Cyclotron Resonance of Piezoelectric Polarons

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The magnetic field dependence of the energy and linewidth of the transition from the $n=1$ to the $n=0$ Landau level of a piezoelectric polaron has been calculated numerically for polarons at zero temperature. A weak isotropic piezoelectric coupling between the electron and the acoustic phonon modes is assumed, and is treated as a perturbation on free-electron magnetic eigenstates. It is found that the shift in the cyclotron resonance frequency due to piezoelectric electron-phonon interaction begins to differ drastically from that expected from the polaron effective-mass theory when $\hbar\omega_c/mc^2>1$, where $\hbar\omega_c$ is the separation in energy of the unperturbed magnetic levels, m is the band mass of the electron, and c is the velocity of sound in the crystal. The semiclassical theory of Mahan and Hopfield is reviewed and shown not to be suitable for interpreting recently reported cyclotron-resonance experiments in CdS, where the Landau-level spacings were substantially greater than the mean thermal energy per electron. Difficulties encountered in extending the present perturbation calculation to finite temperature are pointed out. Finally, the weak-coupling energy shift of the $n=0$ to $n=1$ transition for optical polarons (electrons coupled to longitudinal optical phonons) is evaluated as a function of magnetic field and compared to previous results derived for weak fields. It is suggested that the markedly nonlinear magnetic field dependence of the energy shift found might offer an attractive experimental way of observing optical-polaron effects on the electron self-energy.

I. INTRODUCTION

THE present study of "piezoelectric polarons" (electrons coupled to acoustic phonons via the piezoelectric electron-phonon interaction) is prompted by the suggestion of Mahan and Hopfield[1] that the piezoelectric electron-phonon interaction might play an important role in determining the cyclotron resonance frequency for electrons in the conduction band of piezoelectric semiconductors, e.g., CdS. At the present time, measurements of the effective mass of photo-excited conduction electrons in CdS present something of a mystery. On the one hand, careful cyclotron-resonance measurements by Baer and Dexter[2] (henceforth referred to as BD) give a slightly anisotropic "effective mass," which varies between 0.162 m_0 and 0.171 m_0 (where m_0 is the electron mass in vacuum), depending upon the orientation of the static magnetic field relative to the crystal c axis. On the other hand, a variety of different experiments, though less precise, more or less agree among themselves that the "effective mass" lies somewhere between 0.19 m_0 and 0.21 m_0. None of these latter experiments were cyclotron-resonance measurements and all have either been carried out at much higher temperature than the 1.3°K of the BD experiment or involve very high frequency transitions compared to the 72-Gc/sec transitions observed by BD.

To explain the disagreement between the measured values of "effective mass," Mahan and Hopfield suggested that at moderately low temperatures and frequencies, piezoelectric coupling in CdS could produce the required shift in the cyclotron resonance energy (of the order of 15%), but that in experiments at high temperature or frequencies, piezoelectric-polaron effects are negligible.

Unfortunately the calculation of Mahan and Hopfield (henceforth referred to as MH) is semiclassical. The experiment of BD, on the other hand, is done at low temperature and strong field. One may ask to what extent the semiclassical treatment approximates a fully quantum-mechanical calculation of the line shift.

We have undertaken an investigation of this question for a hypothetical experiment at zero temperature. We find that the agreement between MH and the quantum-mechanical result is very good for fields weak enough that $\hbar\omega_c<\frac{1}{2}mc^2$. However, for $\hbar\omega_c\gg\frac{1}{2}mc^2$ the semiclassical approximation gives a qualitatively incorrect result, predicting an energy shift which is much too large and of the wrong sign. While the deviation of the semiclassical theory from the results of an exact theory may not be as serious at a temperature of 1.3°K as at 0°K, we shall give an argument to show that a fully quantum-mechanical treatment is necessary for a self-consistent interpretation of the BD experiment.

In the present work no weak-field approximations are made. We investigate the line shift and linewidth due to piezoelectric electron-phonon interaction at zero temperature, using the "isotropic interaction" model,[3] defined by the Hamiltonian

$$\begin{aligned}&\tilde{H}=\tilde{H}_0+\tilde{H}_1,\\&\tilde{H}_0=\hbar c\sum l b_{\mathbf{l}}^{\dagger}b_{\mathbf{l}}+(1/2m)(\mathbf{P}_{\mathrm{op}}+(e/\tilde{c})\mathbf{A})^2,\\&\tilde{H}_1=(\kappa/\Omega^{1/2})\sum(1/l^{1/2})(e^{-i\mathbf{l}\cdot\mathbf{R}}b_{\mathbf{l}}^{\dagger}+\mathrm{H.c.}).\end{aligned}\qquad(1)$$

Here $b_{\mathbf{l}}^{\dagger}$ and $b_{\mathbf{l}}$ create and annihilate, respectively, acoustic phonons of wave vector $\mathbf{l}$, c is the velocity of sound in the solid, m is the electron band mass, e is the magnitude of the electron charge, $\tilde{c}$ is the velocity of light, Ω is the crystal volume, $\mathbf{A}$ is the vector potential describing the applied magnetic field, and κ is a coupling

* Operated with support from the U. S. Air Force.
[1] G. D. Mahan and J. J. Hopfield, Phys. Rev. Letters **12**, 241 (1964).
[2] W. Baer and R. N. Dexter, Phys. Rev. **135**, A1388 (1964); K. Sawamato, J. Phys. Soc. Japan **18**, 1224 (1963).
[3] A. R. Hutson, J. Appl. Phys. Suppl. **32**, 2287 (1961).

