Journal of Magnetism and Magnetic Materials 177–181 (1998) 1058–1060

Incommensurate–commensurate lock-in phase transition in $EuAs_3$

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Abstract

We have investigated the incommensurate–commensurate lock-in phase transition in $EuAs_3$ by the relatively new resonant X-ray magnetic scattering technique. Strong resonance enhancements of the magnetic scattering have been observed at the L_{II} and L_{III} absorption edges of Eu. The intensity of the $1, 0, \frac{5}{2}$ magnetic reflection of the commensurate phase decreases continuously with increasing temperature and at $T_L = 10.3$ K drops abruptly to zero. Satellite reflections characteristic of the incommensurate phase appear at $T \geqslant T_L$. By using the excellent Q-resolution of the X-ray magnetic scattering experiment we have determined the temperature variation of both x and z components of the propagation vector of the incommensurate sine-wave phase stable in the temperature range $10.3\ \text{K} \leqslant T \leqslant 11$ K. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Incommensurate phase; X-ray magnetic scattering

Incommensurate magnetic structures can be described as long-period modulated structures with periodicities which are in general incommensurate with that of the crystal lattice (see, e.g., Ref. [1]). These can be mainly of two types, sine-wave or helical and are generally stabilized in a narrow temperature range below the ordering temperature T_N or T_C. As the temperature is lowered the propagation vector $\boldsymbol{k} + \boldsymbol{\delta}$ of the incommensurate structure often changes continuously or discontinuously and at some temperature T_L locks into the commensurate value $\boldsymbol{k}$.

Semimetallic $EuAs_3$ is a model incommensurate magnetic system which has been investigated by neutron diffraction [2]. $EuAs_3$ crystallizes [3] in the monoclinic space group C2/m and is isostructural with $BaAs_3$ with four formula units per C-face centered unit cell with lattice parameters $a = 9.471 \pm 0.002$ Å, $b = 7.598 \pm 0.002$ Å, $c = 5.778 \pm 0.002$ Å, $\beta = 112.53° \pm 0.05°$ at $T = 295$ K. The crystal structure consists of Eu^{2+} ions sandwiched in between quasi two-dimensional puckered layers of As_3^{2-} polyanions. Eu^{2+} ions are in a spherically symmetric $^8S_{7/2}$ ground state with no orbital moment. Although one expects very simple magnetic properties for such a system, the actual magnetic behaviour of $EuAs_3$ is rather complicated presumably due to the p–f hybridization effects in this semimetallic system. $EuAs_3$ orders at $T_N \approx 11$ K to an incommensurate sine-wave phase with the propagation vector $\boldsymbol{k} = (-1, 0, \frac{1}{2} - \delta)$ where δ decreases with temperature and becomes zero at $T_L = 10.3$ K. The temperature variation of δ was determined by neutron diffraction [2]. Although the neutron diffraction results suggested the deviation from unity of the component parallel to $\boldsymbol{a}^*$, its temperature variation could not be measured due to the limited Q resolution of the neutron diffraction experiment. By exploiting the excellent Q resolution of the X-ray magnetic scattering we have been successful in measuring the temperature dependence of the components of the propagation vector parallel to both $\boldsymbol{a}^*$ and $\boldsymbol{b}^*$. The propagation vector was found to be $\boldsymbol{k} = (-1 + \delta_x, 0, \frac{1}{2} - \delta_z)$.

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PII S0304-8853(97)00797-X

A single crystal of $EuAs_3$ of size of about $1 \times 2 \times 5$ mm^3 with its longest dimension parallel to the monoclinic b axis was used for the X-ray diffraction experiment on the wiggler beam line W1 at the HASYLAB, DESY. The crystal was fixed to the cold tip of the specially designed helium cryostat of the four-circle diffractometer.

