High-Pressure Electrical Resistivity Measurements of EuFe$_2$As$_2$ Single Crystals

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Abstract.
High-pressure electrical resistivity measurements up to 3.0 GPa have been performed on EuFe$_2$As$_2$ single crystals with residual resistivity ratios $RRR = 7$ and 15. At ambient pressure, a magnetic / structural transition related to FeAs-layers is observed at $T_0 = 190$ K and 194 K for samples with $RRR = 7$ and 15, respectively. Application of hydrostatic pressure suppresses $T_0$, and then induces similar superconducting behavior in the samples with different $RRR$ values. However, the critical pressure $\sim 2.7$ GPa, where $T_0 \rightarrow 0$, for the samples with $RRR = 15$ is slightly but distinctly larger than $\sim 2.5$ GPa for the samples with $RRR = 7$.

1. Introduction
Since the discovery of superconductivity in LaFeAs(O,F) with $T_c = 26$ K [1], a family of Fe-pnictide superconductors has attracted much attention. In particular, $A$Fe$_2$As$_2$ ($A = $ Ca, Sr, Ba, Eu, etc.) with a tetragonal ThCr$_2$Si$_2$-type structure has been intensively studied because of the availability of stoichiometric single crystals with high quality. It turned out that, in Fe-pnictide compounds, the superconducting (SC) ground state could appear in accordance with the suppression of a magnetic/structural transition by doping [2]. In the phase diagrams, it is argued that the superconductivity could coexist and/or compete with the antiferromagnetism [3, 4]. However, a random potential introduced by doping could smear the intrinsic SC properties. For understanding the origin of the high-$T_c$ superconductivity with $T_c$ up to 55 K [5], it is of considerable importance to probe the systematic change of ground states using high-quality single crystals. An alternative way to tune the ground state is to apply hydrostatic pressure ($P$). For instance, recent high-$P$ ac-susceptibility and resistivity measurements have revealed that $A$Fe$_2$As$_2$ ($A = $ Sr, Eu) exhibits $P$-induced bulk superconductivity by suppressing the magnetic/structural transition [6–10]. Meanwhile, superconductivity under hydrostatic $P$ is absent in CaFe$_2$As$_2$ [11–14], and remains a controversial issue in BaFe$_2$As$_2$ [8, 15–17].

Among the $A$Fe$_2$As$_2$ series, EuFe$_2$As$_2$ is quite unique because the localized Eu$^{2+}$ moments order antiferromagnetically at $T_N \sim 20$ K, in addition to the magnetic/structural transition related to FeAs-layers at $T_0 \sim 190$ K [18–21]. Interestingly, the magnetic order of Eu$^{2+}$ moments...
can be detected even in the SC state induced by doping or application of pressure, which could be a main reason for the novel reentrant-SC-like behavior [9, 10, 22-25].

Here, we report the results of high-\(P\) electrical resistivity measurements in EuFe\(_2\)As\(_2\) using newly grown single crystals with a residual resistivity ratio (\(RRR\)) as high as 15. At ambient \(P\), the magnetic/structural transition occurs at a higher temperature of \(T_0 = 194\) K, compared with 190 K for single crystals with \(RRR = 7\). Consequently, it is found that the higher quality single crystal requires higher-\(P\) to suppress \(T_0\), and to induce the SC ground state in EuFe\(_2\)As\(_2\).

2. Experimental Details

Single crystals of EuFe\(_2\)As\(_2\) were grown by Bridgman method from a stoichiometric mixture of the constituent elements. In this study, we examined several crystals from two different batches with residual resistivity ratios \(RRR = 7\) and 15, where \(RRR\) is defined as \(\rho_{300K}/\rho_{4K}\). Single crystals measured in Ref [10] were taken from a batch with \(RRR = 7\). High-pressure resistivity measurements of samples with \(RRR = 7\) and 15 have been performed simultaneously up to 3.0 GPa using a hybrid-type piston cylinder pressure device [26]. The resistivity was measured by the four-probe method with an ac current \(I = 0.3\) mA in the \(ab\)-plane. To generate hydrostatic pressure, Daphne 7474 (Idemitsu Kosan) oil, which remains in a liquid state up to 3.7 GPa at room temperature [27], was used as a pressure-transmitting medium. Samples were cooled down in Oxford \(^4\)He system, slowly with an average rate of 0.5 K/min. Applied pressure was estimated at 4.2 K from the resistance change of a calibrated Manganin wire [28].

