Josephson tunneling studies of magnetic screening in proximity-superconducting silver

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The diamagnetic properties of silver backed by a thick lead layer have been studied as a function of normal-metal thickness and of temperature. Data were obtained by measuring the period of the magnetic field dependence of the critical Josephson current in $S-I-N-S'$ tunnel junctions. Strong screening has been seen at low temperatures and a thickness-independent penetration depth is indicated as the temperature goes to zero. This characteristic penetration depth is observed to be on the order of 1500 Å for proximity-effect silver. We have used a modified London equation for the case of a spatially varying pair amplitude and have numerically solved for the magnetic field profile in this system. The spatial dependence of the pair amplitude obtained from Landau-Ginzburg theory produces good agreement with thickness- and temperature-dependence data.

I. INTRODUCTION

The study of the superconducting proximity effect allows the exploration of the relationship between superconducting properties and normal-metal properties in materials and at temperatures which are otherwise unsuitable for this work. The induced superconductivity has the additional feature of being spatially varying, which introduces strong temperature and thickness effects. Although this spatial inhomogeneity is an essential aspect of the proximity effect, there have been no experimental studies which test the commonly accepted theoretical predictions.¹

Thermodynamic properties (transition temperatures and critical fields) probe the phase boundary where the superconducting state exists almost entirely in the superconducting metal. Electron tunneling in zero magnetic field measures properties directly at the tunneling interface —induced energy gaps in the case of single-particle tunneling and pair amplitude in the case of Josephson tunneling.

The magnetic screening property, on the other hand, allows one to measure a simple integral of a local property, namely, the magnetic field, and is therefore a sensitive probe of the spatial distribution of the induced superconductivity far into the normal metal.

Previous studies of the magnetic screening properties of proximity-effect superconductors have primarily centered around the magnetic breakdown field. $2-4$ However, some work was done by the Orsay group⁴⁻⁸ on the subject of the diamagnetic response of proximity superconductors. This work studied the properties of thick $(1-2 \mu m)$ normal metals via the dynamic susceptibility technique of Schawlow and Devlin.⁹ The aim of their work was largely to establish that a

proximity material did in fact exhibit a Meissner effect.

The problem may be easily visualized as in Fig. 1. The pair amplitude is strongly decaying in the normal metal as distance increases from the N-S interface. The penetration depth is therefore increasing. The magnetic field penetrates freely into the normal metal while the screening is becoming progressively more effective. Finally, at some point, the screening becomes sufficiently large to cause a rapid decay of the field.

The Orsay group pictured the magnetic field profile as shown by the dashed line in Fig. 1 and interpreted their results in terms of the screening length ρ , taken roughly from where the field drops off sharply near the $N-S$ interface. They observed that the magnetic field was rather weakly screened in the normal metal and that at a given temperature ρ was independent of normal-metal thickness.

In order to compare their results with theory, the

FIG. 1. Orsay-group schematic picture for magnetic field profile in $N-S$ sandwich.

Orsay group used a generalized Landau-Ginzburg theory in which they modeled the magnetic field profile as a step function with the drop occurring at ρ . In the thick-sample regime, the pair amplitude F in the normal metal is expected to decay exponentially as a function of distance from the N-S interface. Therefore a position-dependent penetration depth $\lambda(x)$ [$\alpha(1/F)$] will exponentially increase over this range. The value of ρ was calculated as the point at which the Landau-Ginzburg parameter K_N ($\equiv \frac{\lambda}{\xi_N}$) reaches unity¹⁰ and is given by

$$
\rho = \xi_N \left[\ln \frac{1}{K_N} - 0.116 \right] \ .
$$

This behavior is consistent with the idea that the superconductivity is induced in the material only over a distance on the order of a few coherence lengths. Thus, screening is only effective where the field varies faster than the pair amplitude $(\lambda \leq \xi)$.

