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Dependence of the Energy Gap in Superconductors on Position and Magnetic Field*

Abstract: A review is given of work concerning the decrease of the energy gap in superconductors when a magnetic field is applied. The absence of any observable effect in previous spectroscopic work is explained, and conditions for large effects are outlined. Experimental measurements on thermal conductivity and microwave absorption in films in a magnetic field are described. The results show that in thin films, the gap can be depressed continuously to zero, yielding a second-order phase transition. In thicker films, the gap can be only partially depressed before the transition, and the transition is of first order. These results agree with those obtained theoretically by Douglass from the Ginzburg-Landau-Gor'kov theory, and experimentally, by electron tunnelling. An attempt to generalize the Ginzburg-Landau-Gor'kov theory to cope with the case when $\varepsilon_0/kT \gg 1$ is then indicated. In this phenomenological theory, the normalized gap $\varepsilon(H)/\varepsilon(0)$ is taken as an order parameter ω, and the free energy is assumed to contain a term in $\xi_0^2|\nabla\omega|^2$, as well as a free energy density $f(\omega)$ and magnetic energy $\varkappa(\omega)H^2$. Some success is found.

Introduction

It is a basic part of the BCS theory[1] that stable supercurrents, such as those which arise in producing perfect diamagnetism, can be understood qualitatively by considering the normal BCS ground state composed of pairs with net zero momentum to be transformed into a state in which each has the same non-zero momentum $2m\mathbf{v}_d$, where $\mathbf{v}_d$ is the drift velocity of the entire electron cloud. Although there is disagreement on this point,[2] this state is considered stable against scattering because of the gap energy required to remove each pair and drop it into a state with lower net momentum. A fluctuation taking any macroscopic number of electrons out of the ground state against an energy cost in this way is astronomically unlikely, and we have stable currents. Bogoliubov[3] pointed out in 1958 that the presence of this drift should alter the excitation spectrum of the BCS state by adding a term $\mathbf{p}_F \cdot \mathbf{v}_d$. Evidently, this would cause the current to be unstable if $|p_F||v_d|$ equaled the gap ε_0. Calculations by various authors have shown that this condition is similar to that derived from the macroscopic free-energy balance. Thus the effect should be sizable, leading to perhaps a 50 per cent reduction in the minimum gap, together with a similar increase in gap on the other side of the Fermi surface.

Having devoted considerable effort at Berkeley to measuring the gap spectroscopically, we were intrigued by the possibility of observing such large effects in the presence of a field. However, we had little success doing so in spectroscopic experiments at the time. In Ginsberg's experiments[4] on transmission of far infrared through superconducting films, we intentionally applied 8 kG fields to see what effect this would have. We found no effect outside a few percent. In Richards' experiment[5] on reflection from bulk samples, the geometry prevented any controlled use of magnetic fields, but trapped flux kept $\sim 1/2$ the sample normal and provided fields of the order of the critical field at the surface of the superconducting metal. Yet, he found a very sharp absorption edge with no evidence of a smear or shift of the gap.

How was this to be explained? For one thing, our photons have little momentum; hence we must produce

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pairs of excitations on opposite sides of the Fermi surface, and the first-order gap change should cancel out. Moreover, with the thin film we could reach only a small fraction of H_c, and hence j_c or v_c, with a field of 8000 gauss, so no higher order effects $\sim H^2$ or v^2 should have been observable. In the bulk sample we readily reached H_c, but any sizable second-order gap decrease was prevented by the long coherence length ξ_0 of Pippard,[6] which keeps the surface gap from being greatly affected, as long as there is an interior region of depth ξ_0 from which the field and current are excluded by the surface currents in a depth λ. The question then was how to proceed.

Because of the long coherence length, $\xi_0 \gg \lambda$, it was evident that we would have to use films in which the whole sample felt the field, if we were to find effects greater than the few percent observed by Pippard for the change in λ. Also, evidently, to get a maximum effect, we must be able to reach the critical field of the film. Since $H_c/H_0 \sim \lambda/d$, this limits the film thickness d to $\gtrsim$ 200 A for fields of convenient magnitude. But a film as thick as that is far too good a reflector to be usable in a far-infrared experiment of the sort performed by Ginsberg. Thus we considered other physical effects, for which observations could be made on films of $d =$ 300 to 3000 A. The tunnel experiments[7] described earlier in the sessions are a beautiful example (which we did *not* think of!). However, we had chosen another approach, via thermal properties.

