



Dependence of magnetic ordering temperature of doped and undoped EuFe_2As_2 on hydrostatic pressure to 0.8 GPa

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ARTICLE INFO

Article history:

Received 19 March 2011

Accepted 18 April 2011

Available online 22 April 2011

Keywords:

Fe pnictide compound

Superconductivity

Magnetic order

High pressure

Eu compound

ABSTRACT

The ac susceptibility of single crystalline tetragonal EuFe_2As_2 , $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$, and $\text{EuFe}_{1.715}\text{Co}_{0.285}\text{As}_2$ has been measured over the temperature and hydrostatic (He-gas) pressure ranges 10–60 K and 0–0.8 GPa, respectively. For all three samples the magnetic ordering temperature (17–19 K) from the Eu sublattice increases linearly with pressure, presumably due to the enhanced exchange coupling between Eu-layers. No evidence for a superconducting transition was observed in the susceptibility for any sample over the measured temperature/pressure range.

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1. Introduction

The high pressure variable has proven to be a valuable tool to further our understanding of superconducting materials. The lattice parameters of a single sample can be changed reversibly without introducing the disorder prevalent in doping experiments. However, in the recently discovered Fe-based pnictides the superconducting state appears to react very sensitively to the introduction of lattice strains such as those arising from the shear stresses introduced by nonhydrostatic pressure media [1]. As pointed out by Duncan et al. [2], high pressure studies on the compounds XFe_2As_2 , where X = Ba, Sr, Ca, “have led to a bewildering array of confusing and apparently contradictory results”. Single crystals of CaFe_2As_2 , for example, are reported to become superconducting under a relatively low pressure ≤ 0.4 GPa if pressure media such as Fluorinert or silicone oil are used [3], whereas with much more hydrostatic He-gas pressure no sign of superconductivity could be detected to 0.7 GPa [4].

It is important to note that in such highly anisotropic materials such as these, pressure-dependent shear stress effects may occur in polycrystalline samples even under purely hydrostatic high-pressure conditions; the anisotropic compression of each crystallite in a polycrystalline sample results in shear stresses across each grain boundary to neighboring crystallites with differ-

ent orientations. To establish the intrinsic dependence of the superconducting state on pressure for a given Fe-based material, it would thus appear to be essential to study only high quality single crystals and to apply pure hydrostatic pressure using He gas pressure medium. It is possible that these same considerations may apply in studies of the magnetic properties of Fe-based materials under pressure.

EuFe_2As_2 exhibits spin-density-wave order below $T_s \approx 185$ K involving the Fe-sublattice; under pressure T_s shifts rapidly to lower temperatures, disappearing completely above ~ 2.5 GPa [5]. The interactions between the strong local magnetic moments on each Eu site (Eu is divalent with a $4f^7$ orbital configuration) lead to type A antiferromagnetic order below 19 K, although the positive Curie–Weiss temperature points to predominantly ferromagnetic interactions [6]. Under pressure the magnetic ordering temperature reportedly remains constant to 2.6 GPa [7], increasing slowly at higher pressures before reaching a maximum near 55 K at ~ 9 GPa [5]. Only in a narrow region of pressure around 3 GPa has bulk superconductivity near 30 K been reported for EuFe_2As_2 [8]. The partial substitution of Co for Fe or of P for As in EuFe_2As_2 leads to a ferromagnetic ground state [9–11]; surprisingly, samples of polycrystalline $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$ [10] and single-crystalline $\text{EuFe}_{1.78}\text{Co}_{0.22}\text{As}_2$ [11] have been reported to exhibit at ambient pressure ferromagnetism in coexistence with a superconducting transition near 26 K and 21 K, respectively, although only a very small fraction of full diamagnetic shielding was observed. In very recent work on well characterized single crystals of $\text{EuFe}_2\text{As}_{2-y}\text{P}_y$,

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Jeevan et al. [9] report that bulk superconductivity only exists in the narrow doping range $0.32 \leq y \leq 0.44$, and thus not for $y = 0.6$.

In this paper we measure the temperature-dependent ac susceptibility of EuFe_2As_2 , $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$, and $\text{EuFe}_{1.715}\text{Co}_{0.285}\text{As}_2$ under the application of hydrostatic He-gas pressures as high as 0.8 GPa; we find that the magnetic ordering temperatures increase linearly with pressure at the rates $+0.35(3)$, $+2.4(1)$, and $+2.0(1)$ K/GPa, respectively. No evidence is found for a superconducting transition in the temperature range 10–60 K.

2. Experiment

High quality single crystals with nominal composition EuFe_2As_2 and $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$ were prepared by solid reaction methods involving two steps. Firstly, polycrystalline EuFe_2As_2 and $\text{EuFe}_2\text{As}_{1.35}\text{P}_{0.65}$ were prepared in evacuated quartz tubes at 900 °C for 50 h and then the resulting materials were ground thoroughly, pressed into pellets, and loaded into Al_2O_3 crucibles. Secondly, the crucibles were sealed in evacuated quartz tubes and were heated to 1190 °C, held at this temperature for 24 h, and finally cooled to 1100 °C over 100 h. Many plate-like tetragonal-structure EuFe_2As_2 and $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$ crystals were obtained. As seen in Fig. 1, X-ray diffraction from the (001) planes yields the lattice parameters $c = 1.207(7)$ nm and $1.188(7)$ nm, respectively. Comparison of these value of c with those obtained in the extensive studies by Jeevan et al. [9] on $\text{EuFe}_2\text{As}_{2-y}\text{P}_y$ crystals, where the doping level y for each crystal was determined by EDX, reveals that the actual level of P-doping for our crystal may be somewhat lower than the nominal value $y = 0.6$, but likely in the range $0.45 \leq y \leq 0.6$. As expected for this doping level no sign of a SDW transition was observed in $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$ either in the ac susceptibility or electrical resistivity over the temperature range 4–300 K [12]. Single crystals of $\text{EuFe}_{1.175}\text{Co}_{0.285}\text{As}_2$ were grown by a self-flux method as described elsewhere [13].

