

Doping dependence of Hall coefficient and evolution of coherent electronic state in the normal state of the Fe-based superconductor $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

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We investigated the in-plane transport properties of the Fe-based superconductor $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with a wide composition range $0 \leq x \leq 0.55$. We show that the doping dependence of the Hall coefficient is well described by the Boltzmann equation for a two-band system with a rigid-band approximation. We successfully deduced transport parameters, which suggested that holes with heavier mass conduct more smoothly than electrons. Moreover, the temperature variation of the Hall coefficient indicated that an anomalous coherent state characterized by heavy quasiparticles in hole bands evolved below ~ 100 K, predominantly in the optimal and overdoped regions. We argue that this phenomenon can be understood in relation to the pseudopeak structure observed in angle-resolved photoemission spectroscopy.

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I. INTRODUCTION

In correlated electronic systems, we frequently observe a crossover phenomenon which does not accompany the long-range order associated with symmetry breaking. A famous example is the pseudogap behavior in a normal state of cuprate superconductors, where various quantities show a gap-like feature below a characteristic temperature.¹ Despite the significant effort that has been made to date, there still remains debate concerning the microscopic mechanism.

The pseudogap phenomena are also observed in a canonical system of Fe-based superconductors $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$,² an electronic phase diagram of which is shown in Fig. 1. An angle-resolved photoemission spectroscopy (ARPES) measurement for $x = 0.25$ revealed a spectral weight transfer to a deeper energy level of ~ 18 meV in the innermost hole Fermi surface below ~ 120 K, indicating an opening of the pseudogap.³ On the other hand, a laser ARPES for $x = 0.41$ clarified the evolution of a pseudopeak structure centered ~ 12 meV below the Fermi energy in the three-hole Fermi surfaces below ~ 100 K.⁴ In this course, the density of states at the Fermi energy gradually increased upon cooling, which is in stark contrast to the pseudogap phenomena. In order to unravel the underlying microscopic mechanism of the pseudogap and pseudopeak features, we need to collect detailed information on electronic states over a wide x range.

A sensitive probe of a crossover phenomenon is the Hall coefficient (R_H), which can detect not only single-particle information but also the electron correlation effect. Indeed, the temperature (T) evolution of R_H was one of the earliest pieces of evidence of a pseudogap in cuprate superconductors.⁵ In Fe-based superconductors, however, the multiband nature of Fermi surfaces, which consist of three hole bands and two electron bands (see inset of Fig. 1),⁶ prevents us from providing a straightforward interpretation of R_H . Moreover, most previous studies focused on R_H in alloyed compounds such as $\text{Ba}(\text{Fe}_{1-y}\text{Co}_y)_2\text{As}_2$,⁷⁻¹² where a disorder effect as well as the band reconstruction were not negligible. Hence, a systematic study of R_H for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ has been highly anticipated. Recently, such a study was actually performed; however, it still focused on the underdoped regime.¹³

In this work, we investigated normal state of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ over a wide composition range $0 \leq x \leq 0.55$ by measuring in-plane transport properties with a special focus on R_H . The analysis based on the Boltzmann equation indicated that the quasiparticle mass ratio of holes to electrons increased with decreasing T , which likely causes a decrease in R_H below ~ 100 K for $0.45 \leq x \leq 0.55$. We claim that this behavior is closely related to the evolution of the pseudopeak in the ARPES spectra and argue that its mechanism can be understood in terms of a coherent quasiparticle formation due to the coupling with a boson in the hole bands.

II. EXPERIMENT

Single crystals of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ were grown by the self-flux method as reported in the literature.¹⁴ Since the composition of K has a tendency to be spatially inhomogeneous even within a single crystal, we carefully characterized each crystal. We mounted a crystal oriented along the c axis on an x-ray diffractometer and checked the $0\ 0\ l$ reflection width, which is a measure of x variation according to the linear relationship $c = 13.01 + 0.821x$ Å (c being the lattice parameter). The variation of x in a crystal used for transport measurements was less than ± 0.01 . The in-plane resistivity (ρ) was measured by the standard four-probe method. The Hall resistivity in the plane was measured by using a rotator under a magnetic field of 7 tesla at a fixed value of temperature.

