

## Unconventional Charge Density Wave Order in the Pnictide Superconductor $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$

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$\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  is a structural homologue of the pnictide high temperature superconductor,  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , in which the Fe atoms are replaced by Ni. Superconductivity is highly suppressed in this system, reaching a maximum  $T_c = 2.3$  K, compared to 24 K in its iron-based cousin, and the origin of this  $T_c$  suppression is not known. Using x-ray scattering, we show that  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  exhibits a unidirectional charge density wave (CDW) at its triclinic phase transition. The CDW is incommensurate, exhibits a sizable lattice distortion, and is accompanied by the appearance of  $\alpha$  Fermi surface pockets in photoemission [B. Zhou *et al.*, *Phys. Rev. B* **83**, 035110 (2011)], suggesting it forms by an unconventional mechanism. Co doping suppresses the CDW, paralleling the behavior of antiferromagnetism in iron-based superconductors. Our study demonstrates that pnictide superconductors can exhibit competing CDW order, which may be the origin of  $T_c$  suppression in this system.

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The discovery of Fe-based superconductivity in 2008 [1] uncovered an entirely new and fascinating class of unconventional superconducting materials with transition temperatures rivaling those of the high- $T_c$  cuprates [2]. A central but poorly understood feature of these materials concerns the importance of the magnetic Fe cation: Exchange of another transition metal for Fe either quenches superconductivity altogether or strongly suppresses it [3]. For example, the prototypical Fe-based superconductor,  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , exhibits a maximum superconducting transition temperature,  $T_c = 24$  K when  $x = 0.07$  [2,4,5]. However, its Ni homologue,  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$ , exhibits a maximum  $T_c$  of only 2.3 K [3,6,7]. Understanding why Ni substitution suppresses superconductivity is interesting in its own right and could shed light on the mechanism of superconductivity in Fe-based materials.

The parent material,  $\text{BaNi}_2\text{As}_2$ , has the same tetragonal  $I4/mmm$  structure as  $\text{BaFe}_2\text{As}_2$ , and undergoes a phase transition to a triclinic  $P\bar{1}$  structure at  $T_{\text{tri}} = 136$  K [7,8]. This transition is analogous to the orthorhombic transition in iron-based superconductors [2,4,5], with the exception that no evidence for antiferromagnetism has yet been found in  $\text{BaNi}_2\text{As}_2$  [9]. So the full nature of this triclinic phase is not yet established. Co doping reduces  $T_{\text{tri}}$  and leads to a superconducting dome closely resembling that in  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ , however, with a greatly reduced  $T_c$  [7]. Co-doped  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  is therefore an ideal

system to study the properties and possible origins of  $T_c$  suppression in Ni-pnictide superconductors.

Here, using x-ray scattering, we show that  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  exhibits robust, large-amplitude CDW order exhibiting the same interplay with superconductivity that antiferromagnetism does in iron-based superconductors. The CDW is incommensurate, unidirectional, and emerges in the vicinity of the triclinic transition temperature,  $T_{\text{tri}}$  [8]. At a lower temperature  $T_L$  the CDW exhibits a lock-in transition where it becomes commensurate. The CDW ordering temperature is suppressed by Co doping, leading to a phase diagram with the same dome structure as iron-based superconductors [2,4,5]. Our study shows that a competing CDW phase plays an analogous role in  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  that antiferromagnetism does in iron-based materials, and may be the cause of  $T_c$  suppression in this system.

Single crystal x-ray measurements were carried out using a low-emittance, Xenocs GeniX 3D, Mo  $K_\alpha$  (17.4 keV) microspot x-ray source with multilayer focusing optics, providing  $2.5 \times 10^7$  photons/sec in a divergence of 5 mrad and beam spot of 130  $\mu\text{m}$ . The sample was cooled by a closed-cycle cryostat employing Be domes as vacuum and radiation shields, providing wide angular access and a base temperature of 5 K. Sample motion was done with a Huber four-circle diffractometer supporting a Mar345 image plate detector in which each of  $12 \times 10^6$  pixels exhibits

