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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 $\text{SrFe}_{2-x}(\text{Ni},\text{Co})_x\text{As}_2$ (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 $400 - 700 \mu\Omega\cdot\text{cm}$, consistent with the Mott-Ioffe-Regel (MIR) limit and in contrast to 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.

PACS numbers:

Introduction

The new iron-pnictide family of superconductors is often compared to 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 scattering¹. However, magnetic susceptibility in $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ has been observed to show unusual linear temperature dependence up to 700 K, described as indicating strong magnetic fluctuations². Magnetic fluctuations have also been observed in neutron scattering^{3,4} and 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 of 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 temperatures⁷. This linear $\rho(T)$ cannot continue indefinitely in real crystals as pointed out by Ioffe and Regel⁸ and Mott⁹ due to the finite size of the crystals' interatomic spacing. As 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 saturate¹⁰ at $\rho_{\text{MIR}} = \frac{3\pi^2\hbar}{e^2k_F\ell}$. Typically, metals have a MIR limit around $100 - 1000 \mu\Omega\cdot\text{cm}$; this behavior has been observed in many ordinary metals¹¹.

It is worth noting that 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, like electron-magnon scattering in $\gamma\text{-Fe}_{80-x}\text{Ni}_x\text{Co}_{20}$, can also show Ioffe-Regel resistive saturation¹². Any electron-boson scattering will produce a MIR saturation; however, scattering from other processes that do not depend on boson excitations

will not MIR limit if the scatterers do not depend on the lattice spacing, such as electron-electron interactions in highly correlated systems¹¹. For example, the strong Coulomb interactions in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) allow the t-J model to predict resistive saturation well above the MIR limit due to scattering caused by electronic interactions¹⁰. Experimentally, MIR limit violation is known to occur in the high- T_c cuprates like LSCO^{13,14} and $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO)^{15,16}.

The high-temperature ferropnictide superconductors are believed to be correlated systems, although more weakly correlated than the cuprates. Because 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 paper we report 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.

Experimental Details

We report high temperature resistivity measurements on the electron-doped cuprates, $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ (NCCO) and $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ (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 $0.5 \text{ mm} \times 0.5 \text{ mm} \times 0.03 \text{ mm}$ as described elsewhere¹⁸. Crystals were then reduced in a low oxygen anneal to achieve superconductivity. PCCO thin films were grown by pulsed laser deposition (PLD) using a Lambda Physik KrF excimer laser on $5 \text{ mm} \times 10 \text{ mm}$ STO substrates. The films were grown to a nominal thickness of 300 nm and were vacuum annealed to maximize superconductivity. Single crystals of $\text{SrFe}_{2-x}(\text{Ni},\text{Co})_x\text{As}_2$ were prepared by self flux, as described in ref. 19, with a typical dimension of $2 \text{ mm} \times 0.5 \text{ mm} \times 0.1 \text{ mm}$.

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 measure-

ments; 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 over-anneal 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 4-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 PPMS.

Results

As shown in Figure 1, the ab-plane resistivity of optimally doped PCCO and NCCO violate the MIR limit with T^2 behavior up to 700 K and linear behavior at higher temperatures²⁰. This trend is consistent with previous work on NCCO which showed resistivity increasing up to 600 K²¹. The MIR limit in these materials should be around 700 $\mu\Omega\cdot\text{cm}$, similar to LSCO¹⁰. Optimally-doped NCCO can be fit reasonably well to $\rho = \rho_0 + AT^2$ between T_c and 700 K; above 700 K resistivity is linear in T with a slope of $\sim 5 \mu\Omega\cdot\text{cm}/\text{K}$.

FIG. 1: (color online) The ab-plane resistivity versus temperature of optimally doped ($x=0.15$) and annealed $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ films (■) and $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ 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 fit to T^2 above T_c and below 700 K (+). [$\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ from Ref. 35] The MIR limit in these materials is around 700 $\mu\Omega\cdot\text{cm}$.

The ab-plane resistivity of some 122 Fe-pnictides up to 800 K is shown in Figure 2. The SrFe_2As_2 parent compound and $x(\text{Ni}) = 0.14$, $x(\text{Ni}) = 0.18$, and $x(\text{Co}) = 0.26$ dopings

are shown. The resistivity increases smoothly above the SDW transition or T_c and saturates at high temperatures. Above 650 K the resistivity maintains a constant value of between 400 and 700 $\mu\Omega\cdot\text{cm}$ depending on doping.

FIG. 2: (color online) The ab-plane resistivity, $\rho(T)$, up to 800 K for SrFe_2As_2 (●), $\text{SrFe}_{1.86}\text{Ni}_{0.14}\text{As}_2$ (▲), $\text{SrFe}_{1.82}\text{Ni}_{0.18}\text{As}_2$ (▼), and $\text{SrFe}_{1.74}\text{Co}_{0.26}\text{As}_2$ (◆) single crystals. The resistivity saturates around 400 – 700 $\mu\Omega\cdot\text{cm}$; the MIR limit is around 600 $\mu\Omega\cdot\text{cm}$.

