Suppression of $T_c$ by Zn impurity in the electron-type LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO system

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Abstract

The effect of non-magnetic Zn impurity on superconductivity in electron-type pnictide superconductor LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO is studied systematically. The optimally doped LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO without Zn impurity exhibits superconductivity at $T_c^{\text{mid}}$ of 13.2 K, where $T_c^{\text{mid}}$ is defined as the mid-point in the resistive transition. In the presence of Zn impurity, the superconducting transition temperature, $T_c^{\text{mid}}$, is severely suppressed. The result is consistent with the theoretic prediction on the effect of non-magnetic impurity in the scenario of $s_\pm$ pairing, but it is in sharp contrast to the previous report on the effect of Zn impurity in the F-doped systems. The possible interpretation of the different effects of Zn impurity on superconductivity in different systems is discussed.

Keywords: A. Superconductors, D. Defects, D. Superconductivity, D. Transport Properties, D. Magnetic properties

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1. Introduction

Great progresses have been made since the discovery of the first FeAs-based pnictide superconductor LaFeAsO$_{1-x}$F$_x$ (La-1111 system) [1]. Similar to the high-temperature superconducting (SC) cuprates, the parent compounds of the iron pnictides are antiferromagnetic [2, 3]. Superconductivity can be induced by various chemical doping methods, which introduces charge carriers and suppresses the antiferromagnetic (AFM) order [1, 4]. The relative phases of the SC order parameters in hole or electron pockets can be either positive ($s$-wave pairing) or negative ($s_\pm$-wave pairing) depending on the sign of the inter-Fermi pocket pair scattering amplitude or their Josephson coupling. The $s_\pm$-wave pairing is appealing with some experimental supports [25]. In addition, there are also evidences for nodal SC gap in FeAs-based superconductors [26].

The effect of non-magnetic impurity is dependent on the pairing symmetry and thus it can be used as a powerful probe to detect the pairing symmetry for unconventional superconductors. Non-magnetic impurity does not cause severe pair-breaking effect in a conventional $s$-wave superconductor according to the Anderson’s theorem [27]. In the $d$-wave superconducting cuprates, even a minimal amount of non-magnetic Zn doping can quench $T_c$ quickly [28]. In the $s_\pm$-wave state where the order parameters in hole and electron pockets have opposite signs, theoretical studies [29, 30] have predicted that non-magnetic impurities like Zn can severely suppress the SC transition temperature $T_c$, similar to the high-$T_c$ cuprates with $d$-wave pairing. Zinc element has a stable $d^{10}$ configuration in the alloy [31], and can serve as the best non-magnetic impurity for this study. However, there is a seemingly discrepancy between our early data in LaFeAsO$_{0.9}$F$_{0.1}$, where $T_c$ is robust to Zn-impurity [32], and a following report showed a severe suppression of $T_c$ due to Zn-impurity in the oxygen-deficient LaFeAsO$_{1-y}$ samples [33]. Further studies showed that the effect of Zn impurity on $T_c$ is strongly dependent on the charge doping level, i.e., $T_c$ increases with Zn content in the under-doped regime, remains unchanged in the optimally doped regime, and is severely suppressed in the over-doped regime in the LaFe$_{1-y}$Zn$_y$AsO$_{1-y}$F$_y$ system [34]. This finding reconciles the contradiction in the previous reports of Zn impurity effect and strongly suggests that a switch of pairing symmetry from an impurity-insensitive state to an impurity-sensitive state is possible.

In this paper, we study the effect of Zn impurity on superconductivity in an optimally Co-doped LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO system. We found that $T_c$ is severely suppressed with addition of Zn impurity although the system is in the optimally doped regime. This result is in contrast to the effect of Zn impurity in optimally F-doped La-1111 system [32]. The possible interpretations regarding to the SC pairing state are discussed.