coefficient with dimensions of energy times length. If we use $mc^2/2$ as our unit of energy and $r_0=\hbar/mc$ as our unit of length we can introduce the quantities

$$\begin{gathered}\mathcal{H}=2\tilde{H}/mc^2, \quad \hat{\mathbf{p}}=(r_0/\hbar)\mathbf{P}_{\rm op}=\mathbf{P}_{\rm op}/mc, \quad \mathbf{k}=r_0\mathbf{l}, \\ (4\pi\alpha)^{1/2}=2\kappa/r_0mc^2, \quad \mathcal{S}=\Omega/r_0^3, \\ \mathbf{r}=\mathbf{R}/r_0, \quad y=Y/r_0, \quad \mathbf{A}=(-HY,0,0), \\ \lambda^2=2\hbar(eH/m\tilde{c})/mc^2,\end{gathered} \tag{2}$$

where we have assumed a uniform magnetic field, H, in the z direction and we use the Landau gauge for $\mathbf{A}$ (Y is the Y coordinate of the electron). The dimensionless electron momentum operator is denoted by $\hat{\mathbf{p}}$ and its eigenvalue by $\mathbf{p}$.

II. PERTURBATION THEORY

In terms of the quantities in (2) we can rewrite Eqs. (1) in the form

$$\begin{aligned}&\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1, \\ &\mathcal{H}_0=2\sum k b_{\mathbf{k}}^\dagger b_{\mathbf{k}}+(\hat{p}_x-\tfrac{1}{2}\lambda^2 y)^2+\hat{p}_y^2+\hat{p}_z^2, \\ &\mathcal{H}_1=(4\pi\alpha/\mathcal{S})^{1/2}\sum (1/k^{1/2})(e^{-i\mathbf{k}\cdot\mathbf{r}}b_{\mathbf{k}}^\dagger+\text{H.c.}).\end{aligned} \tag{3}$$

Consider first the case when no magnetic field is present ($\lambda^2=0$). The unperturbed states of interest, eigenstates of $\mathcal{H}_0$, are given by

$$e^{i\mathbf{p}\cdot\mathbf{r}}|0\rangle,$$

where $|0\rangle$ is the phonon vacuum and $p\leq 1$. Treating $\mathcal{H}_1$ as a perturbation we obtain for the energy correction $E_c(p)$

$$E_c(p)=-(4\pi\alpha/\mathcal{S})\sum k^{-1}(2k-2\mathbf{p}\cdot\mathbf{k}+k^2)^{-1}. \tag{4}$$

Expression (4) is not valid for $p>1$ since in that case the denominator of the summand can vanish at nonzero k. With this restriction on p we obtain for the excitation energy in the limit of infinite volume

$$\begin{aligned}&p^2+E_c(p)-E_c(0) \\ &\quad=p^2+\frac{\alpha}{\pi}\frac{1}{p}\left[-2p+\ln\left(\frac{1+p}{1-p}\right)+p\ln(1-p^2)\right] \\ &\quad=p^2-(\alpha/\pi)[p^2/3+p^4/10+p^6/21+\cdots].\end{aligned} \tag{5}$$

Now in the case of electrons coupled to *optical* modes (which we will refer to as "optical polarons") it is shown in[4] I that the zero-field excitation energy analogous to (5) acts as an effective Hamiltonian (in the sense that its eigenvalues when p^2 is replaced by $\hat{p}_y^2+[\hat{p}_x-(\lambda^2/2)y]^2+\hat{p}_z^2$ give the correct energy spectrum for the optical polaron in a magnetic field)—provided that the magnetic field is sufficiently weak. If we compare the dimensionless Hamiltonian $\mathcal{H}$ in I to (3) we find that the only formal difference is in the phonon energy spectrum ($2k$ here versus 1 for optical phonons) and in the wave-vector dependence of the interaction ($1/k^{1/2}$ in (3) versus $1/k$ for electron-optical phonon interaction). Therefore we might expect (5) to serve as an effective Hamiltonian for piezoelectric polarons in a magnetic field.

To investigate this possibility we consider $\mathcal{H}_0$ as the unperturbed Hamiltonian with normalized eigenstates

$$e^{i(p_x x+p_z z)}\phi_n(y+2p_x/\lambda^2)|\{n_k\}\rangle,$$

where

$$(p_y^2+\lambda^4 y^2/4)\phi_n(y)=(n+\tfrac{1}{2})\lambda^2\phi_n(y)$$

and $|\{n_k\}\rangle$ is the phonon state characterized by the set of occupation numbers $\{n_k\}$. Modifying only trivially the relevant expressions in I we can write down the energy, E_n, of the perturbed $n=1$ and $n=0$ states. (Since this is a zero-temperature calculation we can set $p_z=0$ and assume that no phonons are present in the unperturbed states of interest.) The required expressions are

$$\begin{aligned}&E_0=\tfrac{1}{2}\lambda^2+\alpha C_0(\lambda), \\ &E_1=\tfrac{3}{2}\lambda^2+\alpha C_1(\lambda),\end{aligned}$$

$$C_0(\lambda)=-\frac{4\pi}{\mathcal{S}}\sum_{\mathbf{k}}\frac{\exp[-(k_\perp/\lambda)^2]}{k}\sum_{n=0}^{\infty}\frac{1}{n!}\left(\frac{k_\perp}{\lambda}\right)^{2n}\times\frac{1}{2k+(n\lambda^2-k_\perp^2)+k^2}, \tag{6}$$

$$C_1(\lambda)=-\frac{4\pi}{\mathcal{S}}\mathcal{P}\sum_{\mathbf{k}}\frac{\exp[-(k_\perp/\lambda)^2]}{k}\sum_{n=0}^{\infty}\frac{1}{n!}\left(\frac{k_\perp}{\lambda}\right)^{2(n-1)}\times\frac{(n-k_\perp^2/\lambda^2)^2}{2k+[(n-1)\lambda^2-k_\perp^2]+k^2}, \tag{7}$$

where $k_\perp$ is the magnitude of the component of $\mathbf{k}$ in the x-y plane.

