Energy Gap Measurements by Tunneling between Superconducting Films. II. Magnetic Field Dependence

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The magnetic field dependence of the energy gap 2Δ was measured by the electron tunneling technique over the temperature interval $T_c \geq T \geq 0.7\ T_c$ on eight aluminum films ranging in thickness d from 420 to 9850 Å. Measurements of the reduced energy gap versus the reduced field can be represented as a family of curves with a single parameter d/λ. For $d/\lambda > 1$ the $[\Delta(O,T)]^2$ versus $[H/H_c]^2$ curves have an initial small negative curvature and then drop abruptly at H_c. For $d/\lambda < 1$ the curvature is always positive, decreasing in magnitude as the field increases. For $d/\lambda \approx 1$ we obtain an almost straight line. The results are qualitatively as predicted by the Ginzburg-Landau (GL) equations for $d/\lambda \geq 1$ but not for $d/\lambda < 1$. It is suggested that this discrepancy with solutions of the GL equations may arise from the breakdown of the assumption that the order parameter is independent of position. Measurements on a lead film of thickness 1000 Å at $T/T_c = 0.14$ showed that the energy gap goes smoothly to zero with positive curvature as H approaches H_c.

I. INTRODUCTION

THE properties of a superconductor are largely determined, according to the Bardeen, Cooper, Schrieffer (BCS) theory,[1] by the gap in the spectrum of available energy states. The existence of such a gap has been inferred or measured in a number of ways, but perhaps the simplest method which gives precise results is that of electron tunneling between superconductors separated by a thin dielectric layer. Giaever[2] and Nicol *et al.*,[3] in their remarkable experiments showed that certain characteristic points on the current-voltage (I–V) curves between such pairs of superconductors could be related to the energy gap 2Δ of the BCS theory.

Using this method, Giaever and Megerle[4] showed that the energy gap of a thin film of aluminum decreased smoothly as the magnetic field (applied parallel to the film) was increased to its critical value H_c. This behavior was interpreted by Douglass[5] in terms of the Ginzburg-Landau[6] (GL) theory using Gor'kov's[7] result that the order parameter of the GL theory is proportional to Δ. This theory predicts a continuous decrease of Δ to zero as $H \to H_c$ (i.e., a second-order phase transition) for films whose thickness d is less than $\sqrt{5}$ times the penetration depth λ and a discontinuous drop in Δ at H_c (i.e., a first-order phase transition) when $d > \sqrt{5}\lambda$. Tunneling measurements by Douglass[8] and by Douglass and Meservey,[9] as well as interpretations of the thermal conductivity measurements of Morris and Tinkham,[10] all tended to confirm the predictions of this theory in the region of its validity: near T_c and in the local limit of $\lambda \gg \xi$, where ξ is the effective coherence length. No complete microscopic theory of the effect of the magnetic field on the energy gap has been published yet. Gupta and Mathur[11,12] and more recently Nambu and Tuan[13,14] and Maki[15] have given calculations which predict the behavior in certain important limiting cases. Bardeen,[16] using a semimicroscopic approach, has predicted that thin films, which at higher temperatures have a second-order transition, should have a first-order transition at low temperatures.

The present investigation was undertaken to measure the energy gap in thin films as a function of magnetic field, thickness, and temperature by means of the electron-tunneling technique.

II. EXPERIMENTAL MEASUREMENTS

The magnetic field dependence of the energy gap 2Δ was measured in thin evaporated films of aluminum with lead as a reference metal. The eight aluminum films ranged in thickness from 420 to 9850 Å and measurements were made from the transition temperature down to $T = 0.87°$K ($t = T/T_c \approx 0.7$).

The preparation of the junctions and the measure-

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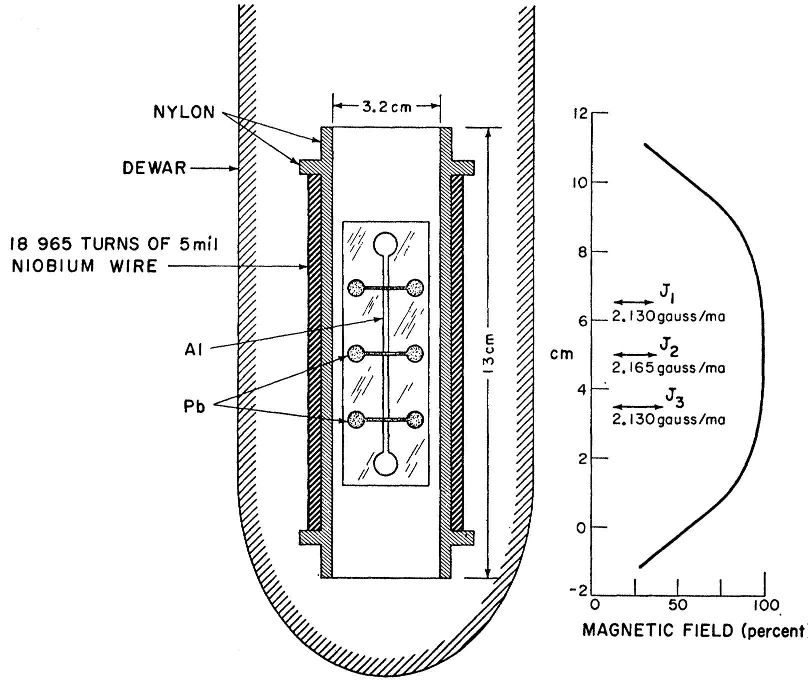

FIG. 1. Experimental configuration showing sample, solenoid, and axial field variation.

ment of the film thickness, the temperature, and the current-voltage characteristic were described in the preceding paper (hereafter referred to as I). The objective criterion for determining the energy gap from the I–V curves is shown in Fig. 4 of I. It is the number of electron volts corresponding to the difference in voltage from the first point of inflection of the I–V curve (starting from $I=V=0$) to the next point of equal slope. This criterion gives the correct limiting value for wide gaps with large values of negative resistance where there is little ambiguity about the magnitude of 2Δ. For small values of 2Δ and where there is no negative resistance region, this criterion is no more than a self-consistent interpolation scheme. A somewhat fuller discussion of the criterion is given in I.

