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Effect of substitution of Ca^{2+} for Eu^{2+} on pressure-induced superconductivity in $EuFe_2As_2$

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To elucidate competition between the Fe-based superconductivity and the antiferromagnetic order of Eu^{2+}, the electrical resistivity of $Eu_{0.85}Ca_{0.15}Fe_2As_2$ under high pressure is presented. Under ambient pressure, the Fe-based spin-density-wave (SDW) transition and the antiferromagnetic ordering (AFM) of Eu^{2+} are observed at $T_{SDW} = 190$ K and $T_N = 17$ K, respectively. With applying pressure, the Fe-based superconductivity with $T_c \sim 25$ K appears above 1.5 GPa, accompanying collapse of the SDW transition. Compared with $EuFe_2As_2$, T_c is close to T_N, which implies strong competition between the superconductivity and the AFM. This competition would lead to non-zero resistivity at lowest temperature.

KEYWORDS: Fe-based superconductivity, pressure, antiferromagnetism, Eu, Ca

1. Introduction

Discovery of the high-T_c superconductivity in $LaFeAsO_{1-x}F_x$ by Kamihara et al.[1] triggered extensive researches on the Fe-based superconductivity. The parent compound, LaFeAsO, which crystallizes in the tetragonal ZrCuSiAs-type structure at room temperature, performs a structural phase transition from tetragonal to orthorhombic symmetry at 155 K and a spin-density-wave (SDW) transition associated with Fe at 137 K. The F^- doping (electron doping) induces the Fe-based superconductivity almost simultaneously with disappearance of both transitions.[1] Interestingly, although a magnetic element has been considered to be destructive to superconductivity, the FeAs layer plays a key role in the emergence of the superconductivity. AFe_2As_2 (A=Ca^{2+}, Sr^{2+}, Ba^{2+}, Eu^{2+}), which crystallizes in the tetragonal $ThCr_2Si_2$ type structure with FeAs layers similar to those in LaFeAsO, also exhibits similar SDW and structural transitions. Substitution of alkali metal for A (hole doping)[2–5] or application of pressure[6–12] induces the superconductivity, accompanied by suppression of the SDW and structural transitions. However, $EuFe_2As_2$ is a little different from the other AFe_2As_2 systems since Eu^{2+} ions with a large magnetic moment of $7\mu_B$ exhibit antiferromagnetic order (AFM), which is expected to be destructive to the Fe-based superconductivity. Actually, the electrical resistivity under pressure of $2\ \mathrm{GPa} \leq P \leq 2.5\ \mathrm{GPa}$ presents a sharp drop, which is reminiscent of the superconductivity, but does not reach zero resistivity at the lowest temperature, which suggests the complete formation of the superconducting state is interrupted by the ordering of Eu^{2+}.[11] A little higher pressure of ~ 2.8 GPa barely achieves zero resistivity.[12] To elucidate such competition between the Fe-based superconductivity and the AFM of Eu^{2+}, we have attempted to weaken the AFM of Eu^{2+}. In the previous paper, we reported that Ca-substitution by 50 % weakens the AFM of Eu^{2+} and results in the pressure-induced bulk superconductivity with zero resistivity in a wide pressure range of $1.2 \sim 2.4$ GPa.[13,14] In the present paper, to clarify more detailed effects of Ca substitution for Eu on the Fe-based superconductivity, the behavior of $Eu_{0.85}Ca_{0.15}Fe_2As_2$ under high pressure is mainly presented.

2. Experimental

The single crystals of $Eu_{1-x}Ca_xFe_2As_2$ ($x = 0$, 0.15, 0.50, 0.65, 1) were grown in a tin flux. A mixture of constituent elements and the tin flux, which was put into an alumina crucible, was heated at 1000°C and cooled at a rate of $-14°\mathrm{C/h}$ under Ar atmosphere, as described in Ref.[13] We obtained many pieces of plate-like single crystals with typical dimensions of $\sim 3 \times 3 \times 0.1$ mm^3. The powder X-ray diffraction (XRD) was carried out at room temperature. It was verified that the samples are a single phase with $ThCr_2Si_2$ type. The lattice constants, a and c, estimated from the XRD pattern, decrease linearly with increasing x, which indicates the samples obeys Vegard's law. The composition of the sample was determined from the energy dispersive X-ray (EDX) spectroscopy. The measurement of the electrical resistivity was performed under high pressure by using an ac resistance bridge (LR-700; Linear Research) in the temperature range of $4-280$ K. The pressure was generated by using a piston-cylinder-type pressure cell, which consists of inner (NiCrAl alloy) and outer (CuBe alloy) cylinders. The sample and a tin manometer were placed into a Teflon cell filled with a pressure-transmitting medium consisting of a mixture of two types of Flourinert in a ratio of FC70 : FC77 = 1 : 1. The Teflon cell was inserted into the pressure cell and pressed by pistons made of nonmagnetic tungsten carbide. The pressure value was estimated from pressure dependence of superconducting transition of the tin.

