Resistivity, magnetic susceptibility and specific heat studies in superconductor LaFePO$_{1-x}$F$_x$

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Abstract. We have investigated the transport, magnetic and thermal properties in LaFePO$_{1-x}$F$_x$ to clarify the superconducting and normal state characters of this system. Above the critical temperature for superconductivity ($T_c$), the low-temperature ($T$) resistivity is proportional to $T^2$, and the electronic specific heat coefficient has large value, indicating the existence of electron-electron correlation in this system. The resistive transition to the superconducting state has been broadened, as the applied magnetic field is increased. This resistive broadening is characteristic of the superconductivity in the two-dimensional electron systems such as high-$T_c$ cuprates.

1. Introduction

Since the discovery of high-$T_c$ superconductivity in cuprates, much experimental and theoretical effort has been made to explore similar phenomena in other transition metal compounds with layered structures, and many superconducting (SC) systems, such as YNi$_2$B$_2$C [1], Sr$_2$RuO$_4$ [2], NaCoO$_2$.H$_2$O [3] and so on, have been found. Recently, superconductivity in the iron- and nickel-based oxypnictides, LaFePO [4] and LaNiPO [5] were reported. In the same family of iron-based oxypnictides, Y. Kamihara and co-workers have found a new superconductor, LaFeAsO$_{1-x}$F$_x$, with $T_c$=26 K [6]. Since then, the research on this system have been intensified, and the superconductivity with higher critical temperature around 50 K have been discovered in other rare-earth systems, RFeAsO$_{1-x}$F$_x$ (R=rare earth elements) [7].

The iron-based oxypnictides, LaFePO and RFeAsO, are composed of alternant stacked Fe-P/As and R-O layers. The charge carrier move in the two-dimensional Fe-P/As layers and the R-O ones play a similar role to that of the insulating block layer in high-$T_c$ cuprates. The substitution of F$^-$ for O$^2-$ or the deficiency of oxygen in the charge reservoir layers of R-O introduces the charge carriers (electrons) to the conductive layers of Fe-P/As. LaFePO shows a SC behavior below $T_c$=4 K [4], and a paramagnetic metallic one above $T_c$. The substitution effect of F$^-$ for O$^2-$ increases $T_c$ in LaFePO$_{1-x}$F$_x$. On the other hand, superconductivity does not appear down to the lowest temperature, but long-range magnetic order occurs around 150 K in RFeAsO, as temperature ($T$) is lowered. In RFeAsO systems, the electron doping causes the magnetic disorder and the superconductivity at low temperatures [6].

In the iron-based oxypnictides, not only the mechanism of superconductivity but also the electronic, magnetic and structural properties have not yet clarified. We have studied the resistivity, magnetic susceptibility and specific heat in LaFePO$_{1-x}$F$_x$ to clarify the electronic and magnetic features of the normal and SC states in this system.
2. Experimental procedures

The polycrystalline samples of LaFePO and LaFePO$_{1-x}$F$_x$ ($x=0.10$) were synthesized by using a two-step solid-state reaction. The powder of precursor LaP was first prepared by solid states reaction using La and P as starting materials. The mixed powders of the starting materials were reacted at 400 and 700 °C in evacuated quartz tubes. Then the prepared LaP powder was mixed with Fe, Fe$_2$O$_3$ and LaF$_3$, and pressed into pellets. The samples were sealed in evacuated quartz tubes and sintered at 1100 °C for 48 hours. The obtained samples were pulverized and checked by powder x-ray diffraction. The results of the powder x-ray diffraction analysis indicated that the samples were a single phase.

Magnetic susceptibility measurements were performed with a SC quantum interference device magnetometer. Measurements of $T$-dependent resistivity were carried out in the magnetic field up to 7 T by a conventional four probe method. Specific-heat measurements were performed by a relaxation technique down to 1.8 K.

3. Experimental results and discussion

Figure 1 presents the $T$ dependence of resistivity for LaFePO$_{1-x}$F$_x$ with $x=0$ (LaFePO) (solid line) and 0.10 (broken line), respectively. The inset shows the plot of $\rho-\rho_0$ vs. $T^2$, where $\rho_0$ is residual resistivity.

Figure 1. Temperature ($T$) dependence of resistivity ($\rho$) for LaFePO$_{1-x}$F$_x$ with $x=0$ (LaFePO) (solid line) and 0.10 (broken line), respectively. The inset shows the plot of $\rho-\rho_0$ vs. $T^2$, where $\rho_0$ is residual resistivity.
Figure 2(d) presents the $T$-dependent specific heat ($C$) for LaFePO$_{1-x}$F$_x$ with $x=0$ and 0.10, plotted as $C/T$ v.s. $T^2$. Above $T_c$, the $T$ dependence can be well described by $C/T = \gamma + \beta T^2$, and the electronic specific heat coefficient $\gamma$ are 9.6 mJ/K mol for $x=0$ and 10.7 mJ/K mol for $x=0.10$, respectively. These values of $\gamma$ for LaFePO$_{1-x}$F$_x$ are similar to those by the previous reports [8,9]. The $\gamma$ of the present result is approximately 2.5 times larger than that estimated from the band calculation [10], indicating the mass enhancement due to the electron-electron correlation. As shown in figure 2(c), the $C/T$ for LaFePO$_{1-x}$F$_x$ has a peak associated with the SC transition. However, the change of the specific heat caused by SC transition is much smaller than that expected from the BCS weak-coupling limit ($\Delta C = 1.43 \gamma T_c$). In addition, the residual $\gamma$ is observed at lowest $T$. These behaviors of $C$ may suggest the existence of minority impurity and/or the coexistence of SC and normal states at low temperatures due to the inhomogeneity.

Figure 3 shows the $T$-dependent resistivity for LaFePO$_{1-x}$F$_x$ with $x=0$ and 0.10 in the magnetic field up to 7 T. With increasing the magnetic field, the SC transition temperature $T_c$ shifts to lower $T_c$ and the transition gradually broadens. The broadening of the resistive transition between the SC and normal states by applying the magnetic field has been often observed in high $T_c$ cuprates. The present results for LaFePO$_{1-x}$F$_x$ have indicated the large anisotropy of the critical field of superconductivity, as expected from the two-dimensional electronic structure [10]. We roughly estimate the upper critical field at zero $T$, $H_{c2} \sim 5$ T for $x=0$ and $H_{c2} \sim 7$ T for $x=0.10$, respectively.

4. Conclusion

We have prepared the polycrystalline samples of LaFePO$_{1-x}$F$_x$ with $x=0$ and 0.10, and investigated the resistivity, magnetic susceptibility, and specific heat to clarify the SC and normal state features of this system. Above $T_c$, the resistivity is distinctly proportional to $T^2$, and the electronic specific heat coefficient $\gamma$ has large magnitude indicating the mass enhancement. The present results reveal that the
electron-electron correlation plays an important role in this system. As applying the magnetic field, the resistive transition to the SC state broadens, indicating the large anisotropy of the critical field for superconductivity.

![Figure 3. Temperature dependence of resistivity in the various magnetic fields for LaFePO$_{1-x}$F$_x$ with (a) $x=0$ and (b) $x=0.10$.](image)

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