The magnetic field was obtained by positioning the junction along the axis of a superconducting solenoid with the field parallel to the plane of the thin film (Fig. 1). The solenoid was 10 cm long with an inside diameter of 3.81 cm. For the earlier measurements it had 18 965 turns of 5-mil-diam unannealed Nb wire and for later measurements 6360 turns of 10-mil-diam Nb-Zr alloy wire (25% Zr). The magnetic field of the solenoid was calibrated at liquid-helium temperatures with a fluxmeter and the calibration agreed with the calculated field for such a solenoid of nonsuperconducting wire within 1%. Neither solenoid showed a significant amount of hysteresis or flux trapping up to the maximum field used in these experiments. The measured variation of the magnetic field along the axis as shown in Fig. 1 was used to determine the actual field applied at the position of the junction.

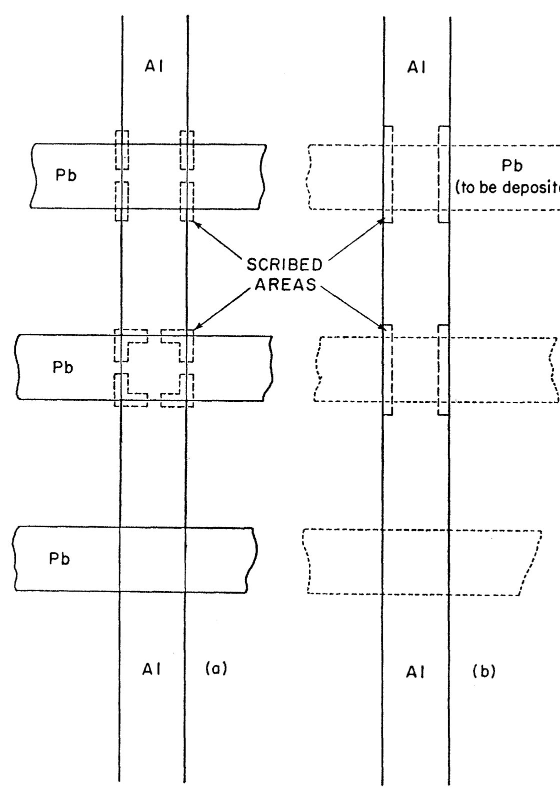

FIG. 2. Tunneling junctions in "edge" experiment. (a) Pb and Al edges "removed." (b) Al edges "removed" prior to Pb evaporation.

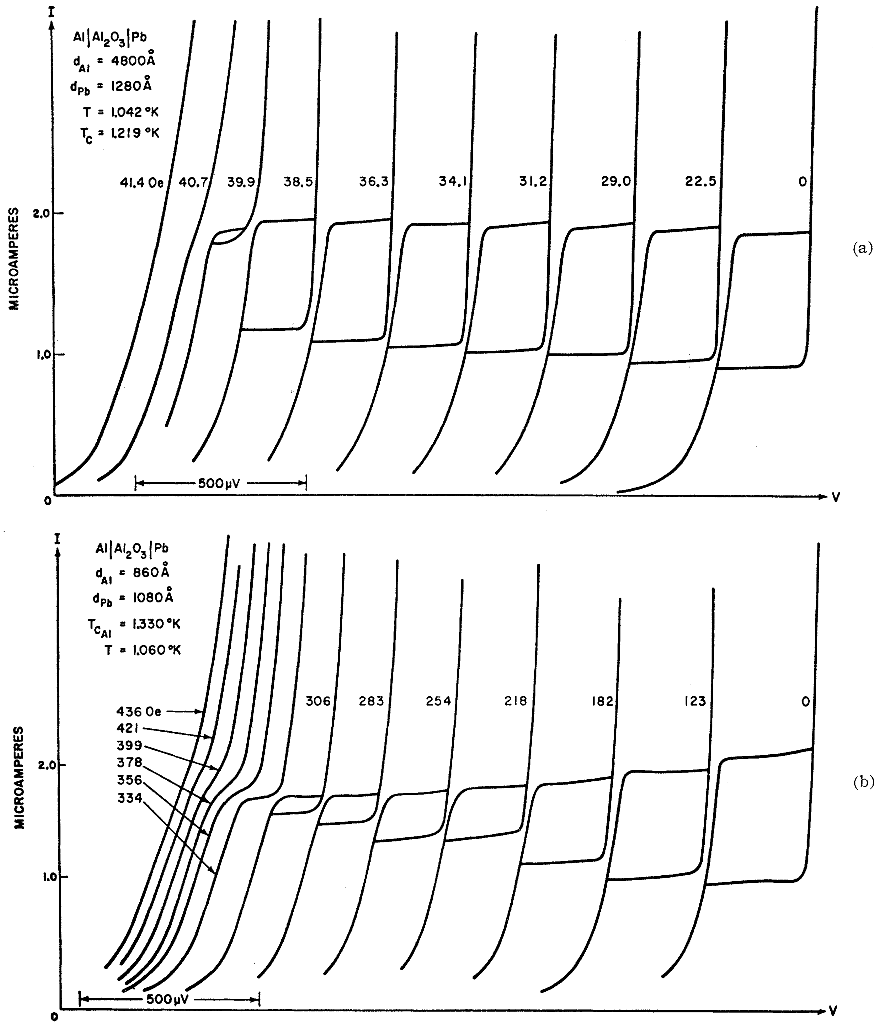

FIG. 3. Current-voltage curves versus magnetic field (using constant-current generator) for (a) $d_{Al}=4800$ Å and (b) $d_{Al}=860$ Å.

