Effect of doping on the anisotropic resistivity in single-crystal $Bi_2Sr_2CaCu_2O_{8+\delta}$ with Fe, Co, and Ni

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Anisotropic transport properties have been investigated for single-crystal $Bi_2Sr_2CaCu_2O_{8+\delta}$ doped with Fe, Co, and Ni. The doping has dramatic effects on both the in-plane resistivity ρ_{ab} and out-of-plane resistivity ρ_c. Upon doping with Fe, Co, and Ni the in-plane resistivity ρ_{ab} increases, accompanied by a decrease in T_c. The out-of-plane resistivity increases in the case of doping with Co and Ni, while monotonously decreasing with increasing Fe concentration. The anisotropy $\rho_c(T)/\rho_{ab}(T)$ decreases with doping. The difference in the effect of doping on transport properties between Fe, Co, and Ni arises from the fact that the Co and Ni preferentially occupy the Cu site, whereas the trivalent atom of Fe possibly resides in the Bi site. The data of ρ_c are well fitted by the activation-type phenomenological formula $\rho_c(T)=(a/T)\exp(\Delta/T)+bT+c$.
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A distinctive property for the high-T_c layered cuprates is their extremely anisotropic resistivity at temperatures above the superconducting transition temperature T_c. In the underdoped regime, the *c*-axis resistivity has ''nonmetallic'' temperature dependence while the in-plane resistivity is metallic in behavior.[1–3] The resistivity anisotropy ρ_c/ρ_{ab} and *c*-axis resistivity ρ_c decrease systematically with increasing carrier concentration.[1,4] In the overdoped regime, the *c*-axis resistivity $\rho_c(T)$ also shows a metallic behavior,[5,6] especially the *c*-axis resistivity of highly oxygenated $YBa_2Cu_3O_{6+x}$ ($x>0.93$) decreases linearly with temperature down to the superconducting transition temperature.[5,7] The resistivity anisotropy in over-doped $La_{2-x}Sr_xCuO_4$ ($x\sim0.34$) is temperature independent, as expected of an anisotropic three dimensional metal.[4,6] This is strong evidence that the transition into the ''overdoped'' regime involves a crossover from two-dimensional superconducting to three-dimensional metallic behavior. In $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), however, the resistivity anisotropy ρ_c/ρ_{ab} is as high as 10^5,[3,8] which is significantly larger than those of other layered cuprates; the insulating trend in $\rho_c(T)$ is observable at temperature as high as 200 K, even in the overdoped regime.[9,10]

The impurity effect on the anisotropic transport properties is a sensitive probe for investigating the interlayer charge dynamics.[11,12] Many research studies on doping the Cu-O planes with Fe, Co, Ni, and Zn in the Bi2212 system have been reported,[13–24] however, in which less work was made for the *c*-axis resistivity and anisotropic transport properties. A sharp contrary result was observed for the samples doped with Co and Zn.[18,19] The doping with Zn led to a reduction of out-of-plane resistivity and anisotropy by about one order of magnitude. The semiconductorlike behavior of the out-of-plane resistivity was strongly suppressed with Zn doping. In contrast, there is an increase in out-of-plane resistivity with Co doping. In addition, a temperature linear dependence of the *c*-axis resistivity has been observed in the overdoped sample of Pb-doped Bi2212, and its magnitude is reduced by four orders, and is nearly the same magnitude as that of in-plane resistivity.[25] It is anomalous that the anisotropy is too small, even much less than that of highly oxygenated $YBa_2Cu_3O_{6+x}$ and overdoped $La_{1.66}Sr_{0.34}CuO_4$. Many models for the out-of-plane conduction have been proposed; however, no consensus has been achieved so far.[26]

To clarify further the *c*-axis conduction and the effect of doping Cu-O planes on the out-of-plane resistivity and anisotropy, we have measured in detail the in-plane and the out-of-plane resistivities for single crystals of Bi2212 doped with Fe, Co, and Ni using the Montgomery method. It is found that the in-plane resistivity for all crystals is linear with temperature, and the residual resistivity increases with doping concentration. The out-of-plane resistivity shows a dramatic change upon doping. In the case of doping with Fe, the *c*-axis resistivity monotonously decreases with Fe doping, and the anisotropic ratio $\rho_c(T)/\rho_{ab}(T)$ apparently decreases, while the out-of-plane resistivity increases upon doping with Co and Ni, and the anisotropic ratio $\rho_c(T)/\rho_{ab}(T)$ decreases compared with the pure Bi2212 crystal. The out-of-plane resistivity can be well fitted by a thermally activated formula.

