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XI-1. Polaron Induced Anomalies in InSb

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We have found striking anomalies in the interband magneto-absorption of InSb which constitute an unambiguous observation of polaron self-energy effects. Examining the magnetic field dependence of the infrared absorption associated with interband transitions to the $n=1$ conduction band Landau levels, we find that the photon energy at the absorption maxima and absorption intensity behave in a manner quite different from that expected on the basis of a rigid lattice model.

Theoretically, polaron effects are expected to be strongly enhanced when the cyclotron energy approaches the energy of a LO phonon. Analysis indicates that enhanced polaron effects are responsible for the observed anomalies.

§ 1. Introduction

Although there exists an extensive literature on large polarons, it has not been demonstrated until very recently that polarons actually exist in real crystals. Certain electrical conductance phenomena observed in III-V tunnel diodes have been attributed to polarons,[1] however, this interpretation now appears to be incorrect.[2] Measurements of the cyclotron mass of carriers in AgBr and KBr[3] have been used to evaluate the accuracy of polaron mobility theories, but they do not give direct or entirely convincing evidence for the existence of polarons.

Recently we have examined in detail the magnetic field dependence of the interband infrared absorption associated with transitions to the $n=1$ conduction band Landau levels in InSb.[4] The purpose of this investigation was to look for large changes in the polaron self-energy in magnetic fields strong enough to cause the energy difference between the $n=1$ and $n=0$ rigid lattice Landau levels to equal $\hbar\omega$, the energy of an LO phonon. The large effects observed had been expected on the basis of the theory of the Fröhlich polaron.

§ 2. Theory

To understand the predicted effects we consider the Hamiltonian (due to Fröhlich) describing the interaction of a conduction band electron with an external magnetic field and the LO phonons. By the methods of reference *I*[5] it can be shown that this Hamiltonian in units of $\hbar\omega$ can be written:

$$\mathscr{H}=\mathscr{H}_1+\mathscr{H}_2+\mathscr{H}_3 \tag{1a}$$

$$\mathscr{H}_1=p_y^2+\frac{1}{4}\lambda^4 y^2+\sum n_k+\sum k_z l_z n_k n_l + \sum k_x l_x n_k n_l \tag{1b}$$

$$\mathscr{H}_2=-\lambda^2 y \sum k_x n_k \tag{1c}$$

$$\mathscr{H}_3=(4\pi\alpha/S)^{1/2}\sum\frac{1}{k}(e^{-ik_y y}b_k^{+}+\text{H.c.}) \tag{1d}$$

where λ^2 is the dimensionless magnetic field defined by the ratio of the unperturbed cyclotron frequency to the optical phonon frequency ($\lambda^2=eH/mc\omega$), p_y and y are y-momentum and y-displacement operators in units of $\hbar/r_0$ and r_0, respectively, b_k^{+} is the creation operator for an LO phonon of wavevector $\boldsymbol{k}$, $n_k=b_k^{+}b_k$, S is the crystal volume in units of r_0^3, $\alpha=(1/2)(e^2/r_0)(1/\hbar\omega)(1/\varepsilon_\infty-1/\varepsilon)$ and $r_0=(\hbar/2m\omega)^{1/2}$. We have set p_z, the total z-momentum of the electron-phonon system, equal to zero. We assume throughout that $\alpha\ll 1$.

If we treat $\mathscr{H}_3$ as a perturbation on eigenstates of $\mathscr{H}_1+\mathscr{H}_2$ we obtain the energy of the $n=1$, $p_z=0$ Landau level formally to order α. In Wigner-Brillouin perturbation theory (WBPT) this is

$$E_{WB}(\lambda^2)-\frac{3}{2}\lambda^2=-\frac{\alpha}{2\pi^2}\mathscr{P}\int d^3k\frac{\exp(-k_\perp^2/\lambda^2)}{k^2} \times\sum_{n=0}^{\infty}\frac{1}{n!}\left(\frac{k_\perp^2}{\lambda^2}\right)^{n-1}\frac{(n-(k_\perp^2/\lambda^2))^2}{1+(1/2)\lambda^2-E_{WB}+n\lambda^2+k_z^2}, \tag{2}$$

where $k_\perp^2=k_x^2+k_y^2$ and $\mathscr{P}$ denotes the Cauchy principal value.

