Temperature Dependence of the Magnetic Penetration Depth and Nodal Gap Structure of UPt$_3$ from Small Angle Neutron Scattering

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We report measurements of the temperature dependence of the different components of the London magnetic penetration depth using small angle neutron scattering from the vortex lattice in a high quality crystal of UPt$_3$. Deconvolution of the contributions to the neutron scattering form factor from currents along the $a^*$ and $c$-axes gives direct information about the nodal structure of the order parameter. Our observations of linear temperature dependence at low temperatures of all components of the penetration depth support the assignment of $E_{2u}$ symmetry to the superconducting state of UPt$_3$.

The heavy fermion compound UPt$_3$ has attracted substantial interest as a paradigm for unconventional superconductivity. Among a host of fascinating physical properties, perhaps most striking is the fact that the $H-T$ superconducting phase diagram has three distinct superconducting vortex phases shown in Fig. 1(a), labeled A, B, and C. Experiments and theory demonstrate that this phase diagram can only be explained by an unconventional superconducting order parameter, a close parallel to superfluid $^4$He. However, a complete theoretical description of the superconducting state of UPt$_3$ has not been settled, and there are several candidate theories that can account for the material's unusual physical properties. The order parameter structure consistent with the widest variety of experiments is an odd-parity, spin-triplet, $f$-wave orbital state of $E_{2u}$ symmetry. With some success, comparisons with experiment have also been made for an even-parity, spin-singlet, $d$-wave orbital state of $E_{1g}$ symmetry. Both of these order parameters break time reversal symmetry in the low temperature B phase. A more recent proposal for an odd-parity, spin-triplet, $f$-wave model with $E_{1u}$ orbital symmetry, is time reversal symmetric in the B phase, in contrast to the other two models.

All of these order parameters have nodes in the superconducting energy gap, each with different nodal structure in the three vortex phases. Consequently, it is of particular importance to explore physical properties that are directly linked to this nodal structure. In this Letter we report our constraints on order parameter symmetry based on small angle neutron scattering (SANS) measurements from the vortex lattice (VL) in the superconducting mixed state. We have measured the temperature dependence of the components of the London penetration depth, $\lambda(T)$, that probe the gap nodal structure along the principal directions of the crystal. We compare our results for $\lambda(T)$ with those from other methods including ac-susceptibility and muon spin rotation ($\mu$SR). In our work we find a linear temperature dependence of $\lambda$ for currents parallel to the $a^*$ and $c$-axes consistent with an $E_{2u}$ order parameter.

Our sample consists of a high-quality, 15 g single crystal (RRR > 600), cut into two pieces, and is described by Gannon et al. The UPt$_3$ crystals were co-aligned, fixed with silver epoxy to a copper cold finger, and mounted to the mixing chamber of a dilution refrigerator with the crystal $a$-axis vertical and the $c$ and $a^*$-axes in the horizontal scattering plane. Rotation of the dilution insert allowed easy reorientation of the $a^*$ or $c$-axes to be parallel to the magnetic field and neutron beam inside a horizontal superconducting magnet on the SANS-I and SANS-II beamlines at the Paul Scherrer Institut in Villigen, Switzerland. For measurements on SANS-I, the neutron wavelength was 6 Å with 11 m of collimation. For measurements on SANS-II, 9 Å neutrons were used with 6 m of collimation.

Fig. 1(b) shows an example of a SANS diffraction pattern from UPt$_3$, with magnetic field $H = 0.3$ T parallel to the $a^*$-axis. As UPt$_3$ has relatively long penetration depths, only the first order Bragg reflections are evident in this image which is a superposition from rocking angles about the horizontal where the Bragg condition was satisfied for diffraction spots above the beam center. For all data discussed in this letter, background scattering measured in zero applied field was subtracted. The diffraction pattern shown in Fig. 1(b) is of a distorted hexagonal VL, similar to, but more anisotropic than, previous SANS measurements in this orientation. By symmetry, there are four additional peaks indicated by red circles that were not imaged in the hexagonal domain in Fig. 1(b). The symmetry of the diffraction pattern is the same as that of the real space VL, rotated by 90 degrees with a rescaling of the axes. The distortion of the VL from a perfect hexagon is a result of penetration depth anisotropy in the plane perpendicular to $a^*$. When a 0.2 T field is applied parallel to the $c$-axis, a perfect hexagonal VL is seen, in agreement with previous measurements in that orientation at a similar field.

In our work we find a linear temperature dependence of $\lambda$
The ground state.

parameter orientation and to ensure that the VL was in the superconducting state with an equilibrium order preparating the VL with this field history was to produce a final measurement field was reached. The motivation for $T$ was performed around the desired field until the final magnetic field dependence in the B-phase that becomes field independent in the C-phase, where the B to C-transition are shown as white arrows. The UPt$_3$ crystal axes are guides to the eye.

Fig. 2 shows the opening angle $2\alpha$ of the VL, defined in the inset, shown as a function of applied magnetic field along the $a^*$-axis measured at $\approx 50$ mK. The B to C-phase transition is given by the vertical dashed line at $H = 0.6$ T. Red lines are guides to the eye.

