Influence of a magnetic field on the antiferromagnetic order in UPt_3

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A neutron-diffraction experiment was performed to investigate the effect of a magnetic field on the antiferromagnetic order in the heavy-fermion superconductor UPt_3. Our results show that a field in the basal plane of up to 3.2 T, higher than $H_{c2}(0)$, has no effect: it can neither select a domain nor rotate the moment. This has a direct impact on current theories for the superconducting phase diagram based on a coupling to the magnetic order. [S0163-1829(96)50238-9]

Most of the heavy-fermion superconductors order antiferromagnetically before the onset of superconductivity, with $T_N \simeq 10\ T_c$. The possible relation between the phenomena is one of the central issues in the field. However, no two compounds have exactly the same magnetic behavior. While both UPt_3 (Ref. 1) and URu_2Si_2 (Ref. 2) show an extremely small ordered moment, of order $0.01\mu_B$/U atom, it is as large as $0.85\mu_B$/U atom in UPd_2Al_3.[3] The specific-heat anomaly at T_N is large in URu_2Si_2 (Ref. 4) yet absent in UPt_3.[5] The ordered structure breaks the hexagonal symmetry in UPt_3 and UPd_2Al_3, with the moments aligned in the basal plane, while the tetragonal symmetry of URu_2Si_2 is preserved. The magnetic order and fluctuations are unaffected by the onset of superconductivity in UPd_2Al_3[6], while a slight decrease in the amplitude of the moment is observed in UPt_3 (Refs. 7 and 8) and a saturation of the moment in URu_2Si_2.[9]

The coexistence of magnetism and superconductivity in these compounds has been viewed as evidence for an unconventional pairing mechanism. Unlike the Chevrel phases, where the electrons responsible for the superconductivity are distinct from those responsible for the magnetism, it appears that in the case of UPt_3, in particular, the same electrons participate in both phenomena. Indeed, in this material a division of labor is implausible in view of the presence of the f electrons at the Fermi level and the fairly uniform effective mass around the Fermi surface.[10,11]

The unconventional nature of the superconducting state in UPt_3 is most strikingly manifest in the existence of several superconducting phases.[12] The magnetic field (H)-pressure (P)-temperature (T) phase diagram shows two distinct transitions at $T_c^+=0.5$ K and $T_c^-=0.44$ K for $H=P=0$.[13] Application of a magnetic field in the basal plane ($\vec{H}\perp\hat{c}$) brings the two transitions together at a tetracritical point,[14] which shows up clearly on the $H_{c2}(T)$ line as a kink at a field H^* of about 0.4 T.[15] Hydrostatic pressure also causes T_c^+ and T_c^- to merge, at a critical pressure of about 3.7 kbar.[16] A complete theory for the phase diagram of UPt_3 has been one of the major pursuits in the field over the past five years. Two main scenarios are currently debated: in the first type, the proximity of T_c^+ and T_c^- is considered accidental and the two zero-field phases are attributed to different representations of the order parameter.[17] In the second type, the double transition is viewed as a splitting resulting from the lifting of the degeneracy of a state (within a single representation for the order parameter) by some symmetry-breaking field.[18,19] An obvious choice for such a field is the antiferromagnetic order, with its moment and propagation vector both lying in the basal plane ($\vec{M}_s \| \vec{q} \| \hat{a}^*$). The moment configuration has been described so far in terms of a single-$\vec{q}$ structure with a given sample in general possessing three equivalent domains.[7,8,20] However, the existing data is also compatible with a triple-$\vec{q}$ structure.

In their neutron study under pressure, Hayden *et al.*[20] found that the antiferromagnetic moment of UPt_3 is fully suppressed by applying 3 to 4 kbar, which is also the critical pressure for the merging of T_c^+ and T_c^-. The parallel disappearance of magnetism and phase multiplicity under pressure is strong evidence in favor of the coupling scenarios (the second type), with the antiferromagnetic order acting as the symmetry-breaking field. Within the coupling scenarios, the kink in the H_{c2} curve is basically the result of a sudden reorientation of the (vector) order parameter $\vec{\eta}$ in the basal plane.[18] Both the moment $\vec{M}_s$ and the field $\vec{H}$ will couple to $\vec{\eta}$, each trying to align it in the minimum energy direction. Without loss of generality, let us consider the case of $\vec{M}_s \perp \vec{H}$, with both couplings to $\vec{\eta}$ favoring *parallel* alignment. At low fields, the coupling to the magnetic order dominates and $\vec{M}_s$ determines the orientation of $\vec{\eta}$. Then, when the field is increased to the point where its coupling dominates, a reorientation of $\vec{\eta}$ occurs, causing a kink in $H_{c2}(T)$. Of course, if the field direction is instead made parallel to $\vec{M}_s$, no kink is predicted, since there is no competition between the two couplings. As a result, within a single antiferromagnetic domain (assuming a single-$\vec{q}$ structure for

