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Pressure effects on the electron-doped high T_c superconductor $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$

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Abstract

Application of pressure or electron doping through Co substitution into Fe sites transforms the itinerant antiferromagnet BaFe_2As_2 into a superconductor with T_c exceeding 20 K. We carried out systematic transport measurements of $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ superconductors in pressures up to 2.5 GPa, and elucidated the interplay between the effects of electron doping and pressure. For the underdoped sample with nominal composition $x = 0.08$, application of pressure strongly suppresses a magnetic instability while enhancing T_c by nearly a factor of two from 11 to 21 K. In contrast, the optimally doped $x = 0.20$ sample shows very little enhancement of $T_c = 22$ K under applied pressure. Our results strongly suggest that the proximity to a magnetic instability is the key to the mechanism of superconductivity in iron-pnictides.

(Some figures in this article are in colour only in the electronic version)

The recent discovery of superconductivity at 26 K in the carrier-doped quaternary iron oxy-pnictide LaFeAsO attracted tremendous interest among the scientific community around the world [1]. More recently, the ternary iron-pnictide family RFe_2As_2 ($\text{R} = \text{Ba}, \text{Sr}, \text{Ca}$) with the tetragonal ThCr_2Si_2 structure also has been shown to become a high T_c superconductor upon doping [2]. The common structural building blocks of the LaFeAsO and RFe_2As_2 families are the square-lattice Fe sheets hybridized with As layers, hence there is no doubt that superconductivity takes place in the FeAs layers. However, the mechanism of superconductivity is highly controversial. The RFe_2As_2 family is a promising model system for a detailed investigation of the physical properties of the new iron-based high T_c superconductors, because large single crystals are available. BaFe_2As_2 is an itinerant antiferromagnet, and exhibits simultaneous structural (tetragonal to orthorhombic) and magnetic phase transitions at $T_{\text{SDW}} \sim 140$ K [3, 4]. Holes doped into the Fe sheets through substitution of K^+ ions into the Ba^{2+} sites suppress

the magnetic instability, and induce superconductivity with T_c as high as 38 K in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with $x = 0.45$ [2]. This strategy of transforming an antiferromagnetic parent phase into a superconductor by doping from the $\text{Ba}_{1-x}\text{K}_x$ charge-reservoir layers is analogous to the recipe used in high T_c copper-oxide superconductors.

However, two recent discoveries show that there are fundamental differences between cuprates and iron-pnictides. First, one can induce superconductivity in the Fe sheets by substituting Co [5–7] or Ni [8] atoms directly into the Fe sites. Co(Ni) atoms have one (two) more electron(s) than Fe atoms, and they donate these extra electrons to FeAs layers as superconducting carriers. This phenomenon is in remarkable contrast with the case of cuprates; doping non-magnetic Zn^{2+} ions with one extra electron into Cu sites induces local magnetic moments and destroys superconductivity [9, 10]. Our ^{59}Co and ^{75}As NMR measurements in $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ showed no evidence for induced magnetic moments [11]. Second, one can also suppress the itinerant antiferromagnetism

of the parent phase RFe_2As_2 and induce superconductivity without doping by applying pressure [12–14]. In cuprates, application of pressure does not transform the Mott insulating state into a metallic state.

To date, the mechanism of carrier *or* pressure-induced superconductivity in RFe_2As_2 is not understood well, calling for further investigations. In this report, we shed new light on this fascinating problem by exploring the interplay between carrier doping *and* pressures through systematic measurements of the electrical resistivity ρ in Co-doped $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ single crystals in pressures up to $P = 2.5$ GPa. We choose two nominal concentrations for the present study: the underdoped $\text{BaFe}_{1.92}\text{Co}_{0.08}\text{As}_2$ ($T_c \sim 11$ K) and optimally doped $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ ($T_c \sim 22$ K). Notice that the Co dopants replace nominally 4% and 10% of Fe sites in $x = 0.08$ and $x = 0.20$ samples, respectively. We will demonstrate that application of pressure drives $x = 0.08$ away from a magnetic instability, while enhancing T_c by nearly a factor of two. The highest $T_c \sim 21$ K observed for $x = 0.08$ in 2.5 GPa is almost as high as $T_c \sim 22$ K of $x = 0.20$, and indicates that the underdoped Fe sheets are as susceptible to pressures as undoped RFe_2As_2 . In contrast, we show that T_c of the optimally doped $x = 0.20$ barely changes with pressure. Given that application of pressure also suppresses the magnetic instability in $x = 0$ [15] and $x = 0.08$, we argue that the proximity to a magnetic critical point plays a crucial role in the superconducting mechanism.