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 ~ 250 K has been known for many years but its exact origin is not known²². For typical Fermi liquid metals, $\rho \propto T^2$ usually comes from electron-electron scattering, but this is not expected above ~ 20 K. At higher temperatures electron-phonon scattering, or electron scattering off of 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, $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ^{11,14}, Li_2VO_4 ^{11,23}, $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$ ²⁴, and the hole-doped cuprate high temperature superconductors. For example, neither YBCO nor LSCO show 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 elec-

trons in doped antiferromagnets²⁶. MIR limit violation consistent with this picture has been reported in LSCO at similar temperatures¹⁰. 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 doping²⁶. This model has been used to analyze LSCO where the MIR limit is violated but resistivity still saturates, although at a higher value, $\rho_{sat} \approx \frac{0.07c}{x(1-x)} \text{ m}\Omega\cdot\text{cm}$ where c is the inter-CuO₂-plane spacing and x is the doping^{10,27}. Unfortunately, for PCCO and NCCO, the saturation value in this model is around 3500 $\mu\Omega\cdot\text{cm}$ 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 to the cuprates, the Sr-122 pnictides do show $\rho(T)$ 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 behavior²⁸; however, there have been observations of non-Fermi-liquid²⁹ and quantum critical³⁰ behavior. Additionally, 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 K to ~ 400 K has a slope of $0.5 - 0.9 \mu\Omega\cdot\text{cm}/\text{K}$, about a tenth that of NCCO, and suggests correspondingly weaker electron-phonon scattering. From transport data, this value can be calculated¹ by $\lambda_{tr} = \frac{\hbar\omega_p^2}{8\pi^2k_B} \frac{d\rho}{dT}$. For SrFe₂As₂, the plasma frequency has been measured³¹ to be $\omega_p = 1.4 \times 10^4 \text{ cm}^{-1}$, giving a $\lambda_{tr} \sim 0.12$. This is too small to reproduce the T_c of the 122s. This mismatch between the electron-phonon coupling constant λ and the expected T_c is similar to calculations done on several 1111 pnictides where $\lambda \sim 0.17 - 0.21$ predict a T_c of a few Kelvin at most³². A $\lambda_{tr} \sim 0.12$ is also much smaller than the $\lambda_{tr} = 1.53$ reported for PrFeAsO¹.

The MIR limit is a stronger indication of charge carrier coupling to phonon excitations. For a 3D system with a spherical Fermi surface (FS), the MIR limit is given¹⁰ by $\rho = \frac{3\pi^2\hbar}{e^2k_F^2\ell}$. For a 2D system, more appropriate for the pnictides, a cylindrical Fermi surface gives a MIR limit¹⁰ of $\rho = \frac{2\pi\hbar c}{e^2k_F\ell}$ with c , the inter-planar spacing, taken to be half of 12.32 Å, SrFe₂As₂'s c-axis lattice parameter¹⁹. Then $\ell \approx a$ gives a MIR limit of $\rho_{MIR}^{2D} = 634 \mu\Omega\cdot\text{cm}$. A 3D spherical FS would have a much higher $\rho_{MIR}^{3D} = 2560 \mu\Omega\cdot\text{cm}$, with $k_F = \left(\frac{3\pi^2N}{V}\right)^{1/3}$. For the MIR limit in the 122s with a quasi-2D FS, ρ_{MIR}^{2D} is a reasonable, but slightly low, estimate. The measured ρ saturation is 400 – 700 $\mu\Omega\cdot\text{cm}$, near $\rho_{MIR}^{2D} = 634 \mu\Omega\cdot\text{cm}$. Electron-phonon scattering, therefore, could explain the value of ρ_{sat} in the 122s 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 $\rho(T)$ ¹⁷. 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 $\mu\Omega\cdot\text{cm}$ ρ saturation; we measured 400 – 700 $\mu\Omega\cdot\text{cm}$. The measured ρ saturation is a factor of $\sim 2 - 4$ less than predicted, implying a larger spin-fermion coupling, $g_0 \gg 1$, and the possible breakdown of the model¹⁷. This model, however, is only a two-band model where the 122 system is known to be more complicated; therefore, some discrepancy in magnitude cannot rule out relatively strong spin-fermion coupling in the 122s. 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 scattering⁴². 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. While there could still be a MIR limit in this model, it would not be the primary cause of resistivity saturation.

In similar systems to the 122s, much higher resistivity values have been reported in high temperature measurements on the 1111 oxypnictides. Large resistivities have been observed in the NdFeAsO³³ and LaFePO³⁴ systems and a resistivity saturation at 2500 – 3000 $\mu\Omega\cdot\text{cm}$ has been reported in PrFeAsO crystals as evidence for a MIR limit¹. This saturation is significantly higher than the 100 – 1000 $\mu\Omega\cdot\text{cm}$ seen in most MIR limited materials. Low carrier concentrations can produce a large MIR limit¹⁰ or a small spin-fermion coupling constant could produce a resistivity saturation at high values¹⁷.

The pnictide $\rho(T)$ saturation could also 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 122s with an energy scale of $E_h = 300 \text{ K}$ ³⁶. Conduction in the 122s might then increase at high T . However, Hall effect measurements have argued for transport dominated by a single charge carrier³⁷, reducing multi-band effects. At elevated temperatures, semimetal resistivity can, indeed, show saturation and even decrease, as in the case of semimetal graphite³⁸ and graphene⁴⁰. However, increase without limit is seen in semimetal CaB₆³⁹ and Bi⁴¹. Although pnictide semimetal behavior could describe the high temperature $\rho(T)$ saturation seen, it is difficult to draw conclusions regarding semimetal behavior in the 122s 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-122s. Saturating resistivity in the Sr-122s does rule out strong electronic correlation effects like those in the cuprates. In the Sr-122s, however, the resistivity saturation cannot by itself provide a conclusive picture of pnictide charge dynamics.

Finally, the similarity in 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.

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 while the 122s saturate at $400 - 700 \mu\Omega\cdot\text{cm}$ at $T > 650 \text{ K}$. 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 in the Sr-122s suggests more weakly correlated physics in the ferropnictides than in the cuprates.

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