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the magnetic order) the upper critical field in the basal plane of UPt_3 is predicted to show a sharp kink only for one direction of the field (say $\vec{H}\|\hat{a}$), and no kink for the $\hat{a}^*$ direction 90° away.[18] Experimentally, however, a kink is observed at $H^* \approx 0.4$ T for any high-symmetry direction (0°, 90°, 120° relative to $\hat{a}$).[21] The theory can be reconciled with a ubiquitous kink by supposing that the moment is not fixed to the lattice but rather follows the field in such a way that $\vec{M}_s \perp \vec{H}$ for all field orientations in the basal plane. This is possible provided the in-plane magnetic anisotropy energy is negligible compared to the Zeeman energy acting on $\vec{M}_s$. Sauls[22] showed that a rotation of $\vec{M}_s$ in the basal plane is accompanied by a modulation of its amplitude M_s with 60° periodicity, which in turn causes $H_{c2}(\theta)$ to exhibit 60° oscillations, such as those observed recently in UPt_3.[23] The first goal of our experiment was to determine whether a magnetic field lower than 1 T can indeed cause the magnetic moment to rotate in the basal plane away from its zero-field configuration ($\vec{M}_s\|\vec{q}\|\hat{a}^*$) and remain perpendicular to $\vec{H}$.

If the magnetic ground state of UPt_3 has only one propagating vector (single-$\vec{q}$), as assumed until now by all authors,[1,7,8,20] then there should in general be three independent domains with $\vec{M}_s$ oriented at 120° with respect to each other. Agterberg and Walker[24] have recently considered the effect of having three possible domains on the H_{c2} curve of UPt_3 in the basal plane. They assume that $\vec{M}_s$ is fixed with respect to the crystal lattice (i.e., parallel to any one of the three a^* axes) but that only the most thermodynamically stable domain will be populated for any given field direction. Within the coupling scenario, the implications are fairly straightforward: the angle between $\vec{M}_s$ and $\vec{H}$ can only range over $\pm 30°$ and the domain selection by the field as it is rotated causes a 60° variation in $H_{c2}(T)$. The limited range of angles could perhaps explain why a straight H_{c2} curve is never observed. The second goal of our experiment was therefore to establish whether a magnetic field of less than 1 T can select a single domain.

We show that a magnetic field of up to 3.2 T in the basal plane—which is greater than $H_{c2}(0)$ and much greater than H^*—has no influence on the antiferromagnetic order: it can neither rotate the moments nor select a domain.

Our neutron-diffraction studies were performed with the DUALSPEC triple-axis spectrometer at the NRU reactor at Chalk River Laboratories with a pyrolytic graphite monochromator, analyzer and filter, and a neutron wavelength of 2.37 Å. The collimation was 0.6° between the monochromator and sample and 0.8° between sample and analyzer. The sample, used in previous neutron experiments,[20] was a high-quality single crystal of UPt_3 that exhibits two sharp successive superconducting transitions, a moment of $0.03\mu_B$/U atom and a Néel temperature of approximately 6 K. It was aligned with its hexagonal plane in the scattering plane of the spectrometer and mounted in a horizontal field cryostat that enabled a field of up to 3.2 T to be applied at any angle in the basal plane.

