Temperature Dependence of the Penetration Depth in Sr_2RuO_4: Evidence for Nodes in the Gap Function

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We report measurements of the magnetic penetration depth in single crystals of Sr_2RuO_4 down to 0.04 K using a tunnel-diode based, self-inductive technique. We observe a power law temperature dependence below 0.8 K, with no sign of a second phase transition nor of a crossover predicted for a multiband superconductor. A power law dependence suggests that the gap function has nodes, inconsistent with candidate p-wave states. We argue that nonlocal effects, rather than impurity scattering, can explain the observed T^2 dependence instead of the T-linear behavior expected for line nodes.

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Recently superconductivity in Sr_2RuO_4 (Sr214) has received considerable attention both theoretically and experimentally. This is because Sr214 is in the same layered-perovskite crystal class as the high-T_c superconductors but without copper and, especially, due to the existence of ferromagnetism in the related compound $SrRuO_3$. Shortly after the discovery of superconductivity in Sr214 [1], it was suggested that Sr214 might have odd-parity (p-wave) pairing symmetry [2]. Indeed, striking evidence has been found for a non-s-wave order parameter [3–6]. Specifically, an odd-parity (spin-triplet) pairing symmetry is required to explain the temperature-independent ^{17}O Knight shift for magnetic fields parallel to the conducting plane [6]. It has been argued that the isotropic (nodeless) p-wave state $\mathbf{d}(\mathbf{k}) = \Delta(0)\hat{\mathbf{z}}(k_x \pm ik_y)$ [2] is the best candidate gap function. In an attempt to explain many experimental results within the context of a nodeless, p-wave gap, an orbital dependent superconductivity (ODS) model was proposed [7]. The ODS approach assumes that of the three Fermi surfaces of Sr214 only one opens up a large isotropic gap, leading to exponential temperature dependence of thermodynamic and transport properties at low temperatures, whereas the other two have a much smaller proximity-induced isotropic gap. According to the model this second gap would lead to a crossover between the exponential temperature behaviors of the mentioned properties at very low temperatures or show up as a second superconducting transition.

However, recent specific heat [8] and NMR [9] experiments on high quality samples show no evidence of a crossover nor of a second transition down to 0.1 K. Moreover, the power law temperature dependences of C_e and $1/T_1$ found in these experiments are the same as those found for high-T_c and heavy-fermion superconductors [10]. Such similarities suggest that the gap function of Sr214 could have nodes which, if proven, would have profound implications for our current understanding of the superconductivity of Sr214.

Magnetic penetration depth $\lambda(T)$ measurements are fundamental probes of the existence of nodes in the gap function of unconventional superconductors. Here we report measurements of $\lambda(T)$ in Sr214 from T_c down to 0.04 K. In clean samples the data display a power law temperature dependence below $\sim 0.6T_c$, which is interpreted as strong evidence for nodes in the superconducting gap of Sr214.

The crystals used in the experiment were grown by a floating-zone method [11] and they come from three batches which differ significantly in quality. The samples have platelike shapes with typical dimensions $0.8 \times 0.8 \times 0.1$ mm^3. We label and summarize the characteristics of the samples in Table I. Samples Nos. 1 and 2 come from the same batch, but the surface of No. 2 was roughened with sandpaper. The T_c's were determined from the sharp peaks of the imaginary component of ac susceptibility measurements. $\lambda(T)$ measurements were performed utilizing a 28 MHz tunnel diode oscillator [12] with a noise level of 1 part in 10^9 and a low drift. The magnitude of the ac field was estimated to be less than 5 mOe.

TABLE I. Characteristics of the samples used in this work. T_{max} is the temperature up to which the data fit very well to $\Delta\lambda \propto T^n$.

Sample No.	T_c (K)	Treatment	n	T_{max} (K)
1	1.39	Not annealed	≈2	0.82
2	1.39	No. 1 with surface ground on sandpaper	≈2	0.82
3	1.44	Annealed in air, 1500 °C, 60 h	≈2	0.82
4	0.82	Annealed in oxygen, 1050 °C, 3 weeks	≈3	0.6

The cryostat was surrounded by a bilayer Mumetal shield which reduced the dc field to less than 1 mOe. Thus the samples were exposed to magnetic fields much smaller than $H_{c1\|} \approx 50$ Oe [13], greatly reducing the possibility of vortex contributions to the measured $\lambda(T)$. The sample was aligned inside the probing coil so that the *ab* plane was perpendicular to the rf field; thus we measured the in-plane penetration depth $\lambda_\|$ which corresponds to screening currents in the *ab* plane. The sample was 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 sample temperature was measured with a calibrated RuO_2 thermometer located at the end of the rod linked to the mixing chamber.

