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Superconductivity and magnetism in K-doped $EuFe_2As_2$

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Abstract

Superconductivity is found in 50% K-doped $EuFe_2As_2$ samples below 33 K. Our results from electrical resistivity, magnetic susceptibility and ^{57}Fe and ^{151}Eu Mössbauer spectroscopy provide clear evidence that the ordering of the Fe moments observed at 190 K in undoped $EuFe_2As_2$ is completely suppressed in our 50% K-doped sample; thus there is no coexistence of the Fe magnetic order and the superconducting state. However, short range ordering of the Eu moments coexists with the superconducting state below 15 K. A bump in the susceptibility well below T_c as well as the broadening of the Fe Mössbauer line below 27 K evidence an interplay between the Eu magnetism and the superconducting state.

1. Introduction

The recent discovery of superconductivity in quaternary rare earth–transition metal oxypnictides has generated immense activity in the field of condensed matter physics. The high superconducting transition temperature of 26 K for F-substituted LaOFeAs [1] was an instant hit as it challenged the monopoly of the copper based oxides as the sole provider of high T_c superconductors. Subsequently it was found that the superconductivity of these oxypnictides could be raised further by replacing La by other rare earths such as Ce, Pr, Nd, Gd, Sm [2–6]. The highest T_c achieved so far is 55 K in SmOFeAs [6]. In all these cases the parent compound has a spin density wave transition at high temperature along with the structural transition which is suppressed by suitable doping and this suppression/weakening of the SDW transition leads to superconductivity. It was soon realized that a related series of compounds, namely AFe_2As_2 (A = Ca, Ba, Sr, Eu), also show similar SDW type anomalies in the resistivity and these could also exhibit superconductivity upon suitable doping or application of external pressure [7–10]. Indeed superconductivity at high temperature has been observed in Na-doped $EuFe_2As_2$ [11], $CaFe_2As_2$ [12] and K-doped $BaFe_2As_2$, $SrFe_2As_2$ as well as $EuFe_2As_2$ [13–15]. Suppression of the SDW transition by the application of pressure has also been observed in AFe_2As_2 (A = Ca, Sr, Eu) [16–19]. Among these, $EuFe_2As_2$ has a special place due to the fact that Eu in this compound is in a divalent state and has a large magnetic moment (7 μ_B) and hence is an ideal candidate to investigate the interplay between superconductivity and magnetism. Many interesting phenomena were discovered from the interplay between local moment magnetism and superconductivity in RRh_4B_4 and RNi_2B_2C [20, 21]. A unique situation was found in $HoNi_2B_2C$ where a double re-entrance is observed in a narrow range of temperature interval which was attributed to the development of a c-axis modulation leading to a ferromagnetic component, thus causing superconductivity to weaken/disappear [22]. In this paper our primary concern is to ascertain the magnetic state of Eu moments and how they affect the superconductivity in this compound.

2. Experimental details

We prepared the polycrystalline $Eu_{0.5}K_{0.5}Fe_2As_2$ using solid state reaction. The starting elements were of high purity. The sample preparation process is similar to that described in [15]. Subsequently the sample was annealed at 800 °C for one week. Phase purity was checked using powder x-ray diffraction and scanning electron microscopy (SEM). Energy dispersive x-ray analysis (EDAX) was used to check the composition of the sample. Magnetization was measured using a SQUID magnetometer (Quantum Design, USA) and electrical resistivity was measured using a Physical

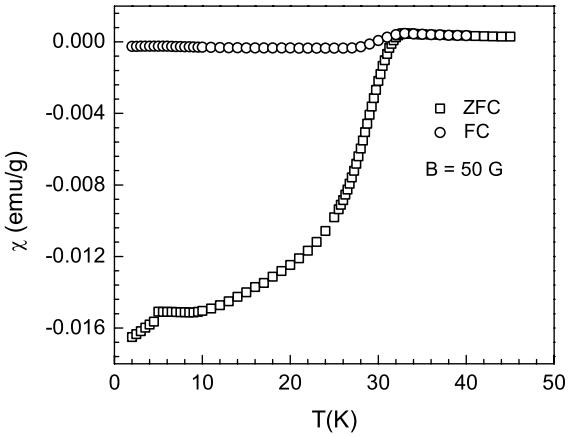

Figure 1. Zero-field cooled (ZFC) and field cooled (FC) magnetization as a function of temperature for $Eu_{0.5}K_{0.5}Fe_2As_2$ measured under an applied magnetic field of 50 G.

