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IMPURITY-STATE–OPTICAL-PHONON COUPLING IN A MAGNETIC FIELD IN InSb

R. Kaplan and R. F. Wallis
Naval Research Laboratory, Washington, D. C. 20390
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A number of peaks in photoconductivity have been observed in n-type InSb when the impurity-shifted cyclotron resonance frequency is near the longitudinal optical phonon frequency. Certain of these peaks are interpreted as due to photon absorption resulting in the creation of an LO phonon and simultaneous excitation of the electronic impurity state. We present a theory based on the Fröhlich-type electron-phonon interaction.

Interactions between optical phonons and free or bound charge carriers are responsible for a number of optical effects in semiconductors. One such effect is the phonon-assisted excitation of charge carriers bound to impurity atoms. In this process, photon absorption leads to the creation of one or more optical phonons, with the simultaneous excitation of the charge carrier to a discrete impurity state. Transitions of this type have been reported previously for the II-VI polar semiconductor ZnTe,[1,2] and for diamond.[3] In both of these cases the impurity binding energy was considerably larger than the optical phonon energy, and the transitions were observed in the absence of a magnetic field.

We wish to report the observation of phonon-assisted donor impurity excitations in InSb, a III-V semiconductor in which the impurity binding energy is nearly an order of magnitude smaller than the optical phonon energies. The excitations could be observed only in the presence of a magnetic field of suitable magnitude. The observed magnetic field dependence of the transition energies provides information pertinent to the problem of polaron coupling in InSb.[4-6]

The spectral dependence of the photoconductivity of relatively pure n-type InSb near 4.2°K has been measured at photon energies between 4 and 45 meV, in magnetic fields up to 55 kG. Previous work[7] has shown that in these circumstances the spectral response of the photoconductivity is identical to that of the absorption constant, within experimental error. Most of the experiments were done on two samples, of thickness 30 and 100 μ, with net donor concentration of approximately 7×10^{13} cm^{-3}. Spectra were obtained using an interferometric spectrometer in conjunction with a superconducting solenoid and light-pipe optics; the apparatus and techniques have been described elsewhere.[7] Apart from overall signal amplitude, no dependence of the photoconductivity on the magnitude of the biasing electric field was observed, for the electric fields used. No magnetic-field-dependent structure in the magnetoresistance of the samples was observed when the latter were shielded from infrared radiation by cold filters.

For magnetic fields smaller than about 25 kG, the photoconductivity spectra contained two sharp peaks, due to cyclotron resonance absorption by free electrons and electrons bound to impurity atoms. As the field was increased to bring the cyclotron resonance and optical phonon energies into proximity, the photoconductivity spectra became increasingly more complex, as shown in Fig. 1. At sufficiently high fields, the spectra again contained only the two cyclotron resonance peaks. By cooling the samples to about 1.5°K and obtaining spectra at small field intervals, it was possible to exclude signal peaks due to free carriers and to plot the energies of the transitions of the bound electrons versus magnetic field. A composite plot for the two samples, each measured using light propagating either parallel or perpendicular to the magnetic field, is shown in Fig. 2. Lines have been drawn through the data points, showing the field dependence of the positions of individual photoconductivity peaks. Peak

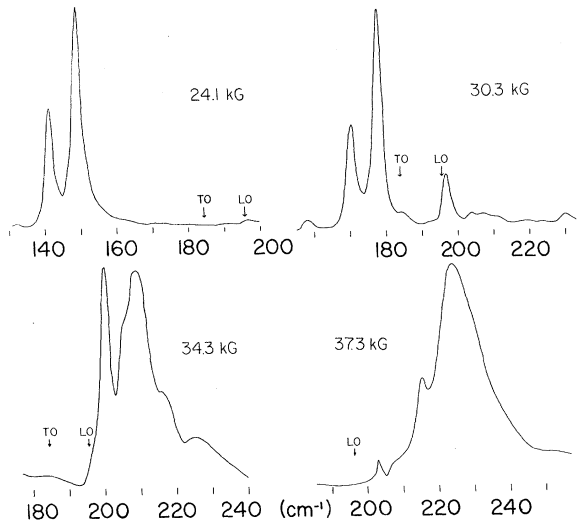

FIG. 1. Typical photoconductivity spectra obtained at the magnetic fields indicated. The spectra at 24.1 and 30.3 kG show the usual cyclotron resonance doublet at 4.2°K due to both free and bound electrons. The two higher-field spectra were obtained at 1.5°K. Arrows identified by the letters TO and LO show the energies of transverse and longitudinal optical phonons, respectively.