Single crystals of Co-doped BaFe_2As_2 were grown out of FeAs flux, and characterized by x-ray diffraction and electron-probe micro-analysis. Further details can be found in [6]. We cut small piece of crystals with typical dimensions $1 \text{ mm} \times 1 \text{ mm} \times 0.15 \text{ mm}$ from a larger piece for the electrical resistivity measurements in applied pressure. Sample contacts were made using Epotek silver epoxy for conventional four-probe ac-measurements. We employed a compact double-layered hydrostatic cylindrical clamp cell with a BeCu outer jacket and a NiCrAl alloy inner core to achieve pressures up to 2.5 GPa. Daphene oil 7373 and 99.99% purity Sn were used as a pressure transmitting medium and a pressure calibrating gauge, respectively. In order to achieve high precision, we used a highly flexible home-built ac-measurement system. We stabilized the temperature of the high pressure cell for every data point, instead of continuously sweeping the temperature. We confirmed the consistency of our results by conducting both cooling and warming measurements for every run.

We begin our discussions with the transport properties of optimally electron-doped $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$. In figure 1, we present the temperature dependence of the in-plane (*ab*-plane) resistivity ρ_{ab} measured for $x = 0.20$ at various hydrostatic pressures. ρ_{ab} decreases monotonically with temperature, and we can fit the temperature dependence with a simple power law, $\rho_{ab} = A + BT^n$, with $n = 1.33 \pm 0.05$ in a broad range of temperatures from ~ 300 K down to ~ 100 K. Below ~ 100 K, the resistivity shows T -linear behavior $\rho_{ab} \sim T$ ($n = 1$) down to T_c [6]. Our recent NMR measurements in the same crystal showed that spin susceptibility χ_{spin} also undergoes a crossover between two different regimes across ~ 100 K; χ_{spin} decreases monotonically with a pseudo-gap $\Delta_{\text{PG}}/k_B \sim$

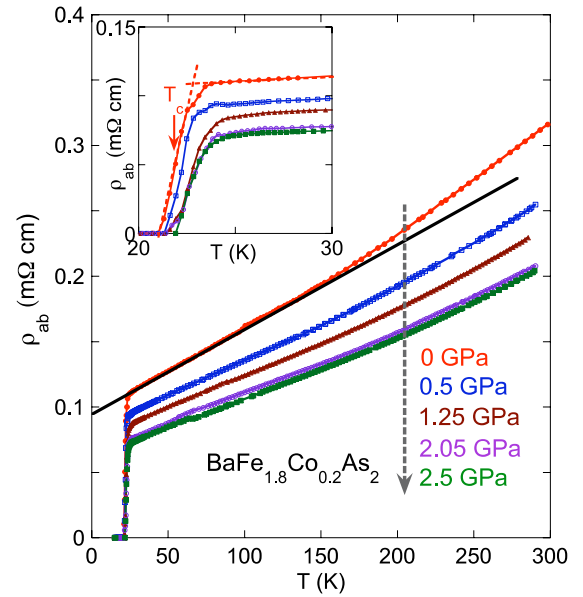


Figure 1. The temperature dependence of the in-plane resistivity ρ_{ab} of a single crystal of $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ at various pressures. A straight line is drawn through the ambient pressure data to show the T -linear behavior up to ~ 100 K. The inset shows the expanded view near T_c . We define T_c as the midpoint of the transition.

