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# Electron doping evolution of the structural and antiferromagnetic phase transitions in $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ iron-pnictides

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We use transport and neutron diffraction to study the electronic phase diagram of  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ . In the undoped state,  $\text{NaFeAs}$  exhibits a tetragonal-to-orthorhombic structural transition below  $T_s$  followed by a collinear antiferromagnetic (AF) order below  $T_N$ . Upon Co-doping to form  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ ,  $T_s$  and  $T_N$  are gradually suppressed leading to optimal superconductivity near  $x = 0.025$ . While transport experiments on these materials reveal an anomalous behavior suggesting the presence of a quantum critical point (QCP) near optimal superconductivity, our neutron diffraction results indicate that commensurate AF order becomes transversely incommensurate with  $T_N > T_c$  before vanishing abruptly at optimal superconductivity. These results are remarkably similar to electron-doping and isovalent doping evolution of the AF order in  $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$  and  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ , thus suggesting a universal behavior in the suppression of the magnetic order in iron pnictides as superconductivity is induced.

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High-transition temperature (high- $T_c$ ) superconductivity in copper oxides<sup>1</sup> and iron pnictides<sup>2-5</sup> generally appears in the vicinity of a magnetically ordered phase. For this reason, magnetism is believed to play a critical role in the pairing mechanisms of high- $T_c$  superconductors<sup>4</sup>. In the case of copper oxides, hole-doping to  $\text{La}_2\text{CuO}_4$  by partially substituting La by Sr will gradually suppress the static antiferromagnetic (AF) order before superconductivity emerges<sup>6</sup>. For iron pnictide superconductors such as electron-doped  $\text{BaFe}_{2-x}\text{TxAs}_2$  ( $T = \text{Co}, \text{Ni}$ )<sup>7-13</sup> and isovalent doped  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ <sup>14-18</sup>, optimal superconductivity appears near the regime where the static collinear AF order [Fig. 1(a)] is suppressed. While transport measurements have suggested the presence of a quantum critical point (QCP) in  $\text{BaFe}_{2-x}\text{TxAs}_2$ <sup>12,19</sup> and  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ <sup>20</sup> as revealed by the linear temperature dependence of the resistivity near optimal superconductivity, neutron scattering experiments instead find that the static AF order in  $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$  and  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  families of materials always has  $T_N$  above  $T_c$  and disappears in a weakly first-order fashion abruptly near optimal superconductivity, suggesting an avoided QCP<sup>12,13,17,18</sup>. On the other hand, recent neutron scattering<sup>21,22</sup> and Mössbauer spectroscopy<sup>23</sup> experiments on hole-doped iron pnictides  $(\text{Ba},\text{Sr})_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2$  family of materials found a new magnetically ordered state with restored tetragonal symmetry near optimal superconductivity, replacing the stripe AF ordered state of the undoped  $\text{BaFe}_2\text{As}_2$ <sup>24</sup>. These results suggest that the static AF order in iron pnictides display a plethora of interesting behavior near optimal superconductivity. In order to understand the relationship between magnetism and su-

perconductivity, it is important to systematically study the electronic phase diagrams of different classes of iron pnictides, and establish their universal features.

In this article, we report systematic neutron diffraction experiments on Co-doped  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  iron-pnictides. Although previous transport,  $\mu\text{SR}$ , and heat capacity experiments have established the phase diagram of  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ <sup>25-30</sup>, there are no systematic neutron diffraction measurements on  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  to determine the Co-doping evolution of the AF order and compare with those of electron, hole, and isovalently doped  $\text{BaFe}_2\text{As}_2$ <sup>12,13,17,18</sup>. Figure 1(c) summarizes the phase diagram of  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  determined from our transport and neutron diffraction experiments on single crystal samples. In addition to the well-known gradual suppression of structural and AF order with increasing Co-doping ( $T_s > T_N > T_c$ ), our data reveal a commensurate to incommensurate transition in static AF order before it vanishes abruptly near optimal superconductivity. Although transport measurements on  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  show linear temperature dependence of the resistivity near optimal superconductivity suggestive of a QCP, the first-order-like disappearing nature of the AF order with increasing Co-doping indicates an avoided QCP. These results are remarkably similar to electron-doped  $\text{BaFe}_{2-x}\text{TxAs}_2$ <sup>7-10,12,13</sup>, thus suggesting a universal electronic phase diagram for electron-doped  $\text{BaFe}_2\text{As}_2$  and  $\text{NaFeAs}$  iron pnictides in spite of their differences in crystal and electronic structures.