In a first measurement, the magnetic field was applied in the basal plane along the [1, 2, 0] direction, which is perpendicular to the a^* direction and to the wave vector of the $\vec{q}_1$

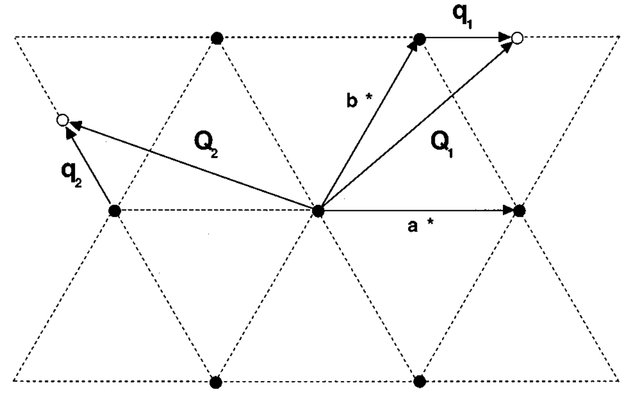

FIG. 1. Reciprocal space diffraction geometry for the two domains investigated here. The $\mathbf{q}_i$ and $\mathbf{Q}_i$ indicate the propagation and scattering vectors, respectively.

$= (\frac{1}{2}, 0, 0)$ domain. This should favor the $\vec{q}_1$ domain and remove the $\vec{q}_2 = (\bar{\frac{1}{2}}, \frac{1}{2}, 0)$ and $\vec{q}_3 = (0, \frac{1}{2}, 0)$ domains, each of which is at 30° to the applied field. The intensity of the $\vec{q}_1$ peak, observable at a scattering wave vector $\vec{Q}_1 = (\frac{1}{2}, 1, 0)$, which is at an angle to $\vec{M}_1\|\vec{q}_1$ in order to sense the moment (see Fig. 1), should then increase by a factor of 3 on application of a sufficiently strong field. Concomitantly, the intensities of the $\vec{q}_2$ domain at $\vec{Q}_2 = (\bar{\frac{3}{2}}, \frac{1}{2}, 0)$ and the $\vec{q}_3$ domain at $\vec{Q}_3 = (\bar{1}, \frac{3}{2}, 0)$ should vanish.

From scans such as those displayed in Fig. 2, in which the crystal angle ψ was rotated through the Bragg position at a fixed temperature of 1.8 K and a fixed field orientation, namely $\vec{H}\perp\vec{q}_1$, we find that the Bragg peaks corresponding to the three wave vectors persist up to a magnetic field of 3.2 T, as shown in Fig. 3. There is no significant increase in the population of what should be the most thermodynamically stable domain ($\vec{q}_1$). A slight increase of order 30% at 3 T is not inconsistent with the error bars in Fig. 3. This would then be compatible with a roughly equivalent decrease observed in the $\vec{q}_2$ intensity, and suggest that complete domain repopulation could be achieved at higher fields. However, as far as the superconducting phase diagram is concerned, it is important to stress that this anisotropy field is larger than $H_{c2}(0)$, so that the sample is multidomain in all superconducting phases.

In order to make $\vec{q}_2$ the least favored domain, we rotated the field by 30° to lie along the $\vec{q}_2$ direction. At 1.6 T, we again observed that both the $\vec{q}_1$ and $\vec{q}_2$ modulations remain present. Within the statistical error of 20%, the integrated intensity of the $\vec{q}_2$ modulation observed at a scattering vector $\vec{Q}_2 = (\bar{\frac{3}{2}}, \frac{1}{2}, 0)$ was unchanged between 0 and 1.6 T. For independent (and weakly pinned) domains the intensity would have vanished. A similar independence of field was observed for the $\vec{q}_1$ modulation seen at $\vec{Q}_1 = (\frac{1}{2}, 1, 0)$, where the peak should have grown by a factor of $\frac{3}{2}$.

This is in contrast with the behavior of UPd_2Al_3,[25] where a field of less than one T in the hexagonal basal plane perpendicular to $\vec{q} = (1,1,0)$ clearly enhances the population of

