force constants were found to fall off quite rapidly. This behavior indicates that the potential energy interaction function, $v(R_{ij})$, has a well-defined minimum at the nearest-neighbor separation and decreases rapidly and has an oscillatory behavior at the second- and third-neighbor positions. These conclusions are consistent with the theoretical analyses of Langer and Vosko[9] which strongly suggest that the interaction potential between a pair of ions in an electron gas would

have an oscillatory behavior as a function of separation distance.

A second application of the A-S model has been carried out on zirconium hydride and deuteride which have tetragonally deformed fluorite structures. We have concluded from this study that the hydrogen-hydrogen interaction is very weak compared to the hydrogen-zirconium interaction and that the six optical branches are nearly degenerate and have frequencies which are independent of the propagation vector to within 3%.

[9] J. S. Langer and S. H. Vosko, J. Phys. Chem. Solids **12**, 196 (1959).

Magnetization of Slow Electrons in a Polar Crystal

R. W. HELLWARTH
Hughes Research Laboratories, Malibu, California

P. M. PLATZMAN
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received April 23, 1962)

An approximate expression for the free energy of an electron which is coupled to a phonon field (a "polaron") and is in a uniform magnetic field is given. This expression is obtained by an extension of Feynman's path integral variational calculation of the polaron binding energy and is valid for all values of the field, the temperature and the electron-lattice coupling strength. The explicit form of the result is given in terms of a model system with an action functional different from that of the actual problem. A minimum-variational method is given for determining the best model to employ in calculations. We have evaluated the magnetic free energy at small magnetic fields using a model of a simple form. The two variable parameters of this model have already been optimized by Feynman and Schultz for Fröhlich's Hamiltonian and zero field. The result is applicable at all coupling strengths and carried to the first two terms in a Laurent series in the temperature. The first (inverse temperature) term yields an approximation to the effective polaron mass that varies, as the coupling is varied, by at most 1.5% from that calculated by Feynman and others. The second (temperature independent) term reflects the internal structure (atomic diamagnetism) of the polaron at large coupling and also the temperature variation of effective mass. The difference between our magnetic mass and that calculated by Feynman is shown to disappear when the model system is fully optimized.

I. FORMULATION AND METHOD OF APPROXIMATION

AN electron in the interior of an ionic crystal surrounds itself with a distorted lattice; a cloud of phonons. This system, commonly called a polaron, has been extensively studied theoretically.[1-6] We present here a calculation for the free energy of this coupled electron-phonon system in a uniform magnetic field. The results are obtained by an extension of Feynman's variational calculation of the polaron binding energy[7] and are applicable at all values of the magnetic field, the temperature and the electron-lattice coupling

strength. The susceptibility χ and other quantities of interest are easily obtainable from the free energy F. We will not attempt here to assess all those effects that would be present in a real material,[4] but rather treat the polaron as an idealized mathematical model. For numerical computations, we will specialize even further and use the Fröhlich model of the polaron[2] in order to compare our results with those of other theories.

To obtain the polaron free energy F as a function of the applied uniform magnetic field H (taken to be in the z direction), we use a path integral formulation equivalent to the $\mathrm{Tr}[\exp(-\beta\mathfrak{IC})]$. This allows the elimination of all phonon coordinates from the expression for F and results in an expression of the form.[1,8]

$$e^{-\beta F} = \sum_{\mathbf{X}(t)} \exp\{S[\mathbf{X}(t)]\}. \tag{1}$$

The symbol $\sum_{\mathbf{X}(t)}$ denotes the "path integral," or sum over all electron trajectories $\mathbf{X}(t)$ for t in the domain 0

[1] R. P. Feynman, Phys. Rev. **97**, 660 (1955); hereafter to be called I.
[2] H. Fröhlich, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1954), Vol. 3, p. 325.
[3] S. I. Pekar, Zhur. Eksp. i Teoret. Fiz. **19**, 796 (1949).
[4] T. D. Schultz, Phys. Rev. **116**, 526 (1960). References 2–4 contain a more complete bibliography of polaron studies.
[5] P. M. Platzman, Phys. Rev. **125**, 1961 (1962).
[6] M. Krivoglaz and S. I. Pekar, Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. **21**, 1, 16, 33 (1957).
[7] Y. Osaka, Progr. Theoret. Phys. Japan **22**, 437 (1959).
[8] R. P. Feynman, Phys. Rev. **84**, 108 (1951).

