Magnetic Order and Fluctuations in Superconducting UPt_3

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Neutron diffraction shows that superconducting UPt_3 can be an antiferromagnet with an ordered moment of $(0.02 \pm 0.01)\mu_B$ and a Néel temperature (T_N) of 5 K. The squared order parameter is a linear function of temperature from T_N to the superconducting transition temperature, $T_c = 0.5$ K, below which it ceases to evolve. Inelastic neutron scattering demonstrates that the fluctuating moments are strongly correlated; the characteristic energy for fluctuations with wave vectors close to the ordering wave vector is 0.2 meV $\approx 4kT_c$.

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There is a growing consensus that the superconductivity of heavy-fermion systems is due to magnetic fluctuations.[1] Until now, however, the magnetic fluctuations in the heavy-fermion superconductors have not been probed at energies comparable to pair-breaking energies. Furthermore, the influence of superconductivity on magnetic correlations has yet to be demonstrated directly. In the present paper, we describe a neutron-scattering experiment which probes the low-frequency fluctuations in UPt_3. In addition, we show that superconducting UPt_3 can be an antiferromagnet, as has been suggested by muon spin-relaxation measurements,[2] and that the evolution of the antiferromagnetic order terminates at the onset of superconductivity.

The experiments were performed with the TAS-7 triple-axis instrument installed at the end of a guide tube emanating from the cold neutron source of the Risø DR-3 reactor. A 20-cm-high segmented pyrolitic-graphite (PG) crystal provides a vertically focused incident beam. For inelastic measurements, a set of six PG crystals, in a horizontally focusing arrangement, function as an analyzer with an angular acceptance of 6°, full width at half maximum (FWHM). For elastic measurements, the analyzer was flat and used in combination with a collimator. The PG crystals were set for their (002) reflections. Various Be, BeO, and PG filters eliminated higher-order contamination of the beam and reduced the elastic scattering admitted by the analyzer system at nominally finite energy transfers. For inelastic scattering, the final energy was fixed at either $E_f = 5.1$ or 3.735 meV. The corresponding energy resolutions were 0.2 and 0.1 meV (FWHM), respectively.

Four approximately cylindrical (6 mm diam) ingots of UPt_3 were grown by the float-zone method. These were divided to yield six crystals, of which the five largest (each of length $\simeq 4$ cm) were mounted together, so as to intercept the neutron beam, on the cold finger of either a dilution or pumped ^{3}He cryostat. The sixth crystal was mounted above the beam and fitted with coils to measure the ac susceptibility ($f = 100$ Hz) during the scattering experiment. The $(h,0,l)$ zones of the hexagonal close-packed crystals (space-group $P6_3/mmc$) coincided with the horizontal scattering plane of the spectrometer. In our experiment, momentum transfers are expressed in reciprocal-lattice units, where $a^* = 4\pi/a\sqrt{3} = 1.261$ Å^{-1} and $c^* = 2\pi/c = 1.285$ Å^{-1}.

Earlier neutron-scattering[3] experiments at relatively large energy transfers ($\hbar\omega \simeq 6$ meV) are consistent with short-range antiferromagnetic correlations where the two U ions in each unit cell tend to be oppositely polarized. Because the long-range order associated with such correlations would yield magnetic Bragg scattering only for integer-valued reciprocal-lattice indices, it came as a surprise that $U_{1-x}Th_xPt_3$ and $U(Pt_{1-x}Pd_x)_3$, which clearly undergo magnetic phase transitions[4,5] for $x \gtrsim 0.02$, display superlattice peaks[5] at reciprocal-lattice positions of the form $(h+\frac{1}{2},0,l)$, where h and l are integers. Particularly strong peaks were found at $(\pm\frac{1}{2},0,1)$ and $(\pm\frac{1}{2},0,2)$. We therefore set out to probe the elastic and inelastic scattering near $(\pm\frac{1}{2},0,1)$ in our nominally (see below) *pure* crystals of UPt_3.

