Small Angle Neutron Scattering Studies of the Vortex Lattice in the UPt_3 Mixed State: Direct Structural Evidence for the $B \rightarrow C$ Transition

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Small angle neutron scattering studies of the flux line lattice (FLL) in UPt_3 for fields $\mathbf{H} \perp \mathbf{c}$ provide direct microscopic evidence for the 5 kOe $B \rightarrow C$ transition. We find a pronounced maximum in the longitudinal correlation length of the FLL at the transition and an abrupt change in the field dependence of the scattered intensity which can be interpreted as a 15% decrease in the coherence length and a 9% increase in the penetration depth, consistent with discontinuities in the critical fields. Finally, in the low field phase, the FLL distortion evolves roughly linearly with field, while in the high field phase it appears to be less field dependent. [S0031-9007(97)02976-1]

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The heavy fermion metal UPt_3 remains the cleanest and most unambiguous example of a non-s-wave superconductor [1]. One consequence of this non-s-wave pairing is the extraordinary and complex phase diagram [2] for the mixed state, shown schematically in the inset to Fig. 3. For s-wave pairing, there can be only one superconducting state. However, in UPt_3, there are field and pressure driven transitions between different superconducting phases [3] as in superfluid ^{3}He. While the complex phase diagram can be reproduced within the framework of a multicomponent order parameter model [4], the theories have many parameters, and detailed microscopic verification has been elusive. Indeed, other competing scenarios have been introduced to explain various aspects of the phase diagram. These include transitions between single and doubly quantized vortices [5], stearic effects [6] due to noncylindrical vortices, and extrinsic effects [7] due to small scale phase separation into distinct superconducting compounds with different values of T_c and H_{c2}.

In this Letter, we report on a detailed small angle neutron scattering (SANS) study of the mixed state flux line lattice (FLL) in UPt_3 as a function of applied magnetic field at $T = 50$ mK. SANS studies of the FLL are a unique, powerful probe of the microscopics of superconductivity. From them, one can extract the field and temperature dependence of the coherence length ξ, penetration depth λ, three FLL correlation lengths, the value of the flux quantum, and the structure of the unit cell of the FLL. Our data show a number of remarkable effects in the FLL at the 5 kOe transition, which separates the low field B phase from the high field C phase. In particular, we find a pronounced maximum in the longitudinal FLL correlation length, changes in both ξ and λ, and a change in the field dependence of the lattice distortion. These measurements are compelling evidence that there exists a change in the microscopic superconducting state in UPt_3 at the $B \rightarrow C$ transition. In the accompanying paper [8] it is shown how these results are consistent with a two-component order parameter model.

The UPt_3 crystal used here has been thoroughly characterized in several previous experiments [9]. It is a right circular cylinder, 5 mm in diameter and 30 mm long, with the **c** axis parallel to the cylinder axis. The 1700 Å mean free path for electronic conduction along the c axis and the split, zero field superconducting transition in the specific heat are indicatative of a high quality sample. The experiments were performed on the SANS beam line in the cold neutron guide hall of the Risø DR3 reactor using an area detector 6 m from the sample. Both the magnetic field and the neutron beam were parallel to the **a** axis. All the SANS data shown here were taken at 50 mK following a zero field cooled process. Field cooled data were found to be similar. The data in this experiment consist of rocking curves together with zero field backgrounds. Care was taken to keep the experimental resolution function field independent. The total rocking curve width is $\sigma^2 = \sigma_i^2 + x^2\sigma_x^2 + y^2\sigma_y^2 + (\theta\Delta\lambda_n/\lambda_n)^2$, where σ_i is the intrinsic width of the FLL and the other contributions are due to beam divergences in the x and the y direction and the wavelength distribution $\Delta\lambda_n/\lambda_n$. The beam divergences were set by fixed pinhole collimation at 6 m. In order to keep the instrumental contributions to σ^2 constant as the applied magnetic field was changed, the neutron wavelength λ_n was varied from 8.5 Å at low fields to 6.4 Å at high fields to keep the scattering angle 2θ fixed, and the beam divergence contributions nearly fixed at $x\sigma_x \sim 0.16°$ and $y\sigma_y \sim 0.12°$. A fixed $\Delta\lambda_n/\lambda_n = 0.36$ gave a constant $\theta\Delta\lambda_n/\lambda_n \sim 0.12°$.

