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Antiferromagnetic order in UPt_3 under pressure: Evidence for a direct coupling to superconductivity

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We use neutron diffraction to study the effect of hydrostatic pressure on the ordered antiferromagnetic moment in the heavy-fermion superconductor UPt_3. It is found that hydrostatic pressure suppresses and eventually destroys the antiferromagnetic order. A comparison of our results with specific-heat measurements made under hydrostatic pressure shows that the splitting of the two anomalies in the low-temperature heat capacity correlates well with the collapse in the magnitude of the ordered moment. This suggests a coupling between the superconducting and antiferromagnetic order parameters.

Superconductivity in heavy-fermion systems has attracted considerable interest because the pairing mechanism may not be directly dependent on the electron-phonon interaction. In its normal state at low temperatures (below 1.5 K), UPt_3 displays a Fermi-liquid behavior[1] characterized by a linear temperature dependence for the heat capacity ($C=\gamma T$), a virtually constant magnetic susceptibility [$\chi(T)=\chi(0)$], and resistivity quadratic in temperature ($\rho=\rho_0+AT^2$). It has well-defined heavy-fermion quasiparticles as established by studies of the de Haas–van Alphen effect.[2] Due to electron-electron interactions, γ, $\chi(0)$, and A are several orders of magnitude larger than in simple metals. The formation of a superconducting state (below 0.5 K) from such a highly correlated Fermi liquid suggests that an unconventional (non-BCS) type of pairing mechanism may be involved. In its superconducting state, UPt_3 exhibits nonexponential temperature dependences in the specific heat, thermal conductivity, ultrasound attenuation, and NMR relaxation rate suggesting the existence of nodes in the superconducting gap function.[1] The case for unconventional pairing in UPt_3 has been considerably strengthened by the discovery of successive superconducting transitions, at T_{c+} and T_{c-}, corresponding to more than one superconducting phase.[3] Of particular interest here are two additional findings. (i) The two superconducting transitions observed in zero field, $H=0$, converge into one with the application of hydrostatic pressures[4] in excess of 4 kbar. (ii) The observation[5-7] of a small ordered antiferromagnetic moment ($\sim 0.02\mu_B$/U atom) in UPt_3. In this paper, we show that the application of hydrostatic pressure also leads to a reduction of the magnitude of the ordered moment, with the moment disappearing altogether at a critical pressure $p_c=5.4\pm2.9$ kbar. The splitting of the two specific-heat anomalies follows the magnitude of the ordered moment.

Neutron-scattering studies have shown that UPt_3 exhibits strong *magnetic fluctuations* on several frequency scales with different characteristic wave vectors.[8] Recently, a number of high-quality crystals of UPt_3 have been found to show *static* antiferromagnetic correlations[5-7,9] with a small ordered moment ($M\approx0.02\mu_B$/U atom) and a Néel temperature $T_N\approx6$ K. The magnetic order in UPt_3 probably arises due to the partial antiferromagnetic alignment of the fluctuating U $5f$ moments. The experimental resolution of μSR measurements[7] provides a lower limit of approximately 10 μs for the time scale on which the order persists. (The corresponding frequency scale is clearly several orders of magnitude smaller than that appropriate for the superconductivity Δ/h.) Small ordered moments have also been reported in other heavy-fermion compounds (e.g., URu_2Si_2, $CeAl_3$, and UBe_{13} with a few percent Th).

We performed our neutron-scattering measurements using the IN12 triple-axis spectrometer on a cold guide at the Institut Laue-Langevin in Grenoble. Pyrolitic graphite served as monochromator and analyzer, and a cooled Be filter was used to eliminate higher-order neutrons reflected by the monochromator. The incident wave vector was $k_i=1.35$ Å^{-1}. The collimation of the neutrons incident on the monochromator (determined by the neutron guide) was 38′, and 60′ collimators were installed before the sample, analyzer, and detector. The sample was mounted in a carefully shielded pressure cell made from a high-strength aluminium alloy. Hydrostatic pressure was provided by a ^{4}He gas compressor, the sample being surrounded by solid ^{4}He during the measurements.