The present work attempts to look at proximity superconductors over a wider thickness and temperature range. Rather than using the dynamic susceptibility technique, which is a relative measurement, we chose to make use of the magnetic field dependence of the dc Josephson current, which yields absolute values of the field-penetration integral via the field periodicity of the Josephson current. The periods, which will be discussed in the next section, are given by

$$
\Delta H = \Phi_0 / [W(\lambda_1 + \lambda_2 + d)]
$$

where Φ_0 is the fluxon, W the junction width, λ_1 and λ_2 the effective penetration depths of the two superconductors making up the Josephson junction (Fig. 2), and d the thickness of the oxide barrier. Since the field does not vary as a simple exponential in the proximity sandwich, λ_{eff} is operationally defined by

FIG. 2. Tunnel junction geometry for tin-oxide silver-lead sandwich.

where the integral is carried out from the N free surface $(x = 0)$ to deep within the superconductor.

The model used by the Orsay group is insufficient to describe the behavior of proximity metals of thickness comparable to the normal-metal penetration depth. In this case the functional dependence of the screening on position is required in some detail, and its determination is the main purpose of the present work. We have therefore developed the appropriate extensions of the London theory to treat the case of an inhomogeneous pair amplitude as well as the boundary condition at the $N-S$ interface. These equations have been solved numerically for comparison with our experiments.

II. EXPERIMENT

In a rectangular geometry, the dc Josephson current is modulated by an applied magnetic field in a Fraunhofer-diffraction-type pattern given by

$$
I_{\max}(H) = I_0 \left| \frac{\sin(\pi \Phi_J/\Phi_0)}{\pi \Phi_J/\Phi_0} \right| ,
$$

where Φ _{*j*} is given by

$$
\Phi_J = H W(\lambda_1 + \lambda_2 + d)
$$

Tunnel junctions with the geometry of Fig. 2 were prepared using conventional techniques. Tin counter electrodes were resistively evaporated at reduced (-250 K) temperatures¹¹ in a vacuum on the order of 1.0×10^{-6} Torr. The width of these films was kept to approximately 100 μ m so that we could avoid self-field corrections to the measurements. The films were masked with silicon dioxide to prevent edge effects and were then oxidized in a dc glow discharge in 50 m Torr of dry oxygen. Exposure times on the order of a minute in a 2-kV glow produce junction resistances from 0.001 to 0.1 Ω . Silver and lead films were then successively evaporated from electron beam and resistive sources, respectively. Film thicknesses were measured with a quartz microbalance calibrated with an optical interferometer.

Measurements were made in a standard helium cryostat with temperatures measured by a calibrated germanium resistor. The probe was magnetically shielded with several layers of μ metal, Currentvoltage characteristics were monitored on an oscilloscope (Fig. 3), and the Josephson current was measured electronically via a technique described elseured electronically via a technique described else-
where.¹² As is usually the case in Josephson experi ments, the amplitude of the Josephson current generally scales linearly with the superconducting energy gap and inversely with junction resistance, but significant enough deviations occur so as to make measurements that depend on the absolute value of the critical current unfeasible. Temperature dependence of

FIG. 3. Oscilloscope trace of 500-A silver proximity junction current-voltage characteristic at 1.2 K. Josephson current amplitude 4 mA; energy gap is 0.95 mV,

the Josephson current was in qualitative accord with earlier work on proximity junctions. 13,14

Effective penetration depths were deduced from the Fraunhofer patterns by averaging over 15 or more periods. Some samples displayed over 50 maxima on each side of zero field. Junctions exhibiting anomalous field dependencies were rejected for this study. Films up to 1 μ m thick were measured, and good quality patterns could be obtained for all thicknesses. A trace of a $1-\mu m$ silver film is shown in Fig. 4. Thicker junctions have not been attempted at this time. The requirement of increasingly small resistances (and thus thinner oxide barriers) and the small critical currents associated with a tiny induced gap in a thicker film make such samples extremely difficult to produce. 1 μ m was chosen as a practical limit, although perhaps another factor of 2 might be achievable.