Single crystals of pure Bi2212 and Bi2212 doped with Fe, Co, and Ni were grown by self-flux using CuO as flux. The single crystal of Bi2212 doped with Zn was also grown, but an intergrowth of Bi2212 and Bi2223 phases was observed, and the resistivity measurement always shows two superconducting transitions at about 90 and 120 K, respectively. Therefore, we will not present the experimental results of single crystal Bi2212 doped with Zn. The composition of Bi2212 crystals was analyzed by electron probe microanalysis (EPMA). The analysis shows the doping concentration up to about 2 at. % of Ni and Co for Cu. The concentration of Fe in the crystals can be much higher, but there exists a critical concentration of about 5 at. % to keep the Bi2212 structure. With increasing Fe concentration, the structure is changed from the Bi2212 to the Bi1212 phase. A similar result has been reported by vom Hedt *et al.*[27] For higher concentrations of Fe, the structure of the single crystal was identified as the

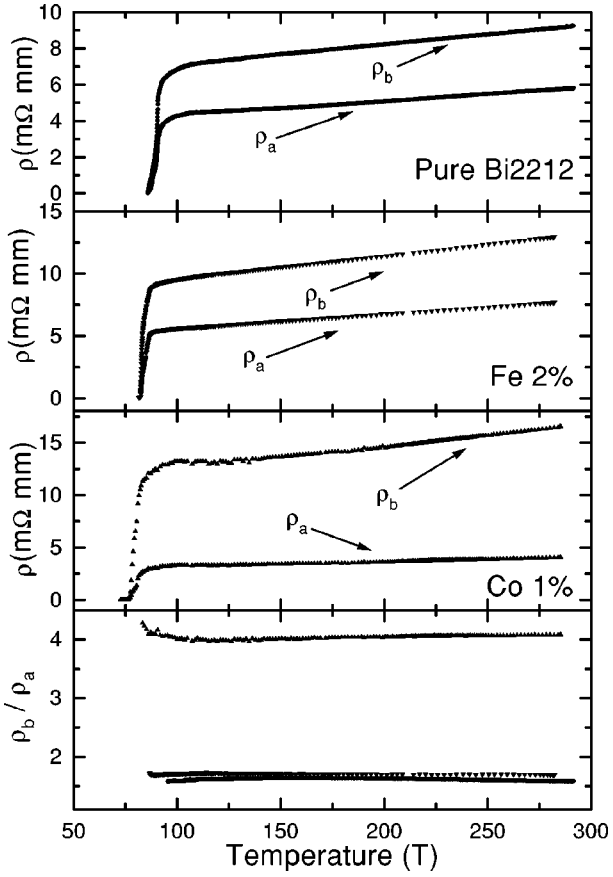

FIG. 1. The temperature dependence of anisotropic in-plane resistivity for the as-grown single crystal (circles) and the single crystals doped with Fe (down triangles) and Co (up triangles), and their resistivity ratio $\rho_b(T)/\rho_a(T)$.

