a degeneracy (or near degeneracy) between the unperturbed adsorbate level ϵ_a and the continuum of metal states ϵ_m. It is a general phenomenon; for example, antiresonances appear in inelastic electron-atom scattering and in atomic ultraviolet light adsorbtion. There, the interference arises from a degeneracy of a discreet two-electron excited atomic state with the continuum of single ionized atomic states. The strength of the antiresonance is directly related to the coupling between the discreet and continuum states, i.e., $V_{m,a}$ in the case of photoemission.

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[1]D. E. Eastman and J. K. Cashion, Phys. Rev. Lett. 27, 1520 (1971).

[2]G. Breit and H. A. Bethe, Phys. Rev. 93, 888 (1954).

[3]I. Adawi, Phys. Rev. 134, A788 (1964).

[4]D. Penn, R. Gomer, and M. H. Cohen, Phys. Rev. Lett. 27, 26 (1971).

[5]G. D. Mahan, Phys. Rev. B 2, 4334 (1970).

[6]Attributing a momentum dependence to λ in $e^{\lambda z}$ would have the effect of multiplying j_0 by a factor of the form $(1-P_c/P)$, in agreement with C. N. Berglund and W. E. Spicer, Phys. Rev. 136, A1030 (1964).

Magneto-optical Study of Hole-Optical Phonon Coupling in InSb

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The interactions of bound holes with TO and LO phonons in InSb have been observed in magneto-optical experiments. Theoretical estimates of the hole-phonon coupling strengths have been obtained.

Magneto-optical studies of polarons have yielded valuable information concerning the resonant electron-optical-phonon interaction in semiconductors.[1] The interaction typically is manifested as a splitting and broadening of a transition such as cyclotron resonance, when the electron excitation and optical-phonon energies are comparable. Previous studies of this type have been restricted to electron excitations in a number of n-type polar semiconductors. In the present work, resonant bound-hole–optical-phonon coupling in p-type InSb is reported. Interactions with both TO and LO phonons have been observed. The observations serve to illustrate several significant differences between the interactions of holes and of electrons with optical phonons.

Before presenting the experimental results it will be useful to discuss briefly two factors which differentiate hole coupling from electron coupling to optical phonons. Firstly, because of symmetry considerations,[2] the interaction of electrons with TO phonons is expected to vanish, whereas hole–TO-phonon coupling is allowed, as confirmed by optical[1,3] and tunneling[4] experiments. Secondly, in contrast to the case for electrons in materials previously studied, free holes occupy states in both light- and heavy-mass bands. This leads to a multiplicity of initial states for magneto-optical transitions, thus tremendously complicating the study of free-hole polaron effects. It is therefore desirable to perform experiments on bound holes which possess a unique state. Another consequence of the valence-band structure is that in the presence of a magnetic field, acceptor excited states exist in association both with the low-lying heavy-hole Landau levels, and with the more widely spaced light-hole levels. In subsequent discussions, the terms "low mass" and "high mass" will be used to distinguish between these two types of acceptor excited states. Transitions involving low-mass excited states in InSb and Ge were previously investigated in some detail.[5,6]

In the present experiments, the first few acceptor excitations to low-mass excited states have been studied in a spectral region encompassing the optical-phonon energies in InSb. Above a few kilo-oersteds the energies of these acceptor excitations vary approximately linearly with magnetic field. Structure in the absorption spectra due to hole-phonon coupling would first be expected when the transition energies approach the optical-phonon energies. However, this spectral region is inaccessible because of strong lattice absorption and reflection. At higher field strengths, the energy separations of the low-mass and high-mass excited states are comparable to the optical-phonon energies. It is this field region in which structure in the acceptor excitation spectra has been observed and studied.

InSb samples used in these experiments were

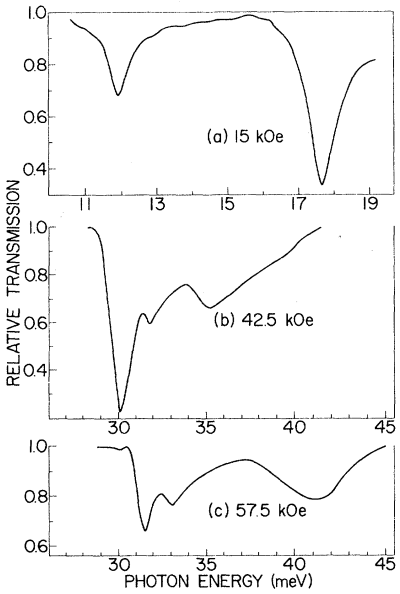

FIG. 1. Transmission spectra, at three magnetic field strengths, for an InSb sample containing 2×10^{15} cm^{-3} Cd acceptor impurities.

plates of thickness 0.5 to 5 mm, doped with Cd or Zn in the range 2×10^{14} to 5×10^{15} cm^{-3}. Carrier mobilities were approximately 10^4 cm^2/V sec at 80°K. The transmission of light through samples at 4.5°K, in the wave-number range 40–380 cm^{-1}, was studied at fixed magnetic fields up to 62.5 kOe. We employed Fourier-transform spectroscopic techniques utilizing a Michelson interferometer. Earlier measurements,[5] using grating optics and a Bitter-type magnet, extended to 700 cm^{-1} and 100 kOe, but did not include the spectral region encompassing the phonon interactions.

