High-temperature resistivity in the iron pnictides and the electron-doped cuprates

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We measured the high-temperature (up to 800 K) resistivity of several dopings of $SrFe_{2-x}(Ni,Co)_xAs_2$ (Sr-122) and compared the results with similar measurements on electron-doped cuprates. We find that the Sr-122 pnictide resistivity saturates above 500 K at around 400–700 $\mu\Omega$ cm, consistent with the Mott-Ioffe-Regel (MIR) limit and in contrast with the MIR-violating behavior of the hole-doped cuprates and our measurements on electron-doped cuprates. This supports the view that electronic correlations in the ferropnictides may be weaker than in the cuprates.

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I. INTRODUCTION

The new iron pnictide family of superconductors is often compared with the cuprate superconductors. Both families have similar phase diagrams, with magnetic ordering in the parent compounds suppressed by doping. Further doping yields a superconducting dome with quite high transition temperatures. However, the cuprate superconductors are doped Mott-Hubbard insulators with strong Coulomb interactions, whereas the pnictide parent compounds are metallic, suggesting that the pnictides might be more weakly correlated than the cuprates. Supporting this picture, resistivity saturation has been reported in PrFeAsO crystals as evidence for electron-phonon scattering.[1] However, magnetic susceptibility in $BaFe_{2-x}Co_xAs_2$ has been observed to show an unusual linear temperature dependence up to 700 K, described as indicating strong magnetic fluctuations.[2] Magnetic fluctuations have also been observed in neutron scattering[3,4] and nuclear magnetic resonance (NMR).[5,6]

Transport measurements at high temperatures could be affected by electronic correlations. In typical (weakly correlated) metals, high-temperature resistivity depends on charge carriers scattering off phonons. In these materials, $\rho(T)$ is described by the Bloch-Grüneisen theory and is predicted to be linear above the Debye temperature up to high temperatures.[7] This linear $\rho(T)$ cannot continue indefinitely in real crystals, as pointed out by Ioffe and Regel[8] and Mott,[9] due to the finite size of the crystals' interatomic spacing. As the temperature increases, charge carriers scatter more frequently; this shrinks the mean free path ℓ and increases $\rho(T)$. This picture breaks down, however, when ℓ becomes smaller than the lattice parameter (a). This is the Mott-Ioffe-Regel (MIR) limit and implies that the resistivity of a metal should saturate[10] at $\rho_{\mathrm{MIR}} = 3\pi^2\hbar/e^2k_F^2l$. Typically, metals have a MIR limit around 100–1000 $\mu\Omega$ cm; this behavior has been observed in many ordinary metals.[11]

The Ioffe-Regel criterion $\ell < a$ affects resistivity dominated by charge carriers scattering off of bosonic excitations. Generally this is electron-phonon scattering; however, bosonic magnetic scatterers such as electron-magnon scattering in γ-$Fe_{80-x}Ni_xCo_{20}$ can also show Ioffe-Regel resistive saturation.[12] Any electron-boson scattering produces a MIR saturation; however, scattering from other processes that do not depend on boson excitations does not MIR-limit if the scatterers do not depend on the lattice spacing, such as electron-electron interactions in highly correlated systems.[11] For example, the strong Coulomb interactions in $La_{2-x}Sr_xCuO_4$ (LSCO) allow the t-J model to predict resistive saturation well above the MIR limit due to scattering caused by electronic interactions.[10] Experimentally, MIR-limit violation is known to occur in the high-T_c cuprates such as LSCO (Refs. 13 and 14) and $YBa_2Cu_3O_7$ (YBCO).[15,16]

The high-temperature ferropnictide superconductors are believed to be correlated systems, although more weakly correlated than the cuprates. Because the MIR-limit-violating behavior is typical in strongly correlated systems, high-temperature resistivity measurements are important for understanding charge-carrier dynamics in the ferropnictides. In this Brief Report we present our observation of resistivity saturation around the MIR limit in the $SrFe_2As_2$ (Sr-122) system and contrast this with a MIR-limit violation that we find in the electron-doped cuprates. These results qualitatively support the picture of modest correlation strength in the ferropnictides.