We use the symbol $\mathcal{P}$ in (7) to mean that in the infinite-volume limit the sum is to be evaluated as a principal-part integral.

It is important to realize that it is always energetically possible for an electron in a Landau level with $n>0$ to drop into a lower level by emitting an acoustic phonon. Therefore no Landau level except $n=0$ can be perfectly sharp, even at zero temperature. This is in marked contrast to the model considered in I, in which the electrons are coupled exclusively to the longitudinal optical phonon modes. In that case perturbation theory on the low-lying levels gives rise to stationary states and no principal-value integral appears in the expression corresponding to (7). Furthermore, variational techniques can be used to advantage in calculating the energy separation of the optical-polaron magnetic levels. No such methods can be trusted in treating piezoelectric-polaron levels except

[4] D. M. Larsen, Phys. Rev. 135, A419 (1964). This paper will be henceforth referred to as I.

possibly in the case of extremely weak or extremely strong magnetic field.

Investigating first the weak-field limit of (6) and (7) we note that in the limit $\lambda \to 0$ for fixed $k_\perp$ the factors $\exp[-(k_\perp/\lambda)^2](k_\perp/\lambda)^{2n}/n!$ are approximately given by

$$[\lambda/(2\pi)^{1/2}k_\perp]\exp[-(n\lambda^2-k_\perp^2)^2/2\lambda^2k_\perp^2]. \quad (8)$$

Expression (8), which is derived by using Stirling's approximation for $1/n!$, shows that $n\lambda^2-k_\perp^2$ can be treated as a small quantity when $\lambda \to 0$ provided that $k_\perp^2 \gg \lambda^2$. This suggests that (6) and (7) can be evaluated for small λ by expanding their energy denominators in powers of $(n\lambda^2-k_\perp^2)/(2k+k^2)$ and $[(n-1)\lambda^2-k_\perp^2]/(2k+k^2)$, respectively. In the case of optical polarons such expansions gave rise to a procedure for expanding the polaron energy in a power series in λ^2 as was indicated in I. However, that procedure fails in the case of piezoelectric polarons, since it leads formally to divergent integrals for the coefficients of terms of order λ^4 and higher. This probably means that corrections to the perturbed energy beyond the λ^2 term have no asymptotic expansion in powers of λ^2.

Nevertheless we can use the method of expansion to prove that, in agreement with the effective-mass theory,

$$\lim_{\lambda\to 0}(E_1-E_0)=\lambda^2-(\alpha/3\pi)\lambda^2. \quad (9)$$

The procedure is to write, for example,

$$\frac{1}{2k+(n\lambda^2-k_\perp^2)+k^2}=\frac{1}{2k+k^2}+\frac{(k_\perp^2-n\lambda^2)}{(2k+k^2)^2}+\frac{(k_\perp^2-n\lambda^2)^2}{(2k+k^2)^3}+R,$$

where

$$R=\frac{(k_\perp^2-n\lambda^2)^3}{(2k+k^2)^3[2k+(n\lambda^2-k_\perp^2)+k^2]}.$$

Inserting this expansion into (6) and the corresponding expansion into (7), one can verify (9) by evaluating exactly all terms except the terms arising from R in (6) and its analog in (7). But these remainder terms can be shown to have an upper bound which is of higher order than λ^2 in the limit $\lambda \to 0$.

Having established the asymptotic form of E_1-E_0 for small λ, we turn our attention to the limit of infinite λ. In this limit the dominant contributions to C_0 and C_1 come from the $n=0$ and $n=1$ terms, respectively. Evaluating the integrals for large λ gives

$$\lim_{\lambda\to\infty}(E_1-E_0)=\lambda^2+(3\sqrt{2}/64)\Gamma(1/4)\alpha\lambda^{1/2}\approx\lambda^2+0.2403\alpha\lambda^{1/2}. \quad (10)$$

We remark in passing that the $n=0$ term in C_0 [see (6)] is an upper bound to $E_0-\frac{1}{2}\lambda^2$ even when α is not small. This can be seen most easily by rewriting $\tilde{H}_0$ using the more symmetrical gauge $\mathbf{A}=\frac{1}{2}H(-Y, X, 0)$, reducing $\tilde{H}$ to dimensionless form, $\mathfrak{IC}$, and minimizing with respect to f_k the expectation value of $\mathfrak{IC}$ in

$$(\lambda/2\pi)^{1/2}\exp(-iz\sum k_z b_k^\dagger b_k)\exp(-\lambda\rho^2/4)\times\exp[\sum f_k(b_k^\dagger-b_k)]|0\rangle, \quad (11)$$

where ρ is the magnitude of the component of the electron coordinate perpendicular to z. This variational procedure is just a generalization of the well-known "product ansatz" of polaron theory[5] to the case in which a magnetic field is present. It is reassuring to note that second-order perturbation theory and the variational product-ansatz method agree in the limit of strong magnetic field.[6]

Comparison of (10) and (9) shows that the polaron correction to the transition energy changes sign as the magnetic field increases. To study this effect in detail it is necessary to resort to numerical calculation of E_1-E_0 for intermediate values of λ.

