

## Evolution from non-Fermi- to Fermi-liquid transport via isovalent doping in $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ superconductors

S. Kasahara,<sup>1,2,\*</sup> T. Shibauchi,<sup>3</sup> K. Hashimoto,<sup>3</sup> K. Ikada,<sup>3</sup> S. Tonegawa,<sup>3</sup> R. Okazaki,<sup>3</sup> H. Shishido,<sup>3</sup> H. Ikeda,<sup>3</sup> H. Takeya,<sup>2</sup> K. Hirata,<sup>2</sup> T. Terashima,<sup>1</sup> and Y. Matsuda<sup>3</sup>

<sup>1</sup>Research Center for Low Temperature and Materials Sciences, Kyoto University, Kyoto 606-8502, Japan

<sup>2</sup>Superconducting Materials Center, National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan

<sup>3</sup>Department of Physics, Kyoto University, Kyoto 606-8502, Japan

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The normal-state charge transport is studied systematically in high-quality single crystals of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  ( $0 \leq x \leq 0.71$ ). By substituting isovalent P for As, the spin-density-wave (SDW) state is suppressed and the dome-shaped superconducting phase ( $T_c \lesssim 31$  K) appears. Near the SDW end point ( $x \approx 0.3$ ), we observe striking linear temperature ( $T$ ) dependence of resistivity in a wide  $T$  range, and remarkable low- $T$  enhancement of Hall-coefficient magnitude from the carrier number estimates. We also find that the magnetoresistance apparently violates the Kohler's rule and is well scaled by the Hall angle  $\Theta_H$  as  $\Delta\rho_{xx}/\rho_{xx} \propto \tan^2 \Theta_H$ . These non-Fermi-liquid transport anomalies cannot be attributed to the simple multiband effects. These results capture universal features of correlated electron systems in the presence of strong antiferromagnetic fluctuations.

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### I. INTRODUCTION

Within the last decade, it has been found that in strongly correlated electron systems various transport properties display striking deviations from the conventional Fermi-liquid behavior, particularly in the vicinity of magnetic instability. It is generally believed that strong-magnetic fluctuations seriously modify the quasiparticle masses and scattering cross section of the Fermi liquid. Very recently discovered Fe pnictides<sup>1</sup> have stimulated great interest because the high- $T_c$  superconductivity with nontrivial Cooper pairing state occurs in the vicinity of the spin-density-wave (SDW) instability. It has been suggested that the antiferromagnetic fluctuations associated with the Fermi-surface nesting between electron and hole pockets are essential for the occurrence of the superconductivity.<sup>2-4</sup> Therefore, clarifying the normal-state electron transport of Fe pnictides is of crucial importance since it might be a key to elucidating the mechanism of high- $T_c$  superconductivity.

In Fe pnictides, systematic transport studies using high-quality single crystals have been fairly limited, while those provide important clues for understanding the nature of superconductivity, as revealed by the research of cuprate and heavy-fermion materials. Although anomalous  $T$ -linear resistivity<sup>5-10</sup> have been reported in single crystals of electron-doped  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , the interpretations are yet controversial as to the importance of magnetic fluctuations and multiband effects. The observation of  $T$ -linear resistivity alone cannot be simply taken as evidence for the deviation from the conventional Fermi-liquid theory because it can appear as a result of multiband effect. Indeed, the  $T$ -linear resistivity, as well as  $T$ -dependent Hall coefficients, have been discussed in terms of the multiband effect inevitably present in the electron-doped  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  system.<sup>8,9</sup> However, the carrier doping, which gives rise to the imbalance between the electron and hole numbers, makes the interpretation of the observed transport properties rather complicated. Be-

sides, the scattering effect of the substituted atoms within the Fe planes is still unclear.