II. EXPERIMENTAL DETAILS

We report high-temperature resistivity measurements on the electron-doped cuprates $Nd_{2-x}Ce_xCuO_{4-\delta}$ (NCCO) and $Pr_{2-x}Ce_xCuO_{4-\delta}$ (PCCO), as well as on Ni- and Co-doped $SrFe_2As_2$. NCCO single crystals were prepared by self-flux with a typical size of $0.5 \times 0.5 \times 0.03$ mm^3 as described elsewhere.[17] Crystals were then reduced in a low-oxygen anneal to achieve superconductivity. PCCO thin films were grown by pulsed laser deposition (PLD) using a LambdaPhysik KrF excimer laser on 5×10 mm^2 $SrTiO_3$ substrates. The films were grown to a nominal thickness of 300 nm and were vacuum annealed to maximize superconductivity. Single crystals of $SrFe_{2-x}(Ni,Co)_xAs_2$ were prepared by self-flux, as described in Ref. 18, with a typical dimension of $2 \times 0.5 \times 0.1$ mm^3.

Low-temperature measurements were performed before and after the high-temperature measurements to be certain that the crystals and films of both types of materials were not damaged by thermal cycling up to 800 K. Neither cuprates nor pnictides decompose at these high temperatures if maintained in an inert environment. We found that any damage due to thermal cycling was minimal and did not affect our measurements; the resistivity did not change when sweeping up and down in temperature. Sweeping down in temperature immediately after reaching the maximum minimized the time spent at high temperatures and did not significantly overanneal

the PCCO films. The samples were contacted to gold wires by electrically conductive silver paint that maintained its bonding at high temperatures and were heated by a Neocera PLD substrate heater. The sample and heater were enclosed in a vacuum chamber with the atmosphere maintained between 8 and 10 Torr of flowing argon to minimize the possibility of oxidation. The crystal geometry allowed a four-probe resistivity measurement; a 100 Hz 1 mA current was applied and measured by a Princeton Applied Research 5210 lock-in amplifier. The phase was stable to less than a degree. Measurements below 300 K were performed in a Quantum Design physical property measurement system (PPMS).

III. RESULTS

As shown in Fig. 1, the *ab* plane resistivity of optimally doped PCCO and NCCO violates the MIR limit with T^2 behavior up to 700 K and linear behavior at higher temperatures.[19] This trend is consistent with previous work on NCCO, which showed the resistivity increasing up to 600 K.[20] The MIR limit in these materials should be around 700 $\mu\Omega$ cm, similar to that in LSCO.[10] Optimally doped NCCO can be fitted reasonably well to $\rho = \rho_0 + AT^2$ between T_c and 700 K; above 700 K the resistivity is linear in T with a slope of $\sim 5\ \mu\Omega$ cm/K.

The *ab* plane resistivity of some Sr-122 Fe pnictides up to 800 K is shown in Fig. 2. The $SrFe_2As_2$ parent compound and $x(\mathrm{Ni}) = 0.14$, $x(\mathrm{Ni}) = 0.18$, and $x(\mathrm{Co}) = 0.26$ dopings are shown. The resistivity increases smoothly above the spin-density wave (SDW) transition or T_c and saturates at high temperatures. Above 650 K the resistivity maintains a constant value between 400 and 700 $\mu\Omega$ cm depending on the doping.

