Copper and Nickel impurities have been doped into the iron pnictide superconductor \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \). Resistivity measurements reveal that Cu and Ni impurities suppress superconducting transition temperature \( T_c \) with rates of \( \Delta T_c / \text{Cu-1\%} = -3.5 \text{ K} \) and \( \Delta T_c / \text{Ni-1\%} = -2.9 \text{ K} \) respectively. Temperature dependence of Hall coefficient \( R_H \) of these two series of samples show that both Cu-doping and Ni-doping can introduce electrons into \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \). With more doping, the sign of \( R_H \) gradually changes from positive to negative, while the changing rate of Cu-doped samples is much faster than that of Ni-doped ones. Combining with the results of first-principles calculations published previously and the non-monotonic evolution of the Hall coefficient in the low temperature region, we argue that when more Cu impurities were introduced into \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \), the removal of Fermi spectral weight in the hole-like Fermi surfaces is much stronger than that in the electron-like Fermi surfaces, which is equivalent to significant electron doping effect. DC magnetization and the lattice constants analysis reveal that static magnetic moments and notable lattice compression have been formed in Cu-doped samples. It seems that the superconductivity can be suppressed by the impurities disregard whether they are magnetic or nonmagnetic in nature. This gives strong support to a pairing gap with a sign reversal, like \( S^\pm \). However, the relatively slow suppression rates of \( T_c \) show the robustness of superconductivity of \( \text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2 \) against impurities, implying that multi-pairing channels may exist in the system.

PACS numbers: 74.20.Rp, 74.70.Dd, 74.62.Dh, 65.40.Ba

I. INTRODUCTION

The study of impurity effect is very important to the understanding of superconductivity. It happens quite often that the impurity induced suppression of superconductivity gives an early hint of the unconventional pairing state.\(^1\) Conventional s-wave superconductors are sensitive to magnetic impurities while robust to nonmagnetic impurities, which could be explained by Anderson’s theorem.\(^2\) But for cuprates with d-wave pairing symmetry, the superconductivity shows little tolerance to both magnetic and nonmagnetic impurities.\(^3\) Nonmagnetic impurities could induce a high density of states (DOS) due to the sign change of the gap on a Fermi surface and cause rapid suppression of superconducting transition temperature \( T_c \). The study of the impurity effect on superconductivity could give insights on the underlying pairing symmetry and superconducting mechanism. For example, rapid suppression of the transition temperature \( T_c \) in Al-doped \( \text{Sr}_2\text{Ru}_4\text{O}_8 \) was the first indication that it is a novel superconductor.\(^4\) The observations of unusual charge localization\(^4\) and enhanced antiferromagnetic (AFM) correlations around Zn impurities\(^5\) in \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) have spurred hot discussions of the relationship between local AFM correlations and superconductivity, which have important implications on the superconducting mechanism in cuprate.

After the discovery of superconductivity in iron pnictides and chalcogenides,\(^6\) a lot of investigations show unconventional superconducting mechanism and complicated gap structures in these new superconductors.\(^7\)\(^-\)\(^12\) Theoretically, an AFM fluctuation mediated fully-gapped sign-reversal \( S^\pm \) pairing state was proposed\(^13\)\(^-\)\(^14\) and received supports from the inelastic neutron scattering experiments\(^15\)\(^-\)\(^18\) and scanning tunneling spectroscopy (STS) measurements.\(^19\) However there were also other theoretical suggestions and experimental evidences for pairing states ranging from \( S^\rightarrow \)-wave to d-wave, or the existence of gap nodes.\(^20\)\(^-\)\(^23\)\(^-\)\(^25\) Theoretical calculations show that the simple version of \( S^\pm \) pairing state should be fragile to impurities,\(^24\)\(^-\)\(^27\) only 1% impurity with moderate scattering potential could induce large in-gap state and completely suppress superconductivity.\(^25\) Therefore plenty of experiments are carried out on the impurity induced suppression of superconductivity in Fe-based superconductors,\(^28\)\(^-\)\(^36\), in order to unravel the superconducting mechanism. Unfortunately the conclusions remain highly controversial. Further experimental and theoretical works are clearly desired to clarify the impurity effect in Fe-based superconductors, which could help us better understand the pairing symmetry and superconducting mechanism.

