Magnetic structure of UPd_2Si_2

B. Shemirani, H. Lin,* M. F. Collins, C. V. Stager, and J. D. Garrett
Department of Physics and Institute for Materials Research, McMaster University, Hamilton, Ontario, Canada L8S 4M1

W. J. L. Buyers
AECL Research, Chalk River, Ontario, Canada K0J 1J0
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Elastic-neutron-scattering and magnetization measurements have been performed on a single crystal of UPd_2Si_2 in the 12–160-K temperature range. Two magnetically ordered phases are observed with the magnetic moment parallel to the c axis. At low temperatures ($T<108$ K), there is a simple body-centered-tetragonal antiferromagnet with an ordered moment of $2.3\pm0.3\mu_B$. At higher temperatures (108 K $<T<$ 136 K), there is an incommensurate spin-density wave with wave vector $\mathbf{q}=(0,0,0.732)$ and magnetic moment $2.3\pm0.3\mu_B$ at 120 K. The phase transition at 108 K is first order while there is a critical phase transition at 136 K to the paramagnetic phase with critical index $\beta=0.30\pm0.03$. The low-temperature phase contains a small amount of a $(0,0,\frac{2}{3})$ phase; the maximum intensity from this phase is about 0.8% of that from the simple antiferromagnetic phases at 90 K. The magnetization in low field shows an analogous peak at 90 K.

I. INTRODUCTION

The ternary compounds UT_2Si_2, with T a $3d$, $4d$, or $5d$ transition-metal atom, attract much interest because of their unusual magnetic and transport properties. They exhibit a wide variety of different ground states,[1] ranging from paramagnetism to Kondo spin fluctuations, antiferromagnetism, and superconductivity. Previous studies on polycrystalline UPd_2Si_2 include magnetic-susceptibility measurements[1] and neutron diffraction.[2] The magnetic-susceptibility measurement suggests antiferromagnetic ordering below 97 K, while powder neutron diffraction shows two coexisting magnetic phases. From these measurements the low-temperature phase ($0<T<40$ K) was believed to give rise to simple body-centered-tetragonal antiferromagnetic reflections that coexist with reflections corresponding to a spin-density wave with wave vector $q=\frac{2}{3}$. At a higher-temperature phase ($40<T<150$ K), the body-centered-tetragonal Bragg peaks disappeared, while the spin-density wave peaks with $q=\frac{2}{3}$ remained. The results in this paper do not confirm this picture. Instead, we find a single structure at the lowest temperature from neutron scattering and susceptibility on a high-quality single crystal. There is no phase transition at 40 K. As the temperature increases, a weak (less than 1%) $q=\frac{2}{3}$ magnetic structure grows in a thermally activated manner and reaches a peak at 90 K.

II. EXPERIMENTAL DETAILS

The single crystal was grown by a modified triple-arc Czochralski method.[3] The crystal was 35 mm long and 4 mm in diameter. Neutron-scattering measurements were carried out on the N5 triple-axis spectrometer at the NRU reactor of Chalk River Laboratories. The sample was aligned with the $(h0l)$ plane horizontal and mounted in a closed-cycle refrigerator. Measurements were made between temperatures of 12 and 160 K, controlled to better than 0.1 K. The neutron wavelength was 2.37 Å, produced by a Si(111) reflection in conjunction with a pyrolytic graphite filter. The analyzer crystal was also Si(111). The sample had a mosaic spread of less than 0.25° [full width at half maximum (FWHM)].

The use of a long wavelength improves the accuracy for measuring the position of the Bragg peaks, but increases extinction effects that reduce the intensity of the strong peaks. Therefore our measurement may underestimate the intensity of strong peaks.

The observed Bragg peaks were close to Gaussian in shape and were analyzed by least-squares fitting to Gaussian functions. A correction to the integrated intensity for measurements made with a triple-axis spectrometer in the elastic mode was applied following Cowley and Bates.[4]

Magnetization measurements were undertaken on a small single-crystal sample of mass 428 mg aligned with the field approximately along the c axis. This crystal was grown in the same furnace with the same technique as the crystal used for the neutron-scattering experiments. The magnetic moment was measured with a Quantum Design [superconducting quantum interference device (SQUID)] magnetometer.