A significant technical improvement resulted from the use of (0001) single crystal sapphire windows in the thermal shields of the dilution refrigerator, reducing the background scattering by a factor of 50. The detailed changes in the FLL unveiled in this experiment were previously [10] obscured by this background. The coarse features of the data are unchanged. The residual background was highly anisotropic [11] with significantly greater intensity along the $\mathbf{c}^*$ direction. This background and the decreased scattering wave vector lead to reduced S/N for the two FLL peaks along $\mathbf{c}^*$. Because of this, we have assumed flux quantization [10] as previously determined and used the remaining four bright, first order peaks to determine the FLL unit cell. The data shown here required one month of reactor time to collect. It will not be possible to significantly improve on this experiment with the present generation of reactor-based sources.

Three correlation lengths [12] can be extracted from a SANS experiment on a FLL. Two of these lengths, $\xi^{\mathbf{r}\|\mathbf{G}}$ and $\xi^{\mathbf{r}\perp\mathbf{G}}$, measure correlations perpendicular to the flux lines in directions parallel and perpendicular to the reciprocal lattice vector $\mathbf{G}$. In this experiment, these two lengths are resolution limited. The third length, ξ_L, measures correlations along the flux lines and is extracted from the FWHM of the rocking curve widths using $\xi_L = 2/\tau\sigma_i$, where τ is the scattering vector and σ_i is the intrinsic FLL width. Raw data from sample rocking curves reveal an abrupt broadening for $H > 4$ kOe as shown in Fig. 1. The intensity data in Fig. 1 are total counts, after subtraction of zero field data, in a 7×7 box ($\sim 35 \times 35$ mm) on the area detector, versus distance from the Bragg condition q_0, as determined from the center of Lorentzian fits. Figure 2(a) shows the inferred values of ξ_L from all rocking curves as a function of field. In the region below 4 kOe, this length shows a gradual increase as the field increases. This effect is due to the increasing dominance of interactions over disorder as the field and, hence, vortex density increases [13]. An unusual and unexpected collapse of ξ_L occurs at $H = 4$ kOe. Collapse of the FLL correlation lengths just below a phase transition is familiar from FLL melting [14] and the "peak effect" [12] below H_{c2}. In those cases, the reductions in $\xi^{\mathbf{r}\|\mathbf{G}}$ and $\xi^{\mathbf{r}\perp\mathbf{G}}$ are associated with a soft mode of the FLL. The situation in UPt_3 is less readily apparent, although the $B \rightarrow C$ transition does coincide with a small change in the amplitude of the staggered magnetization [9], which has been shown [15] to couple to ξ_L in the antiferromagnetic superconductor $ErNi_2B_2C$.