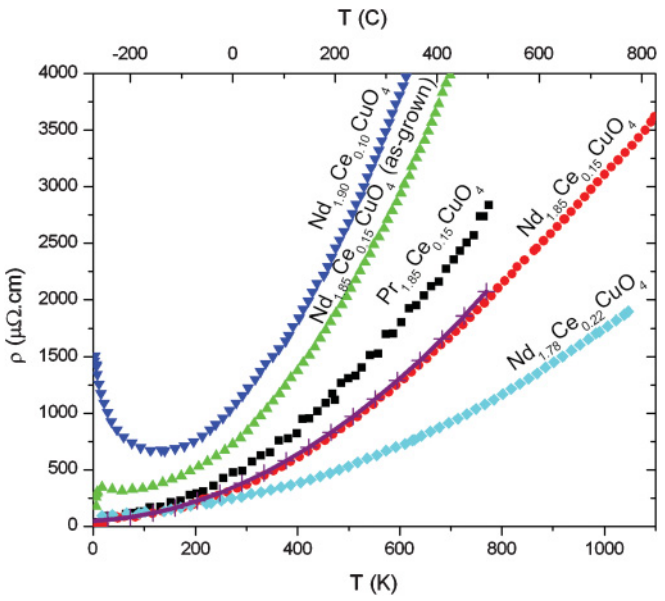

FIG. 1. (Color online) The *ab* plane resistivity versus temperature of optimally doped ($x = 0.15$) and annealed $Pr_{2-x}Ce_xCuO_4$ films (■) and $Nd_{2-x}Ce_xCuO_4$ crystals: optimally doped ($x = 0.15$) and annealed (●), optimally doped ($x = 0.15$) as-grown (unannealed) (▲), underdoped $x = 0.10$ (▼), and overdoped $x = 0.22$ (◆). Optimal crystals are fitted to T^2 above T_c and below 700 K (+). ($Nd_{2-x}Ce_xCuO_4$ from Ref. 21.) The MIR limit in these materials is around 700 $\mu\Omega$ cm.

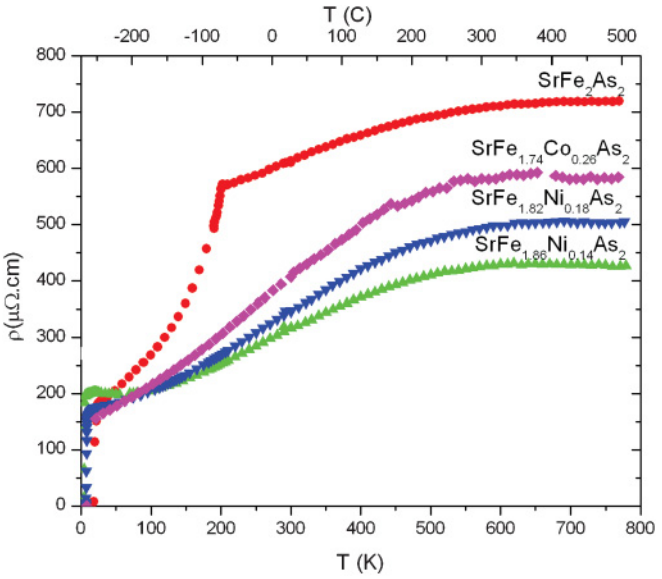

FIG. 2. (Color online) The *ab* plane resistivity $\rho(T)$ up to 800 K for $SrFe_2As_2$ (●), $SrFe_{1.86}Ni_{0.14}As_2$ (▲), $SrFe_{1.82}Ni_{0.18}As_2$ (▼), and $SrFe_{1.74}Co_{0.26}As_2$ (◆) single crystals. The resistivity saturates around 400–700 $\mu\Omega$ cm; the MIR limit is $\sim$600 $\mu\Omega$ cm.

IV. DISCUSSION

In the electron-doped cuprates, the MIR limit is strongly violated with $\rho \propto T^2$ up to 700 K. A $\rho \propto T^2$ above T_c and up to $\sim$250 K has been known for many years but its exact origin is not known.[22] For typical Fermi-liquid metals, $\rho \propto T^2$ usually comes from electron-electron scattering, but this is not expected above $\sim$20 K. At higher temperatures electron-phonon scattering, or electron scattering off other bosons, usually dominates the resistivity. At high temperatures ($T > \Theta_D$) T-linear behavior from electron-phonon scattering usually dominates. This picture, however, assumes a lack of strong correlations and would predict MIR-limit saturation. The lack of resistivity saturation rules out dominant phonon scattering for the electron-doped cuprates.

In many strongly correlated metallic systems, the MIR limit has been shown to be violated, as in $La_{1-x}Sr_xMnO_3$,[11,14] Li_2VO_4,[11,23] $Sr_{n+1}Ru_nO_{3n+1}$,[24] and the hole-doped cuprate high-temperature superconductors. For example, neither YBCO nor LSCO shows resistivity saturation up to 1100 K and LSCO still shows resistivity increasing linearly at those temperatures, well beyond ρ_{MIR}.[25] Additionally, in many cuprates the Ioffe-Regel criterion is violated below room temperature due to the high resistivity. The Ioffe-Regel condition assumes noninteracting electrons, so the electron-doped cuprates' lack of $\rho(T)$ saturation suggests strongly correlated behavior consistent with hole-doped high-T_c cuprates.

