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Anisotropic Electron-Phonon Coupling*

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The interaction between electrons and "pseudolongitudinal" or "pseudotransverse" phonons is derived and used in the calculation of renormalized phonon modes. Coupled modes involving plasmon and optical pseudolongitudinal or pseudotransverse phonons are expected to give rise to observable anisotropic results in Raman scattering.

I. INTRODUCTION

Wave motion in homogenous isotropic media is capable of a convenient representation in terms of transverse and longitudinal modes. In the case of real elastic media, such a decomposition becomes idealized, sound waves moving near to a surface or light passing through an anisotropic medium may not be discussed in terms of purely irrotational or divergence free waves. Similarly the lattice vibrations of many crystals are not purely longitudinal or transverse except in certain particular directions. The treatment of phonons and of electron-phonon interaction in solid-state physics is usually based upon an isotropic model for the crystal. If such a model is augmented in favor of one displaying some on the anisotropies of real crystals it becomes interesting to consider the consequences. In Sec. II of this paper an anisotropic electron-phonon interaction is derived and used in Sec. III to determine the renormalization of the acoustic modes and of the electron gas. Some of the consequences are outlined in Sec. IV together with a discussion of the possibility of the existence of observable effects. Finally in Sec. V the mixed optical phonon and phonon modes are derived and their significance in Raman scattering discussed.

II. ANISOTROPIC ELECTRON-PHONON COUPLING

In a crystal, having a single ion in each cell, the

ith ion is displaced from its equilibrium position R_{i0} by a small distance δR_i as a result of thermal vibrations of the crystal. In such a crystal three normal modes of vibration occur which are characterized by normal mode vectors $\vec{q}_{k\lambda}$ and polarization vectors $\vec{\epsilon}_{k\lambda}$, where $\vec{k}$ is a momentum vector and the index λ labels the three modes; thus

$$\delta R_i \propto \sum_{k\lambda} \vec{q}_{k\lambda} \cdot \vec{\epsilon}_{k\lambda} e^{i\vec{k}\cdot\vec{R}_{i0}} . \tag{1}$$

The polarization vectors are orthogonal $\vec{\epsilon}_{k\lambda}\vec{\epsilon}_{k\lambda'} = \delta_{\lambda\lambda'}$ in the special case of a crystal without a basis. If the crystal lattice were totally isotropic then the direction of propagation of the vibrations $\vec{n}$ would be the same as $\vec{k}$ and the displacements of the ions perpendicular or parallel to this direction of propagation. In a real crystal the ionic displacements do not take place parallel or perpendicular to the direction of propagation, or to $\vec{k}$, unless the vibrational mode happens to propagate along some special symmetry direction. Thus, in general,

$$\vec{\epsilon}_{k1} \cdot \vec{q}_{k1} \neq |q_{k1}| \quad \text{and} \quad \vec{\epsilon}_{k\lambda} \cdot \vec{q}_{k\lambda} \neq 0 \quad (\lambda = 2, 3) ,$$

and such lattice vibrations may be classified as "pseudolongitudinal" $\lambda = 1$, or "pseudotransverse" $\lambda = 2, 3$.

In certain materials the derivations of these modes from the purely longitudinal or transverse cases is by no means negligible; for example, in the case of zinc, Musgrave[1] computes the maximum angle between the displacement vector and the wave normal for a LA wave to be almost 30° and the angle between the ray and the wave normal to be greater than 30°.

Since electrical fields are associated with the lattice vibrations and coupling to the electron gas is a result of the ionic displacements, it is important to investigate any new effects which result when the simple picture of lattice vibrations is modified.

The coupling of electrons to phonons may be written

$$A_{kk'\lambda} = \vec{\epsilon}_{q\lambda}(\vec{k} - \vec{k}') \, V(\vec{k} - \vec{k}') , \tag{2}$$

where $\vec{k}$ and $\vec{k}'$ are the initial and final wave vectors of the scattered electron and $V(\vec{k} - \vec{k}')$ is a matrix element between these initial and final states. If we assume umklapp processes to be negligible, that is confine ourselves to low temperatures and to momenta which are not close to the zone boundary, then we may assume that the matrix element $V(\vec{k} - \vec{k}')$ is a function of $(\vec{k} - \vec{k}') = \vec{q}$ alone. Hence

