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Considerations on the Magnetic Field Problem in Superconducting Thin Films*

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We adapt the results obtained in a previous paper on the magnetic field dependence of the energy gap in superconductivity for bulk specimen to thin-film superconductors, using the model of discrete quantization in momentum space. Only the case of parallel and constant external magnetic field along the film surfaces is considered. A series of elementary theorems and some specific calculations lead to the conclusions: (1) A second-order phase transition should be observed at all temperatures for thin film thicknesses. (2) A simple scaling rule exists concerning the field and temperature dependence on the energy gap. (3) The critical field H_c depends on thickness L and reduced temperature t like $H_c \sim L^{-3/2}[\ln(1/t)]^{1/2}$ for not too thin films ($L \gtrsim 0.5\times10^{-5}$ cm). The behavior changes as the film becomes very thin or as the temperature becomes moderately low. A crude comparison with available experimental data seems to bear out our conclusions qualitatively.

1. INTRODUCTION

IN a previous paper[1] we derived expressions for the magnetic field dependence of the energy gap in superconductivity for bulk matter. The theoretical assumptions here are that we can somehow introduce a varying magnetic field into the bulk medium and that it makes sense to talk about energy gap depending on the field; of course, for a constant imposed field, the Meissner effects tells us that there is in fact no change in the gap due to the field. Though in some sense the discussion of bulk matter calculations is an academic problem, nevertheless it supplies a useful test model from which we can draw physically realistic conclusions about the case of superconducting thin films—which is directly amenable to experimental corroboration.

It was pointed out earlier[2] how we might expect to apply bulk material results to the actual experimental details of thin-film specimen. We can still introduce momentum pairs, though these are now quantized or discrete at least in one direction, and instead of integrating over momentum variable k (as for the case of infinite or bulk medium)—we have summation over momentum states. We are typically dealing with samples of thickness comparable or smaller than the penetration depth ($\sim5\times10^{-6}$ cm), such that it allows the magnetic field to penetrate the body without much attenuation, yet it is still large compared to the atomic scale in order not to alter drastically the basic dynamics of superconductivity. Thus, we shall not concern ourselves with the intrinsic change of properties due to finite thickness, but only the changes induced by the presence of the magnetic field relative to the field-free case.

Under such circumstances, the bulk material results can be applied by properly observing the discrete

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[1] Y. Nambu and S. F. Tuan, Phys. Rev. **128**, 2622 (1962), hereafter denoted I for reference purposes. See also Y. Nambu, Phys. Rev. **117**, 648 (1960). A brief account of the present work is given in Y. Nambu and S. F. Tuan, Phys. Rev. Letters **11**, 119 (1963).

[2] Y. Nambu and S. F. Tuan, in Proceedings of the Eighth International Conference on Low Temperature Physics, London, 1962 (to be published).

momentum for a thin film, but otherwise considering a simulated bulk material formed by a sequence of thin films (of thickness L) placed side by side in which the magnetic field runs parallel to the layers with alternating directions. The Fourier transform $A(q)$ of the vector potential across such superconducting film layers is of the form

$$A(q)\sim 1/q^2, \quad \text{where} \quad q=n\pi/L, \quad n=1, 2, 3, \cdots.$$

Thus, the lowest q is $=\pi/L$, which becomes large as L becomes small. The need for high q values (Pippard limit) is evident. In Sec. 2, this discrete quantization model is formulated for both the case of thick and thin films and the question of phase transition (energy or magnetic field versus gap ϕ diagrams) at $T=0°$K is briefly discussed.

Section 3 treats the perturbation calculation at $T=0$ for superconducting thin films in the framework of the discrete or lamination model. We shall find that it is essential to take into account the correct gauge and electronic boundary conditions to arrive at a result valid for very small gap or thickness.

In Sec. 4 we derive a formula [Eqs. (4.21) and (4.22)] at zero temperature which is based on the previous perturbation result but can be expected to hold for stronger magnetic fields. It shows the energy gap to decrease steadily with increasing field without ever vanishing until the whole calculation breaks down at extremely high fields. We cannot determine the critical field at zero temperature in our framework.

The above formula can be extended to finite temperatures without carrying out calculations in detail. We shall show in Sec. 5 that there exists an approximate scaling rule with respect to the temperature and magnetic field dependence of the gap, which enables one to treat the finite temperature case more easily than at zero temperature. We find a second-order phase transition at a finite critical field H_c, which depends on thickness L and reduced temperature as $L^{-3/2}(\ln 1/t)^{1/2}$ for not too thin films.

In Secs. 6 and 7 our results are compared with available experiments as well as with other theoretical calculations, notably by Bardeen,[3] by Douglass[4] in the Ginzburg-Landau-Gor'kov (G-L-G) theory,[5] and by Mathur *et al.*[6] The measurements of Tinkham and Morris[7] on thermal conductivity of lead and those of Douglass and Meservey[8] using the more direct tunnel approach on lead, do suggest that for film thicknesses in the range 500 to 1000 Å and reduced temperatures down to 0.12, a second-order phase transition is observed. This agrees with our predictions; moreover, the theoretical curves are in reasonable agreement with the data.

The concluding remarks of Sec. 8 evaluate the problems which confront the present study on thin films, in particular the questions of boundary conditions, a proper and more elaborate extension to finite temperatures, and the case of uneven or nonparallel magnetic fields. Suggestions are made which will test most critically the notions here outlined.

[3] J. Bardeen, Rev. Mod. Phys. **34**, 667 (1962).
[4] D. H. Douglass, Jr., Phys. Rev. Letters **6**, 346 (1961); Phys. Rev. **124**, 735 (1961).
[5] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **36**, 1918 (1959) [translation: Soviet Phys.—JETP **9**, 1364 (1959)].
[6] V. S. Mathur, N. Panchapakesan, and R. P. Saxena, Phys. Rev. Letters **9**, 374 (1962).
[7] D. E. Morris, Ph.D. thesis, 1962 (unpublished).
[8] D. H. Douglass, Jr., and L. M. Falicov, Progress in Low Temperature Physics (to be published); also private communications.

2. THE DISCRETE QUANTIZATION MODEL

We consider a parallel thin film of macroscopically large dimensions along Oy and Oz, and of thickness L along Ox [Fig. 1(a)]. Equal and parallel external magnetic fields H are applied in the plane of film surfaces at $x=0$ and $x=L$. With the boundary conditions $\psi_n(x,y,z)=0$ at $x=0$ and $x=L$, the single electron wave functions $\psi_n(x,y,z)$ are

$$\psi_n(x,y,z)=(2/L)^{1/2}\sin(n\pi x/L)\psi(y)\psi(z), \quad (2.1)$$

with

$$p_{nx}=n\pi/L>0, \quad n=1, 2, 3, \cdots, \quad (2.2)$$

and so our discrete jump in p_{nx} is $\Delta p_{nx}=\pi/L$, which is still much smaller than the Fermi momentum p_F for typical thin-film values of L (of order 100 to 1000 Å). Thus, we expect that the concept of a quasi-infinite medium should be applicable.

The Meissner effect relation[9] for the current j is

$$j_i(\mathbf{q})=\sum_{j=1}^{3} K_{ij}(\mathbf{q})A_j(\mathbf{q}),$$

with

$$K_{ij}(\mathbf{q})=-(ne^2/m)\delta_{ij}+K_{ij}^{(2)}(\mathbf{q}). \quad (2.3)$$

Here n is the number of conduction electrons, of both spin directions per unit volume. We are interested in the ϕ (energy gap) dependent part of $K(\mathbf{q})$, viz. $K^{(2)}(\mathbf{q})$;

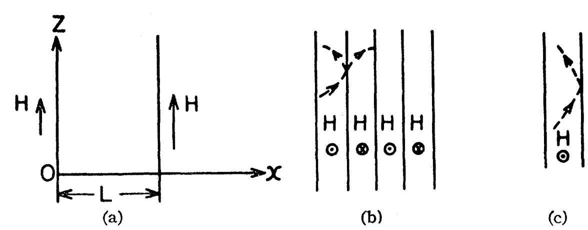


FIG. 1. (a) Thin film of thickness L along Ox, with equal and parallel magnetic fields at film surfaces. (b) Parallel thin films, side by side, with alternating fields H applied in the body of the film. (c) The mathematically equivalent single film, with magnetic field H and specular reflection at film boundaries.

[9] More precisely, Eq. (2.3) must be written

$$j_i(\mathbf{q})=\Sigma_{\mathbf{q}'}\Sigma_{j=1}^{3}K_{ij}(\mathbf{q},\mathbf{q}')A_j(\mathbf{q}').$$

Actually, only $\mathbf{q}'=\mathbf{q}$ gives a dominant contribution as we see from Eqs. (2.6) and (2.7) below. (All unexplained notations are the same as those in Ref. 1 throughout this paper.)

this is given to the second order in field strength by

$$\sum_k K_{ik}^{(2)}(\mathbf{q})A_k(\mathbf{q})=\sum_{\mathbf{p}}\frac{\langle\mathbf{p}|j_i|\mathbf{p}+\mathbf{q}\rangle\langle\mathbf{p}+\mathbf{q}|\mathbf{j}\cdot\mathbf{A}|\mathbf{p}\rangle}{E(\mathbf{p})+E(\mathbf{p}+\mathbf{q})}+\text{crossed terms.}\quad(2.4)$$

$E(p)=[\epsilon(p)^2+\phi^2]^{1/2}$ is the quasiparticle energy. It will be convenient at this stage to work in the London gauge. Since the induced current perpendicular to the plane of film is zero, the external vector potential in this gauge is then just $A(x,y,z)=(0,H(x-L/2),0)$. $A_y(x)$ is chosen to be an odd function with respect to the middle plane of the film. If we want the London equation in the simple form $j\propto A$, this is the proper choice since the induced current should also have the same symmetry when the magnetic field is equal on both sides of the film. However, an additive constant to $A(x)$ does not affect the final result (see below). Equation (2.4) then simplifies to (putting $K_{yy}\equiv K$)

$$K^{(2)}(\mathbf{q})|\mathbf{A}(\mathbf{q})|^2=\sum_{p,\pm}\frac{|\langle\mathbf{p}\pm\mathbf{q}|j_yA_y|\mathbf{p}\rangle|^2}{E(\mathbf{p})+E(\mathbf{p}\pm\mathbf{q})}.\quad(2.5)$$

Here the denominator $E(\mathbf{p}+\mathbf{q})+E(\mathbf{p})$ is just the energy of the quasiparticle pair (hole+particle) created in the intermediate state.