Typical transmission spectra are shown in Fig. 1 for three magnetic field strengths; the spectra are normalized to the transmission in zero field. Figure 1(a) shows the first two low-mass acceptor excitations in a field of 15 kOe. The final states for these excitations are bound to the $a^+(1)$ and $b^+(1)$ light-hole levels. (The notation is that of Ref. 7.) As the field was increased, the $a^+(1)$ transition shifted linearly to higher energy. However, the $b^+(1)$ transition was observed to split into three components, with the absorption strength gradually shifting to the upper component, as shown in Figs. 1(b) and 1(c). The widths of the two lower components were in the range 1–2 meV. The upper component when first observed was about 5 meV wide, but became somewhat narrower at high fields. Accurate determination

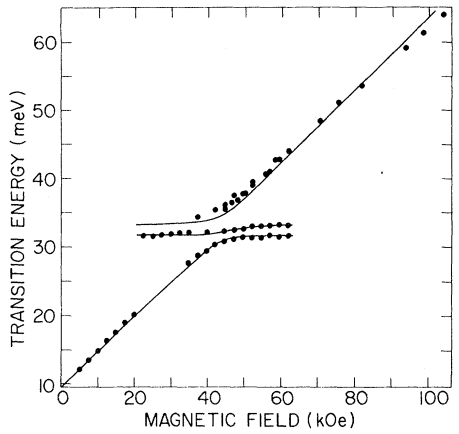

FIG. 2. Observed transition energies (data points) for the $a^+(1)$ and $b^+(1)$ acceptor excitations. Solid lines, a fit to the data using the model described in the text.

of linewidths and intensities was hindered by the strong overlap of the various components.

The field dependence of the $b^+(1)$ transition energies is shown in Fig. 2. Transitions to other low-mass excited states have been omitted for clarity. The three branches of the $b^+(1)$ transition define two field-independent "pinning energies" at 31.5 and 33.0 meV. As in other resonant carrier-phonon interactions, the transition components decreased in intensity as they asymptotically approached the pinning energies. However, the decrease was much slower than that expected for simple polaron coupling. At 60 kOe, or 1.5 times the interaction field, the integrated intensities of the asymptotic components were of order 0.1 that of the upper component.

The observed pinning energies of 31.5 and 33.0 meV do not correspond to any phonon or combination of phonon energies in InSb, and zero-field absorption at these energies was not observed in the spectra. Thus one or more additional states must be involved in the coupling to serve as final states for phonon emission from the $b^+(1)$ acceptor state. As is evident from Fig. 2, the pinning energies do not vary with magnetic field. Therefore, the field dependence of the final states for phonon emission must be similar to the field dependence of the high-mass acceptor ground state, i.e., very weak. The possibility that the observed splitting may be due in part to structure in the acceptor ground or $b^+(1)$ states may be eliminated for the following reasons. Such structure would of necessity have been present at other field values and in other transitions. It was not observed. Furthermore, the 1.5-meV splitting is much too large to be associated with structure in the ground

or $b^+(1)$ excited states. Thus it appears that the observed splitting is due either to structure in the final state for phonon emission, or to the participation of two different phonons in the coupling.

The participation of LO phonons is to be expected in either of the above cases. This conclusion follows from the strength of the observed electron–LO-phonon interaction, and the form of the Frohlich Hamiltonian for polaron coupling, which is independent of the type of charge carrier involved. The only other phonon in InSb whose energy is comparable to that of the LO is the TO phonon. By subtracting the optical-phonon energies from the observed pinning energies, the energies of the final states for phonon emission are obtained. All possible final-state energies determined in this manner lie below the continuum, and therefore represent high-mass acceptor excited states.