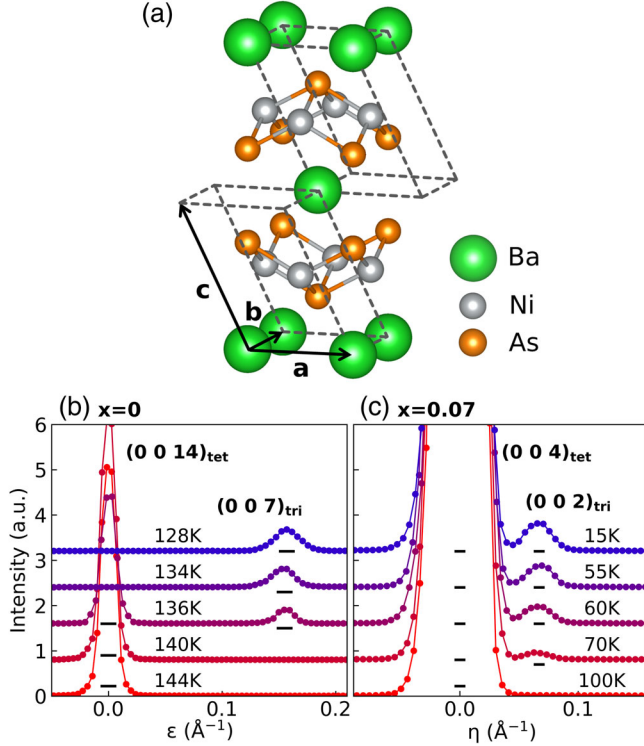


FIG. 1. Tetragonal-to-triclinic structural phase transition in  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$ . (a) Crystal structure showing the triclinic unit cell (gray dashed lines) and basis vectors (black arrows). (b) Line momentum scans through tetragonal  $(0,0,14)_{\text{tet}}$  and triclinic  $(0,0,7)_{\text{tri}}$  reflections from  $\text{BaNi}_2\text{As}_2$  ( $x=0$ ) crystal for a selection of temperatures showing the change in symmetry at  $T_{\text{tri}}$ .  $\epsilon$  is the distance in momentum space from  $(0,0,14)_{\text{tet}}$  along the direction to  $(0,0,7)_{\text{tri}}$ . (c) Same measurement for the  $(0,0,4)_{\text{tet}}$  and  $(0,0,2)_{\text{tri}}$  reflections from a Co-doped crystal with  $x=0.07$ .  $\eta$  is the distance from  $(0,0,4)_{\text{tet}}$  along the direction to  $(0,0,2)_{\text{tri}}$ . Horizontal bars represent the instrumental momentum resolution at the specific scattering geometry [10].

single-photon sensitivity. The instrument allows 3D mapping of momentum space with resolution varying from  $\Delta q = 0.01$  to  $0.08 \text{ \AA}^{-1}$ , depending on the direction of the momentum cut [10].

Single crystals of  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  with  $x = 0, 0.071 \pm 0.003, 0.082 \pm 0.0019$ , and  $0.118 \pm 0.0051$  were grown using the Pb flux method described previously [7,17]. The chemical compositions were determined by

energy dispersive x-ray measurements on multiple regions of each sample [10]. X-ray rocking curves were resolution limited for all samples,  $< 0.2^\circ$ , indicating high crystallographic quality [10]. X-ray absorption spectroscopy measurements at the As  $L_1$  edge, obtained in electron yield mode at beam line 13-3 at the Stanford Synchrotron Radiation Laboratory (SSRL), revealed changes in the As  $p$  density of states [Fig. 4(a) (inset)] similar to those observed in  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  [18].

The tetragonal and triclinic phases of  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  are characterized by distinct sets of x-ray Bragg reflections that index to their respective  $I4/mmm$  and  $P\bar{1}$  space groups [8,10]. The triclinic unit cell used for this study is defined in Fig. 1(a) with refined lattice parameters in Table I [10]. Here, we use  $(H,K,L)_{\text{tet}}$  and  $(H,K,L)_{\text{tri}}$  to denote reciprocal space locations in terms of tetragonal and triclinic unit cells, respectively.

The evolution of the tetragonal-triclinic transition with Co doping is summarized in Figs. 1(b), 1(c). The reflections used for each comparison are unimportant and chosen out of convenience. Figure 1(b) shows line scans through  $(0,0,14)_{\text{tet}}$  and  $(0,0,7)_{\text{tri}}$  reflections for a  $x=0$  crystal at a selection of temperatures. The  $(0,0,14)_{\text{tet}}$  intensity decreases at  $T_{\text{tri}} = 136 \pm 1 \text{ K}$  and the  $(0,0,7)_{\text{tri}}$  appears. After a narrow range of coexistence,  $(0,0,14)_{\text{tet}}$  peak vanishes and the  $(0,0,7)_{\text{tri}}$  grows rapidly. This observation validates previous claims that this transition is weakly first order [8].

The same comparisons for Co-doped samples show that the triclinic phase is suppressed to  $T_{\text{tri}} = 75 \pm 5 \text{ K}$  at  $x = 0.07$  [Fig. 1(c)] and  $T_{\text{tri}} = 74 \pm 2 \text{ K}$  at  $x = 0.08$  [Fig. S3(c) in the Supplemental Material [10]], respectively. The tetragonal phase does not vanish below  $T_{\text{tri}}$  at these compositions, however, but persists down to our base temperature of 5 K. Also, the development of the intensity of the triclinic Bragg reflection is more gradual in these crystals than in the  $x=0$  case. These observations suggest that Co doping suppresses and broadens the triclinic transition and leads to an extended region of coexistence between tetragonal and triclinic phases. No structural phase transition was observed in the  $x=0.12$  crystal [Fig. S3(d) [10]], which remained tetragonal down to 5 K. The behavior for all compositions studied is summarized in Fig. 4(b).