In this paper, we report the doping effect of Cu and Ni impurities on the superconductor \( \text{Ba}_0.6\text{K}_{0.4}\text{Fe}_2\text{As}_2 \). Both of these impurities could suppress superconductivity in certain rates and cause electron-doping effects. According to many other first-principles calculations, although the dopant Cu in Fe-based superconductors is in the valence state of +1 and with a fully occupied d orbit, the doping of Cu seems to introduce more electrons than dop-
ually suppressed and the residual resistivity (RR) rose previously an optimally hole-doped sample. As we can see Cu-doping in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ can induce the magnetic impurities, while the suppression to superconductivity is similar to the Ni-doping, which yields non- or weak magnetic impurity centers. Our results clearly indicate that the superconductivity can be suppressed by impurities at the Fe sites, disregard whether they are magnetic or nonmagnetic in nature.

II. EXPERIMENTAL RESULTS

The Cu-doped and Ni-doped polycrystalline samples Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$TM$_x$)$_2$As$_2$ (TM = Cu and Ni) were fabricated by solid state reaction method, the specific fabrication process was described in our previous paper. The x-ray diffraction (XRD) measurement was performed using an MXP18A-HF-type diffractometer with Cu-K$_\alpha$ radiation. The analysis of x-ray diffraction data was done by using the softwares POWDER-X and Fullprof, the obtained results are consistent with each other. The AC susceptibility measurements were carried out through an Oxford cryogenic system Maglab-EXA-12. The resistivity, magnetoresistance and Hall effect were measured with a Quantum Design instrument physical property measurement system (PPMS), and the DC magnetization by a Quantum Design instrument SQUID (MPMS-7).

The temperature dependence of resistivity of samples Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$TM$_x$)$_2$As$_2$ (TM = Cu,Ni) were shown in Fig.1(a) and Fig.1(b) respectively. The $T_c$ in impurity-free sample Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ is about 38 K, which is obviously an optimally hole-doped sample. As we can see the superconducting transition temperature was gradually suppressed and the residual resistivity (RR) rose upon the doping of either Cu or Ni impurities. The specific values of changing rates for $T_c$ and RR were calculated and presented later. We also notice that the values of normal state resistivity at above 150 K tend to increase with Cu doping while decrease with more Ni doping. This difference could be attributed to the different change of electronic band structures brought by these two different impurities, which would be also discussed in next section.

The x-ray diffraction patterns of Cu-doped samples were shown in Fig.2(a), the rather pure single phase indicate good quality of our samples. Fig.2(b) displays the evolution of lattice parameters for different doping samples. The values of Mn-doped ones were taken from our previous paper. Upon doping transition metal impurities onto the Fe sites, the lattice parameters for a-axis increase while that for c-axis decrease, which indicates that the crystal lattice is gradually compressed along the c-axis direction with impurity-doping. One can see that although the overall changing trend for the three different series of samples are similar, the Cu-doped samples show much stronger lattice compression compared to Ni and Mn doped ones.

![FIG. 1: (color online) (a) Temperature dependence of resistivity of the Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$Cu$_x$)$_2$As$_2$ samples under zero field. (b) Temperature dependence of resistivity of the Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ samples under zero field. With the doping of more Ni impurities, both the suppression rate of $T_c$ and the increasing rate of residual resistivity are relatively slow compared with that of Cu-doped samples.](image1)

![FIG. 2: (color online) (a)The x-ray diffraction patterns for Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$Cu$_x$)$_2$As$_2$, the samples are quite clean and no obvious phase segregation can be detected. (b) Evolution of a-axis and c-axis lattice constants with the doping of Cu, Ni and Mn impurities respectively. All the data are normalized by the values of undoped sample. The data points of Mn-doped samples are derived from our previous report.](image2)

In Fig.3(a) and Fig.3(b), we show the temperature dependence of Hall coefficients $R_H$ for Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$TM$_x$)$_2$As$_2$ (TM = Cu,Ni) respectively. Hall coefficients gradually change from positive values to negative values as either Cu or Ni doping. This indicates that both Cu and Ni introduced electrons into the hole-doped superconductor Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. We also noticed the changing rate of $R_H$ is higher for Cu-doped ones. For Cu-doped samples, the values of $R_H$ become completely negative for $x$=6% in the whole measured temperature region, while for Ni-doping samples, $R_H$ become completely negative at the doping level of $x$=8%. From the temperature dependence of the Hall coefficient $R_H$ for
the slightly doped samples, one can see a non-monotonic temperature dependence in the low temperature region. While it becomes more monotonic like when the electron-like charge carriers dominates. The doping effect of \( R_H \) cannot be easily understood within the rigid band model. In Fig.4(a) and 4(b) we present the doping dependence of the Hall coefficient \( R_H \) measured at 200 K. It is clear that the effective doping of electrons by adding Cu is stronger than that by adding Ni. A simple normalization of the Hall efficient suggest that the rigid model seems working at high temperatures, that is, doping electrons ratio Cu(electrons)/Ni (electrons)=3/2, this is very much like that of the Co, and Ni doping\(^{27}\). This interesting observation should be reconciled with the calculations of electronic structures.

