Evidence for nonlocality and the crossover from nonlocal to local behavior near the critical temperature in superconducting metals

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We have performed high precision measurements of the temperature dependence of the magnetic penetration depth $\lambda(T)$ in pure aluminum and cadmium samples. These superconductors and most of the other superconducting metals are predicted to be in the nonlocal limit of electrodynamics. However, most previous experimental works on $\lambda(T)$ in Al reported large deviations from the expected nonlocal behavior. Here we present experimental data which agree remarkably well with theory, giving very strong evidence that nonlocal superconductivity indeed occurs in both Al and Cd. More strikingly, the cadmium data are consistent with the existence of the crossover from nonlocal to local behavior predicted to occur in all superconductors near T_c.

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For the last three decades it has been assumed, without doubts, that pure aluminum is an extreme Pippard (nonlocal) superconductor, because its coherence length ξ_0 is much larger than its magnetic penetration depth $\lambda(0)$ [$\kappa=\lambda(0)/\xi_0\approx 0.03$]. Aluminum in fact has become the classic example of a nonlocal superconductor, and it is very often used as a reference for calibration purposes and theoretical comparisons. However, nonlocality in superconducting Al surprisingly has not been confirmed experimentally. The most recent experimental works on the temperature dependence of the penetration depth in Al reported that the data did not agree with the behavior predicted by the nonlocal BCS theory.[1,2] Tedrow *et al.*[1] went further and analyzed the data from all previous experiments. They found that such data agree with their own and that all the data were close to the local BCS prediction. More recently, Signore *et al.*,[3] in a penetration depth study on UPt_3 where Al was used as a reference, reported that the temperature dependence of λ in pure Al fitted very well the nonlocal BCS expression in a λ vs T graph. However this result should be taken with care, because it can be shown that in this type of plot both the local and nonlocal BCS approximation can fit equally well the data. Remarkably no attention has been paid to this contradiction between experiment and theory for many years, in spite of the fact that such contradiction questions the basis of the electrodynamics of a superconductor.

Even in clean superconductors for which $\lambda(0)/\xi_0 \ll 1$, nonlocality is not expected to occur throughout the temperature range below T_c. It has been predicted in classic textbooks that *all* superconductors become local very close to T_c where $\lambda(T) > \xi_0$.[4,5] To our knowledge, such a crossover has never been observed. Here we present high precision measurements of the temperature dependence of the magnetic penetration depth $\lambda(T)$ in Al and Cd down to 30 mK. Cadmium is a superconductor in the intermediate nonlocal range ($\kappa \sim 0.5$). We find that for both elements the temperature dependence of the magnetic penetration depth is in excellent agreement with the prediction of nonlocal BCS electrodynamics, and that the high resolution Cd data display the crossover from nonlocal to local behavior expected very near T_c.

The aluminum sample used in the experiment was a 99.9995% sphere of 0.8 mm in diameter, chemically polished with a solution of $3HCl{:}1HNO_3$ to remove the oxide film from the surface. The cadmium sample was a 99.999+% platelike shape of dimensions $0.8\times 0.8\times 0.1\ mm^3$, also chemically polished. $T_c=1.175$ K and $T_c=0.512$ K for aluminum and cadmium, respectively, were determined from the onset of superconductivity in the penetration depth measurements.

$\lambda(T)$ measurements were performed utilizing a 28 MHz tunnel diode oscillator with a noise level smaller than 1 part in 10^9 and a low drift.[6] The magnitude of the ac field was estimated to be less than 5 mOe. The cryostat was surrounded by a bilayer mumetal shield which reduced the dc field to less than 1 mOe. Both of these fields are well below the critical fields 106 and 29 Oe for aluminum and cadmium, respectively.[5] The samples were mounted, using a small amount of vacuum grease, on a rod made of nine thin 99.999% Ag wires embedded in Stycast 1266 epoxy. The other end of the rod was thermally connected to the mixing chamber of a dilution refrigerator. The temperature was measured with a calibrated RuO_2 thermometer located at the end of the rod linked to the mixing chamber, and the temperature error below and above 300 mK is 1–2 and 2–3 mK, respectively.