3. Results and Discussions

Figure 1 shows the temperature (\(T\)) dependence of electrical resistivity scaled at 300 K (\(\rho/\rho_{300K}\)) in EuFe\(_2\)As\(_2\) single crystals with \(RRR = 7\) and 15, where \(RRR\) is determined as \(\rho_{300K}/\rho_{4K}\). The measurement was performed in zero field at ambient pressure outside a pressure device with current direction \(I \parallel ab\). To our knowledge, \(RRR = 15\) is the largest value in EuFe\(_2\)As\(_2\) single crystals [9, 21, 29]. Overall \(T\)-variations of the resistivity in the samples with \(RRR = 7\) and 15

![Figure 1](image)

**Figure 1.** (Color online) The scaled electrical resistivity \(\rho/\rho_{300K}\) versus temperature in EuFe\(_2\)As\(_2\) single crystals with \(RRR = 7\) and 15. The measurement was carried out in zero-field at ambient pressure with the current direction \(I \parallel ab\). Upper left and lower right insets represent the expanded views around \(T = T_0\) and \(T_N\), respectively. The data for the sample with \(RRR = 7\) in the lower right inset is arbitrarily shifted in vertical direction for clarity.
are qualitatively similar to each other, and are consistent with previous results [9, 20, 21, 29]. It is worthwhile to mention that, as shown in the upper left inset, a magnetic/structural transition temperature $T_0 = 194$ K for the sample with $RRR = 15$ is higher than $T_0 = 190$ K for the sample with $RRR = 7$. This would be the reason why samples with $RRR = 15$ needs higher pressure ($P$) to suppress $T_0$, as will be discussed below. The Néel temperature $T_N$ of the localized Eu$^{2+}$ moments for the sample with $RRR = 15$ is slightly higher than the value for the sample with $RRR = 7$, as can be seen in the lower right inset.

Next, we turn to the pressure effect on the electrical resistivity for the samples with $RRR = 7$ (Fig. 2) and 15 (Fig. 3), which are simultaneously measured in the same pressure device. With increasing $P$, the resistivity peak related to the magnetic/structural transition is suppressed to a lower temperature in both samples as shown in Figs. 2(a) and 3(a). For the sample with $RRR = 7$, a reminiscence of the peak is clearly recognized at 2.38 GPa around 100 K, and faintly visible at 2.46 GPa around 70 K as shown in Fig. 2(a). At 2.55 GPa, there is no detectable anomaly, which implies that the critical pressure $P_c$, where $T_0 \rightarrow 0$, may be about 2.5 GPa. For the sample with $RRR = 15$, $P_c$ would be $\sim 2.7$ GPa since the resistivity hump is slightly recognized at 2.69 GPa, but undetectable at 2.77 GPa as shown in Fig. 3(a). It is of interest that the resistivity follows nearly $T$-linear behavior above $T_c$ at 2.55 and 2.77 GPa ($P \sim P_c$) for samples with $RRR = 7$ and 15, respectively, as guided by a dashed line. A similar $T$-variation of resistivity was also reported in several optimally-doped Fe-pnictide superconductors [4, 30–32]. For the sample with $RRR = 7$, a resistivity upturn and a small maximum, as indicated by an arrow in Figs. 2(b), in the broad SC transition below 31 K are observed at $P = 2.38$ GPa ($< P_c$). It suggests that the superconductivity is suppressed by the magnetic order of the Eu$^{2+}$ moments; consequently, reentrant-SC-like behavior appears. A similar behavior is also slightly seen for the sample with $RRR = 15$ (Figs. 3(b)), but more smeared out. At $P > P_c$, resistivity exhibits sharp SC transitions to zero-resistivity with $T_c \sim 30$ K for both samples. With increasing $P$, the SC

![Figure 2. $\rho$ vs $T$ of a EuFe$_2$As$_2$ single crystal ($RRR = 7$) up to 3.0 GPa in the temperature ranges (a) 30 – 120 K and (b) 15 – 40 K. An arrow in (b) indicates an anomaly attributed to $T_N$.](image1)