The effect of a magnetic field on the high-mass excited-state spectrum of acceptors in InSb has not previously been investigated. In zero magnetic field, two prominent high-mass excitation lines, at 7.24 and 7.82 meV, have been reported.[8] These lines have also been observed in the present work, and their behavior studied in a magnetic field. As the field strength was increased, a number of absorption components appeared. One of these, deriving from the 7.82-meV excitation, became clearly dominant as its energy approached that of the 2TA(L) multiphonon absorption line at 9.05 meV. Between 20 and 60 kG its energy increased from 8.1 to 8.9 meV. In this field range it was the only prominant feature in the high-mass excitation spectrum. It is then concluded that the high-mass excited state responsible for this dominant absorption line is the final state for the optical-phonon emission from $b^+(1)$. Details of its coupling to the 2TA(L) multiphonon mode will be given elsewhere. Electron-2TA(L) coupling in InSb has been previously observed in magnetophonon experiments.[9]

When the field-dependent energy of the dominant high-mass acceptor excitation is subtracted from the data of Fig. 2, the resulting pinning energies are 22.8 and 24.3 meV. These values are in excellent agreement with the energies of TO and LO phonons in InSb. Thus it is concluded that the observed structure in the $b^+(1)$ transition is due to the coupling of the $b^+(1)$ acceptor state with TO and LO phonons.

To substantiate this conclusion, a calculation of the $b^+(1)$ magnetoabsorption has been undertaken. Interactions of bound holes with both TO and LO phonons have been included. The acceptor states $|\alpha\rangle$ have wave functions of the form

$$\sum_{j=1}^{4} F_\alpha{}^j(\vec{r})\varphi_j(\vec{r}),$$

where φ_j are degenerate valence band edge functions, and $F_\alpha{}^j(\vec{r})$ are envelope functions. For the bound-hole–LO-phonon coupling the Frohlich interaction, diagonal in the φ_j representation, is used:

$$V_{\beta\alpha}{}^L(\vec{q}) \equiv V^L(\vec{q}) m_{\alpha\beta}{}^L(\vec{q}) = -i(\hbar\omega_0/q)(4\pi\alpha_0/\Omega)^{1/2}\sum_{j,j'}\langle F_\alpha{}^j(\vec{r})|e^{i\vec{q}\cdot\vec{r}}\delta_{jj'}|F_\beta{}^{j'}(\vec{r})\rangle. \quad (1)$$

The bound-hole–TO-phonon interaction is obtained by generalizing the theory of Bir and Pikus[10] to the presence of an external magnetic field. The deformation potential coupling for TO phonons of polarization ν is given by

$$V_{\beta\alpha}{}^{T\nu}(\vec{q}) \equiv V^T(\vec{q}) m_{\alpha\beta}{}^{T\nu} = i\left[\frac{m_{\mathrm{In}}+m_{\mathrm{Sb}}}{2(m_{\mathrm{In}}m_{\mathrm{Sb}})^{1/2}}\right]\left(\frac{\hbar}{\Omega\rho_0\omega_0 a_0{}^2}\right)^{1/2} d_0\sum_{j,j'}\langle F_\alpha{}^j(\vec{r})|e^{i\vec{q}\cdot\vec{r}}\mathfrak{D}(\vec{e}_\nu)|F_\beta{}^{j'}(\vec{r})\rangle, \quad (2)$$

where $\vec{e}_\nu$ is the polarization vector of one of the atoms whose masses are m_{In} and m_{Sb}, ρ_0 is the mass density of the crystal, a_0 the lattice constant, ω_0 the *Reststrahlen* frequency, d_0 the optical deformation potential for InSb, and $\mathfrak{D}(\vec{e}_\nu)$ a 4×4 matrix function of $\vec{e}_\nu$.[10] The methods developed by Ngai[11] were used to calculate the photon absorption cross section $\sigma(\omega)$ with the results

$$\sigma(\omega) = \{\omega/[1-\exp(-\hbar\omega/kT)]\}\,\mathrm{Im}R(\omega), \quad (3)$$

$$R(\omega+i0) = \sum_{\alpha,\beta}|\gamma_{\alpha,\beta}|^2 Q_{\beta\alpha}(\omega+i0) - \sum_\nu\sum_{i=T_\nu,L} U_{\vec{q}}{}^i(\omega+i0)\Big[\sum_{\alpha',\beta'}\gamma_{\alpha'\beta'}{}^* m_{\alpha'\beta'}{}^i Q_{\beta'\alpha'}(\omega+i0)\Big] \times\Big[\sum_{\alpha',\beta'}\gamma_{\alpha'\beta'} m_{\alpha'\beta'}{}^{i*} Q_{\beta'\alpha'}(\omega+i0)\Big]. \quad (4)$$

The notation is the same as used in Ref. 11, except that in the present case

$$\gamma_{\alpha\beta} = \langle\alpha|(e/c)\vec{v}\cdot\vec{A}_\omega|\beta\rangle, \quad U_{\vec{q}}{}^i = \phi_{\vec{q}}{}^i/\epsilon_T, \quad \phi_{\vec{q}}{}^i = |v^i(\vec{q})|^2 D_i(\vec{q},\omega+i0),$$

$$\epsilon_T(\vec{q},\omega+i0) = 1 - \sum_\nu\sum_i \phi_{\vec{q}}{}^i\sum_{\alpha,\beta}|m_{\alpha\beta}{}^i|^2 Q_{\beta\alpha}(\omega+i0),$$

and D_i is the ith-phonon propagator.