High-quality  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  single crystals with nominal doping level  $x$  were synthesized by the self-flux method as described elsewhere<sup>30</sup>. Similar to previous work<sup>31</sup>, we use inductively coupled plasma (ICP) atomic-

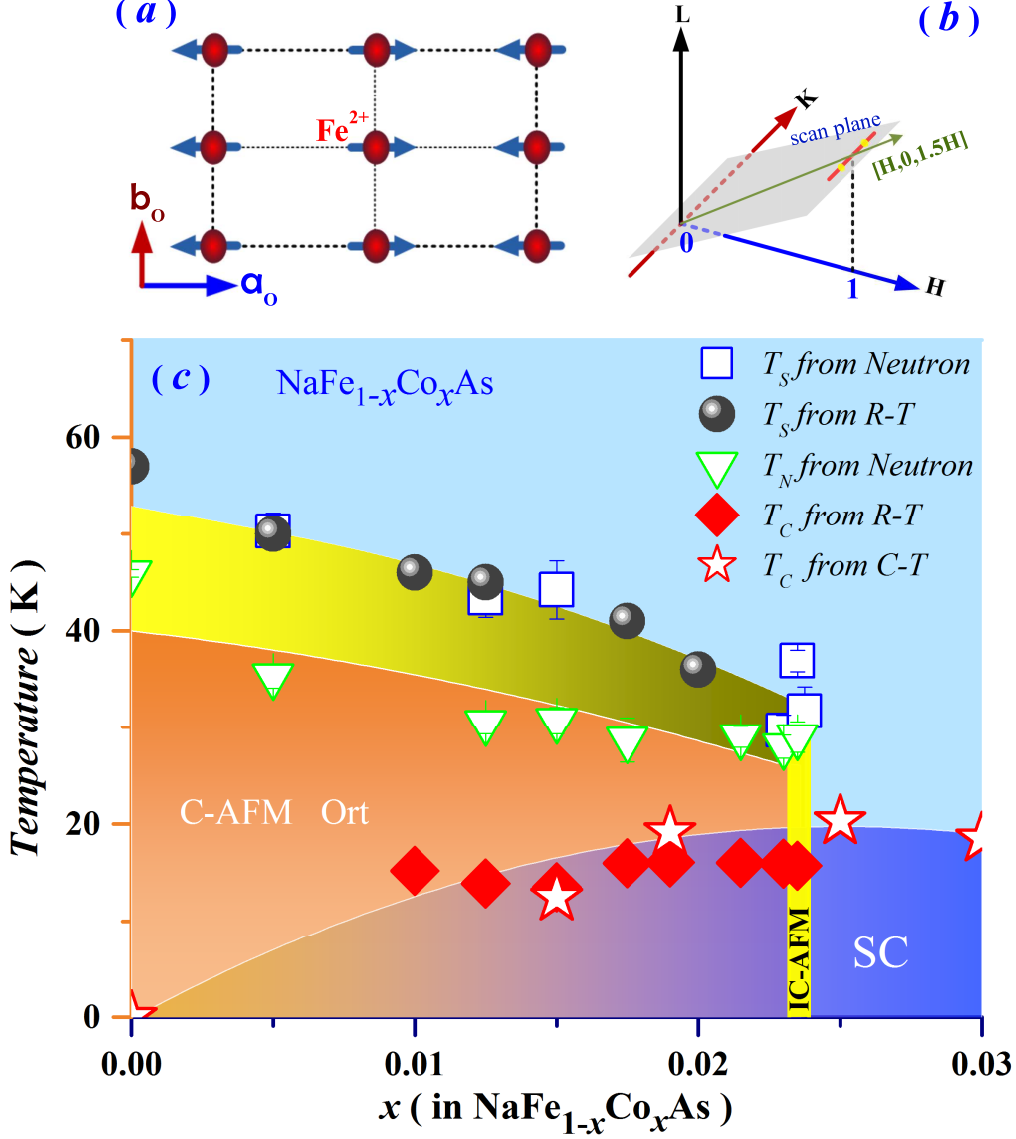


FIG. 1: (color online) The antiferromagnetic structure, neutron scattering geometry, and phase diagram of NaFe<sub>1-x</sub>Co<sub>x</sub>As as determined from transport and neutron diffraction experiments. (a) The in-plane magnetic structure of NaFeAs<sup>32</sup>. The  $a_o$  and  $b_o$  ( $a_o > b_o$ ) are orthorhombic lattice parameters in the AF ordered state. (b) Neutron scattering geometry used to search for transverse incommensurate AF order for samples near optimal superconductivity. The scattering plane is  $[0, K, 0] \times [H, 0, 1.5H]$ , and the scan direction to search for incommensurate magnetic order is along the  $[1, K, 1.5]$  direction as shown in the dashed line. (c) The phase diagram of NaFe<sub>1-x</sub>Co<sub>x</sub>As, where the Ort, Tet, and SC are orthorhombic, tetragonal, and superconductivity phases, respectively. The C-AFM and IC-AF denotes commensurate and incommensurate AF order, respectively. The green down triangles and blue squares are  $T_N$  and  $T_s$  determined from neutron diffraction experiments, respectively. The diamonds and stars are  $T_c$  determined from resistivity ( $R$ - $T$ ) and heat capacity ( $C$ - $T$ ), respectively<sup>30</sup>.

