Evidence of *d*-wave pairing symmetry of the gap of the heavy-fermion superconductor $CeIrIn_5$ from magnetic-penetration-depth measurements

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The heavy-Fermion compounds $CeTIn_5$, where T is Co, Ir, or Rh, exhibit two distinct superconducting regimes upon tuning the lattice density via doping or pressure. $CeIrIn_5$ is close to the boundary and there is some disagreement on whether its superconducting state is the same as $CeCoIn_5$ or not. In this Brief Report, we report the low-temperature penetration-depth $\lambda(T)$ measurements for $CeIrIn_5$. That $\lambda(T) \propto T$ below $0.3T_c$, irrespective of sample orientation, confirms the existence of line nodes in the gap with no evidence for the point node proposed in a hybrid-gap structure.

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The heavy-Fermion family $CeTIn_5(T=\mathrm{Co,Rh,Ir})$ offers rich insights into the interplay among electronic structure, composition, magnetism, and superconductivity. The highest superconducting transition temperature (T_c=2.3 K) among all Ce-based heavy-Fermion superconductors is found for T=Co,[1] for which there is some consensus that the order parameter is of *d*-wave character with line nodes parallel to the *c* axis.[2–4] On the other hand, $CeRhIn_5$ is an antiferromagnet (AFM) and becomes superconducting around a pressure-induced quantum-critical point (QCP),[5,6] showing a shifted, but otherwise similar, superconducting phase diagram to that of $CeCoIn_5$ under pressure. Substitution of Ir for Rh introduces chemical pressure and changes the c/a ratio,[7] suppressing the AFM order and inducing the appearance of a superconducting dome near and above the AFM QCP. Iridium substitution in $CeRh_{1-x}Ir_xIn_5$ above the AFM QCP reduces $T_c(x)$, which reaches a minimum (perhaps zero) around $x=0.9$. On further doping or applying pressure, a second superconducting region appears, continuing to $CeIrIn_5$.[8] The existence of two superconducting domes observed in $CeRh_{1-x}Ir_xIn_5$ (Refs. 6–8) is remarkably similar to that of $CeCu_2Si_2$ under pressure,[9] leading to the speculation that two distinct superconducting states in $CeTIn_5$ are tuned via lattice density or c/a ratio. It is important, therefore, to determine whether the order parameter of $CeIrIn_5$ has the same symmetry as $CeCoIn_5$ or if it is the Rh-rich dome that represents a distinct state. There is no obvious phase boundary in the $CeIr_{1-x}Co_xIn_5$ alloys, yet one thermal-conductivity study[10] suggests a different gap structure for $CeIrIn_5$. As a further puzzle, the resistivity and Hall effect drop sharply in $CeIrIn_5$ near 1.2 K,[11] even though there is no evidence for superconductivity from the magnetic, heat-capacity,[12] or thermal-conductivity data.[13] Other thermal-conductivity data,[10] however, indicate a change in the ratio of *c*-axis and *a*-axis data near 1 K that is absent in the presence of a magnetic field.

Recent measurement of the dependence of the thermal conductivity on orientation of the in-plane magnetic field reveals the existence of line nodes along the *c* axis of $CeIrIn_5$,[14] supporting a *d*-wave picture as in $CeCoIn_5$. However, competing thermal-conductivity data are argued to rule out simple line nodes in favor of a hybrid gap.[10] The proposed gap has a point node along c^* and a line node around the equator of the Fermi surface. In this Brief Report, we employ the temperature dependence of the penetration depth in $CeIrIn_5$ to examine the nature of the superconducting gap, attempting to settle the competing results from thermal conductivity. We also look for evidence for flux expulsion in the vicinity of the 1.2 K transition.

Single crystals of $CeIrIn_5$ were prepared from 99.99% pure materials using a self-flux method with excess In in an Ar atmosphere.[12] The quality of samples prepared in this way is confirmed by the detection of all Fermi surfaces by de Haas–Van Alphen experiments.[15] The as-grown samples were cut to a size appropriate for our millikelvin apparatus. A Van Degrift tunnel-diode oscillator was adapted[16] for use in an Oxford dilution refrigerator, with the sample mounted on a sapphire rod attached to the mixing chamber and positioned in the primary coil, maintained at a constant temperature of about 1 K.[17] The sample temperature could be varied from 80 mK up to 10 K. The change in the frequency of the tunnel-diode-sustained oscillations ($f \approx 23$ MHz) is proportional to the volume of the sample from which flux is excluded and therefore proportional to the penetration depth through the relation $\Delta\lambda(T)=G\Delta f(T)$, where G depends on sample size and geometry and is given by