Our sample was a single crystal of mass 550 mg prepared from high-purity starting materials using a ultra-high-vacuum zoning apparatus based on rf heating and a water-cooled copper crucible. This method of preparation[2] is expected to produce samples with extremely low levels of chemical impurities. However, this does not necessarily imply that the lattice is free from imperfections. Single crystals produced in nominally identical conditions have been used for de Haas–van Alphen (dHvA) studies of the Fermi surface and have shown electron mean free paths in the range 1000–2000 Å (as determined from the damping of the dHvA signal[2]). The residual resistivity $\rho_0(\mathbf{J}\|\mathbf{c})$ of crystals cut from material next to our sample was in the range 0.4–0.8 $\mu\Omega$ cm. The sample used here showed two sharp specific-heat anomalies associated with the successive superconducting transitions at $T_{c+}=0.50$ K and $T_{c-}=0.44$ K.[10]

UPt_3 crystallizes in a hcp structure with space group $P6_3/mmc$. Bragg positions are labeled using reciprocal lattice units, where $a^*=b^*=4\pi/(a\sqrt{3})=1.264$ Å^{-1} and $c^*=2\pi/c=1.283$ Å^{-1}. We study the scattering arising from the interaction of the neutron's magnetic moment with the local magnetic field in the sample due to the electrons. Neutron scattering measures $\mathbf{M}$ the spatial Fourier component of the moment *perpendicular* to the scattering vector $\mathbf{Q}=\mathbf{k}_i-\mathbf{k}_f$, where $\mathbf{k}_i$ and $\mathbf{k}_f$ are the incident and final neutron wave vectors. The magnetic order in UPt_3 corresponds to a doubling of the unit cell along the $\mathbf{a}^*$-type direction,[6] with the ordered moment also lying in the basal plane, parallel to $\mathbf{a}^*$, as shown in Fig. 1(a). The magnetic structure in Fig. 1(a) gives rise to magnetic peaks centered at positions $\mathbf{Q}_0=(\frac{1}{2},1,0)$, $(\frac{3}{2},\bar{1},0)$, $(\frac{\bar{1}}{2},\bar{1},0)$, and $(\frac{\bar{3}}{2},1,0)$, shown by the open circles in Fig. 1(b). For positions such as $(\frac{1}{2},0,0)$ and $(\frac{\bar{1}}{2},0,0)$, $\mathbf{M}$ is parallel to $\mathbf{Q}$ and the reflections have zero intensity. We can describe the structure shown in Fig. 1(a) by its propagation vector $\mathbf{q}_1$. There are two symmetry-related structures $\mathbf{q}_2$ and $\mathbf{q}_3$ corresponding to a rotation of Fig. 1(a) by 120° and 240°, respectively.

Figure 1(c) shows transverse scans (where $\mathbf{Q}=\mathbf{Q}_0+\delta\mathbf{Q}$, and $\delta\mathbf{Q}\perp\mathbf{Q}_0$) through Bragg positions $\mathbf{Q}_0$ corresponding to $\mathbf{q}_1$, $\mathbf{q}_2$, and $\mathbf{q}_3$. For $p=0$ and $T=1.8$ K (closed circles), we observe intensity at each position showing that the overall structure is made up of a combination of structures $\mathbf{q}_1$, $\mathbf{q}_2$, and $\mathbf{q}_3$. Figure 2 shows longitudinal scans $(\delta\mathbf{Q}\|\mathbf{Q}_0)$ through the $\mathbf{Q}_0=(\frac{1}{2},1,0)$ position. From a comparison of the intensity of the magnetic peaks with that of the (1,1,0) nuclear peak, we estimate $M=0.03\pm0.01\mu_B$/U atom, close to the value obtained in previous measurements.[5]

The application of 4.3-kbar hydrostatic pressure causes a suppression of the antiferromagnetic peaks $\mathbf{q}_1$, $\mathbf{q}_2$, and $\mathbf{q}_3$, as can be seen from the open circles in Fig. 1(c). In addition, the lower scan in Fig. 1(c) shows that no peak appears at the $(\frac{1}{2},0,0)$ position under pressure; hence the

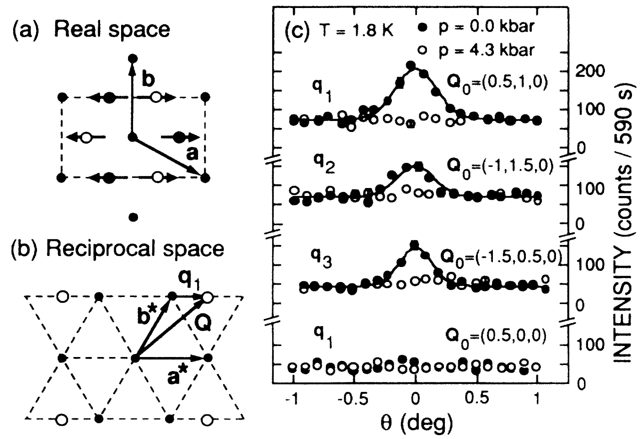

FIG. 1. (a) The antiferromagnetic structure of UPt_3. Larger circles represent the positions of the U atoms, with $z=\frac{3}{4}c$ (closed) and $z=\frac{1}{4}c$ (open). Small closed circles are lattice points. The dashed line encloses the doubled orthorhombic antiferromagnetic unit cell. (b) Reciprocal lattice (closed circles). The structure in (a) is described by the propagation vector $\mathbf{q}_1$ and gives rise to magnetic reflections at the positions of the open circles. (c) The three uppermost traces are transverse scans (with and without applied hydrostatic pressure) through the antiferromagnetic peaks associated with the three possible domains $\mathbf{q}_1$, $\mathbf{q}_2$, and $\mathbf{q}_3$. The lower trace is a scan through a position sensitive to a moment rotation (see text).