In the form given by (6) and (7) the numerical calculation of $C_0(\lambda)$ and $C_1(\lambda)$ for arbitrary λ would involve summing a very large number of two-dimensional integrals. However, we can reduce the problem to calculating only three two-dimensional integrals by the following method. Consider the expression for $C_1(\lambda)$ from (7). For all $n\geq 1$ we can write

$$\frac{1}{2k+(n-1)\lambda^2+k_z^2}=\int_0^\infty dt\exp\{-[2k+(n-1)\lambda^2+k_z^2]t\}.$$

Hence

$$C_1(\lambda)=-\frac{4\pi}{S}\mathcal{P}\sum_k\left[\frac{\exp[-(k_\perp/\lambda)^2]}{k}\left(\frac{k_\perp}{\lambda}\right)^2\times\frac{1}{2k-\lambda^2+k_z^2}\right]+D(\lambda), \quad (12)$$

where

$$D(\lambda)=-\frac{4\pi}{S}\int_0^\infty dt\sum_k\left[\frac{\exp[-(k_\perp/\lambda)^2]}{k}\times\{\exp[-(2k+k_z^2)t]\}\sum_{n=1}\frac{(n-k_\perp^2/\lambda^2)^2}{n!}\xi^{n-1}\right],$$

$$\xi=(k_\perp^2/\lambda^2)e^{-\lambda^2 t}.$$

[5] H. Fröhlich, *Advances in Physics* (Taylor and Francis, Ltd., London, 1954), Vol. 3, p. 325. The product ansatz discussion begins on p. 345.

[6] Strictly speaking, we must introduce a cutoff for the Hamiltonian at the Debye wave number in order to define the ground-state energy of the piezoelectric polaron. However, for calculating the energy difference between the $n=1$ and $n=0$ level we can ignore the cutoff.

Performing the n summation in (12), we obtain

$$D(\lambda)=-\frac{4\pi}{S}\int_0^\infty dt\sum_k\left[\frac{\exp\{-[(k_\perp/\lambda)^2+2kt+k_z^2t]\}}{k}\times\{[1+\xi-2k_\perp^2/\lambda^2+(k_\perp^2/\lambda^2)^2(1/\xi)]e^\xi-(k_\perp^2/\lambda^2)^2(1/\xi)\}\right].$$

Similar manipulations for $C_0(\lambda)$ give

$$C_0(\lambda)=-\frac{4\pi}{S}\int_0^\infty dt\sum_k\left[\frac{\exp\{-[k_\perp^2/\lambda^2+2kt+k_z^2t]\}}{k}e^\xi\right].$$

Changing from summation to integration over k space, using variables of integration u and τ given by

$$u=k/\lambda,\quad \tau=\lambda^2t,$$

and integrating over u, we obtain

$$D(\lambda)-C_0(\lambda)=-\frac{4\lambda^4}{\pi}\int_0^\infty\frac{d\tau}{\tau^4}\sinh^2\left(\frac{\tau}{2}\right)\times\int_0^1 dx(1-x^2)a^4E(a)+\frac{\lambda^4}{\pi}\int_0^\infty\frac{d\tau}{\tau^4}e^\tau\times\int_0^1 dx(1-x^2)b^4E(b),\quad(13)$$

where

$$E(y)=(y^2+1)-(3+2y^2)ye^{y^2}\int_y^\infty e^{-v^2}dv,$$
$$a=\tau/\{\lambda[(1-e^{-\tau})-(1-e^{-\tau}-\tau)x^2]^{1/2}\},$$
$$b=\tau/\{\lambda[1-(1-\tau)x^2]^{1/2}\}.$$

The function $e^{y^2}\int_y^\infty e^{-v^2}dv$ is tabulated in Rosser[7] for $y\leq 6.3$ and an asymptotic expansion for $y\gg 1$ is available. Thus the line shift $\alpha S(\lambda)$ is given in a form relatively convenient for numerical integration by

$$\alpha S(\lambda)=\alpha[C_1(\lambda)-C_0(\lambda)]=\alpha[D(\lambda)-C_0(\lambda)]-\frac{2\alpha}{\pi}\mathcal{P}\int_0^\infty du\,u^3\int_0^1 dx(1-x^2)\times\frac{\exp\{-[u^2(1-x^2)]\}}{u^2x^2+2u/\lambda-1}.\quad(14)$$

The results for $S(\lambda)$ are plotted in Fig. 1.

The straight line appearing in Fig. 1 is a plot of the correction to the $n=1$ to $n=0$ transition energy predicted by the effective-mass theory (or by applying the method of the MH theory to the zero-temperature case).

[7] J. B. Rosser, *Theory and Application of* $\int_0^z e^{-x^2}dx$ *and* $\int_0^z e^{-py^2}dy\int_0^y e^{-x^2}dx$ (Mapleton House, Brooklyn, New York, 1948), p. 190.

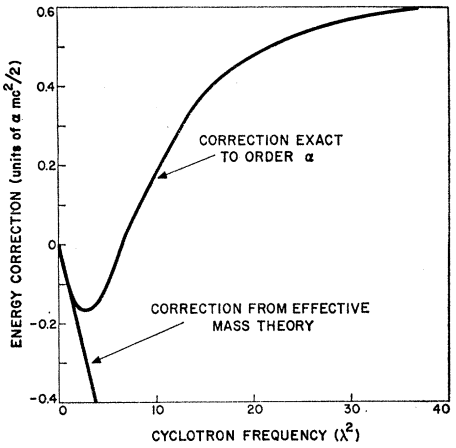

FIG. 1. Correction to the energy of the $n=0$ to $n=1$ cyclotron-resonance transition due to weak piezoelectric electron-phonon interaction at $T=0$.

