observed to approach the energy value 23.9 ± 0.5 meV. The indicated uncertainty represents the maximum total error from all sources, the major contribution to the error arising in the determination of the spin-flip energy.

There is some disagreement in the literature concerning the energies assigned to the zone-center optical phonons in InSb, typical values in meV being $\hbar\omega_{LO}=24.45\pm0.25$, $\hbar\omega_{TO}=22.9\pm0.4$,[11] $\hbar\omega_{LO}=24.2$, $\hbar\omega_{TO}=22.6$,[12] $\hbar\omega_{LO}=24.4\pm0.1$.[13] The interaction energy observed in the present work is consistent with the values quoted for the LO but not TO phonons. This result, together with the absence of a second interaction energy, shows that electron–TO-phonon coupling certainly must be substantially weaker than electron–LO-phonon coupling in InSb. The observed magnetic field dependence of the absorption line-

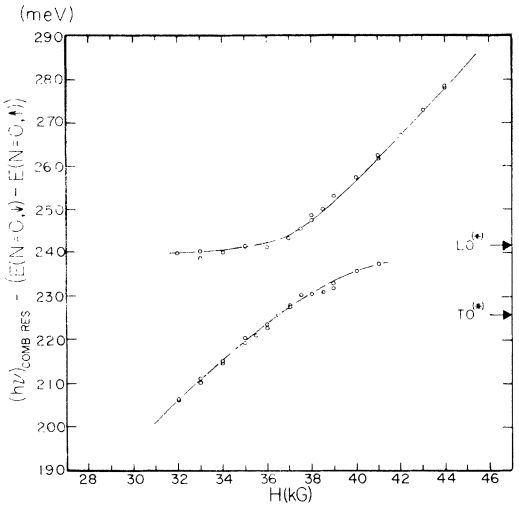

FIG. 2. Combined resonance minus spin-flip energy for free carriers versus magnetic field. Smooth curves have been drawn through the energy values. The symbol (*) identifies the LO and TO phonon energies according to Ref. 12.

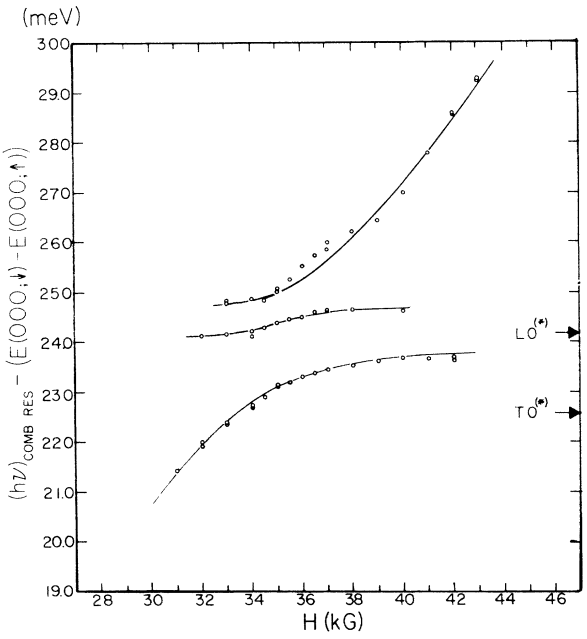

FIG. 3. Combined resonance minus spin-flip energy for localized electrons versus magnetic field. The spin-flip energy was estimated as described in the text. Values of the LO and TO phonon energies according to Ref. 12 are indicated by (*) in the figure.

widths, intensities, and transition energies is consistent with the theoretical picture of polaron anomalies in magneto-optical transitions.[2-6]

Localized-Electron Combined Resonance.—Localized-electron combined resonance spectra were obtained for sample temperatures between 4.5 and 6.5°K. In the strong-coupling region, these spectra exhibited three distinct absorption peaks whose relative intensities varied with field in a manner analogous to that described for the free-carrier case. The lowest two peaks remained sharp at all fields while the highest energy peak broadened considerably in the region of strong coupling.