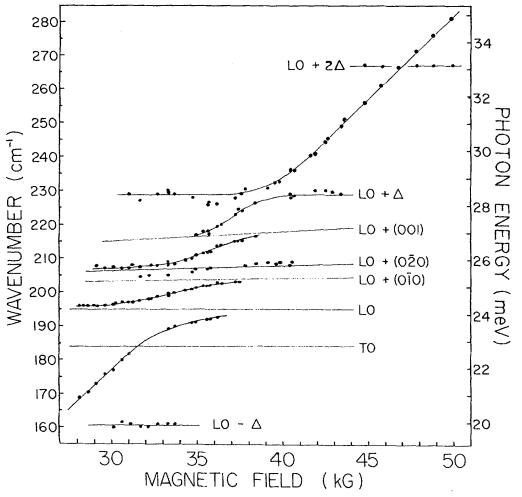

FIG. 2. Magnetic field dependence of the photon energies corresponding to the observed photoconductivity peaks. Smooth curves have been drawn through the data points corresponding to particular peaks. The lines labeled LO + (0$\bar{1}$0) and LO + (001) show the sum of the LO energy and the impurity state energy as determined by far-infrared measurements. The (0$\bar{2}$0) energy was obtained from impurity-state calculations, as described in the text.

positions did not depend on sample thickness or direction of light propagation. However, there was some dependence on these quantities of the relative intensities of the various peaks.

The photoconductive peaks were found to be strongest when their transition energies matched that of the bound-electron, or "impurity," cyclotron resonance. At magnetic field strengths for which this matching occurred, the field dependence of the transition energies of the peaks was a maximum. At higher or lower magnetic fields, the intensities decreased and finally vanished. The photoconductivity also vanished at photon energies in the immediate vicinity of the TO phonon energy.

The widths of the photoconductive peaks were found to vary markedly among themselves, and to depend strongly on magnetic field, as is evident from Fig. 1. In general, in the region between 25 and 29 meV, the peaks were broadest when their positions were most dependent on field. Above 29 meV the impurity cyclotron-resonance linewidth gradually decreased to its low-field value.

The three peaks identified in Fig. 2 as $\pm\Delta$ and $+2\Delta$ appear to belong to a system separate from the others. This is indicated by several features of the data, among which is the fact that, to within the limit of experimental error, the energy separation of these peaks from the LO phonon is an integral multiple of $\Delta = 4.34$ meV. The value of Δ is close to the energy of TA(L) phonons, 4.2 meV,[8] but this relationship may be fortuitous. The three peaks in question will not be considered further in the present work.

We now present a simple theoretical interpretation based on second-order perturbation theory. Such a theory should be valid not too close to the regions of sharp bending in Fig. 2. Let the Hamiltonian be

$$H = H_0 + H_I + H_{EP}, \tag{1}$$

where H_0 is the unperturbed Hamiltonian, H_I is the interaction of the electron with the radiation field, and H_{EP} is the electron-phonon interaction. The eigenstates of H_0 are taken to be those of an impurity electron in a high magnetic field as given by Wallis and Bowlden.[9]

Using semiclassical time-dependent perturbation theory to second order, one can write the absorption coefficient η associated with transitions from an initial state i to a final state k in the form

$$\eta = (4\pi^2\omega/nc\hbar V)\,|\,\vec{n}_0 \cdot \vec{M}_{ki}(\omega)\,|^2 \delta(\omega_i - \omega_k + \omega), \tag{2}$$

where ω is the radiation frequency, $E_i = \hbar\omega_i$ and $E_k = \hbar\omega_k$ are the energies of initial and final states, $\vec{n}_0$ is a unit vector in the direction of the electric vector of the radiation,

$$\vec{M}_{ki}(\omega) = \sum_j \left[\frac{\langle k|H_{EP}|j\rangle \vec{c}_{ji}}{E_i - E_j + \hbar\omega} + \frac{\vec{c}_{kj}\langle j|H_{EP}|i\rangle}{E_i - E_j} \right],$$

$$\vec{c}_{ji} = -\frac{e(E_j - E_i)}{\hbar\omega} \int \psi_j^* \vec{r} \psi_i \, d\tau, \tag{4}$$

ψ_j and ψ_i are the effective mass modulating functions for states j and i, V is the volume, and n is the refractive index.