In the cuprates, the t-J model has been used to explain the normal-state behavior in terms of strongly correlated electrons in doped antiferromagnets.[26] A MIR-limit violation consistent with this picture has been reported in LSCO at similar temperatures.[10] Derived from the Hubbard model, the t-J model describes a system of strongly correlated electrons with antiferromagnetic interactions associated with a Mott-Hubbard insulator state at zero doping.[26] This model has been used to analyze LSCO where the MIR limit is

violated but resistivity still saturates, although at a higher value $\rho_{\rm sat} \approx \frac{0.07c}{x(1-x)}$ mΩ cm, where c is the inter-CuO_2-plane spacing and x is the doping.[10,27] Unfortunately, for PCCO and NCCO the saturation value in this model is around 3500 $\mu\Omega$ cm, and 1100 K is not hot enough to test this model. It would be instructive to look for saturating behavior up to 1300 K; however, it is doubtful that this could be done without complications due to oxygen loss and the crystal melting. In summary, strong correlations seem to dominate the PCCO and NCCO *ab* plane charge transport because of the MIR limit violation.

In contrast with the cuprates, the Sr-122 pnictides do show a $\rho(T)$ saturation at high T. This superficially resembles the picture expected for electron-phonon scattering in typical metals, suggesting weak correlations in the ferropnictides. It has been suggested that the Fermi-liquid picture can explain the pnictide transport behavior;[28] however, there have been observations of non-Fermi-liquid[29] and quantum critical[30] behavior. Several other pnictide models also suggest $\rho(T)$ saturation.

A MIR limit often suggests a typical band metal picture. In that picture, the electron-phonon coupling constant should be a meaningful quantity. The roughly linear $\rho(T)$ from ~200 to ~400 K has a slope of 0.5–0.9 $\mu\Omega$ cm/K, about a tenth that of NCCO, and suggests correspondingly weaker electron-phonon scattering. From transport data, this value can be calculated[1] by $\lambda_{\rm tr} = \frac{\hbar\omega_p^2}{8\pi^2 k_B}\frac{d\rho}{dT}$. For $SrFe_2As_2$, the plasma frequency has been measured[31] to be $\omega_p = 1.4 \times 10^4$ cm^{-1}, giving a $\lambda_{\rm tr} \sim 0.12$. This is too small to reproduce the T_c of the Sr-122's. This mismatch between the electron-phonon coupling constant λ and the expected T_c is similar to calculations done on several 1111 pnictides where $\lambda \sim 0.17$–0.21 predicts a T_c of a few kelvin at most.[32] A $\lambda_{\rm tr} \sim 0.12$ is also much smaller than the $\lambda_{\rm tr} = 1.53$ reported for PrFeAsO.[1]

The MIR limit is a stronger indication of charge-carrier coupling to phonon excitations. For a three-dimensional (3D) system with a spherical Fermi surface (FS), the MIR limit is given[10] by $\rho = \frac{3\pi^2\hbar}{e^2 k_F^2 \ell}$. For a 2D system, more appropriate for the pnictides, a cylindrical Fermi surface gives a MIR limit[10] of $\rho = \frac{2\pi\hbar c}{e^2 k_F \ell}$ with c, the interplanar spacing, taken to be half of 12.32 Å, $SrFe_2As_2$'s c-axis lattice parameter.[18] Then $\ell \approx a$ gives a MIR limit of $\rho_{\rm MIR}^{\rm 2D} = 634$ $\mu\Omega$ cm. A 3D spherical FS would have a much higher $\rho_{\rm MIR}^{\rm 3D} = 2560$ $\mu\Omega$ cm, with $k_F = (\frac{3\pi^2 N}{V})^{1/3}$. For the MIR limit in the 122's with a quasi-2D FS, $\rho_{\rm MIR}^{\rm 2D}$ is a reasonable but slightly low estimate. The measured ρ saturation is 400–700 $\mu\Omega$ cm, near $\rho_{\rm MIR}^{\rm 2D} = 634$ $\mu\Omega$ cm. Electron-phonon scattering therefore could explain the value of $\rho_{\rm sat}$ in the 122's and would suggest that electron correlations do not play an important role in the scattering for transport.