$$A_{q\lambda} = \vec{\epsilon}_{q\lambda} \cdot \vec{q} \, V(q) . \tag{3}$$

Let the angle between $\vec{q}$ and $\vec{\epsilon}_{q\lambda}$ be Δ_λ, then

$$A_{q\lambda} = \cos(\Delta_\lambda) q V(q) , \tag{4}$$

and for arbitrary directions of propagation of phonon modes in the crystal the coupling of transverse phonons to the electrons is not zero. The coupling constant between the electron gas and each vibrational mode depends upon the direction of propagation of that mode. Only in the special case $\Delta_{T_1} = \Delta_{T_2} = \frac{1}{2}\pi$, $\Delta_L = 0°$, does $A_{qT} = 0$, $A_{qL} = qV(q)$. This orientation-dependent coupling will give rise to additional renormalization effects and certain anisotropic phenomena which are discussed in Secs. III–VI.

III. RENORMALIZATION

The dispersion relation for longitudinal phonons, which have been calculated on the basis of an unscreened ion-ion potential, has incorrect asymptotic behavior in the long-wavelength limit, the phonon frequency does not approach zero. However, when the effects of electron scattering from phonons are considered the longitudinal mode becomes renormalized and an effective lattice mediated interaction between the electrons appears.

If the electron-phonon interaction derived in Sec. II is used, then the pseudotransverse as well as the pseudolongitudinal phonon modes will be renormalized. In the harmonic approximation the following Hamiltonian may be assumed:

$$H = \sum_p \epsilon_p C_p^\dagger C_p + \sum_{pp'k} (\tfrac{1}{2} V_K)(C_{p+k}^\dagger C_{p'-k}^\dagger C_{p'} C_p) + \sum \Omega_{k\lambda} b_{k\lambda}^\dagger b_{k\lambda} + \sum_{\lambda, p_1 - p_2 = k} A_{k\lambda}(b_{k\lambda} + b_{-k\lambda}^\dagger) C_{p_1}^\dagger C_{p_2} , \tag{5}$$

where $C_p^\dagger$ and $b_p^\dagger$ are creation operators for electrons and phonons, respectively, the first two terms describe the electron gas, the third term describes the three phonon modes having frequencies $\Omega_{k\lambda}$, and the final term contains the electron-phonon coupling "constant," a function of $\cos\Delta_\lambda$. With the aid of retarded double-time Green's functions the dispersion relations of the various modes may be calculated. The phonon Green's function $\langle\langle B_{k\lambda}, B_{k\lambda}^\dagger \rangle\rangle$ is defined in terms of the operators $B_{k\lambda} = b_{k\lambda} + b_{-k\lambda}^\dagger$, $B_{k\lambda}^\dagger = b_{k\lambda}^\dagger + b_{-k\lambda}$. Similarly a two-particle Green's function for the electron gas $\langle\langle \rho_k, \rho_k^\dagger \rangle\rangle$ is defined in terms of the density operator $\rho_k = \sum_p C_{p-k}^\dagger C_p$.

The equation of motion for the phonon Green's function using the Hamiltonian (5) is

$$(\omega^2 - \Omega_{k\lambda}^2)\langle\langle B_{k\lambda}, B_{k\lambda}^\dagger \rangle\rangle = (\Omega_{k\lambda}/2\pi) + A_{-k\lambda}\Omega_{k\lambda}\langle\langle \rho_{k\lambda}, B_{k\lambda}^\dagger \rangle\rangle . \tag{6}$$

The equation of motion for the mixed Green's function which occurs on the right-hand side of (6) is

$$[1 + V_k \chi(k)]\langle\langle \rho_k, B_{k\lambda}^\dagger \rangle\rangle = \chi(k) \textstyle\sum_{\lambda'} A_{k\lambda'} \langle\langle B_{k\lambda'}, B_{k\lambda}^\dagger \rangle\rangle , \tag{7}$$

where

$$\chi(k)=\sum_p (n_{p+k}-n_p)/(\omega-\epsilon_{p+k}+\epsilon_p)$$

and n_p are the electronic occupation numbers. Using expression (7) one obtains for the phonon Green's function

$$(\omega^2-\Omega_k^2)\langle\langle B_{k\lambda}, B_{k\lambda}^\dagger\rangle\rangle = \frac{\Omega_{k\lambda}}{2\pi}+\frac{A_{k\lambda}\chi(k)}{1+V_k\chi(k)}\Omega_{k\lambda}\sum_{\lambda'} A_{k\lambda'}\langle\langle B_{k\lambda'}, B_{k\lambda}^\dagger\rangle\rangle . \quad (8)$$