Denoting this energy correction when $\alpha=1$ by $S_{\text{e.m.}}$, we obtain from (9)

$$S_{\text{e.m.}}=-(1/3\pi)\lambda^2.$$

Of particular interest is the sharp departure of $S(\lambda)$ from $S_{\text{e.m.}}$ when $\lambda^2>1$. When $\lambda^2=30$ (as in the BD experiment) the actual value of S is less than $\frac{1}{5}$ of the magnitude of the correction predicted by the effective-mass theory and of opposite sign.

A second quantity of interest is the linewidth of the $n=1$ level. We have evaluated numerically the golden-rule formula (15) for the transition probability per unit time γ out of state i into final states labeled by f.

$$\gamma=2\pi\sum_f|\mathcal{H}_{1fi}|^2\delta(E_f-E_i),\quad(15)$$

where i refers to an $n=1$, $p_z^2<1$ Landau level with no phonons present so that

$$|\mathcal{H}_{1fi}|^2=(4\pi\alpha/S)(1/k)(k_\perp/\lambda)^2\exp[-(k_\perp/\lambda)^2],\quad(16a)$$
$$E_f-E_i=-\lambda^2-2p_zk\chi+k^2\chi^2+2k\quad(16b)$$

with $\chi=\mathbf{p}\cdot\mathbf{k}/pk$. In the limit of infinite volume the sum on final states f becomes an integral over the vector momentum $\mathbf{k}$ of the emitted phonon. Taking $\frac{1}{2}\gamma$ as the $\frac{1}{2}$ width αW_P at $\frac{1}{2}$ height of the line and performing an integration we obtain

$$\alpha W_P=\frac{\alpha}{2}\int_{-1}^1 d\chi\frac{k\exp[-(k_\perp/\lambda)^2]}{1+k\chi^2-p_z\chi}\left(\frac{k_\perp}{\lambda}\right)^2,\quad(17)$$

where

$$k=\{-(1-p_z\chi)+[(1-p_z\chi)^2+\lambda^2\chi^2]^{1/2}\}/\chi^2\quad(18)$$

by conservation of energy. W_P is plotted in Fig. 2 for $p_z=0$. If we replace $\mathcal{H}_1$ in (3) by

$$(4\pi\alpha_D/S)^{1/2}\sum k^{1/2}(e^{-i\mathbf{k}\cdot\mathbf{r}}b_{\mathbf{k}}^\dagger+\text{H.c.}),$$

$\mathcal{H}$ becomes the usual model Hamiltonian for electron-phonon interaction via the deformation potential. The

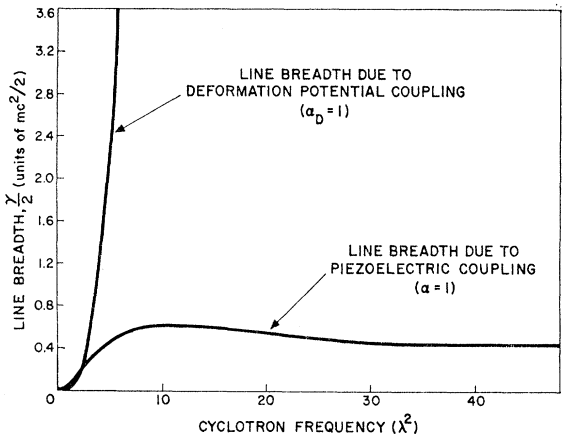

FIG. 2. Natural line breadth ($\frac{1}{2}$ width at $\frac{1}{2}$ height) of the $n=1$ to $n=0$ transition at $T=0$. W_P is represented by the lower curve, W_D by the steeply rising curve.

linewidth expression for deformation-potential coupling in this model is obtained by including an additional factor of k^2 inside the sum in (15). [Expression (16b) remains the same, of course.] The linewidth coefficient W_D obtained this way for deformation coupling is plotted in Fig. 2 to facilitate comparison with W_P.

We remark that the MH theory would predict zero linewidth at zero temperature.

Comparison of Fig. 1 and Fig. 2 indicates that at zero temperature the line shift due to piezoelectric coupling is approximately the same size as the intrinsic piezoelectric linewidth except at very large fields ($\lambda^2 \gg 1$) or very small fields ($\lambda^2 \ll 1$). However, unlike the piezoelectric linewidth, which saturates with increasing field, the line broadening due to deformation potential interaction grows like λ^2 and so eventually becomes dominant. At very small fields one is prevented from exploiting the fact that the line breadth from acoustic phonon emission falls to zero more rapidly than the line shift, by the fact that broadening from impurities and imperfections does not vanish with vanishing field. We conclude that it would be difficult to observe the effect of piezoelectric electron-phonon coupling on the cyclotron resonance frequency of photoexcited carriers in CdS or ZnO at temperatures well below 0.1°K.

III. VALIDITY OF SEMICLASSICAL THEORY

It is not surprising that a semiclassical theory of cyclotron resonance should fail to give the correct magnetic-field dependence of the line shift and line breadth at zero temperature. The more important question is whether we can expect such a theory to be reliable at the relatively low temperatures and high fields where, at present, well-defined cyclotron resonance lines can be observed experimentally in piezoelectric semiconductors. We proceed to study this question in the particular case of the BD experiment.

First, let us express the temperature in terms of the quantity T defined by

$$T=2k_B\tau/mc^2,$$

where k_B is Boltzmann's constant and τ is the absolute temperature. The results of the preceding section apply, presumably, when $T \ll 1$ and $T \ll \lambda^2$. (In CdS, $T=1$ at a temperature of about 0.1°K.) In the BD cyclotron-resonance experiment $T=11$, $\lambda^2=30$, so that a zero-temperature theory does not apply. However, assuming thermal equilibrium in the conduction band, the observed transitions were between the $n=1$ and $n=0$ Landau levels as would be true also at zero temperature. Is the semiclassical description of the electron motion in the MH theory appropriate under the foregoing experimental conditions?