III. RESULTS

UPd_2Si_2 belongs to the $ThCr_2Si_2$ crystal structure type with space group $I4/mmm$. The atomic sites are as follows:

U 2(a): $0,0,0,\ \frac{1}{2},\frac{1}{2},\frac{1}{2}$;

Pd 4(d): $\frac{1}{2},0,\frac{1}{4},\ 0,\frac{1}{2},\frac{1}{4},\ \frac{1}{2},0,\frac{3}{4},\ 0,\frac{1}{2},\frac{3}{4}$;

Si 4(e): $0,0,x,\ 0,0,\bar{x},\ \frac{1}{2},\frac{1}{2},\frac{1}{2}+x,\ \frac{1}{2},\frac{1}{2},\frac{1}{2}-x$.

Nuclear Bragg peaks were observed at $h+k+l=2n$, confirming the body-centered nature of the structure. The lattice constants are $a=4.077\pm0.002$ Å and $c=10.046\pm0.001$ Å at 12 K. From the intensities of the weaker nuclear Bragg peaks, the free positional parameter x is fitted to be 0.383 ± 0.002. From x-ray-powder measurements, Ptasiewicz-Bak, Leciejewicz, and Zygmunt[2] find $x=0.3816\pm0.0010$ at room temperature, a result that is in agreement with our low-temperature value within experimental error.

A series of scans were taken along different directions in reciprocal space at fixed temperatures. The scattering is consistent with the premise that only the uranium atoms carry detectable magnetic moments.[5] In the $[00l]$ scans ($l\leq6$), no magnetic peaks were present, indicating that the spins are completely longitudinal. Figure 1 shows $[10l]$ scans at $T=12$, 120, and 160 K. In the low-temperature phase, in addition to the weak nuclear Bragg peak (101), the magnetic Bragg peaks (100) and (102) are seen, corresponding to the simple body-centered-tetragonal antiferromagnet ($h+k+l=2n+1$). At $T=120$ K, magnetic Bragg peaks are observed at (1,0,0.268) and (1,0,1.732); these peaks are described by the scattering wave vector $\mathbf{Q}=\tau\pm\mathbf{q}$, where τ is the scattering wave vector of a nuclear reflection and $\mathbf{q}$ is (0,0,0.732). This scattering corresponds to an incommensurate longitudinal spin-density wave. No harmonics are observed, and so the wave is purely sinusoidal. A similar high-temperature phase has been reported in the isostructural material UNi_2Si_2.[5] At $T=160$ K, the sample is paramagnetic.

In the high-temperature phase, the position of the magnetic Bragg peak is temperature dependent, varying between (0,0,0.71) and (0,0,0.74) as shown in Fig. 2. In the same figure, we have taken data from Lin *et al.*[5] to plot the temperature variation of q in UNi_2Si_2. There is a notable similarity between the two ternary compounds.

The temperature variation of the intensity of the (100) peak and the incommensurate (1,0,0.268) peak is shown in Fig. 3. There is a first-order phase transition at 108 K and a critical phase transition at 136 K. Near the critical phase transition,[6] the intensity of the magnetic Bragg peak varies as $(T_c-T)^{2\beta}$, from which T_c and β are fitted, giving $\beta=0.30\pm0.03$ and $T_c=135.9\pm0.1$ K. This value is somewhat lower than the value 0.35 ± 0.03 reported by Lin *et al.*[5] for UNi_2Si_2, though both these values are consistent with the three-dimensional Ising model prediction of $\beta=0.326$.

Magnetic moments are determined by comparing the intensities of the magnetic Bragg peaks with those of the weaker nuclear Bragg peaks. The structure factor $F(\mathbf{Q})$ of the magnetic Bragg peak $\mathbf{Q}$ is given by

$$F(\mathbf{Q})=\sum_j \mu_j f(\mathbf{Q})\exp(-2\pi i\mathbf{Q}\cdot\mathbf{R}_j)\ ,$$

