## Anisotropic Temperature Dependence of the Magnetic-Field Penetration in Superconducting UPt<sub>3</sub>

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The anisotropy and temperature dependence of the magnetic-field penetration in superconducting UPt<sub>3</sub> have been measured by muon spin relaxation. The extrapolated zero-temperature values for the penetration depths parallel and perpendicular to the *c* axis are  $\lambda_{\parallel} = 7070 \pm 30$  Å and  $\lambda_{\perp} = 7820 \pm 30$  Å, respectively. The temperature dependences of  $\lambda_{\parallel}$  and  $\lambda_{\perp}$  are different and can both be accounted for by a superconducting gap function with a line of nodes in the basal plane and axial point nodes.

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There are many experiments which give indirect evidence for anisotropic superconductivity in heavy-fermion systems. These fall into two categories, namely, measurements of transport coefficients or specific heat at low temperatures (T), <sup>1,2</sup> and measurements showing transitions between different superconducting states.<sup>3,4</sup> Experiments of the first type have shown that the temperature dependence of bulk properties of heavy-fermion superconductors follow power laws at low temperatures as opposed to the exponentially activated behavior characteristic of conventional superconductors. The absence of activated T dependence implies nodes in the gap functions, which can be due to either unconventional pairing in a clean sample or conventional pairing in the presence of magnetic impurities.<sup>5</sup> In spite of the great activity<sup>5</sup> on heavy-fermion superconductivity, the only experiment which by itself resolves this ambiguity in favor of unconventional pairing is a transverse ultrasound measurement on UPt<sub>3</sub>,<sup>2</sup> where the attenuation was found to rise (from T=0) by  $\Delta \alpha \sim T$  and  $\Delta \alpha \sim T^3$  for sound propagating in the basal plane with polarization parallel and perpendicular to the basal plane, respectively. For conventional gapless superconductors,  $\Delta \alpha \sim T^2$ , independent of polarization direction. Ultrasound attenuation is a nonequilibrium property, and a prerequisite for its interpretation is a detailed understanding of the mean free path for thermally excited quasiparticles. In the present paper, we describe the first demonstration that a static property, namely, the magnetic penetration depth  $\lambda$ , of a heavyfermion superconductor has an anisotropic T dependence. Previous measurements of  $\lambda$  for several heavyfermion systems showed that  $\lambda$  increased from its T=0value in proportion to  $T^2$ , but did not address the issue of anisotropy.<sup>6,7</sup> The technique used here, muon spin relaxation  $(\mu^+SR)$ , is unique in that it both gives absolute values for  $\lambda$  and allows anisotropy to be measured in a single experimental run for a single face of the sample, with no mechanical disturbance of the apparatus. Furthermore, the 4.2-MeV muons used in this experiment penetrate  $\approx 70 \,\mu$ m into UPt<sub>3</sub> and thus probe bulk superconducting material. The important results of our experiment are (i) that  $\lambda(T=0)$  is consistent with the London formula where the effective mass is the renormalized (heavy) mass, and (ii) that the *T* dependence of  $\lambda$  is anisotropic in a manner which implies that the superconducting gap function has a line of nodes in the basal plane and point nodes along c.