One question which has been raised is whether the actual magnetic field present in the dielectric tunneling layer is the same as the applied external field considering that the superconducting films are only separated by about a 10–30 Å dielectric layer. However, since there is no circulating current flowing between the two superconducting films, Ampere's law and the finite width of the film imply that the magnetic field between the films equals the applied field. Thus for tunneling barriers, across which a finite voltage drop exists, this result should be rigorously true. To check this point experimentally, an aluminum strip 4010 Å thick was oxidized and then crossed with a lead strip 1300 Å thick in one place and with a lead strip 3600 Å thick in another. The variation of the aluminum energy gap with magnetic field in these two junctions was found to be essentially identical.

Early in the investigation it was considered likely that the measured energy gap would be effected by the nature of the edges of the evaporated strips. Such effects are well known in measurements of critical currents and critical fields in thin films.[17] Control experiments were tried in the present investigation to determine the importance of edge effects on the measured energy gap. In one instance [see Fig. 2(a)], most of the edge area of a junction was separated from the remaining area by scribing with a diamond. Although the edge area of the film was decreased by a factor of about 5, there was no significant difference in the I–V curves between this junction and a neighboring unscribed junction for fields up to the critical field. In another instance, the edge of an aluminum film, about 3000 Å thick, was scribed as shown in Fig. 2(b) before the lead film was evaporated. Again, no significant difference was found between this junction and a neighboring control junction. Thus it seems reasonable to conclude that the edges of the films had little effect on the energy gaps measured in these investigations.

[17] C. J. Kahan, R. B. Delano, Jr., A. E. Brennemann, and R. T. C. Tsui, IBM J. Res. Develop. 4, 173 (1960).

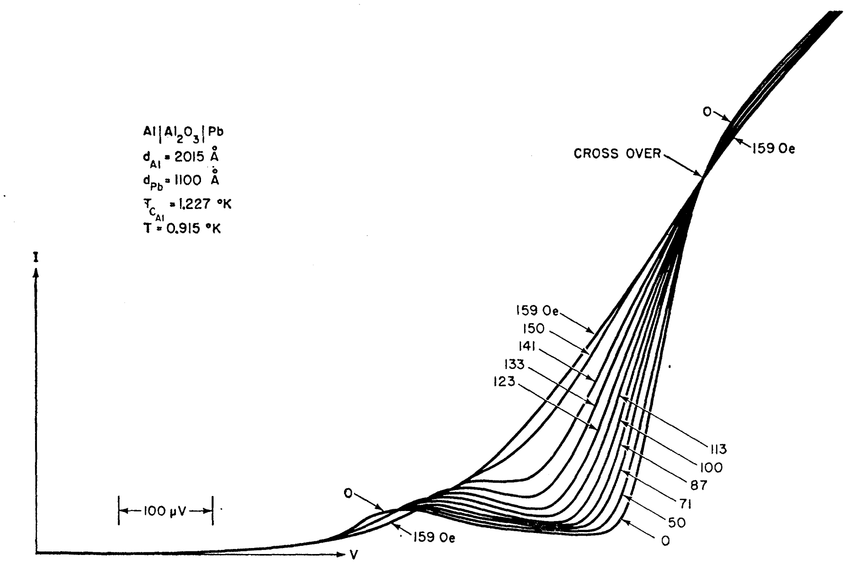

FIG. 4. Current-voltage curves versus magnetic field for d_{Al}= 2015 Å (using constant-voltage generator).

III. EXPERIMENTAL RESULTS

Aluminum. Figures 3 and 4 show typical recorder traces of the $I-V$ curves. Figure 3(a) shows a thick aluminum film (d=4800 Å) and Fig. 3(b) a thin aluminum film (d=860 Å); both these curves were made with a constant-current source. On the other hand, Fig. 4 was made with a nearly constant voltage source so that detail in the negative resistance region is obtained. (Note the crossover points.)

Figure 5 shows the measured energy gap as a function of magnetic field and reduced temperature for eight different film thicknesses. Values of $2\Delta(0,T)$ are given in Table I. The value of the magnetic field for which the energy gap goes to zero is defined as the critical field and was chosen by extrapolation of the curves in Fig. 5. The values of these critical fields are listed in Table I and plotted in Fig. 6 for each film. Figure 7 shows the thickness dependence of H_c for two selected temperatures as obtained from the smoothed curves of Fig. 6.

Lead. Because of the Bardeen calculation[16] predicting a first-order phase transition in a thin film at low temperatures it was decided to prepare a $Pb/Al_2O_3/Pb$ junction to test this. After evaporating a 1000 Å film of Pb on a microscope slide, a small quantity of Al was evaporated and allowed to oxidize completely; then a second 1000 Å Pb film was deposited. The $I-V$ curves that were obtained at T=0.87°K are shown in Fig. 8. The value of the gap was chosen by extrapolation to the V axis and the results are plotted in Fig. 9.

IV. ANALYSIS OF RESULTS

In this section we partially analyze the data and present it in such a form that its general features can be readily grasped. To compare the results in a more compact form, we now wish to normalize with respect to the critical fields, H_c. This quantity was determined by extrapolating the curves in Fig. 5 to the field where the energy gap becomes zero. Actually the values of Δ near the critical field are not very reliable because the negative resistance region in the tunneling characteristics disappears and the somewhat arbitrary criterion of selecting the gap is not necessarily correct. However, in each case it was found that at a somewhat higher field there was no effect on the $I-V$ curve with magnetic field. This evidently sets an upper bound on this definition of H_c which was always less than 10% above the extrapolated value and usually less than 5%. It is expected that there should be some rounding of the energy gap curves near H_c caused by film inhomogeneities, so that it is probable that the extrapolated values rather than this upper bound are closer to the true H_c.