Writing $\bar{A}_{k\lambda}=A_{k\lambda}/[1+V_k\chi(k)]$ as the screened matrix element for electron-phonon coupling, (8) becomes

$$[\omega^2-\Omega_{k\lambda}^2-A_{k\lambda}\bar{A}_{k\lambda}\chi(k)\Omega_{k\lambda}]\langle\langle B_{k\lambda}, B_{k\lambda}^\dagger\rangle\rangle = \frac{\Omega_{k\lambda}}{2\pi}+\bar{A}_{k\lambda}\Omega_{k\lambda}\chi(k)\sum_{\lambda'\neq\lambda} A_{k\lambda'}\langle\langle B_{k\lambda'}, B_{k\lambda}^\dagger\rangle\rangle . \quad (9)$$

In the case of isotropic electron-phonon coupling with $\Delta_L=0$, $\Delta_{T_i}=\frac{1}{2}\pi$, the two transverse modes are uncoupled and unrenormalized and the dispersion relation for the longitudinal mode derived from (9) is just that found by Bardeen and Pines.[2]

In the case of arbitrary directions of propagation of the phonons, as a result of the anisotropic coupling constant (4), the dispersion relations become more complicated; all modes are renormalized and in addition become coupled via the mixed Green's function on the right-hand side of (9). This may be seen explicitly by writing the renormalized phonon Hamiltonian

$$H_{\text{ren}}=\sum_{\lambda'}\Omega_{k\lambda}b_{k\lambda}^\dagger g_{k\lambda}+\tfrac{1}{2}\sum_{\lambda'}\chi(k)A_{k\lambda}\bar{A}_{k\lambda'}\times(b_{k\lambda'}+b_{-k\lambda'}^\dagger)(b_{k\lambda}^\dagger+b_{-k\lambda}) . \quad (10)$$

The dispersion relations for the three phonon modes may be calculated with the aid of the Green's function $B=\langle\langle \vec{B}_k, \vec{B}_k^\dagger\rangle\rangle$, where $\vec{B}_k=(B_{k1}, B_{k2}, B_{k3})$. Using the Hamiltonian (10), the equation of motion is

$$B_0^{-1}B=U/\pi+MB , \quad (11)$$

where B_0 is the unperturbed Green's function,

$$U=\begin{vmatrix}\Omega_{k1}&0&0\\0&\Omega_{k2}&0\\0&0&\Omega_{k3}\end{vmatrix} , \quad (12)$$

and

$$M=\begin{vmatrix}2A_{k1}&\bar{A}_{k1}\chi(k)\Omega_{k1}&2A_{k1}&\bar{A}_{k2}\chi(k)\Omega_{k1}&2A_{k1}&\bar{A}_{k3}\chi(k)\Omega_{k1}\\2A_{k2}&\bar{A}_{k1}\chi(k)\Omega_{k2}&2A_{k2}&\bar{A}_{k2}\chi(k)\Omega_{k2}&2A_{k2}&\bar{A}_{k3}\chi(k)\Omega_{k2}\\2A_{k3}&\bar{A}_{k1}\chi(k)\Omega_{k3}&2A_{k3}&\bar{A}_{k2}\chi(k)\Omega_{k3}&2A_{k3}&\bar{A}_{k3}\chi(k)\Omega_{k3}\end{vmatrix} . \quad (13)$$

Writing $L=B_0^{-1}-M$ one obtains finally for the matrix elements of the Green's function for the renormalized phonon modes

$$B_{ii}=[(L)_{ii}/\det L](U/\pi) . \quad (14)$$

Each renormalized phonon frequency is a function of all three bare-phonon frequencies and coupling constants. At nonzero temperatures one would expect that anharmonic phonon terms would be larger than effects due to the off-diagonal terms occurring in M. That is the anharmonic effects should dominate the phonon-damping mechanisms.

