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Magnetic Field Dependence of the Superconducting Energy Gap in Ginzburg-Landau Theory with Application to Al

Abstract: A theoretical calculation is given of the magnetic field dependence of the superconducting energy gap, using the Ginzburg-Landau theory. In addition to depending upon the size of the specimen, the field dependence of the energy gap depends quite strongly on the nature of the boundary conditions. For the usual case with the magnetic field equal on opposite sides of a film, the calculations show that for a ratio of thickness, d, to penetration depth, λ, less than $\sqrt{5}$, the energy gap goes smoothly to zero as the critical field is approached—a second-order phase transition. When $d/\lambda > \sqrt{5}$, the energy gap approaches a finite value as the critical field is approached—a first-order phase transition. Energy-gap measurements for aluminum agree very well with these calculations. When one changes the boundary conditions so that the magnetic field is constrained to be zero on one side of the film, the theory predicts a very different behavior. For this case, at all thicknesses, the energy gap approaches a finite value as the critical field is approached—a first-order phase transition. An experiment involving cylindrical films is proposed to test this latter case. It is shown that in this proposed experiment these boundary conditions are appropriate for predicting the *current* dependence of the energy gap.

Introduction

It is well known that for a bulk superconductor the application of a sufficiently large magnetic field (the critical field) will cause the superconductor to go into the normal state with a corresponding absorption of a latent heat—a first-order phase transition. In terms of the energy-gap model, this means that at the critical field the energy gap drops discontinuously to zero. Since for small specimens the other magnetic properties of superconductors are very different from those in the bulk state, one might speculate that the field dependence of the energy gap of thin specimens is also quite different. We shall show both theoretically and experimentally that this is indeed true.

In the following section, theoretical calculations will be presented showing that the field dependence of the energy gap does depend quite strongly on the size of the specimen and also upon the nature of the boundary conditions. In the subsequent section, direct experimental measurements on the field dependence of the energy gap of aluminum, obtained by the electron tunneling technique, will be presented.

Field dependence of the energy gap: theory

In the absence of a magnetic field one can calculate the magnitude of the energy gap from the theory of Bardeen, Cooper, and Schrieffer[1] (BCS). Since, however, the BCS theory is capable of handling the application of a magnetic field only as a perturbation, one would expect the direct solution of their microscopic equations for the full field dependence of the energy gap to be quite difficult. On the other hand, Gor'kov,[2] starting from the microscopic theory, has derived a set of equations which are identified with the original nonlinear phenomenological equations of

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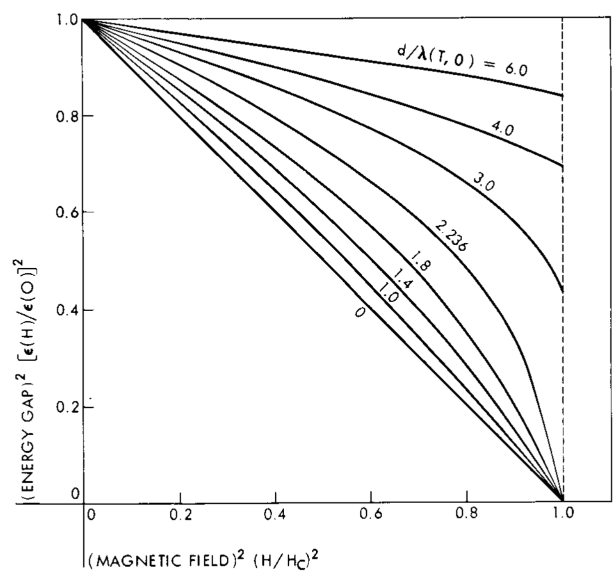
Ginzburg and Landau[3] (GL) and in which he showed that the order parameter ψ of the GL theory is proportional to the energy gap ε. In fact, BCS had previously suggested that the energy gap would be a good order parameter for nonlinear extensions of their theory. Thus the problem of finding the field dependence of the energy gap reduces to solving the GL equations for the order parameter. It is rather a windfall that the application of this suggestion has led to the GL equations because the solutions in various special cases have already been worked out in detail. Also, it should be pointed out that the GL equations are valid for strong fields; therefore one may investigate the behavior of a superconductor near the critical field with confidence. Thus we have a method of calculating the field dependence of the energy gap with as much rigor as would be expected of calculations from the microscopic theory.

