Quasiparticle Magnetospectroscopy in Superconductors: A Detailed Look at the Shape of the Penetration Layer

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Microwave magnetoabsorption peaks in single-crystal superconducting Al have been identified by comparison with a detailed calculation of quasiparticle energy levels. The peaks correspond to resonant transitions between states bound in the magnetic-field penetration layer as predicted by Pincus. By fitting the observed spectra to the calculation, we are able to extract the penetration length λ and a second parameter describing the shape of the nonlocal-field penetration.

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Since the discovery of the Meissner-Ochsenfeld effect[1] the penetration layer of magnetic-field decay in superconductors has been studied extensively.[2] Although theoretical predictions for the depth of penetration and the decay law for the magnetic field have existed for many years, experimental determinations of penetration depths disagree and detailed studies of the shape of the penetration layer are rare. The central reason for the paucity of detailed experimental information is that most penetration values are deduced from observed deviations from perfect diamagnetism, i.e., from a global sample property.

This Letter reports the successful determination of penetration-layer characteristics in Al from an effect whose sensitivity to magnetic-field decay in the surface layer has long been apparent, but for which a realistic theoretical calculation has been lacking. The experiment concerns resonant transitions from surface-bound thermal quasiparticle states first predicted by Pincus.[3] Subsequent experimental work in Sn,[4,5] In,[6] Pb,[7] and Nb[8] verified qualitative expectations of the Pincus model, but quantitative analysis remained elusive partly as a result of insufficient knowledge of normal-state electronic properties in these metals. Not only is Al a simple, well-understood metal, but first experimental results[9] strongly suggested that the resonant quasiparticles have free-electron-like dispersion, a significant simplification for the analysis. Furthermore, the observation of multiple transitions in Al sets a rigid standard for theoretical fitting.

In the experiment one studies the fixed-frequency microwave absorption of a highly polished plane of single-crystal Al as a function of an external magnetic field $\vec{H}$ applied parallel to the surface. (For additional experimental details see Ref. 9.) Quasiparticles traveling parallel to the Meissner current are bound to the surface by the effective magnetic potential. Resonances in the microwave absorption are associated with transitions between quasiparticle levels bound in the well formed by the surface barrier, taken to be infinite, and the effective potential $V_{\rm eff}$. Analogous surface wells and resonances have been exploited both in normal metals[10] and in semiconductors.[11]

In the London limit one can write

$$V_{\rm eff}^{\rm L} = -\frac{ep_{\rm F}H\lambda}{m^*c}e^{-z/\lambda} \qquad (1)$$

where the coordinate z is directed into the superconductor, $p_{\rm F}$ is the Fermi momentum, and m^* is the appropriate effective-mass component. The thickness of the penetration layer is governed by λ. In Al, where the coherence length is much larger than λ, the potential is expected to assume its nonlocal form. To allow for nonlocality, we take

$$V_{\rm eff}^{\rm NL} = V_{\rm eff}^{\rm L}e^{\eta z/\lambda}(2-e^{\eta z/\lambda}). \qquad (2)$$

In this expression the parameter η ($0\leq\eta\leq 0.5$) represents the effect of nonlocality, introducing a repulsive hump in $V_{\rm eff}$ as shown in Fig. 1 (inset). For $\eta=0$, $V_{\rm eff}$ reduces to the London limit [Eq. (1)], and for $\eta=0.32$, the height of the hump corresponds to that which we calculate from BCS theory.[12] In Eq. (2), λ is always the physical penetration depth, $\int_0^\infty H(z)dz/H(0)$, independent of η. Since $V_{\rm eff}$ is proportional to the vector potential, magnetic-field sign reversal occurs at the maximum of $V_{\rm eff}$.

The identification of the resonant quasiparticles with free-electron-like portions of the Al Fermi sur-