The penetration depth which will be used in the analysis is a semiempirical one given by the expression

$$\lambda=\lambda_0(1-t^4)^{-1/2}(1+\xi_0/l)^{1/2},$$

where

$$\lambda_0=5.3\times10^{-6}\text{ cm}$$
$$\xi_0=1.6\times10^{-4}\text{ cm}$$
$$l=\text{mean free path}.$$

Here the value of the bulk penetration depth $\lambda_0(1-t^4)^{-1/2}$ is the expression used by McClean[18] to describe his recent measurements on the penetration depth in aluminum. The expression in the second parenthesis is the approximate correction for mean-free-path effects suggested by Tinkham[19] on the basis of the Pippard theory and the value of ξ_0 is taken from Bardeen and Schrieffer.[20] The effective electronic mean free

[18] W. L. McLean, Proc. Phys. Soc. (London) **A79**, 572 (1962).
[19] M. Tinkham, Phys. Rev. **110**, 26 (1958).
[20] J. Bardeen and J. R. Schrieffer, *Progress in Low Temperature Physics* (North Holland Publishing Company, Amsterdam, 1961), Vol. III, p. 243.

TABLE I. Aluminum properties.

d (Å)	T_c (°K)	$2\Delta(0)$ (μV)	t	H_c (Oe)	$\left.\frac{d(\phi)^2}{d(h^2)}\right\rvert_{h^2=0}$	D_{exp} ($\times10^{-2}$)	$\frac{d}{\lambda}$	$\frac{1}{d^3}\frac{1-t^4}{1+t^4}\left(\frac{d\phi^2}{d(H^2)}\right)_{H^2=0}$ [a]
420	1.405	400	1.624	564	−1.7	0.93	0.117	5.20×10^{10}
			0.874	500	−1.5	0.16	0.082	2.10
			0.943	411	−1.8	0.066	0.058	1.70
			0.981	307	−2.2	0.016	0.034	0.58
			0.991[b]	242	−2.5	...	...	1.00
860	1.333	345	0.660	461	−1.8	1.30	0.330	1.30
			0.795	411	−1.4	0.58	0.280	0.57
			0.878	375	−1.6	0.27	0.230	0.71
			0.950	262	−1.4	0.098	0.160	0.32
			0.990[b]	26.4	−1.2	...	...	...
1280	1.246	351	0.717	232	−1.7	3.8	0.560	0.49
			0.899	154	−1.9	1.7	0.390	0.82
			0.984	57.4	−2.2	0.34	0.160	1.10
1880	1.241	294	0.703	224	−1.5	3.6	1.000	0.28
			0.727	211	−1.4	3.3	0.980	0.26
			0.845	164.5	−1.3	2.0	0.810	0.24
2020	1.227	354	0.746	153	−1.0	4.2	1.060	0.23
			0.852	116	−1.3	3.5	0.880	0.36
			0.915	87.8	−1.0	1.7	0.700	0.32
			0.986	35.7	−1.3	0.41	0.300	0.35
4520	1.217	346	0.740	66.2	−0.38	8.5	3.35	...
			0.751	63.6	−0.37	8.5	3.31	
			0.825	49.5	−0.39	8.0	2.93	
			0.875	39.3	−0.42	8.2	2.58	
			0.898	35.5	−0.46	6.5	2.37	
			0.918	30.0	−0.51	7.1	2.16	
			0.929	28.2	−0.54	6.3	2.02	
			0.941	25.0	−0.62	6.6	1.86	
			0.950	22.5	−0.67	6.5	1.72	
			0.962	20.0	−0.78	5.5	1.52	
			0.967	18.3	−0.80	6.6	1.42	
			0.973	16.4	−0.90	4.8	1.29	
4800	1.219	342	0.736	62.4	−0.31	8.1	3.66	
			0.855	40.7	−0.39	8.3	2.97	
			0.934	24.0	−0.40	5.7	2.13	
			0.979	12.6	−0.97	8.3	1.24	
			0.9966[b]	9.44	−1.4	...	...	
9850	1.225	314	0.739	55.2	−0.22	7.3	9.61	
			0.860	33.4	−0.20	6.0	7.72	
			0.949	14.9	−0.34	7.6	4.99	
			0.9877[b]	6.90	−0.94	...	...	

[a] $\phi=\Delta(T,H)/\Delta(T,0)$.
[b] Data for $t\geq0.99$ were not used in the computations because the uncertainty in the value of T_c leads to excessively large uncertainties in λ.

path in thin evaporated films is known to be approximately equal to the film thickness. For simplicity, we set $l=d$.

It is convenient to present the critical field data by normalizing with the expression for the critical field of a thin film derived from the London equation.[21]

$$H_{cL}=H_0(1-t^2)[1-(2\lambda/d)\tanh(d/2\lambda)]^{-1/2}.$$

Here, H_0 is the bulk critical field at $T=0$°K and $(1-t^2)$ gives the temperature dependence of the bulk critical field with an error of at most 4%. Since the penetration depth λ is the semiempirical one described above, H_{cL} could be called the modified London critical field. Figure 10 shows the normalized values of the critical field obtained from the present energy gap measurements and from the recent resistance measurements of aluminum films by Khukhareva.[22] The values of H_c as measured by the two methods are in generally good agreement with each other. For $d/\lambda>0.1$ the energy gap measurements agree with H_{cL}, and the resistance measurements are only slightly higher. Because of the rather arbitrary nature of the chosen penetration depth, the comparison of the absolute value of H_c with theory is, perhaps, not significant in choosing between the GL and the modified London expression. This uncertainty in λ is especially true for $d/\lambda<0.1$ and probably explains the large scatter and perhaps the trend downward in H_c/H_{cL}. The most important point of the above analysis is, however, that it shows that H_c for $\Delta=0$ is essentially the same quantity as the H_c of resistance measurements.