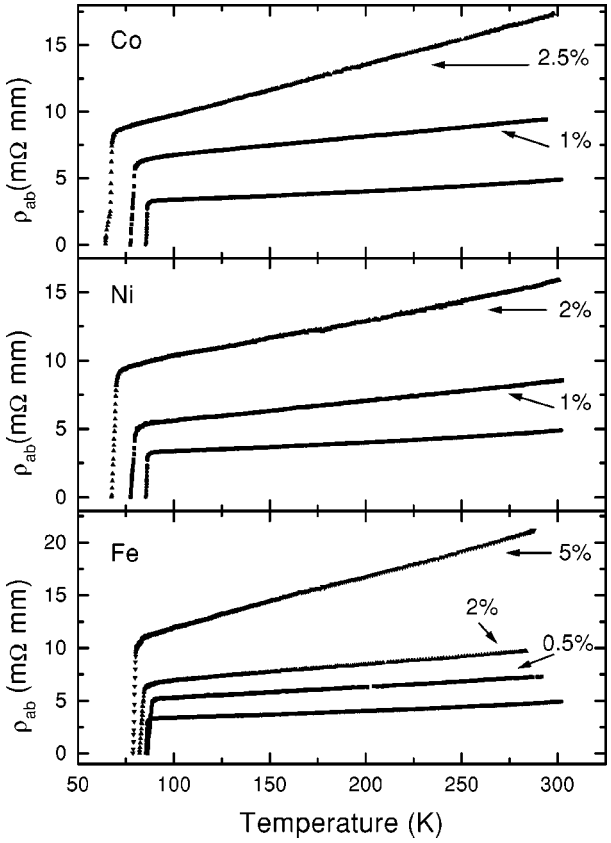

FIG. 2. The temperature dependence of in-plane resistivity for the pure $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal and the crystals doped with Fe, Co, and Ni.

1212 phase, which is similar to that of $FeSr_2YCu_2O_7$ (1212) (Ref. 28) or $(Bi,M)Sr_2YCu_2O_7$ (1212) (M=Cu, Fe, etc).[29] It suggests that the trivalent atom of Fe preferentially occupies the Bi site in Bi-based systems. This is consistent with the case of Fe doping in $YBa_2Cu_4O_8$ (Y2212),[30] in which Fe substitutes preferentially for [Cu(1)] sites because the double Cu(1)-O chains in $YBa_2Cu_4O_8$ (Cu2212) are equivalent to the double Bi-O layers from the viewpoint of crystallography.

We obtained two crystals for Co and Ni (1 at. % Co and 2.5 at. % Co; 1 at. % Ni and 2 at. % Ni) and three crystals for Fe (0.5 at. %, 2 at. %, and 5 at. %) with different doping concentrations, respectively. The crystals used to measure resistivity had a typical size of 1.5 mm×1 mm×0.05 mm with the shortest dimension along the c axis. The thickness of the crystal was determined by a scanning electron microscope. Electrical contacts of less than 2 Ω resistance were established by soldering the copper leads onto the sample surface on which pure silver was evaporated. The Montgomery contact configuration is the same as that reported in our previous paper.[31] The anisotropic in-plane resistivity (ρ_a and ρ_b) and the out-of-plane resistivity ρ_c were separately measured in the different crystals. In order to directly obtain the anisotropic ratio $\rho_c(T)/\rho_{ab}(T)$, the transport along the a axis and b axis is considered to be isotropic when the out-of-plane resistivity ρ_c was measured.

Figure 1 shows the temperature dependence of the anisotropic in-plane resistivity for the as-grown single crystals. It is found that the normal state resistivity behavior is nearly the same, and is linear with temperature for the undoped and doped crystals with Fe and Co. The in-plane anisotropy $\rho_b(T)/\rho_a(T)$ is also shown in Fig. 1. The anisotropy $\rho_b(T)/\rho_a(T)$ is temperature independent for all crystals. It suggests that a common scattering mechanism for a-axis and b-axis transport. It is worth pointing out that the resistivity ratio $\rho_b(T)/\rho_a(T)$ for the crystal doped with Fe is nearly the same as that for the pure crystal, while there is an apparent increase in $\rho_b(T)/\rho_a(T)$ for the crystal doped with Co. For the pure $Bi_2Sr_2CaCu_2O_{8+\delta}$, the in-plane anisotropic ratio was reported to be in the range 2–4,[32–34] depending on the carrier concentration. The anisotropic in-plane resistivity is believed to be due to incommensurate superlattice structure along the b axis.[33,34] The corrugated CuO_2 planes along the b axis would give a larger effective mass of the carrier, leading to a larger resistivity along this direction. The modulation period q_b is decreased by Co doping,[35] resulting in more corrugated CuO_2 planes. Such a structural change would be an origin of the larger in-plane ansotropy $\rho_b(T)/\rho_a(T)$ observed in Co-doped Bi2212 crystals.