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2. Experimental

Polycrystalline samples of LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO ($y = 0, 0.01, 0.02, 0.03, 0.04$) were synthesized by solid state reaction method. Details on the sample preparation can be found in the previous report\cite{32}. The phase purity of the samples was investigated by powder X-ray diffraction (XRD) using a D/Max-rA diffractometer with Cu-K$_\alpha$ radiation and a graphite monochromator. Lattice parameters were calculated by a least-squares fit using at least 20 XRD peaks.

The electrical resistivity was measured on bar-shaped samples using a standard four-probe method. The measurements of resistance and Hall effect were performed on a Quantum Design Physical Property Measurement System (PPMS-9). DC magnetization were measured on a Quantum Design Magnetic Property Measurement System (MPMS-5).

3. Results and Discussion

Figure 1(a) shows the XRD patterns of the LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO samples, and Figure 1(b) shows the variations of lattice parameters as a function of Zn content. All the XRD peaks can be well indexed with the tetragonal ZrCuSiAs-type structure, indicating that all the samples are single phase without foreign phases. Both the $a$-axis and $c$-axis increase slightly with the Zn content, consistent with the fact that the lattice constants of LaZnAsO are larger than those of LaFeAsO\cite{34}. These results indicate that Zn has successfully substituted partial Fe ions in the lattice.

Figure 2 shows the temperature dependence of resistivity for the LaFe$_{0.925-y}$Co$_{0.075}$Zn$_y$AsO samples. The resistivity around the SC transition is enlarged as shown in the Figure 2(b). According to our previous study, the LaFe$_{0.925}$Co$_{0.075}$AsO sample without Zn doping is optimally doped electron-type superconductor\cite{10}. The SC transition temperature $T_c$ (defined as the midpoint in the resistive transition) is 13.2 K. With the presence of Zn impurity, $T_c^{mid}$ is suppressed quickly. Meanwhile the normal state resistivity increases with Zn doping, and there is an obvious upturn at low temperatures just above $T_c$. 4% substitution of Fe by Zn has already quenched superconductivity ($T_c$ at least below 2 K). Recall that superconductivity is found to be robust to the Zn impurity in the optimally F-doped electron-type LaFeAsO system, and also in the optimally K-doped hole-type (Ba,K)Fe$_2$As$_2$, this result is quite surprising and suggests a crucial difference in the Co-doped pnictides where the chemical doping occurs directly in the conducting FeAs layers.

Figure 3 shows the temperature dependence of d.c. magnetic susceptibility measured under $H$ of 10 Oe. Without Zn doping, the 7.5% Co doping makes the system into optimally-doping state with a maximum $T_c^{mid}$ of 13.2 K. The sharp transition in the magnetic susceptibility below $T_c$ suggests bulk superconductivity and high homogeneity, consistent with the previous report\cite{10}. Actually the volume fraction of superconducting magnetic shielding even exceeds 100 % for this samples. The demagnetizing factor $N$ has not been taken into account, thus the volume fraction could be much over-estimated. With addition of Zn impurity, the transition in magnetic susceptibility moves to lower temperatures, consistent with the resistivity data. Meanwhile, the volume fraction of superconducting magnetic shielding becomes smaller.
Figure 4 shows temperature dependence of Hall coefficient $R_H$ for LaFe$_{0.925-x}$Co$_{0.075}$Zn$_y$AsO samples. For the optimally doped samples without Zn impurity, $R_H$ shows a sharp transition when it enter into superconducting state. There is a large negative peak just below $T_{c,mid}$, and then $R_H$ goes to zero with further decreasing temperature. Such a negative peak in $R_H$ below $T_c$ is often observed in high-quality high-$T_c$ cuprates and it is ascribed to the contribution from the motion of vortices [35]. The presence of Zn impurity diminishes this peak. Meanwhile, the high-temperature $R_H$ remains almost unchanged with Zn doping, indicating that Zn doping does not change the charge carrier density. This is consistent with the band calculation result which predicts that energy level of Zn 3d electrons are deep below the Fermi level[31]. Unlike Co doping, Zn doping does not induce extra mobile electrons into the conducting FeAs layers.