emission spectroscopy measurements to determine the chemical compositions of the samples. For the nominal NaFe<sub>0.975</sub>Co<sub>0.025</sub>As sample, which contain an unknown amount of NaAs flux, ICP measurements suggest a total chemical composition of Na<sub>0.94</sub>Fe<sub>0.89</sub>Co<sub>0.02</sub>As. Since the Na concentration of 0.94 is similar to the

combined Fe and Co concentration of 0.91, we assume that the actual chemical compositions of NaFe<sub>1-x</sub>Co<sub>x</sub>As are similar to the nominal doping levels<sup>31</sup>. Resistivity measurements were carried out with the four probe method using a physical property measurement system (PPMS). Neutron diffraction experiments were carried

out on HB-1A thermal triple-axis spectrometer at High-Flux Isotope reactor (HFIR), Oak Ridge National Laboratory. We define the wave vector  $\mathbf{Q}$  at  $(q_x, q_y, q_z)$  as  $(H, K, L) = (q_x a_o/2\pi, q_y b_o/2\pi, q_z c/2\pi)$  in reciprocal lattice units (rlu), where  $a_o \approx b_o \approx 5.56$  Å, and  $c = 6.95$  Å. In this orthorhombic unit cell notation, the collinear AF order in NaFeAs and Co-doped samples should occur at  $\mathbf{Q}_{AF} = (1, 0, L)$  where  $L = 0.5, 1.5, \dots$  rlu<sup>32</sup>.

