

Resistivity of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$: Evidence for a broad composition range of non-Fermi-liquid behavior

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The resistivity vs temperature of annealed and unannealed single crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ over a broad range of nominal compositions between $x = 0.05$ and 0.30 , with special focus on compositions around that (x_{opt}) of the optimal superconducting transition temperature, is reported. Above any transition temperature, either superconducting (T_c) or structural (T_S) for $x < x_{\text{opt}}$, the low temperature resistivity, ρ , behaves approximately linearly with temperature from $x = 0.05$ up to at least $x = 0.095$, with $\rho \propto T^2$ starting for $x = 0.14$. The apparent constancy of the temperature dependence over a broad composition range argues strongly for the presence of a quantum critical point (QCP) underneath the superconducting dome. However, the temperature dependences measured down to T_c leave the exact location of the QCP open. Interestingly, although the superconducting transition temperature rises by ≈ 1.5 K with annealing, the temperature dependence of ρ appears unaffected by annealing, i.e., the QCP appears insensitive to small amounts of lattice disorder.

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I. INTRODUCTION

The source of the superconducting pairing mechanism in the iron pnictide and chalcogenide superconductors (FePn/Ch) remains a focus of research.^{1,2} There have been numerous suggestions that the fact that the magnetic spin density wave (SDW) transition temperature, T_{SDW} , goes to $T = 0$ at around the maximum superconducting transition temperature, T_c , of many FePn/Ch suggests a quantum critical point (QCP). The fluctuations associated with a QCP could play a dominant role in the superconducting mechanism. Two of the FePn/Ch for which quantum critical behavior has been suggested are P-doped $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ and Co-doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$.

In the initial work³ on the $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ superconducting system, Jiang *et al.* note that the low temperature resistivity for $x = 0.32$ and 0.45 , near the maximum (optimal) T_c of 30 K, varies as T^α , $\alpha = 0.9 \pm 0.1$, from T_c up to 50 K. For the other compositions ($x \leq 0.2$, where $T_c \leq 6$ K, i.e., almost to the left of the superconducting dome and $x \geq 0.55$, where $T_c \leq 22$ K, i.e., starting to fall off the peak of $T_c^{\text{opt}} = 30$ K), α values mostly clustered around 2 ± 0.4 , much closer to the normal Fermi liquid behavior of $\rho \propto T^2$. The authors then suggest that there is a QCP present near optimal doping based on the non-Fermi liquid, $\rho \approx \rho_0 + T^1$, behavior in the resistivity. Since P is isoelectronic with As, later work⁴ showed that single crystals can be clean and have a relatively high (≈ 25) residual resistivity ratio ($= R(300)/R(T \rightarrow 0)$, a metric of sample quality) since there is no doping in the Fe layer. Hashimoto *et al.*⁵ in single-crystal $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ report a sharp peak in the zero temperature penetration depth, $\lambda(0)$, at the optimal doping, $x = 0.3$ and $T_c = 30$ K, and interpret this as further evidence for a QCP at $x = 0.3$. Shibauchi, Carrington, and Matsuda also interpret⁶ this result as “clear and direct evidence” of a QCP under the superconducting dome. However, Chowdhury *et al.*⁷ put forth a theoretical argument that the QCP lies not at the point where, at optimal doping, $T_{\text{SDW}} \rightarrow 0$ but rather further to the left (i.e., $x < 0.3$) in the phase diagram where magnetism and superconductivity still coexist.

In the initial work⁸ on the superconducting system $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, Sefat *et al.* report resistivity data for $x = 0.1$, $T_c = 22$ K, but do not comment on the exponent α . Ahilan *et al.*⁹ point out the possibility of a critical point in the phase diagram and find that $\rho = \rho_0 + AT^1$ for $x = 0.1$ from $T_c = 22$ K up to 100 K. A following work¹⁰ reported the resistivity for $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ for nine compositions in the range $0 \leq x \leq 0.114$, with again no discussion of the temperature exponent α nor of a QCP.

