Pressure-induced superconductivity in lightly doped RFeAsO$_{1-x}$F$_x$ (R=Sm and Nd)

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Abstract. We performed the measurements of electrical resistivity ($\rho$) and dc magnetization ($M$) as a function of temperature ($T$) under pressure to investigate whether or not the superconductivity is induced by the application of pressure in lightly doped RFeAsO$_{1-x}$F$_x$ (R=Sm and Nd) using pulse current sintered (PCS) high density polycrystalline specimens. We have successfully observed pressure induced superconductivity with $T_c$ of $\sim 6$ K ($\sim 8$ K) above $\sim 4$ GPa ($\sim 1$ GPa) for R=Sm (R=Nd). An anomaly corresponding to a magnetic phase transition into spin density wave (SDW) state was observed in the $\rho(T)$ curves in the entire pressure range, indicating that the superconducting and SDW states coexist in the $T$-$P$ phase diagram. Volume fraction of the superconductivity is found to be very small. This is consistent with the coexistence of the superconducting and SDW phases.

1. Introduction

Since LaFeAsO$_{1-x}$F$_x$ has been found to exhibit superconductivity below $T_c$$\sim 26$ K[1], intensive studies have been made and superconductivity has been discovered in materials which have iron-pnictide (Fe-Pn) layers consisting of edge-sharing FePn$_4$ tetrahedron, such as RFeAsO$_{1-x}$F$_x$ (R=lanthanide), Ba$_{1-x}$K$_x$Fe$_2$As$_2$, AFeAs (A=Li, Na), FeSe. It is important to establish temperature ($T$) versus pressure ($P$) phase diagrams of iron-pnictide superconductors, where superconducting, antiferromagnetic (AFM) and nematic states coexist or compete, to throw further light on the superconducting mechanism. Indeed, in the $T$-$P$ phase diagram of FeSe, superconducting phase coexists with nematic phase for $0 \leq P \leq 1.5$ GPa and with dome-shaped AFM phase for $1.5 \leq P \leq 6$ GPa but highly enhanced above 6 GPa[2, 3]. In RFeAsO$_{1-x}$F$_x$ (R=lanthanide), the $T$-$P$ phase diagram however remains solved, although several studies have been conducted for R=La[4, 5, 6], Ce[7, 8]. To study the relationship between the superconducting and other two phases in the phase diagram of RFeAsO$_{1-x}$F$_x$, we need to prepare lightly doped specimens so that the superconductivity is induced only above a critical pressure $P_c$, since the SDW and nematic phases do not appear in optimally doped superconducting specimens. Thus, to discover pressure induced superconductivity in RFeAsO$_{1-x}$F$_x$ is the first step to establish the $T$-$P$ phase diagram of RFeAsO$_{1-x}$F$_x$. In the present work, we performed the measurements of electrical resistivity and dc magnetization under pressure for lightly doped RFeAsO$_{1-x}$F$_x$ with R=Sm and Nd using high density polycrystalline specimens obtained by pulse current sintering (PCS) and report the pressure induced superconductivity.
2. Experiment
High density PCS specimens of RFeAsO$_{1-x}$F$_x$ (R=Sm and Nd) were synthesized by the same procedure adopted in the previous study[6]. dc magnetization measurements under pressure were performed by using a miniature diamond anvil cell[9] with glycerin as pressure transmitting media (PTM), which was combined with a sample rod of commercial SQUID magnetometer. A small piece of high-purity lead (Pb) to determine the pressure from the $T_c$ shift and RFeAsO$_{1-x}$F$_x$ were loaded into a sample room of 0.3-0.4 mm in diameter and 0.2 mm in height in a CuBe gasket. The magnetization data for RFeAsO$_{1-x}$F$_x$ and Pb were obtained by subtracting the magnetic contribution of the DAC measured in an empty run from the total magnetization[3, 11, 10]. Electrical resistivity under pressure was measured by using an indenter-type pressure cell for R=Sm and piston-cylinder cell for R=Nd with Daphne oil 7373 as PTM.

3. Results and Discussion

To observe pressure induced superconductivity in RFeAsO$_{1-x}$F$_x$, we need to prepare the specimens with proper F-content $x$. As seen in a previous study on LaFeAsO$_{1-x}$F$_x$[6], if $x$ is much lower than the critical value $x_c$ above which the specimens exhibits superconductivity at ambient pressure, then we would fail to observe pressure induced superconductivity in the specimens, since the pressure we can generate is actually limited within $\sim 10$ GPa. According to our verification of superconductivity at ambient pressure for several specimens with various $x$, we assumed that $x_c$ is around 0.10 for both R=Sm and Nd. Thus, we prepared the specimens with $x=0.09$ for R=Sm and $x=0.08$ for R=Nd and verified the pressure induced superconductivity for the specimens in the present study.