560 ± 150 K, then almost levels off below ~ 100 K [11]. This means that low energy spin excitations are abundant in the high temperature regime above ~ 100 K, and the power $n = 1.33 \cong 4/3$ may be associated with the scattering of electrons by these excitations. We emphasize that although the temperature independence of χ_{spin} alone might suggest that $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ is a Fermi-liquid system below ~ 100 K, the T -linear behavior of ρ indicates the contrary. We recall that Fermi-liquid systems would satisfy $\rho_{ab} \sim T^2$. Instead, the T -linear behavior is the benchmark of strongly correlated phenomena in copper-oxide high T_c cuprates. For example, optimally doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ exhibits $\rho_{ab} \sim T$ for a wide range of temperatures from T_c to as high as ~ 1000 K [16].

Upon applying hydrostatic pressure, the magnitude of ρ_{ab} in $x = 0.20$ decreases substantially in the entire temperature range above T_c , but the temperature dependence shows very little change. To illustrate this point, in figure 2 we plot ρ_{ab} measured for various pressure values by normalizing the magnitude at 290 K to the result obtained for 2.5 GPa. All the data collapse on top of each other except near T_c . The pressure effect on T_c is also weak. T_c increases only by ~ 1 K from $T_c \sim 22$ K at $P = 0$ to $T_c \sim 23$ K at $P = 1.25$ GPa, then almost saturates, as summarized in figure 3. The pressure coefficient below 1.25 GPa, $dT_c/dP \sim 0.65 \text{ K GPa}^{-1}$ is by an order of magnitude smaller than that of the undoped antiferromagnetic phase BaFe_2As_2 [14]. In the latter case, $T_c(x = 0) = 0$ K increases quickly to 29 K in a narrower pressure range from 2.5 to 4 GPa as shown in figure 3. These contrasting results indicate that once the magnetic instability is completely suppressed by 10% Co substitution into Fe sites, application of pressure has little effect on T_c in the optimally electron-doped system. It remains to be seen whether applying higher pressures than 2.5 GPa

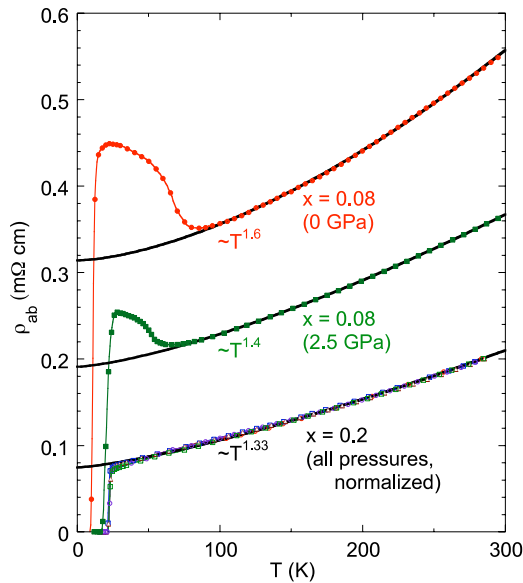


Figure 2. Representative fits of the resistivity data above ~ 100 K with a power law, $\rho_{ab} = A + BT^n$, where A and B are constants. For the optimally doped $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$, we normalized all the resistivity data at 290 K to the result obtained at 2.5 GPa. For clarity, we show one data point of ρ_{ab} for every four temperatures.

would further enhance T_c , or even suppress T_c to show a ‘dome shape’ in the P versus T_c phase diagram. However, a naive extrapolation of T_c in $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ to higher pressure values does not seem to achieve as high a $T_c \sim 29$ K as in the undoped parent phase. One obvious possibility is that the disorder induced by Co substitution into Fe limits the highest T_c , but this scenario seems unlikely. The normal state residual resistivity at 2.5 GPa estimated from the linear extrapolation to $T = 0$ is $\rho_{ab}(T \rightarrow 0) \sim 0.06$ m Ω cm, and this value is about the same as that of undoped BaFe_2As_2 at 13 GPa [15]. It is also interesting to notice that the sign of the pressure coefficient dT_c/dP is different between the present case of electron-doped $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ and the hole-doped $\text{Ba}_{0.55}\text{K}_{0.45}\text{Fe}_2\text{As}_2$. The latter has a negative coefficient, $dT_c/dP = -1.5$ K GPa $^{-1}$ [17]. This may be an indication that the Fermi energy E_F of the undoped BaFe_2As_2 is located near a singularity of the density of states, and electron and hole-doping place E_F on different slopes.