Thermal conductivity measurements

Even if the sample had to be too thick for a sensitive far-infrared gap measurement, a simple thermal property could still reveal gap changes. Specific heat seemed hard to measure in a thin film, but thermal conductivity seemed more promising. Morris and I set up a simple experiment[8] with an evaporated film on a thin glass substrate, with carbon resistance thermometers at both ends. One resistor served as heat source, as well as thermometer, and the other was attached at the temperature bath end of the film.

Figure 1 shows results on thin ($\sim$700 A) films of tin and indium. For both metals, the thermal conductivity K increases nearly as H^2 up to the critical field. At the critical field, the thermal conductivity in the superconducting state joins smoothly onto the field-independent conductivity of the normal metal. The location of the shoulder at this point fixed H_c for the purposes of normalizing the data. The absence of any discontinuity at H_c indicates that the superconducting transition in a magnetic field is second order for films of this thickness, in contrast to the first-order transition of bulk superconductors in a field. In other words, since K is related directly to the gap width, we can infer that the gap closes continuously to zero when a field is applied to a thin film, whereas in a bulk sample it drops discontinuously from nearly the full value ($\sim$98 percent even at the surface) to zero.

In view of the theoretical prediction of a $\mathbf{v}_d \cdot \mathbf{p}_F$ term

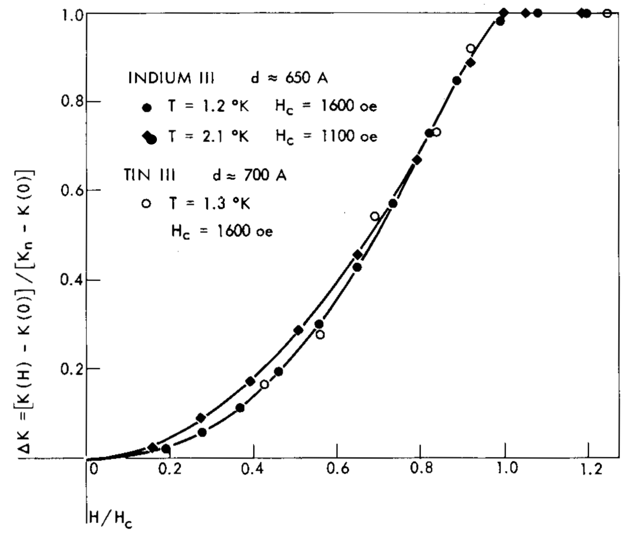


Figure 1 **Change of thermal conductivity of thin superconducting films with magnetic field.**

in the excitation spectrum, we varied the orientation of the field in the plane of the film from parallel to perpendicular to the direction of heat flow in the film. This change produced no effect on K to within experimental accuracy of a few percent. Since the induced diamagnetic currents are orthogonal to the field, this measurement also shows the absence of a dependence on the angle between heat current and diamagnetic current. Thus, if we interpret our results in terms of a decrease of the energy gap with increasing field, the modified gap appears to remain essentially isotropic. Of course, even if there were a $\mathbf{v} \cdot \mathbf{p}$ term, first-order effects would cancel in an average transport effect like K. However, as the change approaches 100 percent, noncancelling second-order effects would appear, and these would be expected to differ by a factor of several for the parallel and perpendicular cases, because of a different average of $\cos^2 \theta$. Thus another factor must enter to explain the lack of anisotropy. It may well have to do with the fact that in these films, the two equal and opposite surface currents are only slightly separated in space (compared to ξ_0, for example). Alternately, the short mean free path in these "dirty" samples may upset the simple prediction. Both of these characteristics of the experimental situation contrast strongly with the Bogoliubov idealization, which corresponds to a spatially uniform current arising from a displaced sphere of momentum eigenfunctions characteristic of an ideal sample.