3. Results and Discussions

Figure 1 shows temperature dependence of the electrical resistivity of $Eu_{1-x}Ca_xFe_2As_2$ ($x = 0$, 0.15, 0.50, 0.65, and 1) under ambient pressure. All $\rho(T)$ curves have a peak at around $170 \sim 190$ K, which is associated with the SDW

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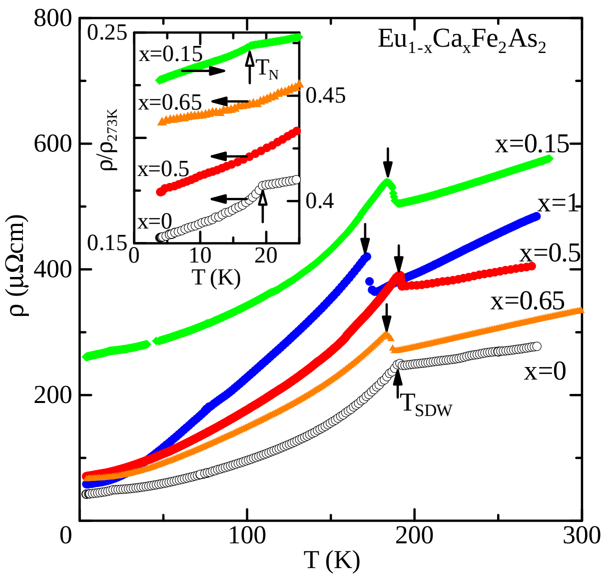

Fig. 1. Temperature dependence of the electrical resistivity under ambient pressure of $Eu_{1-x}Ca_xFe_2As_2$ (x = 0, 0.15, 0.50, 0.65, and 1). The inset shows the resistivity normalized by the value at 273 K as a function of temperature. The kink (open arrows) exhibits the Néel temperature of Eu^{2+}.

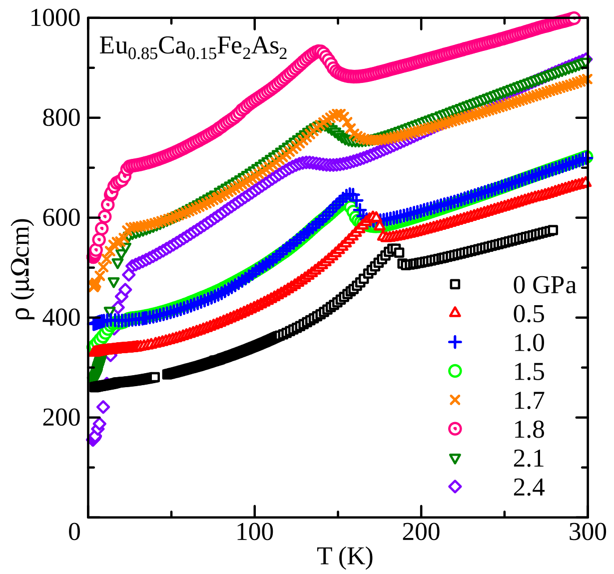

Fig. 2. Temperature dependence of the electrical resistivity under high pressures of $Eu_{0.85}Ca_{0.15}Fe_2As_2$.