If a phase transition due to a complex superconducting order parameter was responsible for the feature at 5 kOe, it would be expected to induce changes in λ and ξ. The intensity of the FLL Bragg reflections can be used to measure these microscopic lengths. The first order Ginzburg-Landau correction [16] to the London form factor gives a reflectivity $R = (2\pi\gamma^2\lambda_n^2 t/16\phi_0{}^2\tau)H_1^2$ with sample thickness t and form factor $H_1 = (\phi_0/\lambda^2)(\sqrt{3/8\pi^2})\exp(-4\pi^2 B\xi^2/\sqrt{3\phi_0})$. A semilog plot of the reflectivity versus field should be linear with a slope determined by ξ and an intercept related to λ. Shown in Fig. 2(b) is the average intensity of the four bright first order reflections as a function of applied field. There is a clear break in slope at 5 kOe, which we have used to define the location of the $B \rightarrow C$ transition for our sample, as this transition is known to be sample dependent. Averaging all the data gives $\xi \sim 110$ Å and $\lambda \sim 6000$ Å as in previous studies [10,17]. The break in slope implies a change in ξ at the transition with $\xi^2_{\mathrm{high}} = 0.71\xi^2_{\mathrm{low}}$. This change in ξ is consistent [18] with the change in slope of the upper critical field $\partial H_{c2}/\partial T = \phi_0/(2\pi\xi_0^2)$. The change in the intercepts similarly gives $\lambda^2_{\mathrm{high}} = 1.19\lambda^2_{\mathrm{low}}$. Changes in the fundamental length scales, λ and ξ, of a superconductor are convincing evidence for a microscopic, thermodynamic transition. The changes in λ and ξ imply that the thermodynamic critical field $H_c \approx 350$ G increases with $\Delta H_c = 30$ G at the $B \rightarrow C$ transition. The corresponding energy change of 125 erg/cm^3 is similar to the 100 erg/cm^3 as measured by studies of the specific heat [19]. The Clausius-Clapeyron relations can be used to estimate the change in the internal field $\Delta B = -(\Delta H_c^2/2T)(dH/dT) \approx 1$ G, where dH/dT is the slope of the $B \rightarrow C$ transition line. This change was obscured by critical currents in global magnetization data [20], but should be observable with a local probe [21].

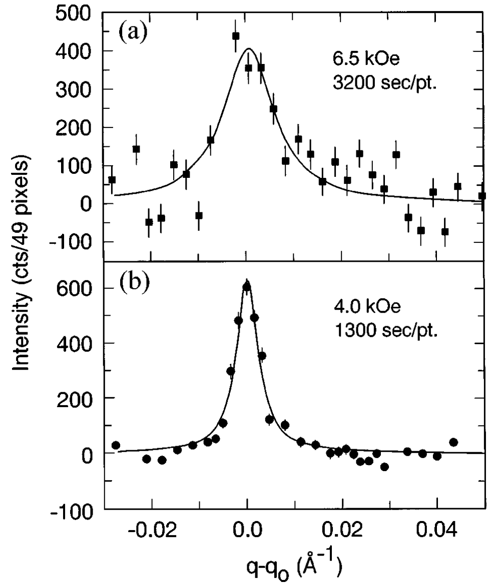

FIG. 1. Shown are the raw data from rocking curves for the FLL at $T = 50$ mk for fields of 6.5 kOe (a) and 4.0 kOe (b), straddling the $B \rightarrow C$ transition. Zero field data were subtracted as background. The figure shows total counts in a 7×7 box on the detector. The scattered wave vector is shown as $q - q_0$, where q_0 is the Bragg condition, determined from the fitted center of a Lorentzian line shape. Because of the rapid decrease in reflectivity with field, the 4 kOe data were taken with $\sim$1300 seconds per data point (sec/pt), whereas the 6.5 kOe data required $\sim$3200 sec/pt. Despite reduced statistics at the higher field, the dramatic broadening is readily apparent.