Recently, however, it has been proposed by Prelovšek and Sega that strong spin fluctuations couple the electron and hole bands in the system and produce a saturating $\rho(T)$.[33] In this model, the magnitude of ρ is primarily tuned by the spin-fermion coupling constant g_0. A $g_0 \approx 1$ corresponds to a 1000–1500 $\mu\Omega$ cm ρ saturation; we measured 400–700 $\mu\Omega$ cm. The measured ρ saturation is a factor of ~2–4 less than predicted, implying a larger spin-fermion coupling, $g_0 \gg 1$, and the possible breakdown of the model.[33] This model, however, is only a two-band model where the Sr-122 system is known to be more complicated; therefore, some discrepancy in magnitude cannot rule out a relatively strong spin-fermion coupling in the 122's. As the pnictides are known to show spin fluctuations from neutron scattering[3,4] and NMR,[5,6] coupling to spin fluctuations could produce MIR-like $\rho(T)$ saturation in the pnictides.

Similar models predict $\rho(T)$ saturation due to two bands with unequal scattering.[34] If the two bands have different transport parameters, the more conductive band can "shunt" the less conductive band, saturating the high-temperature resistance. The more conductive band, therefore, would begin to dominate transport properties and the material would appear less resistive. Although there could still be a MIR limit in this model, it would not be the primary cause of resistivity saturation.

In systems similar to the Sr-122's, much higher resistivity values have been reported in high-temperature measurements on the 1111 oxypnictides. Large resistivities have been observed in the NdFeAsO (Ref. 35) and LaFePO (Ref. 36) systems and a resistivity saturation at 2500–3000 $\mu\Omega$ cm has been reported in PrFeAsO crystals as evidence of a MIR limit.[1] This saturation is significantly higher than the 100–1000 $\mu\Omega$ cm seen in most MIR-limited materials. Low carrier concentrations can produce a large MIR limit[10] or a small spin-fermion coupling constant could produce a resistivity saturation at high values.[33]

The pnictide $\rho(T)$ saturation could also be due to the arsenides being semimetals. With multiple bands near the Fermi energy, thermally activated behavior would then increase the charge-carrier density. Sales *et al.* have proposed this behavior in the Sr-122's with an energy scale of $E_h = 300$ K.[37] Conduction in the Sr-122's might then increase at high T. However, Hall- effect measurements have argued for transport dominated by a single charge carrier,[38] reducing multiband effects. At elevated temperatures, semimetal resistivity can indeed show saturation and even decrease, as in the case of semimetal graphite[39] and graphene.[40] However, an increase without limit is seen in semimetal CaB_6 (Ref. 41) and Bi.[42] Although pnictide semimetal behavior could describe the high-temperature $\rho(T)$ saturation seen, it is difficult to draw conclusions regarding semimetal behavior in the Sr-122's from high-temperature resistivity.

MIR-limited behavior in a weakly coupled system, resistive saturation due to relatively strong spin-fermion coupling, or thermally activated charge carriers could explain the high-temperature resistive saturation in the Sr-122's. Saturating resistivity in the Sr-122's does rule out strong electronic correlation effects such as those in the cuprates. In the Sr-122's, however, the resistivity saturation cannot by itself provide a conclusive picture of pnictide charge dynamics.

Finally, the similarity in the resistivity saturation between undoped $SrFe_2As_2$ and Co- and Ni-doped samples reveals a lack of doping dependence of the saturation mechanism. If spin fluctuations are responsible, as in the model of Prelovšek and Sega or more generally as the bosonic scatterer, it may be expected that the suppression of AFM order with doping should result in a modification of this scattering and therefore

a change in the resistivity saturation. Similarly, semimetal behavior is often quite sensitive to doping. However, the lack of change in the saturation temperature of $\rho(T)$ suggests that either the spin-fluctuation spectrum or thermally activated behavior does not change at high temperatures with doping, or that phonon-scattering is indeed dominant at high temperatures in the iron pnictides.

V. CONCLUSIONS

We measured the *ab* plane resistivity of several Sr-122 superconductors and electron-doped cuprates. The electron-doped cuprates strongly violate the Mott-Ioffe-Regel limit whereas the 122's saturate at 400–700 $\mu\Omega$ cm at $T > 650$ K. Strong electronic correlations can explain the cuprate behavior; however, pnictide saturation can be explained either by electron-phonon scattering, by coupling between spin fluctuations and charge carriers, or by thermally activated charge carriers. The observed high-temperature resistivity saturation suggests more weakly correlated physics in the ferropnictides than in the cuprates.

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