Magnetic Order in the Different Superconducting States of UPt_3

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We use neutron diffraction to show that superconductivity affects the magnetic order in UPt_3. The different superconducting states identified in previous bulk measurements can be associated with different behaviors of the magnetic order parameter. The data suggest that the coupling between the multicomponent superconducting and magnetic order parameters leads to the variety of superconducting phases of UPt_3.

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UPt_3 continues to fascinate because of its unusual superconductivity,[1] simultaneous antiferromagnetic tendencies,[2] and large carrier masses.[3] Early experiments were concerned primarily with the superconducting properties in zero magnetic field, with results strongly indicating anisotropic *d*-wave pairing. More recently, attention has shifted to anomalies found within the superconducting phase for nonzero applied fields H by torsional-oscillator,[4] ultrasound,[5] and specific-heat[6] measurements. In the present paper, we describe neutron-diffraction data which show that these anomalies are best understood in the context of coupled superconducting and (static) magnetic order parameters. Specifically, we show that the $(1,\frac{1}{2},0)$ magnetic Bragg intensity decreases with decreasing temperature for $T \lesssim 0.4$ K and $H=0$. Even more interesting is that it can be made to increase at $T=0.1$ K by applying a field *perpendicular* to the basal planes and ordered moments of UPt_3. However, the intensity is T and H dependent only in the low-H, low-T superconducting state delineated by the most pronounced bulk anomalies.

Before presenting our results, we recall that UPt_3 is hexagonally close packed (space group $P6_3/mmc$) with two U atoms per unit cell. The locations of Bragg peaks are expressed in reciprocal-lattice units, where $a^* = b^* = 4\pi/(a\sqrt{3}) = 1.264$ Å and $c^* = 1.285$ Å. The magnetic order[2,7] in UPt_3 involves a doubling of the unit cell along an a^*-type direction in the basal planes, with the ordered moment also parallel to this direction. The ordered moment is small $[(0.02 \pm 0.01)\mu_B/\text{U atom}]$ but comparable to that for other superconducting heavy-fermion systems, such as URu_2Si_2[8] and $U_{1-x}Th_xBe_{13}$.[2,9] The Néel temperature is $T_N \approx 5$ K, which is close to where several transport and thermodynamic anomalies occur.[10] The $(\frac{1}{2},0,1)$ intensity was previously measured for $T \geq 0.25$ K: It rises in proportion to $T_N - T$ until it reaches a plateau below $T \approx 0.5$ K.

We performed our neutron scattering experiments using the triple-axis spectrometer TAS7 installed in the cold-neutron guide hall of the DR-3 reactor at Risø National Laboratory, Denmark. Both the monochromator and analyzer were pyrolitic graphite, set for the (002) reflection, while cooled Be filters eliminated higher-order contamination of the incident beam. Except where explicitly mentioned, no collimators were installed; we note, however, that the divergence of the beam incident on the monochromator was 20′ as given by the neutron

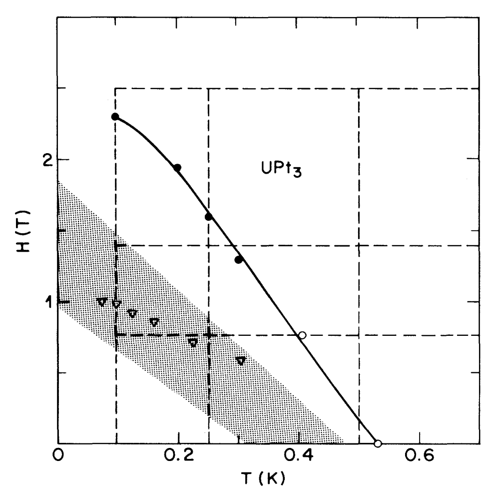

FIG. 1. Phase diagram for superconducting UPt_3. Open and closed circles represent $H_{c2}(T)$ found from constant-field and -temperature sweeps, respectively, of ac susceptibility measured *in situ* as described in text. Open triangles are locations of torsional-oscillator anomalies in Ref. 4. Shaded area represents boundary between states where antiferromagnetic intensity varies with field and temperature and where it is H and T independent. Dashed lines are trajectories followed to accumulate diffraction data.

guide. The neutron energy was 5 meV. We mounted four UPt_3 crystals, each a cylinder of roughly 4 cm in length (c‖ growth direction) and 6 mm in diameter, so that their crystallographic axes coincided to within better than 1°, on a copper sample holder. A fifth crystal mounted on the holder was the core of a mutual inductance coil used for monitoring the ac susceptibility during the neutron scattering experiment. The assembly was attached to the cold finger of an Oxford Instruments dilution refrigerator such that the c axes of the crystals were parallel to the (vertical) field produced by a split-coil superconducting magnet. The entire refrigerator was installed on the sample table of the spectrometer whose (horizontal) scattering plane coincided with the $(hk0)$ zone of UPt_3.