III. RESULTS AND ANALYSIS

A. Resistivity

Figure 2 represents the T variation of ρ scaled by the 350 K value, which somewhat depends on x (inset of Fig. 2). The quantity ρ at $x = 0$ was weakly T dependent in the paramagnetic state and showed an abrupt decrease below 137 K, which corresponded to the antiferromagnetic transition temperature (T_N). Upon K substitution, the anomaly appeared at a lower T in a subtle manner and completely disappeared at $x = 0.26$. In the wide composition range $0.14 \leq x \leq 0.55$, the zero resistivity was observed at low T . The superconducting

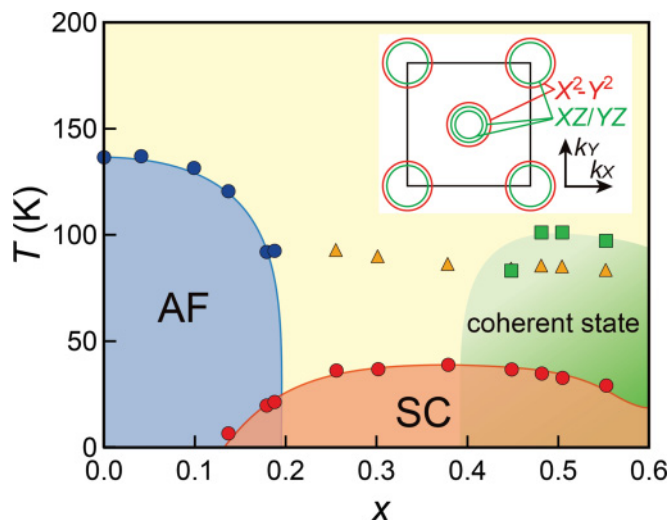


FIG. 1. (Color online) Electronic phase diagram of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ in the K content (x) and temperature (T) plane, where AF and SC stand for antiferromagnetic and superconducting phases, respectively. Triangles and squares indicate the crossover temperature obtained by the resistivity and Hall coefficient, respectively. The inset is a schematic view of Fermi surfaces. The orbital character of each band is also shown.⁶

transition temperature (T_c) was estimated by the midpoint of the ρ drop; a typical T_c width was 0.6 K. We then obtained an electronic phase diagram (Fig. 1) which was in perfect agreement with the phase diagram for polycrystalline samples.¹⁵ The quantity ρ for $0.26 \leq x \leq 0.55$ exhibited a crossover from a high- T upward-concave to a low- T downward-concave behavior; the characteristic T (80–95 K) are plotted as open triangles in Fig. 1.

The power-law behavior of ρ is a key ingredient in relation to the possible quantum critical phenomena near

