Studies of the Phase Transitions in UAs with Neutron Scattering

CONF-800417--3

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Abstract**MASTER**

Uranium arsenide is known to order with the type-I antiferromagnetic (AF) structure at ~ 126 K, and exhibit a first-order transition to the type-IA AF structure at $T_N/2$. We have now reexamined these transitions with a single crystal. Above T_N UAs exhibits critical scattering suggesting a tendency to order with an incommensurate wavevector, but then suddenly orders with the AF-I structure. The analysis of the data shows the need to consider anisotropic exchange interactions of cubic symmetry between U moments.

*Work supported by the U. S. Department of Energy, under Contract No. DE-AC02-76CH00016.

Paper presented at the "Symposium on the Physics of Actinides and Related 4f Materials", Zurich, 9-11 April 1980.

To be published in Physica B.

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Introduction

From studies of the critical scattering in UN [1] and USb [2] these materials have been shown to exhibit strong anisotropic magnetic interactions despite their cubic NaCl crystal structure. Uranium arsenide undergoes a paramagnetic $\rightarrow$ antiferromagnetic transition (type I structure) at ~ 126 K and at lower temperature ($\sim 1/2 T_N$) exhibits a first-order transition to the type-IA structure [3]. These structures can be characterized by (001) ferromagnetic sheets stacked in a certain sequence. The moment direction is perpendicular to the sheets, i.e., parallel to the propagation direction. The type-I structure consists of alternating sheets $+-+-$, and has $\vec{q} = 2\pi/a (0,0,1)$, where $a = 5.78 \text{ \AA}$ is the lattice parameter. The IA structure $(++--)$ has $\vec{q} = 2\pi(0,0,0.5)$. The object of these studies is to characterize the competing exchange interactions.

Experiment and Results

Experiments were performed at both the Argonne CP-5 and Brookhaven High-Flux Beam Reactors using 3-axis neutron spectrometers and an incident neutron energy of 13.5 meV ($\lambda = 2.46 \text{ \AA}$). Both monochromator and analyzer crystals were graphite (002) and a pyrolytic graphite filter was used to reduce higher-order contamination. The single crystal used had a volume of 0.034 cm^3 and was oriented with the scattering vector in the $(1\bar{1}0)$ plane. The study of the diffuse scattering was carried out both as a function of momentum and energy transfer, but in the latter case no inelasticity was detected within the instrumental resolution ($\Delta E < 0.4 \text{ meV}$).

To visualize the experimental information obtained we show a schematic 3-dimensional diagram in Fig. 1. The point at the origin is $\vec{Q} = 2\pi/a(1,1,0)$,

where $\vec{Q}$ is the momentum transfer, and ordering at this point corresponds to an ordered wavevector $\vec{q}_0$ given by $\vec{Q} = \vec{\tau} + \vec{q}_0$, where $\vec{\tau}$ is the reciprocal lattice point of the NaCl lattice. In this case $\vec{\tau} = 2\pi/a(1,1,1)$ and $\vec{q}_0 = 2\pi/a(0,0,1)$, defining the type I structure. Figure 1 shows contours of intensity at a given temperature, $T_N + 1.2$ K. First notice the intensity in the $[00\eta]$ direction. This peaks at $\eta = 0.3$, but has a long tail extending to $\eta = 0$. In the $[\xi\xi 0]$ direction the contours have their smallest full-width at half maximum (FWHM) for the peak along η (i.e., $\eta \sim 0.3$) and are more extended as the peak intensity, at $\xi = 0$, decreases. (Note the different scales in the ξ and η directions in Fig. 1). The instrumental resolution functions are $\Delta\xi_r = 0.012$ and $\Delta\eta_r = 0.005$ units.

The temperature dependence of the scattering along η (with $\xi = 0$) is shown in Fig. 2. These scans suggest the material will order with a wavevector $\vec{q} \sim 2\pi/a(0,0,0.7)$, i.e., with an incommensurate sine-wave modulation of the moments. This is not the case. At $T = 123.3$ K the intensity at $[1,1,0]$ suddenly starts to increase (Fig. 2) and within 0.2 K a sharp Bragg peak appears at $[1,1,0]$ indicating long-range order of the first kind ($q = 1$). The transition may be characterized as weakly first order since 70% of the ordered moment appears by $T_N - 0.1$ K. Simultaneously the scattering at $\eta = 0.3$ rapidly disappears. A further important observation is that, as for USb [2], the diffuse scattering corresponds to fluctuations of the z component of the spin for $\vec{q} \parallel [001]$, with corresponding permutations for $\vec{q}$ along the other cube axes.

Discussion

The anisotropy in the critical scattering arises because correlations in the (001) planes are strong, leading to a relatively sharp ridge around $\xi = 0$, whereas correlations between the planes are weak, leading to a diffuse peak around $\eta = 0.3$. To analyze these results we use a Hamiltonian in which the exchange interactions between the z components of the moments are J_0 (nearest neighbor) and J_2 (next nearest neighbor) and the corresponding interactions between the x and y components are J_1 and J_3 . A mean-field calculation of the susceptibility then yields:

$$\chi^{zz}(q) = a_1 [1 + (a_2 - a_3 \cos aq_{||}/2) a^2 q_{\perp}^2 / 4 + a_4 (1 - \cos aq_{||}) - 4a_3 (1 - \cos aq_{||}/2)]^{-1}$$

where $a_1 = C/(T - T'_N)$, $a_2 = D(J_0 + 2J_3)/(T - T'_N)$,

$$a_3 = DJ_1/(T - T'_N), a_4 = DJ_2/(T - T'_N),$$

$D = 2CV/g_J^2 \mu_B^2$, C is proportional to the Curie constant and V is the crystal volume. The components $q_{\perp}$ and $q_{||}$ of the wavevector $(\vec{q} - \vec{q}_0)$ are in the directions normal and parallel to the z direction, respectively. T'_N represents an effective ordering temperature.

The solid lines in Fig. 2 are the results of a fit to the data and clearly reproduce the data qualitatively. The resulting parameters show that the z - z exchange interaction within the (001) planes is ~ 40 as strong as the x - x or y - y coupling between the spins. Moreover, the tendency to antiferromagnetic ordering due to the z - z coupling between adjacent

(001) planes of spins, as given by J_1' (which is -ve or AF) is opposed by the $z - z$ coupling between next-nearest (001) planes of spins given by J_2 (which is -ve and almost equal to J_1). Thus the system experiences "frustration" in deciding between AF-I or a sinusoidal spin arrangement [4]. The critical behavior of a system with cubic anisotropy in the vicinity of a Lifshitz point has been discussed by Bruce and Aharony [5], who show that such systems in general exhibit 1st order transitions unless an external stress is applied.

As is the case of the other U pnictides, the strong cubic anisotropy is interpreted as arising from bonding effects between the $5f$ wavefunctions and the anion p orbitals.

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Figure Captions

Fig. 1. Schematic representation of the diffuse scattering in UAs at

$T_N + 1.2$ K.

Fig. 2. Temperature dependence of the intensity along $[11\eta]$ near

T_N ($= 123.5$ K).

Fig. 1. Schematic representation of the diffuse scattering in UAs at $T_N + 1.2 \text{ K}$.

Fig. 2. Temperature dependence of the intensity along [11n] near T_N (≈ 123.5 K).