Evolution of superconductivity and ferromagnetism in Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$

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Abstract. Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ crystals were studied by resistivity and magnetization measurements. It is found that the SDW transition is gradually suppressed with the Ru doping, and superconductivity with $T_{sc} \sim 22$ K emerges when $x \geq 0.2$. The magnetic order of the Eu sublattice changes from $A$-type antiferromagnetic to ferromagnetic within $x \leq 0.2$, making superconductivity and ferromagnetism coexist in a broad regime of $0.2 < x < 0.55$.

1. Introduction
Since the discovery of superconductivity (SC) in LaFeAsO$_{1-x}$F$_x$[1], iron-based high temperature superconductors have attracted great much attention.[2] Among them, $AT_2$As$_2$ (so-called 122 phase, $A$ = Ca, Ba, Sr, Eu, $T$ = Fe) system has been more intensively investigated, because high-quality large single crystals can be grown. EuFe$_2$As$_2$ (Eu122) is a special member in the 122 system, since the Eu atoms carries a spin of $S=7/2$ that is ordered at about 20 K.[3] This unique feature makes Eu122 as a possible platform for investigating the interplay between magnetism and Fe-based SC.

By the hole doping with partial substitution of K for Eu, SC over 30 K was realized in Eu$_{1-x}$K$_x$Fe$_2$As$_2$.[4] However, the possible magnetic ordering of Eu$^{2+}$ spins was also lost due to the dilution in the Eu sublattice. To keep the Eu sublattice intact, one has to perform doping study at either As site or Fe site in Eu122. By the phosphorus doping at the As site, we found SC at 26 K and ferromagnetism (FM) at 20 K in EuFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$.[5, 6] Another choice is to dope electrons at the Fe site. Using cobalt as the dopant, superconducting transition appears at 21 K, and the Eu spins are helimagnetically ordered at 17 K in Eu(Fe$_{0.89}$Co$_{0.11}$)$_2$As$_2$.[7] Very recently, with the isoelectronic Ru doping, SC at 23 K and FM at 20 K was obtained in Eu(Fe$_{0.75}$Ru$_{0.25}$)$_2$As$_2$, where an unprecedented anisotropic superconductivity was observed.[8] In this paper we report how the SC and FM are evolved in the Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ system.

2. Experiments
A series of Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ crystals were grown by self-flux method using (Fe$_{1-x}$Ru$_x$)$_2$As$_2$ as solvent, analogous to our previous method.[8] The as-grown crystals were annealed in vacuum to homogenize the Eu distribution. The real composition for each crystal obtained was determined by a Phoenix EDAX x-ray spectrometer (EDX). X-ray diffraction demonstrated high quality for the crystals (not shown here). Electrical resistivity was measured using a standard four-terminal...
Figure 1. (a) In-plane electrical resistivity (normalized at 300 K) of Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ crystals ($x$ as indicated in the plot). (b) and (c) Lower-temperature range of normalized resistivity. $T_{SC}$ and $T_C$ refer to superconducting and magnetic transition temperatures, respectively.

Method. Magnetization was measured on a Quantum Design Magnetic Property Measurement System (MPMS-5).

3. Results and discussion

Fig 1. shows temperature dependence of normalized resistivity [with the same current (20 mA) flowing in the $ab$-plane] for Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ crystals. SDW anomaly associated with magnetic transition of Fe sublattice is gradually suppressed from $T = 186$ K ($x=0$) down to $T = 102$ K ($x=0.14$). The $x=0.14$ crystal has signature of both superconducting and SDW transitions. For $x \geq 0.14$, no sign of SDW ordering can be seen. The crystals with $x=0.2$, 0.21 and 0.25 show superconducting transition at $T_{SC} \sim 22$ K with zero resistance. For $x > 0.25$, SC is also signatured by the resistivity drop at $\sim 23$ K, although no zero resistivity could be achieved. The absence of zero resistance is evidently owing to the magnetic ordering of Eu spins. The enlarged plots for low temperature data enable us to see the anomalies in connection with magnetic transition at $\sim 17$ K. Worthy to note is that the superconducting transition temperatures varies little with changing the Ru content. This is in stark contrast with the case in Ru-doped Ba122.[9]