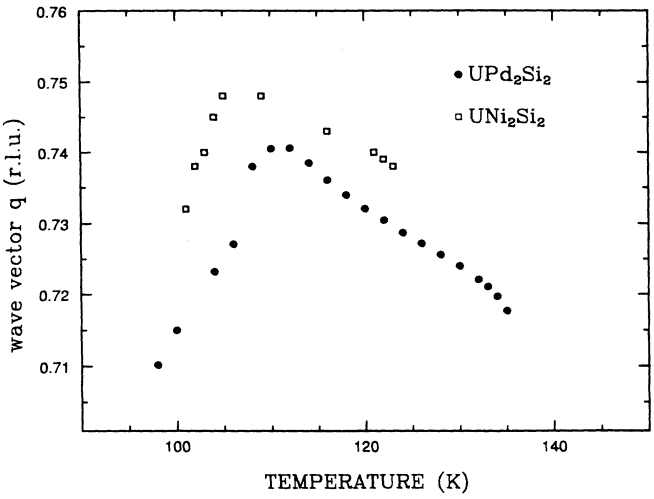

FIG. 2. Temperature variation of the incommensurate wave vector q in UPd_2Si_2 and UNi_2Si_2 as measured from the position of the $(1,0,1-q)$ Bragg peak.

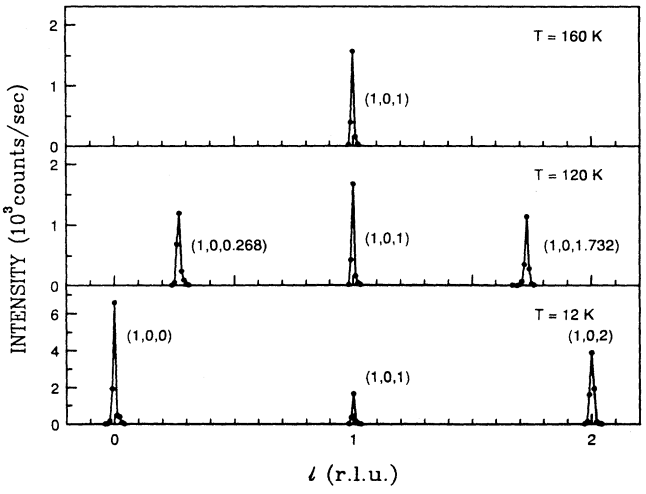

FIG. 1. Elastic scans in the $[10l]$ direction for the paramagnetic phase and the two magnetic phases.

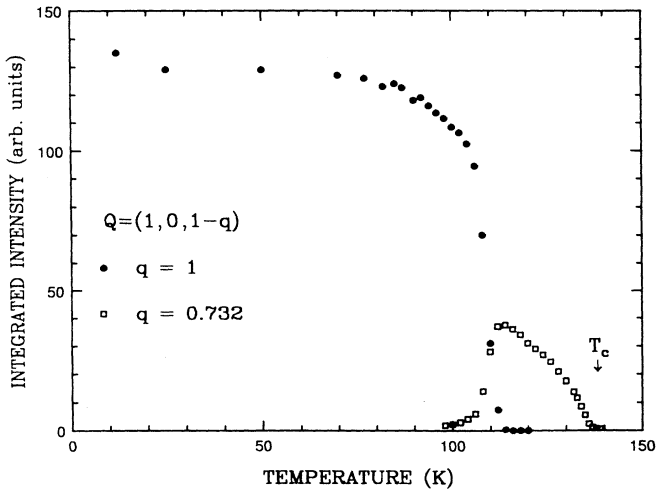

FIG. 3. Integrated intensity of the (1,0,0) and (1,0,0.268) magnetic Bragg peaks as a function of temperature. There is a critical phase transition at 136 K and a first-order phase transition at 108 K.

where μ_j is the magnetic moment on the jth uranium atom at position $\mathbf{R}_j$ in the unit cell. For both structures we can express the moment by

$$\mu_j = \mu_0 \cos(2\pi \mathbf{q}\cdot\mathbf{R}_j) .$$

The magnetic form factors $f(\mathbf{Q})$ are taken from Freeman *et al.*[7] Least-squares analysis gives $\mu_0 = 2.4\mu_B \pm 0.3\mu_B$ at low temperatures and $2.3\mu_B \pm 0.3\mu_B$ for the high-temperature phase at $T = 120$ K.