The deviation of the penetration depth from its value at $T=0.03$ K, $\Delta\lambda(T)=\lambda(T)-\lambda(0.03\text{ K})$, was obtained from the change in the measured resonance frequency $\Delta f(T)$ through the expression $\Delta\lambda(T)=G\Delta f(T)$. Here G is a factor

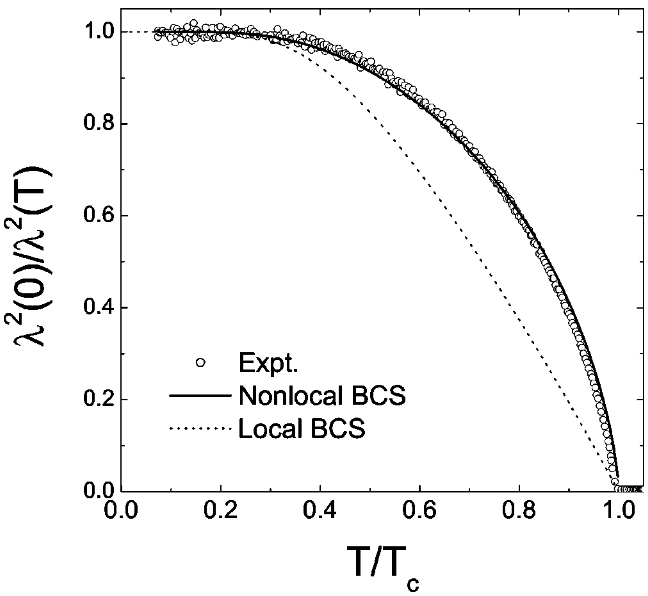

FIG. 1. $[\lambda(0)/\lambda(T)]^2$ against T/T_c up to T_c for the aluminum data and the numerical evaluation of the BCS nonlocal and local expressions of the penetration depth.

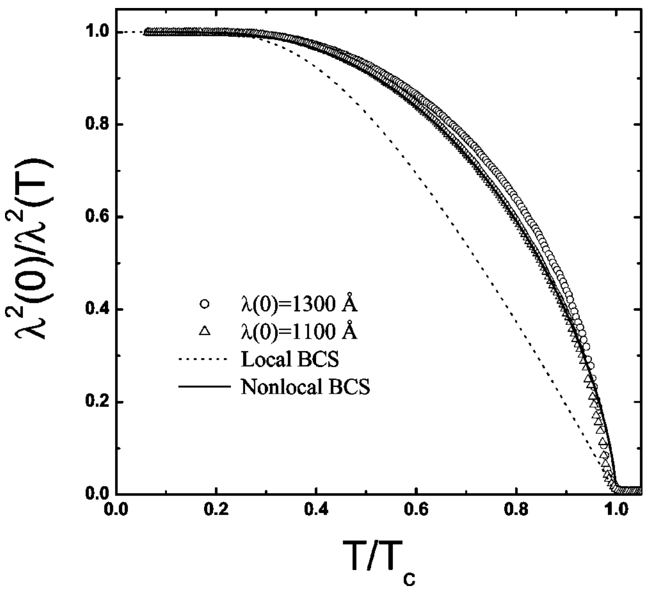

FIG. 2. $[\lambda(0)/\lambda(T)]^2$ against T/T_c up to T_c for the cadmium data and the numerical evaluation of the BCS nonlocal and local expressions of the penetration depth.

which depends on the sample and coil geometries and includes the demagnetizing factor of the sample. We determined G by two procedures: (a) by measuring the Cd sample at the lowest temperature where an exponential behavior is expected independently of the electrodynamic limit, and in this case G for Al was found from the value of the different shape relative to the Cd sample and (b) by an independent calibration of the system using a technique employed in several others penetration depth measurements.[7] The values of G obtained from these methods agree within 10%.

We compared the data to the theoretical penetration depth expressions of the local and nonlocal BCS electrodynamics. For a local s-wave superconductor we have

$$\frac{\lambda^2(0)}{\lambda^2(T)}=\left[1+2\int_0^\infty d\epsilon\frac{\partial f}{\partial E_k}\right]. \quad (1)$$

Here the total energy $E_k=\sqrt{\epsilon^2+\Delta_0^2}$, where ϵ is the single-particle energy measured from the Fermi surface. Δ_0 is the energy gap a $T=0$. In the case of a nonlocal s-wave superconductor the expression above changes to

$$\frac{\lambda^2(0)}{\lambda^2(T)}=\left[\frac{\Delta(T)}{\Delta_0}\tanh\frac{\Delta(T)}{2k_BT}\right]^{2/3}. \quad (2)$$

We evaluated numerically Eqs. (1) and (2) assuming for the T-dependent gap function the weak-coupling interpolation formula $\Delta(T)=\Delta_0\ \tanh[(\pi k_BT_c/\Delta_0)\sqrt{a(T_c/T-1)}]$ with $a\approx0.953$ (Ref. 8) and $\Delta_0=1.76k_BT_c$.