The measured dependence of K upon H may be used to compute the dependence of the energy gap on H in a simple way if the electronic term, K_e, is dominant in K, and if K_e is primarily limited by elastic scattering of electrons by lattice imperfections. Under these conditions, the ratio of thermal conductivity in the superconducting state, K_{es}, to that in the normal state, K_{en}, has been given by Bardeen, Rickayzen, and Tewordt[9] as

$$\frac{K_{es}}{K_{en}} = G(\varepsilon_0/kT) = \frac{\int_{\varepsilon_0}^{\infty} E^2(\partial f/\partial E)\, dE}{\int_0^{\infty} E^2(\partial f/\partial E)\, dE}, \tag{1}$$

where $f(E/kT)$ is the Fermi function, and $2\varepsilon_0$ is the energy gap. If we assume that the effect of a field upon the superconducting state can be adequately represented as a change in the ε_0 of BCS, then this relation may be used to make a point-by-point inversion of experimental data to determine $\varepsilon_0(H)/\varepsilon_0(0)$. Starting from the data on the thin indium film shown in Fig. 1, we find the results shown in Fig. 2. The error bars represent the uncertainty in normalization to the critical field, H_c, because of a slight rounding of the transition region in the data on K. This uncertainty is magnified by the fact that $G(\varepsilon_0/kT)$ initially drops only quadratically as ε_0/kT increases from zero. This would produce a rounded approach even if the gap dropped to zero linearly in $(H_c - H)$. For the same reason, it requires the rather steep final drop $\sim(H_c - H)^{1/2}$ in the gap near H_c to reproduce the almost linear approach of K_{es} to K_{en} observed at that point.

Our results for the dependence of the energy gap upon magnetic field in a thin film are qualitatively similar to those from the more direct tunnel experiments of Giaever and Megerle[7] and of Douglass,[7] although their data were taken at higher reduced temperature and in a different metal (aluminum). From our data shown in Fig. 2, we see that $\varepsilon_0(H)/\varepsilon_0(0)$ seems to approach $1 - h^2$ $(h = H/H_c)$ at low temperatures, but it is moderately well fitted at $T/T_c = 0.63$ by $(1 - h^2)^{1/2}$. The latter form is that given by Douglass[10] based on the Ginzburg-Landau-Gor'kov (GLG) theory,[11] which is expected to hold near T_c. We also note that our data at $T/T_c = 0.36$ deviate from $1 - h^2$ and approach $(1 - h^2)^{1/2}$ when the gap has dropped so that $\varepsilon_0(H) \sim kT$. This behavior indicates that the GLG approach becomes successful, as expected, when there is a large amount of thermal excitation present.

With thicker films, new features appear, as shown in Fig. 3. The thickest film, tin II with $d \approx 2800$ A, has $\Delta K \sim H^2$ up to the vicinity of H_c, where it has only reached $\sim$25 percent. Then K appears to rise almost discontinuously to K_n, indicating that the transition is first order in films of this thickness, if we assume that the finite slope is entirely due to the measuring temperature gradient and the nonuniformity of the film. Applying our method of analysis, we find that $\varepsilon_0(H)/\varepsilon_0(0)$ drops linearly with $(H/H_c)^2$, then abruptly from $\sim$0.83 to zero at H_c. An analysis based on the GLG theory leads to $\varepsilon_0(H_c)/\varepsilon_0(0) = 0.78$ for a film with this thickness [as related to H_c/H_c (bulk)]. This agreement is really quite good, considering the accuracy and the fact that at H_c the surface gap (which governs the penetration law) should be somewhat less than the interior value for a film as thick as this one. The film indium II, of intermediate thickness $d \approx 1800$ A, is still thin enough to display a second-order transition, but the increase in K near H_c is steeper than H^2. This qualitative behavior is also predicted by the GLG theory, and by various phenomenological models

Figure 2 **Magnetic field dependence of superconducting energy gap computed from data of Fig. 1.**