It is evident from Eq. (2.5) that we need to evaluate general matrix elements of the form $\langle\mathbf{p}'|(x-L/2)j_y|\mathbf{p}\rangle$. Using Eq. (2.1) we get

$$\begin{aligned}\langle\mathbf{p}'|(x-L/2)j_y|\mathbf{p}\rangle&=\frac{2p_y}{L}\int_0^L\sin(p_x'x)\sin(p_xx)(x-L/2)\\&\qquad\times dx\,\delta(p_y'-p_y)\delta(p_z'-p_z)\\&=-\frac{p_y}{L}\left\{\frac{1}{(p_x'-p_x)^2}-\frac{1}{(p_x'+p_x)^2}\right\}\{1-(-1)^{n'-n}\}\\&\qquad\times\delta(p_y'-p_y)\delta(p_z'-p_z).\quad(2.6)\end{aligned}$$

Comparison of Eqs. (2.5) and (2.6) gives, putting $p'=p+q$, the only nonvanishing $A_y(\mathbf{q})=A_y(q_x,0,0)$:

$$A_y(\mathbf{q})=\frac{-2H}{L}\left\{\frac{1}{q_x^2}-\frac{1}{(2p_x+q_x)^2}\right\}\approx-\frac{2H}{L}\frac{1}{q_x^2},\quad(2.7)$$

$$q_x=\pm(2r+1)\pi/L,\quad r=0,1,2,3,\cdots.$$

The term $q_x=0$, or $p=p'$ (no scattering) does not appear, and the approximate equality in Eq. (2.7) is generally satisfied for $(p_x+p_x')^2\gg(p_x-p_x')^2$. Note that for the smallest $|q_x|=|p_x'-p_x|=\pi/L$, $(p_x'+p_x)^2>9(p_x'-p_x)^2$.

We have here evaluated the matrix elements in the single electron picture. However, it is completely evident that the transition p to p' does not depend on the details of paired or unpaired states, but rather it is a special nature of the wave function and properties of Fourier transform in going from space variables to momentum variables. Information about the Cooper pairing is inherent in the kernel $K(\mathbf{q})$.

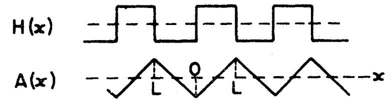


FIG. 2. The distribution of external field and vector potential as a function of distance along Ox for the laminated bulk medium.

Equation (2.7) is actually an expression for the external vector potential $\mathbf{A}^{\mathrm{ex}}(\mathbf{q})$. The physically interesting expression is that of the energy change $\mathcal{E}_A$ due to the magnetic field given by Eq. (5.1) of I

$$\mathcal{E}_A=-\tfrac{1}{2}\sum_{\mathbf{q}}\frac{K(\mathbf{q})}{1-K(\mathbf{q})/q^2}|\mathbf{A}^{\mathrm{ex}}(\mathbf{q})|^2\quad(2.8)$$

to the second order in $A^{\mathrm{ex}}(\mathbf{q})$. We see at once that the component $\mathbf{A}(\mathbf{q}=0)$ does not contribute because $K(0)\neq0$ and, hence,

$$K(q)/[1-K(q)/q^2]\rightarrow0$$

as $q^2\rightarrow0$. This means that the ambiguity of a constant additive term in $A(x)$ is effectively eliminated.

The discussion thus far is restricted to a single thin film. In order to establish connection with the bulk matter case, we prove the following theorem.

Theorem 1. Bulk medium, consisting of parallel layers of thin films placed side by side with alternating magnetic fields (uniform within its periodicity), is *mathematically* equivalent to a single thin film in a uniform magnetic field H, where the electrons confined in the film undergo specular reflection scattering at the film boundaries, except for the difference that in the latter the electron motion has discrete quantization across the film.

Figures 1(b) and 1(c) exhibit the mathematical equivalence in diagrams. An intuitive *physical* interpretation of the theorem is as follows. In the classical sense, an electron follows a curved path inside a film and then reflected at the surface. If we take the mirror image of the reflected motion with respect to the surface, the electron effectively passes into the next film without suffering reflection, but there the curvature is reversed, i.e., the magnetic field within the next film appears to be reversed. We remark that this picture is valid under the adiabatic condition where we neglect scattering of electrons at points of periodicity.

Proof. Figure 2 shows the distribution of external field and vector potential as a function of distance along Ox for the laminated bulk medium. It is evident that (writing $A_y\equiv A$)

$$A^{\mathrm{ex}}(x)=1/L\sum_{n=0}^{\infty}C_n\cos(n\pi x/L).\quad(2.9)$$

Since $A^{\mathrm{ex}}(x)$ is an even function, we have

$$C_n=(2/L)\int_0^L (x-L/2)\cos(n\pi x/L)dx$$
$$=-2/L\left(\frac{L}{n\pi}\right)^2[1-(-1)^n]. \qquad (2.10)$$

We see that the even terms vanish identically. The running wave form of the Fourier expansion of $A^{\mathrm{ex}}(x)$ is

$$A^{\mathrm{ex}}(x)=(1/L)\sum_{n=-\infty}^{\infty}(C_n/2)e^{in\pi x/L}. \qquad (2.11)$$

Thus, it is $C_n/2$ which is to be equated with the usual Fourier component $A(q_{nx})$ of $A^{\mathrm{ex}}(x)$. Hence,

$$A(q_{nx})=C_n/2=-2/L(L/n\pi)^2H,$$
$$n=2r+1,\ r=0,1,2,3,\cdots. \qquad (2.12)$$

Equation (2.12) is completely equivalent to Eq. (2.7), provided the term $(2p_x+q_x)^{-2}$ can be neglected, Q.E.D.

We have established the theorem under the assumption that H is constant within its periodicity; in fact the theorem holds for an arbitrary field $H(x)$,[10] as is physically desirable since for films of finite thickness the field inside is generally x-dependent and different from the surface field. The necessary modification requires us to replace $A^{\mathrm{ex}}(x)$ by a generalized function $\tilde{A}(x)$, such that

$$\tilde{A}(x)=(2/L)\sum_q A(q)\cos qx.$$

$\tilde{A}(x)$ is an even function of x, with periodicity $2L$, so that the general field $H(x)$ is odd (and hence alternating) but also of period $2L$.

The study of phase transition at $T=0°\mathrm{K}$ proceeds along the lines of an investigation of the total energy $\mathcal{E}$ versus energy gap parameter ϕ. Here $\mathcal{E}=\mathcal{E}_0+\mathcal{E}_A$, where $\mathcal{E}_0$ is the nonmagnetic BCS ground-state energy relative to the energy of the Fermi sea and $\mathcal{E}_A$ is the magnetic energy calculated from Eq. (2.8). It can be readily shown that $\mathcal{E}_0(\phi)$ in the weak-coupling approximation ($\phi\ll$the Debye frequency cutoff ω) is

$$\mathcal{E}_0(\phi)=N\phi^2\{\ln(2\omega/\phi)-\tfrac{1}{2}\}-N\rho\phi^2\{\ln(2\omega/\phi)\}^2. \qquad (2.13)$$

N is the density of Bloch states of one spin per unit energy at Fermi surface, and $\rho=N\bar{V}$ ($\bar{V}$ the effective electron-electron interaction). Using Eqs. (2.7) and (2.8), $\mathcal{E}_A$ can be written as

$$\mathcal{E}_A=\tfrac{1}{2}\left(\frac{2H}{L}\right)^2\sum_{q=\pm(2r+1)\pi/L}\left[\frac{1}{q^2}-\frac{1}{q^2+f(q)/\lambda^2}\right]. \qquad (2.14)$$

In Eq. (2.14) we have written, following I, $K(\mathbf{q})=(-1/\lambda^2)f(q)$, where λ is the London penetration depth at $T=0°\mathrm{K}$: $\lambda=(m/ne^2)^{1/2}$, and $f(q)$ describes the nonlocal nature of the relation between j and A with a characteristic bulk coherence length $\xi_0=v_F/\pi\phi$. In other words, f can be expanded as

$$f(q)=1-C(\xi_0q)^2+O[(\xi_0q)^4]. \qquad (2.15)$$

The function $f(q)$, in general, decreases with increasing q, and the sum in Eq. (2.14) converges rapidly. For a thick film such that

$$\lambda\pi/L\ll1 \quad\text{and}\quad \pi\xi_0/L\ll1,$$

the contribution predominantly comes from the first term in the summand only. Thus,

$$\mathcal{E}_A=\left(\frac{2H}{L}\right)^2\sum_{r=0}^{\infty}\left(\frac{L}{\pi}\right)^2\frac{1}{(2r+1)^2}=\tfrac{1}{2}H^2. \qquad (2.16)$$

Typically, $\lambda\sim5\times10^{-6}$ cm and $\xi_0\sim10^{-4}$ cm, so this result applies to thicknesses large compared to 10^{-4} cm.

For such thick films, we see that a first-order phase transition is expected for $\phi=\phi_c$ ($\neq0$) such that $\mathcal{E}_0(\phi_c)+\mathcal{E}_A(\phi_c)=0$ [since $\mathcal{E}_A(\text{normal})\sim0$], i.e.,

$$\tfrac{1}{2}H_c^2=-\mathcal{E}_0(\phi_c). \qquad (2.17)$$

Qualitatively, this type of behavior can be understood from a study of the $\mathcal{E}$ versus ϕ diagram shown in Fig. 3(a). $\mathcal{E}_A(\phi)$ is insensitive to ϕ for a thick film (London limit in bulk matter), so point C is the minimum position (little changed from zero-field value ϕ_0) for $\mathcal{E}$ and point O is the normal state position. As the external field H increases, point C rises above point O, so an abrupt change of phase from superconducting to normal state takes place at $\phi_c\sim\phi_0$.