Here we present a systematic study of the dc resistivity, Hall effect, and magnetoresistance in the normal state of high-quality single crystals of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ , ranging from the undoped SDW ( $x=0$ ) state to the overdoped Fermi-liquid state ( $x \approx 0.71$ ) through the highly unusual non-Fermi-liquid state. By substituting isovalent P for As, the SDW state is suppressed and the superconducting phase ( $T_c \lesssim 31$  K) appears.<sup>11</sup> We consider that  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  is the most suitable system to study the transport properties of Fe pnictides because of the following reasons. First, the system can be assumed as a compensated metal, i.e., essentially the same number of electrons and holes,  $n_e = n_h = n$  for any  $x$  value, which is supported by the band-structure calculations.<sup>12</sup> Therefore we can only tune magnetic character without nominally changing charge-carrier concentrations. Indeed, it is theoretically suggested that P doping of As presents a means to access a magnetic quantum criticality in an unmasked fashion.<sup>13</sup> Second, we succeeded to grow very clean single crystals with large residual resistivity ratio values ( $\approx 25$  for  $x=0.41$ ). In fact, in recent high-field studies clear quantum oscillations are observed in our doped crystals at  $x \gtrsim 0.4$ ,<sup>14</sup> indicating that the substitution in the pnictogen sites induces less impurity scattering than that in the Fe planes.<sup>7-10</sup> By using these clean crystals, we observe that all of the transport coefficients exhibit striking deviations from the conventional Fermi-liquid behaviors near the SDW end point. These anomalies become less pronounced as the nesting conditions degrade with overdoping. We show that these non-Fermi-liquid transport anomalies cannot be attributed to the simple multiband effects. Several noticeable features highlight common non-Fermi-liquid behaviors in strongly correlated electron systems in the vicinity of antiferromagnetism.

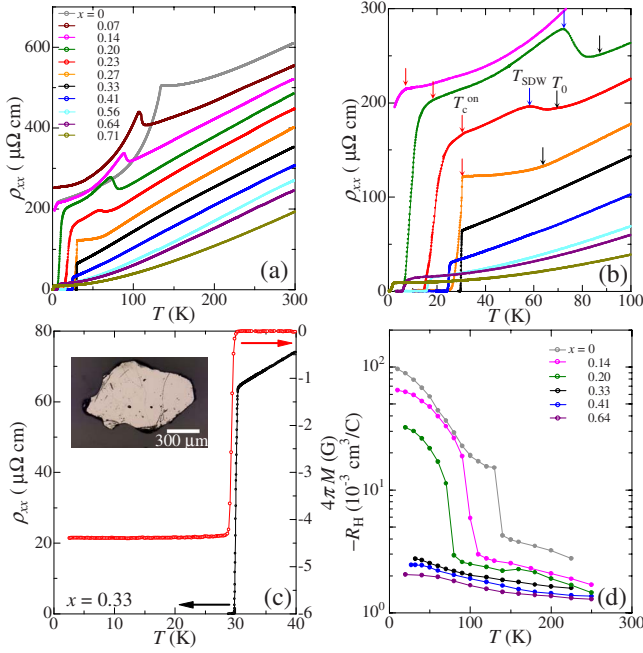


FIG. 1. (Color online) (a)  $\rho_{xx}(T)$  curves of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  in zero field. (b) The same data below 100 K. (c)  $\rho_{xx}(T)$  and de-magnetization curve,  $M(T)$ , measured by superconducting quantum interference device magnetometer, recorded for zero-field-cooling process for a  $x=0.33$  crystal. For  $M(T)$ , a small magnetic field (3 Oe) is applied along the  $c$  axis. The inset is a photograph of the (001) surface of a single crystal. (d)  $T$  dependence of the Hall coefficient  $R_H(T)$  for various doping levels.

## II. EXPERIMENTAL

Single crystals of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  were grown from stoichiometric mixtures of Ba (flakes), and FeAs, Fe, P, or FeP (powders) placed in an alumina crucible, sealed in an evacuated quartz tube. It was heated up to 1150–1200 °C, kept for 12 h, and then cooled slowly down to  $\sim 800$  °C at the rate of 1.5 °C/h. Platelet crystals with shiny (001) surface were extracted [inset of Fig. 1(c)].  $x$  values were determined by an energy dispersive x-ray analyzer.