Next, we turn our attention to the pressure effects on the underdoped, Co 4% doped $\text{BaFe}_{1.92}\text{Co}_{0.08}\text{As}_2$. In figure 4, we show ρ_{ab} measured for $x = 0.08$ at various hydrostatic pressures. The temperature dependence above ~ 100 K is qualitatively similar to that of the optimally doped $x = 0.2$. An analogous power-law fit to $\rho_{ab} = A + BT^n$ yields a somewhat larger value of the power $n = 1.60 \pm 0.03$ at ambient pressure, as shown in figure 2. Unlike the optimally doped $x = 0.20$, a monotonic decrease of ρ_{ab} from ~ 300 K down to ~ 95 K is followed by a step-like increase near ~ 70 K. The sharp dip centered around 66 K in the derivative presented in figure 2(b), $d\rho_{ab}/dT$, hints at the presence of a phase transition. To clarify the origin of the anomaly, we also carried out NMR measurements on the same batch of crystals [18]. The dramatic NMR line broadening and a divergence of the nuclear spin-lattice relaxation rate $1/T_1$ below $\lesssim 70$ K indicate that this

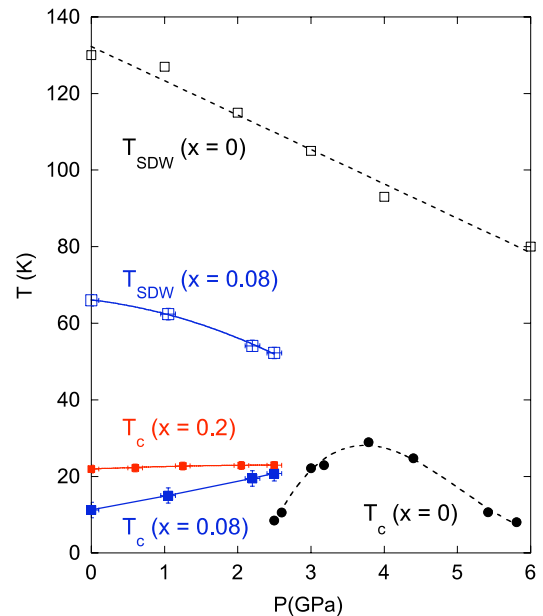


Figure 3. A summary of the P - T phase diagram of magnetism and superconductivity in $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$ ($x = 0.20, 0.08$, and 0). The results of T_c and T_{SDW} for $x = 0$ are by Alireza *et al* [14] and Fukazawa *et al* [15], respectively. All lines are a guide for the eyes.