While it can be shown that (2) is not truly correct to order α when $1-0(\alpha)<\lambda^2<1+0(\alpha)$, the behavior of solutions of (2) provide the key

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to a qualitative understanding of some of the polaron induced anomalies in InSb.

First we note that as E_{WB} approaches $1+(1/2)\lambda^2$ from below, the right hand side of (2) diverges to $-\infty$ because of the divergence of the $n=0$ term in the sum. This means that as $\lambda^2\to\infty$, E_{WB} approaches $1+(1/2)\lambda^2$ from below. We describe this situation by saying that in high magnetic field the perturbed $n=1$ eigenvalue is pinned or clamped to the unperturbed ground state energy plus the energy of an LO phonon. Second, for $\lambda^2\lesssim 1$ the solution of (2) is double-valued. In addition to the branch of E_{WB} which remains always less than $1+(1/2)\lambda^2$, there is a branch on which E_{WB}, when it exists, is always greater than or equal to $1+(1/2)\lambda^2$. We have plotted in Fig. 1 the two branches of the polaron energy spectrum derived from a better theory than WBPT, however the spectrum in Fig. 1 may also be considered to be a qualitative plot of E_{WB}.

A theory correct to order α for the lower branch of the polaron spectrum can be developed by a generalization of the method previously described for the free polaron.[6]

Any eigenfunction of $\mathscr{H}$ can be expanded in eigenfunctions of $\mathscr{H}_1+\mathscr{H}_2$; these expansion functions are simply products of an electronic displaced oscillator function $\phi_n(y-2\sum k_x n_k/\lambda^2)$ with eigenvalue $(n+1/2)\lambda^2$ and a phonon eigenfunction of $\sum n_k$ and $\sum \boldsymbol{k} n_k$. We denote the phonon states by

$$b_k^+|0>=|\boldsymbol{k}>,\quad b_k^+ b_l^+|0>=|\ \boldsymbol{k},\boldsymbol{l}>,\quad \text{etc.}$$

where $|0>$ is the phonon vacuum state.

It can be shown that to obtain the energy to order α, it is sufficient to take for the $n=1$ eigenfunction of $\mathscr{H}$

$$\Psi=c\phi_1(y)|0>+\sum_{n,k} c_n(\boldsymbol{k})\phi_n(y-2k_x/\lambda^2)|\boldsymbol{k}> + \sum_{n,k,l} d_n(\boldsymbol{k},\boldsymbol{l})\phi_n(y-2(k_x+l_x)/\lambda^2)|\boldsymbol{k},\boldsymbol{l}> . \quad (3)$$

Components Ψ containing three or more phonons contribute to higher order than α in the energy, while terms of the form $\phi_m|0>$, $m\neq 1$, do not appear because, as can be verified a' posteriori, $\mathscr{H}$ does not connect any term on the right-hand side of (3) to $\phi_m|0>$ if $m\neq 1$.

We require $\mathscr{H}\Psi=E\Psi$ and operate with $\mathscr{H}$ on the right hand side of (3). This gives the following equations for the coefficients.