Figure 2 summarizes temperature dependence of the in-plane resistivity measurements for NaFe<sub>1-x</sub>Co<sub>x</sub>As with  $x = 0.01, 0.0125, 0.015, 0.0175, 0.019, 0.02, 0.021$ , and  $0.023$ . The resistivity is normalized to the room temperature value per  $\rho_{ab}(T)/\rho_{ab}(300\text{ K})$  from 2 to 300 K in the inset of Fig. 2(a). For all measured Co-doping levels,  $\rho_{ab}(T)/\rho_{ab}(300\text{ K})$  is metallic in the paramagnetic state, and the upturns above  $T_c$  for lightly Co-doped samples are due to structural and magnetic phase transitions<sup>12,19</sup>. To compare the resistivity behavior in NaFe<sub>1-x</sub>Co<sub>x</sub>As with those of doped BaFe<sub>2</sub>As<sub>2</sub> family of materials<sup>12,19,33</sup>, we performed fits for the data from  $T_s + 10\text{ K}$  to  $220\text{ K}$

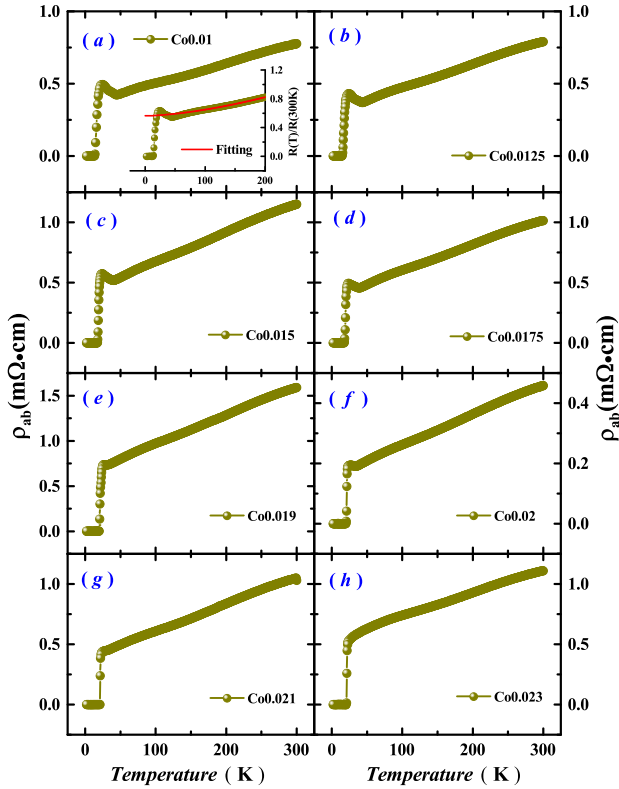


FIG. 2: (color online) Temperature dependence of the resistivity as a function of increasing Co-doping for NaFe<sub>1-x</sub>Co<sub>x</sub>As with (a)  $x = 0.01$ , (b)  $0.0125$ , (c)  $0.015$ , (d)  $0.0175$ , (e)  $0.019$ , (f)  $0.02$ , (g)  $0.021$ , and (h)  $0.023$  as obtained by PPMS. The red solid line in the inset is a fit to the model:  $\rho(T) = \rho_0 + AT^n$  up to  $220\text{ K}$  in the normal state.