III. ANALYSIS AND DISCUSSIONS

A. Magnetic moments introduced by doping Cu ions in Ba\(_{0.6}K_{0.4}Fe_2As_2\)

In order to investigate the impurity effect in Ba\(_{0.6}K_{0.4}Fe_2As_2\), we measured the magnetization of the doped samples in a magnetic field of 1 T. The data of Cu-doped samples were shown in Fig.5(a). The magnetization curve exhibits a typical T-linear behavior for undoped Ba\(_{0.6}K_{0.4}Fe_2As_2\) sample in the high temperature region. This linear behavior was interpreted as the origin of the short range AF correlation\(^{38}\). However with more Cu-doping, a Curie-Weiss like behavior emerges and gets more and more strong. For all Cu-doped samples, the data of magnetic susceptibility at the temperature below 150 K could be well fitted by the Curie-Weiss law. Based on the fitting results, we calculated the average magnetic moments of one Ba\(_{0.6}K_{0.4}(Fe_{1-x}Cu_x)_{2}As_2\) molecule, which is presented in Fig.5(b). It is clear that the doping of Cu gradually introduces magnetic moments into this system and the magnetic moment is getting stronger with more Cu doping. In the inset of Fig.5(b), the fitted curve of Cu-doped 8% sample was shown as an example of the fitting. Fe-based superconductors are in proximity to magnetism, so the disruption of the electronic structure by scattering would be expected to lead the formation of local moments around Cu sites\(^{39}\). Through the fitting results of the M-T curve with Curie-Weiss law:

\[
\chi = \chi_0 + \frac{C}{T + T_N}
\]  

where \( C = \mu_0\mu_J^2/3k_B \), we can get the magnetic moment \( \mu_J \) for each Ba\(_{0.6}K_{0.4}(Fe_{1-x}Cu_x)_{2}As_2\) molecule, which is shown in Fig.5 (b). The values of magnetic moments gradually increase with more Cu-doping and seem to get saturated at the doping level of 6%. Because of the \( d^{10} \) configuration of Cu, as claimed by the first-principles calculations, it is quite difficult to understand why a magnetic moment is induced at the Cu-sites. One possible picture would be that these magnetic moments may not exist at the Cu site, but could be distributed over the Fe ions around the Cu site, like the case in the cuprate superconductors near a Zn impurity.

B. Suppression to \( T_c \) by the impurities

From the x-ray data, we can see that transition metals such as Cu, Ni and Mn could be easily doped on the Fe sites in Fe-based superconductor Ba\(_{0.6}K_{0.4}Fe_2As_2\). The substitution of these impurities could all suppress superconductivity and raise the values of residual resistivity (RR), which means that these transition metals act as scattering centers. We should mention that, although the doping can induce the partial charge doping to the system, while the suppression to superconductivity here is mainly induced by the impurity scattering. This can be corroborated by the simple linear relation between
the residual resistivity and the normalized $T_c$ suppression as shown in Fig.1, Fig.6(b) and Fig.7. According to the Abrikosov-Gorkov formula, if the impurities act as strong pair breakers, the $T_c$ suppression due to pair breaking is essentially related to the impurity scattering rate $k_B \Delta T_c \approx \pi \hbar / 8 \tau_{imp} \propto \rho_0$, where $\rho_0$ is the residual resistivity. In Fig.6(a), the doping dependence of $T_c$-suppression rates for Cu, Ni and Mn were presented, $T_c$ decreases almost linearly with increasing the nominal impurity doping level. Through the calculation of the slopes, we can get the average change of $T_c$ values per doping percent: $\Delta T_c/\text{Mn-1}\% = -4.2$ K, $\Delta T_c/\text{Cu-1}\% = -3.5$ K, $\Delta T_c/\text{Ni-1}\% = -2.9$ K. While in Fig.6(b), we show the corresponding doping dependence of residual resistivity. Similarly we got $\Delta \rho_0/\text{Mn-1\%} = 0.107$ mΩ cm, $\Delta \rho_0/\text{Cu-1\%} = 0.093$ mΩ cm, $\Delta \rho_0/\text{Ni-1\%} = 0.071$ mΩ cm. From these data, it is clear that the impurity-doped samples which have higher rising rate of RR could cause a more rapid $T_c$ suppression. Thus Mn and Cu are relatively strong scattering centers, while Ni causes relatively weak impurity scattering effect.