Figure 2 shows the temperature dependence of in-plane resistivity $\rho_{ab}(T)$ for the pure Bi2212 single crystal and the crystal doped with Fe, Co, and Ni. All samples show the well-known linear resistivity $\rho_{ab}(T)=\rho_0+aT$.[36] We fit the data of $\rho_{ab}(T)$ to the formula for all samples. The results are listed in Table I. For the pure Bi2212 crystal, the temperature dependence is nearly linear, but contains a small quadratic

TABLE I. The fitting parameters of in-plane resistivity of normal state with $\rho_{ab}(T)=\rho_0+bT$ for the undoped Bi2212 crystal and the doped Bi2212 crystals with Fe, Co, and Ni.

Samples	ρ_0 (mΩ mm)	b (mΩ mm/K)
Pure B2212	2.598 ± 0.005	$0.0071\pm2.5\times10^{-5}$
1% Co	5.347 ± 0.005	$0.0139\pm2.8\times10^{-5}$
2.5% Co	5.807 ± 0.005	$0.0380\pm2.5\times10^{-5}$
1% Ni	4.162 ± 0.003	$0.0144\pm1.2\times10^{-5}$
2% Ni	7.512 ± 0.009	$0.0272\pm4.4\times10^{-5}$
0.5% Fe	4.237 ± 0.003	$0.0104\pm1.6\times10^{-5}$
2% Fe	5.347 ± 0.007	$0.0154\pm4.5\times10^{-5}$
5% Fe	7.015 ± 0.012	$0.0488\pm7.3\times10^{-5}$

component, suggesting the crystal is in the slightly overdoped regime. The residual resistivity was estimated from the intercept of ρ_0. Table I shows that the residual resistivity monotonously increases with increasing the doping concentration for all crystals with a different dopant. The monotonous increase in residual resistivity indicates a decreased mean free path λ caused by scattering from the dopant impurities. It is found in Table I that the doping with Fe, Co, and Ni leads to a slightly monotonous increase with doping concentration in the slope of $d\rho_{ab}/dT$ from about 0.007 mΩ mm/K for the pure crystal to 0.038 mΩ mm/K for the crystal doped with 2.5 at. % Co, to 0.027 mΩ mm/K for the crystal doped with 2 at. % Ni, and to 0.049 mΩ mm/K for the crystal doped with 5 at. % Fe, respectively. It implies that the doping results in a decrease in carrier concentration. The superconducting transition temperature T_c decreases monotonically as the amount of Ni and Co in the crystals is increased until the saturation of the doping concentration. We find the magnitude of dT_c/dx caused by Ni substitution in B2212 is nearly the same as that by Co substitution. In present results, $dT_c/dx=-900$ K for Ni and Co substitution, which is nearly the same as the result reported by Kuo er al.[20] The change of T_c caused by Fe substitution is different from that by Co and Ni substitution. $dT_c/dx=-100$ K for Fe substitution is much less than -900 K for Ni and Co substitution. Especially, the T_c has a slight increase at the low concentration of Fe. It could be because the low concentration doping of Fe results in a decrease in carrier concentration to make the crystals from the overdoped regime to the optimally doped regime. The difference of the effect of doping on T_c between Fe, Co, and Ni is also indicative that Fe preferentially occupies the Bi sites, being different from the case of Co and Ni. This is consistent with the structural analysis mentioned above. Therefore, it is easily understood that Fe substitution has a weak destruction on T_c compared to the case of Co and Ni substitution.

