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Antiferromagnetic order in $U_{1-x}Th_xPt_3$

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Neutron-diffraction studies show that the phase transition at 6.5 K in $U_{0.95}Th_{0.05}Pt_3$ is to an antiferromagnetic ground state. The magnetic unit cell is obtained from doubling the nuclear unit cell along one direction in the basal plane. The ordered moment is $(0.65 \pm 0.1)\mu_B$/U atom, which is well below the moment of $2.6\mu_B$/U atom extracted from the high-temperature, Curie-Weiss susceptibility of UPt_3.

A central issue in the field of heavy-fermion systems (HFS) concerns the interactions which lead to superconductivity. Recent theoretical studies[1] suggest a connection between the antiferromagnetic (AFM) correlations observed in neutron-scattering measurements on UPt_3 (Ref. 2) and the superconductivity of this compound. The introduction of small concentrations of impurities into UBe_{13} and UPt_3 results in new and interesting low-temperature behavior. For instance, the addition of several percent Th into UBe_{13} yields a new anomaly in the specific heat at a temperature below the superconducting transition.[3] It has been proposed that this second transition is to a magnetically ordered ground state.[4] However, neutron diffraction[5] has failed to produce evidence for this, perhaps because the ordered moment is unobservably small. Recent thermodynamic and transport studies on UPt_3 doped with small amounts of Th and Pd[6–9] reveal a phase transition at 6.5 K, which has been interpreted as arising from a Fermi-surface instability resulting in a spin-density wave.[6] Frings, Renker, and Vettier[10] have studied magnetic order in $U(Pt_{1-x}Pd_x)_3$. Here, we report neutron-diffraction measurements leading to the discovery of magnetic order in $U_{0.95}Th_{0.05}Pt_3$.

We performed our neutron-diffraction measurements with a triple-axis spectrometer at the Brookhaven High-Flux Beam Reactor using pyrolitic graphite crystals, set for their (002) reflections, as monochromator and analyzer. The neutron energies and collimations for the two spectrometer configurations we employed were 14.7 meV and 20′-20′-20′-40′, and 42 meV and 20′-10′-10′-40′, respectively. In both configurations, two graphite filters, one mounted before the monochromator and the other installed after the analyzer, suppressed higher-order contamination of the neutron beam.

The sample, grown by the Czochralski method, was an approximately cylindrical ingot with a length of 2 cm and a diameter of 6 mm. The cylinder axis was roughly parallel to the crystallographic c axis of the material. We mounted the sample in a $(h0l)$ zone, where h and l are measured in units of $a^* = 4\pi/a\sqrt{3} = 1.263$ Å^{-1} and $c^* = 2\pi/c = 1.282$ Å^{-1}. UPt_3 crystallizes in the Ni_3Sn structure, with space group $P6_3/mmc$.[11] Figure 1(a) shows the arrangement of the U atoms in the $2(c)$ sites of this structure, and Fig. 2 displays the location of the nuclear Bragg peaks in the $(h0l)$ zone of the reciprocal lattice.

In order to locate the magnetic superlattice reflections, we searched the shaded portion (see Fig. 2) of the $(h0l)$ zone by measuring intensities at reciprocal-lattice points on a grid with a mesh size ($\Delta h = 0.02a^*$, $\Delta l = 0.02c^*$) comparable to the instrumental resolution. The only appreciable (intensity greater than 20 counts/min) temperature-dependent scattering was associated with the $(\frac{1}{2},0,1)$ point, for which the intensity at 1.5 K was 880 counts/min). Figure 3 displays the temperature dependence of the $(\frac{1}{2},0,1)$ intensity, which vanishes at

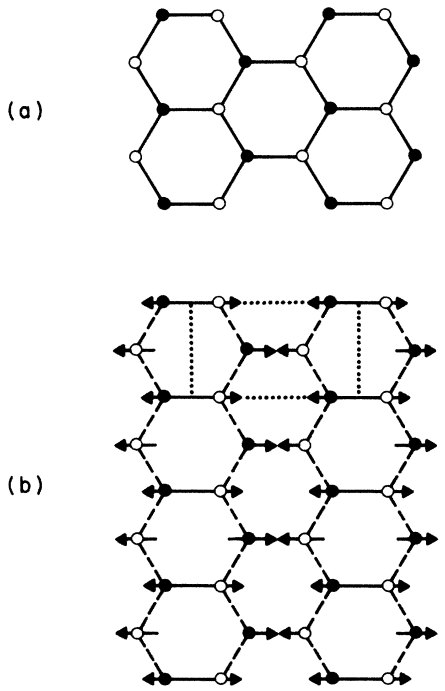

FIG. 1. (a) The open and closed circles denote the positions of U atoms in two adjacent basal planes, respectively, of UPt_3 (Ref. 11). The planes are a distance $c/2$ apart. (b) The magnetic structure of $U_{0.95}Th_{0.05}Pt_3$ is determined in this work. Arrows denote the spin directions.