to β with the boundary condition $\mathbf{X}(0)=\mathbf{X}(\beta)$. The appropriate "action functional" S to employ is determined by the Hamiltonian chosen to describe the system.[1,4,7,8]

The Hamiltonian characterizing the interaction of a single nonrelativistic electron in a uniform magnetic field with a longitudinal phonon field is taken as

$$\mathcal{3C}=K_m+N+\mathcal{3C}_I, \qquad (2a)$$

where

$$K_m=(\mathbf{P}-Hx\hat{y})^2/2, \qquad (2b)$$

$$N=\sum_\mathbf{K}\omega_K a_\mathbf{K}^\dagger a_\mathbf{K}, \qquad (2c)$$

$$\mathcal{3C}_I=\sum_\mathbf{K} V^{-1/2}C_K{}^* a_\mathbf{K}^\dagger \exp(-i\mathbf{K}\cdot\mathbf{X})$$
$$+C_K a_\mathbf{K}\exp(+i\mathbf{K}\cdot\mathbf{X}). \qquad (2d)$$

The momentum of the electron is $\mathbf{P}$ and its coordinate is $\mathbf{X}$ with Cartesian components x, y, z. The operator $a_\mathbf{K}^\dagger$ is the creation operation for a phonon of wave vector $\mathbf{K}$. Our units are such that $\hbar$, the mass of the electron, and some convenient frequency ω, are set equal to unity.[9] The magnetic field H is measured in units of the ratio of the cyclotron frequency to the reference frequency ω.

The appropriate S to employ in (1) is[1,7,8,10]

$$S=-\int_0^\beta\left[\frac{1}{2}\left(\frac{d\mathbf{X}(t)}{dt}\right)^2+iHx(t)\dot{y}(t)\right]dt+\sum_\mathbf{K} V^{-1}|C_K|^2$$

$$\times\frac{1}{2}\int_0^\beta dt\int_0^\beta ds\,\exp\{i\mathbf{K}\cdot[\mathbf{X}(t)-\mathbf{X}(s)]\}$$

$$\times[\exp\{-\omega_K|t-s|\}+2P(\omega_K)\cosh\omega_K(t-s)], \qquad (3)$$

where $P(\omega_K)$ is the Planck function $[\exp(\beta\omega_K)-1]^{-1}$. The action S of Eq. (3) reduces to the one used in I if we set $H=0$, $\beta=\infty$, $\omega_K=1$ and $C_K=i2^{3/4}\alpha^{1/2}\pi^{1/2}/K$. This choice gives Fröhlich's Hamiltonian[2] for characterizing the interaction of an electron with the optical branch of a continuous ionic lattice.

The variational principal employed in I (and extended to finite temperates by Osaka[7]) was based on the reality of S when $H=0$. The addition of the magnetic field H contributes a term to S of the form $i(d\mathbf{X}/dt)\cdot\mathbf{A}(\mathbf{X})$, which destroys the explicit reality of S. In this case an estimate of the free energy by the method employed in I is no longer an upper bound and the great accuracy attained through this variational approach[4] might not be maintained when $H\neq0$. However, for small magnetic fields the accuracy of a perturbed value of F from an F calculated at $H=0$, by the minimal principle, might be expected to be quite satisfactory.

These same considerations arose in I in the calculation of the effective polaron mass, where the action to be used required the addition of a term $i\mathbf{U}\cdot d\mathbf{X}/dt$. However, for small velocities U, the effective mass obtained in I by essentially calculating $F(iU)$, $(\beta\to\infty)$, has been shown independently to be quite accurate.[10] Expressing matters in another way, we may consider $F(H)$ to be an analytic function of complex H. The variational principle of I still holds for H imaginary (S is then real) and the analytic continuation of this result to real H may be expected to be accurate for small H.