$$G = \frac{-2\bar{R}(1-N)V_c}{V_s f_0}. \tag{1}$$

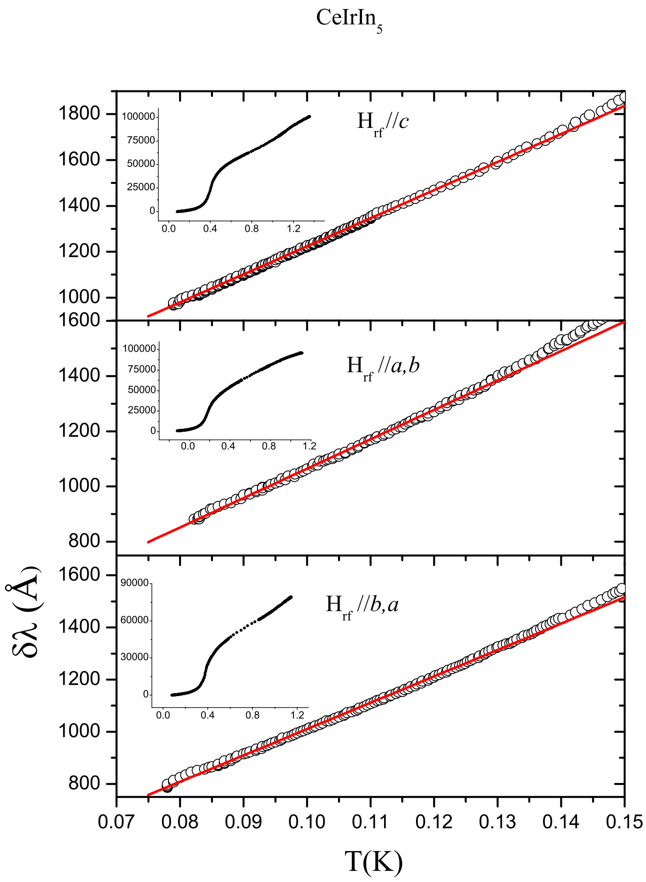

FIG. 1. (Color online) Penetration depth with the rf field along various axes. Inset: penetration depth over a wide temperature range. Solid lines are linear least-squares fits.

Here N, f_0, and $\bar{R}$ are the demagnetization factor, the operating frequency, and a geometrical correction,[18] respectively. V_s and V_c represent the sample and coil volumes. In this Brief Report, the sample G factor was experimentally determined from the ratio of Eq. (1), using the dimensions of an Al sample and the $CeIrIn_5$ samples, and the value G_{Al} obtained from a BCS fit to the Al data. We approximate the demagnetization factors for each sample from their dimensions using[19]

$$N \simeq 1 - (1 + w/2d)^{-1}, \tag{2}$$

where the thickness of the sample in the field direction is d and the area normal to the field is w^2.

The insets in Fig. 1 show the data over a wide temperature range from a $CeIrIn_5$ single crystal with the rf field along the c axis (G=9.6 Å/Hz) and two directions in the basal plane (G=15 Å/Hz); d is smaller than w in the c-axis orientation. The main panels in Fig. 1 demonstrate that the penetration depth measured in all three directions is linear in temperature from the base temperature of about 80 up to 140 mK (approximately 1/3 of T_c), indicating the existence of line nodes in the superconducting energy gap. The hybrid gap has a point node on the Fermi surface along the c^* direction which should give a T^2 contribution to c-axis screening currents. We used least-squares fits, shown as lines in Fig. 1, to extrapolate to $T=0$ in order to define the change in the penetration depth as $\delta\lambda(T)=\lambda(T)-\lambda(0)$. Other samples, from different batches, also show a linear dependence on temperature over a comparable interval.