From the lattice parameters point of view, as displayed in Fig.2(b), one can see that the changing tendency of lattice constants upon impurities is almost the same for all three series of samples, we also noticed the values of a-axis and c-axis in Cu-doped samples change faster than all the other impurity-doped samples. This means that the crystal lattices of Cu-doped samples are most strongly compressed and suffer stronger lattice change than the others.

It has been realized earlier that the main features of band structure and electronic density of states (DOS) remain the same after either cobalt or nickel was doped in the iron-site in iron-pnictides. A lot of researches also point out that, in the parent compound of Fe-based superconductors, the substitution of one Fe ion with one Co ion means the doping of one more electron, while doping one Ni ion can provide two electrons. A simple rigid band model could explain the experimental results. Since Cu is the element behind Ni in the periodical table and has one more $d$ electron than Ni, a straightforward thinking is that the doping of Cu means adding three more electrons. From the Hall data in Fig.3, people could easily have the first feeling that the values of $R_H$ in Cu-doped samples change to be negative more quickly than that in Ni-doped samples. This trend is further illustrated in Fig.4(b), where we plot the $R_H$ values at the temperature of 200 K for the two series of samples. One can also see that the evolution of $R_H$ with doping could be scaled together for Cu and Ni doped samples if we assume the equivalent doping of one Cu is adding three electrons while two electrons for Ni. The similar data and interpretation have also been reported by Canfield et al. However according to the results of many first principles calculations of the electronic density of states in Cu-doped iron pnictides and chalcogenides materials, unlike the case of Co-doping or Ni-doping, Cu exhibits split-band behavior with the $d$-electrons situated well below (3 to 4 eV) $E_F$, the $d$ bands of Cu are fully occupied for a nominal $d^{10}$ configuration. Therefore a state of Cu$^+$ would be expected, which indicates effective hole doping through the substitution of Fe$^{2+}$ by Cu$^+$. However from the shifts of the Fe density of states after Cu doping and the Hall data in this paper, an electron doping picture was suggested. This controversial situation could be reconciled by the analysis of the Fermi surfaces evolution. Chadov et al calculated the Fermi surfaces of Fe$_{1-x}$Cu$_x$Se, where they show that Cu-doping is highly disruptive to the electronic structure of FeSe superconductor and caused strong loss of Fermi spectral weight for the hole-like Fermi surfaces. Based on this picture, we could provide a possible explanation for the effective strong electron doping effect caused by Cu:

![Fig. 5: (color online) (a) DC magnetic susceptibility as a function of temperature of Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x$Cu$_x$})$_2$As$_2$ samples. A DC magnetic field of 1 T was applied in the measurement. (b) The average magnetic moments of one Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x$Cu$_x$})$_2$As$_2$ molecule were calculated according to the results of Curie-Weiss fitting. The inset shows the fitting result of M-T curve in low temperature region of sample Cu=8\% using the Curie-Weiss law.](image-url)
Cu was doped into the lattice, both the hole pockets at around Γ point and the electron pockets at around M point are gradually destroyed. This loss of Fermi spectral weight is especially rapid and stronger for the hole pockets, which could result in an equivalent electron doping effect for Cu-doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. However, the remaining controversy is that why in the high temperature region, the simple rigid model seems working very well, as displayed in Fig.4(b).

Considering the strong impurity scattering effect brought by Cu, the disorder induced localization is usually expected. Through the comparison of the normal state resistivity between Cu-doped and Ni-doped samples, we noticed that with more impurities, the normal state resistivity between Cu-doped and Ni-doped samples, we noticed that with more impurities, the normal state resistivity at high temperature region (150 K to 300 K, approximately) go higher with Cu-doping and have a trend of going down with Ni-doping. This phenomenon is consistent with the above discussions, namely the doping of Ni leads to the formation of a coherent electronic structure with the original Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, while the doping the Cu is destructive to the original electronic structure and causes localization effect. However, we should emphasize that, the suppression to the superconductivity is not induced by the localization of the electrons, but rather by the impurity scattering. This statement is specially valid below a moderate doping level. Actually in our data shown in Fig.1(a), we cannot see the up-turn of resistivity in the low temperature region, which should be the case if the localization would behave an important role here.