The method of I was to approximate (1) using

$$\sum_{\mathbf{X}(t)}\exp S\sim\exp\langle S-S_0\rangle\sum_{\mathbf{X}(t)}\exp(S_0), \qquad (4)$$

where the "average" of any functional A (denoted by $\langle A\rangle$) is defined by

$$\langle A\rangle\equiv\sum_{\mathbf{X}(t)}A[\mathbf{X}(t)]$$
$$\times\exp\{S_0[\mathbf{X}(t)]\}/\sum_{\mathbf{X}(t)}\exp\{S_0[\mathbf{X}(t)]\}. \qquad (5)$$

When S,S_0 are real, it is easily shown that the left-hand side of (4) is always larger than the right-hand side. This is the variational principle of I we have referred to. In the spirit of I and reference 10, we will compute $F(H)$ by using in (4) a trial action S_0 that is equal to S except that the part of S corresponding to the electron lattice interaction is replaced by a quadratic functional[1,8,10]

$$S_0=-\int_0^\beta\left[\frac{1}{2}\left(\frac{d\mathbf{X}(t)}{dt}\right)^2+iHx(t)\dot{y}(t)\right]dt$$

$$-\frac{1}{2}\gamma\int_0^\infty d\omega\,C(\omega)\int_0^\beta dt\int_0^\beta ds\,[\mathbf{X}(t)-\mathbf{X}(s)]^2$$

$$\times[e^{-\omega|t-s|}+2P(\omega)\cosh\omega(t-s)]. \qquad (6)$$

Because we have chosen an S_0 that is quadratic in $\mathbf{X}(t)$, the right-hand side of (4) can be evaluated exactly.[8]

One may think of the interaction term in S (at zero temperature) as indicating that at a time t the particle acts as though it were in a potential.

$$\sum_\mathbf{K} V^{-1}|C_K|^2\int_0^\infty ds\,\exp\{i\mathbf{K}\cdot[\mathbf{X}(t)-\mathbf{X}(s)]-\omega_K|t-s|\},$$

resulting from the electrostatic interaction of the electron with its mean charge density of its previous positions [the weight for different times and different modes being $\exp(-\omega_K|t-s|)$]. The assumption then is that such a potential may be roughly replaced by a superposition of parabolic potentials centered at the mean position of the electron in the past.[10]

[9] This frequency would be the phonon frequency in Fröhlich's one-frequency model of the polaron.

[10] R. P. Feynman, R. W. Hellwarth, C. K. Iddings, and P. M. Platzman, Phys. Rev. **127**, 1004 (1962).

II. CALCULATION OF F

To evaluate the RHS of (4), we will repeatedly make use of the formula (for one dimension)[8,11]

$$\sum_{x(t)} \exp\left[i\int_0^\beta x(t)f(t)dt - \frac{1}{2}\int_0^\beta dt \int_0^\beta ds \, x(t)x(s)a(t-s)\right]$$

$$= N \exp\left[-\frac{1}{2}\int_0^\beta dt \int_0^\beta ds \, f(t)f(s)a^{-1}(t-s)\right.$$

$$\left. -\frac{1}{2}\sum_n \ln A_n\right]. \quad (7)$$

This expression is true if $x(0)=x(\beta)$, $a(0)=a(\beta)$, and $a(t-s)$ is an operator with positive nonzero eigenvalues A_n, hence with a well-defined inverse $a^{-1}(t-s)$. The operator S_0 is of such a form. The $\dot{x}(t)^2$ term is obtained by using an $a(t-s)$ of the form $\sum_n \delta''(t-s-n\beta)$, the inverse of which may be obtained by considering it to be the limiting case of a zero frequency harmonic oscillator. The normalization factor N is independent of the operator $a(t-s)$ and $f(t)$ and will never appear in a calculation of that part of the free energy which depends on the magnetic field H or the coupling constants C_K.

