Incommensurate Magnetic Order in the Heavy Fermion Superconductor UNi_2Al_3

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Neutron scattering measurements show UNi_2Al_3 to be unique among heavy fermion superconductors in that below $T_N \sim 5.2$ K it displays long range magnetic order which is incommensurate with its chemical lattice. The ordering wave vector is $(\frac{1}{2} \pm \delta, 0, \frac{1}{2})$ where $\delta = 0.110 \pm 0.003$. The measured superlattice intensities are well described by the presence of a longitudinal spin density wave within the hexagonal basal plane, polarized along a^*. The magnitude of the maximum ordered moment is found to be very small; $\mu_{ord} = (0.24 \pm 0.10)\mu_B$.

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The interplay between magnetism and superconductivity is a central feature in the description of several classes of strongly correlated electron systems. The heavy fermion metals, a subset of which exhibits the apparent microscopic coexistence of f-electron magnetism and superconductivity at sufficiently low temperatures, form such a class of materials [1,2].

At present, there exist four such uranium-based compounds which display antiferromagnetic order characterized by small ordered moments. UPt_3 [3] forms a hexagonal-close-packed structure and orders magnetically below $T_N \sim 6$ K. It undergoes at least one and likely two superconducting transitions at $T_C \sim 0.5$ K [4]. URu_2Si_2 forms a body-centered-tetragonal crystal structure and undergoes magnetic and superconducting transitions at 17 and ~ 1.2 K, respectively [5]. A remarkable feature of the magnetism in these two compounds is that the magnitude of the ordered moment is extremely small ($\mu_{ord} \sim 0.02\mu_B$ for UPt_3 [3], and $\mu_{ord} \sim 0.04\mu_B$ for URu_2Si_2 [6]) leading to speculation that these materials in fact display nontrivial ordering phenomena [7] distinct from simple antiferromagnetic ordering of the local magnetic dipoles.

Recently, two new heavy fermion metals which display coexistence of superconductivity and antiferromagnetic order have been discovered. These are the simple hexagonal ternary metals UPd_2Al_3 [8] and UNi_2Al_3 [9] which undergo magnetic phase transitions at $T_N \sim 15$ and ~ 5 K, and superconducting transitions at $T_C \sim 2$ and ~ 1 K, respectively. Considerably less is known about these new compounds, compared with the extensive literature which exists on UPt_3 and URu_2Si_2. Neutron powder diffraction measurements indicate that UPd_2Al_3 [10] orders into a simple, $\mathbf{Q} = (0,0,\frac{1}{2})$, antiferromagnetic structure in which the moments lie ferromagnetically aligned within the basal plane, and the moment direction rotates by π from one basal plane to the next. Recent measurements [11,12] suggest a more complicated magnetic phase diagram. However, all measurements are consistent with an ordered magnetic moment of almost conventional size, $\mu_{ord} \sim 0.85\mu_B$.

In this Letter we report on the first observation of magnetic neutron scattering from UNi_2Al_3 and determine the magnetic structure below T_N. As we will discuss, the magnetic order is characterized by a very weak ordered moment and an incommensurate ordering wave vector. This combination made these experiments very difficult. They required detailed, systematic searches through reciprocal space, as well as the sensitivity that is made possible by the use of diffraction from single crystal samples. Earlier neutron diffraction measurements [10] performed on powder samples of UNi_2Al_3 detected no evidence of magnetic order and placed an upper limit on the size of a possible magnetic moment of $0.2\mu_B$. Previous muon spin rotation (μSR) [13] experiments on UNi_2Al_3 provided evidence for antiferromagnetism with an ordered magnetic moment of the order of $0.1\mu_B$. More recently [14] it has been noted that the zero field μSR signal from the organic, spin density wave system $(TMTSF)_2PF_6$ is similar to that from UNi_2Al_3, suggesting the possibility of an incommensurate spin arrangement in UNi_2Al_3. These new measurements [14] were performed on a crystal cut from the same boule as the crystal which is the subject of the present work.