Transport measurements were performed using an ac-resistance bridge (LR-700, Linear Research Inc.) or a nanovoltmeter (Model 2182A/6221, Keithley) equipped with Delta mode. For Hall/magnetoresistance measurements, we took data by sweeping positive and negative magnetic fields up to 5/14 T generated by a superconducting magnet at each temperature. The Hall coefficient  $R_H$  is defined as the field derivative of Hall resistivity  $\rho_{xy}$ ,  $R_H \equiv d\rho_{xy}/dH$ , at  $\mu_0 H \rightarrow 0$  T.<sup>15</sup> Band structure was calculated by density-functional theory implemented in the WIEN2K code.<sup>16</sup>

## III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the in-plane resistivity  $\rho_{xx}(T)$  in zero field below 300 K and 100 K, respectively. An anomaly in  $\rho_{xx}(T)$  in the parent  $\text{BaFe}_2\text{As}_2$  at  $T_0=137$  K corresponds to the structural and simultaneous SDW transitions ( $T_0=T_{\text{SDW}}$ ), consistent with the previous studies.<sup>17,18</sup> With

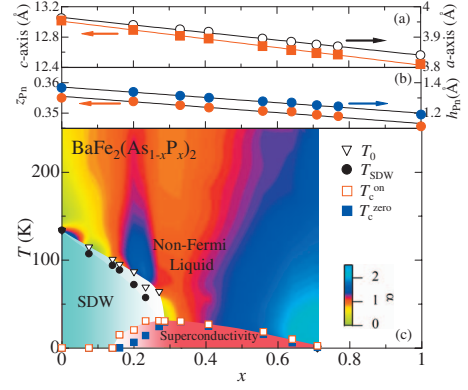


FIG. 2. (Color) (a) Lattice constants determined from x-ray as a function of  $x$ . (b) The  $z$  coordinate of pnictogen atoms in the unit cell  $z_{\text{Pn}}$  and the pnictogen height from the iron plane  $h_{\text{Pn}}=(z_{\text{Pn}}-0.25)\times c$ . (c) Phase diagram of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  against the P content  $x$ . The open triangles show the structural transition at  $T_0$ . The closed black circles show  $T_{\text{SDW}}$ , where the resistivity shows reductions due to the reduced spin scattering at the SDW transition. The onset of superconductivity,  $T_c^{\text{on}}$ , and the zero resistivity temperature,  $T_c^{\text{zero}}$ , are displayed as open red and closed blue squares. Colors in the nonmagnetic normal state represent evolution of the exponent  $\alpha$  in resistivity fitted by Eq. (1).

increasing  $x$ , the anomaly is replaced by a step like increase at  $T_0$ , followed by a sharp peak at  $T_{\text{SDW}}$ . The increase in  $x$  suppresses these anomalies toward lower temperatures. At the same time, the resistivity shows a drop at lower temperatures and zero resistivity is attained at  $x \geq 0.20$ , indicating a coexistence of SDW and superconductivity. The coexistence is observed up to  $x=0.28$  and no anomaly associated with the SDW transition is observed for  $x \geq 0.33$ . As shown in Fig. 1(c), the crystals exhibit a very narrow superconducting transition width ( $\Delta T_c < 0.4$  K for  $x=0.33$ ). dc magnetization,  $M(T)$ , recorded for zero-field-cooling process shows fully shielded Meissner signal. Specific heat of a single crystal from the same batch also shows sharp jump at  $T_c$ , indicating excellent quality of our crystals.<sup>19,20</sup>

Figure 1(d) depicts the temperature dependence of the Hall coefficient  $R_H$  at various doping levels.  $R_H$  is negative, indicating the dominant contribution of electrons. For  $x \leq 0.14$ , with decreasing temperature  $|R_H|$  jumps to higher values at  $T_{\text{SDW}}$ , indicating a reduction in carriers due to the development of SDW gap in the Fermi surface.  $R_H$  is strongly temperature dependent even in the nonmagnetic phase.

Figure 2(c) displays the  $T$ - $x$  phase diagram of the  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  system obtained in the present study.  $T_{\text{SDW}}$  decreases rapidly with  $x$ . At  $0.14 \leq x \leq 0.30$ , both the SDW and superconducting transitions are observed, suggesting that the superconductivity may not have a bulk character. In the vicinity of  $x \approx 0.30$ , the SDW transition disappears suddenly and a rapid growth of bulk superconductivity appears. The superconducting transition shows a maximum  $T_c=31$  K at  $x=0.26$  and further increase in  $x$  leads to the reduction in  $T_c$ . The obtained phase diagram bears striking resemblance to that of the pressure dependence of  $\text{BaFe}_2\text{As}_2$ ,<sup>21</sup> indicating that the isovalent substitution of P for As is identical to the pressure effect. In fact, the lattice constants as well as the