Using a similar procedure, the density-fluctuation Green's function for the electron gas is

$$\left(1+V_k\chi(k)-\sum_\lambda\frac{2A_{k\lambda}^2\Omega_{k\lambda}\chi(k)}{(\omega^2-\Omega_{k\lambda}^2)\pi}\right)\langle\langle\rho_k\rho_k^\dagger\rangle\rangle=\frac{\chi(k)}{\pi} . \quad (15)$$

In the absence of any coupling to the phonons, one obtains the familiar dispersion relation

$$0=1+V_k\sum_p\frac{n_{p+k}-n_p}{\omega-\epsilon(p+k)+\epsilon(p)} , \quad (16)$$

and collective density oscillations occur at a characteristic plasmon frequency ω_p. Switching on the electron-phonon interaction acts as a small perturbation to the plasmon mode since $\omega_p\gg\Omega_{k\lambda}$; however, in the case of semiconductors the optical phonon modes may in certain cases have a large effect upon the plasmon modes.

In a normal metal the effect of the LA phonons upon the electron gas is to produce a very small effective interaction; for sufficiently small differences $\epsilon_{p+k}-\epsilon_p<\omega_c$ this interaction becomes attractive. In the present case, the magnitude of ω_c and the value of the matrix elements of the attractive interaction depend on all three phonon modes and coupling constants, that is, the critical superconducting temperature would not be simply related to the frequency and coupling constant of the longitudinal mode alone.

IV. EFFECTS

It is of interest to decide if such anisotropies in the coupling of electrons to acoustic phonons are manifest in any observable solid-state phenomena. One may immediately predict anisotropies in electron scattering, ultrasonic attenuation, and magnetoresistance. However, the discussion below indicates that any of the existing experimental results does not have a clear interpretation. Anisotropies in the scattering rate should occur, however such effects are sensitive to the shape of the Fermi sur-

face and unless this is known with sufficient accuracy, any attempt to discuss the effect in terms of anisotropies in electron-phonon coupling would be futile.

The attenuation of ultrasonic waves at sufficiently low temperature is a result of interactions involving the conduction electrons and any faults in the crystal. This attenuation is a function of the orientation of the crystal but one supposes that such effects are dominated by anisotropies of the Fermi surface. In the case of copper, however, experimental results cannot be fully understood as a result to calculations based on deviations of the Fermi surface from a spherical shape.[3]

Magnetoresistance is a highly anisotropic phenomena, the transverse magnetoresistance of a crystal vanishing in a free-electron model; in a real metal it is found that the magnetoresistance does not vanish and its temperature dependence is sensitive to the orientation of the magnetic field. A number of suggestions as to such orientation dependence have been offered.[4] However, until accurate calculations are made it does not seem possible to identify the relative magnitudes of such effects.

In the case of acoustic phonons one finds the renormalization and coupling of the phonon modes and the modification of the lattice mediated electronic interaction leading to a more complicated expression for the superconducting critical temperature.

Finally, no anisotropic effects due to acoustic phonon-electron coupling appear to be at present detectable. Nevertheless, in the analysis of the accurate experimental data considerations of phonon anisotropies should be included.

V. PLASMON-PHONON MIXED MODES

In general, plasmon and optical phonon modes have frequencies which differ by several orders of magnitude. However if a semiconductor is doped in a particular manner the plasmon frequency may decrease below that of the optical phonon frequency. Thus it becomes possible to look for plasmon optical phonon mixed modes which propagate through the semiconducting material. Such modes have been detected by Mooradian and Wright[5] using Raman scattering methods.

The derivation of an electron-phonon matrix in the case of a crystal containing two oppositely charged ions in the primitive cell is not as simple as that discussed in Sec. III.

The simple orthogonality condition on the polarization vectors $\vec{\epsilon}_{k\lambda}\vec{\epsilon}_{k\lambda'} = \delta_{\lambda\lambda'}$ is replaced by

$$\sum_{b=1}^{2} \vec{\epsilon}_{k\lambda b}\vec{\epsilon}_{k\lambda' b} = \delta_{\lambda\lambda'}, \quad b = 1, 2$$

where b labels the branches, optical and acoustic.

In addition the approximation $V(\vec{k}-\vec{k}') = V(q)$, where $q = |\vec{k}-\vec{k}'|$, is more difficult to justify. If one assumes that umklapp processes are again negligible and the optical and acoustic branches are well separated then the matrix element for electron optical phonon coupling may be approximated by

$$W_{q\lambda 2} = \vec{\epsilon}_{q\lambda 2}\vec{q}\,V(q)\,, \tag{17}$$

where $\vec{\epsilon}_{q\lambda 2}$ is the polarization vector for optical modes. Since only optical modes will be referred to below the index b will be dropped.