The deviation $\Delta\lambda(T)$ of the penetration depth from its $T = 0.04$ K value, $\Delta\lambda(T) = \lambda(T) - \lambda(0.04 \text{ K})$, 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 which depends on the sample and coil geometry and includes the demagnetizing factor of the sample. We determined G by measuring a 99.9995% Al sample that was cut to the same dimensions as one of the Sr214 crystals. G for the other crystals was scaled using the values of the slightly different thicknesses relative to the Al sample. The whole experimental system was checked via $\lambda(T)$ for 99.999% Al, Zn, and Cd samples. All yielded the expected BCS temperature dependence.

Figure 1(a) shows $\Delta\lambda(T)$ as a function of temperature for sample No. 1. The other samples showed an overall similar behavior, with no sign of a second phase transition nor of a crossover between two activated behaviors. This is consistent with low magnetic field specific heat [8] and thermal conductivity [14] measurements, and with NMR [9] results. Figure 1(b) shows $\Delta\lambda(T)$ against $(T/T_c)^2$ for samples Nos. 1 and 3 up to $0.78T_c$, and the inset to this figure displays the same data in the low temperature region $T \leq 0.2T_c$. Sample No. 2 has the same qualitative behavior observed in Fig. 1(b) but because of surface damage displays a larger field penetration. The slight change in slope observed at intermediate temperatures in the main body of Fig. 1(b) can be attributed to experimental errors, since several runs were taken for each of the samples and no reproducible and systematic changes in slope were found.

For comparison we evaluated numerically several superconductivity models. Except for sample No. 4 the others have the in-plane coherence length $\xi_0 \sim 660$ Å [13] and a mean free path $l > 5000$ Å [5], and therefore are in the clean limit ($l > \xi_0$). However, unlike cuprates and other unconventional superconductors, these samples are only marginally in the local limit since the Ginzburg-Landau parameter $\kappa \simeq \lambda(0)/\xi_0 \sim 2.89$, where the estimated zero-temperature in-plane penetration depth $\lambda(0) \sim 1900$ Å [4]. Nonetheless, to be consistent with the proposed models we assumed for the numerical calculation that Sr214 is a clean, local superconductor for which

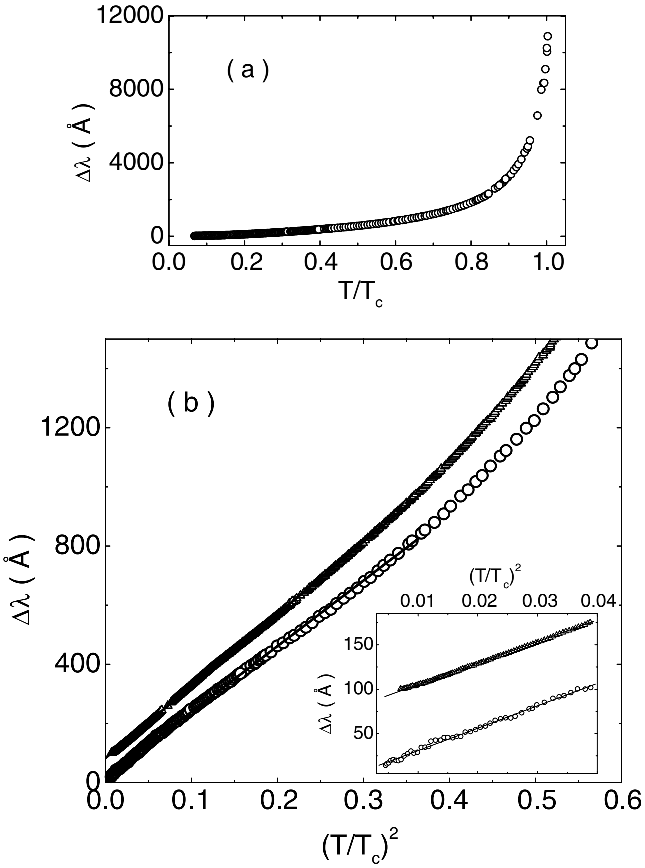

FIG. 1. (a) $\Delta\lambda(T)$ vs T/T_c for sample No. 1. (b) $\Delta\lambda(T)$ against $(T/T_c)^2$ in the temperature range (0.04–1.16) K for samples No. 1 (○) and No. 3 (△), and the inset shows the low temperature regime $T \leq 0.2T_c$ for the same data. All straight lines are fits to the data. The data have been shifted for clarity.