There have been many descriptions of transverse-field (TF)  $\mu^+$ SR<sup>8</sup> as applied to superconductors. The method yields a time-dependent asymmetry function A(t)which is directly proportional to the  $\mu^+$  spin autocorrelation function,  $\langle \mathbf{S}_{\mu}(t) \cdot \mathbf{S}_{\mu}(0) \rangle$ , where  $\mathbf{S}_{\mu}(0)$  represents the known initial spin of the muons implanted in the sample. For static internal fields  $\mathbf{H} \perp \mathbf{S}_{\mu}(0)$ , A(t) is simply the Fourier transform (in time) of the field distribution  $\rho(H)$ . Thus,  $\mu^+$ SR is an excellent probe of the field inhomogeneities associated with vortex lattices in type-II superconductors.<sup>9</sup> The second moment of  $\rho(H)$  is  $\langle |\Delta H|^2 \rangle = 3.706 \times 10^{-3} \Phi_0^2 \Lambda_{\text{eff}}^{-4}$ , where  $\Phi_0 = 2.068 \times 10^{-7}$  $G cm^2$  is the magnetic flux quantum, and  $\Lambda_{eff}$ , which has dimensions of a length, is determined by the flux lattice constant d, magnetic penetration depth  $\lambda$ , and pair coherence length  $\xi$ , all measured in the plane perpendicular to H. Barford and Gunn<sup>10</sup> showed that for uniaxial superconductors, as long as  $2\pi\Lambda_{\rm eff} \gg d \gg \xi$ ,  $\Lambda_{\rm eff}$  is very simply related to the magnetic penetration depths  $\lambda_{II}$  and  $\lambda_{\perp}$  parallel and perpendicular to the symmetry axis c. In particular, for  $\mathbf{H} \perp \mathbf{c}$ ,  $\Lambda_{\text{eff}} = \sqrt{\lambda_{\parallel}\lambda_{\perp}}$ , while for  $\mathbf{H} \parallel \mathbf{c}$ ,  $\Lambda_{\rm eff} = \lambda_{\perp}$ . Thus  $\mu^+ SR$  allows us to determine  $\lambda_{\parallel}$  and  $\lambda_{\perp}$  in the same crystal without even reorienting the surface relative to the muon beam:  $\langle |\Delta H|^2 \rangle$ , and thus  $\Lambda_{\rm eff}$ , in any plane is measured by applying the field perpendicular to that plane, and choosing the polarization of the incident muon beam to be in the plane.

The single crystals used here were fifteen polished disks each 5 mm in diameter and 1 mm thick. The c axis of the resulting composite UPt3 sample was normal to the disks with a characteristic mosaic of 5°, while the basal-plane mosaic was 10°. The horizontal muon beam was incident parallel to c, and a was vertical. The disks were cut from the ingots studied in previous neutronscattering<sup>11</sup> and specific-heat<sup>12</sup> experiments. The residual resistivity  $\rho(0)$  of our samples, determined by fitting data between 3 and 1.8 K by the form  $\rho(T) = \rho(0)$  $+AT^2$ , is  $3.1 \pm 0.5 \ \mu \Omega$  cm along **a** and  $0.8 \pm 0.1 \ \mu \Omega$  cm along c. Combining our results for  $\rho(0)$  with the carrier density of 1e/(formula unit) derived from Hall-effect measurements<sup>13</sup> yields estimates of electronic mean free paths of  $l_{\parallel} = 2700 \pm 300$  Å and  $l_{\perp} = 660 \pm 100$  Å parallel and perpendicular to c, respectively. Both values are much larger than the superconducting coherence length  $\xi$ , <sup>11,14</sup> so we can neglect conventional impurity effects in the analysis of  $\lambda(T)$ . The experiment was performed at the M15 secondary channel of the TRIUMF cyclotron facility using a dilution refrigerator to cool the sample. The temperature was measured with a carbon-glass resistor which below 100 mK was calibrated by <sup>60</sup>Co nuclear orientation thermometry.

Figure 1 shows A(t) for  $\mathbf{H} \parallel \mathbf{c}$ , H = 183 G, in (a) the normal state, T = 650 mK >  $T_c$ , and (b) the superconducting state, T = 40 mK  $\ll T_c$ . The incident muon spin



FIG. 1. Muon spin precession amplitude for transverse field  $H \parallel c$ , H = 183 G, and temperatures (a) above and (b) below  $T_c$ . Solid lines are from fits described in text.

was along  $\mathbf{a}$ . The relaxation of the coherent muon precession is substantially larger in the superconducting state than in the normal state, which shows that, as expected, the internal field in the vortex lattice is inhomogeneous.