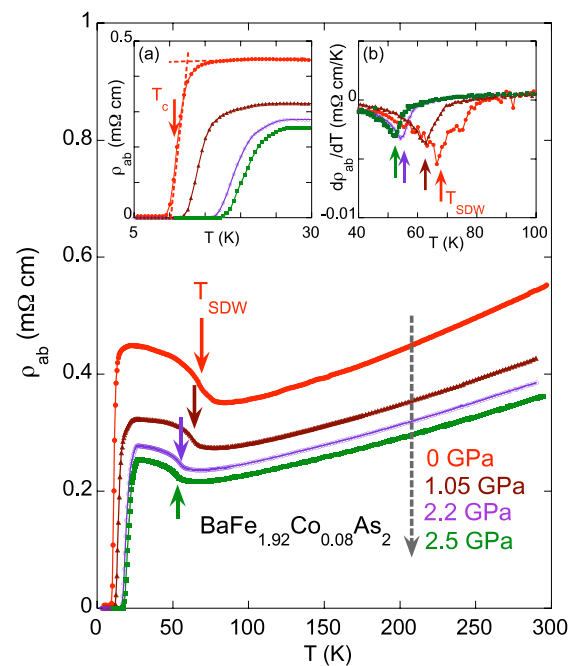


Figure 4. Main panel: the temperature dependence of the in-plane resistivity ρ_{ab} of a single crystal of $\text{BaFe}_{1.92}\text{Co}_{0.08}\text{As}_2$ at various pressures. The vertical arrows show the magnetic ordering temperature at various pressures. Inset: (a) the expanded view near T_c , (b) the derivative $d\rho_{ab}/dT$.

anomaly is caused by a magnetic instability. In what follows, we tentatively represent the magnetic anomaly temperature using the same symbol T_{SDW} commonly used for the undoped BaFe_2As_2 , but we need to keep in mind that the nature of the magnetic instability in the electron-doped $x = 0.08$ may

be somewhat different. In particular, whether a structural phase transition accompanies the magnetic instability at or near 70 K requires further investigation. Regardless, it is worth pointing out that signatures of the critical slowing down observed above 70 K in our NMR data indicate that this magnetic instability is not strongly first order. Since we do not observe any sharp paramagnetic NMR signals below T_{SDW} , we can also conclude that the magnetic instability at T_{SDW} affects the entire $\text{Fe}_{1.92}\text{Co}_{0.08}\text{As}_2$ layers. In passing, a similar step-like resistivity increase is also reported for Co-doped $\text{LaFe}_{1-x}\text{Co}_x\text{AsO}$ [5], $\text{SrFe}_{2-x}\text{Co}_x\text{As}_2$ ($x = 0.1$ and 0.15) [7], and Ni-doped $\text{BaFe}_{1.95}\text{Ni}_{0.05}\text{As}_2$ [8].

With decreasing temperature, $x = 0.08$ also exhibits a superconducting transition to a zero resistivity state below $T_c \sim 11$ K. Given our NMR finding that the magnetic instability affects all sites in the $\text{Fe}_{1.92}\text{Co}_{0.08}\text{As}_2$ layers, we rule out the possibility that this superconducting transition is caused by macroscopic inhomogeneity in Co concentrations. Since the magnetic instability precedes the superconducting transition, the superconducting state should be considered granular, in analogy with the case of underdoped high T_c cuprates. We may naturally understand the apparent coexistence of T_{SDW} and T_c based on the following simple argument. The key point is that the superconducting coherence length $\xi \sim 3.5$ nm [19] is very short in iron-pnictide superconductors. Assume that each Co atom donates an electron to nearby Fe sites, destroys magnetism locally, and creates a superconducting island. As a ballpark figure, one may assume that the diameter of the island is roughly equal to the in-plane superconducting coherence length, ξ , and involves more than ten Fe sites. Then the Co doping level of 4% is more than enough to achieve the classical percolation limit of 50% for two-dimensions, and hence zero resistance. This situation of the ‘reverse Swiss cheese model’ is the opposite to the so-called ‘Swiss cheese model’. The latter describes the destruction of superconductivity in Zn-doped high T_c cuprates [10].