A rather simple Hamiltonian which reduces to that of Singwi and Tosi[6] in the case of an isotropic lattice will be used to represent the coupling of optical phonons to plasmons:

$$H = \sum_{p\lambda} \Omega_{p\lambda} b^{\dagger}_{p\lambda} b_{p\lambda} + \sum_{p} \omega_p a^{\dagger}_p a_p + \sum_{p\lambda} W_{p\lambda}(a^{\dagger}_p + a_{-p})(b_{p\lambda} + b^{\dagger}_{-p\lambda})\,, \tag{18}$$

where $b_{p\lambda}$ creates an optical phonon of frequency $\Omega_{p\lambda}$ and polarization λ and $a^{\dagger}_p$ creates a plasmon of frequency ω_p and is assumed to obey Bose commutation relations. The plasmon frequency is proportional to the electron carrier concentration. Introduction of the operators $\alpha_p = a^{\dagger}_p + a_{-p}$ and $B_{p\lambda} = b_{p\lambda} + b^{\dagger}_{-p\lambda}$ enables the Green's functions $\langle\langle \alpha_p, \alpha^{\dagger}_p \rangle\rangle$ and $\langle\langle B_{p\lambda}, B^{\dagger}_{p\lambda} \rangle\rangle$ to be evaluated. The plasmon Green's function yields the dispersion relation

$$\omega^2 - \omega_p^2 - 4\omega_p \sum_{\lambda} \frac{W^2_{p\lambda}\Omega_{p\lambda}}{\omega^2 - \Omega^2_{p\lambda}} = 0\,, \tag{19}$$

the plasmon being coupled to all three optical phonon modes. In the case of a plasmon propagating along a lattice plane such that the ionic displacements are perpendicular to the direction of propagation, (19) simplifies to the familiar form

$$\omega^2 - \omega_p^2 - 4\omega_p W^2_{pL}\,\Omega_{pL}/(\omega^2 - \Omega^2_{pL}) = 0\,. \tag{20}$$

In other directions the dispersion relation is more complicated and depends upon the relative magnitudes of coupling constants and frequencies.

Proceeding in a similar manner to Sec. III the renormalized optical phonon modes are associated with the Hamiltonian

$$H_{\text{ren}} = \sum_{\lambda} \bar{\Omega}_{p\lambda} b^{\dagger}_{p\lambda} b_{p\lambda} + \sum_{\lambda\lambda'} Z_{p\lambda\lambda'} B_{p\lambda'} B^{\dagger}_{p\lambda}\,, \tag{21}$$

where

$$\bar{\Omega}^2_{p\lambda} = \Omega^2_{p\lambda} + 4W^2_{p\lambda}\omega_p\Omega_{p\lambda}/(\bar{\Omega}^2_{p\lambda} - \omega_p^2) \tag{22}$$

are the renormalized frequencies and

$$Z_{p\lambda\lambda'} = 2W_{p\lambda'}W_{p\lambda}\omega_p/(\omega^2 - \omega_p^2)\,. \tag{23}$$

The dispersion relation (19) may also be written

$$\omega^8 - \omega^6 \sum_{i=1}^{4} \omega_i^2 + \omega^4 \left(\sum_{i \neq j=1}^{4} \omega_i^2 \omega_j^2 - 4\omega_1 \sum_{i=2}^{4} W_i^2 \omega_i \right)$$

$$- \omega^2 \left(\sum_{i \neq j \neq k=1}^{4} \omega_j^2 \omega_j^2 \omega_k^2 - 4\omega_1 \sum_{i \neq j \neq k=2} W_i^2 \omega_i (\omega_j^2 + \omega_k^2) \right)$$

$$+ \omega_1^2 \omega_2^2 \omega_3^2 \omega_4^2 - 4\omega_1 \sum_{1 \neq j \neq k=2} W_i^2 \omega_i \omega_j^2 \omega_k^2 , \qquad (24)$$

where the notation $\omega_1 = \omega_p$, $\omega_i = \Omega_{\lambda k}$, $W_i = W_{\lambda k}$ $i = \lambda = 2, 3, 4$ is used for convenience.

Further investigation of this dispersion relation is possible once a specific form for the electron-phonon interaction is proposed. In the long-wavelength approximation such an interaction may be derived electrostatically.