For the electron-phonon interaction we use the Fröhlich Hamiltonian

$$H_{EP} = \sum_{\vec{q}} \hbar\omega_{\vec{q}} \left(\frac{4\pi\alpha}{S} \right)^{1/2} \frac{1}{r_0 q} \left(e^{-i\vec{q}\cdot\vec{r}} b_{\vec{q}}^{\dagger} + \text{H.c.} \right),$$

where $S = V/r_0^3$, $r_0 = (\hbar/2m\omega_{\vec{q}})^{1/2}$, m is the band mass of the electron, $b_{\vec{q}}^{\dagger}$ is the creation operator for longitudinal optical phonons of wave vector $\vec{q}$, and α is the polaron coupling constant. We shall restrict ourselves to the situation in which the initial state always involves the phonon ground state. It is assumed that the longitudinal optical phonons all have the same frequency ω_{LO}.

The matrix elements of H_{EP} involve integrals of the form

$$I = \int \psi_{lm\lambda}^* e^{-i\vec{q}\cdot\vec{r}} \psi_{l'm'\lambda'} \, d\tau, \tag{6}$$

where $lm\lambda$ are the quantum numbers used by Wallis and Bowlden in specifying the electronic impurity states. We have evaluated the integrals in Eq. (6) in closed form for a number of electronic states. These results were then used with Eq. (2) and a summation over wave vectors $\vec{q}$ to give final expressions for the absorption coefficient of the phonon-assisted transitions. Use was made of the experimental fact that $\omega \approx \omega_{ICR}$, the impurity-shifted cyclotron frequency, to restrict the intermediate states to those satisfying a near-resonant condition.

Physically, our results correspond to the excitation of the impurity electron by the electromagnetic field to an intermediate state. The electron-phonon interaction then causes a transition to the final electronic state together with the emission of a longitudinal optical phonon. Energy conservation requires that

$$\hbar\omega = \hbar\omega_{LO} + E_f^{(e)} - E_i^{(e)}, \tag{7}$$

where $E_i^{(e)}$ and $E_f^{(e)}$ are the initial and final electronic state energies. Since for the low-lying excited electronic states, the condition $\hbar\omega_{LO} \gg E_f^{(e)} - E_i^{(e)}$ can be satisfied, the theory predicts a number of subsidiary peaks in the vicinity of ω_{LO} as seen experimentally. If some low-lying excited impurity states are thermally populated, they can act as initial states and one can have $E_f^{(e)} - E_i^{(e)} < 0$. This leads to subsidiary peaks with frequencies below ω_{LO} rather than above ω_{LO} as is the case when the initial state is the ground electronic state. Indications of one or more peaks corresponding to transitions for which the initial state is an excited impurity state are present in the data. However, the peaks were extremely weak, and were not included in Fig. 2.

To get an idea of the intensity of these peaks, let us consider the case where the initial electronic state is (000), the intermediate state is (010), and the final state is $(0\bar{1}0)$. The ratio of the integrated intensity of the subsidiary peak to that of the impurity-shifted cyclotron resonance peak is given approximately by

$$R = \frac{\alpha}{8\sqrt{\pi}} \left(\frac{\hbar\omega_L}{E_i - E_j + \hbar\omega} \right)^2 \left(\frac{r_0}{a_0} \right) \gamma^{1/2} \epsilon, \tag{8}$$

where ϵ is the variational parameter for the states (010) and $(0\bar{1}0)$, γ is the ratio of one-half the Landau level spacing to the effective Rydberg, and a_0 is the effective Bohr radius. For InSb and typical experimental conditions, $\alpha = 0.02$, $\gamma = 14$, and $\epsilon = 0.4$. Taking $E_i - E_j + \hbar\omega = E_{000}^{(e)} - E_{010}^{(e)} + \hbar\omega = 0.1\hbar\omega_{LO}$, we find from Eq. (8) that $R \approx 0.04$. For final states $(0\bar{2}0)$ and (001), the values of R are about the same as that for $(0\bar{1}0)$ at the same value of γ. It is difficult to make a precise comparison of these results with the experimental photoconductivity data, but the order of magnitude seems reasonable.