We notice three striking features in the pressure effects in figure 4. First, with applying pressure, T_{SDW} is markedly suppressed from $T_{\text{SDW}} \sim 66$ K to ~ 52 K at 2.5 GPa. This strong pressure effect on T_{SDW} is comparable to the case of the undoped BaFe_2As_2 . In the latter, $T_{\text{SDW}} \sim 130$ K is suppressed to ~ 110 K at 2.5 GPa [15]; see the phase diagram in figure 3 for comparison. Second, as the magnetic instability is suppressed, the temperature dependence of ρ_{ab} becomes increasingly similar to that of $x = 0.2$. We show the representative fits of ρ_{ab} by a power law in figure 2, and summarize the pressure dependence of the power n in figure 5. The power n gradually decreases with increasing pressure from $n = 1.6$ at 0 GPa to 1.4 at 2.5 GPa. Figure 5 suggests that the power n for $x = 0.08$ approaches $n = 1.33$ in higher pressures, which is the value observed for the optimally doped $x = 0.20$ sample. It is tempting to interpret this as an indication that $n = 4/3$ is the universal value in the metallic state above ~ 100 K of Co-doped superconducting $\text{BaFe}_{2-x}\text{Co}_x\text{As}_2$. We caution, however, that a similar power-law fit of the resistivity data reported by Fukazawa *et al* [15] for undoped BaFe_2As_2 at 13 GPa yields a smaller value, $n \sim 1.1$. Third, application of pressure results in a very strong enhancement of T_c from 11 to

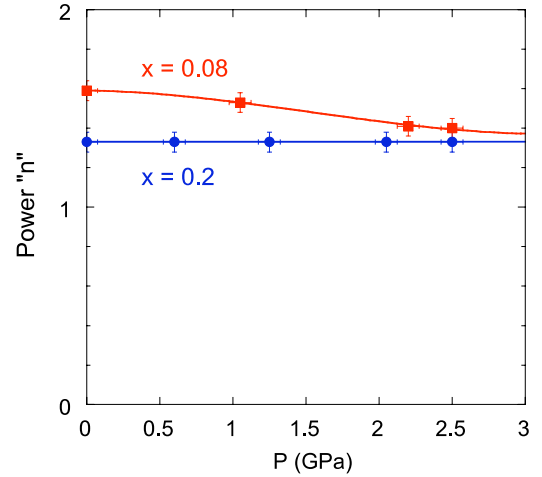


Figure 5. The pressure dependence of the power n for the power-law fit of $\rho_{ab} = A + BT^n$ above ~ 100 K. The straight line across the data points for $x = 0.2$ underscores the pressure independence of n . The solid curve for the $x = 0.08$ data is a guide for the eyes.

21 K in 2.5 GPa. That is, one can enhance T_c by nearly a factor of two. We emphasize that the observed pressure coefficient, $dT_c/dP \sim 4$ K GPa^{-1} is quite sizable, and is nearly an order of magnitude larger than 0.65 K GPa^{-1} of the optimally doped $x = 0.20$ sample. Equally interesting is our finding that T_c of the $x = 0.08$ sample shows no sign of saturation up to 2.5 GPa, unlike the case of $x = 0.20$. In fact, a naive extrapolation of T_c in figure 3 shows that T_c of $x = 0.08$ may exceed that of $x = 0.20$ and approaches the maximum T_c of $x = 0$. We recall that many earlier studies of pressure effects on T_c in undoped or hole-doped iron-pnictides showed either a dome-like shape in the T_c versus P phase diagram, or a linear decrease of T_c at elevated pressures [12–14, 17, 20–24].

To summarize, we demonstrated contrasting behaviors of pressure effects on superconductivity between optimally electron-doped $\text{BaFe}_{1.8}\text{Co}_{0.2}\text{As}_2$ and underdoped $\text{BaFe}_{1.92}\text{Co}_{0.08}\text{As}_2$. We showed that once the magnetic instability is completely suppressed by electron doping, application of high pressures has very little effect on superconductivity and the qualitative behavior of the normal state resistivity. On the other hand, the suppression of a magnetic instability with pressure results in a large enhancement of T_c by nearly a factor of two. This finding strongly suggests that magnetism and superconductivity are strongly correlated. Needless to say, it does not necessarily mean that the glue of the superconducting Cooper pairs is magnetic fluctuations. Instead, one can certainly view figure 3 as evidence for competition between magnetism and superconductivity. In any event, our new results on underdoped $x = 0.08$ establish a link between superconductivity and magnetism, and show that fine tuning of magnetic fluctuations with pressure is as effective as doping more carriers by additional Co doping.

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