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Neutron studies of the magnetic phase transition in UAs

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ABSTRACT

Uranium arsenide undergoes a first order phase transition to a type I antiferromagnetic (AF-I) structure at $T_N \sim 124$ K. Above T_N , UAs exhibits strongly anisotropic critical scattering suggestive of pseudo two-dimensional behavior. The temperature dependence suggests a tendency to order with an incommensurate wave vector but then suddenly orders with the AF-I structure. A mean-field theory is proposed based on an anisotropic Hamiltonian and provides a reasonable description of the results.

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The uranium monpnictides UX ($X = N, P, As, Sb, \text{ and } Bi$) crystallize in the rock-salt structure and form a family of antiferromagnetic compounds in which the Néel temperature and ordered moments steadily increase from UN (53 K and $0.75 \mu_B$) to UBi (285 K and $0.3 \mu_B$). In their ordered state, all compounds exhibit the type I antiferromagnetic structure in which ferromagnetic (001) planes are stacked antiferromagnetically into a (+ - + -) sequence along [001]. The moment direction is parallel to [001] and therefore perpendicular to the ferromagnetically coupled (001) planes. In UAs, $T_N \sim 126$ K and an additional transition occurs at $T_N \sim 0.5 T_N$ to a structure where the moment direction remains the same, but the stacking of the sheets changes to (+ + - -) (1,2). In this paper, we describe the critical scattering associated with T_N (3). An unusual shape of the critical scattering is observed and it can be explained with a mean-field treatment where a Hamiltonian is defined which explicitly accounts for the cubic anisotropy. The results show that UAs at high temperatures is near a Lifshitz point at which commensurate and incommensurate phases are in equilibrium with the paramagnetic phase (4).

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The neutron scattering experiments were performed both at the Argonne CP-5 reactor and the Brookhaven High-Flux Beam reactor. No energy widths were observed with the best resolution used: $\delta E = 0.4$ meV. Measurements were made on single crystals and polycrystalline samples.

In UAs, as in USb (5), no (001) magnetic Bragg peak is observed which implies that the spins are parallel to the [001] direction. The temperature dependence of the (110) type-I magnetic reflection is given in Fig. 1. In this crystal, $T_N \sim 127$ K and the transition is first order. The ordered moment at 78 K is $1.93 \mu_B/\text{U atom}$. At the transition, there is an abrupt change in intensity such that 70% of the total moment is achieved by $T = T_N - 0.1$ K.

The study of the critical scattering above T_N revealed an interesting behavior. Figure 2 shows scans parallel to the moment direction ($q_{\parallel}$) [001] and Fig. 3 shows scans perpendicular ($q_{\perp}$) to [001]. The distribution of intensity is schematically shown in the top part of Fig. 2. It is cigar-like with $q_{\parallel} \gg q_{\perp}$, which is suggestive of two-dimensional behavior. It peaks near $q = 0.7 c^*$ as measured from the (111) zone center. The temperature dependence is seen in Fig. 2. As T is lowered towards $T_N = 123.50$, the divergence of intensity at $q = 0.7 c^*$ suggests that the system would like to order at the incommensurate wave vector $q = 0.7 c^*$, but at T_N , the system suddenly orders into the type I structure with $q = 1.0 c^*$ ($\eta = 0$ in Fig. 2).

These results can be understood by defining the following Hamiltonian:

$$\mathcal{H} = - \sum_{i,j,\alpha} J^{\alpha\alpha}(R_{ij}) S_i^{\alpha} S_j^{\alpha} - v \sum_{i,\alpha} (S_i^{\alpha})^4 \quad (1)$$

where $J^{\alpha\alpha}(R_{ij})$ is the anisotropic exchange interaction which transforms with R_{ij} according to the full cubic symmetry, S_i^{α} is the α component of the spin operator for the i^{th} ion, and v is a measure of the single ion anisotropy. The exchange interactions between the z components of the spins are J_0 (nearest neighbor-nn) and J_2 (next nearest neighbor-nnn); and between the x and y components are J_1 (nn) and J_2 (nnn). The mean-field calculation of the susceptibility yields

$$\chi^{zz}(\vec{q}) = a_1 \left\{ 1 + [a_2 - a_3 \cos(\frac{1}{2} a q_{\parallel})] \frac{a^2 q^2}{4} + a_4 [1 - \cos(a q_{\parallel})] - 4a_3 [1 - \cos(\frac{1}{2} a q_{\parallel})] \right\}^{-1} \quad (2)$$

$$\text{with } a_1 = \frac{C}{T-T_0}, \quad a_2 = \frac{2CV}{(g\mu_B)^2} \frac{J_0+2J_3}{T-T_0},$$

$$a_3 = \frac{2CV}{(g\mu_B)^2} \frac{J_1}{T-T_0}, \quad \text{and } a_4 = \frac{2CV}{(g\mu_B)^2} \frac{J_2}{T-T_0}. \quad C \text{ is the}$$

Curie constant and V is the volume. T_0 is the mean-field ordering temperature.

The observed neutron intensity is proportional to $\chi^{zz}(\vec{q})$ and the curves in Figs. 2 and 3 are fits to the data obtained by varying T_0 and the exchange parameters. $T_0 \approx 116$ K which is less than T_N indicative of the first order nature of the phase transition. The ratio $J_0/J_1 = 37.4$ which states that the zz exchange is much larger than the xx or yy exchange interactions between the spins. This large cubic anisotropy is responsible for the diffuse scattering being elongated along q and may be interpreted as arising from the bonding effects between the f orbitals on the U atoms and p orbitals of the anions in the (001) sheets (5).

The ratio J_1/J_2 varies between 0.62 and 0.69 as compared to the value 1.0 as the condition for a Lifshitz point. The tendency to antiferromagnetic ordering due to the z - z coupling between adjacent (001) planes of spins is opposed by the z - z coupling between nnn (001) planes. This almost cancellation of effects in UAs leads to "frustration" and results in the commensurate and incommensurate phases being in equilibrium with the paramagnetic phase: the definition of a Lifshitz point. The first order nature of the transition also arises due to complications in being in the vicinity of a Lifshitz point.

The process of magnetic ordering in UAs is closely analogous to the case of first order structural phase transitions occurring in some perovskite compounds (6,7). Interestingly enough, similar diffuse scattering has been observed in the study of magnetite near the Verwey transition (8).

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Fig. 1. Integrated intensity of the (110) magnetic reflection as a function of temperature. Closed points are taken in cooling, open in heating.

Fig. 2. The upper portion shows a schematic of the diffuse scattering intensity around (110). The lower section shows the intensity of the scattering along $[00\eta]$ from (110) as a function of temperature. The resolution function is shown by the horizontal bar. The solid lines are fits to the data described in the text.

Fig. 3. Observed (points) and calculated (full lines) diffuse intensity in different directions around the (110) at 124.75 ($T_N + 1.25$ K). The horizontal bars indicate the width of the resolution function.