Consider the response of the lattice to the charge density $\rho(\vec{r}, t)$,

$$\vec{\nabla} \cdot \vec{D} = 4\pi\rho . \qquad (25)$$

In the long-wavelength approximation, the polarizations of the lattice combine additively, thus

$$\vec{P} = b_{22} \vec{E} + b_{21} \textstyle\sum_\lambda \vec{u}_\lambda , \qquad (26)$$

where $\vec{u}_\lambda$ are the relative displacements of the ions. The electric field is thus given by

$$\vec{\nabla} \cdot \vec{E} = \frac{4\pi}{1 + 4\pi b_{22}} \left(- b_{21} \sum_\lambda \vec{\nabla} \cdot \vec{u}_\lambda + \rho(\vec{r}, t) \right) . \qquad (27)$$

If the longitudinal vacuum field caused by $\rho(\vec{r}, t)$ is $\vec{E}_{vac}$ in the direction $\vec{n}$, which is the direction of propagation of the phonons, then the solution to Eq. (27) is

$$E\vec{n} = \frac{-4\pi b_{21}}{1 + 4\pi b_{22}} \sum_\lambda u_\lambda \vec{n} \cos\delta_\lambda + \frac{\vec{E}_{vac}}{1 + 4\pi b_{22}} , \qquad (28)$$

where δ_λ is the angle between $\vec{n}$ and $\vec{u}_\lambda$.

If the orientational dependence of the unrenormalized optical phonon frequencies is ignored then b_{21} and b_{22} may be taken as constant and the familiar identification

$$b_{12} = \left(\frac{\epsilon_0 - \epsilon_\infty}{4\pi} \right)^{1/2} \omega_0 \quad \text{and} \quad b_{22} = \frac{\epsilon_\infty - 1}{4\pi}$$

made, where ϵ_∞ and ϵ_0 are the high-frequency and static dielectric constants, respectively, and ω_0 is the infrared dispersion frequency. The electric field becomes

$$E\vec{n} = - \omega_l \left[4\pi \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \right]^{1/2} \sum_\lambda u_\lambda \vec{n} \cos\delta_\lambda + \frac{1}{\epsilon_\infty} \vec{E}_{vac} , \qquad (29)$$

where ω_l is taken to be the value of Ω_{Lk} in the case $\delta_L = 0 (= \Delta_L)$. The interaction term introduced into the Hamiltonian for the system by the coupling of $\rho(\vec{r}, t)$ to the λ optical mode is

$$H_\lambda^{int} = - \omega_l \left[4\pi \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \right]^{1/2} u_\lambda E_{vac} \cos\delta_\lambda . \qquad (30)$$

In the case that $\vec{E}_{vac}$ represents the longitudinal field of the electron plasma[6] the interaction is

$$W_{k\lambda} = \tfrac{1}{2} [\omega_l \omega_p (1 - \epsilon_\infty/\epsilon_0)]^{1/2} \cos\delta_\lambda . \qquad (31)$$

The dispersion relation (19) may be evaluated in the approximations stated above using (31), in addition to simplify the expression and to render its behavior more transparent δT_2 is set equal to $\frac{1}{2}\pi$, that is one of the transverse modes is assumed to be isotopic. Setting $\Omega_{T_2}^2 = \omega_{T_2}^2 = (\epsilon_\infty/\epsilon_0)\omega_L^2$ the dispersion relation becomes

$$\omega_2^2 = \omega_{T_2}^2$$

and

$$\omega^6 - \omega^4(\omega_p^2 + \omega_L^2 + \omega_T^2) + \omega^2[\omega_L^2 \omega_T^2 + \omega_p^2 \omega_L^2 \sin^2\delta_L + \omega_p^2 \omega_T^2 (1 + \cos^2\delta_L) - \omega_p^2 \omega_T \omega_L (1 - \epsilon_\infty/\epsilon_0)\cos^2\delta_T]$$

$$- \omega_p^2 \omega_L^2 \omega_T^2 \sin^2\delta_L - \omega_p^2 \omega_L^4 \cos\delta_L + \omega_p^2 \omega_T \omega_L^3 (1 - \epsilon_\infty/\epsilon_0)\cos^2\delta_T = 0 . \qquad (32)$$

