The magnetic structure of $EuFe_2P_2$

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(Dated: July 3, 2013)

The pnictide intermetallic compounds RT_2Pn_2 (R = rare earth; T = transition metal Fe, Co, Ni, Pd; Pn = pnictide P, As) crystallise in the tetragonal $ThCr_2Si_2$−type structure (space group *I4/mmm*: #139) [1] and exhibit a rich variety of magnetic and transport phenomena. Following the initial characterisation of these materials over twenty years ago [2, 3], interest in the pnictide members of this much larger 1–2–2 compound family has been revived by the recent discovery of a large group of iron–pnictide-based superconductors.

Single crystal neutron diffraction at a neutron wavelength of 0.85 Å showed that the Eu moments adopt an incommensurate planar spiral structure in $EuCo_2P_2$ [4], while both hot neutrons [5] and resonant x-ray scattering [6] showed $EuFe_2As_2$ to be a planar antiferromagnet (AFM). A recent resonant x-ray scattering of $EuRh_2As_2$ showed evidence for both commensurate and incommensurate planar ordering [7]. Remarkably, an extensive study of the magnetic and transport properties of $EuFe_2P_2$ [8] has shown that the iron does not order and that not only is this system probably ferromagnetic, but that the Eu moments are ordered close to the c−axis, *i.e.* essentially perpendicular to the ordering plane seen in all of the related europium pnictides studied so far.

The polycrystalline sample of $EuFe_2P_2$ was synthesised by solid state reaction between EuP and Fe_2P. The details of the preparation and pre-synthesis of the phosphides have been reported previously [8, 9]. The sample was almost single-phase, with about 2 wt.% of an Fe_3P impurity observed in the neutron diffraction patterns.

For the neutron diffraction measurements, 1.7 g (slightly less than a $1/e$ thickness for absorption) was spread across a 2 cm by 8 cm area on a 600 μm thick single-crystal silicon wafer and immobilised using a 1% solution of GE-7031 varnish in toluene/methanol (1:1) [10]. Neutron diffraction experiments were carried out on the C2 multi-wire powder diffractometer. A relatively long neutron wavelength of 2.3672(1) Å was used so that low-angle peaks due to any AFM ordering would be well clear of any direct beam contamination. All full-pattern magnetic and structural refinements employed the FullProf/WinPlotr suite [11, 12] with neutron scattering length coefficients for natural Eu taken from the tabulation by Lynn and Seeger [13].

We found that $EuFe_2P_2$ is indeed ferromagnetic with Eu^{2+} moments of 6.6(3) μ_B oriented close to the c−axis, and confirmed the canted structure implied by local (^{57}Fe and ^{151}Eu Mössbauer spectroscopy) measurements. [8]. However, we found no evidence for a more complex helimagnetic structure [8].

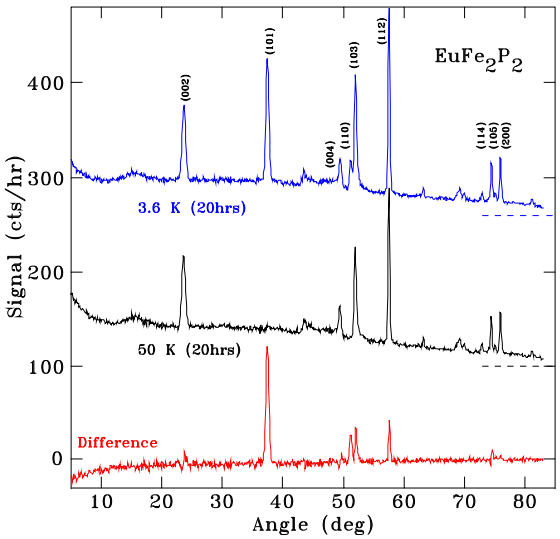

FIG. 1: Powder neutron diffraction patterns for $EuFe_2P_2$ taken at a wavelength of 2.3672(1) Å above (50 K) and below (3.6 K) T_C showing the development of magnetic scattering on cooling. The intensity changes can be seen more clearly in the difference pattern shown at the bottom of the figure.

The diffraction pattern taken at 50 K (*i.e.* above T_C) is presented in Fig. 1 and clearly shows the nuclear Bragg peaks of the $EuFe_2P_2$ phase, demonstrating that neutron powder diffraction does indeed yield a significant signal from europium-based compounds. The lattice parameters at 50 K are a = 3.8165(7) Å and c = 11.166(3) Å. The conventional 'R-factors' are R(Bragg) = 7.6% and R(F) = 7.0%.