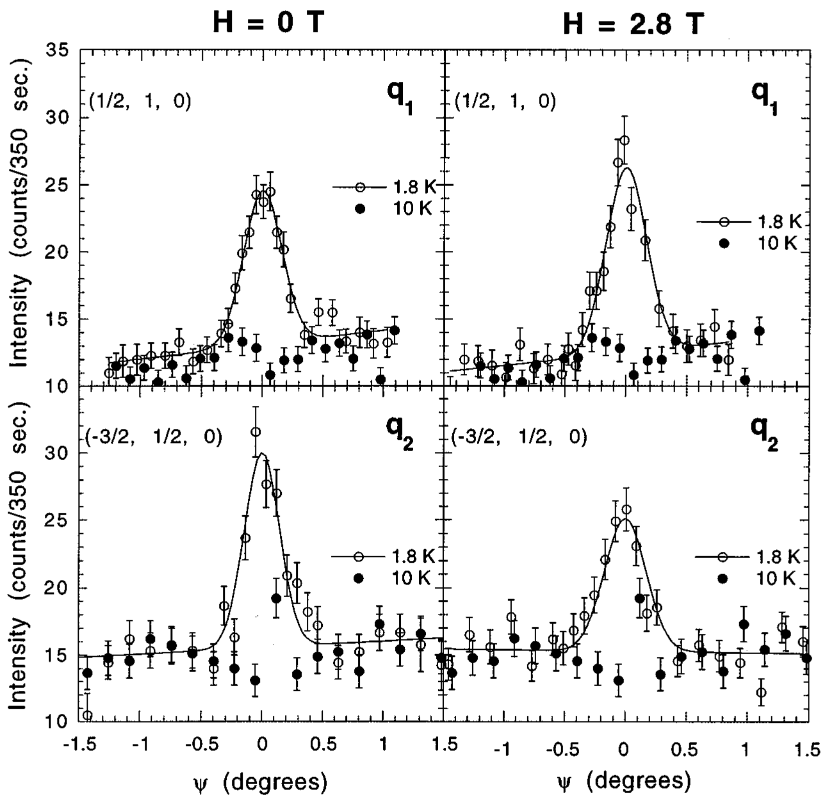

FIG. 2. Magnetic Bragg peaks at $\vec{q}_1$ and $\vec{q}_2$ for $H=0$ and 2.8 T, with $\vec{H}\perp\vec{q}_1$. Complete selection of a single domain by the 2.8 T field would eliminate the $\vec{q}_2$ Bragg peak and increase the intensity of the $\vec{q}_1$ peak by a factor of three.

that particular domain to the detriment of the other two. If a similar effect occurred in UPt_3, the relative intensities of the $\vec{q}_1$ and $\vec{q}_2$ domains would be expected to follow the solid lines shown in Fig. 3.

In UNi_2Al_3, where the moment is $0.12\mu_B$/U atom, intermediate between that of UPt_3 and that of UPd_2Al_3, the propagation vector (0.61,0,0.5) also has a component in the basal plane but it is incommensurate with the crystal lattice.[26] In this case, a field of 3 T is insufficient to produce a monodomain.[27]

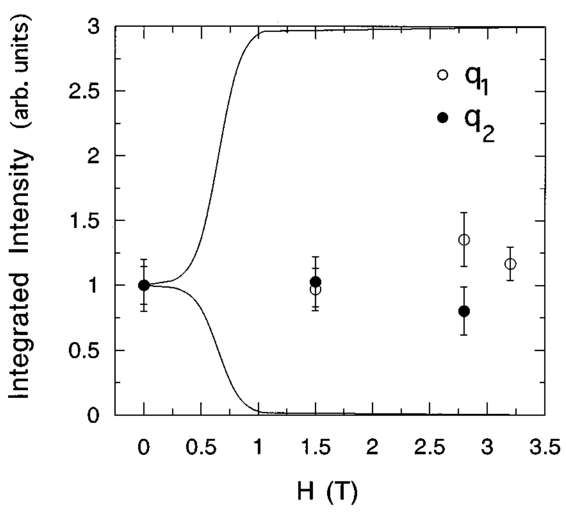

FIG. 3. Integrated intensity as a function of field for $\vec{q}_1$ (open circles) and $\vec{q}_2$ (solid circles) with $\vec{H}\perp\vec{q}_1$. The solid lines show the expected behavior for both magnetic domains for an anisotropy field of order 0.5 T [as observed in UPd_2Al_3 (Ref. 25)].