transition. This result suggests the Ca- substitution does not suppress the SDW transition unlike the K- substitution. The SDW transition temperature, $T_{\rm SDW}$, is almost independent of Ca concentration, x, except for $x = 1$. In the inset of Fig. 1, the resistivity normalized by that at 273 K is plotted as a function of temperature for clarity. There exists a kink in the $\rho(T)/\rho(273\ {\rm K})$ curves of $x = 0$ and 0.15, which corresponds to the Néel temperature of Eu^{2+}, $T_{\rm N}$. The $T_{\rm N}$ value of $x = 0.15$ (17 K) is a little lower than that of $x = 0$ (19 K). This behavior is consistent with the previous data of magnetic susceptibility.[14] On the other hand, one cannot see such a kink for $x \geq 0.5$. For $x = 0.50$, a clear bend in the $\chi(T)$ curve, which is associated with $T_{\rm N}$, is observed at 9 K.[14] This possibly indicates that the AFM of Eu^{2+} for $x = 0.50$ is weakened compared with $x = 0$ and 0.15, and that the magnetic susceptibility is more sensitive to the AFM than the electrical resistivity. For $x = 0.65$, there exists no anomaly of the AFM of Eu^{2+} in the electrical resistivity and the magnetic susceptibility, which implies the AFM of Eu^{2+} is suppressed below 4 K.

Figure 2 depicts temperature dependence of the electrical resistivity of $x = 0.15$ under high pressures. The same data from 4 K to 40 K are plotted in Fig. 3 for clarity. With increasing pressure, $T_{\rm SDW}$ is lowered gradually and the peak of $T_{\rm SDW}$ is broadened. For $0 \leq P \leq 1.0$ GPa, the $\rho(T)$ curves are qualitatively similar to each other and there is no evidence of the superconductivity. As shown in the inset of Fig. 3, the $\rho(T)$ curve has a shallow kink at $T_{\rm N}$, which is almost independent of pressure. For $P \geq 1.5$ GPa, on the other hand, a drop of the electrical resistivity, which is reminiscent of the superconductivity, appears at around $T_{\rm c} = 25$ K, as clearly shown in Fig. 3. The drop temporarily stops at around $T_{\rm N}$. Below $T_{\rm N}$, the resistivity decreases again with decreasing temperature, but does not reach zero resistivity at lowest temperature. These results suggest that the superconducting transition is interrupted significantly by the AFM of Eu^{2+}. With increasing pressure, the drop becomes larger, which implies the superconducting domain becomes larger with pressure. Compared with $EuFe_2As_2$ ($x = 0$), these features are qualitatively similar, but a little different quantitatively. For $x = 0.15$, the superconductivity begins to appear at around $P_{\rm c} = 1.5$ GPa, which is lower than $P_{\rm c} = 2.03$ GPa for $x = 0$.[11] This is possibly caused by chemical pressure applied by Ca-substitution, as discussed in Ref.[13] It was roughly estimated that 50 % substitution of Ca for Eu corresponds to an external pressure of ~ 1 GPa.[13] Therefore, the difference in $P_{\rm c}$ between $x = 0$ and $x = 0.15$ is likely to be explained semi-quantitatively by the chemical pressure. In that case, the sample with $x = 0.15$ deserves to exhibits the superconductivity with zero resistivity since $EuFe_2As_2$ performs the superconductivity with zero resistivity at around 2.8 GPa.[12] However, the zero resistivity is not observed up to 2.4 GPa, which is possibly due to lower $T_{\rm c}$ value of $x = 0.25$ (~ 25 K) than that of $x = 0$ (~ 30 K). This means that the Ca-substitution weakens not only the AFM of Eu^{2+} but also the Fe-based superconductivity. For $x = 0.25$, larger decrease in $T_{\rm c}$ than in $T_{\rm N}$ possibly produces more competitive situation between the Fe-based superconductivity and the AFM of Eu^{2+} than that for $x = 0$. For $x = 0.50$, $T_{\rm N}$ (9 K) is much lower than that for $x = 0.15$, which means that the AFM of Eu^{2+} is more weakened than that for $x = 0.15$, while $T_{\rm c}$ (23 K) is as high as that for $x = 0.15$. Therefore, the superconductivity is superior to the AFM of Eu^{2+}, which gives rise to the superconductivity with zero resistivity in a wide pressure range. For $x = 0.15$, however, it is expected that applying higher pressure than 2.4 GPa would induce the zero resistivity because the resistivity shows a declining tendency with pressure at around 2.4 GPa. In addition, the slight anomaly associated with the SDW remains at 2.4 GPa. To examine these points in detail, the experiments under higher pressures are now in progress.