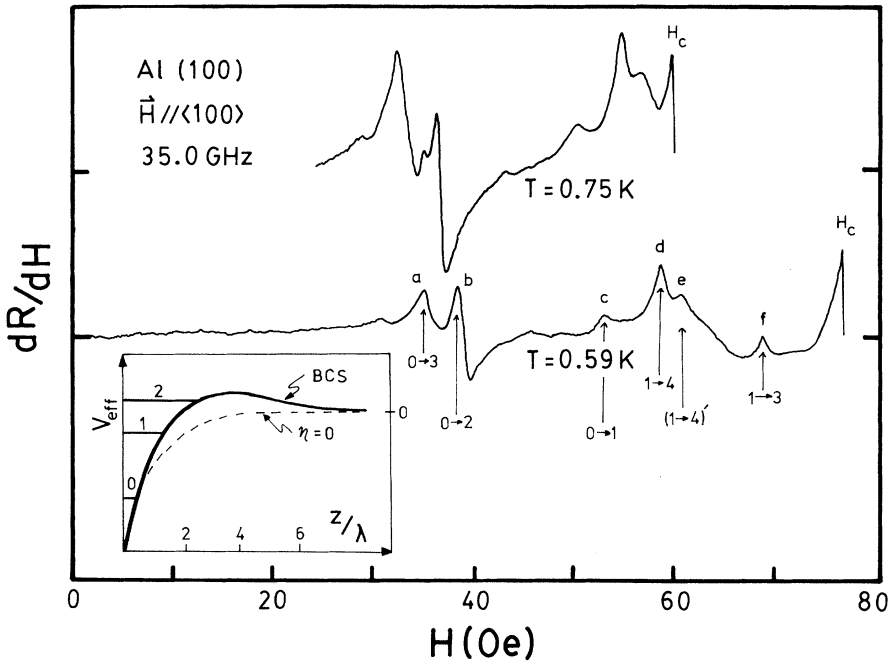

FIG. 1. Magnetoabsorption spectra in superconducting Al. The field derivative of the real part of the surface impedance (arbitrary units) is plotted vs magnetic field. Resonance identifications from the calculation are indicated by arrows. Inset: labeling scheme and effective potential. The gap edge is at $V_{\text{eff}}=0$.

face was based on the variation of resonance fields with field direction in the (110) plane.[9] There a one-to-one correspondence between free-electron normal-state signals[13] and superconductor signals was found. We have since observed the same correspondence in the (100) plane. The identification of the resonant quasiparticles is therefore well established. Improvements in experimental technique have allowed us to study the magnetoabsorption at temperatures as low as 0.5 K yielding spectra such as shown in Fig. 1 for the (100) plane.

These spectra are of particular importance. In addition to the doublet peak (labeled a and b) also seen in the (110) plane,[9] several new transitions are observed. All peaks in Fig. 1 follow the same free-electron-like anisotropy for field direction about a ⟨100⟩ axis. We therefore conclude that all transitions in the trace correspond to a single quantum well, i.e., to a single group of quasiparticles on the Fermi surface. From normal-state work[13] we know that these quasiparticle states are located at the intersection of a ⟨100⟩ axis with the Fermi surface having $p_F/\hbar = 1.69\times 10^8$ cm^{-1} and $m^*/\hbar = 1.32$ sec cm^{-2}.

Energy levels and wave functions for quasiparticle motion normal to the sample surface are described[3] by the one-dimensional Bogoliubov equations,

$$\begin{pmatrix} \mathcal{T}+V_{\text{eff}}-\epsilon & \Delta \\ \Delta & -\mathcal{T}+V_{\text{eff}}-\epsilon \end{pmatrix}\begin{pmatrix} u(z) \\ v(z) \end{pmatrix}=0, \quad (3)$$

where $u(z)$ and $v(z)$ are the electronlike and holelike amplitudes, respectively. The kinetic-energy operator $\mathcal{T}$ is given by $-(\hbar^2/2m^*)d^2/dz^2-\alpha$, where α measures the transverse kinetic energy with respect to the Fermi level: $E_F-(\hbar^2/2m_{\parallel}^*)\times(k_x^2+k_u^2)$. Energy eigenvalues ϵ are also defined relative to E_F. In the above equations, terms second order in V_{eff} have been neglected; we also assume the energy gap to be independent of z and H, and at $T=0$ given by its measured[14] value $\Delta/h = 42$ GHz. These restrictions are well justified because of aluminum's long coherence length.

We have solved the Bogoliubov equations to yield values of the resonance fields H_{mn}. These are the field values at which the energy levels ϵ_m and ϵ_n, determined by the magnetic-field–induced V_{eff}, are separated by the experimentally fixed photon energy $\hbar\omega$. The levels ϵ_n ($n=0, 1, 2, \ldots$) and the eigenvectors $u(z)$ and $v(z)$ are found numerically. The resulting resonance fields H_{mn} are functions of both the wave-vector component, k_H, along the ap-

plied field direction, and of the variable α. We assume that the experimentally observed $m \rightarrow n$ resonance position is determined by the value of H_{mn} which is stationary with respect to k_H and α. Such a condition is familiar from normal-state resonances and from Azbel'-Kaner cyclotron resonance. At the Fermi-surface location identified above for the geometry corresponding to the spectrum of Fig. 1, H_{mn} is already stationary with respect to k_H. To determine the expected resonance positions, therefore, we use the values of p_F and m^* quoted above for this location to determine $H_{mn}(\alpha)$. The stationary values with respect to α are then the predicted resonance fields.