We show temperature dependence of resistivity ($\rho$) at various pressures for R=Sm and Nd in Figs. 1(a) and 1(b), respectively. In Fig. 1(a), the $\rho(T)$ curve for R=Sm at ambient pressure shows a rapid decrease below $T_{\text{SDW}} \sim 120$ K, corresponding to the transition into the SDW phase, which is known to be $\sim 140$ K for a non-doped specimen[12]. As shown in the inset, a sharp drop begins to appear in the $\rho(T)$ curve above 3 GPa below $T_{\text{onset}}$, which is $\sim 5$ K but increases as increasing pressure, suggesting a pressure induced superconducting transition. The $\rho(T)$ curve for R=Nd at ambient pressure in Fig. 2(b) is qualitatively similar to that for R=Sm. In the inset, a downturn probably corresponding to the superconductivity is seen in the $\rho(T)$ curve even at ambient pressure below $T_{\text{onset}} \sim 8$ K, which gradually increases as increasing pressure.

To confirm the superconductivity observed by the $\rho(T)$ measurements under pressure, zero-field cooled dc magnetization ($M$) measurements were performed under pressure. The results are shown in Figs. 2(a) and 2(b). In Fig. 2(a), the $M(T)$ curve for R=Sm is found to exhibit a sharp drop below $\sim 7$ K at ambient pressure, which corresponds to a superconducting transition.
of Pb used as a manometer. At 4.4 GPa, the $M(T)$ curve however appears to exhibit a small downturn below $T_{c\text{dia}} \sim 8$ K, just above the $T_c$ of Pb, which is similarly seen in each $M(T)$ curve above 4.4 GPa, suggesting the onset of the superconductivity induced in the specimen by the application of pressure. A similar behavior is observed in the specimen with R=Nd in Fig. 2(b), where the superconductivity is found to be induced by the pressure above 0.74 GPa. As seen in the figures, it is found that the amplitude of diamagnetic response of the specimens is much smaller than that of Pb, even though the volume of the specimens is much larger than that for Pb. Thus, we roughly estimated the volume fraction of the superconductivity $p$ from the diamagnetic amplitude obtained with extrapolating it to $T=0$. Assuming that the sample room is half filled with the sample, we obtained $p \sim 5\%$ for R=Sm and $\sim 2.5\%$ for R=Nd, while $p \sim 10\%$ for R=La in a previous study[6]. These results suggest that the pressure induced superconductivity in RFeAsO$_{1-x}$F$_x$ is filamentary one.

Figures 3(a) and 3(b) show plots of $T^{\text{onset}}$, $T_{c\text{dia}}$ and $T_{SDW}$ as a function of pressure. $T_{SDW}$ was estimated by extrapolating the initial slope of the $\rho(T)$ curve just below $T_{SDW}$ to that above $T_{SDW}$. In the figures, $T_{SDW}$ for R=Sm decreases slightly as increasing temperature with a pressure coefficient of $dT_{SDW}/dT \sim -2.5$ K/GPa, whereas $dT_{SDW}/dT$ is $+1.4$ K/GPa for R=Nd. $T^{\text{onset}}$ and $T_{c\text{dia}}$ agree with each other, suggesting that both temperatures well correspond to the onset of the superconductivity. We note that $T^{\text{onset}}$ is always higher than $T_{c\text{dia}}$ and a resistive drop begins to appear at lower pressure than the pressure at which a diamagnetic drop appears. Thus, we could say that electrical resistivity is more sensitive for detecting a formation of superconducting network. The superconductivity with small volume fraction is probably due to the coexistence with the SDW phase. We could expect that the SDW phase disappears by the application of much higher pressure especially for R=Sm, since $dT_{SDW}/dT$ is negative for R=Sm. In this case, the superconducting phase would be enhanced having a maximum $T_c$ at the pressure...
where the SDW phase disappears due to the enhancement of the antiferromagnetic fluctuation, if SmFeAsO$_{1-x}$F$_x$ hosts a quantum critical point, such as SrFe$_2$As$_2$[13, 14], BaFe$_2$As$_2$[14] and BaFe$_2$(As$_{1-x}$P$_x$)[15]. Although $dT_{SDW}/dT$ is very small but positive for R=Nd and La[6], similar features are expected under much higher pressure. Experiments under high pressure above 10-20 GPa are necessary to establish the $T-P$ phase diagram and verify the quantum criticality.

4. Summary
In the present work, we have performed dc magnetization and electrical resistivity measurements under pressure for lightly doped RFeAsO$_{1-x}$F$_x$ (R=Sm and Nd) using PCS specimens. Pressure induced superconductivity is successfully observed above 3.5–4.5 GPa for R=Sm and 0.5–1.0 GPa for R=Nd, but the volume fraction of the superconductivity is very small less than 5 % probably due to the coexistence of the SDW phase. The pressure coefficient is found to be very small so that much higher pressure is necessary to establish the $T-P$ phase diagram.

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