There are two limitations of the GL theory. The first is that it is true only in the local limit. This means that the penetration depth λ must be larger than the coherence length ξ, which is not usually true in the bulk state of a superconductor. However, one would expect that for a thin evaporated film the coherence length would be limited by the size, d, of the crystallites. Thus the condition of locality would be $\lambda > d$, which is much easier to satisfy. The second limitation concerns the fact that Gor'kov derived the Ginzburg-Landau equations from the microscopic theory under the condition that the energy gap be small compared to its value at zero temperature and zero field. The theory is therefore valid near the transition temperature, where the gap is small; and for thin films, under suitable boundary conditions, it can be made valid even at $T = 0$ by applying a magnetic field.

Figure 1 **Theoretical energy gap vs magnetic field for the case of equal fields on opposite sides of the film.**



In the GL theory ψ is a function of temperature, magnetic field, and coordinates; therefore, so is the energy gap. In general the energy gap will depend upon position within the superconductor; however, if $d/\lambda(T) \ll 1/\kappa \approx 10$, where κ is the nonlinear coupling constant of the theory, the gap becomes independent of coordinates. In the present paper we will restrict ourselves to this case. It should be remarked here that since the GL equations are differential equations, the field dependence of the energy gap will depend upon the boundary conditions. We shall show that the case of a film with an equal magnetic field on opposite sides is entirely different from the case of a film with unequal fields on opposite sides. Let us take the case of equal fields first.

- *Film with external field equal on opposite sides*

Consider a plate of thickness d with an external field H_0 applied parallel to both surfaces. The GL equations have already been solved for this case by Ginzburg,[4,5] mainly in connection with calculations of the critical field. The two independent equations of condition found by him are

$$H_0^2 = \frac{4\phi_0^2(\phi_0^2 - 1)\cosh^2[\phi_0 d/2\lambda]}{1 - [\lambda/\phi_0 d]\sinh[\phi_0 d/\lambda]} H_{cb}^2 \tag{1}$$

$$H_c^2 = \frac{\phi_c^2(2 - \phi_c^2)}{1 - [2\lambda/\phi_c d]\tanh[\phi_c d/2\lambda]} H_{cb}^2 , \tag{2}$$

where $\phi = \psi(H)/\psi(0)$, λ is the temperature-dependent London penetration depth (or perhaps an effective penetration depth), H_{cb} is the bulk critical field, ϕ_0 is the equilibrium value of ϕ in the presence of H_0, and ϕ_c and H_c are the critical values. Gor'kov's result can be stated as

$$\varepsilon(H)/\varepsilon(0) = \psi(H)/\psi(0) = \phi , \tag{3}$$

where $\varepsilon(H)$ is the energy gap. Equation (1) describes a smooth decrease in the energy gap as the field is increased. The field dependence of the gap for this case has been calculated numerically and is shown in Fig. 1 for various values of d/λ. As $H_0 \to H_c$ the gap approaches a critical value. For this critical gap, the superconductor is in equilibrium with the normal state. Setting (1) equal to (2) gives ϕ_c, from which one can obtain H_c.

For $d/\lambda \ll 1$, the solutions of Eqs. (1) and (2) are

$$\varepsilon(H_c)/\varepsilon(0) = 0 , \tag{4}$$

$$H_c^2 = 24[\lambda/d]^2 H_{cb}^2 \tag{5}$$

$$\left[\frac{\varepsilon(H_0)}{\varepsilon(0)}\right]^2 = 1 - \frac{1}{24}\left(\frac{d}{\lambda}\right)^2\left(\frac{H_0}{H_{cb}}\right)^2 , \tag{6}$$

or, using (5),

$$\left[\frac{\varepsilon(H_0)}{\varepsilon(0)}\right]^2 = 1 - \left(\frac{H_0}{H_c}\right)^2 . \tag{7}$$

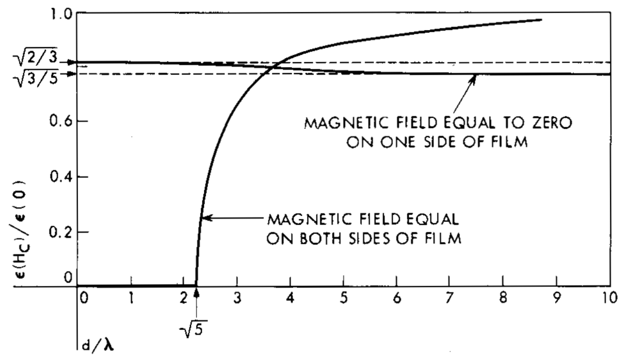


Figure 2 **Theoretical critical energy gap vs d/λ for the cases of equal and unequal fields on opposite sides of the film.**