In summary, the T-P phase diagram of $Eu_{0.85}Ca_{0.15}Fe_2As_2$ is presented in Fig. 4. With increasing pressure, the SDW transition shifts lower, but is not

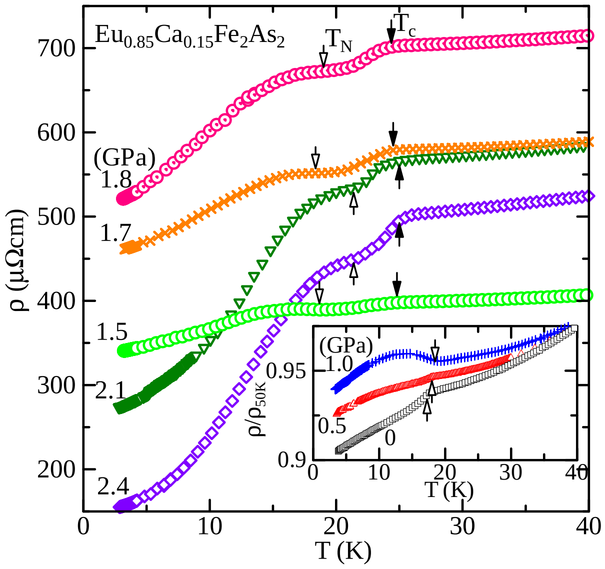

Fig. 3. Temperature dependence of the electrical resistivity under ambient pressure of $Eu_{0.85}Ca_{0.15}Fe_2As_2$ in the temperature range between 4 K and 40 K. The inset shows the resistivity normalized by the value at 50 K as a function of temperature. The onset of the drop (solid arrows) shows the superconducting transition temperature, T_c. The kink (open arrows) exhibits the Néel temperature, T_N, of Eu^{2+}.

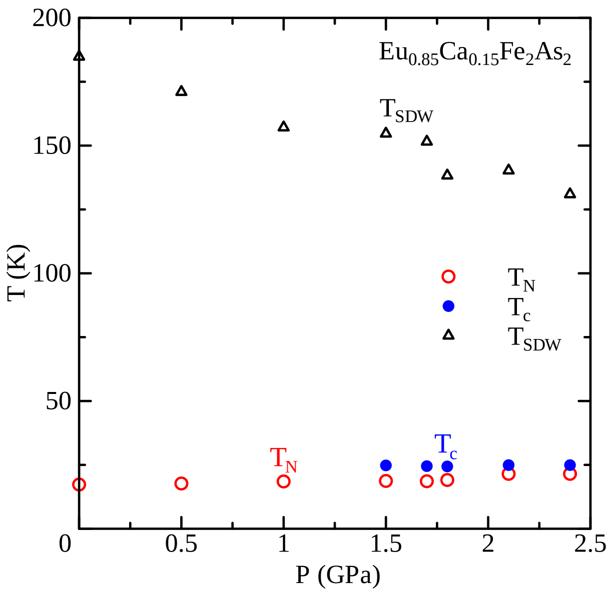

Fig. 4. The T–P phase diagram of $Eu_{0.85}Ca_{0.15}Fe_2As_2$.

collapsed completely in the pressure range of 0–2.4 GPa. Accompanying the collapse of the SDW transition, the Fe-based superconductivity with $T_c \sim 25$ K emerges above 1.5 GPa. On the other hand, T_N of Eu^{2+} remains $\sim$ 20 K up to 2.4 GPa. T_c is close to T_N, which suggests that the Fe-based superconductivity is more competitive with the AFM of Eu^{2+} compared with $EuFe_2As_2$. This results in a finite resistivity value at lowest temperature.

4. Conclusions

To elucidate competition between the Fe-based superconductivity and the AFM of Eu^{2+}, we examined the temperature dependence of the electrical resistivity of $Eu_{0.85}Ca_{0.15}Fe_2As_2$ under high pressure. Similar to $EuFe_2As_2$, the sample exhibits the SDW transition at 190 K and the AFM of Eu^{2+} at 17 K under ambient pressure. With increasing pressure, the Fe-based superconductivity is induced, accompanying collapse of the SDW. In spite of diluting Eu^{2+}, more competitive situation between the Fe-based superconductivity and the AFM of Eu^{2+} is realized. The experiments under higher pressure are desired.

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