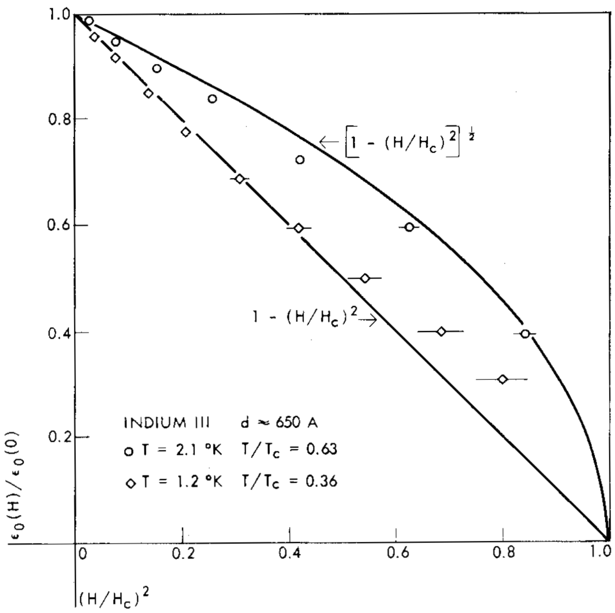
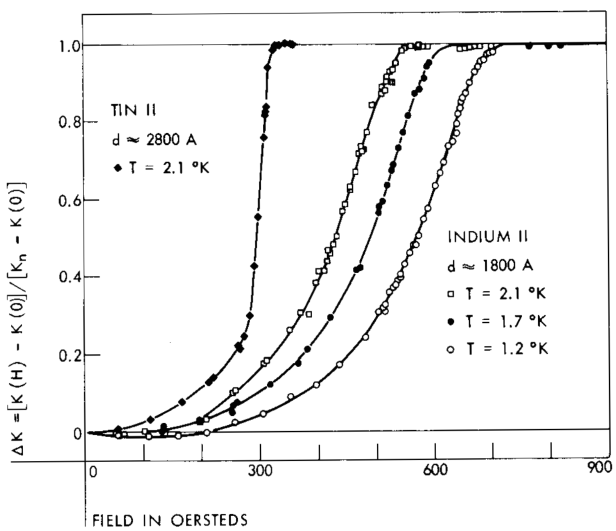


Figure 3 **Change of thermal conductivity of superconducting films of intermediate thickness with magnetic field.**



which take account of the dependence of the penetration depth and free energy of a superconductor upon the energy gap width.

We conclude, then, that our selection of film thicknesses spans the region from second- to first-order superconducting phase transitions in the presence of a field, and that the GLG theory gives a good account of the gap variation with field at the higher temperatures but that it is less successful at lower temperatures. The actual form of $\varepsilon_0(H)/\varepsilon_0(0)$ depends on film thickness, and approaches a definite limit only for films so thin that $d \ll \lambda$.

Microwave absorption

If, indeed, we are able to force the gap down to zero by applying a magnetic field to a film, we should be able to detect this spectroscopically. As remarked above, the critical field of a film thin enough for far-infrared study is unattainably high. However, at 4 to 8 mm microwaves, we have plenty of power available to detect calorimetrically the small fraction of power absorbed by a superconducting film with $d \sim 500$ A. Thus, if we monitor the absorption of microwave power as a function of H, we should see evidence for the same gap decrease observed above.

Morris and White, in our laboratory, have done this on a 120 A indium film at 4 and 8 mm, using matched carbon resistors in a bridge, as above, to measure the heating. Qualitatively, the observed behavior shown in Fig. 4 is similar at both wavelengths, namely a continuous rise in absorption, roughly $\sim H^2$, up to the normal value, after which it remains constant. This behavior simply reflects the increase in absorption by "normal electrons" excited across the gap as the gap decreases. In addition to this common behavior, there seems to be a slight downward shift of the apparent H_c (as measured by the shoulder on the curve) for the 4 mm radiation compared to the 8 mm radiation. This shift can be explained by noting that the 4 mm photons can span the gap as soon as it is reduced to $\sim 1/4$ its field-free value, whereas it must be reduced by another factor of 2 before the 8 mm photons can span it. Because of the steep drop of $\varepsilon_0(H)$ near H_c, however, the shift is small. Also, even the 4 mm photons only have $h\nu = kT$ for 3.5°K. Since the half-gap ε_0 enters the exponential governing thermal excitations, thermal excitations are so prominent that no sharp absorption edge is observed when $h\nu = 2\varepsilon_0(H)$ even at our lowest temperature, 1.2°K. A further reduction in temperature with He^3 refrigeration, or a rise in microwave frequency, would enable us to see whether the absorption edge is still as sharp as in the field-free case, or whether it has changed shape. Because of the sum-rule relation[12] between this absorption spectrum and the supercurrent properties, this data would be a valuable link in clarifying the mechanism by which the modified wave-function associated with the field-reduced $\varepsilon_0(H)$ brings about the increased penetration depth λ.