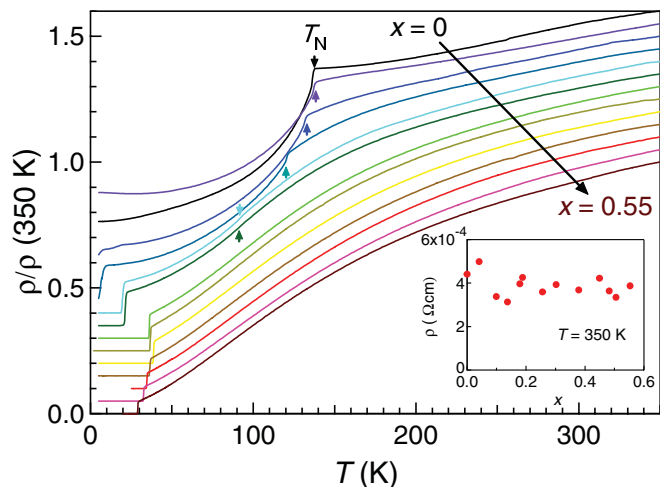


FIG. 2. (Color online) Temperature (T) dependence of resistivity (ρ) scaled by the 350 K value for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$. The composition is $x = 0, 0.04, 0.10, 0.14, 0.18, 0.19, 0.26, 0.30, 0.38, 0.45, 0.48, 0.51,$ and 0.55 from the top to the bottom. Note that each curve is shifted by 0.05 for clarity. The arrows indicate the antiferromagnetic transition temperature (T_N). The inset shows the x dependence of ρ at 350 K.

an antiferromagnetic-paramagnetic phase boundary. We fit the data for $0.26 \leq x \leq 0.55$, where the antiferromagnetic state disappears down to the lowest T , with the function $\rho = \rho_0 + AT^n$. When we take a fitting range as 40–80 K, this fitting yields $n = 1.62$ for $x = 0.26$ and $n = 1.47$ for $x = 0.55$. The deviation from the Fermi liquid value $n = 2$ seemingly suggests an influence of critical antiferromagnetic fluctuations; however, we need special care. The ρ_0 value becomes negative at $x \geq 0.38$, indicating that the actual n is much larger than n deduced by the fitting. One cannot make conclusions about the quantum critical behavior in the present system until the normal-state ρ is acquired down to lower T by eliminating the superconducting phase with a high magnetic field. We also point out the possibility that earlier studies reporting $n < 2$ in Fe-based superconductors overlooked the large- T variation of ρ at low- T ranges owing to a large residual resistivity.

B. Hall coefficient

R_H displayed in Fig. 3 exhibits a significant variation as a function of x and T . At $x = 0$, a negative R_H gradually increased in magnitude upon cooling, showed a discontinuous jump at T_N , and reached a constant value at 5 K. When holes are doped, R_H changes its sign. In a wide T range, R_H still showed a monotonic increase in magnitude upon cooling; however, such a tendency became more moderate with increasing x . For $0.45 \leq x \leq 0.55$, we found a crossover phenomena from the high- T -increasing to the low- T -decreasing behavior in R_H at $T^* \sim 100$ K, which is indicated by triangles in Fig. 3. The crossover temperature T^* is plotted as open squares in Fig. 1,

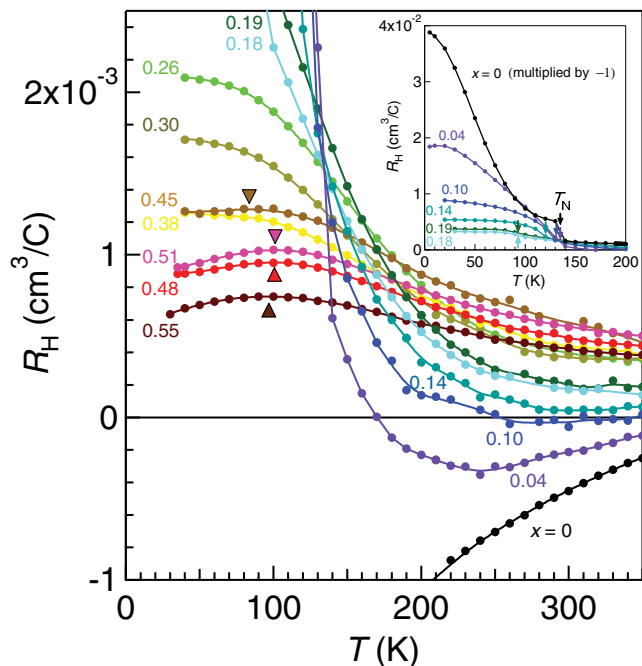


FIG. 3. (Color online) Temperature (T) dependence of Hall coefficient (R_H) for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($0 \leq x \leq 0.55$). The triangles indicate the crossover temperature, where R_H turns to a decrease upon cooling. The inset shows R_H for samples which undergo the antiferromagnetic transition at low T in a full scale. The arrows indicate the antiferromagnetic transition temperature (T_N). Note that the data for $x = 0$ are multiplied by -1 .