A number of works since Ahilan *et al.*'s mention of $\rho \propto T^1$ (or non-Fermi-liquid behavior at least indicative of a QCP) have also discussed evidence of quantum critical behavior in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ based on various measurements, including data on divergent nematic susceptibility.¹¹ Since lattice disorder may affect resistivity, the present work reports on the resistivity of both annealed and unannealed single crystal samples of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ with particular emphasis on annealed compositions near the optimal concentration. The goal is to map out the temperature dependence of ρ vs composition near the peak of the superconducting dome, i.e., near x_{opt} , to investigate the possibility and location of a QCP.

II. EXPERIMENTAL

Single crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, $x = 0.05, 0.0725, 0.075, 0.0766, 0.0783, 0.08, 0.0825, 0.083, 0.085, 0.0875, 0.09, 0.095, 0.14, 0.15, 0.19, 0.20,$ and 0.30 , were grown using the self-flux method.¹² Samples were placed in alumina crucibles and sealed in a Nb cylinder with welded end caps. These Nb containers were then placed in a flow of ultra-high purity Ar in a horizontal furnace tube and heated to 1200°C , followed by slow cooling. Single-crystal samples were separated using mechanical means. In order to optimize T_c in the samples, a study¹³ of T_c onset and width vs annealing was undertaken. The samples were annealed at $650, 700, 800,$ and 900°C for 1, 2, and 4 weeks in sealed quartz tubes with an As vapor source¹⁴ present. Due to this vapor source, samples remained shiny in appearance after annealing. The results were that the increase in T_c onset came rather rapidly

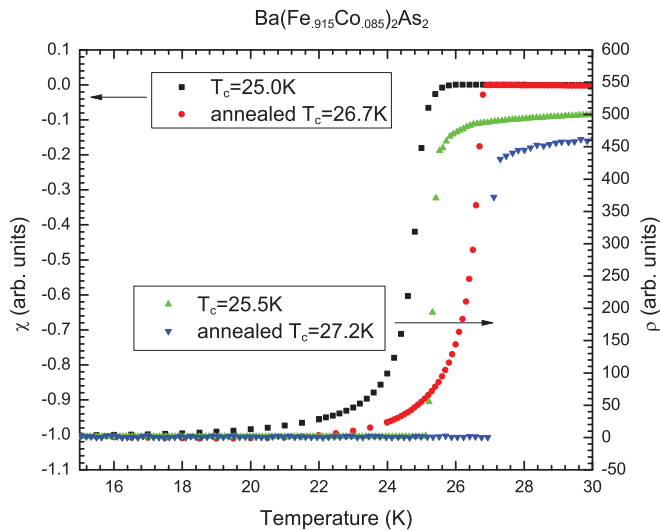


FIG. 1. (Color online) Comparison of χ (left-hand axis) and ρ (right-hand axis) for annealed and unannealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, $x = 0.085$. Note that, as is typical of such measurements, the resistivity data for a given sample has a higher T_c^{onset} and a narrower transition width than the χ data for the corresponding sample. Note that the T_c^{onset} determined from the ρ data from the annealed single crystal of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, $x = 0.085$ is 27.2 K, a record for BaFe_2As_2 samples doped on the Fe site.

with annealing, and that either 1 or 2 weeks at either 700 or 800 °C produced essentially the same T_c onset increase (≈ 1.5 K) vs the unannealed superconducting samples. Figure 1 shows a comparison of the magnetic susceptibilities, χ , and resistances, ρ , of annealed (2 weeks at 800 °C) and unannealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, $x = 0.085$. Note that T_c^{onset} for the annealed sample determined by the resistivity is 27.2 K, an increase of 5 K over that reported in the initial work. Figure 2 shows a picture of typical annealed and unannealed crystals, showing that the annealed sample still displays the original mirrorlike surface of the unannealed sample.

III. RESULTS AND DISCUSSION

The resistivity of annealed single crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ for the various compositions in the present work is shown in Fig. 3.