$$\left(E-\frac{3}{2}\lambda^2\right)c=\sum \nu_k c_n(\boldsymbol{k})M_{1n}(\boldsymbol{k}_\perp) \quad (4a)$$

$$D_n c_n(\boldsymbol{k})=c\nu_k M_{1n}^*+2\sum_{l,n'}\nu_l d_{n'}(\boldsymbol{k},\boldsymbol{l}) \times\exp(+2ik_x l_y/\lambda^2)M_{nn'}(\boldsymbol{l}_\perp) \quad (4b)$$

$$\mathscr{D}_n d_n(\boldsymbol{k},\boldsymbol{l})=(1/2)\nu_l\sum_m c_m(\boldsymbol{k})M_{mn}^*(\boldsymbol{l}_\perp) \exp(-2ik_x l_y/\lambda^2) +(1/2)\nu_k\sum_m c_m(\boldsymbol{l})M_{mn}^*(\boldsymbol{k}_\perp) \exp(-2ik_y l_x/\lambda^2) \quad (4c)$$

where

$$M_{mn}(\boldsymbol{k}_\perp)=\int_{-\infty}^{\infty} dy\phi_m^*(y)e^{ik_y y}\phi_n(y-2k_x/\lambda^2) =\kappa^{m-n}(\boldsymbol{k})\exp(ik_x k_y/\lambda^2)f_{nm}(k_\perp^2)$$

$$D_n=(E-(n+1/2)^2-(1+k_z^2))$$

$$\mathscr{D}_n=(E-(n+1/2)\lambda^2-2-(k_z+l_z)^2)$$

$$\nu_k=(4\pi\alpha/S)^{1/2}1/k$$

$$\kappa(\boldsymbol{k})=k_x+ik_y$$

and we have taken $d_m(\boldsymbol{k},\boldsymbol{l})$ symmetric in $\boldsymbol{k}$ and $\boldsymbol{l}$ for all m. When E is close to $1+(1/2)\lambda^2$, D_0 becomes very small for small k_z. Under these circumstances it is not self-consistent to neglect the term $\sum_l \nu_l d_0(\boldsymbol{k},\boldsymbol{l})$ on the right-hand side of the equation for $c_0(\boldsymbol{k})$. Evaluating $d_0(\boldsymbol{k},\boldsymbol{l})$ for small k_z to lowest order in α, we find that the effect is to change (4b) to

$$(E-E_0-1-k_z^2)c_0(\boldsymbol{k})=c\nu_k M_{10}^* \quad (5)$$

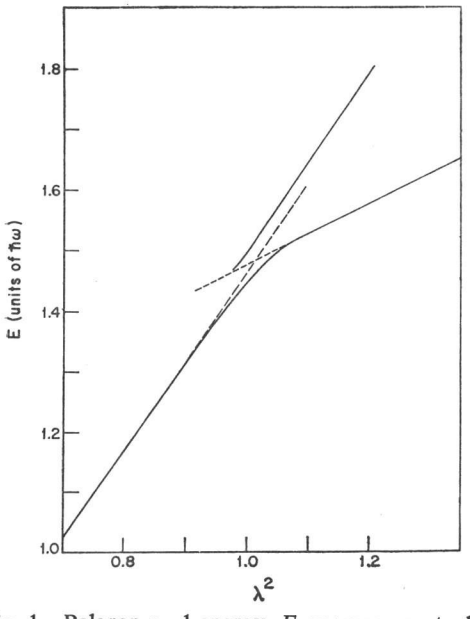

Fig. 1. Polaron $n=1$ energy, E, versus unperturbed cyclotron frequency, λ^2, from a parabolic band with $\alpha=0.02$. E is measured from the bottom of the conduction band in the absence of the magnetic field and the electron-phonon interaction.

where E_0 is the ground state energy of the polaron as computed in WBPT. It is self-consistent to neglect all the sums in (4c) when (5) holds, namely, when $E-E_0-1\geq 0(\alpha)$.

Solving (4a), (5) and (4c) we obtain, to order α,

$$E-(3/2)\lambda^2 = \frac{S}{(2\pi)^3}\mathscr{P}\int d^3l \sum_{n'} \frac{\nu_l{}^2|M_{n'1}(\boldsymbol{l}_\perp)|^2}{[E-E_0-(1+n\lambda^2+l_z{}^2)]} \quad (6)$$

which is identical to (2) except that the unperturbed ground state energy, $(1/2)\lambda^2$, has been replaced by the perturbed ground state energy E_0. We have inserted the principal part symbol in (6) to cover the case in which $E-E_0-1>0$ and $E-E_0-1\geq 0(\alpha)$.