In zero-field-cooled (ZFC) experiments, such as those described above, it is possible that domains, having already formed, cannot attain the new thermodynamic equilibrium associated with the applied field. To check for this possibility, we slowly cooled the sample through its 6 K magnetic transition in a field of 3.2 T along the $(1,\bar{2},0)$ direction. All three wave-vector modulations were found to have condensed with the same intensity as for cooling in zero field. For the $\vec{q}_1$ modulation we can exclude at the 2σ level any increase in peak intensity beyond 30% relative to the ZFC intensities; field selection of one domain would have produced a threefold intensity increase. These results exclude the possibility that an energy barrier, arising from the reduced orthorhombic symmetry of single-$\vec{q}$ ordered state, might have prevented the attainment of an equilibrium domain configuration at low temperature. We therefore conclude that in UPt_3 the three modulations are present with roughly equal importance for all field strengths at which the superconducting state exists.

Even if all three wave vectors survive the application of a magnetic field, the moments themselves might still rotate away from being longitudinal $(\vec{M}_s \| \vec{q})$. To test this possibility, we monitored the scattering wave vector $\vec{Q} = (\bar{\frac{1}{2}},\frac{1}{2},0)$, where neutron diffraction senses the $\vec{q}_2$ spatial periodicity, but where, in the absence of a field, the scattering amplitude is zero because the moment is parallel to $\vec{Q}$. Moment canting in the field would then give a nonzero amplitude. Applying a field of 2.8 T along $(1,\bar{2},0)$—perpendicular to $\vec{q}_1$ and at

30° to $\vec{q}_2$—we observed no measurable growth in intensity above background. The statistics allow us to put an upper bound of 26° on any rotation at the σ confidence level (a realignment of the $\vec{M}_s$ moment of domain $\vec{q}_2$ by the field would have meant a 60° rotation). This shows that the moment does not follow the field as the latter is rotated in the basal plane, and this for field strengths much greater than H*=0.4 T. This suggests that $\vec{M}_s$ is strongly coupled to the crystal lattice, in agreement with the observation that $\vec{M}_s$ does not rotate upon entering the superconducting state at 0.5 K.[8]

Let us look more closely at the single-$\vec{q}$ assumption. Isaacs *et al.*[8] have shown that a collinear structure with three separate domains gives a diffraction pattern consistent with the observed structure factors. The question is: Why are all three domains equally favored upon cooling in a field of 3.2 T which is only perpendicular to one of the associated moments? For a collinear antiferromagnet, the fact that the transverse susceptibility is larger than the longitudinal susceptibility should lead to the selection of the domain perpendicular to the applied magnetic field, as is seen in UPd_2Al_3. A simple explanation for the ubiquitous presence of all three wave vectors is that the magnetic structure might be triple-$\vec{q}$. With a symmetric superposition of three equivalent modulations, the diffraction pattern would be the same as with three single-$\vec{q}$ domains. A magnetic field would have no effect at low fields; it would only produce a single-$\vec{q}$ domain sample when the Zeeman energy developing from distortion of the 3-$\vec{q}$ structure exceeded the binding energy of the 3-$\vec{q}$ state. Triple-$\vec{q}$ structures are known to occur in uranium compounds, such as USb (Ref. 28) and UPd_3,[29] and are characterized by an insensitivity to applied magnetic fields and uniaxial stress.[28] Now, it is far from obvious that such a magnetic order could break the hexagonal symmetry (in zero field), and even more so that a coupling to the superconducting order can lead to a split transition. Therefore, if such a structure is the correct one for UPt_3, a major reassessment of the coupling theories mentioned above is needed.

In conclusion, we have shown that basal plane magnetic fields of up to 3.2 T have no effect on the magnetic order in UPt_3, whether it be in rotating the moments or in selecting a domain with a single wave vector. Because the upper critical field of UPt_3 is less than 3.2 T, the absence of rotation makes it difficult to reconcile the fact that experimentally a kink in $H_{c2}(T)$ is observed at 0.4 T (Refs. 14, 15, 21, 23) for various field directions in the basal plane with the prediction of current theories[18,19,22] that it should only occur for one direction of $\vec{H}$ with respect to $\vec{M}_s$. In this respect, a calculation with *three* fixed domains would prove helpful. Our results also invalidate the respective assumptions (moment rotation and domain selection) underlying two recent explanations[22,30] for the slight 60° variation of H_{c2} in the basal plane.[23] Finally, there is a distinct possibility that the antiferromagnetic order in UPt_3 has a triple-$\vec{q}$ structure, as opposed to the single-$\vec{q}$ structure assumed until now, which would require a major reassessment of current theories for the superconducting phase diagram.

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