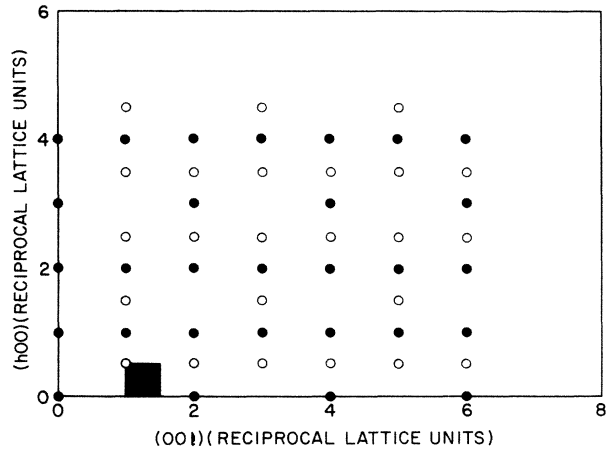

FIG. 2. The $(h0l)$ zone of the reciprocal lattice for UPt_3 (Ref. 11). Closed circles indicate the positions of the allowed nuclear reflections. The open circles denote the positions of the observed magnetic superlattice reflections for $U_{0.95}Th_{0.05}Pt_3$.

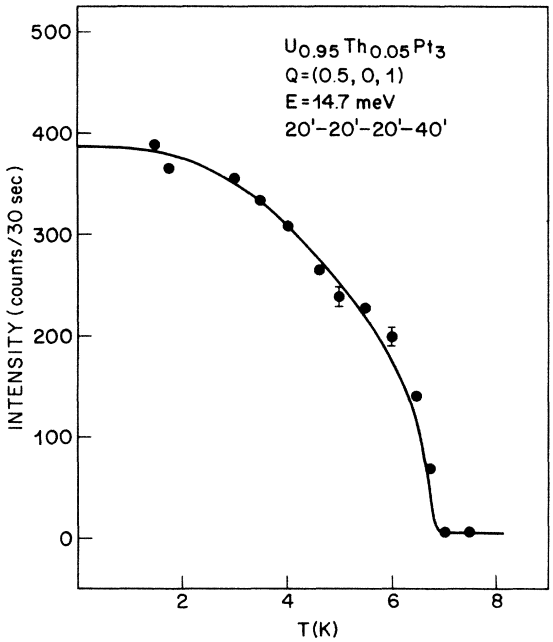

FIG. 3. Temperature dependence of the magnetic Bragg intensity measured at the $(\frac{1}{2},0,1)$ reciprocal-lattice point for $U_{0.95}Th_{0.05}Pt_3$.

$T_N = 6.8 \pm 0.5$ K, in agreement with the bulk ordering temperature of 6.5 K.[6]

An extended search of the reciprocal-lattice points $(h0l) = (0.25n,0,0.25m)$ with $n^2+m^2 \leq 25$ showed that the only new reflections which appear for $T < T_N$ are located at points where h is a half integer and l is an integer subject to the following conditions: (i) $l \neq 0$, (ii) if l is even, $2h$ must not be divisible by three. In other words, reflections at $(\frac{3}{2},0,2)$ and $(\frac{3}{2},0,4)$ are forbidden. The open circles in Fig. 2 indicate the positions of the allowed superlattice reflections.

Both surveys described above were carried out with $E = 14.7$ meV. To reach larger momentum transfers (Q), where a magnetic-form-factor dependence of the scattering might be observed, we performed additional measurements at $E = 42$ meV. For $Q > 5$ Å^{-1}, the intensities are of a level indistinguishable from background. Furthermore, even for $Q < 5$ Å^{-1}, the Bragg intensities (see Table I) are lower for the higher-order reflections. Therefore, the phase transition at $T_N = 6.5$ K in $U_{0.95}Th_{0.05}Pt_3$ is to a magnetically ordered ground state.

