Pressure effect on superconducting properties of LaO$_{1-x}$F$_x$FeAs ($x = 0.11$) superconductor

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Abstract. Diamagnetic susceptibility measurements under high hydrostatic pressure (up to 1.03 GPa) were carried out on a newly discovered Fe-based superconductor LaO$_{1-x}$F$_x$FeAs ($x = 0.11$). The transition temperature $T_C$, defined as a point at the maximum slope of superconducting transition, was enhanced almost linearly by hydrostatic pressure, yielding a $dT_C/dP$ of about 1.2 K GPa$^{-1}$. Differential diamagnetic susceptibility curves indicate that the underlying superconducting state is complicated. It is suggested that pressure plays an important role on pushing the low $T_C$ superconducting phase toward the main (optimal) superconducting phase.

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1. Introduction

A newly discovered LaO$_{1-x}$F$_x$FeAs superconductor with critical temperature $T_C$ of 26 K has stimulated extensive interest in the layered rare-earth metal oxypnictide family which is free of copper, aiming at pursuing higher $T_C$ or uncovering new evidence on its mechanism [1]–[8]. The fact that $T_C$ varies, via lattice site substitution, from 4 to 26 K and especially to the very recently reported 43 K [8] further implies a possible rich superconducting phase diagram of this new layered superconductor family of ZrCuSiAs-type structure. Besides element substitution, high pressure has often served as an effective parameter to raise the $T_C$ by varying carrier concentration through compressing the lattice. In addition, high-pressure experiments, by changing the interaction between particles, often play a very important role in providing new information on pairing interaction and testing of a proposed theory [9]. Here, we report the pressure effect on the newly discovered LaO$_{1-x}$F$_x$FeAs superconductor through magnetic susceptibility measurements. The transition temperature $T_C$ was defined as a point where maximum slope of superconducting transition is achieved, i.e. the peak of differential diamagnetic susceptibility. $T_C$ was found to increase almost linearly upon increasing the hydrostatic pressure, showing a $dT_C/dP$ value of about 1.2 K GPa$^{-1}$. Differential diamagnetic susceptibility curves indicate that the underlying superconducting state is complicated, at least two superconducting transitions coexist. The lower $T_C$ is dramatically enhanced by pressure, while the higher one shows little pressure dependence. The role of pressure is hence discussed based on its different effects on the superconducting transitions that exist.

2. Experimental details

The F-doped LaO$_{1-x}$F$_x$FeAs polycrystalline sample was synthesized by a two-step method. La$_2$O$_3$, Fe, As, LaF$_3$ powders and La pieces (the purities of all chemicals were higher than 99.99%) were used as starting materials and weighed according to the chemical stoichiometry of LaO$_{1-x}$F$_x$FeAs. As a first step, the precursor LaFeAs alloy was prepared by the argon arc-melting method. The weighed Fe and As powders (here, 20% As was added to compensate for the volatilization during alloying) were pressed into pellets and put on a water-cooled copper stage together with the La pieces. The mixtures were then melted and alloyed for 2 min. The alloyed ingot became brittle and was ground to fine powder. The second step was performed through a solid-state reaction process. The prepared LaFeAs powder was mixed with weighed LaF$_3$ and La$_2$O$_3$ powders, then ground thoroughly and pressed into pellets. After being enclosed in evacuated quartz tubes (with a vacuum better than 10$^{-3}$ Pa), the sealed sample was sintered at 1240°C for 50 h in a muffle furnace, with heating and cooling rates of 100°C h$^{-1}$. The sample for measurements was processed into approximately 1.34 × 1.1 × 0.4 mm$^3$ rectangular shape.

The structure of the synthesized samples was characterized by high-power x-ray diffraction (XRD, an 18KW MXP18A-HF diffractometer with Cu–Kα radiation). Scanning electronic microscopy (SEM) analysis was performed by using a Philips XL30 S-FEG microscope. The resistivity data were measured by the standard four-probe technique.

The hydrostatic pressure was generated by a commercial pressure cell (Mcell 10) which was especially designed for magnetic property measurement using a Quantum Design magnetic property measurement system (MPMS). The pressure was measured in situ by recording the $T_C$ shift of a small piece of Sn included in the Mcell 10. The diamagnetic susceptibilities ($\chi$) were
Figure 1. Left: powder XRD patterns of F-doped La[O$_{0.89}$F$_{0.11}$]FeAs. The results from our experiment and theoretical simulation are in good agreement except for two additional phases from the impurities LaOF and FeAs$_2$. Right: SEM image of F-doped La[O$_{0.89}$F$_{0.11}$]FeAs showing the layered structure feature with a grain size of about 10 $\mu$m.

obtained by magnetization measurements carried out during the warming cycle under fixed magnetic field after zero-field cooling (ZFC) or field cooling (FC) using a Quantum Design MPMS XL-1. Background signals were subtracted simultaneously during the measurements through a commercial functional program. All the data reported here were corrected for the demagnetization factor [10].