The temperature dependence of out-of-plane resistivity $\rho_c(T)$ is shown in Fig. 3 for the $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal and the single crystals doped with Fe, Co, and Ni. For all crystals, the resistivity temperature dependence $d\rho_c(T)/dT$ is negative in the measuring temperature range between the superconducting transition temperature (T_c) and 300 K. It implies that the crystals are in the optimally doped or slightly overdoped regime since a positive $d\rho_c(T)/dT$ at high temperature has been observed in the heavily overdoped

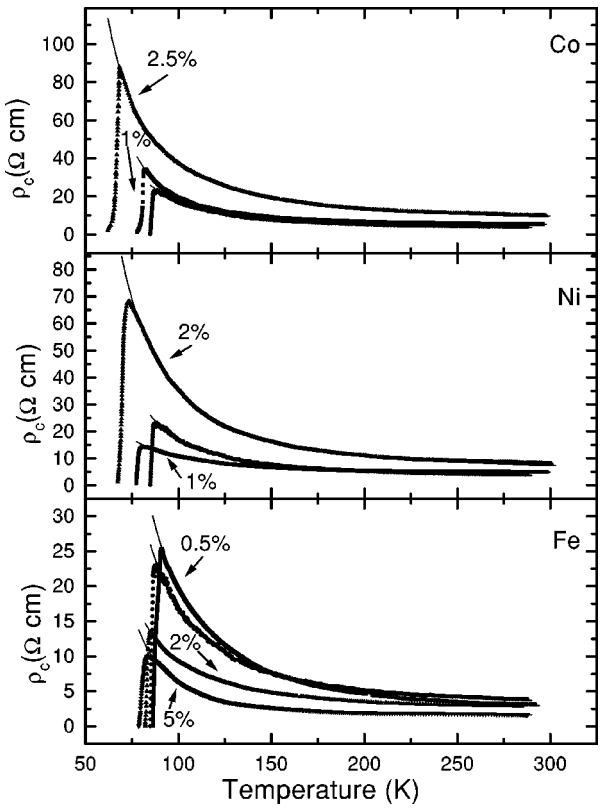

FIG. 3. The temperature dependence of out-of-plane resistivity for the pure $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal and the crystals doped with Fe, Co, and Ni. The solid curves are fits of $\rho_c(T)$ data to Eq.(1).

regime.[10,31] In the case of Co doping, the $\rho_c(T)$ increases as the amount of Co is increased, which is consistent with the result reported by Watanabe and Matsuda.[18] But the crystal doped with 1 at. % Co shows nearly the same behavior of $\rho(T)$ as that for the pure Bi2212 crystal except for a reduction of T_c. The effect of doping with Ni on the out-of-plane resistivity is similar to that for the doping with Co. It is worth noting that the semiconductorlike behavior of the out-of-plane resistivity was suppressed with doping 1 at. % Ni compared to pure Bi2212 crystals, but the temperature gradient of the resistivity was still slightly negative. A similar behavior has been reported for the Bi2212 single crystal doped with 1 at. % Zn, in which the reduction of the out-of-plane resistivity is more than one sixth.[19] Jeon *et al.* explained their results by the elongation of the out-of-plane coherence length ξ_c based on the analysis of the superconductivity fluctuation. In the case of Fe doping, the out-of-plane resistivity monotonically decreases as the amount of Fe in the crystals is increased. This is completely different from the case of the doping with Co and Ni. It could be understood that Fe substitution for the Bi sites of the weakly coupled BiO-BiO double layers leads to an increase in the interlayer coupling strength between the adjacent blocks of the CuO_2 planes, subsequently resulting in an elongation of the out-of-plane coherence length ξ_c.

There have been many explanations for the temperature dependence of *c*-axis resistivity $\rho_c(T)$.[26] Kumar and Jayannavar[37] proposed a model in which the *c*-axis confine-