We now proceed to normalize the magnetic field to the experimental critical field and replot the results. In Fig. 11 plots for three different films show the three

[21] We use the expression for the critical field of a thin film with zero surface energy given in F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1960), Vol. I, p. 131.

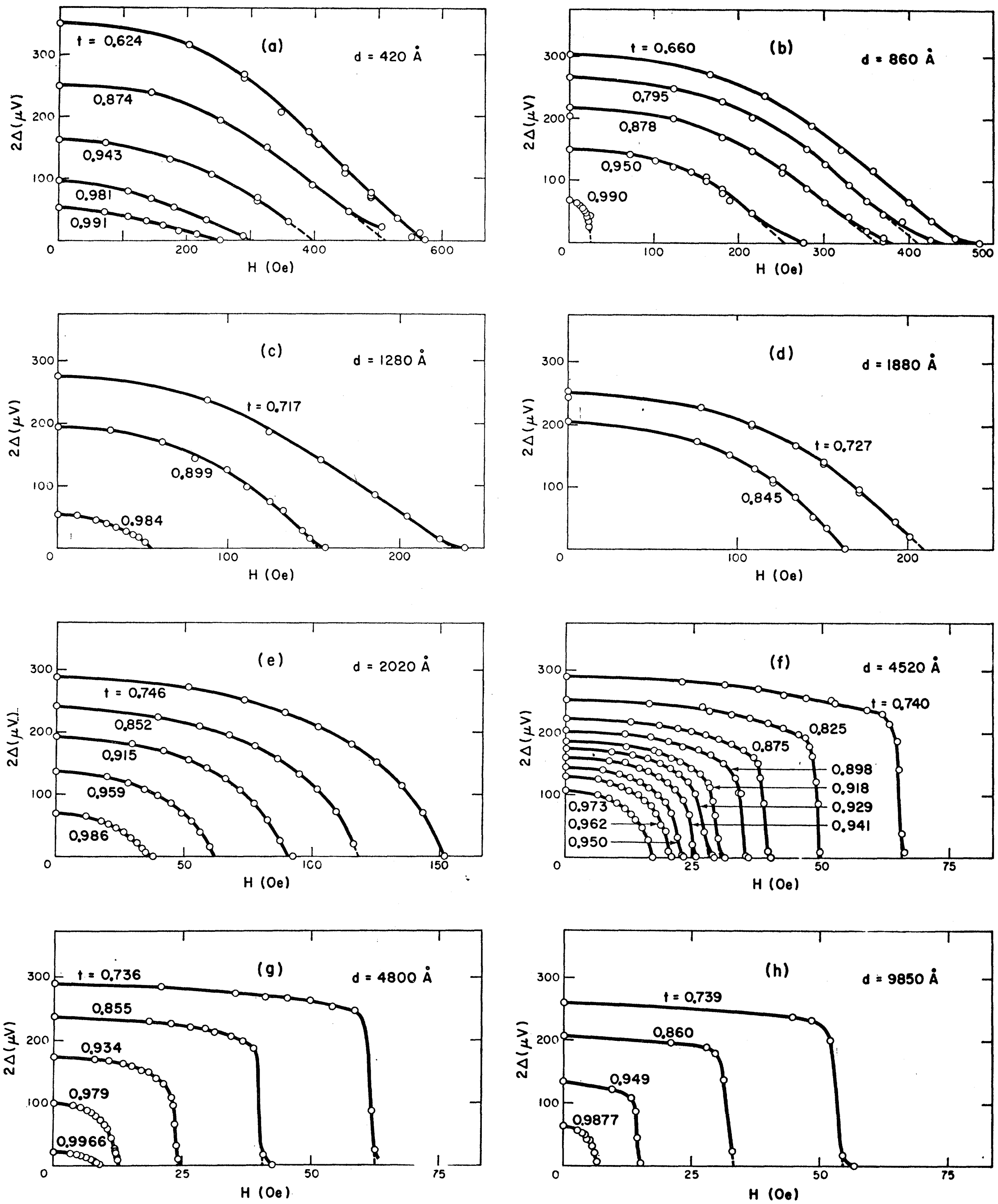

FIG. 5. Energy gap of Al versus magnetic field at various temperatures for eight different Al thicknesses.

characteristic shapes which are obtained. Study of such curves shows that all the results fall very close to a set of universal curves characterized by the single parameter d/λ. For large values of d/λ the curves have negative curvature and for small values positive curvature; whereas for values of $d\approx\lambda$, we obtain almost-straight lines. To test the adequacy of this single parameter, the value of $[\Delta(h)/\Delta(0)]^2$ for $(H/H_c)^2=0.5$ is plotted as a function of d/λ. This is, of course, an arbitrary criterion, but a convenient one which differentiates strongly between the various curves. Figure 12 demonstrates that within the scatter of the data the curves are ordered monotonically in the parameter d/λ and presumably $\Delta(H)/\Delta(0)=f(H/H_c,d/\lambda)$.