The simplest AFM structure for the honeycomb lattice of Fig. 1(a) is where the spins are parallel to all of the other spins in the same basal plane and antiparallel to the spins in the neighboring planes, displaced by $c/2$. This configuration does, in fact, adequately describe the character of the short-range antiferromagnetic fluctuations observed in UPt_3.[12] However, this yields a propagation vector along c which is inconsistent with the results of this study on $U_{0.95}Th_{0.05}Pt_3$. Figure 1(b) illustrates an ordered magnetic structure consistent with the presence of reflections with half-integer h and the extinction rules (i) and (ii). The magnetic unit cell is orthorhombic and results from doubling the hexagonal unit cell along one in-plane direction. Rule (i) is satisfied when the spins lay along the direction in which the unit cell is doubled; this follows because magnetic scattering is never observed when $\hat{\mathbf{Q}}$ is parallel to the spin direction $\hat{\mathbf{S}}$. In Table I the observed Bragg intensities, divided by $1-(\hat{\mathbf{Q}}\cdot\hat{\mathbf{S}})^2$ and the square of the U $5f$ form factor,[13] are compared to the square of the structure factor calculated for the magnetic unit cell of

TABLE I. Observed and calculated magnetic intensities for $U_{0.95}Th_{0.05}Pt_3$ at $T = 1.5$ K. The observed intensities are expressed in barns per nuclear unit cell (b/uc); the (002) and (200) intensities were used for the conversion to absolute units. The calculated intensities are for the magnetic structure shown in Fig. 1(b) and an ordered moment of $0.65\mu_B$ per U atom. The magnetic form factor is from Ref. 13.

$h0l$	Q (Å^{-1})	I_{obs} (10^{-2} b/uc)	$1-(\hat{\mathbf{Q}}\cdot\hat{\mathbf{S}})^2$	$\lvert f(Q)\rvert^2$	$\lvert F\rvert^2_{obs}$ (10^{-2} b/uc)	$\lvert F\rvert^2_{calc}$ (10^{-2} b/uc)
$\frac{1}{2}01$	1.43	1.13 ± 0.03	0.80	0.9	1.57 ± 0.05	1.0
$\frac{1}{2}02$	2.63	2.00 ± 0.06	0.94	0.67	3.17 ± 0.10	3.1
$\frac{1}{2}03$	3.89	0.28 ± 0.07	0.97	0.42	0.70 ± 0.15	1.0
$\frac{3}{2}01$	2.27	1.02 ± 0.04	0.31	0.74	4.45 ± 0.15	4.1
$\frac{5}{2}01$	3.38	0.03 ± 0.03	0.14	0.46	0.47 ± 0.47	1.0
$\frac{5}{2}02$	4.02	0.38 ± 0.04	0.39	0.38	2.55 ± 0.25	3.1

Fig. 1(b). The agreement is quite satisfactory. From normalizing to the (002) and (200) nuclear Bragg intensities, we estimate that the ordered moment is $\mu_0 = (0.65 \pm 0.1)\mu_B$/U atom. During the course of our investigation we were informed of a similar study[10] on $U(Pt_{1-x}Pd_x)_3$. The magnetic structure and ordered moment for this system are the same as that reported here for $(U_{1-x}Th_x)Pt_3$.

Since the magnetic structures and ordered moment of the $U_{0.95}Th_{0.05}Pt_3$ and $U(Pt_{1-x}Pd_x)_3$ systems are the same, it does not seem that antiferromagnetism in these compounds arises from simple dilution (e.g., the substitution of a nonmagnetic species on the U sublattice), but rather more from changes in the unit-cell dimensions upon doping. The substitution of small concentrations of Th for U[6] or Pd for Pt[7] increases the unit-cell volume, decreases the c/a ratio, and leads to magnetic order while suppressing superconductivity. In contrast, the substitution of small amounts of Au for Pt[9] or the application of pressure[14] decreases the unit-cell volume and suppresses superconductivity, but does not promote magnetic order.

In the absence of additional information, the observation of magnetic Bragg peaks, whose intensities are modulated by the U $5f$ form factor, does not distinguish between the ordering of local moments or a spin-density wave involving the f-like quasiparticles on some region of the Fermi surface. However, taken together with our results, resistivity measurements[6] strongly suggest the presence of a spin-density wave. From their thermodynamic data, Ramirez, Batlogg, Cooper, and Bucher[6] estimated that a fraction ($\rho \sim 0.1$) of the Fermi surface is removed upon passing through T_N. An independent measure[15] of ρ is the ratio of μ_0^2 to the mean-square fluctuating moment μ_{eff}^2 found both in the high-temperature bulk susceptibility[16] and low-temperature neutron-scattering measurements.[17] We find that $\mu_0^2/\mu_{\text{eff}}^2$ is 0.05, which, given the crude nature of the assumptions inherent in such comparisons, is in good agreement with the bulk result.

We have shown that the phase transition at 6.5 K in $U_{0.95}Th_{0.05}Pt_3$ is to an AFM state. The magnetic superlattice is commensurate with the underlying nuclear lattice, and the ordered moment is well below the effective moment deduced from the high-temperature susceptibility data. Measurements of the spin dynamics in the magnetically ordered phase are underway.

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