When the rf field is along the c axis, the screening supercurrents flow in the ab planes, making the penetration depth thus measured simply the field penetration into the basal plane with no contribution from nodal quasiparticles at the proposed point node along c^*. Line nodes are present in both d-wave and hybrid scenarios and will give a linear temperature dependence in this orientation. However, with H_{rf} in the basal plane, screening currents flow across the ab plane and along the c axis. In the hybrid-gap picture, the latter currents would be dominated by quasiparticles excited at the point node, and these should exhibit a T^2 temperature dependence. In the absence of that signature of a three-dimensional point node, our data suggest that the quasiparticles that are thermally excited are found at line nodes along the c axis. Because the basal-plane data involve a geometric admixture of currents along and normal to c, $\lambda(0)$ will reflect differences in effective mass.

The experimental method used here measures changes in penetration depth with great precision but does not yield absolute values of $\lambda(0)$ nor are there data available from muon resonance or other techniques to ascertain it. For a d-wave superconductor with line nodes on a cylindrical Fermi surface, the simple BCS expression[20] for the low-temperature behavior of the penetration depth is given by

$$\lambda_{BCS}(T) \simeq \lambda(0)\left(1 + \frac{\ln 2}{\Delta_0} k_B T\right). \tag{3}$$

Using the weak-coupling d-wave value[21] $\Delta_0=2.14k_BT_c$ and $T_c \simeq 0.4$ K, we estimate $\lambda(0) \simeq 10^4$ Å from the slopes of the linear fits in Fig. 1. The actual value depends on the shape of the Fermi surface and the slope of the quasiparticle density of states vs energy near the node.

The quantity of fundamental interest is the fraction of electrons that are superconducting, often termed the superfluid fraction $\rho_s(T)$. Using the usual London expression for the penetration depth, it is straightforward to see that

$$\rho_s(T) = \left(\frac{\lambda(0)}{\lambda(T)}\right)^2 \simeq \left(1 + \frac{\delta\lambda(T)}{\lambda(0)}\right)^{-2}. \tag{4}$$

The temperature dependence of superfluid density is sensitive to the value of $\lambda(0)$. If the thermally excited quasiparticles are entirely associated with line nodes, it should be possible to collapse the data in all three directions using suitable values of $\lambda(0)$. If, on the other hand, a point node is present with quasiparticle momentum primarily along c^*, data collapse would not be possible. Rather than relying on Eq. (3) for each direction, we treat $\lambda(0)$ as an adjustable parameter for each data set. We expect the values for the two directions measured in the basal plane to be similar but not necessarily the same as that measured with the field along the c axis, reflecting the admixture of screening-current directions. Figure 2 shows that this process is successful if the penetration depth along the c axis is set at 9000 Å and the effective value in the basal plane is set at 7500 Å. Super-

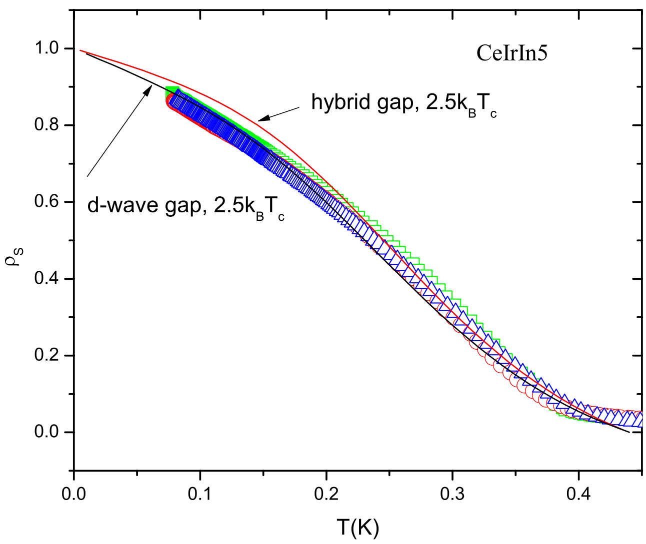

FIG. 2. (Color online) Superfluid fraction calculated to superpose data in all three directions. Red circles—c axis, $\lambda(0)=9000$ Å; basal plane: blue triangles—7500 Å; green squares—7500 Å. The solid lines are calculations for $d_{x^2-y^2}$ and a hybrid gap with $T_c=0.44$ K.

posed on the data is a curve expected for a line nodes in a d-wave picture with $\Delta_0/k_BT_c=2.5$. (See below for this choice of zero-temperature gap; we cannot distinguish between $d_{x^2-y^2}$ and d_{xy}.) The c-axis value of $\lambda(0)$ is in reasonable agreement with the estimate of Eq. (3). We note that the value $T_c=0.44$ is above the peak observed in the heat capacity but close to the onset temperature.