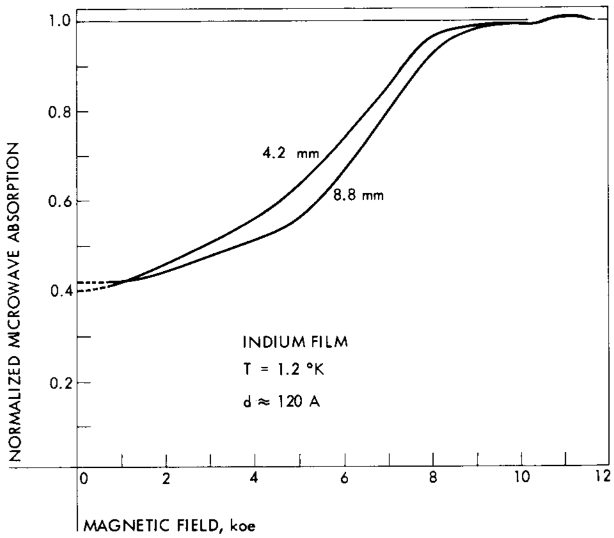


Figure 4 **Change of microwave absorption of a thin superconducting indium film at wavelengths of 4.2 mm and 8.8 mm.** *The zero of absorption is displaced a large, but unknown, amount by absorption in the sample support.*

Discussion

We have seen that the GLG theory gives a fair account of our observations at the higher temperatures, but it seems to be failing at lower temperatures as long as the gap is much greater than kT. For example, our thermal conductivity data shows that $\varepsilon_0(H)/\varepsilon_0(0)$ appears to approach $1 - h^2$ rather than $(1 - h^2)^{1/2}$ at $T \to 0$. This failure is not surprising, since the GLG theory is based on an expansion of the free energy near T_c, which cannot be expected to be valid also when $(\varepsilon_0/kT) \gg 1$. Since there seems to be no reliable *a priori* theory for handling these nonlinear effects over the entire temperature region, we have given thought to formulating a simple phenomenological theory, based on BCS for the zero-field limit, with which to guide our experimental work. We assume that there is a well-defined gap, which we can use as an order parameter, even in situations far from the equilibrium situation treated by BCS. Parmenter[13] has approached the problem in a manner somewhat similar to ours, but he has excluded consideration of magnetic field effects (and hence of many interesting cases).

To shorten notation, let $\omega = \varepsilon_0(H, T)/\varepsilon_0(0, T)$ be an order parameter which goes from 0 to 1 as the gap goes from 0 to its equilibrium value at a given temperature. Also, let $f_T(\omega)$ be the free-energy density at ω and temperature T, normalized to the equilibrium value $H_c(T)^2/8\pi$. [In all the following work, H_c refers to bulk samples; H_T is the transition field of the film.] Similarly, let κ be the susceptibility normalized to $\frac{1}{4}\pi$. Then if, for definiteness, we consider a slab of thickness $2a$ placed in a magnetic field H_a parallel to its surface, we approximate the overall free energy per unit area at given temperature T by

$$G = (H_c^2/8\pi)\left\{ \int_{-a}^{a} [f(\omega) + \beta(\omega)\xi_0^2|\nabla\omega|^2]\, dx + 2a\kappa(H_a/H_c)^2 \right\} . \quad (2)$$

We can evaluate κ approximately using the London theory expression

$$\kappa = 1 - (\lambda/a)\tanh(a/\lambda) . \quad (3)$$

In this, λ is an effective value, as calculated from BCS when $\omega = 1$, but which increases as ω decreases in a manner which is *a priori* uncertain, but expected to lie between $\omega^{-1/2}$ and ω^{-1}. The value, ω_s, of ω at the surface, $x = \pm a$, is the appropriate one to use in this evaluation, in case the slab is thick enough to allow substantial gap variation.