Let us now consider two features of the resistivity data: (1) the development of the high temperature magnetic and

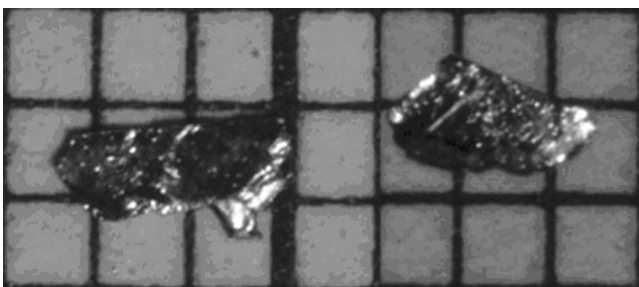


FIG. 2. Picture of unannealed (left) and annealed pieces of single crystal of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ with a nominal concentration of $x = 0.083$. The lines are 1 mm in spacing.

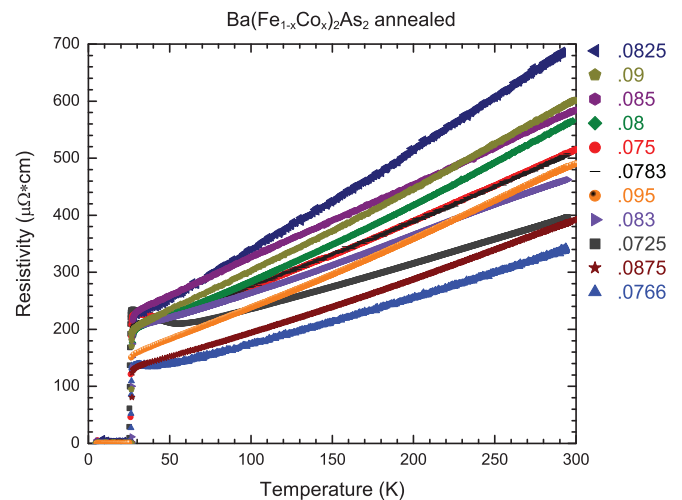


FIG. 3. (Color online) Resistivity vs temperature for annealed single crystal samples of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, for x around x_{opt} , where the compositions given are nominal. Visually, it is clear—as will be discussed quantitatively below—that the temperature dependence is approximately linear, with some (\sim factor two) variation in the slope. As has been pointed out (Ref. 10) in the literature previously, the actual composition, as measured by microscopic analysis such as wavelength dispersive x-ray spectroscopy, is approximately 0.74 times the nominal concentration. Thus, for example, our nominal concentration of $x = 0.085$ would correspond to an actual composition of $x = 0.063$. Unless otherwise mentioned, nominal compositions are used in the discussion.

structural transitions (T_{SDW} and T_S , respectively) present in the ρ data for $x = 0.05$, 0.0725, 0.075, and 0.0766; and (2) the temperature dependence of ρ over the whole doping range with attention both on non-Fermi-liquid behavior and the development of T^2 (Fermi-liquid behavior) in the resistivity at higher doping concentrations. The dependence of the behavior of ρ with temperature as a function of annealing will also be discussed.

The SDW and the structural (tetragonal \rightarrow orthorhombic) transitions that occur in many of the FePn/Ch superconductors have been thoroughly studied via neutron scattering and x-ray diffraction experiments.^{1,2} These two transitions occur at the same temperature (≈ 142 K) in the undoped BaFe_2As_2 but have been known to separate upon doping of the Fe site since the first doping studies.^{1,2} Once the microscopic origins of these transitions were determined by neutron and x-ray studies, various other (simpler) methods to determine T_{SDW} and T_S in the FePn/Ch as a function of doping have been used, with resistivity being relatively straightforward.^{10,15} Obviously, looking for features in the resistivity to determine these two transition temperatures only works when they occur at different temperatures. Since we have focused in this work on $x \geq 0.05$, T_{SDW} and T_S are easily distinguishable. We show an example in Fig. 4, where T_S for our annealed single-crystal $\text{Ba}(\text{Fe}_{0.9275}\text{Co}_{0.0725})_2\text{As}_2$ is taken as the point where the slope of ρ with decreasing temperature starts to decrease more rapidly, and T_{SDW} is taken as the inflection point (minimum in $d\rho/dT$) of the ρ vs T data. The drawback of this method of determining T_{SDW} and T_S (a weakness shared by several other measurement techniques like magnetic susceptibility as well)