V. COMPARISON WITH THEORY

Ginzburg-Landau-Gor'kov Theory

The Ginzburg-Landau phenomenological theory supplemented by the result of Gor'kov that the order parameter is proportional to the energy gap has been used by Douglass[5] to predict the variation of the energy gap with magnetic field. Under the assumption that the gap is independent of position the field dependence is

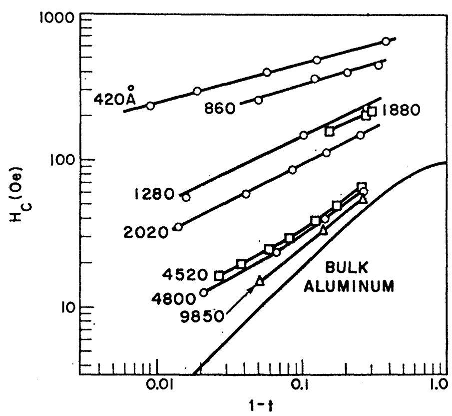

FIG. 6. Critical field versus $1-t$ for Al in various thicknesses.

given implicitly by the expression

$$H^2=\frac{4\Phi^2(\Phi^2-1)\cosh^2[\Phi d/2\lambda]}{1-(\lambda/\Phi d)\sinh[\Phi d/\lambda]}H_{cb}^2,$$

and the critical field by

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

Here, $\Phi=\Delta(H)/\Delta(0)$, H_{cb} is the bulk critical field, H_c is the critical field of the film, and Φ_c is the value of Φ at $H=H_c$.

These nonlinear equations predict that for thick films (actually $d/\lambda>\sqrt{5}$) the energy gap decreases with magnetic field, but is still finite at the critical field, at which point it drops discontinuously to zero (first-order transition). For $d/\lambda<\sqrt{5}$ the energy gap is depressed continuously and reaches zero at the critical field (second-order transition). This theoretical behavior is shown in Fig. 13 and certainly is qualitatively similar to the observed behavior for the thicker films [see Fig. 11(c)]. The experimental results are not discontinuous but for comparison with the theory we define the experimental

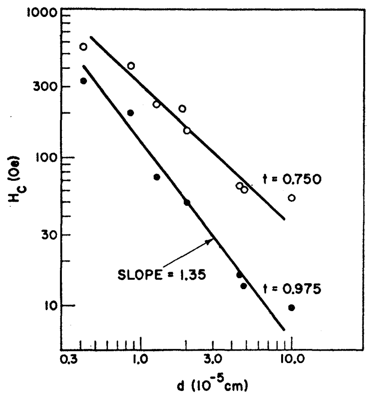

FIG. 7. Critical field versus d for $t=0.75$ and 0.975.

critical energy gap by extrapolating the low-field values of these curves until they intersect the ordinate at the critical field. A comparison with the Ginzburg-Landau prediction is shown in Fig. 14. For values of $d/\lambda>1$ the behavior is qualitatively as predicted although the change from first-order to second-order transitions occurred at a value of d/λ less by a factor of 2 than the predicted $\sqrt{5}$. This numerical disagreement may not be significant because of the uncertainties in the choice of λ. For values of $d/\lambda<1$, however, the prediction is qualitatively wrong, a result corresponding to the positive curvature shown in Fig. 11(a). In previous publications[8,9] it was assumed that in such thin films the proper critical field for normalization was to be obtained by extrapolation of the initial linear portion of the Δ^2 versus H^2 curves and that the long concave "tail" was caused by edge effects. However, in view of the subsequent control

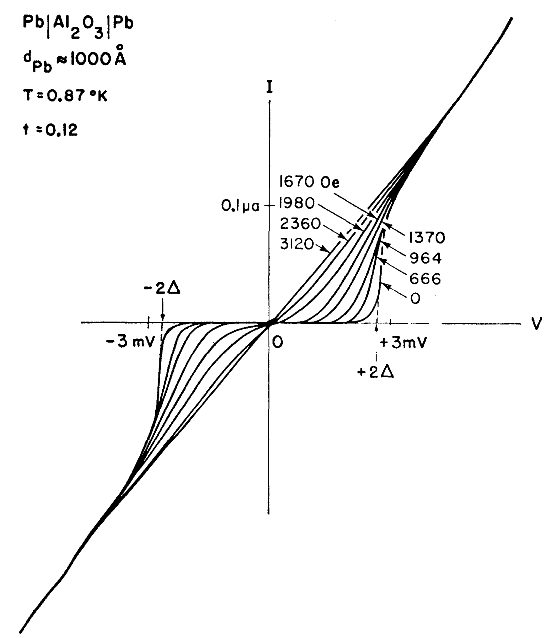

FIG. 8. Current-voltage curves for a $Pb/Al_2O_3/Pb$ junction for various magnetic fields.

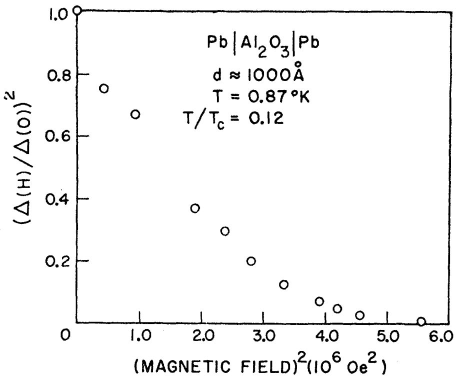

FIG. 9. Energy gap versus magnetic field for Pb thickness of 1000 Å.

experiments on edge effects and the comparison of the critical fields with other results, we believe that these "tails" are not a spurious, secondary effect, but show the true behavior of the energy gap in thin films which does *not* agree with the above equations.

Another quantity to be compared with theory is the change of the energy gap with magnetic field for small fields. The above equations predict

$$\frac{\Delta(H)}{\Delta(0)}=1-D\left(\frac{d}{\lambda}\right)\left(\frac{H}{H_{cb}}\right)^2,$$

where

$$D\left(\frac{d}{\lambda}\right)=\frac{1}{8}\frac{\sinh(d/\lambda)-d/\lambda}{(d/\lambda)\cosh^2(d/\lambda)}.$$

Figure 15 compares the experimental results with this theoretical expression and shows a qualitative agreement.