For thin films such that $L/\lambda\pi\lesssim1$ or $L\lesssim10^{-5}$ cm, we have to use the Pippart form of kernel $K(q)$ given by[1]

$$K(q)=(-1/\lambda^2)f(q,\phi),$$
$$f(q,\phi)=\frac{3\pi^2\phi}{4qv_F}\left[1-\frac{16\phi}{\pi^2qv_F}\ln(qv_F/\phi)\right], \qquad (2.18)$$

which is valid for $q\xi_0\gtrsim1$. For small ϕ we may ignore the logarithm term, and approximate thus

$$\sum_q\frac{1}{q^2+f(q,\phi)/\lambda^2}=\sum_q\frac{1}{q^2}-\sum_q\frac{1}{q^4}\frac{3\pi^2\phi}{4\lambda^2qv_F}.$$

The general expression for $\mathcal{E}_A$, Eq. (2.14), becomes

$$\mathcal{E}_A\approx\frac{3\phi H^2}{\lambda^2v_F}\left(\frac{L}{\pi}\right)^3\sum_{r=0}^{\infty}\frac{1}{(2r+1)^5}. \qquad (2.19)$$

For small ϕ, the leading term of $\mathcal{E}_0(\phi)$ is of order $\phi^2(\ln\phi)^2$ and hence $|\mathcal{E}_0|<\mathcal{E}_A$ for sufficiently small ϕ. We expect on this basis that the $\mathcal{E}$ versus ϕ diagram will again be of the form that will yield a first-order phase transition [Fig. 3(b)] for a finite $\phi=\phi_c$ at $H=H_c$. This

[10] We wish to thank Professors H. Y. Fan and S. Gartenhaus for an interesting discussion on this point.

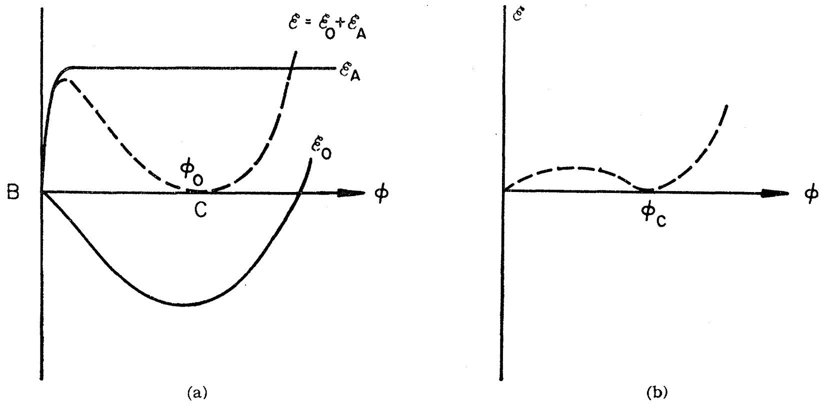


FIG. 3. (a) The energy versus energy gap ϕ diagram for a thick film; ϕ_c is the critical gap for a first-order phase transition. (b) The energy versus energy gap ϕ diagram for a thin film, according to Eq. (2.19).

is the conclusion reached by Bardeen in a recent paper.[3] *It is, however, misleading.* We shall presently prove the following:

Theorem 2. In a thin film which allows complete penetration of the magnetic field, the superconductive ground-state energy is lower than that of the normal state within the framework of diagonal quasiparticle approximation, the meaning of which is specified below. From this follows that no first-order phase transition from superconductive to normal state can be produced at least in this approximation.

The proof is simple. In I, a general approach was developed whereby one sets up the energy gap equation in the presence of an external field. It consists in first considering single electrons as under the influence of the external field and then performing the BCS type pairing in the presence of the field. This alternative was studied using the techniques of field theory in which Green's functions for quasiparticles in a magnetic field was constructed. When an expansion in A was made, it was shown there that the reduction of energy gap to the second order in A was equivalent to the variational result.

We now use this formalism without perturbation expansion. Let the single electron eigenstates and energies in the presence of the field A be labeled by n and ϵ_n. Since the film is thin, A is equal to the external field A^{ex}. We then introduce a pairing between n and its counterpart $\bar{n}$. We may take $\bar{n}$ to be the spin- and space-inversed state of n since the Hamiltonian as well as the boundary conditions remain invariant under spin inversion and $x-L/2\rightarrow -x+L/2$, $y\rightarrow -y$, $z\rightarrow -z$. (It is beyond the scope of this paper to discuss the relation between n and $\bar{n}$ under more general Hamiltonian and geometry.) The only difference from the free-field case is that instead of $\epsilon(p)$ and $\phi(p)$, we use ϵ_n and ϕ_n in setting up the BCS variational procedure.[11] In particular, the energy gap equation becomes

$$\phi_n=\sum_m \frac{\phi_m}{2E_m}V_{nm},\quad E_n=[\epsilon_n^2+|\phi_n|^2]^{1/2} \tag{2.20}$$

and the ground-state energy (relative to the normal state) is

$$\mathcal{E}=\sum_n\left(|\epsilon_n|-\frac{\epsilon_n^2}{E_n}\right)-\frac{1}{4}\sum_{n,m}\frac{\phi_n^*}{E_n}V_{nm}\frac{\phi_m}{E_m}=\sum_n\left\{|\epsilon_n|-\frac{\epsilon_n^2}{E_n}-\frac{1}{2}\frac{|\phi_n|^2}{E_n}\right\}, \tag{2.21}$$

which is <0 unless all $\phi_n=0$.

In the field theoretical treatment of quasiparticles, the energy gap parameter ϕ need not be diagonal in the states n which make the single-particle energy diagonal, but ϕ can be, in general, a matrix. But from the variational point of view, our estimation still gives an upper bound to the total energy $\mathcal{E}$. So as long as this diagonal quasiparticle approximation produces a superconductive state, its true energy is *a fortiori* lower than that of the normal state. This theorem can be extended in a straightforward fashion to finite temperature by replacing $\mathcal{E}$ with thermodynamic free energy.

Bardeen's result is not necessarily contradicted by the above theorem since the above method may not be able to give a superconductive solution when there is actually one in a different treatment such as the perturbation theory. This, however, is unreasonable because the diagonalization with respect to the magnetic eigenstates n will be more justifiable as the field increases.

In the following sections we shall find that a detailed analysis of the implication of our discrete quantization model on the form of the kernel $K(\mathbf{q})$ in perturbation theory actually leads to a different result from

[11] P. W. Anderson, J. Phys. Chem. Solids **11**, 26 (1959); see also *Proceedings of the Seventh International Conference of Low-Temperature Physics, 1960* (University of Toronto Press, Toronto, 1960), p. 298. It might appear that our approach does not allow for a spatially varying energy gap in the presence of the magnetic field, in contrast to the G-L-G theory. This, however, is not true. With our pairing scheme, the pair correlation function in the sense of G-L-G is given by $F(\mathbf{x},\mathbf{x}')=\sum_n(\phi_n/E_n)\psi_n(\mathbf{x})\psi_{\bar{n}}(\mathbf{x}')$, where ψ_n, $\psi_{\bar{n}}$ are the magnetic eigenfunctions. In the G-L-G approximation one defines from this the local gap parameter $\Delta(\mathbf{x})\propto F(\mathbf{x},\mathbf{x})$ which is actually x-dependent.

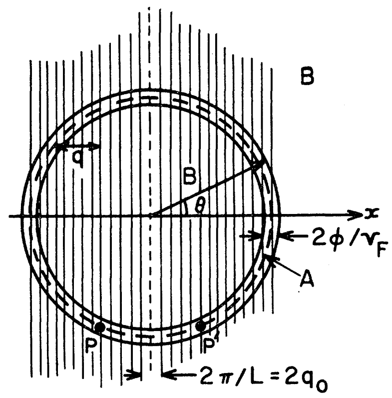


FIG. 4. Cross section of Fermi sphere, with discrete slabs separated by π/L along the x direction. The annulus region represent the BCS band of interaction of width $2\phi/v_F$.

Bardeen's and that the above theorem can be reinforced in the case of a separable potential.

3. PERTURBATION CALCULATION IN THIN FILMS

In this section we shall examine carefully the kernel $K(\mathbf{q})$ under the boundary conditions of a thin film elaborated in Sec. 2. The relation between $K(\mathbf{q})$ and the self-consistent self-energy in the field theoretical method has been established in I. The self-energy equation in a magnetic field reads, to the second order in A,

$$\phi=\Sigma(\phi,A),\quad \Sigma=\Sigma^{(0)}(\phi)+\Sigma^{(2)}(\phi,A). \tag{3.1}$$

Then

$$\begin{aligned}&\Sigma^{(0)}(\phi)\sim\phi\rho\ln(2\omega/\phi),\\ &\Sigma^{(2)}(\phi,A)\sim\textstyle\sum_{\mathbf{q}}[\partial K(q,\phi)/\partial\phi]|A(\mathbf{q})|^2.\end{aligned} \tag{3.2}$$

According to Eq. (2.18), $K(\mathbf{q})\sim\phi$, so that $\partial K/\partial\phi$ is independent of ϕ. This suggests the possibility that the expansion parameter of the perturbation theory for Σ for small ϕ is something like $|A|^2/\phi$ or $|A|^2/(\phi\ln\phi)$. If this is the case, obviously we cannot terminate our expansion at the second-order terms when ϕ becomes small, but we must include all higher order terms, the sum of which may exhibit a quite different analytic behavior.

The above situation is somewhat paradoxical. For a normal metal state we can stop the expansion at terms proportional to A^2, since in a small sample the electrons do not make circular orbits at such magnitudes of the field as are concerned here. We can start from electrons weakly perturbed by H, then introduce the pairing. It is difficult to imagine how this would introduce higher powers of $|A|^2/\phi$.