The UPt_3 crystals were the same as those used in the earlier cold-neutron scattering study; growth procedures and experiments relating to their quality and purity are described elsewhere.[7] Figure 1 shows the superconducting phase diagram for these samples. The solid and open circles represent $H_{c2}(T)$ found from the ac susceptibility χ_{ac} measured *in situ* for constant fields and temperatures, respectively. The phase transition was identified as occurring where χ_{ac} reached 10% of its diamagnetic limit. For $H=0$, the corresponding transition temperature is $T_c = 0.53$ K.

Figure 2 shows the $(1,\frac{1}{2},0)$ Bragg intensities and ac susceptibilities measured along the constant-H and constant-T trajectories indicated by the dashed lines in Fig. 1. At all fields for $T > T_c$, the intensity increases monotonically with decreasing T until $T \approx T_c$, in agreement with the measurements of the $(\frac{1}{2},0,1)$ reflection.[7] The most important new result is that at the lowest fields and temperatures, the $(1,\frac{1}{2},0)$ intensity is actually reduced (by ~5%) with respect to its value at T_c. Another remarkable result is that fields larger than 1 T restore the full Bragg intensity (observed at T_c) even at the lowest temperature (0.1 K). Finally, the Bragg intensity is independent of field at T_c. This observation, together with the knowledge that the ordered moments are within the basal plane and that fact that the magnetic susceptibility is lowest when measured parallel to **c**,[11] the direction of the magnetic field in this experiment, clearly implies that the anomalous behavior of the magnetic intensity is due to the superconductivity of UPt_3. Interestingly, however, the Bragg intensity becomes independent of field and temperature when according to the ac susceptibility data, the samples are still well within the superconducting regime. While occurring far from the boundary defined by $H_{c2}(T)$, the Bragg intensity saturates near anomalies found in the superconducting state by torsional-oscillator and ultrasound-attenuation measurements; the arrows in Fig. 2 represent the reported positions of torsional-oscillator anomalies.[4]

To obtain a quantitative summary of how the magnetic intensity behaves in the H-T plane, we have fitted our data for $T < 0.55$ K by the form

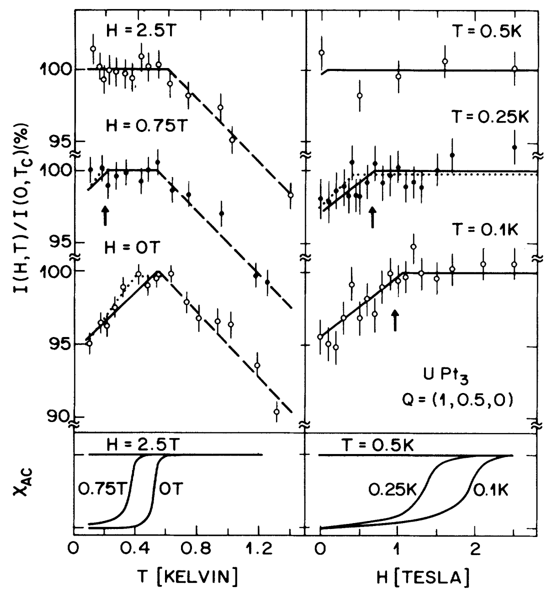

FIG. 2. Normalized and background-corrected temperature-dependent (left-hand column) and field-dependent (right-hand column) $(1,\frac{1}{2},0)$ magnetic Bragg intensities (upper frames) and ac susceptibilities (lower frames). Temperature sweep for $H=1.4$ T is not shown because it is indistinguishable from that for $H=2.5$ T. Solid and dotted lines are derived from fits described in text. At $H=0$ and $T=0.5$ K, the observed peak intensity is 245 counts/min on a background of 48 counts/min.

$$I(H,T) = \begin{cases} I_0 \text{ for } H > H'(T), \\ I_0 - c[H'(T) - H] \text{ for } H \le H'(T), \end{cases} \quad (1)$$

where $H'(T) = H_0'(1 - T/T_c')$.