Arguing against a d-wave state, Shakeripour *et al.*[10] noted that the thermal conductivity along the c axis decreases more rapidly than that along a. They propose, instead, a hybrid gap of the form $\Delta(T)=2\Delta_0(T)\cos\theta\sin\theta e^{i\varphi}$, with $\Delta_0(0)$ set at $2.5k_BT_c$ to match the low-temperature slope of the a-axis thermal conductivity. We have plotted the superfluid density expected for that gap function in Fig. 2, using the interpolation expression,

$$\Delta_0(T) = \Delta_0(0)\tanh\left[\frac{\pi k_BT_c}{\Delta_0(0)}\sqrt{a\frac{\Delta C}{C}\left(\frac{T_c}{T}-1\right)}\right]. \quad (5)$$

Here we have[22] $\Delta C/C=0.76$ and $a=2/3$ for weak-coupling superconductors. The low-temperature curvature reflects the presence of a T^2 temperature dependence of quasiparticle excitations near the point node. We note that the hybrid gap breaks time-reversal symmetry which requires either that the superconducting dome in the $Ce(Ir,Co)In_5$ alloy system represents a broken time-reversal phase or that there is a second-phase transition at some Ir/Co ratio. More recent thermal-conductivity data by Kasahara *et al.*[14] exhibit fourfold oscillations in the a-axis thermal conductivity as a magnetic field is rotated in the basal plane, but no variation when the field is rotated within the ac plane. These researchers conclude that the gap structure for $CeIrIn_5$ is of d-wave form, as for $CeCoIn_5$, and our results support that conclusion.

We return now to the question of the resistive transition at 1.2 K. While there is clearly a sharp drop in resistance and

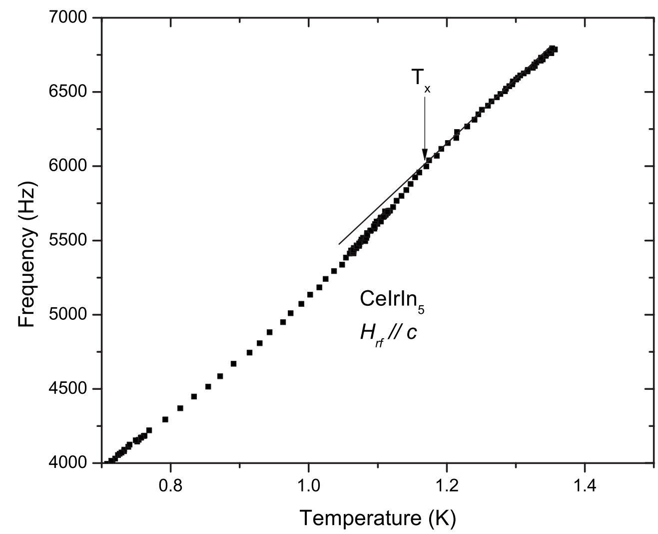

FIG. 3. Hint of a decrease in field penetration below $T_x\approx 1.2$ K for H_{rf} along c.

the disappearance of a Hall signal below that temperature,[11] no heat capacity or magnetic anomaly has been reported. We have examined the raw data for our $CeIrIn_5$ sample in this temperature range, with the result shown in Fig. 3. In this temperature range, we detect the temperature dependence of the normal skin depth at 23 MHz and are outside the range where calibration with data from Al can be used to determine G. Nonetheless, we can see a discernable break in the frequency data below ~ 1.2 K, meaning that the effective volume of the primary coil has decreased and therefore that there is less penetration of the rf field into the sample. Similar breaks in the data are seen in other samples from the same and different batches of $CeIrIn_5$ and in different runs on the same sample. While it is tempting to assign the decrease in field penetration to the appearance of a Meissner effect below 1.2 K, the effect seen here remains within our experimental uncertainty.