The asymptotes to this dispersion relation become

$$\omega_1^2 = 0, \quad \omega_2^2 = \omega_3^2 = \omega_T^2 = \omega_4^2 = \omega_L^2 \qquad (33)$$

and

$$2\omega_{1,3}^2 = \omega_L^2 \sin^2\delta_L + \omega_T^2 (1 + \cos^2\delta_L) - \omega_L \omega_T (1 - \epsilon_\infty/\epsilon_0) \cos^2\delta_T$$

$$\pm \{ [\omega_L^2 \sin^2\delta_L + \omega_T^2 (1 + \cos^2\delta_L) - \omega_L \omega_T (1 - \epsilon_\infty/\epsilon_0)\cos^2\delta_T]^2$$

$$- 4[\omega_L^2 \omega_T^2 \sin^2\delta_L + \omega_T^4 \cos^2\delta_L - \omega_L \omega_T (\omega_L^2 \omega_T^2)\cos^2\delta_T] \}^{1/2} \quad (\omega_2^2, \omega_T^2, \omega_4^2 \to \omega_p^2). \qquad (34)$$

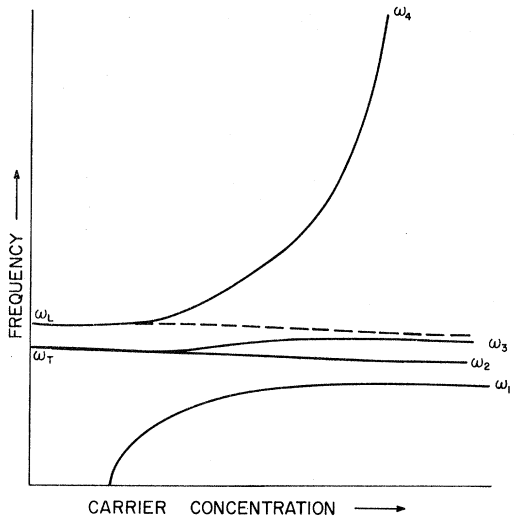

FIG. 1. Behavior of the mixed modes, from Eq. (32), with increasing electronic carrier concentration.

As the plasma frequency increases, the pseudotransverse mode ω_3 increases in frequency from ω_T to a limiting value below ω_L. The behavior of these modes is illustrated schematically by Fig. 1. In the case in which $\delta T_1 = \delta T_2 = \frac{1}{2}\pi$, $\delta_L = 0$,

$$2\omega_{1,4}^2 = \omega_p^2 + \omega_L^2 \pm [(\omega_p^2 + \omega_L^2)^2 - 4\omega_p^2\omega_T^2]^{1/2},$$
$$\omega_2^2 = \omega_3^2 = \omega_T^2 \qquad (35)$$

and the TO phonons are unperturbed by the plasmon.

The angular dependence of the frequencies of the modes was evaluated using the frequencies $\omega_T = 268\ \text{cm}^{-1}$, $\omega_L = 291\ \text{cm}^{-1}$, and $\omega_p = 700\ \text{cm}^{-1}$ and is displayed in Fig. 2; to facilitate the numerical solution of Eq. (32) it was assumed that $\delta_L = \frac{1}{2}\pi - \delta T_2$.

Hence in a Raman scattering experiment as the crystal is rotated from an orientation involving scattering from the mixed plasmon pure-LO-phonon modes the pseudotransverse phonon line will shift towards higher frequencies while the pseudolongitudinal optical phonon line is depressed to lower frequencies. The magnitude of the effect of course depends upon the degree of anisotropy of the sample.

In making a calculation of the cross sections for Raman scattering several refinements may be included. For example, the propagating electromagnetic wave couples to the TO phonon and in the anisotropic crystal there will be some degree of dispersion of the light from all modes. Strictly the Raman scattering of light is from the mixed photon-plasmon-phonon modes. If the crystal exhibits a high degree of anisotropy it is no longer possible to treat the electromagnetic wave as purely transverse; that is the ray vector and the wave are not parallel; the electrical displacement is not perpendicular to the direction of propagation. Anisotropies in the plasmon mode alone, in which coupling to the electromagnetic field results in mixed plasmon-photon modes have been discussed by Foo and Tzoar.[7]

VI. CONCLUSION

The anisotropic coupling between the electron gas and each of the acoustic phonon modes has been found to lead to a coupling of these modes and additional renormalization. These effects are not expected to be appreciable unless accurate experiments may be unambiguously analysed. In the case of optical phonons coupling of plasmons to all three lattice modes occurs and it would appear to be a simple experimental procedure to observe such mixed modes. These considerations indicate an experimental investigation of anisotropic effects could be performed, in particular using a material whose vibrational characteristics are sufficiently well known so that comparison with a theoretical prediction could be made.