Nambu and Tuan[13] have recently performed a calculation of the field dependence of the gap in the thin limit ($d<\lambda$) using a discrete quantization model. Their expression is

$$\Delta(T,H)=\Delta_{00}R(H^2)F(X),$$

where Δ_{00} is the value of the gap at $T=H=0$, $X=t/R(H^2)$, $t=T/T_c$, $F(t)$ is the expression for the temperature dependence of the reduced gap from the BCS theory, and $R(H^2)$ is given by

$$\exp\left[-\frac{8}{\pi^4}\left(\frac{e}{\hbar c}\right)^2\xi_0 d^3H^2\right].$$

They have considered the limit of these equations for

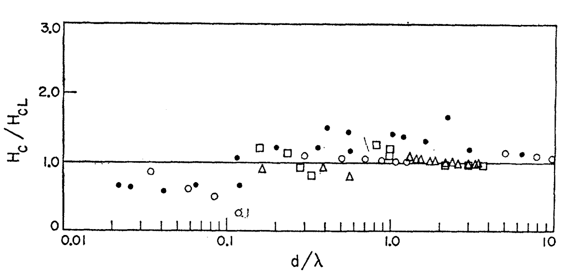

FIG. 10. H_c/H_{cL} versus d/λ (open points). H_c is the magnetic field at which the energy gap$=0$. Solid points give the critical field from Khukhareva's resistance measurements.

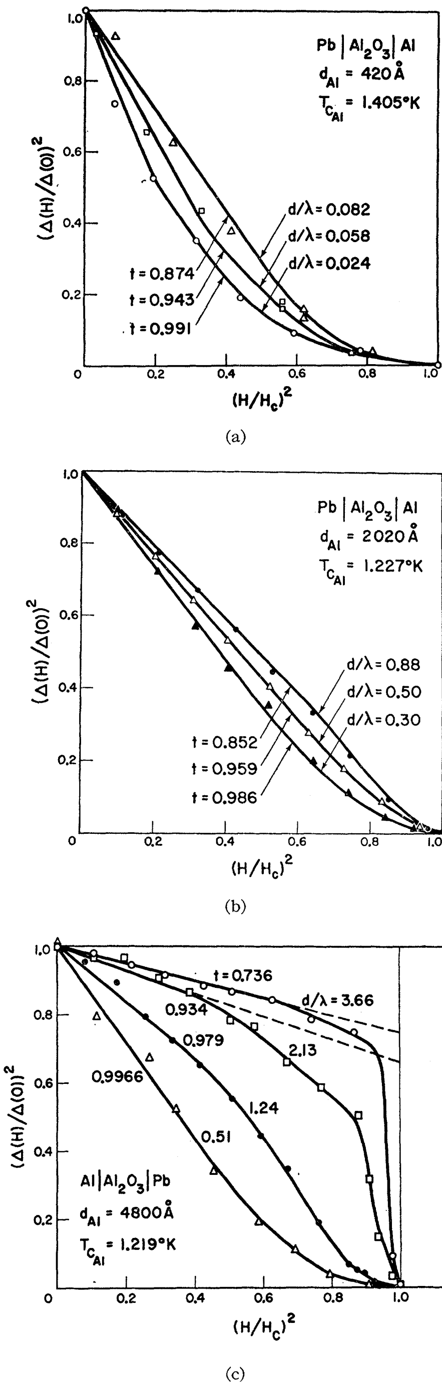

FIG. 11. Reduced gap versus reduced magnetic field of Al for (a) $d=420$ Å, (b) $d=2020$ Å, and (c) $d=4800$ Å.

$T \to 0$ and have made a comparison to the lead data of Fig. 9 and find reasonable agreement.

Defining a quadratic coefficient Q by

$$\Delta(T,H)/\Delta(T,0) = 1 - QH^2 + \cdots,$$

then Q is easily shown to be

$$Q = -\frac{1}{\Delta(T,0)}\left(\frac{\partial\Delta(T,H)}{\partial H^2}\right)_{H^2=0}$$

$$= \frac{8}{\pi^4}\left(\frac{e}{\hbar c}\right)^2 \xi_0 d^3 \left\{1 - \frac{t}{F(t)}\frac{\partial F(t)}{\partial t}\right\}.$$

The temperature dependence of the gap $F(t)$ can be approximated quite accurately by $(1-t^4)^{1/2}$. Using this, one obtains

$$Q = \frac{8}{\pi^4}\left(\frac{e}{\hbar c}\right)^2 \xi_0 d^3 \frac{1+t^4}{1-t^4}.$$

This means that the quantity

$$2\frac{1-t^4}{1+t^4}\frac{1}{d^3}Q = \frac{16}{\pi^4}\left(\frac{e}{\hbar c}\right)^2 \xi_0$$

$$= 0.62\times10^{10}\ \mathrm{G}^{-1}\ \mathrm{cm}^{-1}\ \text{(aluminum)}$$

is a constant. Experimental measurements of this

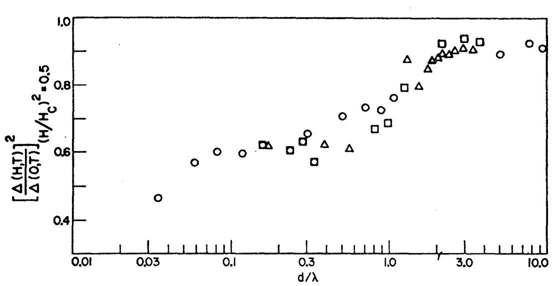

FIG. 12. Reduced gap at H/H_c versus d/λ.

quantity for the thinnest films, for which this calculation is presumably good, are listed in Table I. The values range from 0.2×10^{10} to 5.0×10^{10} which indicates agreement in order of magnitude.