In the mixed state of a type-II superconductor, the field distribution probed by the muons is a convolution of the effective-field distribution characteristic of relaxation processes also present in the normal state, and  $\rho(H)$ , the field distribution for the vortex lattice. Thus, by the Fourier product rule,  $A(t) = G_n(t)G_{sc}(t)$ , where  $G_n(t)$  is the normal-state relaxation function, and

$$G_{\rm sc}(t) = \int_{-\infty}^{\infty} dH \rho(H) \cos(\gamma_{\mu} H t + \phi_0) \, dt$$

Choosing

$$G_n(t) = \exp[-(\sigma_{g1}t)^2] \{\beta + (1-\beta)\exp[-(\sigma_{g2}t)^2]\},\$$

good fits are obtained above  $T_c$  with  $\beta = 0.83(2)$ ,  $\sigma_{g1} = 0.056(1) \ \mu \text{s}^{-1}$ ,  $\sigma_{g2} = 0.32(2) \ \mu \text{s}^{-1}$  for  $H \parallel c$  and  $\beta = 0.78(1)$ ,  $\sigma_{g1} = 0.050(1) \ \mu \text{s}^{-1}$ ,  $\sigma_{g2} = 0.29(1) \ \mu \text{s}^{-1}$  for  $H \parallel a$ . UPt<sub>3</sub> [Fig. 1(a)] has weak antiferromagnetic order<sup>11</sup> which accounts for some of the TF relaxation<sup>15</sup> in the normal state. On the basis of previous  $\mu$ SR and neutron-diffraction work, the changes in the antiferromagnetic contributions to  $G_n(t)$  are negligible for  $T \lesssim T_c$ , so we take  $G_n(t)$  to be T independent. We choose  $G_{sc}(t) = \exp[-(\sigma_{sc}t)^2]\cos(\gamma_{\mu} \langle H_i \rangle t + \phi_0)$  which, when combined with the form selected for  $G_n(t)$ , gives an excellent account of the data below  $T_c$  [Fig. 1(b)].



FIG. 2. T dependence of the Gaussian relaxation rate obtained from transverse field. Solid lines are from fits described in the text.

Figure 2 shows the T dependence of  $\sigma = (\sigma_{sc}^2 + \sigma_{g1}^2)^{1/2}$ for H||c and H||a obtained by fitting data similar to those of Fig. 1. Above 450 mK,  $\sigma$  is T independent, corresponding to the normal-state relaxation parameter  $\sigma_{g1}$ . Below 450 mK,  $\sigma_{sc}$  increases by approximately 0.15  $\mu s^{-1}$  for H||c and H||a. For both field orientations,  $\sigma_{sc}(T)$  is not exponentially flat for  $T \rightarrow 0$ , as is the case for conventional s-wave superconductors. Instead, for H||c, it appears that  $\sigma_{sc}$  falls linearly with T from its T=0 value. For H||a, however, curvature considerably beyond the statistical errors is apparent for 40  $\leq T \leq 200$  mK.

The magnetic penetration depth can be written as  $\lambda^{-2}(T) = \lambda^{-2}(0)\rho_s(T)$ , where  $\rho_s(T)$  is the normalized superfluid density. If the superconducting gap has nodes at particular points  $\mathbf{k}_n$  of the Fermi surface  $\{\mathbf{k}_F\}$ , normal quasiparticles with momenta close to such nodes are most easily excited and  $\lambda^{-2}$  decreases more rapidly along directions parallel to  $\mathbf{k}_n$ . In Fig. 3 we plot  $\lambda^{-2}$ along two principal directions as extracted from the data of Fig. 2 and the theory of Barford and Gunn:  ${}^{10} \lambda_{\perp}^{-2}$  is now clearly seen to have a stronger T dependence than  $\lambda_{\parallel}^{-2}$  as  $T \rightarrow 0$  which leads us to conclude that the density of gap nodes is higher for  $\mathbf{k}_F \perp \mathbf{c}$  than for  $\mathbf{k}_F \parallel \mathbf{c}$ . Indeed, if the data for  $T \leq 300$  mK are fitted by simple power laws  $\lambda^{-2}(T) = \lambda^{-2}(T=0)$ , then  $\lambda_{\perp}^{-2}, \lambda_{\parallel}^{-2} \sim T^{\alpha}$  with  $\alpha = 1.3 \pm 0.1$  and  $2.4 \pm 0.2$ , respectively. The results are thus consistent with expectations based on transverse ultrasound measurements.<sup>2</sup>