suppression is not due to a rotation of the ordered moments away from $\mathbf{M}\|\mathbf{a}^*$. The integrated intensity of the peak is a measure of the square of the *ordered* moment M^2 (or $|\langle M\rangle|^2$), its variation with pressure is shown in Fig. 3(a), for $T=1.8$ K. The functional form for $M(p)$ is unknown *a priori:* the solid line in Fig. 3(a) is a fit to the

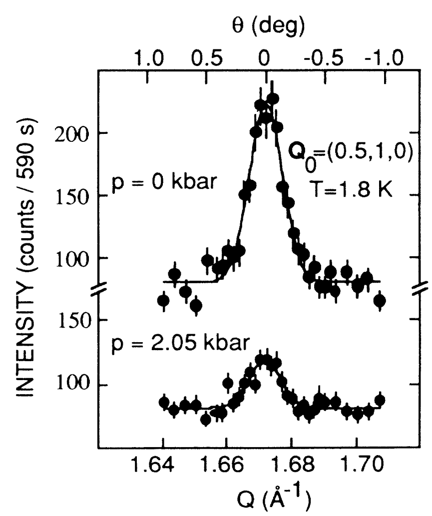

FIG. 2. (a) Longitudinal scans through the $(\frac{1}{2},1,0)$ magnetic peak of domain $\mathbf{q}_1$ without hydrostatic pressure and with $p=2.05$ kbar. The full widths at half maximum for the magnetic peaks are 0.0126(6) Å^{-1} and 0.0116(15) Å^{-1}, for $p=0$ and $p=2.05$ kbar, respectively.

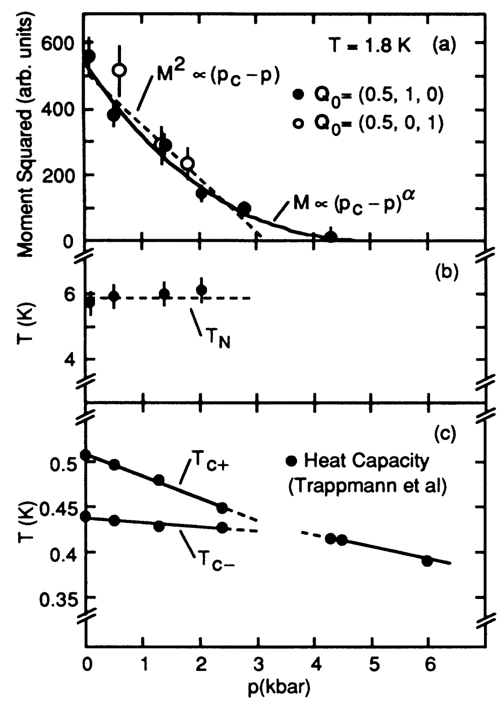

FIG. 3. (a) The variation of the integrated intensity of the magnetic peaks $(\frac{1}{2},1,0)$ (closed circles) and $(\frac{1}{2},0,1)$ (open circles) with hydrostatic pressure. This is a measure of M^2. The solid line is a fit of $M \propto (p_c - p)^\alpha$ and the dashed line $M^2 \propto (p_c - p)$, these yield $p_c = 5.4 \pm 2.9$ kbar and $p_c = 3.2 \pm 0.2$ kbar, respectively. (b) The Néel temperature determined from data such as in Fig. 4(a). (c) Low-temperature specific-heat anomalies in UPt_3, from Trappmann and co-workers (Ref. 4).

general form $M \propto (p_c - p)^\alpha$ for $p < p_c$ and $M = 0$ for $p > p_c$, which gives $\alpha = 2.6 \pm 1.9$ and $p_c = 5.4 \pm 2.9$. [A fit of $M^2(p) \propto (p_c - p)$ gives a critical pressure $p_c(T = 1.8$ K$) = 3.2 \pm 0.2$ kbar, in agreement with the preliminary value[11] ($p_c = 2.9 \pm 0.4$ kbar) obtained by extrapolating low-pressure data using this form.]