The UNi_2Al_3 single crystal used in the experiments described here was grown by the Czochralski technique in a tri-arc furnace. The starting materials were of high purity; the same ultrahigh purity uranium used in previous studies of URu_2Si_2 [15], 99.9999% aluminum, and 99.999% nickel. Other details of the growth were similar to those reported previously for uranium intermetallic crystal growth [15]. The crystal was subsequently annealed under argon at 900 °C for one week. Part of this melt was crushed in order to produce a powder for neutron diffraction and susceptibility characterization, while a cylindrical sample of approximate dimensions 6 mm diameter by 25 mm long was extracted and used for the single crystal neutron measurements. The UNi_2Al_3 crystal, belonging to the hexagonal space group $P6/mmm$, displayed lattice parameters at 4.2 K of $a = 5.204$ Å and

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$c = 4.018$ Å, similar to those reported previously [9]. It was comprised of three closely aligned grains, characterized by a mosaic spread of $\sim 1°$.

The powder neutron diffraction measurements were carried out on the C2 High Resolution Powder Diffractometer at Chalk River Laboratories. The powder was set in a vanadium can in the presence of a helium exchange gas and mounted in a pumped ^{4}He cryostat. Measurements taken at temperatures as low as 1.5 K showed no indication of the development of any scattering which could not be assigned a temperature-independent nuclear origin. There was no evidence of additional impurity phases found in this powder sample. Susceptibility measurements made on the powder sample showed behavior very similar to that reported previously [9]; a broad peak near ~ 90 K, a sharp peak near ~ 5 K, with a broad minimum between them at ~ 35 K.

More sensitive single crystal neutron measurements were carried out on the E3 triple axis spectrometer at the Chalk River Laboratories. This sample was mounted in a pumped ^{4}He cryostat with a helium exchange gas present. Its temperature could be varied from room temperature to ~ 1.5 K. The integrated intensities of several weak nuclear Bragg peaks were determined in order to compare to calculated intensities for pure UNi_2Al_3 as a measure of the quality of the single crystal. This comparison is good, and all indications are that the quality of this UNi_2Al_3 sample is very good.

The search for magnetic Bragg scattering as well as measurements of nuclear Bragg peaks for the purpose of magnetic intensity calibration were done with a variety of spectrometer configurations. All measurements were performed with the triple axis spectrometer set for elastic scattering, with a Si(111) monochromator and a pyrolytic graphite (002) analyzer. Data were collected at neutron energies of 8.23 and 3.52 THz with a pyrolytic graphite filter in the scattered beam, and at 4.12 THz without filters. We investigated three high symmetry planes in the single crystal: (h,h,l), $(h,k,0)$, and $(h,0,l)$.

Our observations of magnetic neutron scattering from UNi_2Al_3 are shown in Fig. 1. Figure 1(a) shows portions of a scan along the $(h,0,\frac{1}{2})$ direction at 20 K (open circles), and then at low temperature, 1.8 K (solid circles), both performed with 3.52 THz neutrons. These panels show clear magnetic Bragg peaks arising at $h = 0.39$ and $h = 0.61$ at low temperature. The intensity of these magnetic Bragg peaks is very low, down by 3 orders of magnitude from the intensity of a typical nuclear Bragg peak such as (1,0,0). The width of the magnetic Bragg peaks is that determined by the resolution of the instrument, and the asymmetric shape of some of the peaks is due to the mosaic of our crystal. The magnetic order displayed by UNi_2Al_3 appears to be true long range order, with correlation lengths exceeding 400 Å, in contrast to that reported in both UPt_3 [3] and URu_2Si_2 [15].