We plotted in Fig. 1 $[\lambda(0)/\lambda(T)]^2$ vs T/T_c for the Al data along with the numerical evaluation of Eqs. (2) and (1). In computing $[\lambda(0)/\lambda(T)]^2$ for the Al data we used a mid-range value $\lambda(0)=515$ Å, which is in between the BCS theoretical value of 530 Å[5] and the experimental estimations of 490–515 Å.[9,10] The small differences of these values do not make a significant change in the temperature dependence of $[\lambda(0)/\lambda(T)]^2$, as we shall see below for the case of Cd. The difference between $\lambda(0)$ and $\lambda(0.03)$ was taken as zero for this s-wave superconductor. We chose to plot $[\lambda(0)/\lambda(T)]^2$ because both it enhances the details of the curves at all temperatures and it is the most direct way to compare penetration depth data to theory. This type of plot has been used to present most of the penetration depth calculations in standard, classic textbooks on superconductivity.

The overall excellent agreement between our data and Eq. (2) is a strong evidence for nonlocality in superconducting aluminum. This contrasts to the results of Tedrow *et al.*[1] and Behroozi *et al.*,[2] which showed significant deviation from both the nonlocal BCS superconductivity and the two-fluid model. Since slightly different values of $\lambda(0)$ do not drastically change the temperature behavior of $[\lambda(0)/\lambda(T)]^2$, the origin of the disagreement between the present and previous results may be related to the treatment of the samples. We believe, however, that the treatment of the samples should not be the cause of the disagreement. In each individual experiment performed previously (see Tedrow *et al.*[1] and references therein) the samples were treated in different ways, but in all these experiments the results and conclusions were qualitatively the same. We should mention that we did not anneal the Al sample, as done in some of the other works, but the fact that the transition was quite sharp and near the well-established critical temperature somewhat indicates a long mean free path and that our sample is in the pure limit. It should be pointed out that experiments on Al have been performed at microwave and radio frequencies, and the results have shown to be independent of the frequency range.[1]

In Fig. 2 we show $[\lambda(0)/\lambda(T)]^2$ against T/T_c for the Cd data along with the numerical evaluation of Eqs. (2) and (1). There is no consensus in the value of $\lambda(0)$ for Cd, with experimental values ranging from 900 to 1340 Å.[11,12]

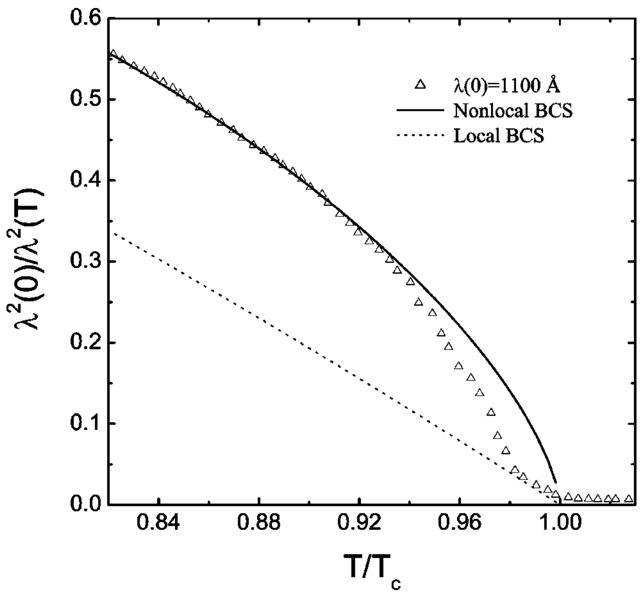

FIG. 3. A blowup of Fig. 2 near T_c. The data of the magnetic penetration depth in superconducting cadmium display the crossover from nonlocal to local behavior expected for all superconductors around T_c. The inset depicts the Al data near the critical temperature.