Our approximate F, in analogy with I, will consist of three terms,

$$F = F_0 - B - A \quad (8)$$

which from (4) we write as

$$F_0 = -\beta^{-1} \ln \sum_{\mathbf{X}(t)} \exp S_0[\mathbf{X}(t)], \quad (9)$$

$$B = \frac{1}{2}\beta^{-1}\gamma \int_0^\beta dt \int_0^\beta ds \int_0^\infty C(\omega)d\omega \left[e^{-\omega|t-s|}\right.$$

$$\left. +2P(\omega)\cosh\omega(t-s)\right]\langle[\mathbf{X}(t)-\mathbf{X}(s)]^2\rangle, \quad (10)$$

$$A = \beta^{-1}V^{-1}\sum_K |C_K|^2 \int_0^\beta \int_0^\beta dt ds$$

$$\times\left[\exp-\omega_K|t-s|+2P(\omega_K)\cosh\omega_K(t-s)\right]$$

$$\times\langle\exp\{i\mathbf{K}\cdot[\mathbf{X}(t)-\mathbf{X}(s)]\}\rangle. \quad (11)$$

In order to calculate F_0, (the z components of all terms are unaltered by H and are the same as in reference 7), we sum over all functions $x(t)$ in (9) using (7) with $f(t)=H\dot{y}(t)$. Since we are using the trial action S_0, the eigenvalues A_n take on the particular values b_n. These quantities may be obtained most easily by Fourier transforming the kernel in (6). We find that

$$b_n = \omega_n^2 + 4\gamma \int_0^\infty d\omega \, C(\omega)\omega^{-1}[\omega_n^2/(\omega_n^2+\omega^2)], \quad (12)$$

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[11] B. Friedman, *Principles and Techniques of Applied Mathematics* (John Wiley & Sons, Inc., New York, 1956) Chap. 2, p. 107.

where $\omega_n \equiv 2\pi n/\beta$ and n takes all positive and negative integral values. There is no term for $n=0$ and so all subsequent sums over n are assumed to omit $n=0$.

After summing over $x(t)$ the action functional is still quadratic in $y(t)$ and (7) may be used again to perform the sum over $y(t)$. The resulting expression for F_0 is

$$\beta F_0 = \frac{3}{2}\sum_{n=-\infty}^{+\infty}\ln(b_n) + \frac{1}{2}\sum_{n=-\infty}^{+\infty}\ln(1+\omega_n^2 H^2/b_n^2). \quad (13)$$

The easiest way to obtain B is to differentiate (9) with respect to γ, for from (9), (10), and (5)

$$B = \gamma\partial F_0/\partial\gamma. \quad (14)$$

Therefore, by differentiating (13) we find that

$$B = \frac{3}{2}\gamma\beta^{-1}\sum_n \left[c_n/b_n - \frac{2}{3}\gamma\omega_n^2 H^2 c_n/(b_n^3+\omega_n^2 H^2 b_n)\right], \quad (15)$$

where $\gamma c_n \equiv b_n - \omega_n^2$. In order to compute the A term, we need to evaluate $\langle\exp\{i\mathbf{K}\cdot[\mathbf{X}(t)-\mathbf{X}(s)]\}\rangle$. Since this term adds only a linear driving force to the action S_0, all the path integrals are easily evaluated using (7):

$$\langle\exp\{i\mathbf{K}\cdot[\mathbf{X}(t)-\mathbf{X}(s)]\}\rangle$$

$$= \exp[-K^2 D + H^2(K_x^2+K_y^2)G]. \quad (16)$$

The functions D and G are functions of $t-s\equiv u$:

$$D(u) = \beta^{-1}\sum_n (1-\cos\omega_n u)/b_n, \quad (17)$$

and

$$G(u) = \beta^{-1}\sum_n \omega_n^2(1-\cos\omega_n u)/(b_n^3+\omega_n^2 H^2 b_n). \quad (18)$$

Equations (13), (15), (16), (17), (18) give our general approximation to F at all temperatures, magnetic fields and coupling constants. The most accurate result that could be obtained from our formulas would come from considering H to be pure imaginary so that F_0-B-A is an upper bound to F and then minimizing our result by varying the function $C(\omega)$. Although the solution of the full variational problem is not beyond the means of machine computation, here, we will use a simpler two-parameter function for $C(\omega)$ for which the best parameters have already been tabulated[1,4] for all coupling strengths (and $H=0$, $\beta=\infty$, and Frohlich's polaron). In Sec. IV, we will give evidence that the errors added by this simplification are small (for small H and β^{-1}).