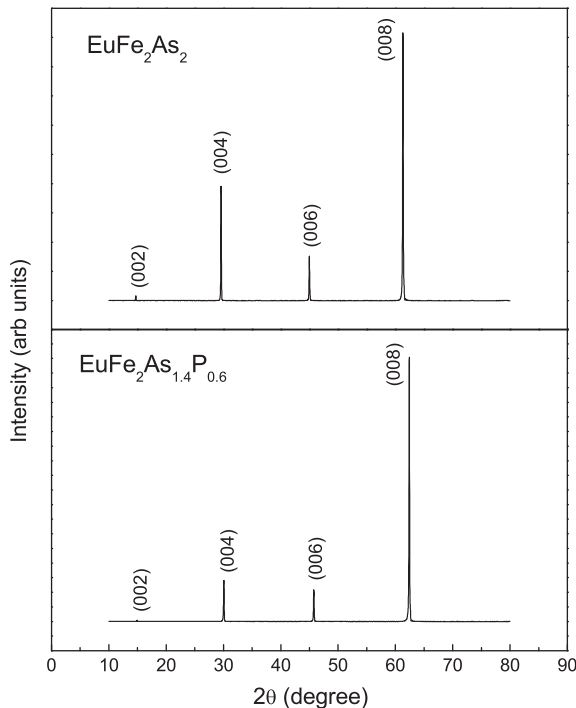


Fig. 1. X-ray diffraction pattern ($\lambda = 0.154$ nm) for (001)-plane oriented single crystals of tetragonal EuFe_2As_2 and $\text{EuFe}_2\text{As}_{1.4}\text{P}_{0.6}$; the data yield the c lattice parameters $1.207(7)$ nm and $1.188(7)$ nm, respectively. No impurity phases are visible.

To generate hydrostatic pressures as high as 0.8 GPa, a He-gas compressor system (Harwood Engineering) was used in combination with a CuBe pressure cell (Unipress). Pressures were changed near room temperature and held for 30–45 min before cooling down to 10 K to measure the temperature-dependent ac susceptibility $\chi(T)$ at 0.1 Oe rms and 1023 Hz applied magnetic field. These measurements were carried out under pressure to the same high accuracy as measurements at ambient pressure by surrounding the sample with a calibrated primary/secondary compensated coil system connected to a Stanford Research SR830 digital lock-in amplifier via a SR554 transformer preamplifier. A two-stage closed-cycle refrigerator (Balzers) was used to cool the pressure cell to low temperatures; measurements were carried out by slowly warming up through the transition at the rate ~ 100 mK/min.

All susceptibility measurements were repeated at least once to verify that the reproducibility of the ferromagnetic or antiferromagnetic transition temperature $T_{C,N}$ was within 20 mK. The temperature-dependent background signal from the empty coil system was measured and subtracted from the sample data. Unfortunately, due to the pressure- and temperature-dependent background signal, it was not possible to reliably extract the pressure dependence of the Curie constant or Curie-Weiss temperature from the paramagnetic ac susceptibility in the temperature region well above $T_{C,N}$. An approximate Curie-Weiss analysis is given below at ambient pressure over the limited temperature range 30–60 K.

All pressures were determined for the sample at a temperature near $T_{C,N}$ using a calibrated manganin gauge located at room temperature near the compressor system and connected via a 3 mm dia. CuBe capillary tube to the pressure cell at low temperature. Further details of the He-gas techniques used are given elsewhere [14].

3. Results and discussion

In Fig. 2 the real part of the ac susceptibility is plotted versus temperature to 60 K at ambient pressure for the doped and undoped EuFe_2As_2 crystals studied. Fitting the data for each in the temperature range 30–60 K results in the Curie-Weiss equations given in the figure and plotted as solid lines. The Curie constant

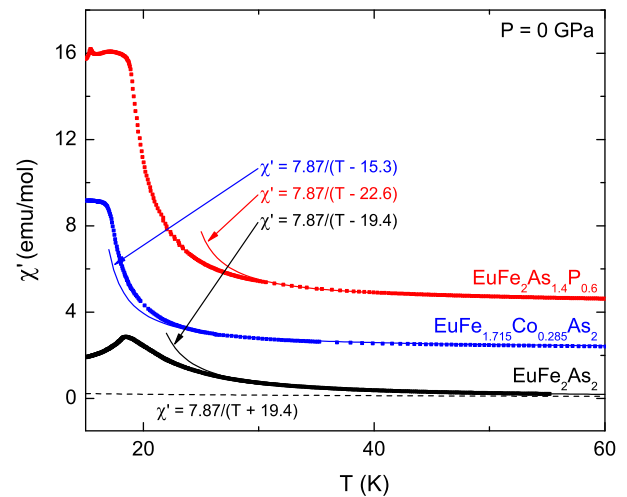


Fig. 2. Real part of the ac susceptibility versus temperature at ambient pressure for the three crystals studied. Ordinate scale applies to EuFe_2As_2 . Data on doped crystals are shifted vertically for clarity. Equations are connected by arrows to respective data fits given by solid lines. Dashed line is plot of equation below it (see text).

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