Figure 1 shows $(h,0,l)$ scans sensitive to magnetic fluctuations [for a fixed energy transfer of 0.5 meV, Fig. 1(a)] and static correlations [$\hbar\omega = 0$, Fig. 1(b)]. Well above 5 K, the two scans are featureless, while at low temperatures, there are well-defined maxima at $Q = (\pm\frac{1}{2}, 0, 1)$. The elastic peak has widths (FWHM) of 0.01 reciprocal-lattice units both parallel and perpendic-

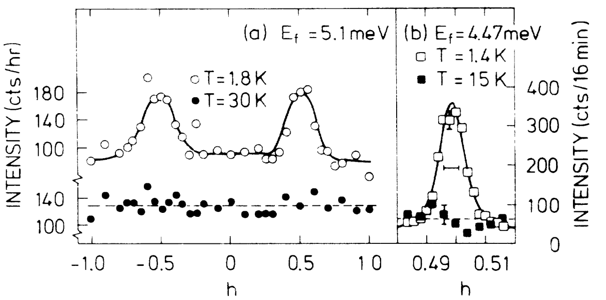

FIG. 1. Constant-energy scans along $(h,0,1)$ above and below T_N. (a) Taken at an energy transfer of 0.5 meV. Note that while the scattering at 30 K no longer peaks at $(\pm\frac{1}{2},0,1)$, its level is higher than that seen between the maxima for $T=1.8$ K. (b) Elastic scattering with a PG and Be filtered beam. The line through these points is a resolution-corrected Lorentzian. The horizontal bar indicates the spectrometer resolution as measured by $\lambda/2$ scattering from the nuclear (102) Bragg reflection.

ular to the basal planes; the corresponding resolution widths are 0.006 reciprocal-lattice units in both directions. A survey in the $(h,0,l)$ plane revealed superlattice reflections at other reciprocal-lattice points of the form $\mathbf{Q}=(h+\frac{1}{2},0,l)$ (h and l are integers). The associated intensities decrease with $|\mathbf{Q}|$ as expected for magnetic reflections; furthermore, they are consistent with the magnetic structure found for $U_{1-x}Th_xPt_3$ and $U(Pd_{1-x}Pt_x)_3$.[5] For this structure, the unit cell is doubled in the basal planes and the ordered moment μ_0 is parallel to the doubling direction. We have determined the ordered moment via normalization with respect to the weak nuclear Bragg peak at (1,0,1). The result, $(0.02\pm0.01)\mu_B$, is consistent with the less exact estimate $0.01\mu_B<\mu_0<0.1\mu_B$ based on the muon spin-relaxation measurements[2] and is much smaller than that $(0.7\pm0.1)\mu_B$ for the doped compounds.[5]

To test whether the magnetic order is intrinsic to UPt_3, we have separately examined the four boules which were grown at different times and with different defect (stacking fault) densities, as established by neutron diffraction. The ordered moments and Néel temperatures [$T_N=5$ K, see Fig. 2(a) and the discussion below] are all identical to within experimental error. The Néel temperature of our samples is higher than that (3.5 K) of the most lightly doped compound, $U(Pt_{1-x}Pd_x)_3$ with $x=2\%$, for which bulk measurements[4] yield clear signatures of a magnetic transition and no sign of superconductivity. Electron microprobe analysis on a segment of one of our samples revealed no impurities with concentrations beyond the detectability limit (0.1 wt.%). According to the more sensitive technique of inductive coupled plasma mass spectrometry, the only appreciable contamination is due to a Pb isotope ($\lesssim 10$ ppm by weight), probably the result of radioactive decay; Pd and Th levels were $\lesssim 1$ ppm by weight. In regard to the purity of our samples, we also recall that the superconducting transition temperature and width are very sensitive to chemical and other defects.[1] Our ac susceptibility data, shown in Fig. 3, indicate a relatively high T_c ($=0.5$ K) and a narrow transition. We conclude that the magnetic order reported here is a property of superconducting UPt_3. Even though by most standards, our samples are exceptionally clean, *some* disorder must be present because the magnetic diffraction peaks are not resolution limited. Interestingly, the resolution-corrected widths of the diffraction peaks correspond to distances comparable to the electronic mean free path ($\simeq 250$ Å) established for some,[6] but not all,[7] crystals of UPt_3.