III. EVALUATION OF F WITH A TWO-COUPLED MASS MODEL

We will use a b_n obtained from (12) by setting $\gamma=1$ and $C(\omega)=C\delta(\omega-w)$. This choice corresponds to the S_0 used in I and in references 4 and 7. In fact, it describes the properties of an electron coupled harmonically to a second particle of mass $M=4C/w^3$ with a spring constant $k=Mw^2$. For this model

$$b_n = \omega_n^2(\omega_n^2+v^2)/(\omega_n^2+w^2), \quad (19)$$

where

$$v^2 \equiv w^2 + 4C/w. \quad (20)$$

With the choice (19), all sums over n may be evaluated by using the expression for the inverse Fourier transform of a general denominator of the form $(\omega_n^2+a^2)$ ($n=0$ included here)

$$\beta^{-1}\sum_{n=-\infty}^{+\infty}\frac{\cos\omega_n u}{\omega_n^2+a^2}$$

$$=[\exp(-a|u|)+2P(a)\cosh au]/2a. \quad (21)$$

For example, the first sum in (13) may be done by differentiating it with respect to v, making a partial fraction decomposition, using (21), and integrating the result over v. This gives the zero field result which we will not reproduce here (see references 1, 4, 7). The second sum in (13) may be done by differentiating with respect to H, using (21) and then integrating the result with respect to H. This procedure requires, in general, a knowledge for arbitrary H of the roots of the cubic equation,

$$\omega(\omega^2+v^2)/(\omega^2+w^2)+i\omega H=0. \quad (22)$$

If we turn off the phonon coupling, the best value of v approaches w^1 and this equation becomes linear. For this case $A=B=0$, and an evaluation of (13) using (21) causes our approximation to give exactly the well-known free energy of a free electron in a magnetic field.[12] That is, in this limit

$$F_0=-\beta^{-1}\ln\{(2\pi\beta)^{-3/2}(H\beta/2)[\sinh(H\beta/2)]^{-1}\}. \quad (23)$$

At small magnetic fields the quadratic term in the magnetic field (in our units) is,

$$F_0=\beta H^2/24. \quad (24)$$

For small magnetic fields and finite phonon coupling, all sums of Eqs. (13)–(18) may be expanded to order H^2 and then evaluated as a power series in the temperature $T\equiv\beta^{-1}$, the leading term being a $1/T$ term corresponding to the usual "free-particle diamagnetism" such as exhibited in (24). We will evaluate this and the next term which is independent of temperature. The latter term may be thought of as arising from several sources which will be discussed in Sec. IV.

To evaluate F_0, we expand the logarithm in Eq. (13) to order H^2 and evaluate the sums using (21). If we retain only terms of the order of the inverse first power and the zeroth power of the temperature, the magnetic change in F_0 is

$$\delta F_0=(H^2/2)[(\beta/12)(w/v)^4$$
$$+(v^2-w^2)(v^2+3w^2)/4v^5]. \quad (25)$$

For extremely low temperatures, the first term dominates and is, in fact, just the expected effective mass correction for the model system since the total mass of the model is (v^2/w^2).

The second term is the temperature-independent diamagnetic piece for the model two-particle system. Using conventional methods, this term is "not easily" obtained. However, in the limit that the mass of the second particle becomes infinite the results are in agreement with a standard perturbation treatment of the isotropic three-dimensional oscillator.[13] In order to see this, we rewrite (25) in terms of the spring constant k and the various combinations of masses in the problem.