To determine consistency with particular forms for the gap function, we use the full expression for  $\zeta_{s\parallel,\perp}(T)$  (Ref. 7), valid in the clean limit which is realized for our samples,

$$\rho_{s\parallel,\perp}(T) = 1 - \frac{3}{n(0)} \sum_{\mathbf{k}} \hat{\mathbf{k}}_{\parallel,\perp}^2 \left[ -\frac{\partial f}{\partial E_{\mathbf{k}}} \right],$$



FIG. 3. Temperature dependence of  $\lambda_{\parallel}^{-2}$  (open circles) and  $\lambda_{\perp}^{-2}$  (solid circles) for  $T \leq 300$  mK. Lines are fits assuming various nodal structures in the superconducting gap and  $T_c = 474$  mK.

where n(0) is the density of states at the Fermi surface,  $f = [\exp(E_k/k_BT) + 1]^{-1}$  is the Fermi function,  $E_k$   $= [\varepsilon^2(\mathbf{k}) + \Delta(\mathbf{k}, T)^2]^{1/2}$ , and  $\varepsilon(\mathbf{k})$  is the quasiparticle energy measured from the Fermi surface. Following Gross et al.<sup>7</sup> we approximate the T-dependent gap function by  $\Delta(\hat{\mathbf{k}}, T) = \Delta(T)\delta(\hat{\mathbf{k}})$  and assume a spherical Fermi surface. For  $\Delta(T)$  we use the weak-coupling gap interpolation formula  $\Delta(T) = \Delta_0 \tanh(a\sqrt{T_c/T-1})$ . The constant  $a \approx 1.6$  is only weakly dependent<sup>7</sup> on the symmetry of the superconducting state and was fixed at 1.6 throughout. Guided by our conclusions above, we investigate the polar state  $\delta^2(\hat{\mathbf{k}}) = \hat{\mathbf{k}}_{\parallel}^2$ , which has a line of nodes in the basal plane. This model (dashed line in Fig. 3) can account for the linear T dependence of  $\lambda_{\perp}^{-2}$  but predicts a much weaker T dependence of  $\lambda_{\parallel}^{-2}$  than experimentally observed. A gap node for  $\mathbf{k} \parallel \mathbf{c}$  can account for  $\lambda_{\parallel}^{-2}$  as shown by the dotted line, calculated for the axial state  $\delta^2(\hat{\mathbf{k}}) = \hat{\mathbf{k}}_{\perp}^2$ . Not surprisingly, however, this state cannot reproduce the linear T dependence of  $\lambda_{\perp}^{-2}$ . When combining the axial and polar nodes, taking  $\delta^2(\hat{\mathbf{k}}) = 2\hat{\mathbf{k}}_{\parallel}^2 \hat{\mathbf{k}}_{\perp}^2$  (solid line) excellent agreement for both principal directions is obtained not only to 300 mK (Fig. 3), but to  $T_c$ , as the accord between the solid lines and the data in Fig. 2 demonstrates.

In most theories  $^{16,17}$  of the superconductivity of UPt<sub>3</sub>, a two-dimensional complex vector  $\eta = \eta_1 + i \eta_2$  represents the order parameter. If we ignore the effects on  $\Delta(\hat{\mathbf{k}}, T)$ of strongly coupled superconducting and (changing) magnetic order parameters,<sup>16</sup> the corresponding gap function is proportional to  $\delta^2(\hat{\mathbf{k}}) = \hat{\mathbf{k}}_{\parallel}^2 [(\eta_1 \cdot \hat{\mathbf{k}}_{\perp})^2 + (\eta_2 \cdot \hat{\mathbf{k}}_{\perp})^2]$  $(\hat{\mathbf{k}}_{\perp})^2$ ]. In the case  $\eta_1 \parallel \eta_2$ ,  $\delta(\hat{\mathbf{k}})$  has nodal lines out of the basal plane for  $\hat{\mathbf{k}}_{\perp} \perp \boldsymbol{\eta}_1$ , a situation which our data exclude. The gap function used to fit our data corresponds to  $\eta_1 \perp \eta_2$ , a state which maintains sixfold symmetry. More general situations in which  $\eta_1 \times \eta_2 \neq 0$  also have polar lines of nodes and axial point nodes, and so may also be consistent with our data. Such states break sixfold symmetry, a feature of several explanations proposed for the splitting of the zero-field superconducting transition found in specific-heat measurements.<sup>4</sup>