Thus for very thin films the critical gap is zero, and the field dependence of the gap is independent of thickness when the external field is normalized to the critical field of the film. A closer examination of Eqs. (1) and (2) shows that the critical gap remains zero all the way up to $d/\lambda = \sqrt{5}$. Therefore, the superconducting phase transition should be second order for films thinner than this value. This result was previously given by GL. The value of the critical gap for these boundary conditions has been calculated as a function of d/λ and is shown in Fig. 2.

For small H_0, Eqs. (1) and (2) can be solved to give an expression for the initial decrease of the energy gap which is valid[6] for all d and is shown in Fig. 3:

$$\frac{\varepsilon(H_0)}{\varepsilon(0)} = 1 - D(X)\left(\frac{H_0}{H_{cb}}\right)^2 \tag{8}$$

where

$$D(X) = \frac{1}{8}\,\frac{\sinh X - X}{X\cosh^2 X} \tag{9}$$

$$X = d/\lambda\,.$$

• *Film with the external field equal to zero on one side and equal to H_0 on the other*

Although the case considered previously is the one usually encountered, this particular case is of interest because it will point out how the boundary conditions influence the field dependence of the gap. Also, this analysis will describe the current dependence of the energy gap in a cylindrical film.

This particular case has also been solved by Ginzburg[7] and only the results will be presented here. The first equation of condition corresponding to Eq. (1) is

$$H_0^{\,2} = \frac{4\phi_0^{\,2}(1-\phi_0^{\,2})\sinh^2(\phi_0 d/\lambda)}{1+(\lambda/2\phi_0 d)\sinh(2\phi_0 d/\lambda)}\,H_{cb}^{\,2}\,. \tag{10}$$

The second equation of condition corresponding to Eq. (2) is obtained from Eq. (10):

$$\frac{\partial H_0}{\partial \phi_0} = 0\,. \tag{11}$$

Equations (10) and (11) are easily solved for the limiting case when $d/\lambda \ll 1$:

$$\varepsilon(H_c)/\varepsilon(0) = \sqrt{2/3} \tag{12}$$

$$H_c^{\,2} = (2/3)^3(d/\lambda)^2 H_{cb}^{\,2} \tag{13}$$

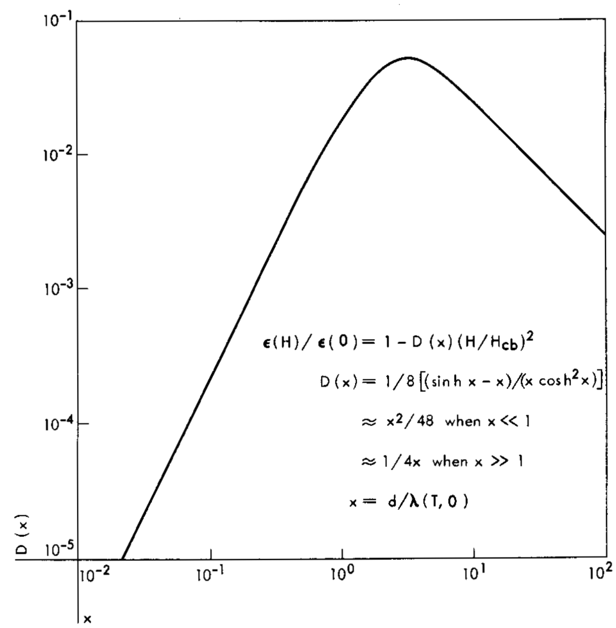
$$2\left[\frac{\varepsilon(H_0)}{\varepsilon(0)}\right]^4\left[1-\left(\frac{\varepsilon(H_0)}{\varepsilon(0)}\right)^2\right]\left(\frac{d}{\lambda}\right)^2 H_{cb}^{\,2} = H_0^{\,2} \tag{14}$$

or using (13)

$$2(3/2)^{3/2}\left[\frac{\varepsilon(H_0)}{\varepsilon(0)}\right]^4\left[1-\left(\frac{\varepsilon(H_0)}{\varepsilon(0)}\right)^2\right] = \left(\frac{H_0}{H_c}\right)^2. \tag{15}$$

