Anomalous impurity effects in the iron-based superconductor KFe$_2$As$_2$

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High-quality K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals have been grown by using KAs flux method. Instead of increasing the superconducting transition temperature $T_c$, through electron doping, we find that Co impurities rapidly suppress $T_c$ down to zero at only $x \approx 0.04$. Such an effective suppression of $T_c$ by impurities is quite different from that observed in Ba$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ with multiple nodeless superconducting gaps. Thermal conductivity measurements in zero field show that the residual linear term $\kappa_0/T$ only change slightly with 3.4% Co doping, despite the sharp increase of scattering rate. The implications of these anomalous impurity effects are discussed.

PACS numbers: 74.70.Xa, 74.25.fc, 74.62.Dh

The iron-based superconductors, as a second high-$T_c$ superconducting family after cuprates, have stimulated great interest since the symmetry and structure of the superconducting gap reflect the underlying electron pairing mechanism, extensive works have been done to clarify them. However, so far there is still no consensus on this issue, particularly the origin of the nodal gaps in KFe$_2$As$_2$ and its related compounds. In this paper, we try to recover the electron Fermi surface in KFe$_2$As$_2$ alternatively by Co doping, and expect the enhancement of $T_c$ and a crossover from nodal to nodeless superconducting state. However, unexpected doping effects are found in K(Fe$_{1-x}$Co$_x$)$_2$As$_2$. First, Co impurities suppress $T_c$ very rapidly, and at only $x = 0.042$ no superconductivity can be observed down to 50 mK. This rapid suppression of $T_c$ by impurities is different from that in Ba$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ with nodeless gaps. Secondly, thermal conductivity measurements show that the residual linear term $\kappa_0/T$ does not change much with 3.4% Co doping. These anomalous impurity effects may reflect the underlying nodal $s$-wave gap structure in KFe$_2$As$_2$.

K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals were grown by using KAs flux method. The starting materials for single crystal growth are KAs, Fe and Co powders. KAs flux was prepared by reacting stoichiometric K pieces and As powders at 200 °C for 4 hours. KAs, Fe and Co powders were carefully weighed according to the ratio KAs: Fe: Co = 12: 2-2x: 2x, with $x = 0.005, 0.010, 0.025$ and 0.050. After thoroughly grounding, the mixtures were put into alumina crucibles and then sealed in iron crucibles under 1.5 atm of pure argon gas. The sealed crucibles were heated to 900 °C in a tube furnace filled with inert gas and kept at 900 °C for 6 hours, then cooled slowly to 600 °C at 3 °C/h to grow single crystals. The
KAs flux was washed out by alcohol and shiny black crystals with typical dimensions of $2 \times 1 \times 0.03$ mm$^3$ were obtained. The single crystals are stable in air or alcohol for several days. The actual chemical compositions were determined by energy dispersive x-ray spectroscopy to be 0, 0.017, 0.034, 0.042 and 0.050 for the five nominal compositions, with a standard instrument error 10%. Below we will use the actual compositions. Resistivity and Hall coefficient were measured in a Physical Property Measurement System (PPMS, Quantum Design). Resistivity below 2 K and thermal conductivity were measured in a dilution refrigerator.

![FIG. 1](image.jpg)

**FIG. 1:** (Color online) The XRD patterns for K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals with $x = 0 - 0.050$. The inset shows $x$ dependence of the lattice parameter $c$ estimated from the data in the main panel.

Figure 1 plots the x-ray diffraction (XRD) patterns for K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals. Only reflections of (0 0 2$l$) show up, suggesting good orientation along $c$-axis for all the crystals. The lattice parameter $c$ was estimated from the XRD data and plotted in the inset of Fig. 1. For the pure KFe$_2$As$_2$, $c = 13.876$ Å is obtained, which is consistent with previous reports.²²,²³ It is found that $c$ decreases linearly with increasing Co doping. At $x = 0.050$, $c$ is reduced by ~0.1%.

Figure 2 shows the temperature dependence of resistivity for K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals. The data were taken down to 50 mK for $x = 0.042$ and 0.050 samples. The transition temperature $T_c$ is defined when the resistivity drops to 90% of the normal-state value. For the pure KFe$_2$As$_2$, $T_c = 4.4$ K and resistivity reaches zero at 4.2 K. The power-law fit $\rho(T) = \rho_0 + bT^\alpha$ between 5 and 25 K yields a residual resistivity $\rho_0 = 0.11 \pm 0.01$ $\mu\Omega$ cm and $\alpha = 1.89$, from which a residual resistance ratio RRR $= \rho(300K)/\rho_0 = 2017$ is estimated. The extremely low $\rho_0$ and high RRR show that this crystal is very close to the clean limit. The RRR decreases dramatically from 2017 to 19 with increasing Co doping from 0 to 0.050 (see Table I), suggesting that the Co doping leads to large impurity scattering. At the same time, $T_c$ decreases rapidly to zero at $x \approx 0.04$.