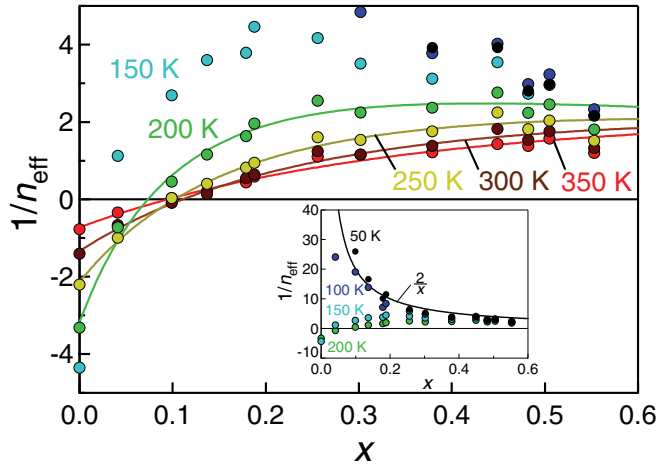


FIG. 4. (Color online) The doping dependence of the inverse effective carrier density (n_{eff}) for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($0 \leq x \leq 0.55$). The high- and low-temperature (T) data are shown in the main panel and inset, respectively. The solid lines in the main panel represent results of fitting, whereas the solid line in the inset is an $n_{\text{eff}} = x/2$ curve.

where we call the low- T phase a coherent state. From now on, we concentrate on R_H in the paramagnetic phase.

In order to interpret the complicated behavior of R_H , we first focus on the doping dependence. We introduce the effective carrier density $n_{\text{eff}} = \frac{V}{e} \frac{1}{R_H}$, where V is the volume per Fe atom and e is the elementary electric charge, and plot them against x in Fig. 4. This quantity becomes $x/2$ if the parent compound ($x = 0$) is a band insulator. Actually, the low- T data seemingly obeyed this relationship (inset of Fig. 4); however, this was an accidental agreement caused by the enhanced R_H in the underdoped region. Instead, intrinsic information could be deduced from the high- T data, where $1/n_{\text{eff}}$ increased from a negative to a positive value with increasing x . We analyzed the 350 K data by adopting the Boltzmann equation for a two-band system,¹⁶

$$\frac{1}{n_{\text{eff}}} = \frac{n_h(\tau_h/m_h)^2 - n_e(\tau_e/m_e)^2}{(n_h\tau_h/m_h + n_e\tau_e/m_e)^2},$$

where n_h (n_e), m_h (m_e), and τ_h (τ_e) are the carrier density per Fe atom, the quasiparticle mass, and the relaxation time for the hole (electron) band, respectively.^{7,9,10,17} In contrast to an alloyed system, a rigid band shift of the Fermi level from a compensated metal with $n_h = n_e = n_0$ at $x = 0$ is reasonably anticipated in the current system. Then, the carrier density changes in form in which $n_h = n_0 + \frac{x}{2} \frac{m_h}{m_h + m_e}$ and $n_e = n_0 - \frac{x}{2} \frac{m_e}{m_h + m_e}$. Substituting these expressions into the n_{eff} formula, we obtained a fitting function. We performed a fitting with the data at 350 K under the assumption that the electron band vanishes at $x = 1$ [$2n_0(1 + m_h/m_e) = 1$].¹⁸ The fitting quality was fairly good, when $n_0 = 0.12$, $m_h/m_e = 3.1$, and $\tau_h/\tau_e = 2.6$ (Fig. 4).