The term in $(\nabla\omega)^2$ arises from the extra kinetic energy which arises from the modulation of plane wave functions when the gap, and hence the occupation numbers $h_k(\varepsilon_k/\varepsilon_0)$ in the BCS wave function, vary in space. The factor $\beta(\omega)$ is of order unity, but probably contains a factor ω^{-n}, where $n = 1$ to 2. To see that ξ_0^2 is the appropriate length factor, one must take account of the orthogonalization to other occupied states implicit in the use of antisymmetrized functions. Then if one considers a modulation, with wave vector q, of the electron wave functions at the Fermi surface, one finds an energy increase of order $\hbar^2 qk_F/2m$ for a fraction $\sim q/k_F$ of the electrons. This raises the system energy by $\sim n\hbar^2q^2/2m$. Since $H_c^2/8\pi \sim nm\varepsilon_0^2/\hbar^2k_F^2$, this energy can be written

$$\sim (H_c^2/8\pi) \times (\hbar v_F/\varepsilon_0)^2 q^2 \sim (H_c^2/8\pi)\xi_0^2(\nabla\omega/\omega)^2 .$$

In view of this result, it seems curious that Parmenter found a characteristic length $\delta \sim (\xi_0/k_F)^{1/2} \sim 100$ A, rather than $\xi_0 \sim 10^4$ A. We might also note that the coefficient of $|\nabla\psi|^2$ in the GLG theory is essentially equivalent to our result.

The free-energy term $f(\omega)$ is approximated in the GLG theory by a two-term power series, $f = -2\omega^2 + \omega^4$, when normalized in our notation, where $\omega \sim \varepsilon_0 \sim \psi$. Although Gor'kov has established this GLG form from the BCS theory near T_c where the gap is small, it cannot be expected to be generally valid. If instead one computes the binding energy at $T = 0$ of the BCS ground state, but with a constrained gap which can be varied all the way down to zero by adjustment of a Lagrange multiplier, one finds the leading term for small gaps to be $-\omega^2/N(0)V$, where $N(0)V$ is the usual coupling-strength parameter, typically 0.2 to 0.4. Of course, the absolute minimum energy occurs at $\omega = 1$, by definition of ω. The function $f(\omega)$ is obtained only implicitly over the full range of ω. It is shown in Fig. 5 for a typical value $N(0)V = 1/3$. Evidently it resembles the GLG form, but departs significantly. One might presume that at intermediate temperature $f_T(\omega)$ might lie between these extremes.

Given $G[\omega(x)]$, one uses a variational principle to find $\omega(x)$ to minimize the free energy for a given value of H_a. All other properties may then be computed.

Evidently the scheme outlined above has so much freedom in it that any number of combinations of assumptions may reproduce a limited amount of data. With enough data, however, or more reliable *a priori* calculations, we might delimit it considerably, and see if it is really capable of describing all observations.

One simple limiting case is that of the very thin film, in which $a \ll \lambda$ or ξ_0. Then the term in $(\xi_0\nabla\omega)^2$ prevents any sizable change in ω over the film thickness. [This case is similar to one treated by Pippard for colloids.[14]] Thus, setting $\omega(x) =$ const.,

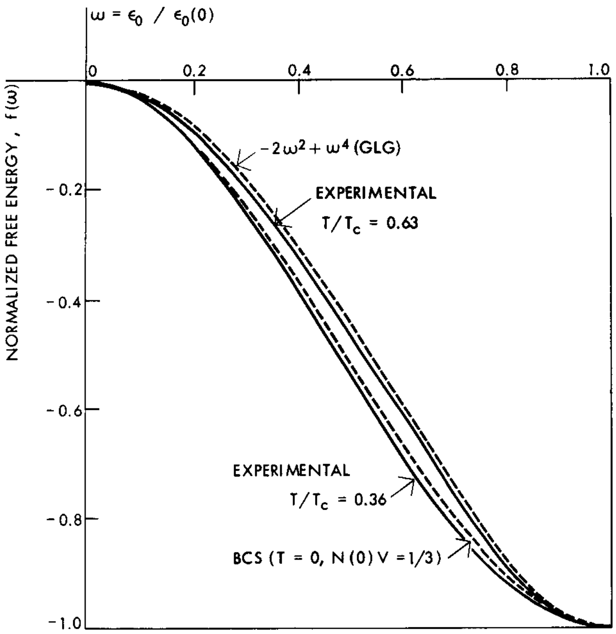
$$G = 2a(H_c^2/8\pi)[f(\omega) + \kappa(\omega)(H_a/H_c)^2] . \quad (4)$$

Then $\partial G/\partial\omega = 0$ implies

$$(H_a/H_c)^2 = -f'(0)/\kappa'(\omega) . \quad (5)$$

Now we know experimentally that as H_a is increased, ω falls continuously from 1 to 0 at a finite transition

