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Pressure tuned superconductivity and normal state behavior in $Ba(Fe_{0.943}Co_{0.057})_2As_2$ 1 near the antiferromagnetic boundary 2

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Superconductivity in iron pnictides is unconventional and pairing may be mediated by magnetic fluctuations in the Fe-sublattice. Pressure is a clean method to explore superconductivity in iron based superconductors by tuning the ground state continuously without introducing disorder. Here we present a systematic high pressure transport study in $Ba(Fe_{1-x}Co_x)_2As_2$ single crystals with x = 0.057, which is near the antiferromagnetic instability. Resistivity $\rho = \rho_0 + AT^n$ was studied under applied pressure up to 7.90 GPa. The parameter n approaches a minimum value of $n \approx 1$ at a critical pressure $P_c = 3.65$ GPa. Near P_c , the superconducting transition temperature T_c reaches a maximum value of 25.8 K. In addition, the superconducting diamagnetism at 2 K shows a sudden change around the same critical pressure. These results may be associated with a possible quantum critical point hidden inside the superconducting dome, near optimum T_c .

Introduction Α.

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Unconventional superconductivity observed in iron-16 based superconductors is in close proximity to an 17 antiferromagnetically ordered state.¹ Superconductivity 18 emerges as antiferromagnetism is suppressed by pressure 19 or chemical doping, $^{2-4}$ and the superconducting critical 20 temperature T_c forms a dome shape. In the Ni-, Co-, P-, 21 Rh- and Pd-doped BaFe₂As₂ system, the antiferromag-22 ²³ netic phase boundary crosses the superconducting dome $_{24}$ near optimal doping.^{2,5–10} Hence, there is a region in the phase diagram where antiferromagnetism and super-25 conductivity coexist. Neutron scattering measurements 26 on $Ba(Fe_{1-x}Ni_x)_2As_2$ observed short range incommen-27 surate antiferromagnetic order coexisting with supercon-28 ²⁹ ductivity near optimal doping, where the first-order-like ³⁰ antiferromagnetism-to-superconductivity transition suggests the absence of a quantum critical point (QCP).⁶ 31 Notably, it has been reported that the magnetic pene-32 tration depth in $BaFe_2(As_{1-x}P_x)_2$ shows a sharp peak 33 34 at optimal doping, possibly due to quantum fluctuations 35 associated with a QCP.⁷

In particular for $Ba(Fe_{1-x}Co_x)_2As_2$, the physical prop-36 erties have been widely studied close to optimal dop-37 ³⁸ ing and the antiferromagnetic phase boundary. Neutron diffraction measurements indicate Co doping rapidly sup-40 presses antiferromagnetism, with the antiferromagnetic 41 order vanishing at $x \approx 0.055$.¹¹ For x = 0.06, it is sug-⁴² gested that superconductivity coexists with a spin den-⁴³ sity wave (SDW).¹² For thin films of $Ba(Fe_{1-x}Co_x)_2As_2$, ⁷⁵ superconducting diamagnetism at 2 K shows a sudden ⁴⁴ the exponent n in the temperature dependence of the $_{76}$ change at a critical pressure of P = 3.5 GPa, in accor-45 resistivity is minimum namely, close to unity at $x \approx \pi$ dance with changes in resistivity. These results may be

 $_{46}$ 0.05 and $x \approx 0.07$ for MgO and CaF₂ substrate, re-47 spectively, which may be associated with an antifer-⁴⁸ romagnetic QCP.¹³ Furthermore, a sign change in the ⁴⁹ electronic-magnetic Gruneisen parameter is observed for $_{50} x = 0.055$ and x = 0.065, consistent with the expected ⁵¹ behavior at a QCP.¹⁴ In addition, a critical concen-52 tration of $x_c \approx 0.065$ is determined from the analy- $_{53}$ sis of $1/T_1T$ in NMR measurements.¹⁵ Considerably en-⁵⁴ hanced flux-flow resistivity ρ_{ff} was also detected for x =55 0.06, perhaps due to enhancement of spin fluctuations ⁵⁶ near QCP.¹⁶ Thermopower(S) measurements reported $_{57}$ a maximum S/T in proximity to the commensurate-58 to-incommensurate SDW transition for $x \approx 0.05$, close $_{\rm 59}$ to the highest superconducting $T_c.^{17}$ However, the su-⁶⁰ perconducting magnetization appears nearly unchanged ₆₁ across the dome in Ba(Fe_{1-x}Co_x)₂As₂.²