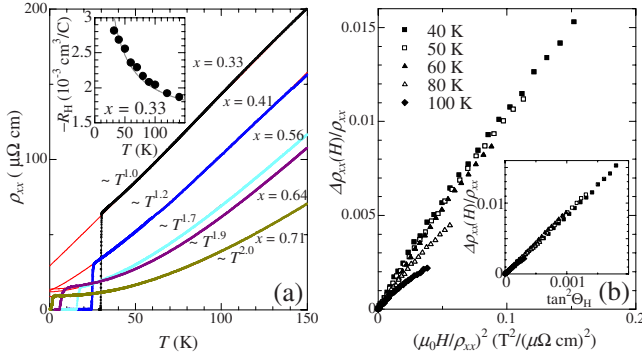


FIG. 3. (Color online) (a) Normal-state  $\rho_{xx}(T)$  for  $x=0.33, 0.41, 0.56, 0.64,$  and  $0.71$  at low temperatures can be fitted by the power law [Eq. (1)]. The inset shows  $T$  dependence of  $-R_H(T)$  for  $x=0.33$ . Solid line is a fit to the data by  $-R_H(T)=C_1/T+C_2$  with  $C_1=0.048 \text{ Kcm}^3/\text{C}$  and  $C_2=1.5 \times 10^{-3} \text{ cm}^3/\text{C}$ . (b) Magnetoresistance  $\Delta\rho_{xx}(H)/\rho_{xx}$  plotted as a function of  $(\mu_0 H/\rho_{xx})^2$  for  $x=0.33$ . The inset shows  $\Delta\rho_{xx}(H)/\rho_{xx}$  plotted as a function  $\tan^2 \Theta_H$ .

pnictogen height decrease linearly with  $x$ , as shown in Figs. 2(a) and 2(b).

Now we discuss the normal-state transport properties focusing on the nonmagnetic regime. At  $x=0.33$  just beyond the SDW end point,  $\rho_{xx}(T)$  exhibits a nearly perfect  $T$ -linear dependence in a wide  $T$  range above  $T_c$  as shown in Fig. 3(a)

$$\rho_{xx}(T) = \rho_0 + AT^\alpha \quad (1)$$

with  $\alpha=1.0$ , where  $A$  is a constant. Thus, the resistivity exhibits a striking deviation from the standard Fermi-liquid theory with  $\alpha=2$ . Based on the two-dimensional (2D) electron-gas model, the conductivity due to five Fermi surfaces is roughly estimated as  $\sigma=5 \times (e^2/h)k_F l_{\text{mfp}}$ , where  $k_F$  is Fermi wave number and  $l_{\text{mfp}}$  is mean-free path of carriers.<sup>22</sup> Thus the Ioffe-Regel limit, which corresponds to  $k_F l_{\text{mfp}} \sim 1$ , is roughly estimated as  $\rho \sim 350 \mu\Omega\text{cm}$ . This may be relevant to the deviation from the  $T$ -linear behavior of  $\rho(T)$  at high  $T$  ( $>150 \text{ K}$ ). With increasing  $x$ ,  $\alpha$  increases and the Fermi-liquid behavior is recovered at  $x=0.71$ . The contour plot of  $\alpha$  in Fig. 2(c) demonstrates this evolution in the phase diagram, indicating that the deviations continue to lower temperatures as  $x \rightarrow 0.33$ . This sheds light on the V-shaped region where anomalous  $T$ -linear behavior takes place, which suggest a strong similarity to the non-Fermi-liquid behaviors governed by quantum fluctuations in strongly correlated electron systems.<sup>23</sup> For  $x=0.33$ ,  $R_H$  exhibits a marked  $T$  dependence that is approximated as  $-R_H(T)=C_1/T+C_2$ , where  $C_1$  and  $C_2$  are positive constants, as depicted in the inset of Fig. 3(a). Similar anomalous behaviors of  $\rho_{xx}$  and  $R_H$  are reported in other Fe pnictides.<sup>5-9</sup> It is well known that temperature dependent  $R_H$  can be obtained in the Bloch theory when multiple bands are involved. Then an important question is whether the most fundamental transport properties described above can be accounted for by the conventional multiband model or can be indicative of unconventional transport properties inherent to the Fe-based systems. We show that the former is highly unlikely for the following reasons.