Previously, the major role of Co doping in iron-based superconductors is to introduce electrons into the FeAs layer, which is evident from the evolution of electron and hole pockets with $x$ in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$.³⁴,³⁵ The impurity effect of Co dopants is less concerned. That is why we try to use Co doping to recover the electron Fermi surfaces in KFe$_2$As$_2$ and expect the increase of $T_c$. However, the rapid suppression of $T_c$ with $x$ indicates that the impurity effect of Co dopants is playing the major role in K(Fe$_{1-x}$Co$_x$)$_2$As$_2$.

Recently, a weak $T_c$ suppression effect from transition-metal impurities was observed in Ba$_{0.5}$K$_{0.5}$(Fe$_{1-x}$M$_x$)$_2$As$_2$ ($M = \text{Mn, Ru, Co, Ni}$,...
Cu, and Zn)\textsuperscript{36}. In Fig. 3, we plot the normalized $T_c$ versus Co content for K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Ba$_{0.5}$K$_{0.5}$(Fe$_{1-x}$Co$_x$)$_2$As$_2$\textsuperscript{36}. For comparison, similar data of $d$-wave superconductors YBa$_2$(Cu$_{1-x}$Zn$_x$)$_2$O$_{6.93}$ ($T_c(x=0) = 92$ K), La$_{1.65}$Sr$_{0.15}$Cu$_{1-x}$Ni$_x$O$_4$ ($T_c(x=0) = 37.7$ K), and CeCo(Im$_{1-x}$Sn$_x$)$_3$ ($T_c(x=0) = 2.25$ K) are also plotted\textsuperscript{32–34}. It is clearly seen that the impurity effect on $T_c$ in K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is very different from that in nodeless $s$-wave superconductor Ba$_{0.5}$K$_{0.5}$(Fe$_{1-x}$Co$_x$)$_2$As$_2$, but mimics in those $d$-wave superconductors.

![FIG. 3: (Color online) The normalized $T_c$ vs impurity content \(x\) for K(Fe$_{1-x}$Co$_x$)$_2$As$_2$, s-wave superconductor Ba$_{0.5}$K$_{0.5}$(Fe$_{1-x}$Co$_x$)$_2$As$_2$, d-wave superconductors YBa$_2$(Cu$_{1-x}$Zn$_x$)$_2$O$_{6.93}$, La$_{1.65}$Sr$_{0.15}$Cu$_{1-x}$Ni$_x$O$_4$, and CeCo(Im$_{1-x}$Sn$_x$)$_3$\textsuperscript{32–34}. It is clearly seen that the impurity effect on $T_c$ in K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is very different from that in nodeless s-wave superconductor Ba$_{0.5}$K$_{0.5}$(Fe$_{1-x}$Co$_x$)$_2$As$_2$, but mimics in those d-wave superconductors.]

The rapid suppression of $T_c$ by Co doping in KFe$_2$As$_2$ is very unusual among all iron-based superconductors. As described earlier, recent ARPES and thermal conductivity experiments support nodal s-wave gap in KFe$_2$As$_2$. Theoretically, for a nodal s-wave superconductor, the impurities will make the gap more isotropic and do not effectively reduce the gap magnitude.\textsuperscript{40} This means its $T_c$ should not decrease in such a rapid manner. However, recently we also find similar rapid suppression of $T_c$ by Co doping in LiFeP and LaFePO. Therefore, the anomalous impurity effect on $T_c$ may be an intrinsic property of these stoichiometric iron-based superconductors with nodal gap. In fact, there was no any real nodal s-wave superconductor before the discovery of iron-based superconductors. In this sense, further theoretical investigation is needed to revisit the impurity effect on the $T_c$ of nodal s-wave superconductor.

An extraordinary feature of $d$-wave superconductor is the universal heat conduction upon increasing the impurity level, which has been clearly demonstrated in YBa$_2$(Cu$_{1-x}$Zn$_x$)$_2$O$_{6.9}$\textsuperscript{32,34}. The universal heat conduction results from the compensation between the increase of nodal quasiparticle density induced by impurities and the decrease of mean free path. For an accidental nodal s-wave superconductor, the heat conduction should not be universal.\textsuperscript{32} Reid et al. observed universal heat conduction by comparing clean and dirty KFe$_2$As$_2$ single crystals with 10 times difference of $\rho_0$, and argued that it is a compelling evidence for $d$-wave gap.\textsuperscript{32} With the intentional Co doping, here we also check whether the heat conduction in K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is universal.