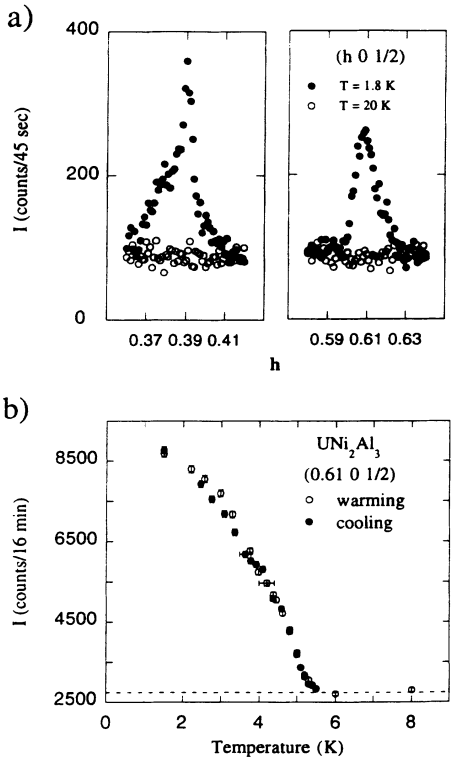

FIG. 1. (a) Sections of an $(h,0,\frac{1}{2})$ scan performed with 3.52 THz neutrons at temperatures above (○) and below (●) $T_N \sim 5.2$ K are shown. The asymmetry in the line shape is due to sample mosaic. (b) The intensity of the $\mathbf{Q} = (0.61,0,\frac{1}{2})$ magnetic Bragg peak is shown as a function of temperature. The dashed line indicates the background scattering level.

The wave vector characterizing this magnetic structure is $\mathbf{Q} = (\frac{1}{2} \pm \delta, 0, \frac{1}{2})$, with $\delta = 0.110 \pm 0.003$. The component of this wave vector within the basal plane is incommensurate, $q_{IC} = \frac{1}{2} \pm \delta$. It can be approximated as $q_{IC} = \frac{1}{2} \pm \frac{1}{9}$, but there is certainly no simple relationship between it and the periodicity of the nuclear lattice. The magnetic structure along the c direction corresponds to a simple antiferromagnetic stacking of the basal plane structure, as in UPd_2Al_3. No additional superlattice Bragg peaks appeared at low temperature in any of the three scattering planes we examined. These planes were carefully examined with both two-dimensional, grid scans, and finer line scans along special high symmetry directions.

Longitudinal scans of the superlattice Bragg peak position $(0.61,0,\frac{1}{2})$ were performed for several temperatures in the range from 2.5 to 6 K. These scans are reasonably described by Gaussian line shapes with temperature-independent peak position and widths. Additional transverse scans of the position of the maxima confirm that the incommensurability is temperature independent to within the tolerance of the measurement, $\Delta\delta = 0.003$. The intensity at this position, and also at $\mathbf{Q} = (0.61,0,\frac{3}{2})$, was

monitored in both warming and cooling runs, over the range $\sim 1.5\ \mathrm{K} < T < 12\ \mathrm{K}$, for the purpose of extracting the intensity of the superlattice Bragg peak as a function of temperature. This is shown in Fig. 1(b) for the $(0.61,0,\frac{1}{2})$ peak. The temperature dependence of the $\mathbf{Q}=(0.61,0,\frac{3}{2})$ superlattice intensity was identical with that at $\mathbf{Q}=(0.61,0,\frac{1}{2})$ to within the precision of the measurement. The onset of the superlattice peak intensity is at ~ 5 K, which corresponds well to the position of bulk anomalies associated with the magnetic phase transition in this compound [9]. As will be discussed, the wave-vector dependence of the intensity of equivalent superlattice peaks is consistent with that expected from magnetic ordering.

The magnetic superlattice intensity decreases monotonically with increasing temperature. However, it does not display the simple linear temperature dependence seen in UPt_3 [3] and URu_2Si_2 [15]. The intensity, shown in Fig. 1(b), can be described by $I \sim (T_N - T)^{0.6}$ down to 2 K, with $T_N = 5.2$ K. To within the sensitivity of these measurements, there is no hysteresis near T_N, consistent with a second order phase transition. Also, it can be seen there is no saturation in the intensity for temperatures as low as 1.5 K. Clearly, it is of interest to extend these measurements to lower temperatures, into the superconducting state.