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Despite extensive studies in $Ba(Fe_{1-x}Co_x)_2As_2$ close 62 ⁶³ to optimal doping, there had been no systematic study ⁶⁴ on how the normal state evolves across the antiferromag-⁶⁵ netic phase boundary. Here we probe the phase diagram ⁶⁶ close to the antiferromagnetic boundary through mea-67 surements of resistivity and magnetization by tuning the 68 applied pressure in a sample with x = 0.057. Normal 69 state resistivity changes from non-Fermi liquid to Fermi 70 liquid with increasing pressure. It shows almost linear ⁷¹ temperature dependence at a critical pressure of P = 3.65 $_{72}$ GPa, where T_c is maximum. In addition, the residual re-⁷³ sistivity ρ_0 and the resistivity at T_c all change around the ⁷⁴ same critical pressure. From the magnetization data, the

⁷⁸ due to a possible QCP at optimum T_c , similar to the case ⁷⁹ of BaFe₂(As_{1-x}P_x)₂⁷ and hole doped cuprates.¹⁸

> **Experimental Details** в.

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Single crystals of $Ba(Fe_{1-x}Co_x)_2As_2$ with x = 0.05781 were synthesized by a flux method.² Electrical resistivity 82 was measured using a Quantum Design Physical Prop-83 erty Measurement System (PPMS). The electronic trans-84 port properties were measured using four-probe electri-85 cal conductivity in a diamond anvil cell made of CuBe 86 alloy. The diamond culet was 800 μ m in diameter. Mag-87 ⁸⁸ netic measurements were performed in a superconduct-⁸⁹ ing quantum interference device (SQUID magnetome-⁹⁰ ter). Pressure was applied using a diamond anvil cell ⁹¹ made of CuBe alloy with the diamond anvil culet of $_{92}$ 500 μ m. In both cases, Daphne oil 7373 was used as ⁹³ a pressure-transmitting medium. Above its solidification at 2.2 GPa,¹⁹ non-hydrostaticity may develop and lead 94 to inhomogeneous pressure distribution inside the sam-95 ple chamber. Pressure was calibrated by using the ruby 96 ⁹⁷ fluorescence shift at room temperature. For resistivity, ⁹⁸ the superconducting transition temperature T_c is defined ⁹⁹ as the temperature for the appearance of zero resistance 100 state (Fig. 1(b)); for magnetization, T_c is the tempera-¹⁰¹ ture we observe a sharp drop in M (inset to Fig. 3(a)).

Results and Discussion С.

103 ¹⁰⁵ at different applied pressures namely, P = 0, 1.25, 2.69, ¹⁴⁰ pressure, reaching 2 at P = 7.90 GPa. 106 3.65, 5.26, 6.87 and 7.90 GPa. The resistivity curve for 141 $_{107}$ P = 7.90 GPa was shifted downward by 0.05 m Ω cm for $_{142}$ at $P_c \approx 3.5$ GPa. This is similar to the heavy fermion ¹⁰⁸ clarity. Note that the large decrease of ρ_{300K} with pres-¹⁴³ superconductor CeCoIn₅, where ρ_0 and n change at $P_c =$ ¹⁰⁹ sure (inset to Fig. 1(b)) is very similar to the changes ¹⁴⁴ 1.6 GPa.²³ We ascribe the decrease in ρ_0 with increas-¹¹⁰ occurring with Co doping in Ba(Fe_{1-x}Co_x)₂As₂.²⁰ By ¹⁴⁵ ing pressure to a change in inelastic scattering.²³ The 111 comparing the data we find that an increase in doping 146 pressure dependence of ρ at T_c shows a change in slope ¹¹² level by 1% is roughly equivalent to 1.2 GPa of pressure, ¹⁴⁷ at P_c , similar to the behavior of the normal state rewhich is comparable with previous report.¹⁴ 113