$$\delta F_0=H^2[\beta m_0^{-2}+3\mu(\mu+4/m_0)(\mu/k)^{1/2}]/24 \quad (26)$$

Here, $m_0=M+1$ the total mass of the model, M is the mass of the second particle, k is the spring constant and μ is the reduced mass of the system. In the limit of $M\rightarrow\infty$ the free-particle term (term proportional to β) vanishes and the temperature-independent piece becomes,

$$\delta F_0=\tfrac{1}{8}H^2 k^{-1/2} \quad (27)$$

In these units $\langle r^2\rangle_{\mathrm{av}}=\tfrac{3}{2}k^{-1/2}$, where $\langle r^2\rangle_{\mathrm{av}}$ is the expectation value of the electron coordinate in the ground state of the oscillator. Thus,

$$\delta F_0=H^2\langle r^2\rangle_{\mathrm{av}}/12 \quad (28)$$

which is the usual atomic temperature independent diamagnetic part.[13] For finite M, the problem does not simply break up into a center of mass mode and a relative mode, since the magnetic field couples only to a single particle. A more detailed discussion of the types of terms appearing in a perturbation treatment of the problem may be found in reference 14.

According to Eq. (14), we may obtain δB by differentiating Eq. (25). With our choice (19), (14) indicates that now $B=C\partial F_0(C,w)/\partial C$, and therefore

$$\delta B=-\tfrac{1}{2}H^2\frac{(v^2-w^2)}{v^3}\left[\frac{\beta w^4}{6v^3}+\frac{1}{8}\left(1+\frac{6w^2}{v^2}-\frac{15w^4}{v^4}\right)\right]. \quad (29)$$

To calculate the correction to the A term of (11) at small H, we need the function $G(u)$, of Eq. (20) to zeroth order in H^2 whence

$$G(u)\rightarrow\beta^{-1}\sum_n\frac{(1-\cos\omega_n u)}{\omega_n^4}\left(\frac{\omega_n^2+w^2}{\omega_n^2+v^2}\right)^3. \quad (30)$$

The summand may be expanded into partial fractions. Then using (21) all sums may be performed. The result to terms of order T^0 is

$$G(u)=(w/v)^6 u^2\beta/24+E(u), \quad (31)$$

where

$$E(u)=-u^3(w/v)^6/12+a_1 f(v,u)$$
$$+a_2 f'(v,u)+a_3 f''(v,u)+a_4 u. \quad (32)$$

In Eq. (32),

$$f(v,u)=(1-e^{-vu})/2v \quad (33)$$

[12] A. H. Wilson, *Theory of Metals* (Cambridge University Press, New York, 1934), p. 164.

[13] Van Vleck, *Electric and Magnetic Susceptibilities* (Oxford University Press, New York, 1952).

and the primed notation indicates differentiation with respect to v. The parameters a_1, a_2, a_3, and a_4 are expressed in terms of v and w:

$$a_1 = -3w^4(v^2-w^2)v^{-8} = -2a_4 \qquad (34a)$$

$$a_2 = \tfrac{3}{8}(v^2-w^2)^2(v^2+3w^2)v^{-7} \qquad (34b)$$

$$a_3 = \tfrac{1}{8}(w^2-v^2)^3v^{-6} \qquad (34c)$$

If we expand (16) to first order in H^2 and substitute it with Eq. (31) into Eq. (11) for A, we obtain a magnetic contribution δA to A:

$$\delta A = H^2 V^{-1} \sum_K |C_K|^2$$

$$\times \int_0^\beta du \ (k_x{}^2+k_y{}^2)G(u)\exp{-K^2D(u)} \quad (35)$$

Using (21) and (12), we find (to order $1/\beta$) that (17) gives

$$D(u) = \tfrac{1}{2}(w/v)^2[R(1-e^{-v|u|})+u-u^2/\beta], \quad (36)$$

where $R = (v^2-w^2)/(w^2v)$.