$$\frac{\lambda^2(0)}{\lambda^2(T)} = \sum_l N_l \left[1 + 2 \left\langle \int_0^\infty d\epsilon_l \, \frac{\partial f_l}{\partial E_{kl}} \right\rangle \right]. \qquad (1)$$

Here $\langle \cdots \rangle$ represents an angular average over the Fermi surface, N_l is the normalized electron density in band l, and f is the Fermi function. The total energy $E_l(k) = \sqrt{\epsilon_l^2 + |\mathbf{\Delta}_l(k)|^2}$, and ϵ_l is the single-particle energy measured from the Fermi surface. We assumed a cylindrical Fermi surface, a T-dependent gap function $\mathbf{\Delta}(k, T) = \Delta(T)\mathbf{d}(k)$, and used the weak-coupling gap interpolation formula $\Delta(T) = \Delta(0)\tanh(a\sqrt{T_c/T - 1})$ with $a \approx 1.6$ and $\Delta(0) = 1.76k_BT_c$. In computing $[\lambda(0)/\lambda(T)]^2$ from our data we used the value of $\lambda(0) = 1900$ Å given above, and we estimated the difference between $\lambda(0)$ and $\lambda(0.04)$ from the fit to T^2.

Along with the ODS and the s-wave weak-coupling BCS superconductivity models, we evaluated the anisotropic p-wave order parameter $\mathbf{d}(\mathbf{k}) = \Delta(0)\hat{\mathbf{z}}[\sin k_x \pm i \sin k_y]$ proposed by Miyake and Narikiyo (MN) [15]. In order to compare to a line-node-gap type of behavior we also evaluated the d-wave weak-coupling BCS model. For MN's model we used Eq. (5) (with the diameter of the Fermi circle parametrized by $R = 0.9$) of Ref. [15] for the gap amplitude and $N = 1$, and for the ODS approach we assumed that the small gap ($N_1 = 1/3$) is one-tenth of the large gap ($N_2 = 2/3$). The results of the calculations and $[\lambda(0)/\lambda(T)]^2$ of sample No. 1 are shown in Fig. 2. The inset shows the low temperature regime of $[\lambda(0)/\lambda(T)]^2$ of sample No. 1. A slight change in $\lambda(0)$ does not change the qualitative behavior of $[\lambda(0)/\lambda(T)]^2$ in any temperature region. A clear disagreement is seen from this figure between the experimental data and the ODS and the MN's anisotropic gap models.

From the inset to Fig. 1(b) it is clear that $\Delta\lambda(T)$ follows a T^n law, where $n \approx 2$, in the very low temperature region. This is remarkably different from the exponential behavior expected for a nodeless, isotropic gap function. Qualitatively any nodeless, anisotropic gap would yield a power law behavior for temperatures above the gap minimum, with an exponential dependence rising for temperatures below the minimum. The very low temperatures ($\sim$0.04 K) of our data indicate that a hypothetical anisotropy of the gap of Sr214 would need to be larger than 36 ($\sim$1.44 K/0.04 K) assuming the weak-coupling limit.

A power law temperature dependence of $\lambda(T)$ at the lowest temperatures would result only if the order parameter vanishes somewhere on the Fermi surface or in case of conventional s-wave gapless superconductivity due to strong impurity effects. Mackenzie *et al.* [5] showed that by increasing the concentration of nonmagnetic impurities in Sr214 superconductivity is eventually destroyed. From this we can rule out conventional gapless superconductivity, which is not destroyed by nonmagnetic impurities. Moreover, for conventional gapless superconductivity a linear temperature dependence is expected for C_e and $1/T_1$ [16]. However, it has been found experimentally that $1/T_1 \propto T^3$ [9] and $C_e \propto T^2$ (plus a small linear contribution) [8].

We conclude then that the power law temperature dependence of $\lambda(T)$ of Sr214 suggests nodes in the gap function. Indeed in high-T_c superconductors and UPt_3 low temperature dependences of $C_e \propto T^2$, $1/T_1 \propto T^3$, and $\lambda \propto T^n$, where $n = 1$ or 2, have been taken as indicative of line nodes [10].