Therefore UNi_2Al_3 is the first heavy fermion superconductor to display incommensurate magnetic order. Examples of incommensurate magnetic ordering and fluctuations are known in other superconducting materials such as the Chevrel-phase superconductors, exemplified by $HoMo_6S_8$ [16], as well as in the high temperature superconductors [17]. However, there are important differences in each case. The moments in the Chevrel-phase superconductors arise from well-localized $4f$ electrons which interact only weakly with the conduction electrons. Incommensurate fluctuations have been observed in some high temperature superconductors [17], but there is no indication of long range order. The prototypical itinerant, incommensurate spin density wave system is chromium [18]. It is worthwhile to note that, in chromium, both the incommensurability as well as T_N are very sensitive to small levels of impurities. To the extent that this comparison is relevant, we may expect similar sample and impurity dependencies in UNi_2Al_3.

At low temperature, measurements were made in the $(h,0,l)$ plane at several magnetic Bragg peak positions as well as at several relatively weak nuclear positions in order to determine the magnetic structure and to estimate the size of the ordered moment. Comparison of the measured intensities of the magnetic peaks (Fig. 2) shows a strong angular dependence within the $(h,0,l)$ plane that cannot be properly accounted for by a dependence on the form factor, $f(|\mathbf{Q}|)$, alone. Figure 2 displays radial scans through three magnetic Bragg peak positions: $(0.39,0,\frac{1}{2})$, with $|\mathbf{Q}|=0.95$ Å^{-1}, $(0.39,0,\frac{3}{2})$, with $|\mathbf{Q}|=2.41$ Å^{-1}, and $(1.39,0,\frac{1}{2})$, with $|\mathbf{Q}|=2.09$ Å^{-1}. As can be seen most clearly by comparing the intensity at the latter two ordering wave vectors, for which $|\mathbf{Q}|$ is very similar, the intensity correlates strongly with the angle between $\mathbf{Q}$ and the $\mathbf{c}^*$ axis, with stronger intensities when $\mathbf{Q}$ tends to be along $\mathbf{c}^*$. The neutron scattering cross section is sensitive to the component of moment which lies in a plane perpendicular to the scattering wave vector. It preferentially senses moments along $\mathbf{a}^*$ as $\mathbf{Q}$ tends towards the $\mathbf{c}^*$ direction. These results provide strong evidence, therefore, for the magnetic moment direction being predominantly along $\mathbf{a}^*$.

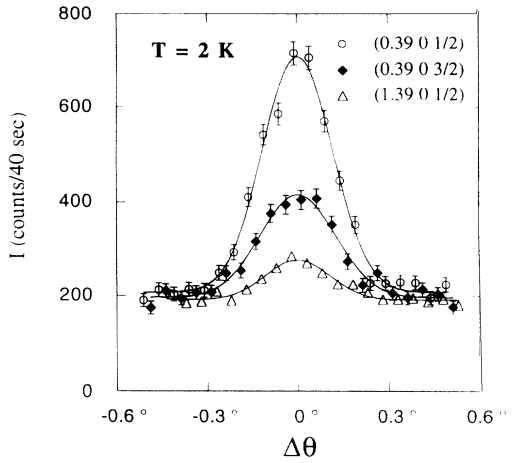

FIG. 2. Radial scans (plotted vs sample angle) of three magnetic Bragg peaks at $T=2$ K are shown. These are $\mathbf{Q}=(0.39,0,\frac{1}{2})$, $|\mathbf{Q}|=0.95$ Å^{-1}, $\alpha=34.8°$; $\mathbf{Q}=(0.39,0,\frac{3}{2})$, $|\mathbf{Q}|=2.41$ Å^{-1}, $\alpha=13.0°$; and $\mathbf{Q}=(1.39,0,\frac{1}{2})$, $|\mathbf{Q}|=2.09$ Å^{-1}, $\alpha=68.0°$. α refers to the angle between $\mathbf{Q}$ and the $\mathbf{c}^*$ axis.

We have modeled the integrated intensities of the magnetic Bragg peaks according to

$$I(\mathbf{Q}) = \frac{F(\mathbf{Q})^2}{\sin(2\theta)} \sin^2(v) f^2(Q) ,$$

where $F(\mathbf{Q})$ is the structure amplitude for the Bragg peak, v is the angle between $\mathbf{Q}$ and the moment direction, and $f^2(Q)$ is the magnetic form factor. Both $F(\mathbf{Q})$ and v depend on the model for the ordered magnetic structure. The magnetic form factor $f^2(Q)$ was described by a form which had previously been used to account for magnetic properties in several uranium based compounds [19].