To answer this question we must review briefly the MH theory.

In a uniform, time-independent magnetic field, the magnitude of the momentum p, of a classical charged particle remains constant in time. If we consider a group of such particles and assume they do not interact with each other, we find that in thermal equilibrium the probability of finding a particle with momentum p is proportional to

$$p^2 \exp(-p^2/2mk_B\tau). \tag{19}$$

The cyclotron frequency is $eH/m\tilde{c}$, independent of p.

In the MH theory, electrons in the conduction band of CdS behave like the classical particles described above, except that their mass is assumed to be a function of p. Thus the cylotron frequency, $\tilde{\omega}_c$, of an electron depends upon the value of p with which the electron moves according to

$$\tilde{\omega}_c=eH/m^*(p)\tilde{c}.$$

An immediate consequence is that electrons of different p have different resonance frequency in a given field, or for a given resonance frequency the number of electrons in resonance varies as a function of the field. This gives rise to the linewidth predicted by the MH theory, in view of the distribution of electrons given by (19). Most electrons will be in resonance when $\tilde{\omega}_c$ is close to $\tilde{\omega}_{0c}$, where

$$\tilde{\omega}_{0c}=eH/m^*((2mk_B\tau)^{1/2})\tilde{c}.$$

The shift in the cyclotron frequency is then

$$\Delta W=\tilde{\omega}_{0c}-eH/mc,$$

where m is the band mass of the electron.

To calculate $m^*(p)$ according to MH, one first calculates $E_c(p)$, which is the correction to the electron energy due to electron-phonon interaction in the

absence of external fields.[8] Then $m^*(p)$ is determined by

$$1/m^*(p)=1+(1/2p)\partial E_c(p)/\partial p=1+\zeta/p^3,$$

where ζ is independent of p, and proportional to T, according to MH.

We are interested in examining the validity of this theory for describing transitions between the $n=1$ and $n=0$ Landau levels. It is easy to show that the smallest mean-square momentum uncertainty that a wave packet constructed from the degenerate $n=0$ Landau states can have is given by

$$\min\langle\hat{p}_x^2+\hat{p}_y^2-\langle\hat{p}_x\rangle^2\rangle_0=\lambda^2/4,$$

where the brackets indicate expectation value in any eigenstate with eigenvalue $\lambda^2/2$. An identical calculation for wave packets constructed solely from the degenerate $n=1$ eigenstates gives an uncertainty in $p_\perp^2$ of $0.75\lambda^2$. Assigning to p_z^2 the typical value $T/2$ for thermal equilibrium, the total magnitude of momentum of a typical particle in the $n=1$ level must have an uncertainty of the order of

$$\Delta p=(T/2+0.75\lambda^2)^{1/2}-(T/2)^{1/2}.$$

Inserting $\lambda^2=30$, $T=11$ we find that Δp is almost as large as the total magnitude of momentum $T^{1/2}$ attributed to the electron in the semiclassical theory. More important, the uncertainty in $p_\perp$ gives rise to an uncertainty in the effective mass

$$\Delta\frac{1}{m^*(p)}=\frac{\zeta}{(T/2)^{3/2}}-\frac{\zeta}{(T/2+0.75\lambda^2)^{3/2}},$$

which is larger than the MH correction to the effective mass, $\zeta/(T)^{3/2}$. Therefore we conclude that the semiclassical MH theory does not give a consistent interpretation of the BD experiment.

We consider it probable, in view of the extreme smallness of the piezoelectric correction to the cyclotron resonance frequency at $T=0$, $\lambda^2=30$, that the MH theory overestimates the size of the frequency shift due to piezoelectric coupling under the conditions of the BD experiment.

Finally, we remark that since ζ is proportional to T in the MH theory, an important experimental check would be to measure the cyclotron frequency as a function of temperature. The MH theory predicts a $1/T^{1/2}$ dependence of the line shift.

IV. PERTURBATION THEORY AT FINITE TEMPERATURE

A more fundamental approach to the theoretical interpretation of the BD experiment would be to extend to finite temperature the perturbation theory described earlier. Unfortunately, this is not a straightforward procedure.

When $T\gg1$ and $\lambda^2>T$, the large majority of electrons are in states which correspond to unperturbed states with $p_z^2>1$. These unperturbed states behave in a very interesting fashion in the presence of piezoelectric electron-phonon interaction.

Let us examine, for example, the probability of decay by phonon emission of the $n=0$ Landau level with $p_z>1$, $T>0$. If we apply the "golden rule" expression (15) using

$$\begin{aligned}&|\mathcal{H}_{1fi}|^2=(4\pi\alpha/\mathcal{S})(1/k)e^{-(k_\perp/\lambda)^2}(1+n_k),\\&E_f-E_i=k(2-2p_z\chi+k\chi^2),\\&n_k=[\exp(2k/T)-1]^{-1},\end{aligned}\tag{20}$$

we obtain a divergent integral for the transition probability because of the rapid divergence of $|\mathcal{H}_{1fi}|^2$ as $k\to0$. This situation is remedied if we replace (15) by the more fundamental expression[9]

$$\eta=4\sum_f|\mathcal{H}_{1fi}|^2\sin^2(\omega_{fi}\tilde{\tau}/2)/\omega_{fi}^2,\tag{21}$$

where η is the probability that a phonon has been emitted at some time between 0 and $\tilde{\tau}$, $\omega_{fi}=E_f-E_i$ given in (16b), and $\tilde{\tau}$ is a dimensionless quantity proportional to the time, $\tilde{\tau}=(mc^2/2\hbar)t$. Unlike (15), expression (21) does not assume that the density of states and $|\mathcal{H}_{1fi}|^2$ are slowly varying. The sum on final states in (21) can be replaced by an integral over the wave vector $\mathbf{k}$ of the emitted phonon.