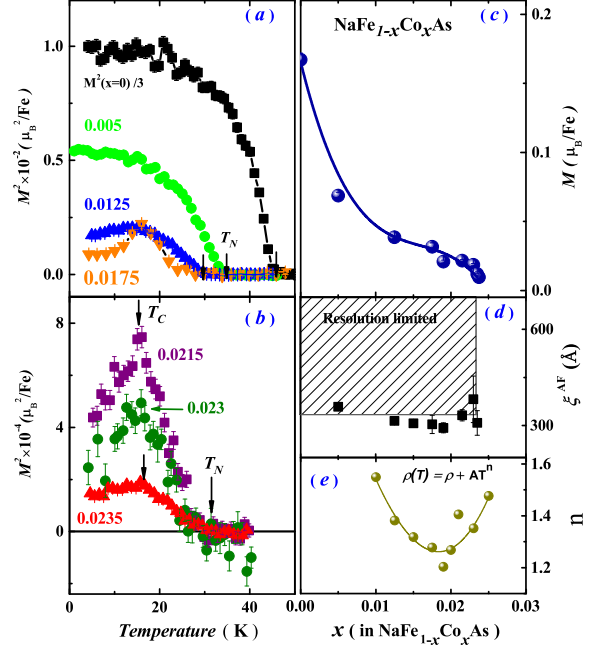


FIG. 3: (color online) Temperature dependence of the magnetic order parameters normalized to a weak nuclear Bragg peak  $(2, 0, 0)$  for NaFe<sub>1-x</sub>Co<sub>x</sub>As with (a)  $x = 0, 0.005, 0.0125, 0.0175$ , (b)  $0.0215, 0.023$ , and  $0.0235$ . The negative values are due to imperfect high-temperature background scattering subtraction. (c) The Co-doping dependence of the ordered magnetic moment. (d) The spin-spin correlation length in Å as a function of Co-doping. The shaded area indicates that spin-spin correlation length is resolution limited. (e) Co-doping dependence of the exponent  $n$  from fits to the resistivity data.

using the empirical model<sup>12,19,33,34</sup>:  $\rho(T) = \rho_0 + AT^n$ , where  $\rho_0$  is a constant,  $A$  is amplitude, and  $n$  is the temperature exponent. For a typical Fermi liquid metal, one expects  $n = 2$ , and a material close to QCP typically has  $n = 1$  with  $n < 2$  suggesting non-Fermi liquid behavior<sup>35,36</sup>. The red solid line in the inset of Fig. 2(a) represents a fit to the resistivity data. The Co-doping dependence of the fitting exponent  $n$  is shown in Fig. 3(e), which has a minimum value of  $n \approx 1.3$  around  $x \approx 0.02$  near optimal superconductivity. These results are similar to previous transport measurements on NaFe<sub>1-x</sub>Co<sub>x</sub>As<sup>28,29</sup>, and suggest the presence of a QCP near optimal superconductivity in NaFe<sub>1-x</sub>Co<sub>x</sub>As similar to that of BaFe<sub>2-x</sub>T<sub>x</sub>As<sub>2</sub>.

Figures 3(a) and 3(b) summarize the magnetic order parameters for NaFe<sub>1-x</sub>Co<sub>x</sub>As with  $x = 0.0, 0.005, 0.0125, 0.0175, 0.0215, 0.021, 0.023$ , and  $0.0235$ . The magnetic scattering has been normalized to a weak nuclear Bragg peak  $(2, 0, 0)$  in the orthorhombic notation, thus giving the ordered moment square  $M^2$  in unit of  $\mu_B^2/\text{Fe}$ . In previous neutron diffraction work on

$\text{Na}_{1-x}\text{FeAs}$ <sup>25,32</sup>, the Fe ordered moment is estimated to be  $M = 0.09 \pm 0.04 \mu_B/\text{Fe}$ , which is somewhat smaller than  $M = 0.17 \pm 0.034 \mu_B/\text{Fe}$  in the present  $\text{NaFeAs}$ . This is most likely due to the fact that previous work was carried out on slightly Na deficient samples, and the air sensitivity nature of the  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  samples was not fully recognized<sup>37</sup>. Figure 3(a) shows the data for  $x = 0.0, 0.005, 0.0125$ , and  $0.0175$ . With increasing Co-doping, we can see a clear reduction in magnetic ordered moment and  $T_N$ . For Co-underdoped bulk superconducting samples including  $x = 0.0175, 0.0215, 0.023$ , and  $0.0235$ , there is also a reduction of the AF ordered moment below  $T_c$ , indicating a competing static AF order with superconductivity similar to underdoped  $\text{BaFe}_{2-x}\text{Te}_x\text{As}_2$ <sup>8,9</sup>. In addition, the magnetic ordered moment decreases systematically with increasing Co-doping, as illustrated in Fig. 3(c). The AF ordering temperature, however, seems to saturate around  $T_N \approx 30$  K on approaching optimal superconductivity with  $x = 0.0235$ . We also note that the AF phase transitions become rounded for samples near optimal superconductivity. This is consistent with temperature dependence of the magnetic order parameters of underdoped  $\text{BaFe}_{2-x}\text{Te}_x\text{As}_2$  near optimal superconductivity, which has been attributed to a cluster spin glass state<sup>38–40</sup>.