The far-infrared photoconductive response of the samples used in the present experiments (Kaplan[10] and subsequent unpublished work) comprised two strong peaks, whose energies increased slowly with increasing magnetic field. These peaks correspond[9] to direct excitation of the bound electrons to the p-like $(0\bar{1}0)$ and (001) excited states. The lines in Fig. 2 labeled LO + $(0\bar{1}0)$ and LO (001) were obtained by adding the

LO energy, 24.2 meV, to the excited-state energies as obtained by the far-infrared experiments. Since the latter did not clearly determine the $(0\bar{2}0)$ energy, the line in Fig. 2 corresponding to the energy LO + $(0\bar{2}0)$ was drawn with the aid of the impurity-state calculations.[9] Agreement with the data of Fig. 2 appears satisfactory, particularly with regard to the characteristic field dependence of the excited state energies. The highest-field spectrum of Fig. 1 clearly shows the excitations for a magnetic field value comparable with that assumed in the calculations. The photoconductivity peak corresponding to the $(0\bar{2}0)$ excitation is relatively weak and broad, and may include excitations to the nearby $(0\bar{m}0)$ states having $m > 2$.

The results of the present experiments may be understood on the basis of electron–LO-phonon interactions alone. The existence of an interaction involving TO phonons[6] would result in appreciable bending of the impurity cyclotron resonance mode in the neighborhood of the TO phonon energy. An effect of this nature is absent from the data shown in Fig. 2. This observation is strengthened if the slight apparent displacement of the observed peaks due to the onset of strong lattice absorption and reflection near $\hbar\omega_{TO}$ is taken into account.

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DIFFUSION OF QUASIPARTICLES IN SUPERCONDUCTING ALUMINUM FILMS

S. Y. Hsieh* and James L. Levine
IBM Watson Laboratory, Columbia University, New York, New York
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The diffusion of quasiparticle excitations in superconducting aluminum films has been observed. The measured diffusion constant $D = 22.5$ cm^2 sec^{-1} is in good agreement with an estimate based on the normal-state resistivity of the films.

We have observed the diffusion of quasiparticle excitations (unpaired electrons) in a superconducting aluminum film. The diffusion constant was obtained from a direct measurement and found to be in excellent agreement with an estimate based on the normal-state resistivity of the film.

The measurements were made using the arrangement of tunnel junctions shown schematically in Fig. 1(a). Quasiparticles are created at a steady rate at one of the aluminum/aluminum-oxide/indium tunnel junctions,[1] diffuse along the aluminum strip, and are detected at junction 6. Because the excitations have a finite τ_γ against recombination with thermally excited quasiparticles,[2] the number reaching the detector decreases with distance from the generator junction as $\exp(-x/\delta)$. Here the diffusion length δ is given by $\delta^2 = D\tau_\gamma$, where D is the diffusion constant, and we have assumed one-dimensional diffusion because of the geometry. The diffusion constant is then determined by measuring the detector response for quasiparticle generation at junctions 1-5 in turn, τ_γ being known from other experiments.[3]

The quasiparticle generation and detection processes have been discussed elsewhere[3,4] and will be described only briefly here. Generation occurs when a junction is biased to a voltage slightly in excess of $(\Delta_{In} + \Delta_{Al})/e$, where Δ_{In} and Δ_{Al} are the energy gaps in the two materials. The rate is proportional to the junction current and independent of the oxide thickness and junction area. Detection occurs at biases below $(\Delta_{In} + \Delta_{Al})/e$, the current flowing in this voltage region being proportional to the quasiparticle den-