A T-linear dependence is expected for line nodes in the clean, local limit. However, for impurity scattering in the unitary limit and line nodes in the gap function it was suggested [17] that a crossover from linear to quadratic behavior should occur in $\Delta\lambda(T)$ at a temperature T^* of order $\Delta(0)\sqrt{(T_{c0} - T_c)/T_{c0}}$, where T_{c0} is the impurity-free T_c. For Sr214 it is reasonable to assume that impurities act as strong scatterers, since impurities strongly modify the low temperature properties even for a small impurity concentration [18]. In Sr214 $T_{c0} \sim 1.5$ K [5], and T^* should be around 0.5 K in sample No. 3 and 0.66 K in samples Nos. 1 and 2. However, for these samples $\Delta\lambda(T) \propto T^2$ up to about 0.8 K and no linear region is found in this variable throughout the whole temperature range. In the local limit, we can analyze instead the superconducting electron density n_s $[n_s(T) \propto [\lambda(0)/\lambda(T)]^2]$. The analysis of $[\lambda(0)/\lambda(T)]^2$ for the experimental data plotted in Fig. 2 yields a crossover from linear to quadratic behavior around 0.25 K. However, this temperature is significantly lower than the T^* expected for sample No. 1. In order to gain further insight into impurity effects on $\lambda(T)$ or $n_s(T)$ we measured a dirty sample (No. 4) with $T_c = 0.82$ K. For this sample the expected $T^* > T_c$, i.e., $\Delta\lambda(T)$ and/or $n_s(T)$ should vary as T^2 up to some high temperature. However, as can be seen from Fig. 3, $\Delta\lambda(T) \propto T^n$ where $n \approx 3$ up

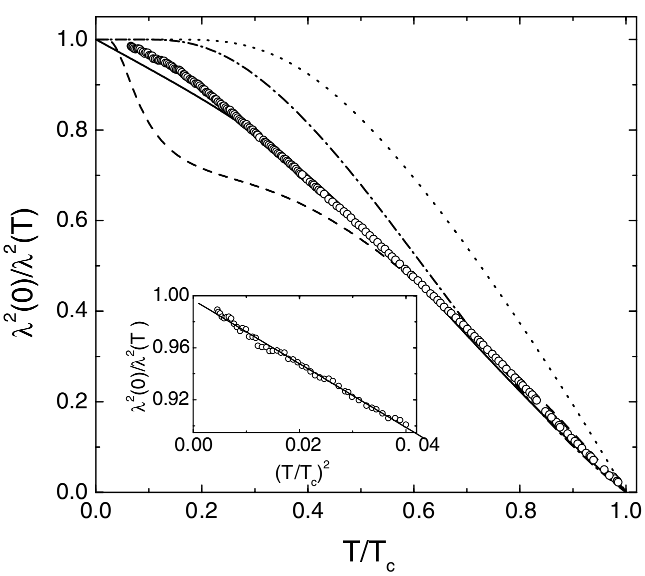

FIG. 2. Results of the calculations and $[\lambda(0)/\lambda(T)]^2$ for sample No. 1 up to T_c. The inset displays the low temperature region of $[\lambda(0)/\lambda(T)]^2$ of sample No. 1. The symbols correspond to (circle) penetration depth data; (dot) s-wave weak-coupling BCS; (solid line) d-wave weak-coupling BCS; (dash dotted line) MN's model; and (dashed line) ODS model.

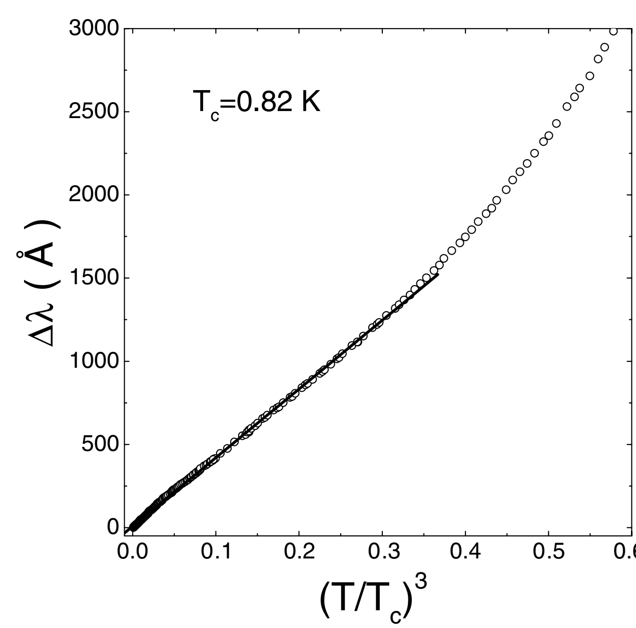

FIG. 3. $\Delta\lambda(T)$ against $(T/T_c)^3$ in the temperature range (0.04–0.69) K for sample No. 4. The line is a fit to the data.

to $0.6T_c$. This particular power law implies that n_s is not quadratic either. These discrepancies suggest that impurities may not be the cause of the T^2 variation.