Our $2\alpha$ data can be best described as having a linear field dependence in the B-phase that becomes field independent in the C-phase, where the B to C-transition occurs between $H = 0.5$ and 0.6 T in this field orientation. Field dependence to the opening angle indicates that there are deviations from the London theory associated with non-local corrections. A change in behavior of the opening angle at the B to C-transition was also reported by Yaron et al.

The intensity in a diffraction peak is related to the Fourier transform of the local field variations from the real space VL. The scattered intensity in a reflection, $I(\phi)$ is shown in Fig. 3 as a function of rocking angle for two different magnetic fields. The scattered intensity decreases as magnetic field is increased since the proportionate increase in vortex density reduces the contrast in local field spatial variations. We measured the first-order Fourier component of the diffraction, called the form factor $|h_1|$, expressed as,

$$|h_1|^2 = R \frac{16\phi_0^2 q}{2\pi \gamma^2 \lambda_n^2 t}.$$  \hspace{1cm} (1)

The form factor is calculated from the reflectivity, $R$, equal to the integrated intensity, multiplied by $\cos \alpha$ (the Lorentz factor), divided by the incident neutron flux. In Eq. 1, $\Phi_0 = 2.07 \times 10^5 T \cdot \AA^2$ is the magnetic flux quantum; $q$ is the magnitude of the scattering vector of the reflection being measured; the gyromagnetic ratio of the neutron is $\gamma = 1.91$; $\lambda_n$ is the incident neutron wavelength; and $t$ is the effective sample thickness which we have taken to be 3.9 mm – the equivalent thickness of a uniform sample with the same width, height, and volume as our sample. The field dependence of our measurements of rocking curve widths, such as those shown in Fig. 3 do not show the sudden broadening at the B-C phase transition reported by Yaron et al. All of our rocking curves are approximately 20% broader than the resolution limit for our experiments. We also do not see a change in slope of the field dependence of $|h_1|$ at the B-C transition as reported earlier. It is likely that absence of these effects can be attributed to the higher quality of our crystal and the oscillatory field procedure which we have used.
to overcome flux pinning.

In London theory, the form factor is related to the material properties through the magnetic penetration depth $\lambda$. For an isotropic superconductor the form factor is given by,

$$|h_1| = \frac{B}{1 + \lambda_2 q^2 e^{-\xi^2 q^2}}$$  \hspace{1cm} (2)

where $\xi$ is the superconducting coherence length and $c$ is a constant, typically taken to be $\frac{1}{2}$. The fractional part of Eq. 2 comes directly from the London equations. The exponential factor is a correction to the London theory to account for the non-zero extent of the vortex cores. For an anisotropic superconductor the form factor can be expressed in terms of the principal values of the penetration depth $\lambda_i$ corresponding to currents flowing along each of the principal directions of the crystal. These components of the penetration depth are related to corresponding diagonal components of the quasi-particle mass tensor $m_i$ in much the same way as for the isotropic London theory, $\lambda_i^2 \propto m_i$, providing a direction-specific measure of the low lying excitations in the superconducting state sensitive to gap nodes.

For uniaxial anisotropy, as for UPt$_3$, $\lambda_1 = \lambda_2 \neq \lambda_3$ where $i = 3$ corresponds to the $c$-axis. The form factor for field along the $a$ or $a^*$-axis becomes

$$|h_1| = \frac{B}{1 + \lambda_2^2 q^2 \sin^2 \alpha + \lambda_3^2 q^2 \cos^2 \alpha} e^{-\xi^2 q^2}. \hspace{1cm} (3)$$

If there is no variation in the VL geometry as a function of temperature – a VL phase transition or temperature driven disorder, for example – then the temperature dependence of the form factor given by Eq. 2 and 3 reflects the temperature dependence of $\lambda_i$. Strictly speaking, the coherence length $\xi$ is also anisotropic and should be taken into account in the exponential factor of Eq. 3. However, the anisotropy in the coherence length is minimal as estimated from $H_{c2}$ measurements and its temperature dependence is sufficiently weak at low temperatures, that the effect of anisotropy is negligible.

We have made measurements of the temperature dependence of the VL scattering for magnetic fields along both the crystal $c$ and $a^*$-axes with the field reduced from above $H_{c2}$, after which the only field changes were damped oscillations before measurement at every temperature. Rocking curves were obtained for each orientation at base temperature and at intermediate temperatures to determine that there was no broadening as temperature was varied. The magnet and sample were rotated to the center of the rocking curve and the scattered intensity $I(T)$ was measured “rocked-on” as a function of temperature. As an example of our raw data, we show $I(T)$ at two fields parallel to the $a^*$-axis in Fig. 4. The diffracted intensity does not approach a constant at the lowest temperatures for either field orientation, indicating the presence of nodes in the superconducting gap.

![Figure 4](image-url)

**FIG. 4.** (color online) The rocked-on scattered intensity as a function of applied magnetic field for fields along the $a^*$-axis at $H = 0.2$ T (red circles) and 0.4 T (blue triangles). Solid red arrow indicates $T_c$ at 0.2 T. Dashed blue arrow indicates $T_c$ at 0.4 T.