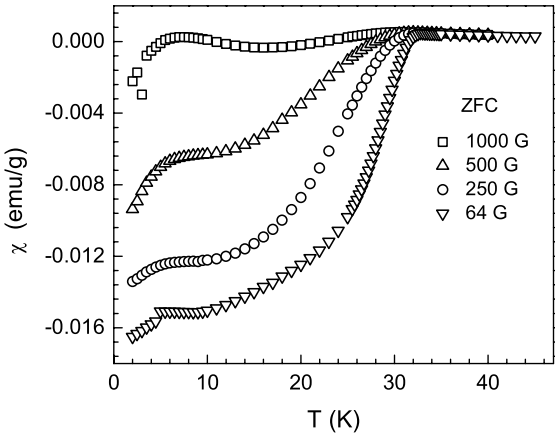

Figure 2. Magnetization of $Eu_{0.5}K_{0.5}Fe_2As_2$ measured under various magnetic fields.

Properties Measurement System (PPMS, Quantum Design, USA). ^{57}Fe and ^{151}Eu Mössbauer spectroscopy measurements were performed at various temperatures between 300 and 4 K using a conventional constant acceleration spectrometer. ^{57}Co and $^{151}SmF_3$ sources were used for the ^{57}Fe and ^{151}Eu Mössbauer spectroscopy, respectively.

3. Results and discussion

Polycrystalline $Eu_{0.5}K_{0.5}Fe_2As_2$ crystallizes in $ThCr_2Si_2$ type tetragonal structure (space group $I4/mmm$) with lattice parameters $a = 3.8656$ Å and $c = 12.962$ Å as confirmed by powder x-ray diffraction data. Our sample forms in single phase with small impurity phases which constitute less than 5%. The lattice parameters are in close agreement with the values as reported in [15]. Also the comparison with lattice parameters of $EuFe_2As_2$ ($a = 3.9104$ Å and $c = 12.1362$ Å [10]) shows that there is contraction of the unit cell along the a-axis and expansion along the c-axis. SEM and EDAX results confirmed the homogeneity with average composition close to the starting composition of $Eu_{0.5}K_{0.5}Fe_2As_2$.

Figure 1 shows the magnetic susceptibility measured under zero-field cooled and field cooled conditions of K-doped $EuFe_2As_2$ for an applied field of 50 G. A clear diamagnetic signal corresponding to a superconducting transition is observed below 33 K. The superconducting signal under the ZFC condition corresponds to that expected for perfect diamagnetism. A reduced diamagnetic signal under the field cooled condition is a hallmark of type II superconductors. In figure 2, we plot the magnetic susceptibility under the ZFC condition for several values of magnetic fields. In addition to the shift of the curves to lower temperature we notice that for 500 G field the magnetic response is similar to that of a re-entrant superconductor like $HoNi_2B_2C$. The strength of the diamagnetic signal increases initially due to superconductivity; below 15 K the signal shows a tendency to flatten out and then below 7 K the diamagnetic signal strength increases again. This kind of behavior was also observed in $HoNi_2B_2C$ which

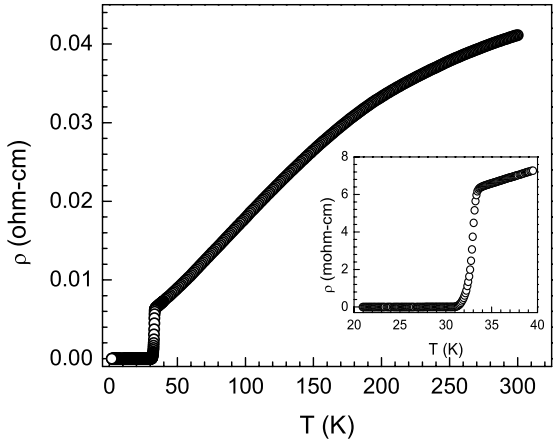

Figure 3. Temperature dependence of the electrical resistivity for $Eu_{0.5}K_{0.5}Fe_2As_2$ at zero field. The inset shows the enlarged view in the low temperature range from 20 to 40 K.

was due to destruction/weakening of superconductivity caused by the appearance of a ferromagnetic component.