We now specialize to the case of the Frohlich Hamiltonian, i.e., we choose $C_K = i\alpha^{1/2}2^{3/4}\pi^{1/2}/K$ and $\omega_K = 1$. The volume is allowed to go to infinity and the sum on K becomes an integral. The integral on K is easily performed and the result for δA is,

$$\delta A = \tfrac{1}{3}\alpha\pi^{-1/2}2^{-3/2}H^2$$

$$\times \int_0^\beta (e^{-u}+2P_1\cosh u)G(u)D^{-3/2}du. \quad (37)$$

At large β, δA may be expanded in a Laurent series in β^{-1}. We will compute the first two terms, one proportional to β, and the next independent of β:

$$\delta A = \delta A^{(1)}+\delta A^{(2)}, \qquad (38)$$

where

$$\delta A^{(1)} = \tfrac{2}{3}\pi^{-1/2}\alpha(w/v)^3H^2\beta$$

$$\times \int_0^\infty u^2 e^{-u}[R(1-e^{-vu})+u]^{-3/2}du/24, \quad (39)$$

and

$$\delta A^{(2)} = \tfrac{2}{3}\pi^{-1/2}\alpha(v/w)^3H^2$$

$$\times \int_0^\infty \{E(u)e^{-u}[R(1-e^{-vu})+u]^{-3/2}$$

$$+\tfrac{1}{16}(w/v)^6u^4e^{-u}[R(1-e^{-vu})+u]^{-5/2}\}du. \quad (40)$$

The integral on the rhs of (39) is easily written in terms of an expression for the effective polaron mass m_F computed in I and (by another approach) in reference 11, and evaluated numerically[4] for all coupling strengths (with the two-parameter model we have employed).

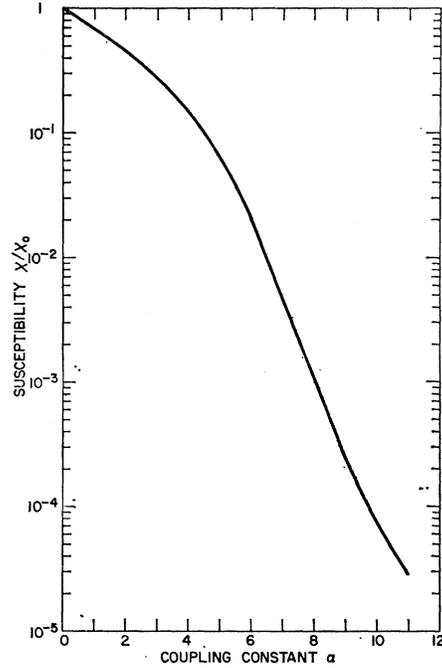
FIG. 1. Plot of leading low temperature term (proportional to β) in polaron susceptibility χ (in units of the free susceptibility χ_0) vs coupling strength parameter α.

This expression was

$$m_F = 1+\tfrac{1}{3}\pi^{-1/2}2^{3/2}\alpha\int_0^\infty u^2e^{-u}D^{-3/2}(u)du \qquad (41)$$

[with $\beta = \infty$ in $D(u)$].

The terms from (25), (29), and (40) are easily combined with (41) to give for the leading term proportional to β in the magnetic free energy

$$\delta F^{(1)} = \beta H^2(w/v)^6(3v^2/w^2-2m_F)/24. \qquad (42)$$

Using the best values of v, w and m_F computed elsewhere[1,4] we have plotted the ratio of the polaron susceptibility χ to that of the uncoupled electron χ_0 as a function of the coupling constant α in Fig. 1. $\chi/\chi_0 = \delta F^{(1)}(\beta H^2/24)^{-1}$.

The terms independent of the temperature are more complicated. We have evaluated them numerically using the same values of v, w computed in references (4) and (1), and plotted $\delta F^{(2)}$ vs α in Fig. 2. This correction ($\delta F^{(2)}$) is further altered by the changes in v, w occurring at small temperatures. We have estimated these corrections at small α and $\alpha = 3$ and found them to be negligible.

IV. DISCUSSION

It has been shown[14] that the complete effect of lattice interactions on the low temperature term proportional to β in the exact magnetic free energy (for small H) is to replace the free mass m by the "exact effective mass"

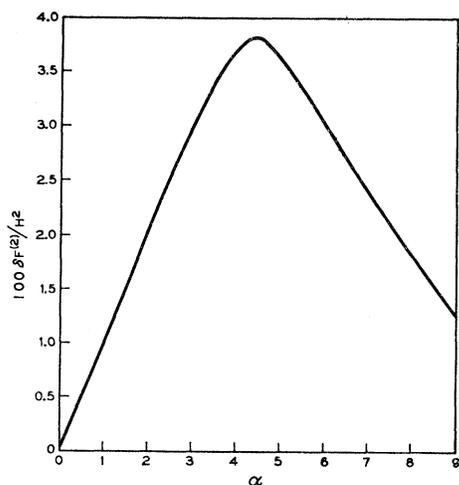[14] E. I. Blount, Phys. Rev. 126, 1636 (1962).