Cooling below T_C leads to many changes in the observed diffraction pattern, and while several peaks clearly grow in intensity, there are no new peaks (*i.e.* none that would be forbidden by the crystallographic *I4/mmm* structure). In particular, there are no new peaks below the (0 0 2) peak near $2\theta = 24°$, which would signal the development of commensurate or incommensurate AFM ordering, nor do we observe any satellite peaks around the main peaks which would indicate helimagnetic ordering, as previously proposed [8]. The changes are emphasised in the difference pattern shown at the bottom of Fig. 1 where the dramatic growth of the (1 0 1) peak is the most striking feature. The much smaller increase in

the intensity of the (0 0 2) peak is less obvious but is far more important as it indicates that the Eu^{2+} moments *cannot* be strictly parallel to the $c-$axis: the FM order must be canted away from the $c-$axis by some small angle as previously deduced [8]. Finally, the downturn in the difference pattern below $2\theta = 30°$ in Fig. 1 is due to the loss of incoherent paramagnetic scattering from the Eu^{2+} ions as the FM order develops below T_C. This serves to underline the strength of the magnetic scattering from this system.

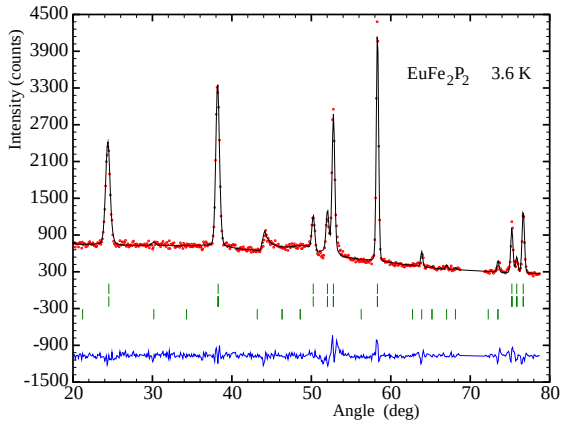

FIG. 2: Refined diffraction pattern for $EuFe_2P_2$ obtained at 3.6 K. The solid line is a full-profile refinement of the nuclear and magnetic contributions. Three rows of Bragg markers are shown (top) nuclear contribution (middle) magnetic contribution, (bottom) Fe_3P impurity of 2 wt.%. The residuals are plotted at the bottom of the figure. Note that since the magnetic order is ferromagnetic, there are no new Bragg peaks associated with it. The instrumental artefacts near $2\theta = 45°$ and 70° were not included in the refinement.

The refinement of the diffraction pattern obtained at 3.6 K is shown in Fig. 2. Earlier ^{57}Fe Mössbauer work showed no evidence for magnetic ordering of the iron [8] so the magnetic contribution from the iron sites was assumed to be zero in our analysis. No misfit resulting from this choice was detected. The 'R-factors' were: R(Bragg) = 6.5%, R(F) = 4.0% and R(mag)=6.3%. We find an Eu^{2+} magnetic moment of 6.6(3) μ_B, close to the 7 μ_B expected for the $J = \frac{7}{2}$ Eu^{2+} ion, confirming the divalence of the Eu ions in $EuFe_2P_2$. Both a full pattern refinement and simulations of the magnetic contributions to the (0 0 2) and (1 0 1) reflections yield a canting angle of 17(3)°. This is fully consistent with the 20(5)° derived from ^{151}Eu Mössbauer and 15(5)° derived from ^{57}Fe Mössbauer [8].

Thus, we find that $EuFe_2P_2$ orders ferromagnetically below a Curie temperature of 30(1) K. The 6.6(3) μ_B Eu^{2+} magnetic moments are canted away from the tetragonal c-axis by 17(3)°.

The magnetic structure of $EuFe_2P_2$ contrasts sharply with that of $EuFe_2As_2$ which serves as a parent material for a family of high temperature superconductors. First, the Fe sublattice in the phosphide is non-magnetic, at least down to 2 K, contrasting with the collinear antiferromagnetic (AFM) ordering below 190 K in the arsenide [5, 6]. Second, the Eu sublattice orders ferromagnetically below 30 K with the moments nearly perpendicular to the basal planes in the phosphide, in contrast to the A-type AFM ordering [14, 15] below 19 K with the moments parallel to the crystallographic $a-$axis [5, 6] seen in the arsenide.

A full description of this work has been published in Phys Rev B[16].

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