The temperature dependence of the electrical resistivity for $Eu_{0.5}K_{0.5}Fe_2As_2$ is shown in figure 3. The electrical resistivity data exhibit a linear behavior in the higher temperature region with an onset of a superconducting transition at 34 K, while the anomaly due to the SDW and structural phase transition as observed in $EuFe_2As_2$ [9, 10] at 190 K is completely suppressed in our 50% K-doped $EuFe_2As_2$. The inset in figure 3 shows the expanded view in the low temperature range 20–40 K. The residual resistivity ratio, RRR ($\rho(300\text{ K})/\rho(34\text{ K})$) is 6.4, which is greater than those for (i) some Na-doped and K-doped 122 iron arsenides and (ii) Eu based 122 intermetallic compounds, indicating the good quality of our sample. Figure 4 shows the electrical resistivity data $\rho(T)$ obtained in the presence of different magnetic fields. As is clear from the figure, with increase in magnetic field the onset of the superconducting transition shifts towards lower temperature. The inset of figure 4 shows the upper critical field $H_{c2}(T)$ and irreversibility field $H^*(T)$ as a function of temperature for $Eu_{0.5}K_{0.5}Fe_2As_2$,

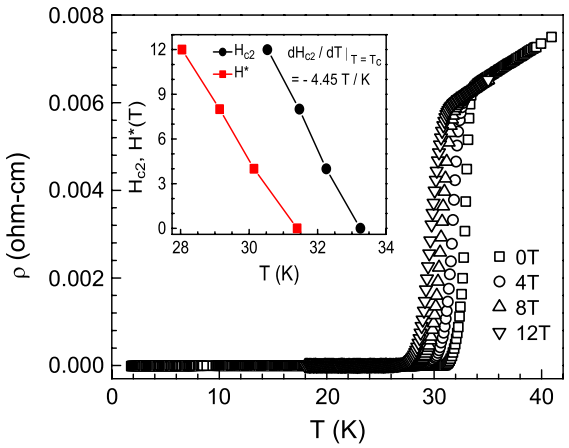

Figure 4. Temperature dependence of the electrical resistivity for $Eu_{0.5}K_{0.5}Fe_2As_2$ at various applied fields. The inset shows the $H_{c2}(T)$ and $H^*(T)$–T (K) phase diagrams.
(This figure is in colour only in the electronic version)

based on resistivity measurements. The corresponding values of $H_{c2}(T)$ and $H^*(T)$ were obtained by using the 90% and 10% values of the normal state resistivity ρ_n at $T = T_c$. The slope of the upper critical field amounts to -4.45 T K^{-1}. We estimated the zero-temperature upper critical field $H_{c2}(0)$ using the Werthamer–Helfand–Hohenberg formula [23], $H_{c2}(0) = -0.693T_c(dH_{c2}/dT)$. Taking $T_c = 33$ K, we obtained $H_{c2}(0) = 101$ T, a value smaller than that reported for $Sr_{0.6}K_{0.4}Fe_2As_2$ (153 T) [24]. This again evidenced the coupling of the Eu magnetic moment with the superconductivity, leading to a smaller value of the upper critical field. The value of the mean-field Ginzburg–Landau coherence length calculated using the relation $\xi = (\Phi_0/2\pi H_{c2})^{1/2}$ ($\Phi_0 = 2.07 \times 10^{-11}$ T cm^2 being the flux quantum) is 18 Å. This value is comparable to the reported coherence length for Na-doped $EuFe_2As_2$ [11].

^{151}Eu Mössbauer spectroscopy results are shown in figure 5. The spectrum at room temperature shows the presence of two single lines with an isomer shifts 1.1 and -11.4 mm s^{-1}. The main line is at -11.4 mm s^{-1}, which confirms the divalent nature of Eu in this compound. The small intensity line at 1.1 mm s^{-1} represents trivalent Eu and this might be due to the formation of a small amount of trivalent impurity phase during the powdering process for preparing the absorber or during the sample preparation. The similar isomer shifts corresponding to divalent and trivalent Eu ions were also observed for EuM_2As_2 (M = Fe, Ni, Pd, Cu) [25, 26]. As the temperature is lowered below 15 K, the line at -11.4 mm s^{-1} broadens without showing a well defined splitting. This indicates the onset of internal fields with some field distribution, evidencing the onset of short range magnetic ordering as could be expected from the diluted Eu sublattice. The least square fit of the spectra at 4.2 K shows a hyperfine field of 11 T at the Eu nucleus. This value of the hyperfine field is substantially smaller (nearly half) than that for $EuFe_2As_2$ ($B_{hf} = 26$ T) which is due to the doping of 50% europium with non-magnetic potassium [25].