Another plausible mechanism is the effect of nonlocal electrodynamics. It was shown by Kosztin and Leggett [19] that in an unconventional superconductor with nodes in the gap under certain conditions at low temperatures nonlocal electrodynamics leads to a $\Delta\lambda(T)$ proportional to T^2 and not T even in the clean limit. They predicted that nonlocal effects should be observable below a temperature $T^* = (\xi_0/\lambda_0)\Delta(0)$. In high-$T_c$ superconductors $T^* \sim 0.02T_c$, and therefore the effects would be observable only in the low temperature region. However in high quality Sr214 samples $T^* \simeq 0.61T_c \sim 1$ K, and we should expect nonlocal effects even in the intermediate temperature range. $\Delta\lambda(T) \propto T^2$ up to $T \sim 0.82$ K in samples Nos. 1, 2, and 3 as mentioned above. On the other hand, the T^3 dependence of $\lambda(T)$ in sample No. 4 could be due to both impurity scattering and nonlocality. Assuming that for this dirty sample in the local limit impurity scattering effects would make $\Delta\lambda_L(T) \propto T^2$, the T^3 behavior could follow from the fact that in the nonlocal limit below T^*, $\Delta\lambda(T) \propto \Delta\lambda_L(T)T$ [19]. These consistencies suggest that nonlocality may be important in Sr214. To our knowledge, there is no previous indication of nonlocality in any unconventional superconductor.

As a test of the significance of nonlocal effects we measured sample No. 3 with the applied magnetic field oriented parallel to the *ab* plane, since nonlocality is expected to be irrelevant in this orientation. No difference with respect to the measurements done with the field parallel to the *c* axis was observed. This result, however, may not rule out nonlocal effects as a possible explanation for the observed temperature dependences of $\Delta\lambda(T)$, because when the field is parallel to the planes the measured signal is a combination of $\lambda_{\parallel}$ and $\lambda_{\perp}$. Here $\lambda_{\perp}$ is the out-of-plane penetration depth, which corresponds to screening currents along the interlayer *c* direction. In the case of Sr214 $\lambda_{\perp}(0) \approx 20\lambda_{\parallel}(0)$ [13], and the signal when the field is parallel to the planes could be dominated by $\lambda_{\perp}$. Because there is no reason to exclude the possibility that $\Delta\lambda_{\perp}(T) \propto T^2$, the intrinsic $\Delta\lambda_{\parallel} \propto T$ in this case could be masked.

Finally, we consider the possible spin-triplet pairing states consistent with nodes in the gap function. All the p-wave states listed in the literature [2,16], including the A phase $[\mathbf{d}(\mathbf{k}) = \Delta(0)\hat{\mathbf{z}}(k_x \pm ik_y)]$ of the E_u representation, have nodeless gaps and are ruled out as the pairing state of Sr214. The B and C phases of the E_u representation [16] have gaps with nodes and are not in contradiction with the penetration depth result, but they are inconsistent with time-reversal symmetry breaking [3]. Nonunitary p-wave states with gapless quasiparticle excitations [20] would yield the same temperature dependences as conventional gapless superconductors for all thermodynamic and transport properties, and we have excluded above the possibility of gapless behavior in Sr214.

Other possible p-wave states, which break time-reversal symmetry and have nodes in the gap, would consist of complex combinations of two states belonging to different representations. This would lead, in general, to double phase transitions for which there is no experimental evidence. We conclude that recent experiments seem to be inconsistent with any p-wave pairing state.

In summary, we reported on rf magnetic penetration depth measurements on single crystals of Sr214 samples. Regardless of whether there is nonlocal or dirty-d-wave behavior, the data require the existence of line nodes, in contradiction to the nodeless p-wave picture. We argued that nonlocality may be relevant in accounting for the T^2 dependence of $\lambda(T)$.

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