We wish to focus our attention on the behavior of (21) for small k. To this end we introduce a wave number k_0 such that $k_0\tilde{\tau}\gg1$, $k_0\ll1$, $k_0\ll T/2$ and break the sum in (21) into two parts. If we denote by η_1 that portion of the sum in (21) for which the emitted phonon has wave number greater than k_0, then

$$\begin{aligned}&\eta=\eta_1+\eta_2,\\&\eta_1=2\pi\tilde{\tau}\sum_f{}^{>k_0}|\mathcal{H}_{1fi}|^2\delta(\omega_{fi}),\\&\eta_2=4\sum_f{}^{<k_0}|\mathcal{H}_{1fi}|^2\sin^2(\omega_{fi}\tilde{\tau}/2)/\omega_{fi}^2,\end{aligned}\tag{22}$$

where $\sum_f{}^{>k_0}$ denotes that the sum is restricted to final states with emitted-phonon wave number greater than k_0; the meaning of $\sum_f{}^{<k_0}$ is obvious.

By virtue of the smallness of k in η_2 we can simplify the integrand, writing

$$\begin{aligned}&|\mathcal{H}_{1fi}|^2\cong(2\pi\alpha/\mathcal{S})(T/k^2),\\&\omega_{fi}\cong2k(1-p_z\chi),\end{aligned}\tag{23}$$

where χ is the cosine of the angle between the field direction and the direction of the wave vector of the emitted phonon. Changing the summation in (22) to

[8] The calculation of $E(p)$, done in MH by treating $\mathcal{H}_1$ of (3) as a perturbation on the free-electron eigenstates at $T>0$ and $p>1$, has been critized by Y. Osaka, J. Phys. Soc. Japan **19**, 2347 (1964) and by Mahan and Hopfield themselves in Ref. 1.

[9] See, for example, *Quantum Mechanics*, L. I. Schiff (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., p. 197.

TABLE I. Shift S_0 of $n=0$ to $n=1$ transition energy from its rigid-lattice value due to weak electron–longitudinal optical phonon coupling (energy shift in units of $\alpha_0\hbar\omega$).

ω_c/ω	0.1000	0.2000	0.3000	0.4000	0.5000	0.6000	0.7000	0.8000	0.9000
$S_0(\omega_c/\omega)$	−0.01830	−0.04057	−0.06823	−0.1035	−0.1502	−0.2153	−0.3138	−0.4851	−0.8903

integration we obtain

$$\eta_2=\frac{\alpha T}{\pi p_z}\int_0^{k_0}\frac{dk}{k}\int_{-2k(p_z-1)}^{2k(p_z+1)}\frac{\sin^2(\omega_{fi}\tilde{\tau}/2)}{\omega_{fi}^2}\,d\omega_{fi}$$
$$=\frac{\alpha T}{2\pi p_z}\tilde{\tau}\left\{\int_0^{(p_z+1)k_0\tilde{\tau}}\frac{dV}{V}\int_0^V dy\frac{\sin^2 y}{y^2}+\int_0^{(p_z-1)k_0\tilde{\tau}}\frac{dV}{V}\int_0^V dy\frac{\sin^2 y}{y^2}\right\}. \quad (24)$$

We can write

$$\int_0^V dy\frac{\sin^2 y}{y^2}=-\frac{\sin^2 V}{V}+\int_0^{2V}\frac{\sin y}{y}dy. \quad (25)$$

The integral on the right side of (25) is the well-known sine integral, which for large V approaches $\pi/2$. Therefore, for

$$(p_z-1)k_0\tilde{\tau}\gg 1,$$
$$\eta_2\sim(\alpha T/2p_z)\tilde{\tau}\ln\tilde{\tau}+\beta\tilde{\tau}, \quad (26)$$

where β is a constant independent of $\tilde{\tau}$. Of course a necessary condition for the validity of the perturbation theory is that $\tilde{\tau}$ be small enough so that $\eta\ll 1$.

We find that a decay of the form (26) results for any level with $p_z>1$, due to those phonon emission or absorption processes at finite temperature which do not involve a change in the magnetic quantum number, n.

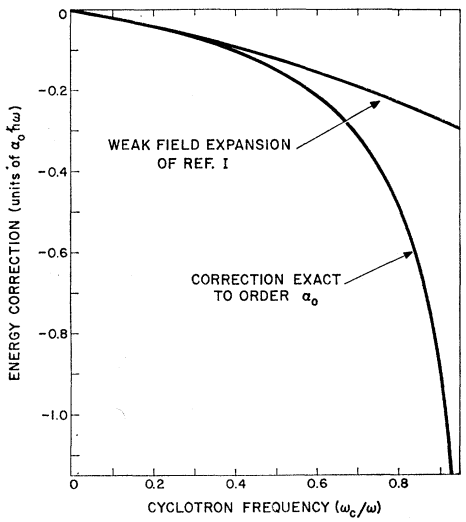

FIG. 3. Correction to the energy of the $n=0$ to $n=1$ cyclotron-resonance transition due to weak electron-longitudinal optical phonon coupling at zero temperature.

At the present time we do not know how to write down the correct expressions for the line shape and line shift for this kind of decay. These expressions are necessary before a complete theory of the cyclotron resonance of the piezoelectric polaron at finite temperature can be given.