The magnetic reflections from the commensurate magnetic phase were readily observed when the X-ray energy E was close to the L_{II} and L_{III} absorption edges of europium. The magnetic reflections were also observed when the X-ray energy was away from these absorption edges. We have measured the resonant enhancements of the X-ray magnetic scattering and also the fluorescence scattering at both these edges which agree well with the previous measurements [4] at NSLS, Brookhaven. Fig. 1 shows the temperature variation of the integrated intensity of the $1, 0, \frac{5}{2}$ magnetic reflection ($E = 7.619$ keV) from the commensurate phase and that of two corresponding satellites from the incommensurate phase close to the lock-in phase transition. The inset shows the same in a larger temperature range. The intensity of the $1, 0, \frac{5}{2}$ magnetic reflection from the commensurate phase decreases with increasing temperature and suddenly drops to zero at the commensurate–incommensurate lock-in phase transition $T_L \approx 10.3$ K. The satellite pair appears at $(Q_x, 0, Q_z) = (1 \mp \delta_x, 0, \frac{5}{2} \pm \delta_z)$ above the lock-in temperature T_L. We have measured the intensity and position of this satellite pair as functions of temperature. The temperature variation of the intensities of the satellite pair is also shown in Fig. 1. Fig. 2 shows the temperature variation of the $Q_x = 1 \mp \delta_x$ and $Q_z = \frac{5}{2} \pm \delta_z$. The Q_x and Q_z or δ_x and δ_z are related almost linearly as shown in Fig. 3. However, s-like deviation of the curve from linearity is evident from the inset of Fig. 3 which shows a plot of ΔQ_x vs. Q_z. ΔQ_x is calculated by subtracting the extrapolated values (obtained from the linear fit) of Q_x from the measured values. The satellite reflections move in such a way that not only does the distance between them in Q-space increase with increasing temperature but also the reciprocal vector joining them changes its orientation with temperature. This is not unexpected because in the monoclinic symmetry all directions in the $h0l$ reciprocal plane is equivalent. In the neutron diffraction investigation [2] the temperature variation of only the δ_z was measured. The accuracy of the present X-ray diffraction measurement of the temperature

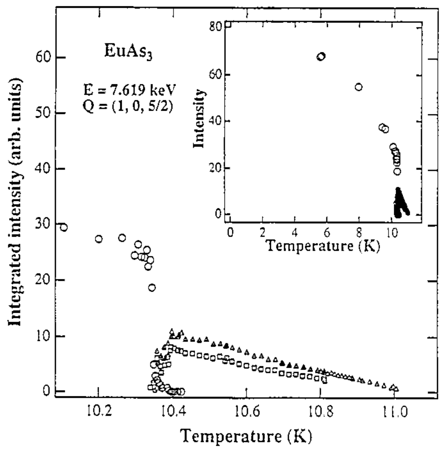

Fig. 1. Temperature variation of the intensity of the $1, 0, \frac{5}{2}$ magnetic reflection from the commensurate phase (circles) and that of two corresponding satellites (triangles and squares) from the incommensurate phase close to the lock-in phase transition. The inset shows the same in a larger temperature range.

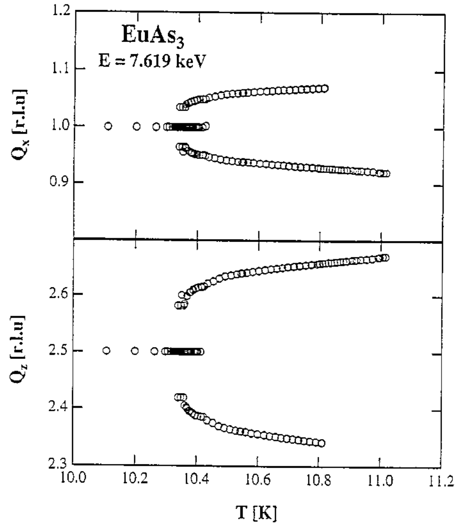

Fig. 2. Temperature variation of the Q_x and Q_z of the satellite pair around $1, 0, \frac{5}{2}$.

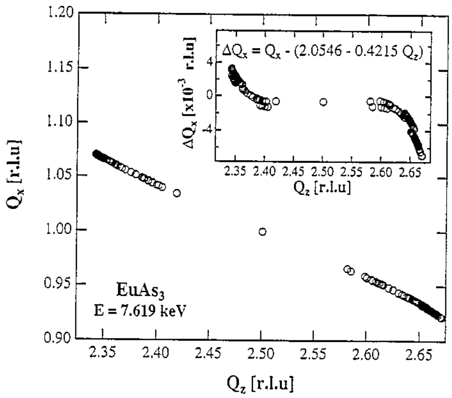

Fig. 3. The plot of Q_x vs Q_z which is approximately linear. The inset shows the plot ΔQ_x vs. Q_z illustrating the deviation from linearity. ΔQ_x is calculated by subtracting from the measured values of Q_x the extrapolated values obtained from the linear fit.

variation of the modulation vector is far superior than those of neutron diffraction investigation. This is due to about an order of magnitude better Q-resolution of the X-ray diffraction experiment.

The present experimental results (Fig. 2) shows that although the temperature variation of the modulation vector is continuous, the lock-in phase transition is ultimately of the first order. The first-order character of the lock-in phase transition is evidenced by the coexistence of both the commensurate and incommensurate phases. We have, however, observed no hysteresis in the temperature variation of the wave vector within our temperature resolution which was better than ± 0.005K. Neither have we observed higher order satellite reflections close to T_L indicating that the incommensurate phase retains its sine-wave structure even close to the lock-in transition.

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