As well as these strong magnetic Bragg peaks, weak Bragg peaks are seen at intermediate temperature corresponding to $q = \frac{2}{3}$. The integrated intensity of the peak $(1,0,\frac{1}{3})$ is shown in the upper panel of Fig. 4 as a function of temperature. The maximum intensity of this peak, at about 90 K, is only 0.8% of the intensity of the (100) peak, so that the magnetism is predominantly concentrated in the $q = 1$ structure. Below 20 K no observable intensity remains. Thus the ground state is a single commensurate phase corresponding to a simple bipartite antiferromagnet.

The lower panel of Fig. 4 shows the magnetization along c in a field of 10 Oe, plotted as a function of temperature. A sharp peak is seen at 90 K. The peak is at the same temperature as the peak in the $(1,0,\frac{1}{3})$ Bragg peak, though the width of the peak is about a factor of 2 less than the width of the Bragg peak. There is hysteresis in the magnetization, indicating the presence of weak ferromagnetism.

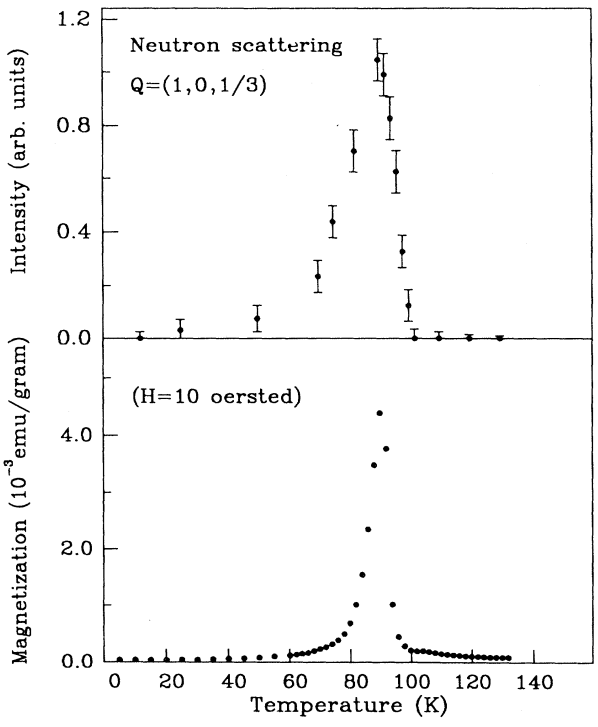

FIG. 4. Upper panel plots the integrated intensity of the $(1,0,\frac{1}{3})$ magnetic Bragg peak as a function of temperature. The intensity scale is the same as that of Fig. 3, and the intensity of the peak shown is at maximum only 0.8% of the intensity of the (1,0,0) peak. The lower panel plots the magnetization in a field of 10 Oe along the c axis against temperature.

IV. DISCUSSION

It is clear that our sample has different magnetic properties to the sample used by Ptasiewicz-Bak, Leciejewicz, and Zygmunt.[2] We do not observe the strong diffraction peaks corresponding to $q = \frac{2}{3}$ that they observe at temperatures below 150 K. For $q = \frac{2}{3}$, we only observe the peaks that are very weak, as shown in Fig. 4. The discrepancy is particularly puzzling in that the crystallographic data (structure, lattice parameters, position parameter x) in the two experiments are in satisfactory agreement. We might point out that a number of compounds of this same type has been grown by one of us (J.D.G.) by the same methods and they have proven to be of high quality.[8,5]

It is interesting to note that Palstra[9] found a susceptibility with a similar temperature dependence to the weak $(1,0,\frac{1}{3})$ peaks that we describe here. He finds hysteresis, indicating a weak ferromagnetism in UPd_2Si_2, just as we do, and a susceptibility that peaks asymmetrically at 97 K. The peak is about a factor of 2 broader than the peak in the $(1,0,\frac{1}{3})$ Bragg peak shown in the upper panel of Fig. 4. The ferromagnetism observed is too weak to be measured directly in the neutron scattering as a change in the intensity of the nuclear Bragg peaks, such as (101). The width of Palstra's susceptibility peak is about 7 times greater than the width of the peak that we observe. We conclude that there is a sample dependence in the amount of this weak coexisting phase, but that our sample and Palstra's have similar qualitative behavior.