The upper branch plotted in Fig. 1 corresponds to solutions of (6) for $E>E_0+1$. We remark that for $\alpha=.02$ (as estimated for InSb) there are no solutions on the upper branch for λ^2 below about .98. The lower branch in Fig. 1 shows the variation of E with λ^2 as computed from (6) for $E<E_0+1$.

The intensity of the absorption to the lower branch is proportional to $|c|^2$ since c is the coefficient of the zero-phonon component of Ψ. By requiring that Ψ be normalized, and solving for the other coefficients in terms of c from (4a), (4c) and (5), one obtains an equation for c. The results show that as $(E-E_0)\to 1$, $\sum_k |c_0(\boldsymbol{k})|^2$ grows rapidly at the expense of $|c|^2$, with the result that the intensity of optical transitions to the eigenstate corresponding to the lower branch should decrease rapidly as E becomes pinned.

§ 3. Experiment

We have performed interband magnetoabsorption experiments in InSb in order to observe the predicted effects. Absorption of infrared radiation by a sample 8μ thick at about 20°K was measured as a function of photon energy and magnetic field. The magnetic field, H, was perpendicular to the propagation direction of the polarized radiation and parallel to both the ⟨110⟩ crystal direction and the radiation E vector. Transitions to the $m_s=\pm(1/2)$, $n=1$ conduction band Landau levels were studied in detail in the regions of field at which these levels were expected to become clamped to the corresponding $n=0$ levels. The transitions observed which have been associated with the band theory assignments of Roth *et al.*[7,9] are indicated in Fig. 2, where the predicted polaron splitting of the $n=1$ levels is indicated schematically.

In Fig. 3 we have plotted the photon energy of the absorption peaks as a function of H for the transitions to the $n=1$, $m_s=+(1/2)$ and to the $n=0$, $m_s=+(1/2)$ conduction band Landau levels. We find that the energy difference between the absorption peaks on the branches marked $B_1{}^+$ and Z_1 for $H\geq 36$ kG, when corrected for the small valence band splitting between the heavy hole $n=2$ and $n=3$ levels (see Fig. 2) is independent of field to within estimated experimental uncertainty (± 0.02 meV). We consider this to be experimental verification of the predicted pinning effect. The corrected splitting between $B_1{}^+$ and Z_1 is 23.55 meV ± 0.02 meV. The value 23.55 is to be compared with LO phonon energies quoted in the litera-

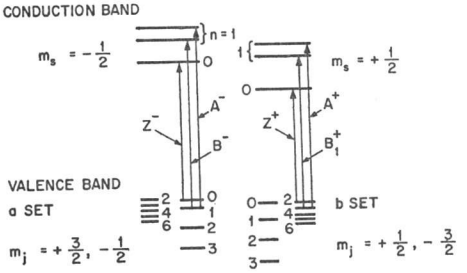

Fig. 2. Schematic energy diagram showing the energy levels for InSb in a magnetic field and the optical transitions observed in the experiments.

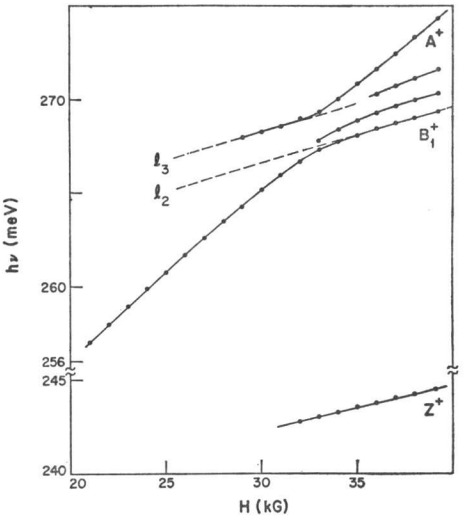

Fig. 3. Variation with magnetic field of the various transmission minima observed in the experiments. The lines l_2 and l_3 correspond to the pinning asymptotes.

ture [1,8] at from 22.8 to 24.4 meV.