If we analyze the form of $K(q,\phi)$ for small ϕ, we find that the dominant contribution comes from the states where both of the intermediate particles are within the energy bandwidth 2ϕ (Fig. 4). This can be seen as follows. We have

$$K(q,\phi)\sim\sum_{\mathbf{p}}\frac{|\langle\mathbf{p}+\mathbf{q}|j|\mathbf{p}\rangle|^2}{E(\mathbf{p})+E(\mathbf{p}+\mathbf{q})}. \tag{3.3}$$

When both intermediate states are in the band, $E(\mathbf{p})+E(\mathbf{p}+\mathbf{q})\approx2\phi$; $|\langle j\rangle|^2$ is essentially constant, while the phase-space volume τ for a q (rather q_x) transition is restricted to

$$|\cos\theta|<2\phi/v_Fq=2/\pi\xi_0q\ll1$$

and $\Delta p<2\phi/v_F$:

$$\tau\approx4\pi p_F^2(2\phi/v_Fq)(2\phi/v_F).$$

Equation (3.3) then gives for the ϕ-dependent part of $K(q)$ a term proportional to

$$16\pi\phi^2p_F^2/2\phi q\sim\phi/q\sim1/\xi_0q,$$

which is precisely the leading term of the BCS kernel (2.18) for small ϕ.

With the discrete electron momenta for a thin film, however, this leading contribution vanishes. We have discrete states separated by $q_0=\pi/L$ along the x direction as shown in Fig. 4, so that for sufficiently thin films the thickness $2\phi/v_F$ of the BCS momentum band becomes small compared to the separation q_0. Since the electronic transition takes place horizontally between different states, there will be a certain critical film thickness below which no transition can take place between two points within the band, except for the special case mentioned below. Simple geometrical considerations show this condition to be

$$(\pi/L)^2=q_0^2\gtrsim m\phi/2=p_F/2\pi\xi_0. \tag{3.4}$$

Taking $\xi_0=10^{-4}$ cm, we have $L\lesssim0.8\times10^{-5}$ cm. (Actually, this thin-film condition will be relaxed later.)

The only situation where both points lie in the bandwidth is when the transition takes place between symmetrical points with respect to the origin O, for instance the p to p' transition shown in Fig. 4. We note, however, that this is forbidden since $q=2p=2r\pi/L$. In fact $K_{ij}{}^{(2)}(\mathbf{q})$ given by Eq. (3.22) of I gives in our case

$$K_{ij}{}^{(2)}(\mathbf{q})=\sum_{p_x}\frac{1}{(2\pi)^2}\frac{e^2}{m^2}\int\frac{p_ip_jdp_ydp_z}{E(\mathbf{p})+E(\mathbf{p}+\mathbf{q})}\times\left[1-\frac{\epsilon(\mathbf{p})\epsilon(\mathbf{p}+\mathbf{q})+\phi^2}{E(\mathbf{p})E(\mathbf{p}+\mathbf{q})}\right], \tag{3.5}$$

so for symmetrical transitions where $\epsilon(\mathbf{p})=\epsilon(\mathbf{p}+\mathbf{q})$, we have

$$1-\epsilon(\mathbf{p})\epsilon(\mathbf{p}+\mathbf{q})/[E(\mathbf{p})+E(\mathbf{p}+\mathbf{q})]=0.$$

We see, therefore, that for thin films the leading contribution comes from states where only one of p and p' lies in the band. It is also clear that $K_{ij}=0$ unless $i=j$, and moreover $i=j=y$ is the only physically relevant part in our gauge.

We need now to study and evaluate semiquantitatively the kernel $K_{ij}{}^{(2)}(\mathbf{q})$ or rather $\partial K_{ij}{}^{(2)}(\mathbf{q})/\partial\phi$, since ultimately we shall be concerned with a self-energy type compensation equation given by Eqs. (3.1) and

(3.2). For convenience of notation write $E(\mathbf{p})=E$ and $E(\mathbf{p}+\mathbf{q})=E(\mathbf{p}')=E'$, then

$$\partial K_{yy}^{(2)}(\mathbf{q})/\partial\phi=-\sum_{p_x}\frac{\phi}{(2\pi)^2}\frac{e^2}{m^2}\int\int\frac{p_y{}^2dp_ydp_z}{EE'(E+E')}\times\left[3-(\epsilon\epsilon'+\phi^2)\left(\frac{1}{E^2}+\frac{1}{E'^2}+\frac{1}{EE'}\right)\right]. \quad (3.6)$$

In Fig. 4, we have labeled the band region by A, and the regions outside by B. Since we have shown that contributions from E, E' belonging to A (ϵA) vanish for sufficiently thin-film slabs, it is evident that the leading contribution must come from transitions with $E\epsilon A$ (B) and $E'\epsilon B$(A). To fix our attention we consider the case where $E\epsilon A$, $E'\epsilon B$; for sufficiently small ϕ we have $E'\approx|\epsilon'|\gg E$. Since $|q|\ll p_F$,

$$|\epsilon'|=|(\mathbf{p}+\mathbf{q})^2/2m-\mathbf{p}^2/2m|\approx|q|v_F\cos\theta, \quad (3.7)$$

where θ is the polar angle shown in Fig. 4. Further,

$$3-(\epsilon\epsilon'+\phi^2)\left(\frac{1}{E^2}+\frac{1}{E'^2}+\frac{1}{EE'}\right)\approx 2$$

effectively since terms proportional to $\epsilon\epsilon'$ vanish upon integration within the narrow band where ϵ runs over both signs while ϵ' remains practically constant. Thus, we get

$$\begin{aligned}\partial K_{yy}^{(2)}(\mathbf{q})/\partial\phi&\approx-\frac{2\phi}{(2\pi)^2}\frac{e^2}{m^2}\sum_{p_x}\int\int\frac{p_y{}^2}{EE'(E+E')}dp_ydp_z\\&\approx-\frac{2\phi}{(2\pi)^2}\frac{e^2}{m^2}\sum_{p_x}\int\frac{p_y{}^2}{E\epsilon'^2}dp_ydp_z\\&\approx-\frac{4\phi}{(2\pi)^3}\frac{e^2}{m^2}\frac{\pi p_F{}^2}{q^2v_F{}^2}\int\int\frac{\tan^2\theta d\cos\theta}{E(l)}l^2dl, \quad (3.8)\end{aligned}$$

where the following crude approximations are understood: (a) The discrete p_x summation is replaced by an integration with cutoff $1\geqslant|\cos\theta|\geqslant\cos\theta_1$ since a large contribution comes from small values of $\cos\theta$, and (b) the l-integration is performed for fixed θ over $0\leqslant|E(l)|\leqslant|\epsilon'|=|qv_F\cos\theta|$ since we have assumed $l=p\epsilon A$, $p'\epsilon B$. A factor of 2 is included to account for the possibility $p\epsilon B$, $p'\epsilon A$. Finally we use Eqs. (3.21), (3.8), and (3.11) of I to obtain

$$\begin{aligned}\Sigma^{(2)}&=\rho\phi\sum_{\mathbf{q}}\int\int\frac{e^2|A(\mathbf{q})|^2\tan^2\theta}{4q^2}\frac{1}{E}d\epsilon d\cos\theta\\&=\rho\phi\sum_{\mathbf{q}}\int\frac{e^2|A(\mathbf{q})|^2}{2q^2}\sinh^{-1}|qv_F\cos\theta/\phi|\times\tan^2\theta d\cos\theta, \quad (3.9)\end{aligned}$$

and

$$\delta\phi=\Sigma^{(2)}/\rho. \quad (3.10)$$

The self-energy of a quasiparticle in general depends on the state one considers, and in particular it will show some anisotropy due to the quantization of p_x. Such an effect is disregarded here. Furthermore, we see that the summation over q in Eq. (3.9) is heavily weighted in favor of small values of q since $|A(\mathbf{q})|^2/q^2\propto 1/q^6$. Thus, it is practically sufficient to keep only the first term: $q=q_0=\pi/L$; the next term will be smaller by a factor $1/3^6\approx 1/600$. So in the following, we shall limit ourselves to the single term $q=q_0$.

With this in mind, the natural cutoff θ_1 in Eq. (3.9) would appear to be given by

$$\cos\theta_1=\pi/Lp_F, \quad (3.11)$$

since this corresponds to states with the smallest p_x. However, the physical situation requires a closer study of the conditions affecting the problem before deciding upon the appropriate cutoff. We will tentatively integrate Eq. (3.9) with an unspecified cutoff $\cos\theta_c\ll 1$ and obtain

$$\delta\phi=-\phi\frac{2e^2}{q_0{}^2}\sinh^{-1}(q_0v_F\cos\theta_c/\phi)\frac{1}{\cos\theta_c}|A(q_0)|^2,\quad q_0=\pi/L. \quad (3.12)$$

A factor of 2 arises when one sums over $q_x=\pm q_0$. This result is free from the objection raised against the earlier formula (3.2). In fact $\delta\phi$ is proportional to

$$\phi\sinh^{-1}(q_0v_F\cos\theta_c/\phi)\sim\phi\ln(2q_0v_F\cos\theta_c/\phi), \quad (3.13)$$

since the argument of $\sinh^{-1}$ is $\gtrsim 1$ in view of our definitions (3.4) and (3.11). So $\delta\phi$ has the same type of ϕ dependence ($\sim\phi\ln\phi$) as $\Sigma^{(0)}(\phi)$ given in Eq. (3.2).