114 ¹¹⁵ ing temperature but shows an upturn just before en- ¹⁵⁰ served in BaFe₂As₂ with Co doping, where the exponent ¹¹⁷ to the structural (T_s) and SDW (T_{sdw}) phase transi-¹⁵² In BaFe₂(As_{1-x}P_x)₂, non-Fermi liquid behavior with n ¹¹⁸ tion, in agreement with earlier studies in underdoped ¹⁵³ close to unity is found around optimal doping x = 0.3, ¹¹⁹ Ba(Fe_{1-x}Co_x)₂As₂.² Both T_s and T_{sdw} can be estimated ¹⁵⁴ with T_c maximum at the QCP.⁷ Similarly, linear resis- $_{120}$ from the first derivative of the temperature dependent $_{155}$ tivity was observed for Ba(Fe_{1-x}Ni_x)₂As₂ with x = 0.05¹²¹ resistivity curve (see inset to Fig. 1(a)).⁵ With further ¹⁵⁶ for which T_c is maximum at a magnetic QCP.²⁴ 122 increase in pressure, the upturn vanishes suggesting sup- 157 The zero field cooled (ZFC) magnetization was mea-¹²³ pression of the T_s and T_{sdw} . Similar changes with pres-¹⁵⁸ sured in a run with increasing pressure for P = 0.6, 1.2,¹²⁴ sure has been reported for Ba(Fe_{1-x}Co_x)₂As₂.²⁰⁻²² The ¹⁵⁹ 2.0, 2.7, 3.5, 4.3, 5.6, 6.4 GPa. The resultant data are $_{125}$ zero resistance transition temperature T_c (solid squares $_{160}$ plotted in Fig. 3(a). Since the sample used in the pres- $_{126}$ in Fig. 2(a)) varies non-monotonically with increasing $_{161}$ sure cell is too small to measure its mass, we show mag- $_{127}$ pressure. For P = 6.87 GPa and above, we observe a $_{162}$ netization data in emu. Another piece of sample is used 128 finite resistivity down to the lowest measured tempera- 163 to obtain the ambient pressure magnetization data (as ¹²⁹ ture. A similar dome shaped variation in T_c is observed ¹⁶⁴ shown in the inset to Fig. 3(a)) to determine T_c at P = 0.



FIG. 1. (a)(b) Temperature T dependence of resistivity ρ under applied pressure P = 1.25, 2.69, 3.65, 5.26, 6.87 and 7.90 GPa. Symbols represent data and solid lines are fits using $\rho = \rho_0 + AT^n$. Note that the resistivity curve for P =7.90 GPa was shifted downward by 0.05 m Ω cm for clarity. Inset to (a) shows the temperature dependence of $d\rho/dT$ at ambient pressure. Inset to (b) shows pressure dependence of resistivity at 300 K, ρ_{300K} .

¹³⁰ in Ba(Fe_{1-x}Co_x)₂As₂ with Co doping.^{21,22}

We fit the resistivity curve under pressure using $\rho =$ 131 ¹³² $\rho_0 + AT^n$ (with fitting parameters ρ_0 , n and A) as shown 133 in Fig. 1, where the symbols represent data points and $_{134}$ the solid lines are fits. The pressure dependence of T_c , $_{135}$ ρ_0 , ρ at T_c and n obtained from Fig. 1 are summarized ¹³⁶ in Fig. 2(a)-(c), respectively. Resistivity can be tuned ¹³⁷ with pressure from a non-Fermi liquid (NFL)(n = 1) to Figure 1 shows the temperature dependence of resis- 138 Fermi liquid (FL) (n = 2) behavior. Note that n = 1.1tivity for $Ba(Fe_{1-x}Co_x)_2As_2$ with x = 0.057 measured ¹³⁹ at P = 3.65 GPa and increases with further increase in