To determine Co-doping dependence of the spin-spin correlation length in the AF ordered phase of  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ , we carried out neutron diffraction measurements in two different scattering geometries. For underdoped sample where we expect the static AF order to be commensurate, we aligned the single crystals in the  $[H, 0, 0] \times [0, 0, L]$  scattering plane. For Co-doping level  $x$  near optimal superconductivity, we aligned the crystals in the  $[0, K, 0] \times [H, 0, 1.5H]$  scattering plane as shown in Fig. 1(b) in order to search for possible transverse incommensurate magnetic order as seen in  $\text{BaFe}_{2-x}\text{Te}_x\text{As}_2$ <sup>11,12</sup>. For lightly Co-doped  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ , Fourier transforms of longitudinal scans along the  $[H, H, 0]$  direction reveal that spin-spin correlation lengths are resolution limited and greater than or equal to 290 Å [Fig. 3(d)]. This is the case for all samples probed.

Figures 4(a) and 4(b) show transverse and longitudinal scans, respectively, for  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  with  $x = 0.0125$  at different temperatures. The scattering is featureless at  $T = 45$  K (above  $T_N$ ) along both directions. On cooling to  $T = 15$  K and 5 K, we see clear magnetic scattering at commensurate positions along the longitudinal and transverse directions [Figs. 4(a) and 4(b)]. The situation is similar for  $x = 0.023$  sample [Figs. 4(c) and 4(d)]. However, the transverse scans for the  $x = 0.0235$  sample show two incommensurate peaks at  $T = 15$  K and 5 K [Fig. 4(e)], while longitudinal scans are commensurate at all measured temperatures [Fig. 4(f)]. The positions of incommensurate peaks are similar to those of electron-doped  $\text{BaFe}_{2-x}\text{Te}_x\text{As}_2$ <sup>11,12</sup>. These results thus suggest that the presence of transverse incommensurate magnetic order before optimal superconductivity may be a common feature of electron-doped  $\text{BaFe}_2\text{As}_2$  and  $\text{NaFeAs}$ .

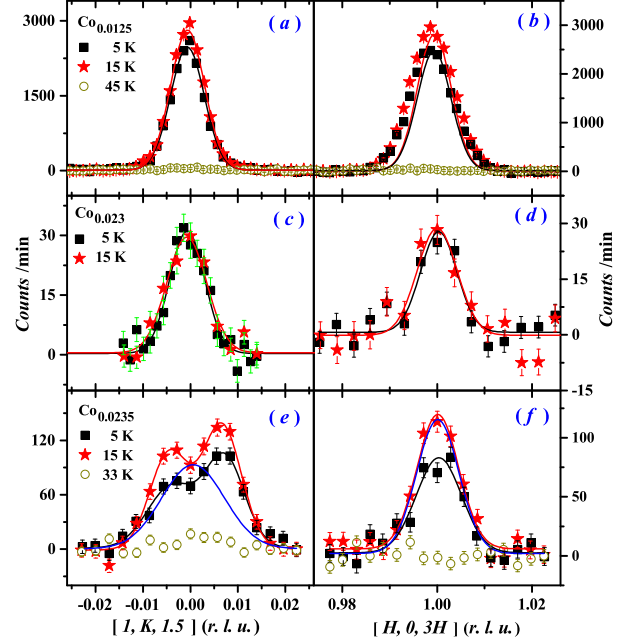


FIG. 4: (color online) Transverse and radial scans through the AF ordering wave vector  $\mathbf{Q}_{AF} = (1, 0, 1.5)$  at different temperatures using scattering geometry as shown in Fig. 1(b). Transverse scans along the  $[1, K, 1.5]$  direction at different temperatures for  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  with (a)  $x = 0.0125$ , (c)  $0.023$ , (e)  $0.0235$ . The corresponding longitudinal scans are shown in (b), (d), and (f), respectively. The blue solid line in (e) shows data obtained using  $\lambda/2$  above  $T_N$ . The solid lines are Gaussian fits to the data on linear backgrounds.