There are several very important differences between this and the previous example. The most striking is that the critical energy gap is finite at all thicknesses—the phase transition is always of first order. The critical field is proportional to the thickness and is less than the bulk critical field; whereas in the first case the critical field is inversely proportional to d and is greater than the bulk critical field (see Eq. (5)). The value of the critical gap for these boundary conditions has been calculated numerically from Eqs. (10) and (11) and is shown in Fig. 2. The variation of the critical gap with d/λ is, however, rather slight; for small d/λ it is $\sqrt{2/3}$ and at large d/λ it is $\sqrt{3/5}$.

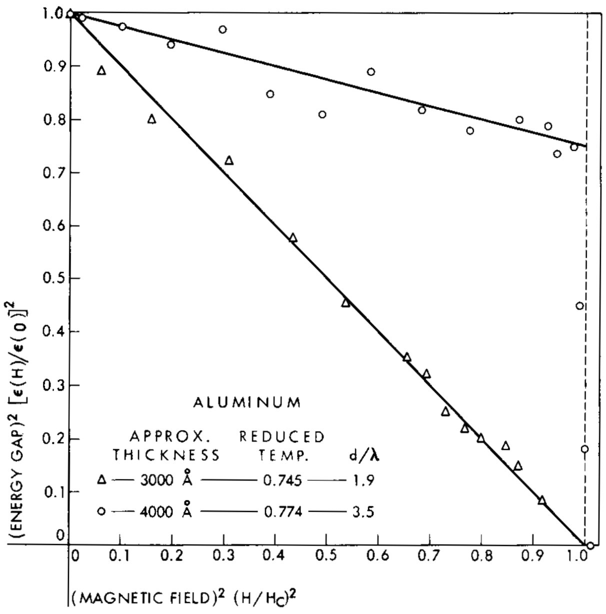
Figure 3 **Theoretical quadratic coefficient vs d/λ for the case of equal fields on opposite sides of the film.**



Direct experimental measurements of the field dependence of the energy gap in aluminum

The energy gap of aluminum was measured directly using the electron tunneling technique[8,9] with sandwiches of Al-Al_2O_3-Pb. The magnetic field was parallel to the plane of the film to within 1°; the experimental conditions correspond to the case of equal fields on opposite sides of the film and the results will be compared with the theory for this case. The normalized energy gap vs reduced magnetic field is shown in Fig. 4 for two films of thickness 3000 A and 4000 A. It should be noticed that for the 3000 A film, the energy gap goes smoothly to zero as the critical field is approached and is described quite well by the equation $[\varepsilon(H_0)/\varepsilon(0)]^2 = 1 - (H_0/H_c)^2$ which agrees with Eq. (7). Measurements on two other films of thickness 500 A and 2000 A gave similar results, and the results on these three films were independent of temperature over the range available ($0.75 < T/T_c < 1.00$). For the 4000 A film an entirely different behavior is observed. The gap initially drops less rapidly with field than in the previous case and, as the critical field is approached, the energy gap drops very abruptly from a finite value to zero; this abrupt change usually occurs within less than 1% of the critical field. The value of the gap just prior to this abrupt drop will be called the *critical gap*. The critical gap of the 4000 A film was found to be a function of temperature.

Figure 4 **Experimental measurement of the energy gap of aluminum vs magnetic field.** *The plotted lines are the best straight lines through the data.* (*Case of equal fields on opposite sides of film.*)



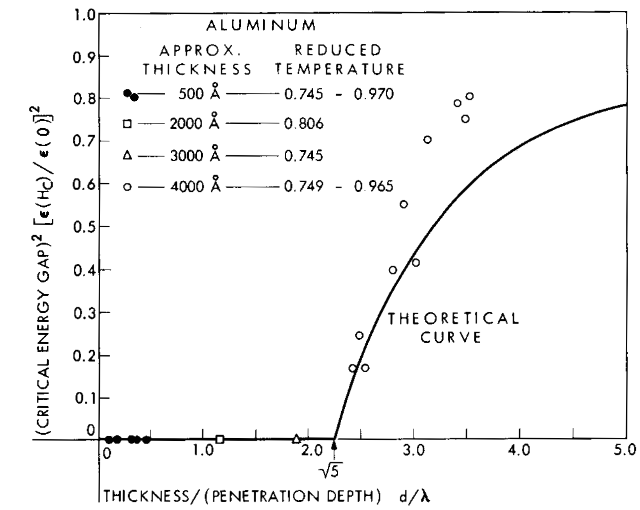


Figure 5 **Experimental measurements of the critical energy gap of aluminum vs d/λ.** (*Case of equal fields on opposite sides of film.*)