With the local potential of Eq. (1), there is one undetermined parameter: the penetration depth λ. By adjusting the value of this parameter, one can shift the predicted spectrum of stationary H_{mn} values in order to position any given H_{mn} at an observed resonance field. All reasonable fits to the observed spectrum of Fig. 1 require identification of the b peak as the $0 \rightarrow 2$ transition to reproduce the featureless field range below peak a and that between b and c. In Fig. 2, the local case ($\eta = 0$ in inset) is compared to experimental resonance fields resulting from extrapolation to $T = 0$. (Experimental field values have been corrected for the demagnetizing factor, 0.04, of the sample.) The fit is unsatisfactory for two reasons. First, the calculation predicts more resonances than observed. Furthermore, the value of λ required for the fit is 572 Å, far outside the range expected[15] for Al.

As the value of the nonlocality parameter η is increased, high-lying levels are no longer bound, causing the disappearance of transitions to such levels (Fig. 2, inset). Only a single new spectral line, labeled $(1 \rightarrow 4)'$, appears. It arises from the development of a second stationary value of $H_{14}(\alpha)$. In the η range 0.25 to 0.35 the number of predicted lines corresponds to that observed in the experiment, and the resonance field values are in reasonable agreement as well. As η is increased much beyond the value 0.32 corresponding to BCS theory, additional lines disappear. The value of penetration depth from the fit is $\lambda = 500 \pm 30$ Å, in good agreement with expectation.[15] The uncertainty in λ reflects the possible variation of η. Adopting a fixed value for η, e.g., that corresponding to BCS theory (giving $\lambda = 484$ Å) would reduce the uncertainty to a few percent. To ensure that the parametrized potential of Eq. (2) adequately mimics BCS theory, we have performed a fit using a numerically accurate BCS form.[12] The fit yields $\lambda = 490 \pm 15$ Å for $T = 0$. The uncertainty accounts

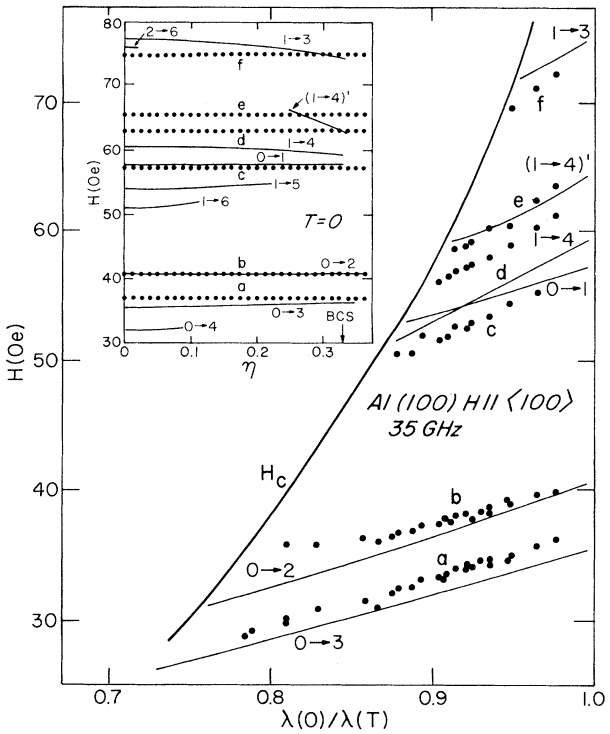

FIG. 2. Temperature dependence of resonance fields at 35 GHz. Experimental peak maxima are given by closed circles; the corresponding sample temperatures determine the $\lambda(0)/\lambda(T)$ values in the BCS extreme anomalous limit (Ref. 17). Theoretical curves are for penetration-layer parameters $\eta = 0.29$ and $\lambda(0) = 518$ Å. Inset compares experimental peak maxima extrapolated to $T = 0$ (dotted lines) with theoretical prediction as a function of η. This shape parameter varies from the purely local case ($\eta = 0$) through the nonlocal BCS value. $\lambda(0)$ is chosen to fit the $0 \rightarrow 2$ transition at each η.