FIG. 4. Maximum josephson current vs applied magnetic field for $1-\mu m$ silver proximity junction.

III. RESULTS

Penetration-depth measurements were performed on samples of varying silver thickness and at various temperatures (iimited by the disappearance of the Fraunhofer patterns at high temperatures). Strong temperature dependence is seen for thick samples. The effective penetration depth in the silver is seen to double over a 1-K temperature change in the $1-\mu m$ sample,

Figure 5 shows data taken at 1.2 K for a number of silver thicknesses. The observed penetration plotted here results from solving the previous equation for an unknown λ_2 with λ_1 known (λ_{SN}) . Error bars primarily reflect a 5% uncertainty in film width.

Qualitatively we may understand this figure in the following way. At low thicknesses the normal metal is too thin to screen the applied field and most of the screening takes place in the lead film which is behind it ($\lambda_{\rho b}$ =400 Å). At large thicknesses the magnetic field penetrates almost to the $N-S$ interface but is excluded from a region near the interface by screening in N . This behavior is very similar to what was seen in the Orsay-group work. It should be noted that in this thick limit, the relationship between the Orsay parameter ρ and our "observed penetration" λ_{eff} should be

$$
\rho = d_N - \lambda_{\rm eff} \quad ,
$$

where d_N is the normal-metal thickness. In a plot like Fig. 5, this would predict, as the asymptotic behavior, a line of unit slope but displaced from the origin by ρ along the x axis.

The. most interesting region in Fig. 5 is the "plateau" between 2000- and 4000- \AA thickness with a λ_{eff} of 1500 A. This is suggestive of a characteristic penetration depth for the normal metal. When the

FIG. 5. Observed penetration in silver-lead sandwich vs silver thickness at 1.2 K (\pm 5% error bars.)

FIG. 6. Observed penetration in silver-lead sandwich vs temperature for 2500, 5000, 6000, 8000 Å, and 1 μ m of silver $(\pm 5\%$ error bars).

thickness is smaller than λ_{eff} the N metal is ineffective in screening, as is seen. The penetration for thicker films should be limited to a distance λ_{eff} for all thicknesses. As we shall see later, this is what results from theory for sufficiently low temperatures. At 1.2 K, the pair amplitude is too small for thick films to screen effectively.

Figure 6 shows the temperature dependence of the effective penetration depth for a number of silver thicknesses. Weak temperature dependence is evident for the thin samples, while the strong changes noted above are observable in the thicker samples. Despite the disparate values of the effective penetration depth for the various thicknesses, all the curves suggest a zero-temperature value of approximately the same magnitude. Least-squares fittings to the 0 curves yield intercepts of ¹²⁰⁰—¹⁶⁰⁰ ^A for all the curves, again suggesting a characteristic penetration depth for the normal metal. We are at this time pursuing further work that will extend these measurements to very low temperatures and determine the zero-temperature limit more exactly.

IV. THEORY

Previous treatments of the magnetic response of proximity superconductors were limited in two important ways: the restriction to thick $(d_N \gg \xi_N)$ samples and the indirect relationship to measured quantities.

The first problem can be eliminated within the context of Landau-Ginzburg theory; the finite d_N case text of Landau-Ginzburg theory; the finite d_N case
has been treated by the Orsay group.¹⁵ Whereas the infinite geometry requires an exponential decay for the spatial dependence of the order parameter, the existence of a free surface imposes a hyperbolic

cosine dependence.

For the geometry shown in Fig. 2, the pair amplitude in the normal metal is written as

$$
F_N = F_0 \frac{\cosh[(d_N - x)/\xi_N]}{\cosh(d_N/\xi_N)}
$$

where F_0 is determined by matching solutions at the $N-S$ interface using a form in the superconductor appropriate to a semi-infinite S layer
 $X_0 - x$

$$
F_S = F_{BCS} \tanh \frac{X_0 - x}{\sqrt{2} \xi_S(T)}
$$

(where X_0 is to be determined from the boundary conditions and F_{BCS} refers to pair amplitude for BCS superconductor).