The temperature dependence of the $(\frac{1}{2},1,0)$ peak intensity is shown in Fig. 4(a) for $p = 0$ and 2.05 kbar. For all pressures investigated, we observe a mean-field behavior $M^2 \propto (T_N - T)$. Aeppli *et al.*[5] have found the same temperature dependence (for $p = 0$) down to T_c, and have observed directly the coexistence of antiferromagnetism and superconductivity below T_c. From Fig. 4(a) one can see that the application of 2.05-kbar hydrostatic pressure causes no observable change in T_N, while the ordered moment M is reduced by a large factor of 0.50 ± 0.04. This is a rather unexpected result, since T_N usually varies strongly with M. The variation of T_N with pressure is shown in Fig. 3(b). If we describe the interaction of the ordered moments with a Heisenberg model then $k_B T_N \approx J M_0^2/3$. The ratio T_N/M_0^2, and hence J is increased by a factor of 3 under 2.05 kbar. Thus, the *effective coupling* between magnetic moments appears to be strongly pressure dependent.

We now discuss the relationship of our results to the superconductivity in UPt_3. Specific heat, thermal expan-

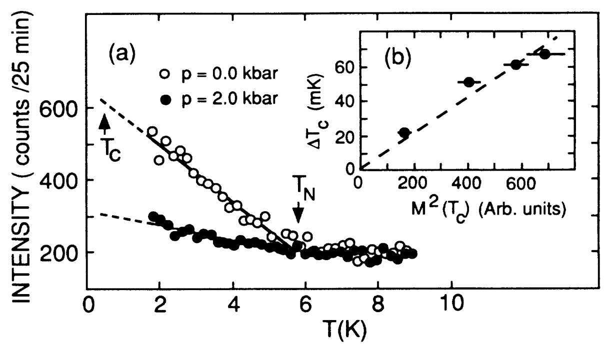

FIG. 4. (a) Variation of the intensity of the $(\frac{1}{2},1,0)$ magnetic peak with temperature for two pressures. Solid lines are fits to $M^2 \propto (T_N - T)$, used to determine T_N. (b) The inset shows the measured variation of the splitting ΔT_c (from Ref. 4) as a function of $M^2(T_c)$. $M^2(T_c)$ has been determined from the integrated intensity of longitudinal scans as in Fig. 2 extrapolated to T_c using the form $M^2 \propto (T_N - T)$.

sion, and sound velocity measurements[1] have all shown that there are two distinct superconducting phase transitions in UPt_3 for $H = 0$. The results of heat capacity measurements under pressure by Trappmann and coworkers[4] are reproduced in Fig. 3(c). These authors find that the two anomalies at T_{c+} and T_{c-} converge, *rather than cross,* under hydrostatic pressure. For $p > 4.2$ kbar, they observe only one transition. Resistivity measurements[12] of $T_{c+}(p)$ are consistent with the specific-heat measurements: in both cases a rapid initial decrease (dT_{c+}/dp) is observed, followed by a slower decrease at higher pressures [see Fig. 3(c)]. From resistivity measurements[12] the change in gradient takes place in the range 2.3–5.3 kbar. Thermal expansion data[13] and heat-capacity measurements made under uniaxial stress[14] have revealed that it is the decrease in c-axis lattice parameter that drives the convergence of the two transitions.

The two transitions in UPt_3 have been interpreted as being due to a splitting by a symmetry-breaking perturbation[15–17] of a doubly degenerate superconducting order parameter. The presence of the antiferromagnetic order, which has a different symmetry to the ionic lattice, can provide such a perturbation. To lowest order, the splitting is proportional to the square of the magnetic moment,[18] in broad agreement with our measurements (see inset of Fig. 4). In its simplest form, the basic model[15–17] fails to account for the observed lack of anisotropy of $T_c(\hat{\mathbf{H}})$, for $\mathbf{H} \perp \mathbf{c}$ and the presence of a tetracritical point for $\mathbf{H} \parallel \mathbf{c}$.[19,20] Whether this is because the symmetry is broken locally[21] remains to be investigated. Other scenarios for phase multiplicity in UPt_3 have also been considered. Two separate but nearly degenerate representations[21] may give rise to a double transition. However, this would probably predict a crossing of $T_{c+}(p)$ and $T_{c-}(p)$ rather than a merging.

In summary, we have used elastic neutron diffraction to study the effect of hydrostatic pressure on the small ordered antiferromagnetic moment in UPt_3. We find that pressure suppresses and eventually destroys the antiferromagnetic order. A comparison of our results with previous specific-heat measurements[4] made under hydrostatic pressure shows that the splitting of the superconducting transition is well correlated with the magnitude of the ordered moment. The critical pressure at which the ordered moment is destroyed, $p_c = 5.4 \pm 2.9$ kbar, is indistinguishable from that at which the two anomalies in the low-temperature heat capacity cease to be resolvable. The antiferromagnetic order appears to be responsible for the double superconducting transition in UPt_3.

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