We plot $T_c$ versus Zn content ($x$) of LaFe$_{0.925-x}$Co$_{0.075}$Zn$_y$AsO and $T_c$ versus Co content ($x$) of LaFe$_{1-x}$Co$_x$AsO in Figure 5 to compare the effect of Zn impurity with that of Co doping. The data of $T_c$ with $x$ of LaFe$_{1-x}$Co$_x$AsO are taken from our previous report[10]. As we pointed out hereinafore, the maximum $T_c$ is reached at the optimally doping level $x = 0.075$ for this Co-doped electron-type La-1111 superconductor. When Co content is greater than 0.075, $T_c$ decreases gradually with a rate ($dT_c/dx$) of about -1.0 K per 1% of Co doping due to over-doping of charge carriers. However, it is obvious that the decreasing rate of $T_c$ with $y$ (Zn content) is much larger, about -3.0 K per 1% of Zn impurity, implied that there is fundamentally different mechanism of Zn doping. The result also supports our conclusion that the Zn doping does not induce extra mobile electrons to the conducting FeAs layers, whereas Co doping does. Actually the resistivity value increases quickly with increasing Zn content, and the upturn in resistivity at low temperature also becomes more obvious, implying that the Zn impurity is a very effective impurity scattering center and it causes severe SC pairing breaking effect in the Co-doped electron-type La-1111 systems. This finding is in sharp contrast to the previous report on the effect of Zn impurity in the optimally F-doped La-1111 system where $T_c$ is robust to the Zn impurity[32]. Nevertheless, a severe suppression of $T_c$ by Zn impurity is indeed observed in over-doped La-1111 systems [33, 34]. In previous study, a switch of pairing state from impurity-insensitive state (such as usual d-wave) to impurity-sensitive state (such as d-wave or $s_\pm$-wave) has been proposed when the F doping level is increased. In the Co-doped electron-type systems, the pairing state could become impurity-sensitive (e.g., d-wave or $s_\pm$-wave) even in the optimally doped level. Such a change in pairing state with different chemical doping (especially on different lattice site) strongly suggests that the even a very subtle change in the electronic structure could result in a different pairing state, i.e. the pairing state could be dependent on the chemical doping. This proposal is also supported by the previous reports on the measurements of SC gap. A nodal gap has been observed for the P-doped BaFe$_2$As$_2$ system[37] and also in over-doped KFe$_2$As$_2$ system[38] while full gaps have been observed in the optimally doped BaFe$_2$As$_2$ system[15]. Furthermore, we should also point out another possibility that the nature of Co doping could be disturbed by Zn impurity since both are substituted on the FeAs layer. Furthermore studies are required to clarify the different mechanisms of chemical doping.

4. Conclusion

In summary, we find a severe suppression of superconductivity by non-magnetic Zn impurity in the Co-doped electron-type La 1111 system LaFe$_{0.925-x}$Co$_{0.075}$Zn$_y$AsO. This finding is in contrast to the previous studies of Zn impurity effect in optimally F-doped system, although it is consistent with the theoretical prediction on the effect of non-magnetic impurity in the scenario of $s_\pm$ pairing. These results on the effects of Zn impurity strongly suggest that the pairing state could be different in the systems with different chemical doping. The difference in the electronic structure due to different chemical doping should be taken into account by the theoretical models.

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Figure 5: Variation of $T_{\text{mid}}$ with Zn content for the LaFe$_{1-x}$Co$_x$AsO system. The $T_{\text{mid}}$ versus Co content in the over-doped regime of the LaFe$_{1-x}$Co$_x$AsO system is also shown for comparison. The $T_{\text{mid}}$ data for the LaFe$_{1-x}$Co$_x$AsO system are taken from Ref. [10].

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