Although Eqs. (3.9) and (3.12) have been obtained under the thin-film condition (3.4), it appears to be valid also for thicker films. In the latter case, there will be contributions to the kernel $K(q)$ coming from transitions between states within the band A, and one expects the result to agree with the old formula (2.18). This is in fact so except for a minor difference. Observing that $\sinh^{-1}(qv_F\cos\theta_c/\phi)\sim qv_F\cos\theta_c/\phi$ in the "thick" case, we find from Eq. (3.9) that

$$\delta\phi=-\sum_{\mathbf{q}}\frac{e^2}{q}v_F|A(\mathbf{q})|^2, \quad (3.14)$$

which is to be compared with the leading term in Eq. (3.25) of I

$$\delta\phi=-\frac{\pi^2}{8}\sum_{\mathbf{q}}\frac{e^2v_F}{q}\left[1-\frac{16\phi}{\pi^2qv_F}\{2\ln(\pi q\xi_0)-1\}\right]\times|A(\mathbf{q})|^2. \quad (3.15)$$

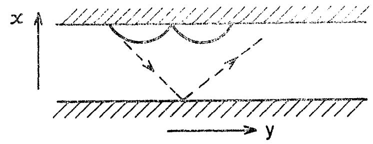


FIG. 5. Situations whereby the electrons are localized to one side of the film.

The correction factor $\pi^2/8=1.24$ may be related to the geometry.

We may therefore regard Eq. (3.12) to be valid irrespective of the restriction (3.4) as long as the thickness is small compared to the penetration depth. In spite of this reasonable behavior of our result we have not yet solved the problem. We need a more critical examination from a nonperturbative point of view before coming to the discussion of phase transition.

4. EXTENSION OF THE PERTURBATION FORMULA

It was remarked earlier that there is in principle no need for perturbation expansion with respect to the magnetic field. Within the framework of diagonal quasiparticle approximation defined there, an exact solution can be obtained by pairing appropriate electron eigenstates in the presence of the magnetic field H.

As was originally assumed in the BCS theory and also adopted in other calculations, let us suppose the interaction to be separable. For its general form take

$$V_{p'p}=\langle p'\bar{p}'|V|p\bar{p}\rangle=S_{p'}S_p^*V_0,$$

where p and $\bar{p}=-p$ are the paired momenta. The energy gap equation takes the form

$$\begin{aligned}1&=\sum_p\frac{V_p}{2E_p}\equiv V_0\sum_p\frac{|S_p|^2}{2E_p},\\ E_p&=[\epsilon_p^2+|\phi_p|^2]^{1/2},\\ \phi_p&=S_p\phi.\end{aligned}\tag{4.1}$$

In particular, $|S_p|=1$ if $V_{pp'}$ is constant within a domain D and zero otherwise. In the presence of the field, we obtain the same equation if we label the states by n instead of p, namely

$$\begin{aligned}1&=\sum_n\frac{V_n}{2E_n}=V_0\sum\frac{|S_n|^2}{2E_n},\\ E_n&=[\epsilon_n^2+|\phi_n|^2]^{1/2},\quad \phi_n=S_n\phi\\ S_n&=\sum_p u_{np}u_{\bar{n}\bar{p}}S_p,\quad u_{np}=\langle n|p\rangle.\end{aligned}\tag{4.2}$$

This may be cast into

$$\begin{aligned}1&=\bar{V}\sum_n\frac{1}{2E_n'},\\ E_n'&=[\epsilon_n^2+\bar{\phi}^2]^{1/2},\end{aligned}\tag{4.2'}$$

by introducing the averages $\bar{V}$ and $\bar{\phi}$. The magnetic field dependence of $\bar{\phi}$ will come from ϵ_n and $\bar{V}$. Let us first consider ϵ_n.

For most of the electrons near the Fermi surface, the magnetic energy is small compared to the kinetic energy, and perturbation theory should be adequate since the classical electron orbits do not deviate much from straight lines. We may still label the states by the unperturbed momenta p, and $\epsilon(H)$ takes the form

$$\epsilon(p,H)=\epsilon(p,0)+e^2H^2L^2(1+p_y^2/p_x^2)/24m,\tag{4.3}$$

as can be seen using the WKB method. This represents the diamagnetic energy increase for electrons confined to within the film. Equation (4.3) is not applicable when $|p_y/p_x|$ becomes very large. This is because these electrons run nearly parallel to the film and get localized to one side of the film by the magnetic field (Fig. 5). The geometrical condition for this to happen is

$$2eHLp_y/p_x^2>1.$$

Those "boundary electrons," however, would not contribute to the pairing energy of superconductivity since we have paired space-reversed states which are now separated to different sides of the film. We shall see this later explicitly.

Equation (4.3) represents an expansion and distortion of the Fermi surface. We must recall here that ϵ must be measured relative to a chemical potential so as to keep the average $\bar{\epsilon}=0$. Hence, only the distortion will affect Eq. (4.1), and this is a fourth-order effect on the effective density of states N at the Fermi surface. The entire picture breaks down only when H is so strong as to make the radius of curvature comparable with the thickness. For $L\sim10^{-5}$ cm, we get $H=p_F/eL\sim10^6$ G.

We see, therefore, that the main effect of the field arises through the change of the matrix element of V, and in going back to Eq. (4.2), we come to the following interesting assertion:

Theorem 3. Under the assumption of separable potential and essentially continuous single-particle energy spectrum, the energy gap in the presence of the field is reduced, but superconductivity is never broken unless $S_n=0$ for all states n near the Fermi surface (except for points of measure zero). In the latter case, the minimum energy gap defined by $\mathrm{Min}_n\{E_n\}$ is zero, but a superconductive solution to Eq. (4.2) may still exist ($\phi_n=S_n\phi$, $\phi\neq0$).

The proof will be rather obvious. When $S_n=0$ for those states with $\epsilon_n=0$ (Fermi surface), Eq. (4.2) does not become singular as $\phi\rightarrow0$ since those states do not contribute, so there may or may not be a superconductive solution. On the other hand, if $S_n\neq0$ in some portion of the Fermi surface, one can always find a solution by making ϕ sufficiently small unless the discreteness of ϵ_n becomes important. The minimum energy gap can still be zero. (This observation brings in the necessity of distinguishing the vanishing of energy gap from the vanishing of "superconductive" state and consequent phase transition to the normal state. A "superconductive" state with a vanishing gap will have different physical properties from the ordinary one. In this paper a superconductive state is meant to be a state with $\phi_n \neq 0$ for *some n*.)

Under a condition like our magnetic field problem, it would actually be difficult to realize the special case of theorem 3 since the system is anisotropic, which means that it would be difficult to destroy superconductivity completely by a magnetic field. It is true that the actual interaction does not exactly have the property assumed, so the problem of critical field may depend on the details of interaction. It is also true that even though S_n of theorem 3 may not exactly vanish, it may become so much reduced everywhere that the gap vanishes for practical purposes.

At any rate, it is clear that the effect of the magnetic field, whatever the dependence, may be regarded as a change in the effective coupling parameter $\rho = N\bar{V}$. Namely, ρ will now become a function of H and $\bar{\phi}$. (It depends on $\bar{\phi}$ because $\bar{V}$ depends on the weight function $1/E_n'$ which involves $\bar{\phi}$.) Consequently, we may write the solution to Eq. (4.2′) as

$$\bar{\phi} = 2\omega \exp[-1/\rho(H,\bar{\phi})], \tag{4.4}$$

which is a transcendental equation. If ρ decreases smoothly to zero for some H, $\bar{\phi}$ vanishes at this point and we shall have a second-order transition to the normal state in view of theorem 2.

Setting up Eq. (4.2) and solving it in the magnetic field is an involved task. Furthermore, there is not much sense in doing it since the real potential will be more complicated. We can, however, compare our perturbation formula (3.12) with Eq. (4.4) and thereby identify $\rho(H,\bar{\phi})$. This is legitimate for weak fields, but it seems reasonable to expect that Eq. (4.4) has a larger domain of validity than the simple perturbation formula.

In order to carry out this program, let us take a differential of Eq. (4.2)

$$0 = -\sum_n V_n \frac{\phi_n \delta\phi_n}{2E_n^3} + \sum \frac{1}{2E_n}\delta V_n. \tag{4.5}$$

By the standard technique, the first sum reduces to

$$\begin{aligned} &\approx -\bar{\phi}\delta\bar{\phi} \sum_n \frac{\bar{V}}{2E_n'^3} = -\bar{\phi}\delta\bar{\phi}(\rho/\bar{\phi}^2) \\ &\qquad = -\rho\delta\bar{\phi}/\bar{\phi}. \end{aligned} \tag{4.6}$$

Comparing this with Eq. (3.9) we immediately obtain

$$\begin{aligned} &\sum_n \frac{1}{2E_n}\delta V_n \\ &\quad = -\rho \sum_q \iint \frac{1}{2E} \frac{e^2|A(\mathbf{q})|^2}{q^2} \tan^2\theta d\epsilon d\Omega/4\pi \\ &\quad \approx -\rho \int \frac{d\Omega}{4\pi} \int_{-\kappa}^{\kappa} d\epsilon \frac{\tan^2\theta}{2E} \frac{2e^2|A(q_0)|^2}{q_0^2}, \end{aligned} \tag{4.7}$$

where $\kappa = q_0 v_F \cos\theta_c$. Hence, from Eq. (4.2) we find

$$\begin{aligned} 1-|S_n|^2 = -\delta V_n/V_0 &= \frac{2e^2|A(q_0)|^2}{q_0^2} \tan^2\theta f_\kappa(\epsilon), \\ f_\kappa(\epsilon) &= 1, \quad |\epsilon| < \kappa, \\ &= 0, \quad |\epsilon| > \kappa. \end{aligned} \tag{4.8}$$

Here V_0 is the potential for the field-free case ($V_{p'p} = V_0$ if p, $p' \epsilon D$, and zero otherwise).