The $b^+(1)$ transition spectrum is obtained from Eq. (4) by choosing $\alpha = b^+(1)$ and $\beta = g$, the acceptor ground state. In calculating the proper self-energy part $\Sigma_{b^+(1)}$ of the state $|b^+(1)\rangle$, only the heavy-mass excited state f, located at 8.7 meV from g, is considered. According to the experimental results, this is the only final state involved in the resonant phonon emission. Singularities occur in $\Sigma_{b^+(1)}(\omega)$ at $\hbar\omega = \epsilon(f) + \hbar\omega_i$, where $\hbar\omega_i$ is the TO- or LO-phonon energy. These two singularities lead to the appearance of three branches in the $b^+(1)$ spectrum as a function of magnetic field.

Calculation of the strengths of the hole-phonon interactions is hampered by the lack of information regarding the acceptor state wave functions for InSb. This necessitates a model approximation to the actual envelope wave functions for $b^+(1)$ and f. $b^+(1)$ should be well approximated by the variational functions $F_{lm\lambda}(\sigma, \varphi, z) = \Phi_{lm}(\sigma, \varphi) P_\lambda(z) \exp(-\gamma\epsilon^2 z^2/4)$ calculated for donor states.[12] Therefore the selection $F_{b^+(1)}(\vec{r}) = F_{010}(\sigma, \varphi, z)$ is made. Since f is a high-mass excited state, it could be approximated by a typical variational function obtained for Ge,[13] since the acceptor states of InSb and Ge should be similar. However, for analytical expediency, we choose $F_f(\vec{r}) = N_f \exp(-\beta r^2)$, with $\beta = 0.283(a^*)^{-2}$,[12] and a^* obtained by scaling the high-mass effective Bohr radius $a_0^* = D\hbar^2/m^*e^2$ such that the corresponding effective Rydberg is the binding energy of f. With these functions, the matrix elements $\langle F_{b^+(1)}| \times \exp(i\vec{q}\cdot\vec{r})|F_f\rangle$ and hence σ can be evaluated analytically. A good fit to the data has been obtained for a value of the optical deformation potential d_0 twice that given in the literature,[14] and for a ratio of hole-TO to hole-LO coupling strength of approximately one half. In view of the highly approximate nature of the calculation, the estimate for d_0 is considered satisfactory. Preliminary results employing a more realistic description of the acceptor wave functions indicate that a reliable determination of d_0 will be possible from these experiments.

[1]D. M. Larsen, in *Proceedings of the Tenth International Conference on the Physics of Semiconductors, Cambridge, Massachusetts, 1970,* edited by S. P. Keller, J. C. Hennel, and F. Stern, CONF-700801 (U.S. AEC Division of Technical Information, Springfield, Va., 1970), p. 145.

[2]H. Ehrenreich and A. W. Overhauser, Phys. Rev. 104, 331 (1956).

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

[4]D. C. Tsui, Phys. Rev. Lett. 21, 994 (1968).

[5]R. Kaplan, Phys. Rev. Lett. 20, 329 (1968).

[6]R. Kaplan, S. G. Bishop, and B. D. McCombe, in *Proceedings of the Ninth International Conference on the Physics of Semiconductors, Moscow, U. S. S. R., 1968* (Nauka, Leningrad, U.S.S.R., 1968), p. 317.

[7]C. R. Pidgeon and R. N. Brown, Phys. Rev. 146, 575 (1966).

[8]V. N. Murzin, A. I. Demishina, and L. M. Umarov, Fiz. Tekh. Poluprov. 3, 434 (1969) [Sov. Phys. Semicond. 3, 367 (1969)].

[9]R. A. Stradling and R. A. Wood, J. Phys. C: Proc. Phys. Soc., London 3, 2425 (1970).

[10]G. L. Bir and G. E. Pikus, Fiz. Tverd. Tela 2, 2287 (1960) [Sov. Phys. Solid State 2, 2039 (1960)].

[11]K. L. Ngai, Phys. Rev. B 3, 1303 (1971).

[12]R. F. Wallis and H. J. Bowlden, J. Phys. Chem. Solids 7, 78 (1958).

[13]K. S. Mendelson and H. M. James, J. Phys. Chem. Solids 25, 729 (1964); see also P. J. Lin-Chung and R. F. Wallis, J. Phys. Chem. Solids 30, 1453 (1969).

[14]J. D. Wiley, Solid State Commun. 8, 1865 (1970); see also P. Lawaetz, Phys. Rev. B 4, 3460 (1971).