If this ferromagnetism corresponds to the squared-off structure found in UNi_2Si_2,[5] where two planes of uranium atoms with moment along z are followed by one plane with moment along $-z$, then the amplitude of the spin-density wave with $q = \frac{2}{3}$ should be 4 times that with $q = 0$. The amplitude of the $q = \frac{2}{3}$ oscillation at 80 K for the data shown in Fig. 4 is $0.23\mu_B \pm 0.04\mu_B$, which is indeed 4 times the moment reported by Palstra.

Palstra[9,1] assigns the peak in the susceptibility of UPd_2Si_2 at 97 K to the Néel temperature. We believe that this peak is associated with the weak squared-up phase rather than with the disordering of the major component of the ordered moments, because susceptibility measurements are more sensitive to even weak ferromagnetic components than to antiferromagnetic components.

The presence of weak Bragg peaks corresponding to $q = \frac{2}{3}$ in the phase with $q = 1$ is unexpected. However, this same coexistence is seen in UNi_2Si_2 in the presence of a magnetic field.[10] There the effect is connected with the presence of an unusual reentrant triple point in the magnetic phase diagram. It is likely that UPd_2Si_2 is reflecting a similar competition between phases.

The competition between competing $q = 1$ and $\frac{2}{3}$ wave vectors in UNi_2Si_2 and UPd_2Si_2 is unusual. The rapid activation like growth with temperature of the $q = \frac{2}{3}$ phase suggests it may be due to an excited state. Thermal excitation across a (001) spin gap in the electronic bands

TABLE I. Magnetic structure of UNi_2Si_2, UPd_2Si_2, UPt_2Si_2, URh_2Si_2, UPd_2Ge_2, and UNi_2Ge_2. There are similarities between the properties of these isostructural materials.

Material	Reference	$q=1$ structure	$q=\frac{2}{3}$ structure	q incommensurate (~ 0.74)	Moment, low T (μ_B)
UNi_2Si_2	5	$53<T<103$	$0<T<53$[a]	$103<T<124$	2.7 ± 0.3
UPd_2Si_2	this work	$T<108$	b	$108<T<136$	2.4 ± 0.3
UPt_2Si_2	11	$T<35$			1.67 ± 0.04
URh_2Si_2	2	$T<137$			1.6
UPd_2Ge_2	2			$T<140$	1.9
UNi_2Ge_2	12	$T<80$			2.35

[a]Traces of this phase persist in the $q=1$ phase.
[b]Traces of this phase exist in the $q=1$ phase, with maximum strength at 90 K and minimum strength at 0 K.

could sense a different nesting wave vector than the ground state, and thus the excited states could develop a different spin gap of $(0,0,\frac{2}{3})$. For example, if temperature populates excited electronic states near $\mathbf{k}=(\pm\frac{1}{2},0,0)$ so that $\mathbf{q}=\mathbf{k}_+-\mathbf{k}_-=(100)$, then minima in the excited band just off the (100) axis at $(\pm\frac{1}{2},0,\pm\frac{1}{6})$ would be sufficient to produce a thermally activated $q=(1,0,\frac{1}{3})\equiv(0,0,\frac{2}{3})$ phase which would coexist with the $q=(001)$ phase. Above 90 K the (001) gap collapses rapidly and destroys the excited-state nesting before the disappearance of the ground-state nesting at 108 K. Because of the presence of more than one ordered wave vector, Landau theory predicts a first-order phase transition, just as we observe.

Table I lists the magnetic properties of the isostructural antiferromagnets UNi_2Si_2, UPd_2Si_2, UPt_2Si_2, URh_2Si_2, UPd_2Ge_2, and UNi_2Ge_2. The low-temperature structure of UPd_2Si_2 with $q=1$ is found in all these materials except UPd_2Ge_2. The high-temperature incommensurate longitudinal spin-density wave structure also occurs in UNi_2Si_2 and UPd_2Ge_2.

It is clear that the compounds listed in Table I are similar in nature. In all cases the magnetism lies predominantly, if not entirely, on the uranium atoms.

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*Present address: Department of Physics, University of Rhode Island, Kingston, RI 02881.

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