Since $|S_n|^2 \geqslant 0$, the perturbation result certainly loses its meaning if the right-hand side of (4.8) exceeds 1. We must then choose a cutoff θ_2 according to

$$\alpha \equiv 2e^2|A(q_0)|^2/q_0^2 = \cot^2\theta_2,$$

or

$$\cos^2\theta_2 = \alpha/(1+\alpha). \tag{4.9}$$

We have ignored here the complication arising from $f_\kappa(\epsilon)$. Equation (4.9) is similar to Eq. (3.11) but more stringent. Beyond this angle up to θ, we shall set simply

$$1-|S_n|^2 = f_\kappa(\epsilon). \tag{4.10}$$

Equation (4.2) now may be written

$$1 = \sum \frac{V_n}{2E_n} = V_0 \sum_n \frac{1}{2E_n} + V_0 \sum_n \frac{|S_n|^2-1}{2E_n}. \tag{4.11}$$

Consulting Eqs. (3.12), (4.2), (4.8), and (4.10), we get

$$\begin{aligned} 1 = \rho_0 \sinh^{-1}(\omega/\bar{\phi}) - \rho_0(r_2^{\mathrm{I}} + r_2^{\mathrm{II}})\sinh^{-1}(\kappa_2/\bar{\phi}), \\ \cos\theta_2 > \cos\theta_1. \end{aligned} \tag{4.12}$$

Here $\rho_0 = NV_0$; r^{I} and r^{II} come from the two regions corresponding to Eqs. (4.8) and (4.10), respectively:

$$\begin{aligned} r_2^{\mathrm{I}} &\approx \alpha[\cos\theta_2 + (\cos\theta_2)^{-1} - 2], \\ r_2^{\mathrm{II}} &\approx \cos\theta_2 - \cos\theta_1, \end{aligned} \tag{4.13}$$

and $\kappa_2 = \kappa(\cos\theta_2)$. Equations (4.12) and (4.13) are primarily designed for small $\cos\theta_2 \ll 1$ and large $\kappa_2/\bar{\phi}$ (thin film), but should be reasonable for the entire range. Especially r_2^{I} is made to vanish correctly for $\cos\theta_2 = 1$.

In case $\cos\theta_2 < \cos\theta_1$, there is no region II, so that

$$\begin{aligned} 1 &= \rho_0 \sinh^{-1}(\omega/\bar{\phi}) - \rho_0 r_1^{\mathrm{I}} \sinh^{-1}(\kappa_1/\bar{\phi}), \\ \kappa_1 &= \kappa(\cos\theta_1), \\ r_1^{\mathrm{I}} &= \alpha/\cos\theta_1. \end{aligned} \tag{4.14}$$

Since

$$1/\rho_0 = \sinh^{-1}\omega/\phi_0 \sim \ln(2\omega/\phi_0)$$

and $\sinh^{-1}(\omega/\bar{\phi}) \sim \ln(2\omega/\bar{\phi})$, Eqs. (4.12) and (4.14) may also be written (dropping the bar)

$$\begin{aligned} \ln(\phi/\phi_0) &= -(r_2^{\mathrm{I}} + r_2^{\mathrm{II}})\sinh^{-1}(\kappa_2/\phi), \\ \ln(\phi/\phi_0) &= -r_1^{\mathrm{I}} \sinh^{-1}(\kappa_1/\phi) \end{aligned}$$

or

$$\phi/\phi_0 = \exp[-(r_2^{\mathrm{I}} + r_2^{\mathrm{II}})\sinh^{-1}(\kappa_2/\phi)], \tag{4.15a}$$

$$\phi/\phi_0 = \exp[-r_1^{\mathrm{I}} \sinh^{-1}(\kappa_1/\phi)], \tag{4.15b}$$

which are transcendental equations for ϕ. For simplicity, we shall write them as a single equation

$$\phi/\phi_0 = \exp[-r \sinh^{-1}(\kappa/\phi)]. \quad (4.16)$$

For sufficiently weak fields Eq. (4.15b) applies, but as the field increases and/or the thickness decreases, we go over to Eq. (4.15a). The transition takes place at $\cos\theta_1 = \cos\theta_2$, or

$$(\pi/p_F L)^2 = \alpha_0/(1+\alpha_0) \approx \alpha_0 \ll 1, \quad (4.17)$$

since the left-hand side is $\ll 1$. In terms of H and L, this means

$$\pi/p_F L = 8^{1/2} eHL^2/\pi^3$$

or

$$8^{1/2} eHL^2 (p_F L)/\pi^4 = 1. \quad (4.18)$$

For $L = 10^{-5}$ cm, it corresponds to $H \approx 25$ G, and for $L = 0.5 \times 10^{-5}$ cm, $H \approx 200$ G. Fields stronger than these will lead into the domain of Eq. (4.15a). For very strong fields, ϕ becomes small so that $\sinh^{-1}(\kappa/\phi) \approx \ln(2\kappa/\phi)$. Thus, Eq. (4.16) reduces to

$$\phi/\phi_0 = (2\kappa/\phi)^{-r} = (2\kappa/\phi_0)^{-r/(1-r)}$$
$$= \exp[-(r/1-r)\ln(2\kappa/\phi_0)]. \quad (4.19)$$

For large α, then, it decays exponentially like

$$\phi/\phi_0 = \exp[-2\alpha \ln(2\kappa_2/\phi_0)] = (2\kappa_2/\phi_0)^{-2\alpha} \quad (4.20)$$

according to Eq. (4.13), but will not vanish at a finite magnetic field.

It is not possible to solve Eq. (4.16) explicitly for ϕ over the entire range, but the following formula

$$\phi/\phi_0 = \exp[-(r/1-r)\sinh^{-1}(\kappa/\phi_0)], \quad (4.21)$$
$$r = \alpha/\sqrt{\alpha_0},$$
$$\kappa/\phi_0 = \pi q_0 \xi_0 \sqrt{\alpha_0} \quad \text{for} \quad \alpha < \alpha_0; \quad (4.21a)$$
$$r = \alpha\{(\alpha/1+\alpha)^{1/2} + (1+\alpha/\alpha)^{1/2} - 2\}$$
$$+ (\alpha/1+\alpha)^{1/2} - \sqrt{\alpha_0},$$
$$\kappa/\phi_0 = \pi q_0 \xi_0 (\alpha/1+\alpha)^{1/2} \quad \text{for} \quad \alpha > \alpha_0; \quad (4.21b)$$
$$\sqrt{\alpha_0} = \pi/p_F L,$$
$$\alpha = 8e^2 H^2 L^4/\pi^6$$

turns out to be a fairly good representation of the solution to Eq. (4.16).

For a relatively thick film and weak field, $\sinh^{-1}\kappa/\phi_0$ may be replaced by κ/ϕ_0, and Eq. (4.21) reduces for $\alpha \ll 1$ to

$$\phi/\phi_0 = \exp[-r\kappa/\phi_0]$$
$$= \exp[-\pi q_0 \xi_0 \gamma \alpha]$$
$$= \exp[-(8/\pi^4)\xi_0 L^3 (eH)^2 \gamma], \quad (4.22)$$

where $\gamma(r,\kappa/\phi_0) = 1$ for $\alpha < \alpha_0$, and $\gamma \sim 1$ for $\alpha > \alpha_0$. Since our calculations have been based on thin-film conditions, perhaps the exact value of γ following from Eq. (4.21b) is not to be trusted. But Eq. (4.22) above is a rather convenient way of seeing the qualitative behavior of ϕ/ϕ_0 since $\gamma(r,\kappa/\phi_0)$ defined in this fashion turns out to be not too wildly varying over the entire range of α. In fact, we know from Eq. (4.20) that $\gamma \to 2\ln(2\pi q_0 \xi_0)/\pi q_0 \xi_0$ as $\alpha \to \infty$, which is ~ 0.1 for $L = 10^{-5}$ cm and $\xi_0 = 10^{-4}$ cm. In the intermediate range $\alpha = 0(1)$, γ can be > 1.

The fact that our result does not produce a phase transition at a finite field is in accordance with our general considerations, but cannot be taken literally since it is certainly not correct to extrapolate our crude theory to arbitrarily high magnetic fields. It only demonstrates, within our model, the difficulty of completely destroying superconductivity at zero temperature.

We conclude therefore that at zero temperature the gap will decrease steadily with increasing magnetic field, and will undergo a second-order phase transition at a critical field H_c which is probably very high but cannot be estimated within our framework. H_c may be as high as the field necessary to produce complete circular orbits within the film ($H \sim p/eL$). We must, however, also take into account the spin paramagnetic energy[12] which tends to destroy the BCS-type pairing:

$$\frac{eH}{2mc}\Big/ kT \approx 6 \times 10^{+5} \text{ G/°K}.$$

Finally, we would like to emphasize that the energy gap will be anisotropic in a magnetic field, and may actually go to zero around the direction perpendicular to the field and parallel to the film since those electron states are most disturbed by the field. The tunnelling and the thermal conductivity experiments will measure different quantities in such a case. If, however, impurity scattering is important ("dirty" superconductor), the anisotropy tends to be smoothed out.[11]

5. PHASE TRANSITION AT FINITE TEMPERATURES

The treatment of the magnetic field problem at finite temperature goes in much the same way as in the previous sections, and qualitatively speaking it is even easier than at zero temperature. The problem of phase transition in these two cases can be treated separately.

The basic procedure is variational calculation where one minimizes the thermodynamic free energy in the presence of the magnetic field. This was carried out in detail by Bardeen[3] in perturbation theory. The objection raised against his calculation of the kernel $K(\mathbf{q})$ still holds, but it does not look as serious as at $T = 0$. This is because both the H-independent and dependent parts of thermodynamic free energy F_s behave alike as functions of the variational parameter ϕ [3]:

$$F_s^{(0)} \propto \phi^2 \ln t(1-\rho \ln t)$$
$$F_s^{(2)} \propto \phi_0 \phi \tanh(\beta\phi/2) H^2 \quad (5.1)$$
$$\sim \phi^2 H^2, \quad \beta\phi \ll 1,$$

[12] A. M. Clogston, Phys. Rev. Letters 9, 266 (1962).

where $t=T/T_c$, $\beta=1/kT$; ϕ_0 is the gap at $T=0$. This is the reason why he obtains a second-order transition at higher temperatures, $t\gtrsim 0.3$.