Figure 5 **Comparison of free energy curves $f(\omega)$.** *In our normalization, the GLG theory (valid near T_c) gives $f(\omega) = -2\omega^2 + \omega^4$. The BCS curve is calculated at $T = 0$ for $N(0)V = 1/3$. The experimental curves are derived from the results of Figure 2. As might be hoped, the agreement is better between GLG and the higher temperature data, and between BCS ($T = 0$) and the lower temperature data.*



field $H_T > H_c$. Thus the ratio f'/κ' must rise continuously and monotonically to a finite maximum given by

$$(H_T/H_c)^2 = -f'(0)/\kappa'(0) . \qquad (6)$$

Therefore, if $f(\omega)$ and $\kappa(\omega)$ are expandable in power series in ω, the leading terms for both must have the same power of ω. Since we expect $f(\omega)$ to start as ω^2, $\kappa(\omega)$ must also. Now for a thin film, from (3),

$$\kappa = \frac{1}{3}\frac{a^2}{\lambda^2} . \qquad (7)$$

Thus, $(1/\lambda^2) \sim \omega^2$ for $\omega \to 0$. The GLG theory assumes this dependence throughout. If we do the same, and take $f = -2\omega^2 + \omega^4$, we naturally reproduce the GLG result

$$\omega = (1 - h^2)^{1/2} .$$

If we retain the assumption $(1/\lambda^2) \sim \omega^2$, and instead use (5) to (7) to invert the smoothed $\omega(h)$ data of Fig. 2, we find the $f(\omega)$ curves shown as solid lines in Fig. 5. For comparison, we also show the GLG- and BCS-based $f(\omega)$ functions as mentioned above. We note that our higher temperature experimental curve agrees best with the GLG curve, whereas the lower temperature data agree better with the BCS curve, as was to be expected if the model is at all correct. Moreover, if one considers the *difference* between the two temperatures, theory and experiment are in still better agreement. This general agreement is encouraging, but it may be fortuitous. Further work will be required to explore its extent.

Concluding remarks

It would seem overly speculative to proceed further along this line here, in view of the fact that it just may not prove possible to obtain a sufficiently correct description of the situation in this way. However, in view of the difficulty of applying the rigorous Gor'kov theory to a nonhomogeneous superconductor, especially in the nonlinear regime, it has seemed worth exploring these views so long as they lead to even a qualitative picture which is helpful.

This picture is that we can think of a reasonably well-defined local gap which characterizes the superconducting wavefunction more or less as in BCS. If this gap varies substantially in a distance of order ξ_0, the associated increase in energy density is of the order of the condensation energy density. Thus, such rapid variations will occur only for good reason. Examples are in the intermediate state in the presence of a magnetic field, or in the junction region between a superconductor and a nonsuperconducting metal. In the latter case, one might expect a nonzero gap to extend a distance $\sim\xi_0$ through a nonsuperconducting material, as observed in experiments of Meissner[15] and others. Since it seems reasonable to assume that in a nonsuperconducting metal the BCS parameter V may have the wrong sign for superconductivity, the gap could presumably be driven to zero in a distance shorter than ξ_0, with the rate depending on the metal concerned, as observed. (Parmenter[13] reached a similar numerical conclusion, even with his surprisingly short characteristic length $\delta \approx 100$ A.)

If the discontinuity is really great, as at the interface with an insulator, the conduction electrons are simply reflected there. In this case, the "localization energy" is already in the wavefunctions in the normal state, and hence it does not inhibit superconductivity even if there is an almost discontinuous change in gap at the interface. Thus, there is no difficulty in understanding why lead and tin films only 20 A thick have nearly the same T_c as bulk samples, though enormously increased critical fields. This consideration might conceivably bear on the thin persistent filaments in high-field superconductors, if they lie along regions of highly strained material.

If we confine our attention to homogeneous material, then the picture is relatively simple. When a field is applied, it depresses the surface gap to increase the penetration depth and reduce the magnetic energy. If the sample is thin, the gap is decreased throughout, and may be depressed to zero. In a bulk sample, however, the interior must retain the full gap to retain the full condensation energy. The resistance to change of gap in a distance less than ξ_0 then keeps the surface gap from dropping by more than a few percent for typical parameter values.

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