ment and the semiconducting *c*-axis resistivity in the layered cuprates was attributed to anisotropic weak localization. However, this model gives the same temperature dependence for the *c*-axis resistivity as that for the in-plane resistivity, in contrast to our experimental results. On the other hand, Anderson[38] argues that the observation of localization behavior in only one direction is not possible. The famous ''confinement'' theory proposed by Anderson[38,39] assumes that *c*-axis confinement and the ''semiconducting'' *c*-axis resistivity in the layered cuprates derive from properties of the spin-charge-separated Luttinger liquid normal state. This theory gives a semiconducting T dependence of *c*-axis resistivity with $\rho_c(T) \sim 1/T$, and can qualitatively explain the nonmetallic $\rho_c(T)$ behavior. Unfortunately, it cannot be quantitatively well fit to our data. Levin and co-workers[40] proposed an interlayer scattering model. According to this model, the *c*-axis conductivity is contributed by three hopping processes: direct interlayer quasiparticle hopping, hopping assisted through static disorder (impurities), and hopping assisted through inelastic boson-mediated scattering (phonon). According to the analysis of Ref. 41, in the frame of the interlayer scattering model the *c*-axis resistivity can be given by $\rho_c(T) = a_0 + aT + 1/(c + bT)$ for the extremely anisotropic Bi-based cuprates. Although our data can be well fitted to the above formula, the unphysical fitting parameters of a_0 and c, being negative for all crystals, are obtained. The ''semiconducting'' ρ_c in underdoped $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+\delta}$ has also been attributed to a gap in the spin density of states.[6] However, recently it is reported that the onset of the semiconducting $\rho_c(T)$ does not coincide with the opening of the spin gap seen in the $\rho_{ab}(T)$ in the Bi2212 system.[10] In the present experimental results, no opening of spin gap is observed in the $\rho_{ab}(T)$, while the $\rho_c(T)$ shows a semiconducting behavior. It suggests that the semiconducting behavior of $\rho_c(T)$ could not be explained by the spin gap. The bipolaron theory proposed by Alexander and co-workers[42] can be fitted to the present data as reported by us.[31] However, we find that the fitting by the activation-type phenomenological formula

$$\rho_c(T) = (a/T)\exp(\Delta/T) + bT + c \qquad (1)$$

is much better than the fitting by the bipolaron theory. In the formula, a, Δ, b and c are constants, and T is independent. The fitting curves are also shown in Fig. 3 as solid lines, and the parameters obtained are listed in Table II. The doping affects the activated term strongly. The fitting parameter b to the data of all crystals was set to zero except for the crystal doped with 1 at.% Ni, since the fitting gave it a negative value which was unphysical. However, the fitting becomes surprisingly better by the setting $b = 0$. But the parameter b is positive for the crystal doped with 1 at.% Ni, and the fitting is much better than that of setting b zero. It has been reported[10,31] that the out-of-plane resistivity of the underdoped and optimally doped samples can be well reproduced with the above activation-type phenomenological formula and the parameter b was set to zero, while the out-of-plane resistivity of the overdoped samples cannot be fitted by setting $b = 0$. Therefore, the nonzero parameter b of the crystal

TABLE II. The fitting parameters a, Δ, b, and c of the formula $\rho_c(T) = (a/T)\exp(\Delta/T) + bT + c$ to the data of the out-of-plane resistivity for the pure and doped Bi2212 crystals with Fe, Co, and Ni.

Sample	a (Ω cm/K)	Δ (K)	b (Ω cm/K)	c (Ω cm)
Pure B2212	270±4.1	171±1.3	0	2.01±0.03
1% Co	181±1.7	214.3±0.8	0	4.07±0.02
2.5% Co	887±22	135±1.8	0	5.1±0.2
1% Ni	366±10.8	85±1.9	0.0063±0.0003	1.4±0.11
2% Ni	865±14.8	131±1.3	0	2.8±0.1
0.5% Fe	391±5.9	160±1.3	0	0.26±0.05
2% Fe	156±1.9	157±1.0	0	1.84±0.02
5% Fe	53±2.2	227±3.4	0	1.10±0.03

doped with 1 at.% Ni suggests that the crystal is in the overdoped regime, in which a weakly semiconducting behavior of out-of-plane resistivity is observed.[10,31] This is consistent with our experimental results. According to the arguments of Anderson,[43] doping the CuO_2 planes with spin impurities will rapidly destroy the unique Luttinger liquid properties by smearing the spinon Fermi surface, reintroducing residual resistance and metallic transport along the *c* axis as well as reducing T_c. Although the spin impurity Ni doping with low concentration leads to an apparent suppression in semiconducting behavior of out-of-plane resistivity, no metallic out-of-plane resistivity is observed. Further research on the arguments of Anderson is being carried out.