We obtain the best results for $T_c' = 0.39$ K and $H_0' = 1.4$ T. The statistically weighted χ^2 increases by 10% from its minimum if T_c' is raised (lowered) by 0.09 (0.08) K, or if H_0' is raised (lowered) by 0.5 (0.4) T. The shaded region in Fig. 1 separates the regimes where the Bragg intensity is saturated and where it is still H and T dependent. The shading is used to indicate the uncertainty, established by the error analysis just described, in the location of the boundary between the two regimes. All of the "phase boundaries" reported previously for superconducting UPt_3 fall within the shaded region. This is true not only for $H \gg H_c$ (see, e.g., open triangles which mark positions of maxima in dissipation measured by the use of a torsional oscillator[4]), but also for $H=0$, where the data are slightly more consistent with Eq. (1) when $T_c' \neq T_c$ (dotted lines in Fig. 2) rather than when $T_c' = T_c$ (solid lines in Fig. 2), in agreement with specific-heat data indicating two superconducting transitions.[6] Thermodynamic measurements on crystals of the type used here are underway and will help to de-

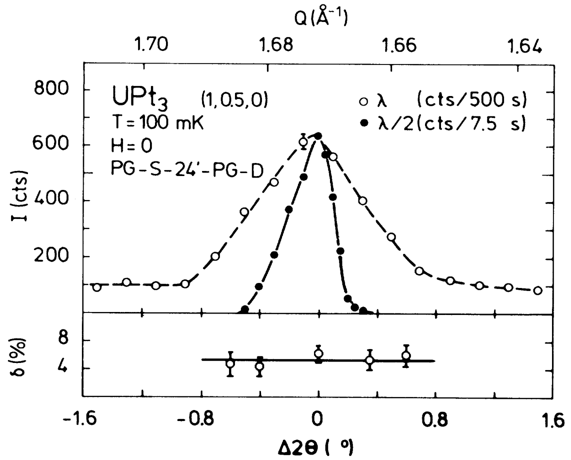

FIG. 3. Upper frame shows longitudinal (θ–2θ) scan through magnetic $(1,\frac{1}{2},0)$ and nuclear (2,1,0) reflections observed with first- and second-order neutrons, respectively, using the same spectrometer configuration, which differs from that of Fig. 2 by the addition of a 24′ collimator after the monochromator. $\Delta 2\theta$ is the deviation in scattering angle from peak position; Q is the momentum transfer. Lower frame represents the Q dependence of the ratio of magnetic intensities I_N and I_S at 540 mK and 100 mK, respectively, with $H=0$: $\delta=(I_N-I_S)/(I_S-I_B)$, where I_B =background signal = 12 counts per minute.

cide where bulk superconductivity sets in (χ_{ac} generally gives a higher value for T_c) and whether our samples undergo two transitions in zero field.

Figure 2 shows intensities measured for constant scattering angle. To determine whether the new effects are due to a change in the overall amplitude, or to a broadening or a shift of the Bragg peak, we have measured the shape of the magnetic scattering peak with higher momentum resolution. In the upper frame of Fig. 3, the magnetic peak is compared to the (2,1,0) nuclear Bragg profile determined using $\lambda/2$ neutrons. On account of the wavelength-dependent divergence of the neutrons in the guide, the $\lambda/2$ profile does not give the resolution width for λ directly. However, it can be used to make a rough estimate (0.016 Å^{-1} full width at half maximum) which implies that as for the $(\frac{1}{2},0,1)$ magnetic reflection,[7] the $(1,\frac{1}{2},0)$ peak is not resolution limited; its width corresponds to a magnetic correlation length of order 150 Å. The lower frame of Fig. 3 shows the effect of reducing the temperature to 0.1 K. We have plotted the fractional increase δ of background-corrected intensities (the background was measured to be temperature independent) from 0.1 to 0.5 K, respectively. To eliminate the influence of small irreproducibilities in the spectrometer setting, the data were taken at both temperatures for each point in the scan before moving to the next point. The constant δ seen in Fig. 3 implies that the line shape and position of the $(1,\frac{1}{2},0)$ magnetic peak remain unchanged between the superconducting and normal states of UPt_3. In other words, the intensity changes associated with superconductivity are due to changes in the overall amplitude of the elastic magnetic scattering. This result excludes scenarios where magnetic order is nucleated at discrete defects, and the finite range associated with the penetration of the order into the otherwise unperturbed host is modified by the superconductivity.