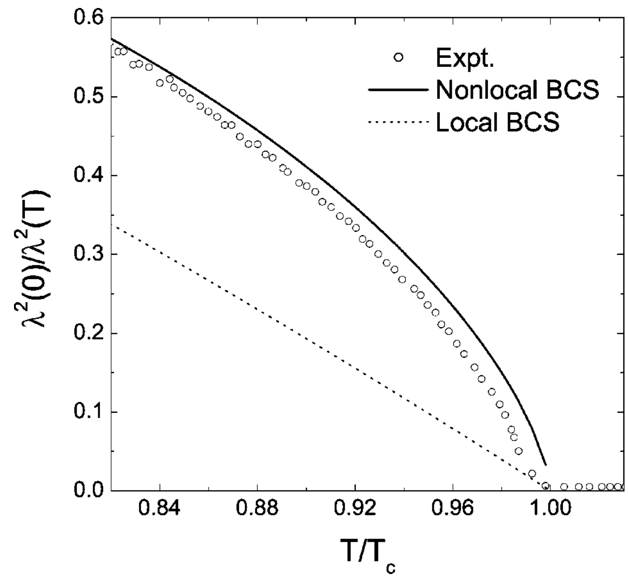

FIG. 4. A blowup of Fig. 1 near T_c. The data of the magnetic penetration depth in superconducting cadmium near the critical temperature.

Because of this, we chose again a midrange value $\lambda(0)$: 1100 Å and used the extreme value of 1300 Å to demonstrate that slightly different values of $\lambda(0)$ do not introduce significant changes in the temperature dependence of $[\lambda(0)/\lambda(T)]^2$. The agreement with the nonlocal BCS theory is extremely good for $\lambda(0)=1100$ Å. As can be interpreted from Fig. 2, a variation of 20% in the value of $\lambda(0)$ just slightly degrades the agreement.

Let us now consider the temperature range significantly close to T_c. The nonlocal generalization of the electrodynamics of a superconductor establishes that the supercurrent $\mathbf{j}(\mathbf{x})$ is determined as a spatial average of the vector potential $\mathbf{A}(\mathbf{x}')$ in a region of dimension r_0

$$\mathbf{j}(\mathbf{x}) = -\frac{n_s e^2}{mc}\frac{3}{4\pi\xi_0}\int d^3x' \frac{\mathbf{X}[\mathbf{X}\cdot\mathbf{A}(\mathbf{x}')]}{X^4} e^{-X/r_0}, \qquad (3)$$

where $\mathbf{X}=\mathbf{x}-\mathbf{x}'$. n_s is the superfluid density, e and m are the electron charge and mass, respectively, and c is the light velocity. The characteristic length r_0 is defined as $1/r_0 = 1/\xi_0 + 1/l$, where l is the mean free path. From the BCS theory the *temperature-independent* coherence length $\xi_0 = \hbar v_F/\pi\Delta_0$, where v_F is the Fermi velocity. In pure superconductors $l \gg \xi_0$, and $r_0 \simeq \xi_0$. Since the vector potential varies with the penetration length $\lambda(T)$, in pure samples the average of $\mathbf{A}(\mathbf{x})$ needs to be carried out when $\xi_0 > \lambda(T)$. Therefore, this last condition defines the nonlocal limit of electrodynamics, and the opposite relation $[\xi_0 < \lambda(T)]$ determines the local limit. Because the magnetic penetration depth increases very rapidly as $T \to T_c$ in all superconductors, we expect that at temperatures very close to T_c the penetration depth would be larger than the coherence length. Therefore, if a superconductor is nonlocal for $T \ll T_c$, it should experience a crossover to a local behavior around the temperature at which $\lambda \approx \xi_0$.[4,5]

Figure 3 is a blowup of the temperature region near T_c for the Cd data. The expected crossover from nonlocal to local BCS behavior is observed. From this figure we can see that the data start deviating from the nonlocal curve at around $T/T_c \approx 0.94$, where the penetration depth and the coherence length are supposed to become similar. Taking $\xi_0 \approx 2200$ Å, which was estimated from shear-wave ultrasound attenuation,[11] and using $\lambda(0)=1100$ Å we get $[\lambda(0)/\lambda(T)]^2 = 0.25$ at $\lambda \simeq \xi_0$. In Fig. 3 this value corresponds to $T/T_c \approx 0.95$, which agrees remarkably well with the observed value of 0.94 mentioned above. Such an agreement gives further support to the all around consistency of our results.

In Fig. 4 we have plotted the Al data near T_c. We could see that the data seem to move toward the local curve just around T_c, even though the effect is somewhat masked by the fact that in this temperature region the data do not fall right onto the nonlocal curve. In any case, the crossover in Al should be much less evident than in Cd, since for Al the crossover should appear at $[\lambda(0)/\lambda(T)]^2 = 0.0009$; that is, very close to T_c. This makes it quite difficult to observe.

To conclude, from high precision magnetic penetration depth measurements we have found strong evidence that both Al and Cd are certainly nonlocal superconductors as theory predicts. We also found that the Cd data are consistent with the existence of the crossover from nonlocal to local electrodynamics theoretically expected near T_c for all superconductors.

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