In the compensated two-band metal with equal number of electrons and holes, the Hall coefficient is described simply as

$$R_H = \frac{1}{ne} \times \frac{\sigma_h - \sigma_e}{\sigma_h + \sigma_e}, \quad (2)$$

where  $\sigma_e$  ( $\sigma_h$ ) is the conductivity of electron (hole) band. The fact that  $R_H$  is negative indicates that the electron band dominates transport properties ( $\sigma_e > \sigma_h$ ). Strong evidence against the simple multiband explanation is obtained from the amplitude of  $R_H$ . From Eq. (2),  $|R_H|$  cannot exceed  $1/ne$ . Band calculations reveal that  $\text{BaFe}_2\text{As}_2$  has  $\sim 0.15$  electrons per Fe (Ref. 8) and that the electron Fermi surface is not seriously influenced by the P replacement (see below). This electron density corresponds to  $1/ne \approx 0.98 \times 10^{-3} \text{ cm}^3/\text{C}$ . However, it is clear from the inset of Fig. 3(a) that the observed magnitude of  $R_H$  becomes considerably larger than this value especially at low temperatures. These results lead us to conclude that the simple multiband picture cannot explain the transport coefficients in the present system.

Another anomalous feature is also found in magnetoresistance (MR). In the conventional Fermi-liquid state, the MR,  $\Delta\rho_{xx}(H)/\rho_{xx} \equiv [\rho_{xx}(H) - \rho_{xx}(H=0)]/\rho_{xx}$ , due to an orbital motion of carriers is simply scaled by the product of cyclotron frequency  $\omega_c$  and scattering time  $\tau$  as  $\Delta\rho_{xx}(H)/\rho_{xx} = F(\omega_c \tau)$ . This is so-called Kohler's rule. Since  $\omega_c \propto H$  and  $\tau \propto \rho_{xx}^{-1}$ , the Kohler's rule is rewritten as  $\Delta\rho_{xx}(H)/\rho_{xx} = F(\mu_0 H/\rho_{xx})$ , where  $F(y)$  is a function of  $y$  depending on the electronic structure. Figure 3(b) is the transverse MR plotted against  $\mu_0 H/\rho_{xx}$  for  $x=0.33$  in  $\mathbf{H} \parallel c$ . The data at different temperatures are on distinctly different curves, indicating apparent violation of the Kohler's rule. The result means  $\omega_c \tau$  is no longer a scale parameter for MR due to an orbital motion of carriers.

It has been proposed<sup>24,25</sup> that the MR in the non-Fermi-liquid regime may be scaled by the Hall angle  $\Theta_H (\equiv \tan^{-1} \frac{\rho_{xy}}{\rho_{xx}})$  as  $\Delta\rho_{xx}(H)/\rho_{xx} \propto \tan^2 \Theta_H$  (modified Kohler's rule). To examine this relation, we plot the MR as a function of  $\tan^2 \Theta_H$  in the inset of Fig. 3(b). Obviously, the MR data at different temperatures collapse into the same curve, indicating a distinct Hall-angle scaling of the MR.

It should be noted that the modified Kohler's rule as well as the  $T$ -linear  $\rho_{xx}$  and the low-temperature enhancement of  $|R_H| (\gg 1/ne)$ , distinct from the standard Fermi-liquid theory of metals, have also been reported in other strongly correlated electron systems including high- $T_c$  cuprates<sup>26</sup> and 2D heavy-fermion compounds.<sup>27</sup> The simultaneous understanding of these anomalies has been a subject of intense research.<sup>24,25,28,29</sup> Among others, one may involve different quasiparticle scattering times  $\tau$  at different parts of Fermi surfaces.<sup>25,28</sup> The effects of band curvature and Fermi velocity anisotropy on  $\tau$  can account for the enhancement of  $|R_H|$ . Another important effect is the vertex corrections to the longitudinal and transverse conductivities due to large antiferromagnetic fluctuations,<sup>24</sup> which modify the current at the Fermi-surface spots connecting with the nesting vectors, resulting in enhanced  $|R_H|$ .<sup>27</sup> Although quantitative analysis of these effects in the Fe-based superconductors deserves fur-