The magnetic susceptibility data for $x=0$, 0.05 and 0.21 are plotted in Fig 2. The data of the parent compound are consistent with the $A$-type antiferromagnetism in which the Eu spins
Figure 2. Temperature dependence of magnetic susceptibility of Eu(Fe\textsubscript{1-x}Ru\textsubscript{x})\textsubscript{2}As\textsubscript{2} under magnetic fields parallel to the c-axis. \(H = 1000\) Oe for \(x = 0\) (a), \(H = 10\) Oe for \(x = 0.05\) (b), and \(H = 10\) Oe for \(x = 0.21\) (c).

lie within ab-plane.[10] For \(x = 0.05\), below \(T_N = 17.1\) K, bifurcation of zero field cooling (ZFC) and field cooling (FC) curves appears around 10 K, indicating the existence of ferromagnetic component along the c-axis (Mössbauer study shows the evidence[8]). SC in \(x = 0.21\) crystal is confirmed by the pronounced diamagnetism below \(T_{SC} \approx 22\) K, consistent with resistivity result in Fig. 1(b). Note that there is a slight peak in the ZFC curve at \(T_M \approx 18.9\) K, which is probably due to FM ordering. On the other hand, no obvious meissner effect (diamagnetism in FC mode) was observed. We believe that this is the intrinsic character of the ferromagnetic superconductor.

The high-temperature susceptibility follows Curie-Weiss law, \(\chi_c(T) = \chi_0 + C/(T + \Theta)\). Table 1 lists the fitted parameters. The effective moments are close to the theoretical value (7.94\(\mu_B\)) of a free Eu\textsuperscript{2+} ion. The Weiss temperature is negative, and tends to increase with \(x\), suggesting enhanced FM correlations between the Eu\textsuperscript{2+} ions.

| fitted parameters | EuFe\textsubscript{2}As\textsubscript{2}[10] | Eu(Fe\textsubscript{0.95}Ru\textsubscript{0.05})\textsubscript{2}As\textsubscript{2} | Eu(Fe\textsubscript{0.79}Ru\textsubscript{0.21})\textsubscript{2}As\textsubscript{2} |
|-------------------|---------------------------------|---------------------------------|---------------------------------|
| \(\Theta\) (K)    | -19.7                           | -20.97                          | -21.1                           |
| \(C\) (emu/K Oe mol) | 8.31                           | 8.00                           | 7.96                           |
| \(\mu_{eff}\) (\(\mu_B\)) | 8.13                           | 8.00                           | 7.98                           |

Fig. 3 shows the isothermal magnetization curves for the three representative samples. The undoped material shows spin reorientation behaviour under external fields. For \(x = 0.05\), a slight magnetic hysteresis can be seen below 10 K, consistent with the above magnetic susceptibility data. The \(M_c - H\) loop for \(x = 0.21\) is particularly interesting. The derivative of magnetization \(dM/dH\) is negative at the temperatures far below \(T_{SC}\). This is consistent with the behaviour
Figure 3. $M_c - H$ curves for Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$. (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.21$.

of a usual superconductor at $H < H_{c1}$ ($M - H$ data is a straight line with negative slopes). However, the difference is obvious. The $M_c - H$ data contain a component of FM loop. This fact clearly indicates the coexistence of SC and FM in Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$. When the temperature is close to $T_M$ and $T_{SC}$, or the magnetic field is increased, more complicated magnetic behaviour is seen.

4. Summary
We have synthesized high-quality Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ single crystals which enable us to systematically investigate the evolution of FM and SC. The resistivity and magnetization of three representative crystals with $x = 0$, 0.05 and 0.21 are reported. The results indicate that the Ru doping first change the magnetism of Eu sublattice into FM (or ferromagnetic component), then SC of Fe 3$d$ electrons emerges. The ferromagnetic superconductor shows some novel phenomena including the absence of zero resistance and/or absence of Meissner effect. Further experiments like Hall and Nernst measurements are promising to reveal the novelty of the coexistence of FM and SC.

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