The intensity of line B_1^+ decreases with increasing field near and in the region of pinning, in qualitative agreement with theory. Similarly the intensity of A^+ decreases with decreasing field in its region of pinning.

Several important differences between the observed and predicted behavior are evident from a comparison of Figs. 1 and 3. First, the A^+ line in Fig. 3 seems to pin at fields below 32 kG, whereas there is no comparable pinning in Fig. 1. Second, the line l_3, which A^+ approaches asymptotically, is not coincident with l_2 but closely parallel to it and displaced upwards by about 1.5 meV. Third, in addition to B_1^+, there are two other lines, labeled B_2^+ and B_3^+ (which we previously were unable to resolve[4]) exhibiting a pinning behavior. Finally, the $n=0$ absorption is not simple; it shows interesting structure (not shown in Fig. 3).

We do not yet understand these phenomena in detail, but we believe they may in part be related to exciton effects. It is known that in a strong magnetic field the presence of relatively weak Coulomb interaction between electron and hole will produce states in which the electron and hole are bound in their relative motion in the z direction.[10] This leads, roughly speaking, to a hydrogenic series of discrete energy levels whose limit is the unperturbed Landau level energy. Such discrete levels associated with the $n=1$ Landau level should be affected by those associated with the $n=0$ Landau level at appropriate magnetic fields in the presence of electron-LO phonon interaction. Further study, both theoretical and experimental, will be necessary before a complete accounting of the phenomena in Fig. 3 can be made.

We have also investigated the $n=1$, $m_s=-1/2$ conduction band level. Here our maximum magnetic field is barely sufficient to produce pinning of the lower branch. We have obtained a value of 23.3 meV for $\hbar\omega$ which we consider to be in satisfactory agreement with the corresponding result from the $m_s=+1/2$ level. A^- and B^- (see Fig. 2) are analogous in their intensity behavior to A^+ and B_1^+ respectively.

Finally, we have looked for effects associated with a hypothetical pinning of the $n=1$, $m_s=-1/2$ level to the $n=0$, $m_s=+1/2$ level due to an optical phonon interaction connecting these states of opposite spin. In our preliminary results we were unable to observe an effect clearly attributable to such an interaction. However, we feel that this problem warrants further investigation.

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COMMENT BY THE AUTHORS

In Phys. Rev. Letters **17** (1966) 436 White and Koonce have tried to give an improved theory of the results reported in ref. 4). Their theory consists, in essence, of omitting the k_z dependence in the denominators in eq. (2) and truncating the sum on n to include only the $n=0$ term. These approximations lead to spurious results for E_{WB} when compared to the exact solution of eq. (2). The fit of the White-Koonce theory to the experimental data

seems acceptable only in the field range of 30–38 kG and requires using a value for the band gap which is smaller than the measured value by approximately 10 meV. It seems strange that in the low-field region where polaron effects are known to be negligible the White-Koonce fit diverges from the experimental data.

DISCUSSION

Smith, S. D.: The existence of the polaron doublet has consequences on non-resonant free carrier dispersive effects such as Faraday rotation and Voigt effect. That this should be so can be seen from Becquerel relation $\theta = \omega\omega_c(dn/d\lambda)$. Calculation of the matrix elements V_+ and V_- for the polaron doublet by simple theory confirms this conclusion. We have observed such effects in InSb and PbTe at $\omega_{LO}=\omega_c$ and $\omega_{LO}=2\omega_c$ and it appears that remote-sensing dispersion methods may well be a useful technique for observing the clamping effect, avoiding as they do, the lattice absorption near ω_{LO}.

Larsen, D. M.: One would expect that the effects described for the $n=1$ level would occur for the $n=2$ level at half the magnetic field. We have not yet succeeded in observing this. Perhaps Dr. Smith has.