The opening angle and the scattering vector $q$ as a function of temperature. For each panel, data is shown for fields along the $a^*$-axis at 0.2 T (red circles) and 0.4 T (blue triangles). For $H||c$ at 0.2 T the data (green squares) are within error bars of being a perfectly hexagonal VL. Solid lines show the average for each data set. Dashed lines in panel (b) show $q$ calculated using the average values of $2\alpha$ form panel (a), assuming $B=H$ and single flux quantization.

![Figure 5](image-url)

**FIG. 5.** (color online) (a) The opening angle $2\alpha$ and (b) the scattering vector $q$ as a function of temperature. For each panel, data is shown for fields along the $a^*$-axis at 0.2 T (red circles) and 0.4 T (blue triangles). For $H||c$ at 0.2 T the data (green squares) are within error bars of being a perfectly hexagonal VL. Solid lines show the average for each data set. Dashed lines in panel (b) show $q$ calculated using the average values of $2\alpha$ form panel (a), assuming $B=H$ and single flux quantization.
Then calculated $\lambda_3(T)$ from Eq. 3 using our values for $\lambda_1(T)$ and our $|h_1|$ values for $H/|a^*|$, with $c = \frac{1}{2}$ and $\xi = 110$ Å. The results of this analysis are shown in Fig. 6.

The limiting, low temperature region of our results appears to be for $T/T_c < 0.3$, where both $\lambda_1$ and $\lambda_3$ are linear. In Fig. 6 we show our fits to the data in this region. Our extrapolation to zero temperature gives $\lambda_1(0) = 6.830 \pm 0.208$ Å and $\lambda_3(0) = 3.990 \pm 0.43$ Å. There is evidence for $\lambda_3$ to deviate from linearity for $T/T_c > 0.3$, in contrast to $\lambda_1$.

On comparing our results with earlier work, we find that interpretation of ac-susceptibility measurements requires an analysis of the real and imaginary parts of the electromagnetic response from which extraction of the penetration depth is not trivial and is necessarily sensitive to surface quality. Signore et al. reported a linear temperature behavior which could not be associated with any specific component of the penetration depth. An early $\mu$SR investigation by Broholm et al. found a penetration depth anisotropy much too small to be consistent with other observations of the superconducting state. In a later $\mu$SR study, Yaouanc et al. found $\lambda_1(0) = 6.040 \pm 0.130$ Å and $\lambda_3(0) = 4.260 \pm 0.150$ Å with $H = 0.018$ T, giving a quasiparticle mass anisotropy of $m_1/m_3 = 2.94 \pm 0.24$ in good agreement with the anisotropy of $2.93 \pm 0.3$ that we calculate from the VL opening angle at $H = 0.2$ T. We infer their mass anisotropies to be 2.66 and 2.15, respectively. It is difficult to make a reliable comparison of our results for $\lambda_{1,3}$ with these two experiments, as they assume field independent penetration depths, inconsistent with measurements of the field dependence of the opening angle, and which might not be theoretically justified.

To interpret our data in terms of the pairing symmetry of UPt$_3$, we provide a brief discussion of the nodal structures of the superconducting gap for the various candidate theories. In the low field and low temperature B-phase where all of the data shown in Fig. 6 are measured, the three predominate pairing models discussed earlier have three different nodal structures. For the $E_{2u}$ model there are point nodes at the poles of the Fermi surface which open with quadratic wave-vector dispersion, and a line node around the equator of the Fermi surface that opens with linear dispersion. The $E_{1g}$ model also has point nodes at the poles, however these nodes open linearly. Similar to $E_{2u}$, the $E_{1g}$ model also has a line node around the equator that opens linearly. The $E_{1u}$ model has a somewhat more complicated gap structure in the B-phase, with point nodes at the poles that have a non-trivial dispersion and an antinode at the equator.

Our measurements of $\lambda_1$ are sensitive to the nodal structure at the equator, while those of $\lambda_3$ are sensitive to the nodal structure at the poles. For a point node with quadratic dispersion, a linear temperature dependence of $\lambda$ is expected in the low temperature limit, while for a point node with linear dispersion, a $T^2$ temperature dependence of $\lambda$ is expected. Our observed linear dependence of $\lambda_3$ is evidence for quadratic point nodes. For a line node with linear dispersion, a linear temperature dependence in $\lambda$ is predicted. And for an antinode along the equator, power-law temperature dependence should not occur. Our observed linear dependence of $\lambda_1$ is evidence for a line node at the equator. Our conclusions of quadratic point nodes and an equatorial line node in the B-phase gap support $E_{2u}$ pairing symmetry.

In summary, we have observed linear temperature dependence for both $\lambda_1$ and $\lambda_3$ at low temperatures, providing strong evidence for an equatorial line node and quadratic polar point nodes in the energy gap of UPt$_3$. Our results support the assignment of $E_{2u}$ pairing symmetry and are consistent with the conclusions from some other recent experiments. However, there are still a number of unresolved questions concerning the nature of the order parameter in UPt$_3$ including the importance of spin-orbit interaction and the spin structure of the order parameter as well as a direct observation of broken time reversal symmetry in the superconducting B-phase.
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