To determine electron-doping evolution of the orthorhombic-to-tetragonal structural transition in  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ , we measured the temperature dependence of the intensity at the  $(4, 0, 0)$  nuclear Bragg reflection position in the orthorhombic notation<sup>7</sup>. From previous neutron diffraction experiments on  $\text{BaFe}_2\text{As}_2$ , we know that the intensity of the  $(4, 0, 0)$  nuclear Bragg reflection [or the nuclear Bragg peak  $(2, 2, 0)$  in the tetragonal notation] shows a dramatic jump at  $T_s$  arising from the neutron extinction release that occurs due to strain and domain formation related to the orthorhombic distortion<sup>41,42</sup>. Figure 5 summarizes the key results for  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  with  $x = 0.005, 0.0125, 0.015, 0.023, 0.0235$ , and  $0.02375$ . In all cases, we see a clear enhancement of the scattering intensity below a characteristic temperature marked by vertical arrows. The resulting  $T_s$  as a function of Co-doping is plotted together with other information shown in Fig. 1(c), thus establishing the Co-doping dependence of the  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  phase diagram.

To summarize, we have mapped out the orthorhombic-to-tetragonal structural and AF ordering phase transition phase diagram in  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$  as a function



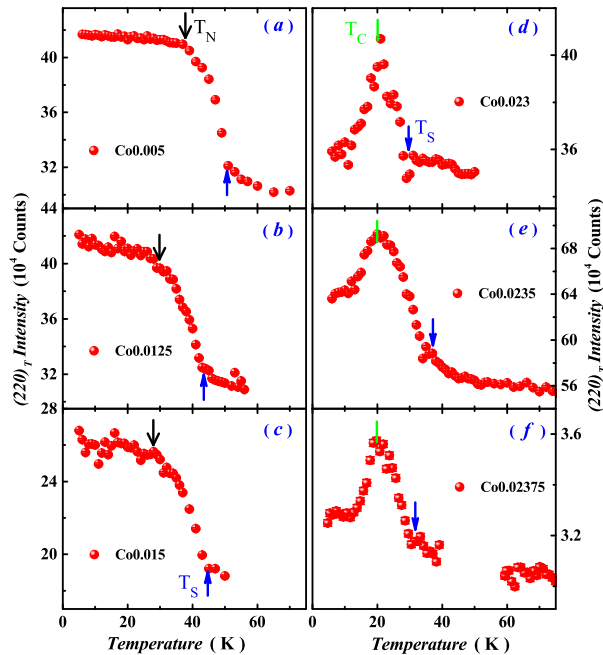


FIG. 5: (color online) Temperature dependence of the strong nuclear Bragg peak scattering  $(4,0,0)$  as a function of Co-doping (a)  $x = 0.005$ , (b)  $0.0125$ , (c)  $0.015$ , (d)  $0.023$ , (e)  $0.0235$ , and (f)  $0.02375$ . The blue vertical arrows mark our estimated  $T_s$ . The black arrows mark  $T_N$  determined from neutron diffraction measurement, and the green arrows are  $T_c$  determined from transport data.

of Co-doping up to optimal superconductivity. In spite of the rather different electron and hole Fermi surfaces between electron-doped  $\text{BaFe}_{2-x}\text{Te}_x\text{As}_2$ <sup>43</sup> and  $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ <sup>44–46</sup>, we show that Co-doping evolution of the structural and magnetic phase transitions in these two classes of materials are remarkably similar. These results confirm the universal nature of the Co-doping dependence of the structure and magnetic phase transitions, and suggest that the AF order vanishes in the first order fashion with an avoided magnetic QCP in iron pnictides. Since inelastic neutron scattering experiments on these two families of iron based superconductors reveal that their Co-doping dependent spin excitations are also similar<sup>47–50</sup>, we conclude that magnetism and spin excitations must play a critical role in the pairing mechanisms of iron-based superconductors<sup>4</sup>.

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