It has been widely discussed, in the only two high-$T_c$ superconductor families discovered by now, the cuprates and iron-pnictides have many similarities, for example, their parent compounds are both AFM ordered. However, they also have a lot of differences. For the impurity doping effect, we would say, these two families act quite differently. For Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, we choose a cuprate analog for comparison, namely La$_{1.85}$Sr$_{0.15}$CuO$_4$. These two superconductors are both hole doped and with almost the same $T_c$ of about 38 K. But for the latter, a d-wave cuprate superconductor, the superconductivity can be killed completely with a slight doping of impurities, for example, only about 3% of Zn doping can diminish the superconductivity, which yields a suppression rate of $\Delta T_c/1\%impurity = -11$ K. This is of course in sharp contrast with the impurity-doping in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$, which is usually below $\Delta T_c/1\%impurity = -4.2$ K. Especially for the case of Cu-doping in this paper, the electron-doping effect itself may result in a partial $T_c$ suppression, therefore the $T_c$-suppression caused by impurity scattering of Cu should be considered as moderate. The different responses to impurities for these two kinds of superconductors indicate that they could have different pairing and superconducting mechanism. One explanation would be that the pairing in the iron pnictide is not induced solely by the simple inter-pocket pair-scattering. The intra-pocket scattering may also contribute significantly to the pairing strength. We also noticed there were reports about more rapid suppression of superconductivity in Zn-doped LaFeAsO$_{0.85}$.$^{32}$ This means that the impurity effect in iron-pnictides is complicated, perhaps it is different for different compounds and doping region.$^{21}$ Theoretically both the S$^\pm$ and d-wave pairing are fragile to impurities, therefore Kontani et al. proposed an s-wave superconducting state without sign reversal (namely the $S_{++}$-wave state) for Fe-based superconductors based on d-orbital fluctuations considering the robustness superconductivity against impurities.$^{22}$ In Fig.7, we present the correlation between $T_c$ and residual resistivity (the scattering rate). One can see that the suppression to $T_c$ is more or less the same. Concerning the different magnetic moments induced by these dopants, our experiment indicates that the superconductivity can be suppressed by impurities disregard whether they are magnetic or nonmagnetic in nature. This phenomenon can certainly place more weight on the side of S$^\pm$ pairing. Further research is highly desired to address why the suppression rate to superconductivity by the impurities in the iron pnictide superconductors is moderate, not as strong as expected by a simplified model of the S$^\pm$ pairing.
FIG. 7: (color online) Correlations between the superconducting transition temperature and the residual resistivity in the Mn, Ni and Cu doped samples. The experiment for Mn doping was done in the system of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. That for Ni and Cu doping was done in the system of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.

IV. CONCLUDING REMARKS

In summary, the impurity doping effects of Cu and Ni on the structure, transport properties, magnetism and superconductivity of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ have been studied. The substitution of these impurities on the Fe sites could all suppress $T_c$ and raise the values of residual resistivity in certain rates. The measurements of Hall coefficients show more electron-like charge carriers have been introduced into the system for Cu-doped samples than Ni-doped ones. This significant electron doping effect in Cu-doped samples could be explained possibly by the relatively stronger loss of Fermi spectral weight in the hole-like Fermi surfaces than the electron-like ones in Ba$_{0.6}$K$_{0.4}$(Fe$_{1-x}$Cu$_x$)$_2$As$_2$. Although the theoretical calculations suggest that the $d$ bands of Cu are fully occupied for a nominal $d^{10}$ configuration, magnetic moments with moderate strength have been found in Cu-doped samples. The common behavior among the samples by doping Mn, Ni and Cu strong suggest that the superconductivity can be suppressed by impurities disregard whether they are magnetic or nonmagnetic in nature. This gives of course more weight to the S$^\pm$ pairing. However, the suppression rate to $T_c$ is moderate, not as strong as expected by the simple model of S$^\pm$ pairing. We attribute this to the multiple pairing channels, such as the inter-pocket or intra-pocket pairing in the samples.

Acknowledgments

We appreciate the useful discussions with D. J. Singh, I. I. Mazin, P. Hirschfeld, D.-H. Lee, Q. H. Wang and G.-M. Zhang. This work is supported by the NSFC of China, the Ministry of Science and Technology of China (973 Projects: 2011CBA001002, 2012CB821403, 2010CB923002). The work at RUC is supported by NSFC (No.11204373) and the Fundamental Research Funds for the central Universities, and the Research Funds of Renmin University of China.

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