The obtained transport parameters are compared with the ARPES results. (1) The n_0 value corresponds to the sum of hole or electron Fermi surface areas at $x = 0$, which is reported to be 0.06–0.10.^{19,20} This value is close to our result, supporting the validity of the present analysis. (2) The quasiparticle mass

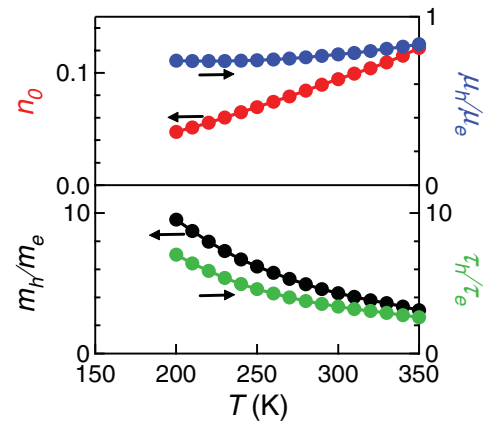


FIG. 5. (Color online) Temperature (T) dependence of transport parameters obtained by fitting with two-band Boltzmann equation for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($0 \leq x \leq 0.55$). n_0 stands for the hole or electron density per Fe atom at $x = 0$. m_h (m_e), τ_h (τ_e), and μ_h (μ_e) are the quasiparticle mass, the relaxation time, and the mobility for the hole (electron) band, respectively.

for each hole and electron band was determined by ARPES measurements for $x = 0.4$;²¹ a simple average among the three hole and two electron bands leads to $m_h/m_e = 4.8$, which is consistent with our result. Although the lighter mass of electrons than that of holes is considered to primarily originate from a strong hybridization between Fe 3d and As 4p orbitals at the zone corner of the Brillouin zone,²² we also speculate that an orbital-dependent correlation effect selectively enhanced the mass of a particular band and resulted in the mass asymmetry.²³ (3) The relaxation time obtained by our analysis pointed to an apparently peculiar conclusion: heavier holes experienced less dissipation than electrons. The relevant scattering source is likely to be phonons;²⁴ the reason why the electron-phonon interaction was so strong in electron bands is unclear at present. A detailed analysis of the quasiparticle lifetime in the ARPES spectra is therefore highly anticipated. (4) The mobility for hole (electron) bands, which are defined as $\mu_h = \tau_h/m_h$ ($\mu_e = \tau_e/m_e$), had the ratio $\mu_h/\mu_e = 0.84$; $\mu_h < \mu_e$ was the reason for the negative value of R_H at $x = 0$.

We move to the T dependence. We performed the above-mentioned fitting with the data at $200 \leq T \leq 350$ K; the results are summarized in Fig. 5. Upon cooling, n_0 showed a decrease, whereas m_h/m_e and τ_h/τ_e increased, keeping μ_h/μ_e nearly constant. We stress that R_H of underdoped samples was so significantly enhanced in magnitude in the low- T region that the present analysis is accurate in the high- T limit. Nevertheless, the global T dependence obtained might be meaningful and is expected to continue to $T < 200$ K. Here, let us divide the R_H behavior with decreasing T into two parts: (1) an increase in magnitude observed in wide x and T regions, and (2) a decrease below T^* observed in the specific composition $0.45 \leq x \leq 0.55$, and identify the transport parameters responsible for these behaviors. We claim that the former behavior was predominantly induced by the decrease in n_0 and the latter behavior was totally induced by the increase in m_h/m_e ; this was concluded by changing a transport parameter in the Boltzmann equation, while other parameters

remained fixed in a relevant parameter region. Which of the two contradictory behaviors is actually observed depends on a competition between the two underlying mechanisms, which we will argue below.