^{57}Fe Mössbauer spectra also reveal important information about this system (figure 6). At room temperature we see a

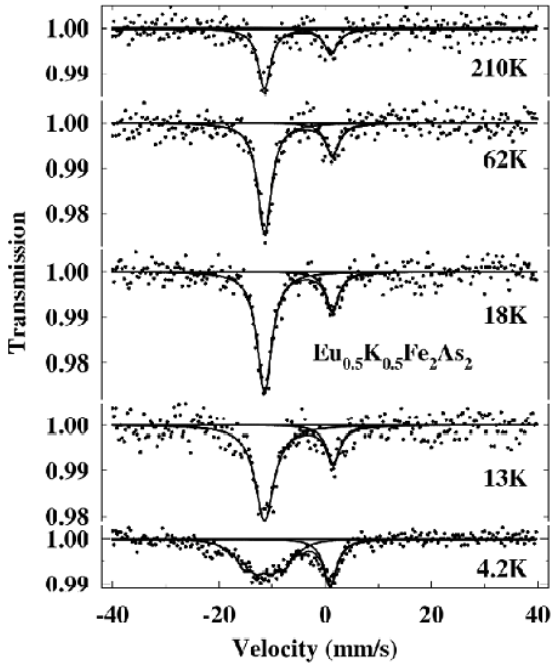

Figure 5. ^{151}Eu Mössbauer spectroscopy in the paramagnetic and magnetically ordered states. Note the broadening of the line at -11.4 mm s^{-1} due to the magnetic order of Eu^{2+} moments.

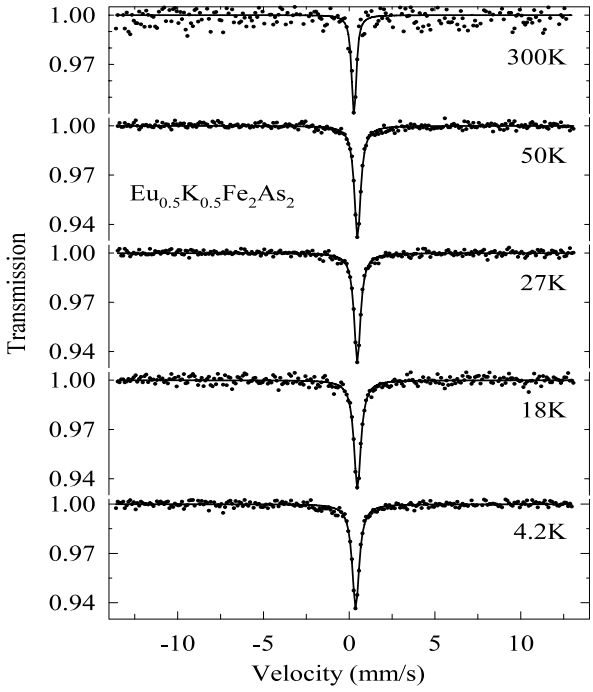

Figure 6. ^{57}Fe Mössbauer spectroscopy of $Eu_{0.5}K_{0.5}Fe_2As_2$ in the paramagnetic state and magnetically ordered state.

single unsplit line with an isomer shift of 0.4 mm s^{-1} and a linewidth (full width at half-maximum) of 0.32 mm s^{-1} confirming the absence of unreacted iron in the sample. The linewidth gradually increases from 0.32 to 0.47 mm s^{-1} at 27 K, suggesting the absence of an SDW transition which was present in the parent compound $EuFe_2As_2$ at 190 K. A clear change in the slope of the linewidth variation is

detectable below about 27 K and the linewidth nearly saturates at 0.5 mm s^{-1} from 18 K downwards showing no tendency of any splitting due to the magnetic hyperfine field. The broadening of the resonance line below 27 K can be attributed to the transferred field from the ordering of Eu moments.

4. Conclusion

To summarize our combined investigation of the magnetic susceptibility, the electrical resistivity and Mössbauer spectroscopy clearly establish the coexistence of Eu short range magnetic order and superconductivity in $Eu_{0.5}K_{0.5}Fe_2As_2$ samples. Superconductivity in this sample occurs below 33 K and magnetic order due to Eu moments occurs below 13 K. The features of the magnetic susceptibility in the temperature range 5–15 K are similar to those found for $HoNi_2B_2C$ which were attributed to the intense competition between Ho magnetic order and superconductivity.

Acknowledgments

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