All these solutions are appropriate to the linearized Landau-Ginzburg equation. Inclusion of the nonlinear term yields elliptic functions rather than hyperbolic functions and leads to considerably different low-temperature behavior; this correction will be neglected for the present purpose.

The second difficulty with the Orsay formulation is that the screening length ρ does not explicitly take into account the actual profile of the magnetic field in the normal metal and therefore does not correspond directly to the measurable quantity: the effective penetration length.

To deal with this problem, we have made use of the idea of a position-dependent penetration depth to derive a modified London equation for the magnetic field. In order to do this, we have assumed a local description of the electrodynamics. This is inherently incorrect as the characteristic penetration depths are small compared with the coherence lengths at typical temperatures. However, we might expect to be able to cast the nonlocal electrodynamics into a local form by postulating an effective penetration depth whose magnitude includes nonlocal corrections. It is in this spirit that we can write a modified London equation for the case of a spatially varying penetration depth¹⁶:

$$
\frac{d^2H(x)}{dx^2} = \frac{1}{\lambda^2(x)}H(x) - \frac{2}{\lambda(x)}\frac{d\lambda(x)}{dx}\frac{dH(x)}{dx}
$$

The first-derivative term results explicitly from the spatial dependence of λ . Looking back at the schematic picture of the field profile in a proximity material proposed by the Orsay group (Fig. 1), it is clear that such a term must appear in the field equation in order to allow for negative curvature to be present.

To solve for the field profile, an explicit form for $\lambda(x)$ must be assumed. Analogous to simple London theory in which $\lambda \sim 1/n_s^{1/2} \sim 1/F$, λ is written as inversely proportional to F , using the form appropriate to Landau-Ginzburg theory in a finite geometry. Hence,

$$
\lambda(x) = \lambda_0 \frac{\cosh(d_N/\xi_N)}{\cosh[(d_N - x)/\xi_N]}.
$$

Here λ_0 must be determined in part from the boundary conditions¹⁷ mentioned previously. However, the characteristic length contained with λ_0 is seen to be independent of the boundary (and of d_N) as the temperature goes to zero. This is consistent with microscopic calculations of superconducting electrodynamics in which the screening is seen to be given by a combination of a diamagnetic response dependent only upon normal-metal properties and a paramagnetic response related to the excitation of the system. 18 We therefore would expect the zerotemperature response of the system to be independent of the boundary and of d_N , the latter suggested by the temperature data shown previously.

Solving the boundary value problem yields a value for λ_0 of

$$
\lambda_0 = \lambda_N \frac{N_S}{N_N} \coth\left\{\frac{1}{2} \sinh^{-1} \left[\frac{\sqrt{2} \xi_N}{\xi_S} \frac{D_S}{D_N} \coth\left(\frac{d_N}{\xi_N}\right) \right] \right\}
$$

For the system studied, the hyperbolic cotangent term is always about unity so that the main contribution to λ_0 comes from the density-of-states factors. Whether these terms should contribute is questionable at this time in light of the previous discussion of the zero-temperature properties. At any rate, the terms only act to renormalize the prefactor in λ_0 .

The resultant differential equation for the field with the explicit spatial dependence included looks like

$$
\frac{d^2H(x)}{dx^2} = \frac{1}{\lambda_0^2} \frac{\cosh^2[(d_N - x)/\xi_N]}{\cosh^2(d_N/\xi_N)} H(x) + \frac{2}{\xi_N} \tanh\left(\frac{d_N - x}{\xi_N}\right) \frac{dH(x)}{dx},
$$

which is not amenable to analytic solution. Therefore, numerical techniques were employed. The N-S interface introduces an additional boundary condition in the problem, namely, that λ^2 is continuous across the interface,¹⁹ and the problem as a whole is then solved on the computer.

The solutions require two parameters, λ_0 and the coherence length. How these are chosen will be discussed in the next section. Solutions generated by this calculation are shown in Fig. 7.