We can also extract quantitative characteristics of the superconducting state from the fit of Fig. 2. First,  $T_c = 474 \pm 7$  mK, consistent with the specific-heat result<sup>12</sup> for the same ingot. Second, the maximum value for the superconducting gap is  $\Delta_0 = 940 \pm 15$  mK, so that the ratio  $\Delta_0/T_c = 2.0 \pm 0.1$  is close to the weak-coupling values 2.03 and 2.46 calculated for axial and polar states, respectively. Third, the T=0 penetration depths are  $\lambda_{\parallel}(0) = 7070 \pm 30$  Å and  $\lambda_{\perp}(0) = 7820 \pm 30$  Å. Correcting for the finite mean free path using  $\lambda_{L\parallel,\perp} \approx \lambda_{L\parallel,\perp} (1 + \xi/l_{\parallel,\perp})^{-1/2}$  and the estimate  $\xi = 120$  Å based on  $H_{c2}(0)$  measurements<sup>11</sup> yields clean-limit values  $\lambda_{L\parallel} = 6920 \pm 40$  Å and  $\lambda_{L\perp} = 7200 \pm 100$  Å. The values are remarkably close to  $c/\omega_p$ , where  $\omega_p = 4.8 \times 10^{14}$  s<sup>-1</sup> is the plasma frequency deduced from infrared reflectivity data.<sup>18</sup> The entire low-frequency por-

tion of the optical conductivity is thus removed below  $T_c$ , which excludes descriptions of UPt<sub>3</sub> as a conventional gapless superconductor. From  $\lambda_L(0)$  we derive  $H_{c1}(0) = \Phi \ln[\lambda_L(0)/\xi]/4\pi[\lambda_L(0)]^2 \approx 14$  and 13 G parallel and perpendicular to c, respectively. The applied fields in our experiment were therefore well above  $H_{c1}(0)$ , a requirement for the applicability of our analysis. Consistent with these values,  $\mu$ SR internal-field magnetometry yields an upper limit of 30 G on  $H_{c1} \parallel n$ . The discrepancy between our results for  $H_{c1}$  and the higher values inferred from some bulk measurements<sup>19</sup> is probably due to the notorious difficulty of establishing a rigorous criterion for  $H_{c1}$  and to flux pinning both of which affect bulk experiments but not the internal-field inhomogeneity probed by  $\mu^+$ SR.

The magnetic penetration depth at T=0 is generally related by a London formula to the carrier density *n* and effective mass  $m_{\text{eff}}$ :  $\lambda^{-2}(0) = 4\pi ne^{2}/c^{2}m_{\text{eff}}$ . Substituting the previously quoted estimate for *n*, our results for  $\lambda_L$  then imply effective carrier masses  $m_{\text{eff}\parallel} = 260m_e$  and  $m_{\text{eff}\perp} = 280m_e$ . For comparison, the mass enhancement  $m^*/m_e$  from specific heat<sup>12,20</sup> based on the same estimate of the carrier density is 280. The renormalization of  $\lambda^{-2}(0)$  is in disagreement with what occurs for single-component Fermi liquids,<sup>21</sup> but consistent with work<sup>22</sup> on heavy-fermion metals, which are strongly interacting two-component systems.

In conclusion, we have performed the first measurements of the anisotropic magnetic penetration depth  $\lambda$  in a heavy-fermion superconductor. The T=0 values are consistent with a pair condensate of heavy carriers whose effective mass  $m_{\text{eff}}$  is close to the effective mass which can be deduced from specific-heat measurements. While  $\lambda^{-2}(T=0)$  is only slightly anisotropic,  $\lambda^{-2}(T)$  decreases proportionally with T when measured perpendicular to c but with a higher power parallel to c. The data can be understood in terms of weak-coupling superconductivity where the gap has both a line of nodes in the basal plane and axial point nodes.

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