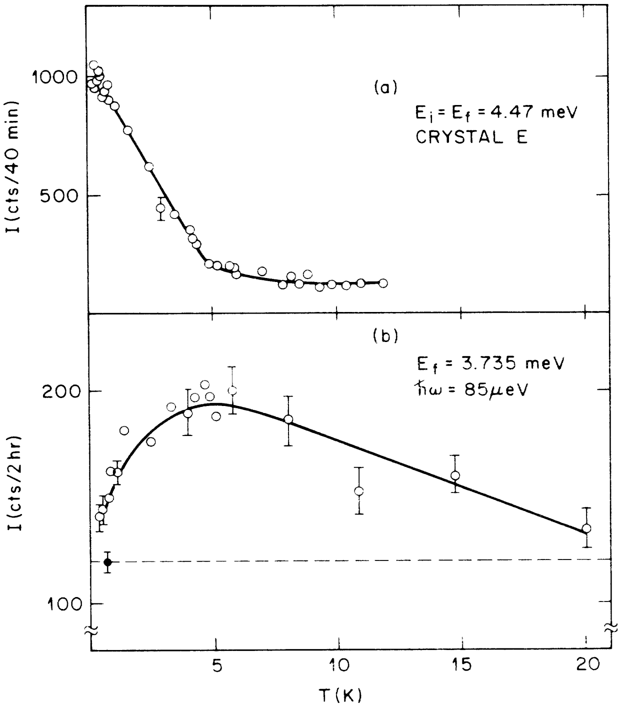

FIG. 2. Temperature dependence of (a) the elastic peak intensity of $Q=(\frac{1}{2},0,1)$ and of (b) the scattering of $Q=(0.52,0,0.99)$ with an energy transfer of 85 μeV.

It is apparent from Fig. 1(b) that there are substantial magnetic fluctuations with wave vectors near $(\frac{1}{2},0,1)$. To find the spectrum of these fluctuations, we performed constant-Q scans as shown in Fig. 4. We have previously[8] reported similar scans performed with coarser energy resolution; more recently, others[9] have also noted enhanced magnetic diffuse scattering and weak magnetic Bragg scattering at $(\frac{1}{2},0,1)$ in nominally pure UPt_3. Figure 4(a) displays raw data for two momentum transfers and temperatures. At $Q=(0.3,0,1.0)$, the signal for $\hbar\omega>0.15$ meV is indistinguishable from the background level, established for neutron energy gain at $T=0.5$ K. The increase for $\hbar\omega<0.15$ is due to incoherent scattering with an intensity of 10 counts/min at $\hbar\omega=0$. We can extract the imaginary part, $\chi''(Q,\omega)$, of

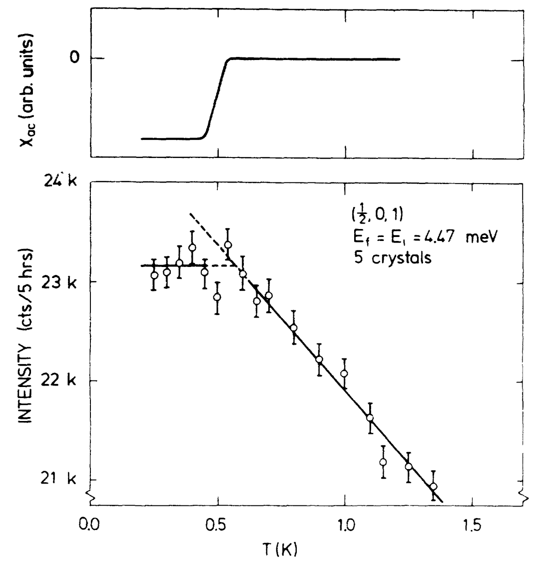

FIG. 3. Temperature dependence of the peak intensity at $Q=(\frac{1}{2},0,1)$ near the superconducting phase transition at $T_c=0.5$ K. The incoherent background at $Q=(\frac{1}{2},0,1)$ and $T=15$ K was 4761 counts/5 h. The top frame shows the temperature dependence of the ac susceptibility measured simultaneously on a similarly mounted UPt$_3$ crystal.