In summary, we have shown that the penetration depth in $CeIrIn_5$ is linear in temperature from our base temperature to $T_c/3$, with no evidence for a T^2 contribution, providing strong evidence for the existence of line nodes in the superconducting energy gap. Accounting only for anisotropy in the zero-temperature penetration depth, data taken with the rf field along c and in two directions in the ab plane converted to superfluid fraction, collapse to a single curve which follows BCS behavior for a d-wave gap. We show that with the parameters suggested,[10] a hybrid-gap function neither follows the data nor provides evidence for a T^2 contribution at low temperatures. We conclude that $CeIrIn_5$ has the same order-parameter symmetry as $CeCoIn_5$, as expected, if there is a single superconducting state in the $Ce(Co,Ir)In_5$ phase diagram. Finally, we provide some evidence for weak Meissner-flux exclusion in the vicinity of 1.2 K, where surface superconductivity is reported.

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[1] C. Petrovic *et al.*, J. Phys.: Condens. Matter **13**, L337 (2001).
[2] E. E. M. Chia, D. J. Van Harlingen, M. B. Salamon, B. D. Yanoff, I. Bonalde, and J. L. Sarrao, Phys. Rev. B **67**, 014527 (2003); R. J. Ormeno, A. Sibley, C. E. Gough, S. Sebastian, and I. R. Fisher, Phys. Rev. Lett. **88**, 047005 (2002).
[3] K. Izawa, H. Yamaguchi, Y. Matsuda, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. Lett. **87**, 057002 (2001).
[4] Y. Kohori, Y. Yamato, Y. Iwamoto, T. Kohara, E. D. Bauer, M. B. Maple, and J. L. Sarrao, Phys. Rev. B **64**, 134526 (2001).
[5] H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).
[6] M. Nicklas, V. A. Sidorov, H. A. Borges, P. G. Pagliuso, J. L. Sarrao, and J. D. Thompson, Phys. Rev. B **70**, 020505(R) (2004).
[7] P. G. Pagliuso, C. Petrovic, R. Movshovich, D. Hall, M. F. Hundley, J. L. Sarrao, J. D. Thompson, and Z. Fisk, Phys. Rev. B **64**, 100503(R) (2001); P. G. Pagliuso, R. Movshovich, A. D. Bianchi, M. Nicklas, N. O. Moreno, J. D. Thompson, M. F. Hundley, J. L. Sarrao, and Z. Fisk, Physica B **312-313**, 129 (2002).
[8] S. Kawasaki, M. Yashima, Y. Mugino, H. Mukuda, Y. Kitaoka, H. Shishido, and Y. Onuki, Phys. Rev. Lett. **96**, 147001 (2006).
[9] H. Q. Yuan *et al.*, Science **302**, 2104 (2003).
[10] H. Shakeripour, M. A. Tanatar, S. Y. Li, C. Petrovic, and L. Taillefer, Phys. Rev. Lett. **99**, 187004 (2007).
[11] S. Nair, S. Wirth, M. Nicklas, J. L. Sarrao, J. D. Thompson, Z. Fisk, and F. Steglich, Phys. Rev. Lett. **100**, 137003 (2008).
[12] H. Shishido *et al.*, J. Phys. Soc. Jpn. **71**, 162 (2002).
[13] R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **86**, 5152 (2001).
[14] Y. Kasahara, T. Iwasawa, Y. Shimizu, H. Shishido, T. Shibauchi, I. Vekhter, and Y. Matsuda, Phys. Rev. Lett. **100**, 207003 (2008).
[15] Y. Haga, Phys. Rev. B **63**, 060503(R) (2001).
[16] C. T. Van Degrift, Rev. Sci. Instrum. **46**, 599 (1975); N. H. Tea *et al.*, Physica C **280**, 281 (1997); R. W. Giannetta *et al.*, *ibid.* **321**, 207 (1999).
[17] I. Bonalde, B. D. Yanoff, M. B. Salamon, D. J. Van Harlingen, E. M. E. Chia, Z. Q. Mao, and Y. Maeno, Phys. Rev. Lett. **85**, 4775 (2000); B. D. Yanoff, Ph.D. dissertation, University of Illinois, 2000.
[18] R. Prozorov and R. W. Giannetta, Supercond. Sci. Technol. **19**, R41 (2006).
[19] R. Prozorov, R. W. Giannetta, A. Carrington, and F. M. Araujo-Moreira, Phys. Rev. B **62**, 115 (2000).
[20] C. Panagopoulos, J. R. Cooper, and T. Xiang, Phys. Rev. B **57**, 13422 (1998).
[21] H. Won and K. Maki, Phys. Rev. B **49**, 1397 (1994).
[22] C. Petrovic *et al.*, Europhys. Lett. **53**, 354 (2001).