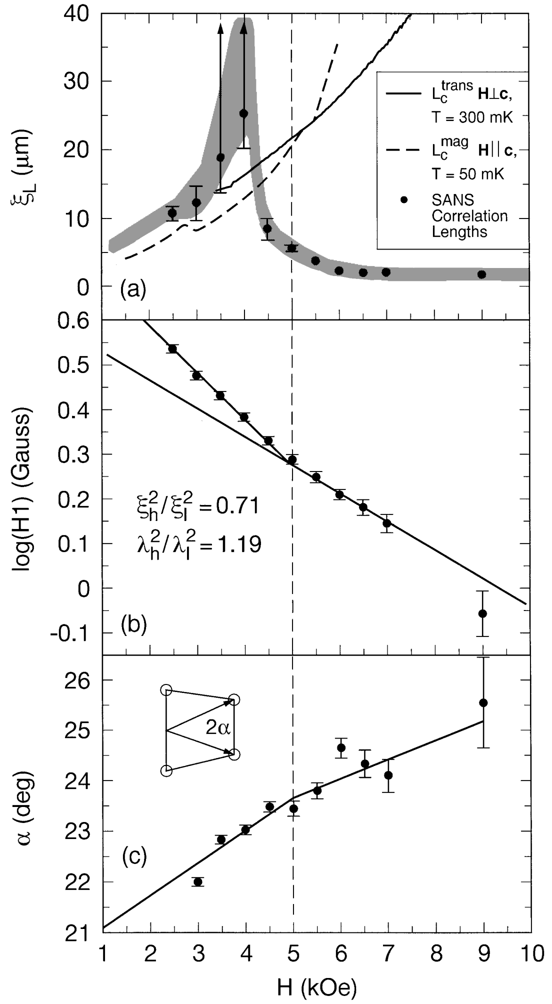

FIG. 2. (a) Shown is the longitudinal correlation length as extracted from the SANS data (circles) and both transport (solid line) and magnetization (dashed line). (b) Shown is a plot of the log of the form factor as a function of field. The break in slope at 5 kOe implies a change in both the coherence length and the penetration depth. (c) Shown is the field dependence of the lattice distortion. In the B phase the distortion is field dependent and in the C phase, less so. The inset defines the angle α. The dashed line across all panels defines the $B \rightarrow C$ transition.

The field dependent distortions of the FLL are an important signature [8,22] of a multicomponent order parameter. To study these, we have done the following self-consistent analysis. We have used the intensity data to define the location of the $B \rightarrow C$ transition for our sample and then done a simple linear least squares fit to the data in both the B and C phases. At low fields, we find the distorted hexagonal lattice sketched in the inset of Fig. 2(c). For a simple hexagonal lattice the angle α (defined in the inset) is 30°. Instead, we find that α starts out close to 20° and grows linearly with a slope of $\sim 0.75 \pm 0.11°$/kOe in the B phase. Within anisotropic Ginzburg-Landau theory [23], the distortion is given by $\tan^2 \alpha = m_3/m_1$, where m_1 and m_3 are the reduced effective masses $\perp c$ and $\| c$, respectively, normalized to $m_1^2 m_3 = 1$. The low field extrapolation $\alpha = 20°$ suggests that $m_1/m_3 \sim 2.5$, while the measured [24] normal state values are 1.5–1.8. The discrepancy is presumably due to gap anisotropy. However, the main point is that the observed field dependence in the B phase must arise from a complex superconducting state. The second observation which cannot by explained within a conventional picture is that, in the C phase, the opening angle appears to change less rapidly with field. In the high field phase, the slope is found to be $\sim 0.37 \pm 0.28°$/kOe, roughly half of the value in the B phase. The implications of this field dependence have been discussed elsewhere [22].

Within the Larkin-Ovchinnikov (LO) model of weak, random pinning, the longitudinal correlation length is related [25] to the critical current by $L_c = A[(c\xi\phi_0)/(\pi^2 a_0^2 J_c 6\sqrt{12})]^{1/2}$. Studies [13] in $NbSe_2$ have verified this expression with $A \sim 1$. To examine this in UPt_3 we have extracted critical currents from two different measurements—transport and magnetization. The transport critical current [26] was measured using a $1.0 \times 0.5 \times 4$ mm^3 bar cut from a nominally identical boule. To minimize the self-heating effects in the flux flow regime, data were taken with the sample immersed in ^{3}He at 300 mK. The data were taken by regulating the dc current to keep $\partial V/\partial I$ fixed, equivalent to a 15 nV/cm criterion for the critical current, as the field was swept. These data are shown in Fig. 3. Somewhat surprisingly, there is no evidence for a feature at 5 kOe in the critical current. One might naively expect that the critical current would change at the $B \rightarrow C$ transition due either to changes in the microscopic lengths, λ and ξ, or the pinning. Over much of the field range, we find that $J_c B^2$ is roughly constant. However, this is consistent with a simple Anderson model [27] of collective pinning, where $J_c B^2 = \phi_0(cp/8\pi l)(H_c^2/\lambda^2) \sim \xi^{-2}\lambda^{-4}$, where l is the average distance between defects, H_c the critical field, and p relates the free energy to $H_c^2/8\pi$. Within this approximation, the observed changes in ξ and λ should

