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NEUTRON DIFFRACTION STUDY OF THE MAGNETIC ORDERING IN $EuAs_3$

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The magnetic structure of one of the two ordered phases has been determined at 5 K. The magnetic structure is found to be antiferromagnetic with a cell doubling in the *c* direction of the monoclinic nuclear cell. The centering in the nuclear cell is replaced by anticentering. The collinear magnetic moments are parallel to the *b* axis. The magnetic moment per Eu atom has been found to be $5.74(6)\mu_B$ which leads to the saturation magnetic moment of $6.5\mu_B$.

1. Introduction

Specific heat, electrical conductivity and susceptibility measurements in $EuAs_3$ [1,2] indicate the presence of two successive phase transitions at 10 and 11 K supposed to be associated with magnetic ordering. The Mössbauer measurements yield a single hyperfine field for all Eu ions below 10 K and the intermediate phase between 10 to 11 K is characterized by two different hyperfine fields [3]. Magnetization measurements indicate antiferromagnetism in both ordered phases at low magnetic fields [3]. We have carried out neutron diffraction measurements for identifying the ordered phases unambiguously and to determine the magnetic structure. We shall describe and discuss in this short communication results of our investigations of the antiferromagnetic phase (AF1) which is stable below 10 K.

2. Experimental

Neutron diffraction measurements have been carried out with the diffractometer D15 situated at the high flux reactor of the Institute Laue–Langevin, Grenoble. A single crystal of the size of $1 \times 1 \times 5$ mm^3 with the longest dimension along the b axis was used for the measurement. Neutror wavelength employed was 1.175 Å. 684 reflexions were collected at 5 K which gave rise to symmetry independent 137 nuclear and 139 magnetic reflexions. We collected data from a second crystal of the size $1 \times 1 \times 5$ mm^3 with the longest dimension along the *a* axis. 22 nuclear and 18 magnetic reflexions were collected from this crystal.

3. Structure of the antiferromagnetic phase (AF1)

The antiferromagnetic phase in $EuAs_3$ which is stable below 10 K will henceforth be called AF1. It has been found that magnetic reflexions appear below 10 K at the reciprocal points corresponding to the doubling of the *c* axis of the monoclinic nuclear cell ($a = 9.43(2)$ Å, $b = 7.50(1)$ Å, $c = 5.75(1)$ Å, $\beta = 112.48(5)$ at 5 K). When indexed on this doubled cell the magnetic reflexions follow the rule $h + k = 2n + 1$ and $l = 2n + 1$, whereas the nuclear reflexions have $h + k = 2n$ and $l = 2n$. No

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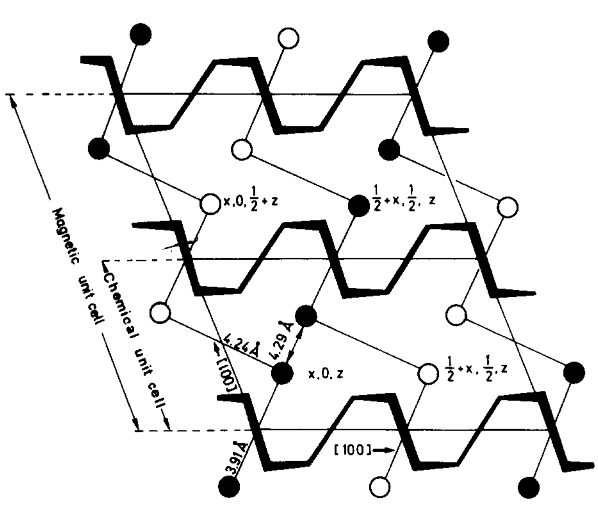

Fig. 1. The magnetic structure of the antiferromagnetic phase of $EuAs_3$ stable below 10 K. The black and white circles represent Eu atoms with opposite moment directions which are parallel to the b axis. The As layers are indicated schematically.