![FIG. 4: (Color online) The thermal conductivity of K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ with (a) \(x = 0\), and (b) \(x = 0.034\) in zero magnetic field. The solid lines are fits to $\kappa/T = a + bT^2$ for $x = 0$ sample, and $\kappa/T = a + bT$ for $x = 0.034$ sample, respectively. The dash lines are the normal-state Wiedemann-Franz law expectation $L_0/\rho_0$, with $L_0$ the Lorenz number $2.45 \times 10^{-8}$ W/K$^2$ and normal-state $\rho_0 = 0.116$ and $3.56$ $\mu$\Omega cm, respectively. For the $x = 0$ sample, its $L_0/\rho_0$ is divided by 5 to put into the panel.]

Fig. 4 shows the thermal conductivity of superconducting K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals with $x = 0$ and 0.034 in zero magnetic field. For the pure KFe$_2$As$_2$ with $\rho_0 = 0.116$ $\mu$\Omega cm, the data below 0.3 K can be fitted by $\kappa/T = a + bT^2$, with the residual linear term $\kappa_0/T = a = 4.94 \pm 0.09$ mW K$^{-2}$ cm$^{-1}$ and $b = 283 \pm 2$. However, for the $x = 0.034$ sample with $\rho_0 = 3.56$ $\mu$\Omega cm, the data below 0.4 K obeys $\kappa/T = a + bT$, with $\kappa_0/T = a = 3.95 \pm 0.01$ mW K$^{-2}$ cm$^{-1}$ and $b = 2.43 \pm 0.06$. Previously, for a dirty KFe$_2$As$_2$ with $\rho_0 = 3.32$ $\mu$\Omega cm, similar temperature dependence of $\kappa/T = a + bT$ was observed, with $b = 3.04$. Therefore, the $T^2$ term of $\kappa$ is attributed to phonons, and the huge $T^3$ term observed in the clean KFe$_2$As$_2$ must come from the nodal quasiparticles. While the origin of this electronic $T^3$ term is still not clear,\textsuperscript{33,31} it is apparently suppressed by impurities in the K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ $x = 0.034$ sample.

From Fig. 4, while the $\rho_0$ is increased by 30 times, the $\kappa_0/T$ of the $x = 0$ and 0.034 samples in zero field remain comparable (4.94 and 3.95 mW K$^{-2}$ cm$^{-1}$). For the $x = 0$ sample, the value is less than 3% of its normal-state
Wiedemann-Franz law expectation $\kappa/T = L_0/\rho_0$, with $L_0$ the Lorenz number $2.45 \times 10^{-8}$ WΩK$^{-2}$. For the dirty $x = 0.034$ sample, the value is more than 50% of its normal-state $\kappa/T$. Therefore, roughly universal heat conduction is observed in K(Fe$_{1-x}$Co$_x$)$_2$As$_2$. Although the theoretical model with some assumed nodal s-wave gap structure suggests that the thermal conductivity is nonuniversal, it is hard to conclude that this comparable heat conduction is an evidence of $d$-wave gap structure in KFe$_2$As$_2$. First, the superconducting gap structure of KFe$_2$As$_2$ is very complex, e.g., the “$\gamma$”-shaped nodal gap at the tip of the off-M-centered hole lobes. Second, the impurity may slightly change the doping level, which can also cause a dramatic change of $\kappa_0/T$. One needs to carefully evaluate whether different effects can finally bring some kind of compensation on thermal conductivity in KFe$_2$As$_2$.

In summary, we have grown high-quality K(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals and find that the superconductivity was suppressed quickly by Co doping at only $x \approx 0.04$. Such an effective suppression of $T_c$ by impurities is very different from that in nodeless s-wave superconductor Ba$_{0.3}$K$_{0.7}$Fe$_2$As$_2$, and it may be an intrinsic property of stoichiometric iron-based superconductors with nodal gap. A comparable heat conduction is observed in the $x = 0$ and 0.034 superconducting samples, however, a careful evaluation of different effects on $\kappa_0/T$ is needed, taking into account the complex nodal s-wave superconducting gap structure of KFe$_2$As$_2$.

This work is supported by National Natural Science Foundation of China (Grant No. 11190021, 51021091, and 91021016), the National Basic Research Program of China (973 Program, Grant No. 2012CB922002, 2011CB001, and 2012CB824102), Chinese Academy of Sciences, and the Program for Professor of Special Appointment (Eastern Scholar) at Shanghai Institutions of Higher Learning.

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