FIG. 2. Plot of the temperature independent correction to the low temperature free energy at small magnetic fields vs the coupling strength parameter α.

m^* in the free-particle expression (24) (which with $m \neq 1$ reads $H^2 \beta m^{-2}/24$). Therefore, our result (42) for $\delta F^{(1)}$ may be thought of as an approximate expression for the polaron effective mass. If we call this approximate "magnetic mass" m_H, and use the fact that the model mass m_0 equals v^2/w^2, we may rewrite (42) as

$$(m_H)^{-2} = (3m_0 - 2m_F)m_0^{-3} \tag{43}$$

It may be easily shown that our result would remain in this form if we had used a different model S_0.

Since our approach parallels closely that of Feynman et al.,[1,11] why do we not obtain a mass m_H equal to that (m_F) obtained by the two other calculations? Evidently we are using the same methods, but to evaluate a different function of m^*: i.e., m^{*-2} rather than m^* itself. However, if we had used the best model S_0 [having optimized the infinite set of parameters b_n in (13)–(18)] we would indeed have found m_H equals m_F if the latter had also been similarly recalculated with the improved $D(u)$ in (41). We may demonstrate this by recalling that in reference 11 it was shown that solving the full free energy variational problem (at zero temperature and magnetic field) yielded an optimum model S_0 which possessed the same response function (to an applied electric field pulse) as the polaron response

function calculated in the approximation which yields m_F; hence, m_0 approaches m_F upon full optimization of S_0, and from (43)

$$m_H \rightarrow m_F \rightarrow m_0 \tag{44}$$

as our method is pressed to its limit of accuracy.

Nevertheless, our cruder treatment which employs a two-parameter S_0 is quite accurate as it stands in (42). In this case Feynman has shown[1] that the best values of v, w cause m_0 to approach $1 + 4\alpha/27$ at small α and $(4\alpha^2/9\pi)^2$ at large α. In these limits, (42) gives

$$\begin{aligned} m_H &\rightarrow 1 + \alpha/6 && \text{for small } \alpha \\ &\rightarrow (4\alpha^2/9\pi)^2 && \text{for large } \alpha \end{aligned} \tag{45}$$

which are the same limits obeyed by m_F. Furthermore, by comparing Fig. 1 with Shultz' numerical results[4] for m_F at intermediate couplings, we find that m_H is at most about 1.5% lower than m_F at intermediate α, whereas m_0 varies as much as 16% from m_F. Of course, we expect m_F and m_H to be more accurate approximations to m^* than is m_0 because they are computed using corrections (from $\langle S - S_0 \rangle$) to the $\exp S_0$ term from which m_0 is derived: Suppose that m_0 is different by some small fraction ϵ from $m_F[m_0 = m_F(1 + \epsilon)]$. Then (43) gives $m_H = m_F[1 + 0(\epsilon^2)]$.

The first correction for higher temperatures $\delta F^{(2)}$ which is plotted in Fig. 2 is not easy to interpret precisely, it seems to arise from at least two effects. At small couplings, these corrections coincide with the corrections to the effective mass caused by the electron feeling a slightly different curvature at the bottom of its energy band. At high coupling strengths, we find the $\delta B^{(2)}$ nearly cancels $\delta A^{(2)}$ leaving $\delta F^{(2)} \sim \delta F_0^{(2)}$. This indicates that the two-mass model imitates the polaron well there, and since this model is atomic in character (i.e., a bound system with clearly separated excited states) the corrections in this region are atomic in character and are due to the internal structure of the polaron; that is, its long-lived excited states which occur in its own self-induced potential at strong enough couplings.

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