Interestingly, all parameters in Fig. 2 show a change hich is comparable with previous report.¹⁴ At low pressures, resistivity decreases with decreas- 148 sistivity ρ_n at T_c around optimal doping in chemically 148 sistivity ρ_n at T_c around optimal doping in chemically 149 tuned BaFe₂As₂.¹² Similar change in n was also obtering the superconducting state. This upturn is due 151 *n* is minimum namely, close to 1 at optimal doping.¹³



FIG. 2. Pressure dependence of (a) superconducting transition temperature T_c , (b) resistivity at the superconducting onset temperature $\rho(T = T_c)$ and residual resistivity ρ_0 , (c) exponent n.



FIG. 3. (a) Temperature dependence of magnetization measured at P = 0.6, 1.2, 2, 2.7, 3.5, 4.3, 5.6, 6.4 GPa with increasing pressure and 3.6, 1.4 GPa with decreasing pressure, in an applied magnetic field of 10 Oe. The inset shows magnetization data at ambient pressure for both zero field cooled (ZFC) and field cooled (FC) runs. (b) Pressure dependence of the superconducting transition temperature (squares) and the diamagnetic signal M(2K) (circles). Solid and open symbols depict data for experiments performed with increasing 219 and decreasing pressure, respectively.

166 167 2(a)). 168

169 $_{170}$ magnetization at T = 2 K, M(2K) in Fig. 3(b). $_{228}$ tween the two. T_c reaches a maximum at a critical pres-171 172 173 174 175 magnetization since the superconducting transitions are 233 is accompanied by the above mentioned change in the 176 broad and incomplete at high pressures and upon releas- 234 superconducting diamagnetism. Together, these exper- $_{177}$ ing the pressure. Nevertheless, it will give some hint to $_{235}$ imental findings suggest the presence of a QCP at P_c , $_{\rm 178}$ further understand the behavior of the superconducting $_{\rm 236}$ where T_c is maximum. $_{179}$ state evolving across the antiferromagnetic phase bound- $_{237}$ Earlier NMR measurements in Ba(Fe_{1-x}Co_x)₂As₂ re-

¹⁸¹ lowed by a sudden suppression at $P_c = 3.5$ GPa, then ¹⁸² becoming negligible at high pressures. A similar pres-¹⁸³ sure induced suppression in the superconducting volume was observed in the parent compound of BaFe₂As₂ and 184 $SrFe_2As_2$, where a dome like behavior of the pressure ¹⁸⁶ dependent superconducting volume is reported.²⁸ Also, ¹⁸⁷ for $Sr(Fe_{1-x}Ni_x)_2As_2$ and $Ca_{1-x}LaFe_2(As_{1-y}P_y)_2$, the 188 superconducting volume shows a dome behavior with ¹⁸⁹ doping.^{25,27} In addition, a sudden suppression in the su-¹⁹⁰ perconducting volume was observed in high- T_c cuprate ¹⁹¹ La_{2-x}Sr_xCuO₄ at a critical doping level of around x =¹⁹² 0.21,²⁹ which is close to a QCP.³⁰ Thus, the suppression ¹⁹³ of the superconducting volume fraction above the critical pressure observed in present work could reflect a phase 194 195 transition at P_c .

Note that in chemically doped (Co, Rh, Ni) BaFe₂As₂ 196 ¹⁹⁷ at ambient pressure, there is no change in magnetization ¹⁹⁸ across the dome.^{2,5,9} Nevertheless, this difference may ¹⁹⁹ be due to different role played by pressure and chemical tuning. In fact, there is a pressure tuned QCP in pure 200 $CeCoIn_5$,²³ while, there is no signatures of quantum crit-201 ical behavior in Cd-doped $CeCoIn_5$, due to the effect of 202 disorder near a zero temperature magnetic instability.³¹ 203 This suggests that tuning a system with disorder to a 204 presumed magnetic QCP does not necessitate a quantum 205 critical response.³¹ 206