The intensities of six magnetic Bragg peaks, corrected for the Lorentz, $[\sin(2\theta)]^{-1}$, factor and magnetic form factor are shown in Fig. 3 as a function of the angle between $\mathbf{Q}$ and the $\mathbf{c}^*$ direction, α. As previously discussed, all Bragg peaks decrease in intensity with increasing α. We can specifically exclude models in which the moment is predominantly aligned along $\mathbf{c}^*$, as had been previously suggested [13].

Several simple models for the magnetic structure were examined as candidates to describe our results. The one

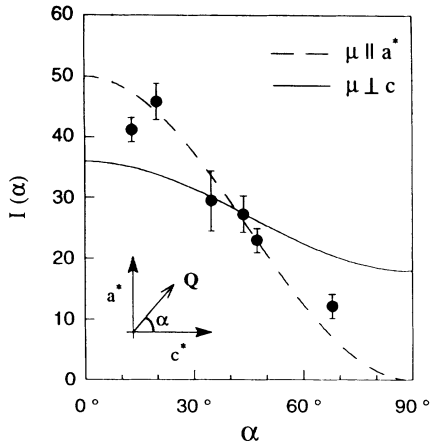

FIG. 3. Corrected (see text) intensity of six magnetic Bragg peaks is plotted vs α, the angle between **Q** and the **c*** axis. The lines are predictions from models for the incommensurate magnetic order with differing polarizations within the basal plane. Shown are predictions for a longitudinal spin density wave structure within the basal plane, in which $\mu \| \mathbf{a}^*$, and also for a planar modulation of the moments perpendicular to **c**.

which best describes these measurements is that of a longitudinal spin density wave (LSDW) within the hexagonal basal plane, in which the moments are parallel to **a***. The full, three dimensional magnetic structure is then formed by a simple antiferromagnetic stacking of this basal plane structure along **c**. The α dependence predicted by this model is shown as the broken line in Fig. 3. This figure compares the observed magnetic intensities corrected for all dependencies other than that on α, with that calculated for two model magnetic structures. A structure in which the moment direction is modulated within the basal plane produces a less-pronounced α dependence, as is shown by the sold line in Fig. 3.

As can be seen in Fig. 3, the LSDW polarized along **a*** provides a very good description of our results. Models in which the moments are polarized along a different basal plane direction, and therefore not longitudinally polarized to the incommensurate component of the ordering wave vector, display less pronounced α dependencies. The key to understanding these dependencies is the fact that measurements at a single ordering wave vector probe only a single domain of any such single-q structure. However, assuming that all possible domains of such structures are equally populated, it is very difficult to distinguish such single-q LSDW structures from multiple-q modulated magnetic structures, as is known, for example, to describe the magnetic structure of neodymium [20].

Comparison of the magnetic Bragg peak intensities to those of the weaker nuclear Bragg peaks allows us to estimate the size of the ordered moment. The weak nuclear Bragg peaks are used in this comparison as they are less likely to be affected by extinction. However, as the role of extinction in these peaks is not completely understood, our estimate of the ordered magnetic moment represents an upper limit. The size of the ordered moment determined from our data depends weakly on the model employed. The model corresponding to a single domain of the LSDW structure with $\mu \| \mathbf{a}^*$, results in $\mu_{\text{ord}} = 0.14 \mu_B$. If, as is physically much more likely, the three possible domains are equally populated, we obtain $\mu_{\text{ord}} = 0.24 \mu_B$. Taken together with the uncertainty from the direct comparison to the nuclear peaks, these model calculations allow us to estimate the magnitude of the ordered moment at $\mu_{\text{ord}} = (0.24 \pm 0.10) \mu_B$. Within this LSDW structure, this ordered moment represents the maximum magnitude of the moment, which is modulated from site to site.

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