Experimental measurements of the critical gap on all four films are shown in Fig. 5 plotted against d/λ. Values of d/λ for the various films were calculated[10] from the measured critical fields using the GL theory. For a given thickness one can vary d/λ over a range of values by changing the temperature since λ is temperature-dependent. The largest attainable value of d/λ is limited by the value of λ at low temperatures, and the smallest value of d/λ is determined by how close one can get to the critical temperature and still have signals big enough to measure. For the 4000 A film, values of d/λ between 2.42 and 3.62 were obtained by varying the reduced temperature between 0.749 and 0.965; and for the four films a total range of d/λ from 0.10 to 3.52 was obtained. It is seen that the data agree very well with the theory when d/λ is less than 2.8. The transition from first to second order is not inconsistent with $d/\lambda = \sqrt{5}$; the data show that it occurs for d/λ between 1.9 and 2.4. For d/λ greater than 2.8 the value of the critical gap deviates from the theoretical curve. This is not surprising, since for aluminum nonlocal effects will become important when d becomes comparable to λ.

Summary and discussion

A theoretical calculation of the magnetic field dependence of the superconducting energy gap was presented. In addition to depending upon the size of specimen, the field dependence of the gap depends quite strongly on the nature of the boundary conditions. The calculations for the case of equal fields on opposite sides of the film were found to be in very good agreement with experimental measurements on films of aluminum. The predictions of zero critical gap for

$d/\lambda \le \sqrt{5}$ and finite critical gap for $d/\lambda > \sqrt{5}$ seem to be entirely borne out.

For the second case, a film with the magnetic field constrained to be zero on one side, the theory predicts that the critical gap will be finite at all thicknesses and the phase transition will be first order. This case has not been examined experimentally. One can set up such boundary conditions by making the film multiply connected. One appropriate experimental arrangement might be an evaporated cylindrical aluminum film on a glass cylinder with a single lead strip evaporated along the axis of the cylinder after oxidation of the aluminum. Then we will have a tunneling sandwich of Al-Al_2O_3-Pb as before, but now we will have the Al in the form of a multiply connected cylindrical film. In the presence of an external axial magnetic field H_0, the field on the outer surface of the aluminum film will be H_0, while the field at the inner surface will be zero since the flux will be excluded. Thus we will have subjected the aluminum film to the boundary conditions of the second case and Eq. (15) will correctly describe the field dependence of the energy gap. As $H_0 \to H_c$, given by Eq. (13), $\varepsilon(H_0) \to \varepsilon(H_c)$ as given by Eq. (12). At H_c the boundary condition $H = 0$ on the inside cannot be maintained since the superconductor goes into the normal state and the flux will leak in. Since the field will then be the same on both sides of the film and less than the critical field for these boundary conditions (see Eq. (5)), the film cannot remain in the normal state. It must return to the superconducting state. Upon further increase of the external field this process will repeat itself, but the field dependence of the energy gap for these latter conditions will have to be calculated anew.

Finally, it should be pointed out that the above boundary conditions can be set up and maintained by passing a current parallel to the axis through the aluminum film in the absence of any external field. This will produce a finite field at the outer surface and zero field at the inner surface. Thus, for this geometry, Eqs. (12) through (15) correctly describe the *current* dependence of the energy gap of the aluminum film (when $d/\lambda < 1$) where H_0 is now equal to $2I/r$, where r is the radius of the cylindrical film. The energy gap will decrease as the current is increased, and as the critical current (given by Eq. (13)) is approached the energy gap will approach the critical value (given by Eq. (12)). At the critical current the energy gap will discontinuously to zero.

Acknowledgments

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References and footnotes

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10. There are as yet no data relating d/λ to d for aluminum. It might be argued that using the Ginzburg-Landau theory to calculate d/λ from the critical field, amounts only to a self-consistency check; however, estimates of d/λ using the approximate thickness and the bulk penetration depth, modified by size effects according to Tinkham [*Phys. Rev.* **110,** 26 (1958)], give values which do not differ by more than 20 to 30% from those quoted here.

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