We also measured two magnetization curves under de-201 208 compression, namely, for P = 3.6 and 1.4 GPa (see Fig. $_{209}$ 3(a)). Interestingly, the superconducting volume fraction $_{210}$ is about the same as compression data, however, the T_c $_{211}$ values are not fully recovered. The different T_c between ²¹² compression and decompression is previously reported in $_{213}$ In₂Se₃, which is intrinsic, as a result of changes in phonon 214 and variation of carrier concentration combined in the ²¹⁵ pressure quench.³² Further measurements are needed to $_{216}$ confirm if there is indeed a suppressed T_c behavior in $_{217}$ Ba(Fe_{1-x}Co_x)₂As₂ during decompression, which is be-²¹⁸ youd the scope of this work.

Figure 4 shows the temperature vs. pressure (T - P)²²⁰ phase diagram of Ba(Fe_{1-x}Co_x)₂As₂ with x = 0.057. ²²¹ The structural phase transition temperature (T_s) , the 222 SDW antiferromagnetic phase transition temperature ¹⁶⁵ The pressure dependence of T_c determined from magne-²²³ T_{sdw} , the superconducting transition temperature T_c and tization measurements is plotted in Fig. 3(b), consistent 224 the exponent n in $\rho = \rho_0 + AT^n$ are summarized. With with the T_c obtained from resistivity measurements (Fig. 225 increasing pressure, we observe a suppression of the anti-²²⁶ ferromagnetic phase whereas, the superconducting tran-We summarize the pressure dependence of the ZFC 227 sition temperature increases, suggesting competition be-Note that the magnetization data at low temperatures 229 sure P_c around 3.5 GPa and decreases with further inwas often used to estimate the superconducting volume $_{230}$ crease in pressure, forming a dome shape. Around P_c , we fraction.^{25–27} In our case, it may not be accurate to 231 observe signature of a non-Fermi liquid namely, n close estimate the volume fraction of superconductivity from 232 to 1, often associated with quantum criticality.^{30,33} This

180 ary. Initially, M_{2K} slightly increases with pressure fol- 238 vealed that the maximum T_c occurs at the antifer-



FIG. 4. Temperature - pressure (T - P) phase diagram of Ba(Fe_{1-x}Co_x)₂As₂ with x = 0.057. The structural phase transition temperature T_s is marked as red hexagon. The SDW phase transition temperature T_{sdw} is marked as orange squares. The superconducting transition temperature T_c is determined from magnetization (solid squares) and resistivity (open circles) measurements. The exponent n is indicated by stars. The light blue and yellow represent the region with ²⁶¹

²⁴⁰ ated superconductivity.³⁴ Such a superconducting pair- ²⁶⁷ the National Research Foundation C NRF Investigator-²⁴¹ ing mechanism may be applicable in several strongly ²⁶⁸ ship (Reference No. NRF-NRFI2015-04). The work at ²⁴² correlated superconducting systems, where fundamental ²⁶⁹ KSU was supported by the National Science Foundation ²⁴³ physical quantities, including the superconducting con- ²⁷⁰ under grant No. DMR-1505826. 244 densation energy, quasiparticle lifetime, and superfluid 271

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²⁴⁵ density show abrupt changes at a QCP.³⁵ Hence, the observation of a linear temperature dependence of resistiv-246 ity at P_c about 3.5 GPa and a possible change in the su-247 perconducting volume fraction, may be associated with 248 a quantum phase transition.

D. Conclusions

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In summary, electrical resistivity and magnetization 251 ²⁵² under pressure were measured in $Ba(Fe_{1-x}Co_x)_2As_2$ with x = 0.057. Resistivity shows linear temperature de-254 pendence around a critical pressure of 3.5 GPa where T_c is maximum. Furthermore, we detected signs of 255 an accompanied change in the superconducting volume. These results are most likely due to a possible pressure 257 258 tuned QCP hidden inside the superconducting dome of ²⁵⁹ Ba(Fe_{1-x}Co_x)₂As₂.

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