Analysis of the localized-electron data is complicated by the lack of an experimental determination of the $N=0$ impurity spin-flip energy, $E(000;\downarrow)-E(000;\uparrow)$. The impurity spin-flip energy may be expected to differ from the corresponding free-carrier quantity, on account of the effects of the conduction-band nonparabolicity on the impurity-state binding energies.[14] An estimate of this difference may be obtained for the $N=0$ case by comparison with an experimental measurement of the corresponding difference for the $N=1$ case. Observation of the combined resonance and spin-up cyclotron resonance for free and localized electrons, at 32 kG, yields the following results: $\{E(010;\downarrow)-E(010;\uparrow)\}=0.981\times\{E(N=1;\downarrow)-E(N=1;\uparrow)\}$; $\{BE(010;\uparrow)-BE(010;\uparrow)\}=+0.136$ meV $=0.065BE(010;\downarrow)$. This may be compared with the results of a simple calculation which assumes that the binding energy varies linearly with mass, and uses the theoretical[15] variation of mass with energy, and the conduction-band energies in a magnetic field.[16] The calculation yields a fractional difference in the binding energy of 0.0392, a factor 1/1.66 smaller than the experimental result of 0.065.

Lacking a direct measure of the $N=0$ impurity spin-flip energy, the following ad hoc approach has been adopted: The fractional difference in binding energies of the $(000;\uparrow)$ and $(000;\downarrow)$ states has been calculated as described above and multiplied by the factor 1.66. From this result the impurity spin-flip energy is obtained as $0.956\times\{E(N=0;\downarrow)-E(N=0;\uparrow)\}$ in the strong-interaction region. Over this region the ratio of impurity to free-electron spin-flip energy is virtually field independent. The localized-electron combined resonance data have thus been analyzed by subtracting from the observed transition energies 0.956 of the measured $N=0$ free-carrier spin-flip energy, which varied from 8.99 meV at 32 kG to 11.50 meV at 43 kG. Figure 3 shows the resulting plot versus magnetic field. The lowest discontinuity occurs at 24 meV, in good agreement with the result obtained for free carriers.

The possibility that electron–TO-phonon coupling may be responsible[1] for the lowest energy discontinuity in the combined resonance of localized electrons may be discounted for the following reasons. First, even assuming that the free- and localized-electron spin-flip energies are equal, the lowest energy discontinuity occurs at 23.5 meV. This is already higher than the published values[11,12] for $\hbar\omega_{TO}$. Allowance for the effects of nonparabolicity further raises this value, placing the discontinuity at 23.7 meV if the measured correction for the $N=1$ levels is applied to the $N=0$ case. In fact, the effects of nonparabolicity are greater in the latter case, and the preceeding analysis places the lowest energy discontinuity at 24 meV. This value, as in the free-carrier case, is consistent with the energy of LO, but not TO, phonons. Second, while the absolute energy values obtained by the analytical procedure used must be viewed with some skepticism, the separation between the energy discontinuities is unaffected by this procedure. The measured separation lies in the range 0.80–0.85 meV. This is substantially smaller than the value $\hbar(\omega_{LO}-\omega_{TO}) \approx 1.55$ meV found in the literature.[11,12]

An explanation of the localized-electron combined resonance results that is consistent with all aspects of the data may be given in terms of phonon-assisted impurity excitations.[8] Figure 2 of Ref. 8, which shows the transition energies versus magnetic field for spin-up localized-electron cyclotron resonance, bears a marked resemblance to Fig. 3 of the present work. The transitions of interest in Ref. 8 occur at photon energies equal to the sum of $\hbar\omega_{LO}$ and the energy required to excite the impurity from its ground state to a discrete excited state. Excitations to the $(0\bar{1}0;\downarrow)$ level would then be responsible for the second discontinuity in Fig. 3. In both the present case and that described in Ref. 8, the lowest two absorption peaks in the strong-coupling region were narrow, while higher lying peaks were broadened. The broadening was probably responsible for the failure to resolve the splitting in the combined resonance spectra at the photon energy corresponding to excitation to the $(001;\downarrow)$ level. In fact, at this energy (~26.5 meV), a broad kink is present in the upper branch of Fig. 3.

The present experiments provide strong evidence against the existence in InSb of an electron–TO-phonon interaction of strength comparable with the electron–LO-phonon interaction. A consistent explanation of the features of the combined resonance spectra in the strong-coupling region has been given in terms of electron–LO-phonon coupling alone for both free and localized electrons. In the latter case the coupling involves excitations to discrete impurity states. The influence of conduction-band nonparabolicity on the binding energies of impurity states has been observed and taken into account in the analysis of the spectra.

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