V. DISCUSSION

This investigation shows that for aluminum films at a temperature $T > 0.7T_c$ the qualitative features of the position-independent solutions of the Ginzburg-Landau theory are observed. Apparently the sudden drop in Δ observed in thick films at H_c corresponds to a first-order phase transition and the smooth approach of Δ to zero observed in thin films at H_c corresponds to a second-order phase transition. On the other hand, the behavior of films with $d/\lambda < 1$ is not as predicted, the values of Δ

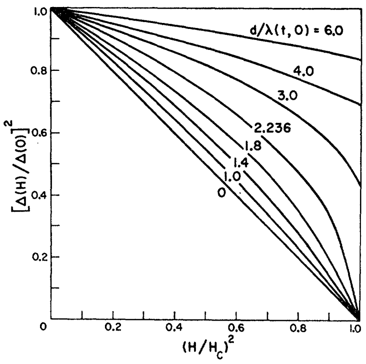

FIG. 13. Reduced gap versus reduced field for various d/λ according to GL theory.

being too low for all values of field up to H_c, which is also significantly less than predicted. This lack of agreement is particularly serious since the limit of $d/\lambda \to 0$ is the one in which these solutions of the GL theory were expected to be most valid.

One possible explanation is that the theory of superconducting tunneling is incorrect when a magnetic field is applied to the sample. In other words it may be an error to assume that the application of a magnetic field affects only the energy gap and not the dynamics of electron tunneling. Such an explanation seems unlikely, however, because of the range of barrier heights used (impedances varying from a few ohms to a few thousand ohms) and the fact that the results are all described by the single parameter d/λ.

A more likely possibility for the above discrepancy is that the assumption of a position-independent energy gap in thin films is incorrect. It was suggested by Gor'kov[23] that in thin films of very short mean-free path, l, the GL coupling constant κ will be greater than $1/\sqrt{2}$ (since $\kappa \approx \kappa_{\mathrm{bulk}}\,[1+\xi_0/l]$), and their behavior would be determined accordingly. In bulk superconductors with $\kappa > 1/\sqrt{2}$ (now called superconductors of the second kind) and in a sufficiently high magnetic field Abrikosov[24] showed that spatial modulation of the order

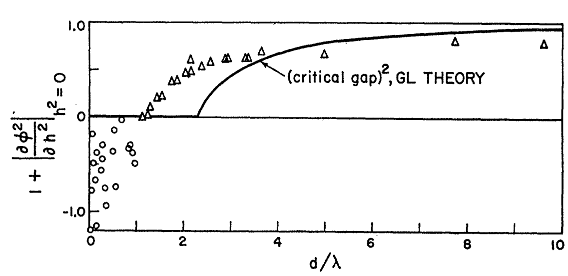

FIG. 14. $1+(\partial\phi^2/\partial h^2)_{h=0}$ versus d/λ, where $\Phi = \Delta(H,T)/\Delta(0,T)$, $h = H/H_c$.

[22] I. S. Khukhareva, Zh. Eksperim. i Teor. Fiz. **43**, 1173 (1962) [English transl.: Soviet Phys.—JETP **16**, 828 (1963)].
[23] L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. **37**, 1407 (1959) [English transl.: Soviet Phys.—JETP **10**, 998 (1960)].
[24] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. **32**, 1442 (1957) [English transl.: Soviet Phys.—JETP **5**, 1174 (1957)].

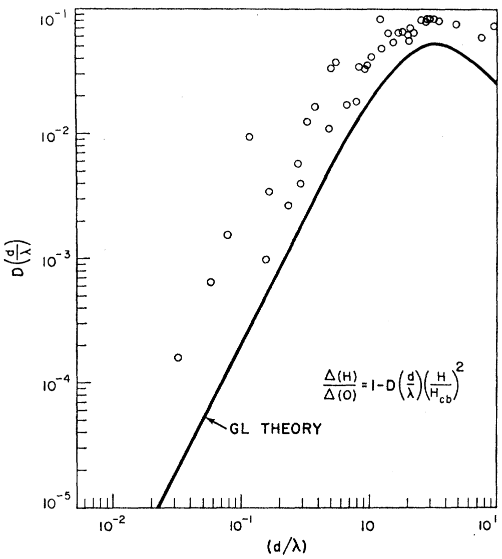

FIG. 15. Quadratic coefficient of the magnetic-field variation of the energy gap versus d/λ.

parameter ψ (or from Gor'kov's result, Δ) led to a state of lower energy because the condition $\kappa > 1/\sqrt{2}$ makes possible a "negative surface energy" between superconducting and normal regions. On the other hand, in solving their equations in the thin-film limit, GL assumed that the order parameter was spatially invariant, but on reflection it appears that this spatial invariance is only reasonable in the direction perpendicular to the plane of the film. Actually, we would expect that the solutions obtained by Abrikosov, in which there are periodic "vortex" solutions in the two directions perpendicular to the applied magnetic field, would reduce to solutions which are singly periodic in the direction of the film width. Although solutions of the GL equations of this form have not yet been found for thin films, they must exist and it is plausible that these solutions will be a state of lower energy and therefore stable.[25] In such a case the tunneling experiments would measure an average energy gap which would decrease rapidly at low fields and decrease more slowly at higher fields. Qualitatively, at least, this agrees with the present results.

In regard to the measurement of the field dependence of the energy gap in the 1000 Å lead film at $T=0.87°K$, no evidence for a first-order phase transition was found. The calculation of Bardeen predicts a discontinuous drop in reduced gap for Pb ($N(0)V=0.39$) from 0.42 to zero. Figure 9 shows that the gap goes smoothly to zero suggesting a second-order phase transition.

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[25] Actually, A. B. Pippard [Phil. Mag. **43**, 273 (1952)] suggested that the thin film magnetization curves measured by J. M. Lock [Proc. Roy. Soc. (London) **A208**, 391 (1951)] (which resemble those of superconductors of the second kind) were to be explained by "negative surface energy."