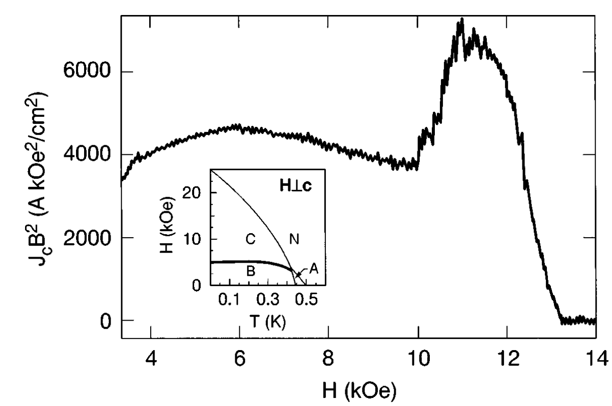

FIG. 3. Shown is the transport critical current as a function of applied field at $T = 300$ mK. The data are plotted as $J_c B^2$ versus applied field following the Anderson model of collective pinning. The broad maximum at 11 kOe is the peak effect for this sample. The inset shows the schematic phase diagram for this field orientation.

cancel, leading to no change in J_c at 5 kOe as we find in the critical current data. The measured J_c implies $l/p = 0.7\ \mu\text{m} \sim \lambda$, a reasonable value. The smooth variations shown are due to flux flow heating effects at high currents. Between 10 and 12 kOe, there is a broad peak effect in the critical current. This is associated with the complete disordering of the FLL close to H_{c2}, where the shear modulus of the lattice softens and the FLL can disorder to accommodate more pinning and the critical current goes through a peak [28]. This scenario is supported by the 9 kOe SANS data, where we find the lattice to be quite disordered with $\xi_L \sim a_0$.

In order to also measure the magnetization of the sample, we have built a custom field gradient magnetometer using a micromachined silicon cantilever whose deflection was measured capacitively [29]. We measured M-H loops at a fixed temperature, 50 mK. The known sample geometry and the critical state model [30] were used to calculate the critical current from M-H loops. Because of the geometry of our sample and the magnetometer, we were restricted to $\mathbf{H} \parallel \mathbf{c}$. However, our transport measurements showed that the anisotropy in J_c is quite small, giving us confidence in comparing these data to the $\mathbf{H} \perp \mathbf{c}$.

Assuming the validity of the LO model, we can calculate the longitudinal correlation lengths from both measured values of the critical currents. These are shown as the dashed and solid lines in Fig. 2(a). In the B phase, the correlation lengths extracted from both SANS and critical currents agree reasonably well. However, in the C phase, the LO model cannot account for the small values of ξ_L as observed in the SANS data. It is unusual to see a disordering of the flux lattice, as measured by ξ_L, with the critical current unchanged. We believe that the drop in ξ_L seen in the SANS data is not a simple pinning effect, but is perhaps due to the thermodynamics of the phase transition at 5 kOe. If the $B \rightarrow C$ transition was first order, this could produce a regime of two-phase coexistence of the B and C phases near the transition. Therefore, the disordering could be driven by the thermodynamics of the transition. Alternatively, the kinetic barriers to transforming the vortex lattice symmetry from that in the B phase to the C phase at low temperatures could also lead to substantial disorder.

In conclusion, we have reported on a detailed SANS study of the FLL in the mixed state of UPt_3. At the $B \rightarrow C$ transition, there are changes in the microscopic length scales λ and ξ, the FLL abruptly disorders, and the field dependent distorted FLL in the B phase gives way to a less field dependent, more isotropic FLL in the C phase. These are the first observations which show how the vortex lattice is directly involved in the $B \rightarrow C$ transition and the first to show that the microscopic lengths change. These results cannot be explained within the framework of a conventional, s-wave superconductor. However, as discussed in the accompanying paper [8], a two-component order parameter model is consistent with our results.

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