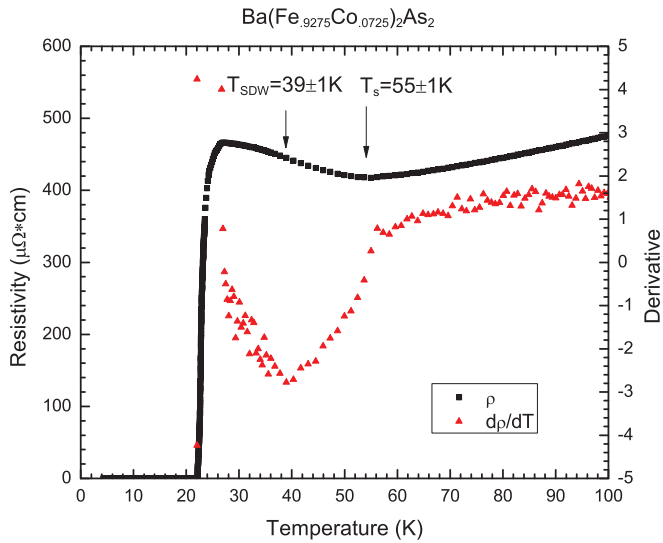


FIG. 4. (Color online) Resistivity (left axis, black square data) and the first derivative of the resistivity with respect to temperature, $d\rho/dT$ (right axis, red triangle data) for $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, $x = 0.0725$. This construction allows a rather precise determination of T_{SDW} and T_S as shown in the figure.

is that the ability to follow the transitions cuts off at the top of the superconducting dome at T_c where $\rho \rightarrow 0$ and χ becomes diamagnetic. Additionally, the presence of the transitions in the resistivity obscures the underlying temperature dependence in the temperature range of the transitions. Thus, as clear from the ρ data in Fig. 4, the temperature dependence of ρ in, e.g., $x = 0.0725$, in our search for the correct location of the possible location of a QCP in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ can only be determined above $T_S = 55$ K.

In Fig. 5, we display the result of this analysis for the four compositions (up to $x = 0.0766$) in the present work, where such a treatment of the resistivity gives a clear result for T_{SDW} and T_S . As can be seen, the results for T_{SDW} and T_S do not strongly depend on annealing. These data presented in Fig. 5 agree well (once the nominal compositions used here are scaled by 0.74 as discussed in Fig. 3) with the published results from Nandi *et al.*¹⁵ on their unannealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. As well, T_c values vs nominal composition for the annealed and unannealed single crystals of the present work are shown in Fig. 5.

Considering now the temperature dependence above T_c (or for $x < 0.08$, above T_S) of the ρ data in Fig. 3, Fig. 6 offers a few examples for five different compositions of the annealed single crystals.

The temperature exponent in $\rho = \rho_0 + AT^\alpha$ is for $x = 0.05$, $\alpha = 1.181$ (fit range 125–300 K); for $x = 0.0825$, $\alpha = 1.050$ (30–300 K); for $x = 0.095$, $\alpha = 1.069$ (30–300 K). For compositions between $x = 0.05$ and 0.095 that are not shown here in Fig. 6(a) (but see Fig. 3 for the raw data), α varies between 0.969 ($x = 0.090$) to 1.272 ($x = 0.0783$), i.e., the exponents cluster rather tightly around the value of $\alpha = 1.1 \pm 0.15$.