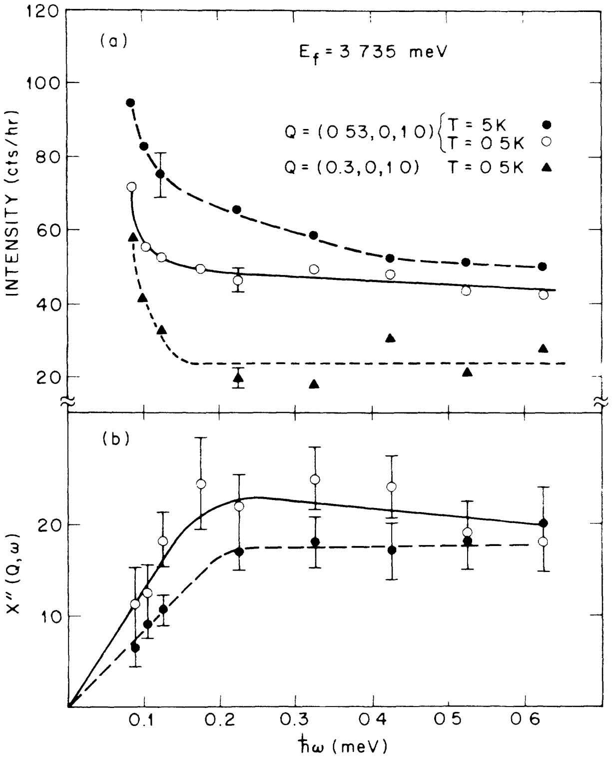

FIG. 4. (a) Constant-Q scans at $Q=(0.3,0,1)$ at $T=0.5$ K and $Q=(0.53,0,1)$ at $T=5$ K and $T=0.5$ K. (b) $\chi''(Q,\omega)$ at the same two temperatures deduced from these data as described in the text.

the magnetic susceptibility using the fluctuation-dissipation theorem,

$$S(Q,\omega)=\chi''(Q,\omega)[1-\exp(-\beta\hbar\omega)]^{-1},$$

where to obtain $S(Q,\omega)$, we have subtracted as background the $Q=(0.3,0,1)$ data for $\hbar\omega<0.15$ meV and a constant of 24 counts/h for $\hbar\omega\geq 0.15$ meV. Figure 4(b) shows the results. The shape of $\chi''(Q,\omega)$ is similar at both temperatures: There is a rise between $\hbar\omega=0$ and 0.2 meV, followed by a plateau. While $S(Q,\omega)$ is considerably larger at the higher temperature, $\chi''(Q,\omega)$ is somewhat smaller. At $T=0.5$ K, $\chi''(Q,\omega)$ is insensitive to small offsets (such as $\Delta h=0.03$ as shown in Fig. 4) from the ordering wave vector. Thus, the result that $\chi''(Q,\omega)$ decreases as ω is reduced below 0.2 meV permits the identification of 0.2 meV as a characteristic energy for fluctuations of moment-containing regions of size given by the inverse resolution volume ($\sim 10^4$ Å^3) of the instrument.

We now describe how the magnetic order and fluctuations vary with T. Figure 2(a) shows the temperature dependence of the magnetic Bragg intensity I_B at $Q=(0.5,1,1.0)$ for one of our crystals. For $T<T_\mathrm{N}=5$ K, I_B rises linearly with decreasing temperature, a feature found for the other two crystals examined in similar detail. Thus, the mean-field expression, $M\sim(T_\mathrm{N}-T)^{1/2}$, adequately describes the temperature dependence of the magnetic order parameter, over a much wider range in T/T_N than is ordinarily the case.

Figure 2(b) shows that $S(Q,\omega)$, measured with Q offset slightly from the ordering vector to eliminate Bragg contamination, has a maximum near T_N. This result is expected for conventional magnets undergoing second-order phase transitions. The decrease for $T<T_N$ is associated with a reduction in the fluctuating moment as the system becomes progressively more ordered. For UPt$_3$, the magnetoresistance $(\partial\rho/\partial H)$ and the temperature derivative $(\partial\rho/\partial T)$ of the resistivity[6] have temperature dependences similar to that of $S(Q,\omega)$ shown in Fig. 2(b). Thus, the hitherto unexplained maxima in $\partial\rho/\partial T$ and $\partial\rho/\partial H$ near 5 K are most likely due to the antiferromagnetic correlations probed in the present experiment.