![Figure 3. $\rho$ vs $T$ of a EuFe$_2$As$_2$ single crystal ($RRR = 15$) up to 3.0 GPa in the temperature ranges (a) 30 – 120 K and (b) 15 – 40 K. Arrows in (b) indicate anomalies attributed to $T_N$.](image2)
transitions persist up to 3.00 GPa although the $T_c$ continuously decreases. Thus, the $P$-variation of the resistive behavior between the samples with different quality is qualitatively similar to each other, and is consistent with the previous result [10]. However, $P_c \sim 2.7$ GPa for the sample with $RRR = 15$ is slightly but distinctly larger than $\sim 2.5$ GPa for the sample with $RRR = 7$, which may be as a consequence of the larger value of $T_0$ for the higher-quality sample at ambient-$P$. We have repeated similar high-$P$ resistivity measurements using several single crystals, and confirmed that the observed difference in the magnitude of $T_0$ and $P_c$ between the samples with $RRR = 7$ and 15 is beyond the error of the pressure estimation ($\pm 2 - 3 \times 10^{-2}$ GPa) [28]. Another meaningful issue, which probably relates to the sample quality, is the width of a SC transition $\Delta T_c$. The minimum values of $\Delta T_c$ are 1 K and 0.8 K for samples with $RRR = 7$ and 15, respectively. These facts suggest that the higher-quality single crystals have larger values of $T_0$ and $P_c$ as well as a sharper SC transition in EuFe$_2$As$_2$.

Until now, there has been no report concerning the quantum oscillation in EuFe$_2$As$_2$, despite the importance for understanding the Fermi surface topology and mass renormalization. In fact, we have already tried de Haas-van Alphen (dHvA) measurements of EuFe$_2$As$_2$ using the samples with $RRR = 7$ at 0.6 K with fields up to 35 T, but could not detect any dHvA oscillation. Given that quantum oscillations were successfully detected in SrFe$_2$As$_2$ ($RRR \sim 8$) [33] and BaFe$_2$As$_2$($RRR = 10$) [34], it is worthwhile to perform the dHvA measurement of EuFe$_2$As$_2$ using the newly grown single crystals with $RRR = 15$.

4. Conclusions
We have performed high-pressure electrical resistivity measurements up to 3.0 GPa in EuFe$_2$As$_2$ single crystals with $RRR = 7$ and 15. At ambient pressure, a magnetic/structural transition occurred at $T_0 = 190$ K and 194 K for the samples with $RRR = 7$ and 15, respectively. Although $P$-induced superconductivity was confirmed in the samples with different $RRR$ values, the critical pressure $P_c \sim 2.7$ GPa for the samples with $RRR = 15$ was slightly but distinctly larger than $\sim 2.5$ GPa for the samples with $RRR = 7$.

References
[1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Ishida K, Nakai Y and Hosono H 2009 J. Phys. Soc. Jpn. 78 062001 and references therein.
[3] Chen H, Ren Y, Qiu Y, Bao W, Liu R H, Wu G, Wu T, Xie Y L, Wang X F, Huang Q and Chen X H 2009 Europhys. Lett. 85 17006
[4] Wang X F, Wu T, Wu G, Liu R H, Chen H, Xie Y L and Chen X H 2009 New J. Phys. 11 045003
[5] Ren Z A, Lu W, Yang J, Yi W, Shen X L, Li Z C, Che G C, Dong X L, Sun L L, Zhou F and Zhou Z X 2008 Chin. Phys. Lett. 25 2215
[6] Alireza P L, Ko Y T C, Gillett J, Petrone C M, Cole J M, Lonzarich G G and Sebastian S E 2009 J. Phys.: Condens. Matter 21 012208
[7] Kotegawa H, Sugawara H and Tou H 2009 J. Phys. Soc. Jpn. 78 013709
[8] Matsubayashi K, Katayama N, Ohgushi K, Yamada A, Munakata K, Matsumoto T and Uwatoko Y 2009 J. Phys. Soc. Jpn. 78 073706
[9] Miclea C F, Nicklas M, Jeevan H S, Kasinathan D, Rossin Z, Rosner H, Gegenwart P, Geibel C and Steglich F 2009 Phys. Rev. B 79 212509
[10] Terashima T, Kimata M, Satsukawa H, Harada A, Hazama K, Uji S, Suzuki H S, Matsumoto T and Murata K 2009 J. Phys. Soc. Jpn. 78 083701
[11] Torikachvili M S, Bud’ko S L, Ni N and Canfield P C 2008 Phys. Rev. Lett. 101 057006.
[12] Park T, Park E, Lee H, Klimczuk T, Bauer E D, Ronning F and Thompson J D 2009 J. Phys.: Condens. Matter 20 322004
[13] Lee H, Park E, Park T, Sidorov V A, Ronning F, Bauer E D and Thompson J D 2009 Phys. Rev. B 80 024519
[14] Yu W, Aczel A A, Williams T J, Bud’ko S L, Ni N, Canfield P C and Luke G M 2009 Phys. Rev. B 79 020511
[15] Ishikawa F, Eguchi N, Kodama M, Fujimaki K, Einaga M, Ohmura A, Nakayama A, Mitsuda A and Yamada Y 2009 Phys. Rev. B 79 172506