For the two strongly overdoped concentrations shown in Fig. 6(a), the Fermi-liquid T^2 behavior starts to come in at low temperatures for $x = 0.14$, where $\alpha = 2$ in the fit range

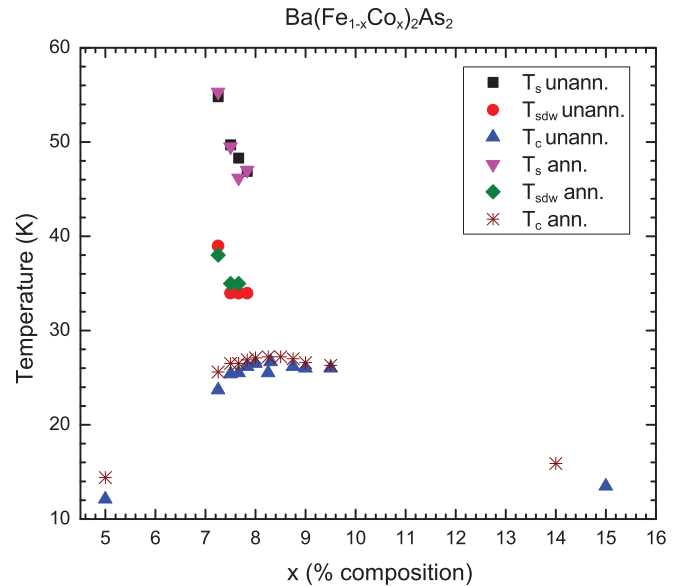


FIG. 5. (Color online) Structural (T_S), magnetic (T_{SDW}), and superconducting (T_c) transitions in annealed and unannealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. As discussed in Fig. 3, our nominal compositions are, as a rough estimate, 1/0.74 times the actual compositions reported by others. This scaling factor between actual and nominal in self-flux prepared crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ was first reported by Ni *et al.* (see Ref. 10).

19–30 K, but $\alpha = 1.49$ from 160–300 K. For $x = 0.20$, an approximate Fermi-liquid behavior dominates over a wide temperature range, with $\alpha = 1.881$ between 50–300 K.

Power laws for fits to the unannealed ρ data, for which the same compositions as shown in Fig. 6(a) are shown in Fig. 6(b), give $\alpha = 1.171, 1.199, 1.006, 2$ (23–40 K)/1.43 (150–300 K), and 1.41 (25–100 K)/1.86 (158–300 K), respectively, i.e., comparable to the annealed results. For the other unannealed compositions not shown in Fig. 6(b) between $x = 0.05$ and 0.095, α varies between 0.992 ($x = 0.090$) and 1.453 ($x = 0.0766$).

Armed with these values, the visual trends in the resistivity dependence on temperature shown for all the compositions in Fig. 3 become more quantitatively clear. From $x = 0.05$ ($T_c = 14.4$ K) through 0.095 ($T_c = 23$ K), with some variation that is clear from Fig. 3, the exponent α remains rather close to 1.0 as was found in the P -doped $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ work.³ Only starting further away from optimally doped does the Fermi-liquid, $\rho = \rho_0 + AT^2$, behavior expected¹⁶ away from a QCP (and observed³ in phosphorous doped away from optimally doped) begins to grow into a short temperature range above T_c with continued temperature range extent out at $x = 0.20$. By $x = 0.30$ (data not shown, no superconductivity detected down to 4 K), T^2 can be fit to the resistivity over the whole temperature range from 4 to 300 K.

Thus, the current work presents results in which, over a fairly large part of the superconducting dome at numerous finely spaced compositions in the phase diagram of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, the resistivity behaves as approximately $\rho = \rho_0 + T^1$, which is an expected¹⁶ non-Fermi-liquid behavior near a QCP. If high field (~ 27 T)¹⁷ resistivity data were taken to follow the exponent down to lower temperature,