reflexions with other combinations of indices were found. The relative intensities of the magnetic reflexions show that Eu atoms related by the centre of symmetry have parallel moments. This, together with the rule established above, leads to the collinear magnetic structure model of fig. 1. The nuclear structure was refined using nuclear reflexions. This enabled us to determine the scale factor of the magnetic reflexions. Now assuming an arbitrary moment orientation with respect to the three crystallographic axes and an arbitrary magnetic moment we have refined four parameters corresponding to the magnetic moment and its three components. It has been found that the magnetic moments are almost entirely oriented along the b axis of the crystal. The magnetic form factor used has been obtained by Freeman and Desclaux [4] from relativistic Dirac–Fock wavefunctions for Eu^{2+} ion. The refined parameters are given in table 1.

Table 1
Refined parameters of the magnetic structure of $EuAs_3$. $\bar{\mu}$ is the average magnetic moment per Eu atom and $\cos\alpha$, $\cos\beta$ and $\cos\gamma$ are the direction cosines of the moment direction with respect to the crystallographic axes

	Crystal 1	Crystal 1 absorption correction	Crystal 2
$\bar{\mu}(\mu_B)$	5.68	5.43(6)	6.05
$\cos\alpha$	−0.01	−0.01(2)	0.06
$\cos\beta$	0.99	0.99(1)	0.99
$\cos\gamma$	0.14	0.14(3)	−0.11
$\bar{\mu}(\mu_B)$ (saturation value)	6.30	6.03	6.90

4. Magnetic phase transition at 10 K

We have determined the temperature variation of the intensities of a few magnetic reflexions near the phase transition temperature 10 K. Fig. 2 shows the variation of intensities of $-10\frac{1}{2}$ magnetic reflexion. The square of the Brillouin function $B(J)$ corresponding to $J = 7/2$ has been plotted in the same figure. It is seen that the intensity variation does not follow the square of the Brillouin function. This is not surprising and similar behavior has been observed in other systems undergoing antiferromagnetic ordering. In our system, however, extra complications arise because of the presence of another intermediate ordered phase between 10 to 11 K. We had tried next to fit these data with $F^2 = K(T_N - T)^{\beta}$. This fits rather well yielding $\beta = 0.31$.

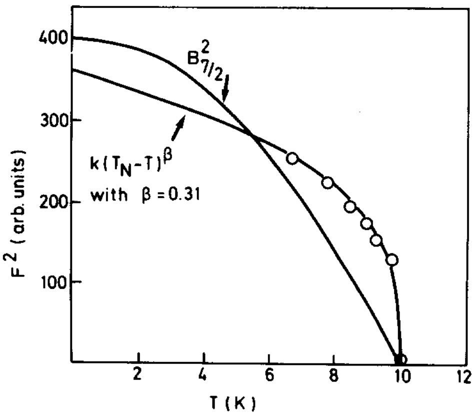

Fig. 2. The variation of the intensity of $-10\frac{1}{2}$ magnetic reflexion with temperature. The critical exponent $K(T_N - T)^{\beta}$ has been fitted to the data yielding $\beta = 0.31$. The square of the Brillouin function $B^2_{7/2}$ does not fit the data.

5. Magnetic moment of Eu atoms

The magnetic moments per Eu atom obtained at 5 K from the refinement of the magnetic structure is $5.7(1)\mu_B$. One expects a saturation magnetic moment corresponding to Eu^{2+} in the ground state $^8S_{7/2}$ to be $7\mu_B$. However, as is evident from fig. 2, the intensities of the magnetic reflexions do not attain saturation value at 5 K; this means that at 5 K magnetic moments are not completely ordered. By extrapolating the intensity variation curve by using the function $F^2 = K(T_N - T)^\beta$, we have determined the saturation value of this magnetic moment to be $6.5\mu_B$ compared to the expected $7\mu_B$. The reason for this small discrepancy is the zero point spin deviation or the presence of Eu^{3+}.

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