Largely because of ultrasound-absorption measurements,[1] UPt_3 has been classified as an anisotropic superconductor where the gap function vanishes at lines on the Fermi surface. The unconventional pairing is commonly thought[12] to be d wave, and mediated by the antiferromagnetic fluctuations observed by inelastic neutron scattering.[2] The corresponding superconducting order parameter ψ can be represented by vectors $\xi_1\hat{\mathbf{x}}-\xi_2\hat{\mathbf{y}}$, where ξ_1 and ξ_2 are complex coefficients, and $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are translations in the basal planes of UPt_3. Theorists have proposed several descriptions of the superconducting phases of UPt_3: (i) The two different superconducting states are characterized by either $\xi_1=i\xi_2$ or $\xi_1\xi_2=0$ (i.e., $\psi=\hat{\mathbf{x}}$ or $\hat{\mathbf{y}}$).[13] (ii) The microscopic superconducting order parameter is the same for both states, which differ only in the nature of the associated vortex cores.[5] (iii) The two states are $\xi_1=i\xi_2$ (for low T and H) and $\xi_1-ir\xi_2$, where r is a nonzero real number.[14]

Specific-heat and ultrasound measurements rule out proposal (i), as described elsewhere.[5,6] Since the magnetic order parameter can only couple to the microscopic superconducting order parameter, the transition between different states as described in (ii) would be invisible in the present neutron scattering experiments. Furthermore, the zero-field transition would not be split as shown by specific-heat measurements[6] and suggested by our data. Thus, (ii) is also ruled out. Proposal (iii), which incorporates the reduced space-group symmetry associated with the magnetic order, does allow for the splitting of the zero-field transition. However, the vector character of the magnetic order parameter and the related broken time-reversal invariance are not taken into account.

In view of the shortcomings of existing theory, not the least of which is the absence of predictions for the field and temperature dependence of the magnetic order parameter, we describe here the framework[15] within which our data might eventually be understood. We begin by noting that the coupling of the vectors representing superconducting (ψ) and magnetic (ϕ) order parameters involves both their amplitudes, as for ordinary s-wave superconductors, and their relative orientation, a quantity which of course does not exist for ordinary superconductors. Couplings involving amplitudes arise from the usual pair-breaking mechanisms which yield competition between superconductivity and magnetism. Taking only such couplings into account, the interpretation of our data would be that there are two different superconducting order parameters, with pair wave functions such that the coupling would be greatly reduced or even zero in the high-field, high-temperature state. Different interpretations are possible when we consider coupling terms involving the relative orientations of ψ and ϕ. These terms appear because of the inevitable and not necessarily pair-breaking interactions between the angular momentum of the Cooper pairs and the ordered moment. As their name implies, they lead to the possibility of orientation of ψ by ϕ when $|\psi|$ is small, which it is for H near $H_{c2}(T)$. On the other hand, when $|\psi|$ is large, ψ can actually *reorient* ϕ. Thus, ψ acts much as an *in-plane* magnetic field on UPt_3: As long as its amplitude (strength) is below a certain critical value (spin-flop field) due to the basal-plane anisotropy, ϕ will maintain its orientation, while for larger $|\psi|$, ϕ will be rotated. In the latter situation, the magnetic Bragg intensities will change because of the dipole selection rules. Measurements of more than a single Bragg peak are needed to determine the relative importance of amplitude and orientation effects. for example, since the rotation of the moments will occur within the basal planes (the in-plane anisotropy energies are small compared to the out-of-plane energies), it would be essentially undetectable at the $(\frac{1}{2},0,1)$ Bragg point studied previously.[7] If the result of Ref. 7, namely that the $(\frac{1}{2},0,1)$ intensity is T independent for $T < T_c$, were to hold to the lower temperatures of the current experiment, we would have strong evidence that the intensity changes observed at $(1,\frac{1}{2},0)$ are due primarily to a reorientation of ϕ.

In summary, we have shown that there are at least two microscopically distinct states of superconducting UPt_3, as characterized by the behavior of the magnetic order parameters. The fact that superconductivity influences the static magnetic scattering and that it does so in a way which changes its overall amplitude but not its shape demonstrates that antiferromagnetism and superconductivity coexist and interact at the microscopic level in our samples of UPt_3.

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