V. OPTICAL POLARONS

The energy difference between the perturbed $n=1$ and $n=0$ levels for optical polarons (electrons coupled to longitudinal optical phonons) in the weak coupling approximation takes the form (in units of $\hbar\omega$, the longitudinal optical phonon energy)

$$\omega_c/\omega+\alpha_0 S_0(\omega_c/\omega), \quad (27)$$

where ω_c is the cyclotron frequency of electrons in the absence of electron-phonon interaction and α_0 is the dimensionless coupling constant introduced by Fröhlich.[5]

In I an expansion procedure, valid when $\omega_c/\omega\ll 1$, was found to give

$$S_0(\omega_c/\omega)=-[\tfrac{1}{6}+(3/20)(\omega_c/\omega)](\omega_c/\omega)+O[(\omega_c/\omega)^3]. \quad (28)$$

We have applied the Laplace transform technique already described to the optical-polaron perturbation expressions analogous to (6) and (7) [see (42) and (53) of I]. For optical polarons the perturbation expressions of I reduce to one-dimensional integrals, with $S_0(\omega_c/\omega)$ given by

$$S_0\left(\frac{\omega_c}{\omega}\right)=-2\left(\frac{\omega}{\omega_c\pi}\right)^{1/2}\int_0^\infty d\tau\left\{\frac{\exp(-\omega\tau/\omega_c)\sinh^2(\tau/2)}{z^2}\times\left[\frac{\tau^{1/2}}{y}-\ln\left(\frac{z+\tau^{1/2}}{y^{1/2}}\right)\right]\right\}, \quad (29)$$

where

$$z=(\tau+e^{-\tau}-1)^{1/2},$$
$$y=1-e^{-\tau}.$$

The integral in (29) has been evaluated numerically. Results are tabulated in Table I and are plotted in Fig. 3. Also plotted for comparison is the expansion of S_0 to order $(\omega_c/\omega)^2$ from (28).

We find that an approximation to $S_0(\omega_c/\omega)$ good to within 5% over the range $0\leq\omega_c/\omega<1$ is given by

$$\frac{1}{\alpha}\left[\frac{p}{2}\frac{\partial}{\partial p}E_{oc}(p)\right]_{p^2=\omega_c/\omega}=\tfrac{1}{2}[\sin^{-1}((\omega_c/\omega)^{1/2})/(\omega_c/\omega)^{1/2}-1/(1-\omega_c/\omega)^{1/2}], \quad (30)$$

where $E_{\infty}(p)=-\alpha \sin^{-1}p/p$, which is the energy analogous to (5) for optical polarons.[5]

As suggested by (30) and Fig. 3, the perturbation result diverges as $\omega_c/\omega \to 1$, which means that second-order Rayleigh-Schrödinger perturbation theory fails when $1-\omega_c/\omega$ is too small. If one applies instead the second-order Brillouin-Wigner perturbation theory one finds that for $\alpha_0 \ll 1$, in the limit $\omega_c/\omega \to 1$ with α_0 fixed, the energy separation between the $n=1$ and $n=0$ states becomes $1-(\alpha_0^2/4)^{1/3}+O(\alpha_0)$, which is a much more reasonable result.

The strong nonlinearity of $S_0(\omega_c/\omega)$ for $\omega_c/\omega>0.3$ suggests that polaron cyclotron-resonance experiments at very high fields would show relatively marked effects due to the electron-phonon coupling. For those materials and field strengths for which α_0 and $\alpha_0 S_0(\omega_c/\omega)$ are sufficiently small that perturbation theory applies, accurate cyclotron resonance frequency measurements at two known, large frequencies would enable one to determine independently the coupling constant α_0 and the band mass of the electron.

For many interesting materials, however, second-order perturbation theory is inadequate. Fortunately, the perturbation result of (29) can be improved upon by variational techniques to include the intermediate coupling case without significant restriction on the size of ω_c/ω. We intend to discuss one such variational technique in a future publication.

VI. DISCUSSION AND SUMMARY

We have shown in a simplified model how the energy of transition between the $n=1$ and $n=0$ Landau levels of a piezoelectric polaron varies with magnetic field. The effect of the piezoelectric electron-phonon interaction at weak fields is to decrease the energy separation between the first two Landau levels, but at stronger fields the separation increases relative to the separation in the absence of electron-phonon interaction. The strength of the magnetic field is measured by the ratio $\hbar\omega_c/mc^2$, strong fields corresponding to $\hbar\omega_c/mc^2 \gg 1$. In CdS, a typical piezoelectric semiconductor, $\hbar\omega_c/mc^2=1$ for conduction-band electrons at applied magnetic fields of approximately 270 Oe.

At zero temperature the intrinsic linewidth of the transition is approximately as large as the shift of the line center from its rigid lattice value except for very weak or very strong fields. However, for weak fields, broadening due to impurities and imperfections becomes important, while for very strong fields, broadening associated with spontaneous emission of acoustic phonons due to deformation-potential interaction may become dominant.

The results of this paper do not rule out the possibility that the line shift for strong fields may grow faster than the linewidth as the temperature increases from zero. This possibility must be realized if the results of the BD experiment are to be understood as a 15% shift in the transition energy relative to the rigid-lattice value due to piezoelectric electron-phonon interaction. However, the semiclassical theory of MH, which supports the above interpretation of the BD experiment, cannot be considered reliable under the experimental conditions of BD because the experiment was carried out in the quantum regime of strong fields and low temperatures.

We have extended the weak-coupling results of Paper I for the energy separation of the $n=1$ and $n=0$ Landau levels of optical polarons to stronger magnetic fields. The results show that relative to its rigid lattice value, the transition energy decreases markedly with increasing field. This effect may offer the possibility of experimentally observing for the first time an electron self-energy change due to the electron-optical phonon coupling.