Figure 7(a) shows a field profile for a thin (1000 A) silver film at intermediate temperature. It looks like two ordinary superconductors with very different penetration depths. Figures $7(b) - 7(d)$, on the other hand, are for a thick sample and show a progressive change in the structure of the field profile with changing temperature. The lowest-temperature plot reflects behavior very similar to an ordinary superconductor while the highest-temperature curve represents the Orsay regime. It is instructive to compare this exact result with Fig. 1.

FIG. 7. Numerical solutions of magnetic field profiles in Ag-Pb sandwiches. (a) 1000 \tilde{A} Ag at 1.2 K, (b) 1.7 μ m Ag at 3.5 K, (c) 1.7 μ m Ag at 1.2 K, (d) 1.7 μ m Ag at 0.2 K.

V. COMPARISON WITH EXPERIMENT

The integral of the field profiles such as shown in Fig. 7 gives the effective penetration depth that would be measured in a Josephson experiment. In each case, there will be a contribution from the silver and from the lead.

To produce the field curves, the approximate magnitude of λ_0 was deduced from the temperaturedependence data. This value was then varied over a range of about 10% to yield the best fit to the data.

The temperature dependence is explicitly built into the theory via the coherence length. A dirty-limit $(1 \ll \xi)$ expression gives the best results although mean-free-path measurements by the technique of Toxen *et al.*²⁰ suggest an intermediate regime between the clean and dirty limits. A typical mean free path so obtained is on the order of 5000 Å as compared with a ξ_N of 6000 Å (see below).

The dirty-limit expression for the coherence length

$$
\xi(T) = \left(\frac{h v_F l}{6 \pi k_B T}\right)^{1/2}
$$

yields a value of around 6000 Å for the silver films at 1.2 K. Varying this value to best fit the data yields a value of 6150 Å^{21}

Figure 8 superimposes the thickness-dependence data shown before upon a theoretical curve generated using the two best-fit parameters discussed above. Agreement is quite good.

Figure 9 shows the temperature-dependence data with theoretical plots obtained by using a dirty-limit temperature dependence ($\propto T^{-1/2}$) normalized to 6150 A at 1.2 K. Agreement is best for lower temperatures. As the temperature is raised, the coherence length decreases and the samples act cleaner. A

FIG. 8. Observed penetration data and theoretical curve vs silver thickness at 1.2 K.

FIG. 9. Observed penetration data and theoretical dirtylimit curves vs temperature for 2500, 5000, 6000, 8000 Å and 1 μ m Ag. Theoretical clean-limit curve (dashed line) for $1 \mu m$ Ag.

clean-limit curve is shown for the $1-\mu m$ sample. It demonstrates clearly that the samples are indeed in an intermediate regime.

It would be fruitful to measure samples intentionally made dirtier in order to examine the validity of the theoretical description in a well-defined limit.

VI. CONCLUSIONS

The work presented here demonstrates that strong magnetic screening is possible in so-called type-II proximity materials (i.e., those without a finite T_c) in the limit of low temperatures. A penetration depth characteristic of the material can be deduced from these experiments; for silver, a value of 1500 A was found. The simple London theory for magnetic penetration depth $[\lambda_L = (mc^2/4\pi ne^2)^{1/2}]$ would predict a value on the order of 400 A for silver. Whether this value must be modified only by a nonlocal correction or by additional factors remains to be seen. Clearly the local description used here is inherently incorrect but until such time as a tractable nonlocal formulation is available, it must suffice.

The property of constant screening length (and thus, continuously increasing penetration with normal-metal thickness) observed by the Orsay group will not be seen unless high enough temperatures are reached for a given thickness sample. Further work will be carried out in the near future at low temperatures to further investigate the strong-screening behavior of thick samples.

In conclusion, the low-field behavior of proximity superconductors is found to be fundamentally similar to intrinsic superconductor screening apart from strong temperature effects.

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