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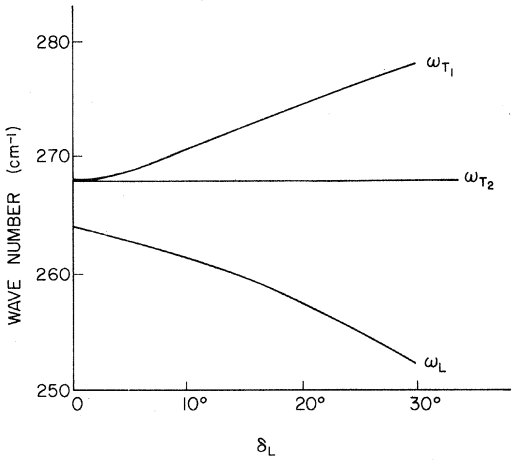

FIG. 2. Dependence of the frequencies of the mixed modes upon angle for $\omega_p = 700\ \text{cm}^{-1}$, $\omega_L = 291\ \text{cm}^{-1}$, $\omega_T = 268\ \text{cm}^{-1}$, and $\delta_{T_1} = \frac{1}{2}\pi - \delta_L$, $\delta_{T_2} = 0$.

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Anomalous Behavior of the Cyclotron Resonance of Holes in Bismuth

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The cyclotron resonance of holes in bismuth has been carefully investigated using microwaves of frequency 50 GHz in the Azbel'-Kaner configuration. It was found that the reflection peak is extremely weak at the fundamental but strong at the second harmonic, and the second-harmonic line shape may be quite complicated. These anomalous effects may be explained if the longitudinal magnetoplasma excitations in the vicinity of the second harmonic (Bernstein mode) are coupled with the electromagnetic waves of a magnetoplasma (Alfvén waves) in the weakly nonlocal regime. As a result, the location of the reflection peaks shifts from that of the exact cyclotron harmonics. Taking into account these shifts, we determined the cyclotron masses of holes to be $(0.210 \pm 0.002)m_0$ for $\vec{B} \perp \vec{E}_{rf}$ and $(0.217 \pm 0.002)m_0$ for $\vec{B} \parallel \vec{E}_{rf}$.

I. INTRODUCTION

A considerable number of experimental and theoretical investigations[1–6] have been performed on magnetoplasmas in bismuth in the microwave frequency region for the Voigt configuration.

The magnetoplasmas in bismuth have been investigated through the tilted-orbit cyclotron resonances,[2] hybrid resonances,[2] Alfvén waves,[7–11] and Azbel'-Kaner cyclotron resonances.[3,4,12] While the first three of these are observed for the classical skin effect condition, the last is observed in the extremely anomalous condition. However, in practice the hole cyclotron resonances in bismuth occur in a weakly nonlocal condition, especially in the case that the hole cyclotron frequency is less than the electron-hole hybrid resonance frequency.

Hebel[6] has discussed a slightly anomalous skin effect for the rf electric fields of the microwaves parallel to the static magnetic field ($\vec{E}_{rf} \parallel \vec{B}$; ordinary configuration). He found that the cyclotron resonance due to the dipole transition arising from the transverse excitation occurs only for the fundamental in this case. However, in the extraordinary configuration the longitudinal component of the dielectric constants should play an important role.

In this paper we shall present both the experimental results and the theoretical calculations for the anomalous line shape of hole cyclotron resonances in bismuth, especially for the extraordinary configuration. Our data indicate that the reflection peak near the second harmonic is stronger than the peak associated with either the fundamental or other harmonics. In addition, the line shape of the reflection peak may be quite complicated. This behavior is quite different from those for metals with large values of $\omega\tau$, in which case the line shapes depend upon the structure of the Fermi surface as was explained by Chambers[13] and experimentally shown by Moore.[14]

II. PLASMA EXCITATION AND PHOTON-PLASMA COUPLED MODE

The Azbel'-Kaner cyclotron resonances for metals have been discussed in terms of surface impedance.[15–17] In Sec. IV we shall analyze the line shapes of the microwave reflection from this viewpoint and compare with experimental results. In this section we shall present a preliminary discussion using the dispersion relations for plasma excitations. We shall assume for simplicity that the energy surfaces are spherical.

Throughout our treatment we assume that the static magnetic field $\vec{B}$ is oriented along the z axis, and the wave vector $\vec{q}$ is parallel to the x axis of a Cartesian coordinate system. The dispersion relations for magnetoplasma excitations as well as the coupled modes of excitations between electromagnetic waves and plasma can be given in terms of the dielectric functions