Figure 4 shows the temperature dependence of the anisot-

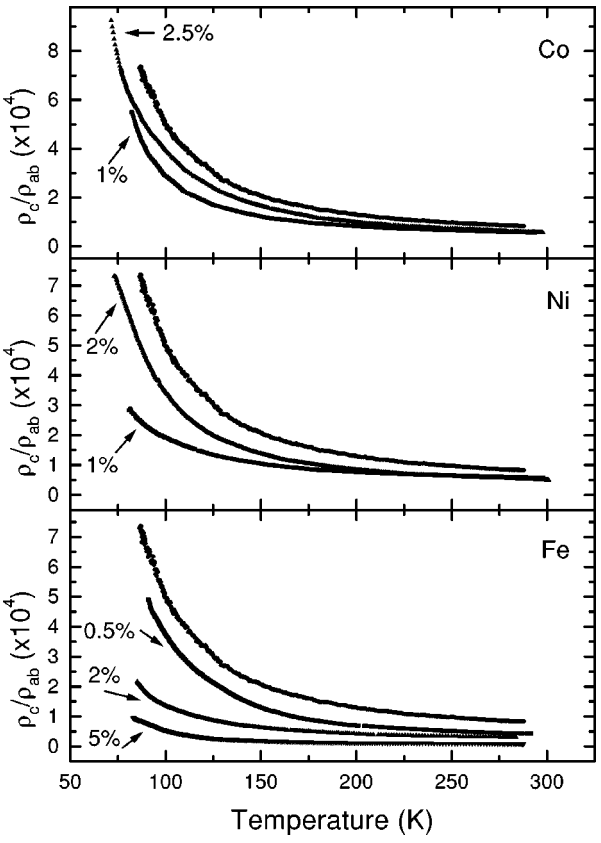

FIG. 4. The temperature dependence of the anisotropy $\rho_c(T)/\rho_{ab}(T)$ for the same crystals as in Figs. 2 and 3.

ropy ratio $\rho_c(T)/\rho_{ab}(T)$ for the same crystals as in Figs. 2 and 3. The anisotropy $\rho_c(T)/\rho_{ab}(T)$ is rather larger; all crystals exceed 10^4 at low temperature. $\rho_c(T)/\rho_{ab}(T)$ depends on temperature for all the crystals. This implies that the mechanisms governing the transport property along and perpendicular to the CuO_2 plane are different. Upon doping the CuO_2 planes with Co and Ni, the anisotropy ratio decreases relative to the pure Bi2212 crystal. The behavior of the anisotropy ratio with doping is the same for Co and Ni doping. This is consistent with the previous reports.[18,19] This behavior has been explained[19] in that doping the CuO_2 planes leads to an elongation of the out-of-plane coherence length ξ_c, while the in-plane coherence length ξ_{ab} was almost unchanged. In the case of Fe doping, the anisotropy ratio $\rho_c(T)/\rho_{ab}(T)$ monotonously decreases as the amount of Fe is increased. The anisotropy ratio $\rho_c(T)/\rho_{ab}(T)$ for the crystal doped with 5 at.% Fe is about one-eighth of that for the pure Bi2212 crystal at low temperature. It further indicates that Fe substitution for Bi greatly strengthens the weak interlayer coupling between the double BiO layers. On the other hand, it supports the observation that the much larger anisotropy ratio $\rho_c(T)/\rho_{ab}(T)$ in Bi-based cuprates than in La-based and Y-based cuprates arises from the very weak bonds between double BiO layers.

In conclusion, single crystals of Bi2212 doped with different concentrations of Fe, Co, and Ni were grown by the self-flux method and their anisotropic transport properties (ρ_{ab} and ρ_c) were examined. With Co and Ni doping, a decrease in the anisotropy ratio $\rho_c(T)/\rho_{ab}$ as well as an increase in out-of-plane resistivity were observed, while the anisotropy ratio $\rho_c(T)/\rho_{ab}(T)$ and the out-of-plane resistivity monotonously decrease as the amount of Fe is increased. The data of $\rho_c(T)$ can be well fitted by the activated-type phenomenological formula $\rho_c(T)=(a/T)\exp(\Delta/T)+bT+c$. An apparent suppression in semiconducting behavior of out-of-plane resistivity was observed for the crystal doped with low concentrations of Ni.

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