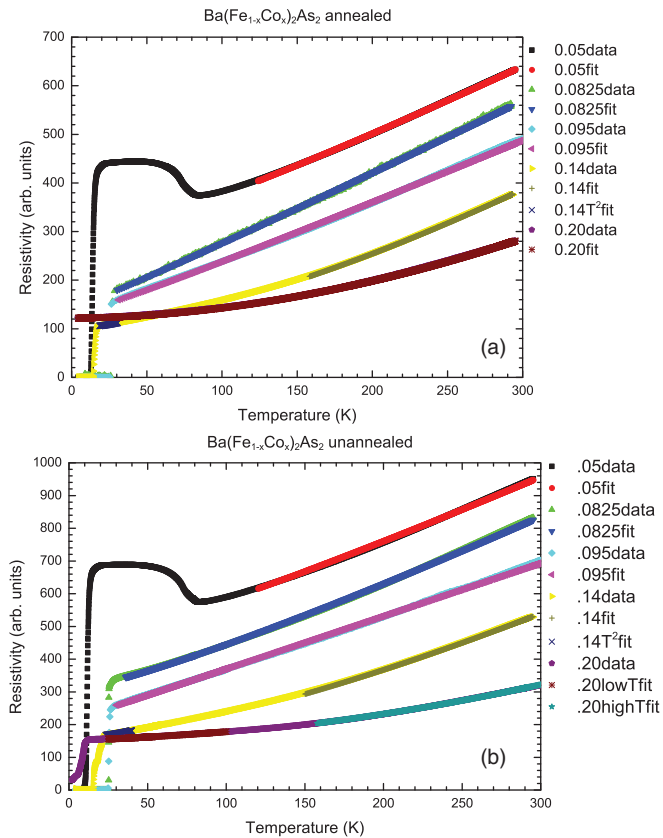


FIG. 6. (Color online) (a) ρ data (arbitrary units to better distinguish the various curves) for annealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ $x = 0.05$ (black), 0.0825 (green), 0.095 (light blue), 0.14 (yellow) and 0.20 (purple.) Clearly, in the neighborhood of the composition, x_{opt} , for the peak T_c (see Fig. 5), the temperature dependence of the resistivity remains fairly constant at about T^1 over a wide range of temperature. To get an idea of the possible practical error bar for these fits, if the fit for $x = 0.095$ is restricted to 70–220 K rather than the 30–300 K shown here, $\alpha = 1.055$ rather than 1.069 for the whole range. The temperature ranges of the various fits (red for $x = 0.05$, blue for $x = 0.0825$, pink for $x = 0.095$, blue/green for low and high temperatures respectively for $x = 0.14$, and brown for $x = 0.20$) are discussed in the text. (b) ρ data for unannealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, $x = 0.05$ (black), 0.0825 (green), 0.095 (light blue), 0.14 (yellow), and 0.20 (purple), i.e., the same compositions as for the annealed data in (a) for comparison. The temperature ranges of the various fits are discussed in the text.

presumably at some temperature away from the QCP composition the approximately T -linear behavior of ρ would change over to a Fermi-liquid, T^2 behavior. Below x_{opt} , however, the presence of the structural and the magnetic transitions (which are more field independent than the superconductivity) could still obscure the underlying temperature dependence at lower temperatures. For $x > x_{\text{opt}}$, T^2 behavior in ρ should be seen at low temperatures if a sufficiently large magnetic field were applied.

IV. SUMMARY AND CONCLUSIONS

Near the peak in T_c measurements down to T_c of the resistivity for annealed and unannealed single crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ shows that $\rho = \rho_0 + AT^\alpha$, where α is close to 1.0. This is consistent with the existence of a QCP underneath the superconducting dome close to the optimal (maximum T_c) doping concentration of Co. It is interesting to note that the width of composition where the linear-with- T resistivity behavior is seen is approximately 5% (from $x = 0.05$ to 0.095), or almost a quarter of the whole superconducting dome width. Presumably,¹⁶ if the temperature dependence of ρ could be probed to lower temperatures with T_c suppressed by applied magnetic field, this width would narrow in composition range as the QCP was approached. Upon cooling, before $\rho \rightarrow T^2$, a crossover regime, with an intermediate range of temperature dependence between T^1 and T^2 , should¹⁶ be found for $x \neq x_{\text{QCP}}$. Field suppression of T_c with available fields (≈ 27 T) could be achieved for $x \geq x_{\text{opt}}$. For $x < x_{\text{opt}}$, the presence of structure and magnetic transitions above T_c would—in available dc fields—prevent determination of α to significantly lower temperatures. Finally, annealing self-flux-grown single crystals of $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ has been shown to raise T_c^{onset} above 27 K, an increase of 5 K above the initial work⁸ on unannealed $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ and an increase of 1 K over a previous annealing work¹⁸ on $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$.

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