From our point of view, we will have to redo all the calculations with our geometrical conditions and using thermal Green's functions. Here let us avoid these troubles and look at the structure of the energy gap equation for finite temperature when the magnetic eigenstates are paired:

$$\phi_n=\sum_m V_{nm}\frac{\phi_m}{2E_m}\tanh(\beta E_m/2). \tag{5.2}$$

E_n and V_{nm} depend on the magnetic field, but not on the temperature. But when we express the solution $\phi_n\approx\phi$ in terms of an effective coupling constant $\rho=N\bar{V}$ after rewriting Eq. (5.2) in the standard form

$$1=\bar{V}\sum_n \frac{1}{2E_n}\tanh(\beta E_n/2), \tag{5.3}$$

a temperature dependence of ρ creeps in through the definition of the average $\bar{V}$ with respect to the weight $\tanh(\beta E_n/2)/E_n$.

We point out, however, that the weight function is rather insensitive to temperature as the actual ϕ changes with temperature to keep the sum in Eq. (5.3) constant. This is also the reason for the well-known fact that the coherence length is nearly independent of temperature. As a result, we may regard ρ as a function of the magnetic field and the gap parameter at $T=0$: $\rho=\rho(H^2,\phi_0)$. This brings in a great simplification of the problem for finite temperatures. Let us plot, in the standard manner, the ϕ versus T curve for the field-free case corresponding to a coupling parameter ρ_0. As the magnetic field is switched on, the effective coupling parameter ρ decreases from ρ_0, but this change is temperature-independent. Consequently, we obtain a family of curves which are only reduced in scale (note that ϕ_0 and the critical temperature T_c are proportional to each other). These curves are shown in Fig. 6 (broken lines).

Now suppose we are operating at a fixed temperature T_1. The energy gap $\phi(H,T_1)$ is given by the intercepts of the family of curves with the vertical plane $T=T_1$. With increasing H, ϕ comes down steadily until it vanishes when T_1 happens to be the critical temperature for a fictitious superconductor with reduced coupling parameter $\rho=\rho(H)$. Hence,

Theorem 4. A thin superconductor in a magnetic field behaves approximately like a fictitious superconductor without the magnetic field, but having a reduced gap and a correspondingly reduced critical temperature. For fixed T and variable H, it undergoes a second-order phase transition when the critical temperature of the fictitious system equals T.

Let us express the above relation quantitatively. For

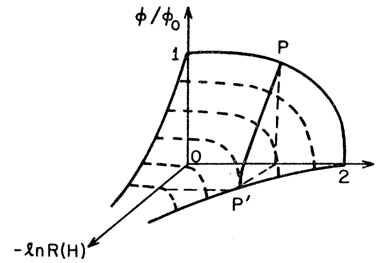


FIG. 6. Three-dimensional plot illustrating the scaling rule. Curve PP' gives the profile of ϕ versus $-\ln R(H)\sim CH^2$ at a fixed t. The letter 2 on the t axis should be read as 1.

$H=0$, the energy gap $\phi(T)$ has the universal form $\phi(t)/\phi(0)=F(t)$,

$$F(0)=1,\quad F(1)=0$$
$$\phi(0)=CkT_c\quad (C=1.75 \text{ in the BCS theory}). \tag{5.4}$$

When $H\neq 0$, $\phi(0)$ and T_c are replaced, respectively, by

$$\phi(0,H)=R(H)\phi(0),$$
$$T_c(H)=R(H)T_c, \tag{5.5}$$

where $R(H)$ is the scaling factor. We get, thus, the general formula

$$\phi(t,H)=R(H)\phi_0F[t/R(H)],$$
$$\phi_0=\phi(T=0,H=0), \tag{5.6}$$
$$t=T/T_c(H=0).$$

Since $F(t)=0$ at $t=1$, it follows immediately that the critical field H_c at which $\phi=0$ is determined by

$$R(H_c)=t. \tag{5.7}$$

Near $t=1$, $F(t)$ behaves like $(1-t)^{1/2}$, so that

$$\phi(t,H)\sim(1-t/R)^{1/2}=[1-R(H_c)/R(H)]^{1/2}. \tag{5.8}$$

$R=\phi(0,H)/\phi_0$ is given by Eqs. (4.21)–(4.22) depending on their applicability. In general, the weak field formula (4.21a) will be valid at high temperatures where $R\approx 1$. At lower temperatures a changeover to Eq. (4.21b) takes place as the field increases. However, this depends on film thickness. We will consider the two cases.

(1) $\pi q_0\xi_0\sqrt{\alpha_0}\lesssim 1$. This happens for a relatively thick film ($L\gtrsim 0.5\times 10^{-5}$ cm for $\xi_0=10^{-4}$ cm). Then Eq. (4.22) (with $\gamma=1$) is certainly valid for temperatures

$$t>t_0=R_0=\exp[-\alpha_0\pi q_0\xi_0]. \tag{5.9}$$

This is very close to 1. For example, with $\xi_0=10^{-4}$ cm, $t_0=1-10^{-3}$ for $L=10^{-5}$ cm and $t_0=0.9$ for $L=0.5\times 10^{-5}$ cm. The ϕ versus H curve following from Eqs. (4.22) and (5.8) becomes then

$$\phi^2(t,H)/\phi^2(t,0)=1-H^2/H_c^2,$$
$$eH_c=(1-t)^{1/2}(\pi^2/\sqrt{8})\xi_0^{-1/2}L^{-3/2}. \tag{5.10}$$

This essentially agrees with the results of Ginzburg-Landau-Gorkov-Douglass theory and of Bardeen near $t=1$.

For temperatures below t_0, there will be a changeover to the region $\alpha>\alpha_0$ at a field determined by Eqs. (4.17)

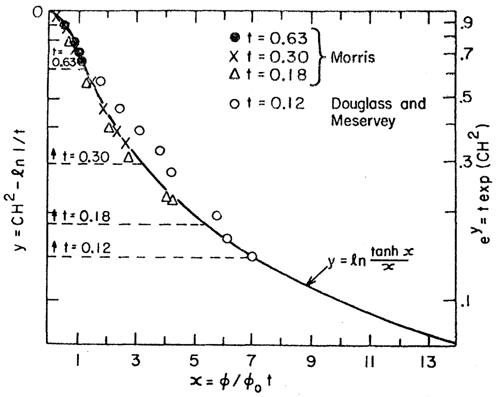


FIG. 7. Universal function $y=\ln(\tanh x/x)$ and experimental points. The experimental points of Morris and those obtained by using the BCS relation $\phi_0=1.75\ kT_c$; $t=T/T_c$ was computed with $T_c=7.2°$K.

and (4.18). As long as $\alpha\ll1$, $\pi q_0\xi_0\sqrt{\alpha}\lesssim1$, Eq. (4.22) is still good, but the ϕ versus H behavior will be somewhat more complicated than Eq. (5.10). The critical field is given by

$$eH_c=(\ln1/t)^{1/2}(\gamma\pi^2/\sqrt{8})\xi_0^{-1/2}L^{-3/2}, \qquad (5.11)$$

which goes over to Eq. (5.10) for $t\approx1$.

As the temperature is further lowered, we begin to deviate from Eq. (5.11). H_c must now be determined numerically from Eq. (4.21). This happens for the temperature ranges of most experiments.

(2) $\pi q_0\xi_0\sqrt{\alpha_0}\gg1$. For this very thin-film case, the changeover between $\alpha<\alpha_0$ and $\alpha>\alpha_0$ will take place if

$$t<t_0=R_0=\exp[-\sqrt{\alpha_0}\ln(2\pi q_0\xi_0\sqrt{\alpha_0})]. \qquad (5.12)$$

Above this, we have then

$$\phi^2(t,H)/\phi^2(t,0)=1-H^2/H_c^2,$$
$$eH_c=(\ln1/t)^{1/2}(\pi^7/8)^{1/2}p_F^{-1/2}L^{-5/2}\times[\ln(2\pi^3\xi_0/p_FL^2)]^{-1/2}. \qquad (5.13)$$

At lower temperatures when Eq. (4.21b) takes over, the behavior again should be determined numerically.

6. COMPARISON BETWEEN THEORY AND EXPERIMENT

In order to express our basic equation (5.6) analytically, it is convenient to use the implicit form[8,13]

$$\phi(t)/\phi_0=\tanh[\phi(t)/\phi_0t]. \qquad (6.1)$$

Introducing the scaling factor ($\phi_0\to R\phi_0$, $t\to R^{-1}t$), we obtain

$$\phi(t,H)R^{-1}(H)/\phi_0=\tanh[\phi(t,H)/\phi_0t], \qquad (6.2)$$

which we can write in the form

$$t/R(H)=\tanh x/x$$

or

$$y=\ln[1/R(H)]-\ln1/t=\ln(\tanh x/x), \qquad (6.3)$$

where

$$x=\phi(t,H)/\phi_0t.$$

We can plot all data on a single y versus x curve if we measure ϕ in units of ϕ_0t and shift the y coordinate by $\ln1/t$ for different t. For those relatively thick films and weak fields where Eq. (4.22) applies, $\ln1/R\sim H^2$, so that Eq. (6.3) becomes of the form

$$CH^2-\ln1/t=\ln(\tanh x/x). \qquad (6.4)$$

Equation (6.3) is plotted in Fig. 7. Also shown are the experimental points of Morris and those of Douglass and Meservey for lead, based on Eq. (6.4). These are well outside the range of applicability of Eq. (4.22), but numerical calculation has shown that $\ln1/R\propto H^2$ is still valid to a good approximation. In addition, we find $H_c=3700$ G under Douglass' condition $t=0.12$ and $L=10^{-5}$ cm. Expermentally H_c is ≈2300 G. Figure 8 shows the same comparison on more conventional plots. We see that the general trends at lower temperatures are correctly predicted by the theoretical curve.

We have a few remarks to make. (1) Lead is an anomalous superconductor with a large coupling parameter ρ whereas our theory is based essentially on the weak coupling. (2) The effect of the finite penetration depth is neglected in our formulas. (3) The experimental conditions are more complicated than those assumed in our model (parallel surfaces with specular reflection, no impurity scattering, etc.). (4) There are certain uncertainties in the interpretation of experiimental data, e.g., the identification of ϕ with measured quantities, and the exact determination of H_c where ϕ vanishes.