for possible errors in p_F, m^*, and Δ as well as in marking the exact resonance position. A successful line-shape calculation would reduce the 15-Å uncertainty by about a factor of 2. Within the framework of BCS theory, therefore, quasiparticle magnetospectroscopy is a precision method for determining penetration depths absolutely.[16]

Further support for the validity of the calculation comes from a comparison of the observed temperature dependence of the peak positions with the calculation. As Fig. 2 shows, except for the nonobserved crossing of the $0 \rightarrow 1$ and $1 \rightarrow 4$ transitions, the agreement is quite good. (The particular values of η and λ used for the figure are characteristic of the entire range quoted above.) In calculating the transition fields it is, of course, necessary to vary the gap parameter as well as $\lambda(T)$ in accord with BCS theory.[17] Neglecting the temperature depen-

dence of Δ fails to reproduce the nearly linear variation of peak position with $1/\lambda(T)$ seen experimentally. In earlier work[4] it was assumed that the singularity in the BCS density of states at the gap edge caused transitions from bound levels to the continuum to dominate the spectrum. In a nonlocal magnetic potential the gap edge ($\epsilon = \Delta$) has no special significance and all our identified transitions occur between discrete quasiparticle levels. In fact, predicted resonance fields resulting from transitions between discrete levels and a fictitious gap-edge level deviate markedly from the observed temperature dependence of Fig. 2.

In summary, we are able to describe the observed magnetoabsorption spectrum with the Pincus theory using a realistic potential. The shape of the penetration layer which we find is consistent with the nonlocal BCS result and in the (100) plane of Al for the $\langle 100 \rangle$ field direction, we find a penetration depth $\lambda = 490 \pm 15$ Å associated with the BCS form. A preliminary line-shape calculation is also in good agreement with experiment, but further refinements are necessary. It would be of interest to make a detailed comparison of wave functions to the semiclassical orbits discussed by Azbel.[18] We point out that our experiment is ideal for investigating anisotropy and, indeed, we have evidence for anisotropy in λ. The anisotropy, studies of the temperature dependence of peak amplitudes, and the dependence of the spectra on microwave power will be treated in future reports.

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[1]W. Meissner and R. Ochsenfeld, Naturwissenschaften **21**, 787 (1933).

[2]For a recent comprehensive review, see A. K. Raychaudhuri, C. Egloff, and L. Rinderer, J. Low Temp. Phys. **53**, 513 (1983).

[3]P. Pincus, Phys. Rev. **158**, 346 (1967).

[4]J. F. Koch and P. Pincus, Phys. Rev. Lett. **19**, 1044 (1967).

[5]J. F. Koch and C. C. Kuo, Phys. Rev. **164**, 818 (1967).

[6]J. R. Maldonado and J. F. Koch, Phys. Rev. B **1**, 1031 (1970).

[7]Y. Onuki, H. Suematsu, and S. Tanuma, J. Phys. Soc. Jpn. **41**, 1313 (1976).

[8]L. Rothkirch, R. Herrmann, H. Dwelk, and U. Preppernau, Phys. Status Solidi (b) **90**, 517 (1978).

[9]S. Whitmore, A. Starr, and R. E. Doezema, Solid State Commun. **41**, 623 (1982).

[10]R. E. Doezema and J. F. Koch, Phys. Condens. Matter **19**, 17 (1975).

[11]T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).

[12]J. Halbritter, Z. Phys. **243**, 201 (1971). For a general discussion, see M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).

[13]T. Wegehaupt and R. E. Doezema, Phys. Rev. B **16**, 2515 (1977).

[14]M. A. Biondi, M. P. Garfunkel, and W. A. Thompson, Phys. Rev. **136**, A1471 (1964). We have learned since completing this work that the tunneling measurements of G. L. Wells, J. E. Jackson, and E. N. Mitchell, Phys. Rev. B **1**, 3636 (1970), would lead to the better choice $\Delta = 45$ GHz for the [100] direction. Fortunately, the resulting change in the inferred penetration depth is small compared with our indicated uncertainty.

[15]T. J. Greytak and J. H. Wernick, J. Phys. Chem. Solids **25**, 535 (1964).

[16]For another very recent new technique, see G. P. Felcher, R. T. Kampwirth, K. E. Gray, and R. Felici, Phys. Rev. Lett. **52**, 1539 (1984).

[17]B. Mühlschlegel, Z. Phys. **155**, 313 (1959).

[18]M. Ya. Azbel', Zh. Eksp. Teor. Fiz. **59**, 295 (1970) [Sov. Phys. JETP **32**, 159 (1971)].