IV. DISCUSSION

A gradual increase of $|R_H|$ upon cooling is widely observed near the antiferromagnetic phase in Fe-based superconductors, and its origin is ascribed to antiferromagnetic fluctuations.^{9,10,25} The present data showing the weakened T variation of R_H , with a departure from the antiferromagnetic phase in the x - T plane, supports this conjecture. Our claim that a reduction in n_0 is responsible for this behavior points to a shrinkage of the Fermi surface areas; however, such a feature is indiscernible in the ARPES spectra.⁴ Instead, a more plausible scenario is that the n_0 decrease is a pretension produced by a phenomenological analysis; we need to directly deal with R_H in terms of the pseudogap formation³ and/or the vertex correction for the current,²⁶ both of which are supposed to originate from antiferromagnetic fluctuations.

The decrease of $|R_H|$ upon cooling is most likely relevant to an evolution of the pseudopeak structure observed in the ARPES spectra.⁴ There are two pieces of supporting evidence for this interpretation. One is that the crossover T of the two phenomena is quite similar at ~ 100 K. Another is that both phenomena are related to the increase of m_h/m_e : since the density of states is proportional to the quasiparticle mass in two dimensions, the evolution of pseudopeak only in hole bands (not in electron bands) hints at the m_h/m_e increase;⁴ we also recall that our analysis connected the $|R_H|$ decrease with the m_h/m_e increase. One might think that the composition at which the pseudopeak structure was observed ($x = 0.41$) is beyond $0.45 \leq x \leq 0.55$, where the R_H decrease was observed. However, the R_H increase upon cooling became moderate below ~ 100 K for $0.26 \leq x \leq 0.38$, hinting that the same phenomenon as observed in $0.45 \leq x \leq 0.55$ happens on the more underdoped side. We conclude that the coherent state is formed in a wider x range, but is most stable in the optimal and overdoped regions.²⁷

We now discuss the underlying microscopic mechanism of the increase of m_h/m_e , which consequently leads to the decrease of $|R_H|$ below T^* . The simplest interpretation is that holes, coupling with a certain boson centered at ~ 12 meV below the Fermi energy, formed heavy quasiparticles and acquired quantum coherence below T^* .⁴ A question to be

answered is what the boson is. A possible candidate is a magnon. However, the stable coherent state separated from the antiferromagnetic phase (Fig. 1) does not match this scenario. Thus, the nature of the coherent state below T^* , as well as its relevance to superconductivity, is unclear at present.

A comparison with other related systems merits a further understanding. Whereas an increase of $|R_H|$ upon cooling is observed in various Fe-based superconductors, including $\text{Ba}(\text{Fe}_{1-y}\text{M}_y)_2\text{As}_2$ ($M = \text{Co}, \text{Ni}, \text{Cu}, \text{and Ru}$)⁷⁻¹² and P-substituted BaFe_2As_2 ,²⁵ the decrease of $|R_H|$ is quite rare. An exceptional case is $M = \text{Co}$, which has a narrow composition range $0.09 \leq y \leq 0.15$, in which a negative R_H decreases at high T and tends to increase at ~ 80 K with decreasing T .⁷⁻¹⁰ A close inspection of the electron-hole asymmetry will open up a route to identify the boson relevant to the formation of the coherent electronic state.

Finally, we briefly comment on the relevance between the crossover phenomenon associated with ρ and R_H (open triangles and open squares in Fig. 1, respectively). Although the two characteristic T s are quite similar in the composition ranges investigated, we concluded that they have distinguishable origins. This is because of a lack of correspondence at $x = 1.0$; that is, ρ exhibited a crossover at ~ 70 K, whereas R_H monotonically increased upon cooling (data not shown). The T dependence of ρ likely reflects the Bloch-Grüneisen formula due to the scattering by lattice vibrations.

V. CONCLUSIONS

We investigated normal state properties of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ($0 \leq x \leq 0.55$) by systematic Hall coefficient measurements. The data analysis based on the Boltzmann equation indicates that heavier holes conduct smoothly while experiencing fewer dissipations than electrons. We also revealed the formation of heavy coherent quasiparticles in hole bands below ~ 100 K, predominantly in the optimally doped and overdoped regions.

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