Each of these points can be taken into account if necessary, but since all of these possibilities may be present and may cause modifications in different directions, we have not attempted to analyze them. In view of the crudeness of our theory, we therefore conclude that the agreement between theory and experiment is at least qualitatively satisfactory.

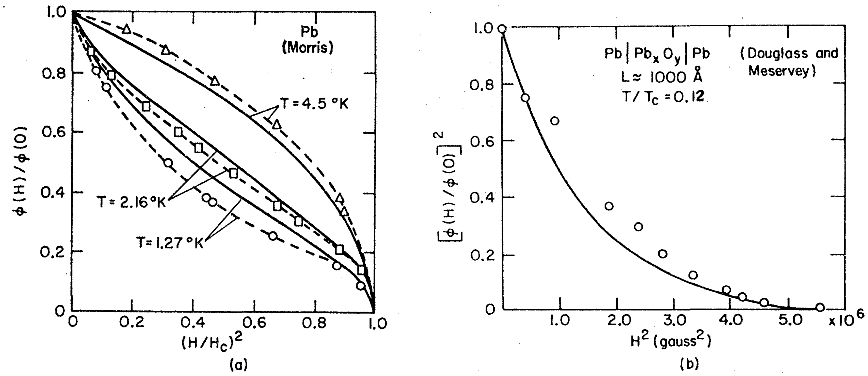
7. COMPARISON WITH OTHER THEORIES

Bardeen,[3] in a recent paper on critical fields in superconductors, concluded that the microscopic theory yielded a first-order phase transition for thin films with reduced temperature $T/T_c\lesssim0.3$. His calculations differ from our approach in two major respects; (a) our use of discrete quantized momentum variables q_x rather than the continuum momentum variable and (b) our choice of the London gauge rather than an arbitrary gauge for purposes of calculations.

Our adoption of the discrete quantization model and the London gauge enables us to conclude that for sufficiently thin films, the leading term in the BCS kernel $K(q)$, ϕ/q, vanishes; this in turn determines in a crucial way in the framework of perturbation theory the question of phase transition at low temperatures. In fact, if we take $\delta\phi$ calculated for the bulk material

[13] D. J. Thouless, Phys. Rev. 117, 1256 (1960).

FIG. 8. Comparison of theoretical and experimental curves on conventional plots taken from Ref. 8.



case in I, apply alternating fields H and other specifics appropriate to the thin film case [cf. Eq. (2.19)], we find complete equivalence with Bardeen's calculations for this case if the ϕ/q term in $K(q)$ is naively retained.

It appears that the choice of gauge is important in our case particularly because there is a degeneracy of magnetic eigenstates, i.e., to one energy ϵ belong an infinity of states with different p_y, p_z and the (discrete) quantum number in the x direction. A different gauge would, in general, lead to a different set of eigenstates, which in turn imply a different pairing scheme of electrons. A wrong choice of the pairing would not maximize the pairing energy, and furthermore might not be adiabatically related to the field-free pairing. In the latter case, the perturbation theory would not work.

For cases when there is a large amount of thermal excitation present, or $T\sim T_c$, the Ginzburg-Landau-Gorkov-Douglas theory[4,5] predicts second-order transition for thin films with $H_c \propto L^{-3/2}$. Despite the fact that experimental data[7] seem to suggest that it might be possible to account for the low-temperature properties along the lines of G-L-G theory as well when a sufficiently strong field is applied so that $\phi(H)\sim kT$, we wish to point out that Gorkov's theory (even with the inclusion of a strong magnetic field) cannot reproduce our results for T near $T=0$°K. This is because the Gorkov theory is essentially a London theory, $\xi_0 \ll \lambda_0$, where ξ_0 and λ_0 are the bulk coherence distance and penetration depth, respectively. Since $\xi_0 = v_F/\pi\phi$, at $T=0$, we have $\xi_0 \to \infty$ as $\phi \to 0$ (near critical field H_c), and the London limit is not satisfied.

Mathur, Panchapakesan, and Saxena[6] arrived recently at the conclusion that a second-order phase transition is expected at $T=0$°K for thin films based on earlier calculations of Gupta and Mathur[14] which used Wentzel's theory of gauge invariance.[15] It appears that Mathur *et al.* took the London limit for bulk specimen parameters ξ_0 and λ_0 in their study. This is quite evidently not satisfied for thin films where the appropriate limit is the Pippard nonlocal form when expressed in terms of bulk material parameters ξ_0 and λ_0. This is to be contrasted with the work of Douglass[4] on thin films, where the London limit is used appropriately in the form $\xi \ll \lambda$, where ξ and λ are the coherence distance and penetration depth for the thin film itself.

[14] K. K. Gupta and V. S. Mathur, Phys. Rev. **121**, 107 (1961).

[15] G. Wentzel, Phys. Rev. **111**, 1488 (1968); a comparison of Wentzel's formulation of gauge invariance with that of the BCS-Bogoliubov approach is given in Ref. 2.

8. CONCLUDING REMARKS

We have adopted specular reflection boundary conditions for electrons at film surfaces. Though a more exact solution, say for an electron in an image potential[16] is certainly more preferable to any artificial boundary condition on some *ad hoc* "surface," we feel that our conclusions are dependent primarily on the London gauge rather than on the details of boundary conditions or the shape of the boundary. In this connection it will be instructive to study the cases where, for example, the magnetic field is not parallel to the film surfaces, or parallel but unequal fields are applied at the opposite surfaces. This will involve us in the proper choice of current distribution and pairing so as to optimize the balance between magnetic and pairing energies. Significant modifications that would ensue are already suggested in the works of Douglass[4] and Tinkham.[17]

A proper extension of the present work to finite temperatures, which evaluates $K(q,T)$ in terms of thermal Green's functions in the presence of magnetic field,[18] is obviously highly desirable since the temperature dependence here is introduced only qualitatively via theorem 4. Such a comprehensive analysis will allow us to compare reliably our results with those of G-L-G at high temperatures.

Finally, experimental work on thin films in the range 100 to 1000 Å, at reduced temperature $T/T_c<0.3$ for a soft (and weak coupling) superconductor like tin will test most critically the present notions. We note

[16] L. A. MacColl, Bell System Tech. J. **30**, 888 (1951).

[17] M. Tinkham, Phys. Rev. **129**, 2613 (1963).

[18] Such a construction may perhaps be obtained analogously to the Green's functions for the thermal conductivity problem. See, for instance, L. Tewordt, Phys. Rev. **128**, 12 (1962), also private communications.

especially that our formulas are generally valid only in the weak coupling case ($N\bar{V}\ll 1$), thus it may perhaps be improper to infer any definitive conclusions from experimental studies[7,8] on an anomalous (strong coupling) superconductor like lead.

Note added in proof. A recent paper by K. Maki, Progr. Theoret. Phys. (Kyoto) **29**, 603, 945 (1963), starts from the Green's function approach of Gorkov, and obtains results similar to ours.

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Bistatic Scattering from a Class of Lossy Dielectric Spheres with Surface Impedance Boundary Conditions*

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Expressions are derived for the bistatic scattering cross sections of spheres which exhibit sufficient electric and/or magnetic loss to permit each modal surface impedance in the Mie formulation to be replaced by a single impedance which is the intrinsic impedance of the lossy medium. Typical bistatic scattering curves are presented for several values of the characteristic impedance of the sphere medium.

IN a recent paper[1] by Wagner and Lynch, sufficient conditions are developed which effect zero electromagnetic backscatter from axially symmetric objects when illuminated along the axis of symmetry. The present paper considers the special case of scattering from spheres which exhibit sufficient loss to permit each modal surface impedance to be replaced by a single surface impedance which is the intrinsic impedance of the lossy medium. If this impedance is that of the ambient medium, the spheres have zero backscatter.

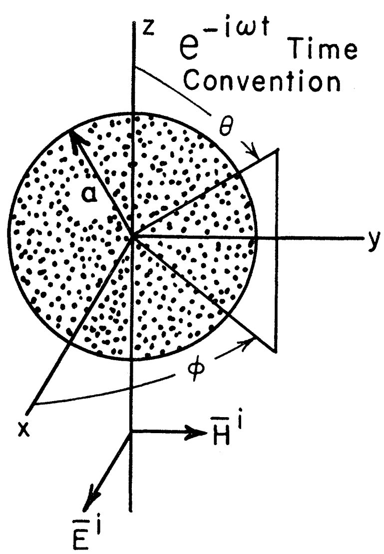

FIG. 1. The sphere geometry.

Referring to Fig. 1, the fields scattered by a sphere due to a plane wave incident from the $-z$ direction are,

$$E_\theta=\frac{-ie^{-i\omega t}e^{ik_2r}}{k_2r}\cos\phi\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\times\left[a_n\frac{P_n^1(\cos\theta)}{\sin\theta}+b_n\frac{\partial P_n^1(\cos\theta)}{\partial\theta}\right], \quad (1)$$

$$E_\phi=\frac{ie^{-i\omega t}e^{ik_2r}}{k_2r}\sin\phi\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\times\left[a_n\frac{\partial P_n^1(\cos\theta)}{\partial\theta}+b_n\frac{P_n^1(\cos\theta)}{\sin\theta}\right], \quad (2)$$

and the resulting normalized scattering cross sections are,

$$\frac{\sigma_E}{\pi a^2}=\lim_{r\to\infty}\left(\frac{2r}{a}\right)^2|E_\theta(\phi=0^\circ)|^2, \quad (3)$$

$$\frac{\sigma_H}{\pi a^2}=\lim_{r\to\infty}\left(\frac{2r}{a}\right)^2|E_\phi(\phi=90^\circ)|^2. \quad (4)$$

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[1] R. J. Wagner and P. J. Lynch, Phys. Rev. **131**, 21 (1963).