3. Results and discussion

The powdered sample’s XRD pattern shown in figure 1(a) indicated a well-indexed tetragonal structure (space group P4/nmm) with $a = 4.030(1)$ Å and $c = 8.706(2)$ Å except for weak impurity peaks from LaOF and FeAs$_2$. An SEM image of our sample is presented in figure 1(b), showing the layered structure feature with a grain size of about 10 $\mu$m. Figure 2(a) shows the temperature dependence of resistivity. A sharp transition starts at 26.3 K and zero resistivity is reached when the temperature is lowered to 22.3 K. The temperature-dependent diamagnetic susceptibility $\chi$ measured under 1 Oe after ZFC is shown in figure 2(b). The onset $T_C$ of 23 K is consistent with the resistivity results, while the 100% magnetic shielding signal is evidence for bulk superconductivity. All these results indicate the good quality of our sample.

Differential diamagnetic susceptibility was focused to monitor any tiny change in order to investigate the pressure effect on the superconducting properties of this newly discovered superconductor. Shown in figure 3 is the differential diamagnetic susceptibility $d\chi/dT$ measured at 1 Oe under hydrostatic pressure. $T_C$, defined as the peak of $d\chi/dT$, showed obvious pressure dependence. With pressure increasing from ambient up to 1.03 GPa, $T_C$ raised almost linearly from 20.3 to 21.5 K, yielding a $dT_C/dP$ value of about 1.2 K GPa$^{-1}$ (the inset of figure 3). To our surprise, the onset $T_C$ showed no obvious change when the pressure was increased up to 1.03 GPa. In contrast, the superconducting transition, illustrated by the $d\chi/dT$
Figure 2. (a) Electrical resistivity ($\rho$) versus temperature. Resistivity begins to drop dramatically below 26.3 K and zero resistivity appears at 22.3 K. (b) Temperature dependence of diamagnetic susceptibility measured at 1 Oe under ambient pressure, the onset $T_C$ is 23 K.

Figure 3. Differential curves of diamagnetic susceptibility ($d\chi/dT$) measured under 1 Oe after ZFC at different hydrostatic pressures. Inset shows the pressure dependence of $T_C$ defined as the peak of ($d\chi/dT$), yielding a $dT_C/dP$ (the slope of the red line) of 1.2 K GPa$^{-1}$. 
Figure 4. Differential curves of diamagnetic susceptibility \(\frac{d\chi}{dT}\) measured under 10 Oe after ZFC at different hydrostatic pressures. The low-\(T\) peak \(T_{PL}\) of \(\frac{d\chi}{dT}\) shifts toward high temperature with increasing pressure and gives a \(dT_p/dP\) value of about 4 K GPa\(^{-1}\) (inset).

peak in figure 3, was narrowed with increasing pressure. In other words, it seems that pressure (below 1.03 GPa) first improves the superconducting correlation network before interfering in the main superconducting phase characterized by the onset \(T_C\) of 23 K.

In order to clarify the pressure effect on both \(T_C\) and the superconducting transition width, the pressure dependence of \(\frac{d\chi}{dT}\) measured at 10 Oe was also investigated, as plotted in figure 4. Although the magnetic field of 10 Oe is rather low, the \(\frac{d\chi}{dT}\) curves largely broadened and extra features occurred, implying a complicated superconducting state. Two peaks are evident, the temperature of the higher peak \(T_{PH}\) is around 19.2 K, whereas the lower one \(T_{PL}\) is located at 12.6 K under ambient pressure, indicating the existence of an inhomogeneous superconducting state. When pressure increases, the higher peak is retained at around 19.2 K with up to 1.03 GPa, showing no obvious pressure dependence. This is consistent with the pressure-insensitive onset \(T_C\) mentioned hereinbefore. However, dramatic pressure dependence is seen for the lower peak. The \(T_{PL}\) shift changes from 12.6 K under ambient pressure to about 17 K at 1.03 GPa, as shown in the inset of figure 4, generating a nearly linear pressure dependence with a large slope, i.e. \(dT_{PL}/dP\), of about 4 K GPa\(^{-1}\) (inset of figure 4). In other words, the low-\(T_C\) phase is more sensitive to pressure compared with the high-\(T_C\) phase, which is also the main (optimal) superconducting phase with the onset \(T_C\) of 23 K as illustrated in figure 2.

Clearly, the onset \(T_C\) of the optimal superconducting phase showed little pressure dependence, implying that a pressure of up to 1.03 GPa had little effect, either by varying carrier concentration or adjusting inter- and intra-plane interactions, via lattice compressing, on the structural frame that bears the optimal superconducting phase. However, notice that the pressure-induced improvement of the superconducting transition is obvious as presented above, we still expect the potential for higher \(T_C\) triggered by pressure above 1.03 GPa. In addition, the

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fact that the superconducting transition is very sensitive to applied field implies that pressure effects such as on lattice defects (especially in La$_2$(O,F)$_2$ layers) and grain boundaries as well as other possible weak links might also be considered.

4. Conclusion

In summary, we have studied the pressure dependence of diamagnetic susceptibilities of the newly discovered Fe-based superconductor LaO$_{1-x}$F$_x$FeAs ($x = 0.11$). The transition temperature $T_C$, manifested by the peak of $d\chi/dT$, increased linearly with hydrostatic pressure, yielding a $dT_C/dP$ value of about 1.2 K GPa$^{-1}$. The differential diamagnetic susceptibility curves indicated that the underlying superconducting state is complicated. Pressure was found to play a key role in pushing the low-$T_C$ superconducting phase toward the main (optimal) superconducting phase. Higher $T_C$ is expected by applying pressures larger than 1.03 GPa.

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