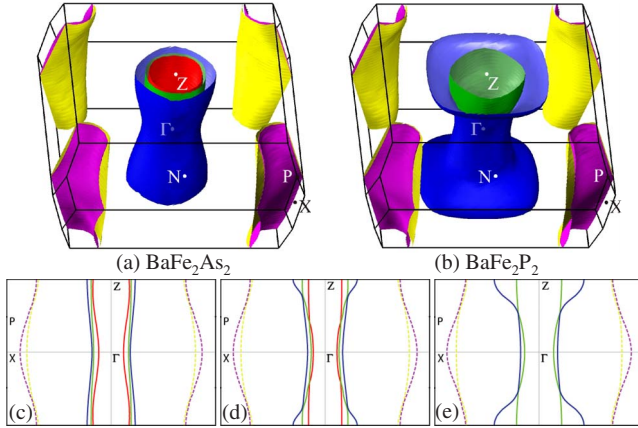


FIG. 4. (Color online) Fermi surfaces of (a)  $\text{BaFe}_2\text{As}_2$  ( $x=0$ ) and (b)  $\text{BaFe}_2\text{P}_2$  ( $x=1$ ). Cross-section views along (110) plane are also shown for (c)  $x=0$ , (d) 0.3, and (e) 1. For  $x=0.3$ , we use the  $\text{BaFe}_2\text{As}_2$  structure with the experimental atomic positions.

ther theoretical studies, the Hall-angle scaling of the MR can be consistently explained by these theories.<sup>24,25</sup> In any case, the observed non-Fermi-liquid properties strongly suggest the significance of antiferromagnetic fluctuations in the transport coefficients. When we go away from the antiferromagnetism by overdoping, these odd properties indeed become less pronounced as shown in Fig. 3(a), consistent with the above view.

The effect of antiferromagnetic fluctuations on the transport properties is also suggested by electronic structure calculations. Figure 4 displays Fermi surfaces calculated for  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  ( $x=0, 0.3$ , and 1). Three hole sheets exist around  $\Gamma$  in  $\text{BaFe}_2\text{As}_2$  while one of them is absent in  $\text{BaFe}_2\text{P}_2$ . Both compounds have two electron pockets around  $X$ . Three dimensionality of the hole Fermi surfaces is quite sensitive to the pnictogen position  $z_{\text{pn}}$ . Similar sensitivity has also been pointed out in  $\text{LaFePO}$  (Ref. 30) and  $\text{SrFe}_2\text{P}_2$ .<sup>31</sup> In the present  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  system, the substitution of P for As is expected to induce a small reduction in  $z_{\text{pn}}$  [Fig. 2(b)],

which dramatically promotes the three dimensionality of the hole sheets. The enlarged Fermi-surface warping upon doping is considered to weaken the nesting along the  $(\pi, \pi)$  direction, minimize the SDW phase, and then induce the superconductivity. In contrast to the significant change in the hole sheets, the electron sheets, which are the dominant carriers for the transport, are almost unchanged. Thus the large change in the antiferromagnetic nesting conditions is the most relevant to the evolution from the non-Fermi- to Fermi-liquid properties by the isovalent doping in this system.

#### IV. CONCLUSIONS

In summary, we have systematically studied the electronic-transport properties of  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  using high-quality single crystals, ranging from nondoped SDW to overdoped Fermi-liquid states. Near the SDW end point, anomalous non-Fermi-liquid behaviors, including a pronounced  $T$ -linear  $\rho_{xx}$ , a striking enhancement of Hall coefficient at low  $T$  ( $|R_H| \gg 1/ne$ ), and violation of the Kohler's rule and Hall-angle scaling in the magnetoresistance, are observed. These cannot be attributed to the simple multiband effect. The doping dependence points to an important role of antiferromagnetic fluctuations in the non-Fermi-liquid transport properties in the Fe pnictides. These highly unusual transport properties, commonly observed in Fe-pnictides, high- $T_c$  cuprates, and heavy-fermion superconductors, very likely capture universal features of correlated electron system near the magnetic instability.

#### ACKNOWLEDGMENTS

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\*kasa@scphys.kyoto-u.ac.jp

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