To determine whether superconductivity influences the magnetic order of UPt$_3$, we have performed a more detailed study, using five crystals, of the $(\frac{1}{2},0,1)$ intensity at low temperatures. Figure 3 shows the results, together with ac susceptibility data collected simultaneously on a sixth crystal not in the neutron beam. The linear rise in I_B persists to T_c, below which I_B becomes temperature independent. While the influence of superconductivity on magnetic order has been noted for other systems,[10] UPt$_3$ is the first system to display such an effect where electrons of the same type are responsible for both the superconductivity and the magnetism. In the superconducting state, electrons are paired so that they can no longer contribute to the antiferromagnetic moment. Further evolution of I_B is suppressed, especially if the same interactions lead to superconductivity as to antiferromagnetic order.[1]

We have shown that UPt_3 is an antiferromagnet with a Néel temperature of 5 K and an ordered moment of $(0.02 \pm 0.01)\mu_B$. This result accounts for hitherto unexplained peaks in the magnetoresistance and temperature derivative of the resistivity. The magnetic order parameter grows as $(T_N - T)^{1/2}$ for $T_c < T < T_N$, after which it ceases to evolve. Thus, we have the first *direct* evidence that superconducting and magnetic order parameters are strongly coupled in heavy-fermion systems. Our inelastic measurements show that there are very strong correlations among the fluctuating moments; these correlations are of the same type as those associated with the magnetic order. The correlated regions fluctuate with a characteristic frequency of 0.2 meV, not far from the pair-breaking energy associated with the superconducting T_c of UPt_3.

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[1] G. R. Stewart, Z. Fisk, J. O. Willis, and J. L. Smith, Phys. Rev. Lett. **52**, 679 (1984); K. Miyake, S. Schmitt-Rink, and C. M. Varma, Phys. Rev. B **34**, 6554 (1986); J. E. Hirsch, Phys. Rev. Lett. **54**, 1317 (1985); M. T. Beal-Monod, C. Bourbonnais, and V. J. Emery, Phys. Rev. B **34**, 7716 (1986); M. R. Norman, Phys. Rev. Lett. **59**, 232 (1987); K. Machida and M. Kato, Phys. Rev. Lett. **58**, 1986 (1987).

[2] R. Heffner, in Proceedings of the Fifth International Conference on Valence Fluctuations, Bangalore, India, 1987, edited by S. Malik and K. Gupta (to be published).

[3] G. Aeppli, A. Goldman, G. Shirane, E. Bucher, and M. Ch. Lux-Steiner, Phys. Rev. Lett. **58**, 808 (1987); A. I. Goldman, G. Shirane, G. Aeppli, E. Bucher, and J. Hufnagel, to be published.

[4] A. de Visser, J. C. P. Klaase, M. van Sprang, J. J. M. Franse, A. Manovsky, and T. T. M. Palstra, J. Magn. Magn. Mater. **54–57**, 375 (1986); A. P. Ramirez, B. Batlogg, E. Bucher, and A. S. Cooper, Phys. Rev. Lett. **57**, 1072 (1986); G. R. Stewart, A. L. Giorgi, J. O. Willis, and J. O'Rourke, Phys. Rev. B **34**, 4629 (1986).

[5] A. I. Goldman, G. Shirane, G. Aeppli, B. Batlogg, and E. Bucher, Phys. Rev. B **34**, 6561 (1986); P. H. Frings, B. Renker, and C. Vettier, J. Magn. Magn. Mater. **63–64**, 202 (1987).

[6] A. de Visser, Ph.D. thesis, University of Amsterdam, 1986 (unpublished).

[7] B. S. Shivaram, T. F. Rosenbaum, and D. J. Hinks, Phys. Rev. Lett. **57**, 1259 (1986).

[8] C. Broholm, J. K. Kjems, G. Aeppli, E. Bucher, and W. J. L. Buyers, in *Magnetic Excitations and Fluctuations II,* edited by Balucani, S. W. Lovesy, M. G. Rasetti, and V. Tognetti (Springer-Verlag, Berlin, 1987), p. 162.

[9] P. Frings, B. Renker, and C. Vettier, to be published.

[10] See, e.g., S. K. Sinha, G. W. Crabtree, D. G. Hinks, and H. Mook, Phys. Rev. Lett. **48**, 950 (1982), and references therein.