TABLE I. Structure parameters and transition temperatures of  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$ . Tetragonal lattice parameters are measured at room temperature and triclinic parameters at 50 K.

$x$	Tetragonal structure		Triclinic structure						$T_{\text{tri}}$ (K)	$T_L$ (K)
	$a$ ( $\text{\AA}$ )	$c$ ( $\text{\AA}$ )	$a$ ( $\text{\AA}$ )	$b$ ( $\text{\AA}$ )	$c$ ( $\text{\AA}$ )	$\alpha$ ( $^\circ$ )	$\beta$ ( $^\circ$ )	$\gamma$ ( $^\circ$ )		
0	4.142(4)	11.650(3)	4.21(3)	3.99(2)	6.31(1)	105.2(3)	108.6(2)	89.3(4)	$136 \pm 1$	$129 \pm 1$
0.07	4.123(4)	11.762(8)	4.15(2)	4.12(2)	6.50(9)	108.8(9)	108.9(9)	89.3(5)	$75 \pm 5$	$47.5 \pm 7.5$
0.08	4.127(4)	11.767(5)	4.14(2)	3.97(12)	6.44(2)	108.6(3)	108.3(8)	88.4(8)	$74 \pm 2$	$25 \pm 5$
0.12	4.114(6)	11.816(6)	...	...	...	...	...	...	...	...

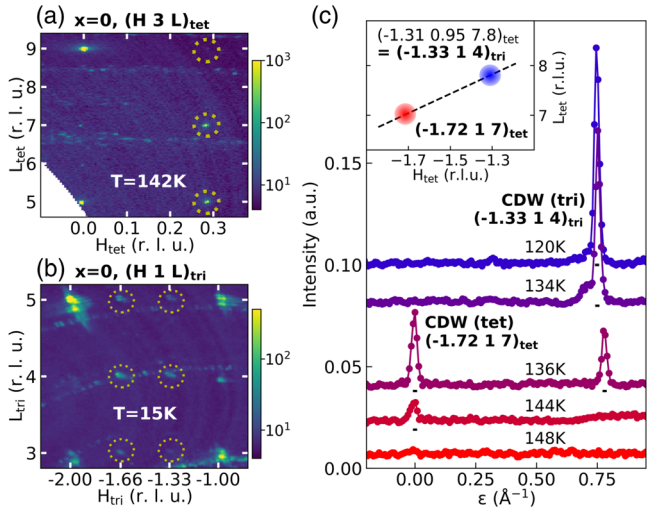


FIG. 2. CDW phases in  $\text{BaNi}_2\text{As}_2$  ( $x = 0$ ). (a) Wide ( $H, L$ ) map of reciprocal space in the tetragonal phase showing the  $(0.28, 0, 0)_{\text{tet}}$  CDW reflections in multiple Brillouin zones (dashed circles), establishing it as a coherent diffraction effect. (b) Similar map in the triclinic phase showing  $(0.33, 0, 0)_{\text{tri}}$  CDW reflections. (c) Line momentum scans through the  $(-1.72, 1, 7)_{\text{tet}}$  and  $(-1.33, 1, 4)_{\text{tri}}$  reflections showing the evolution of the CDW phases with temperature.  $\varepsilon$  is the distance from  $(-1.72, 1, 7)_{\text{tet}}$  along the direction to  $(-1.33, 1, 4)_{\text{tet}}$ . Horizontal scale bars indicate the momentum resolution [10]. (Inset) Schematic of the momentum scan displayed in panel (c).

Our main result is the discovery of a CDW in  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$ . X-ray measurements of the  $x = 0$  crystal are summarized in Figs. 2(a)–2(c) and 3(a), 3(d). While still in the tetragonal phase, as the temperature is lowered toward  $T_{\text{tri}}$ , a weak reflection with propagation vector  $(0.28, 0, 0)_{\text{tet}}$  grows in intensity as the transition is approached. This reflection is visible in multiple Brillouin zones [Fig. 2(a)], identifying it as a coherent superstructure and not an errant reflection from another grain. This reflection is incommensurate, meaning its wave vector does not index to a simple rational fraction. Because this reflection occurs only below 150 K, and is not a property of the room temperature structure, we identify it as a CDW, the first observed in any pnictide superconductor.

Below  $T_{\text{tri}}$  the  $(0.28, 0, 0)_{\text{tet}}$  reflection vanishes and is replaced by a much stronger CDW with incommensurate wave vector  $(0.31, 0, 0)_{\text{tri}}$  [Figs. 2(b), 2(c)]. Evidently the triclinic transition is associated with the formation of this CDW. Note that, despite the similar Miller indices, the vectors  $(0.28, 0, 0)_{\text{tet}}$  and  $(0.31, 0, 0)_{\text{tri}}$  are nearly  $20^\circ$  apart and reside in very different regions of momentum space [Fig. 2(c)]. The wave vector of the CDW shifts as the temperature is lowered and pins to the commensurate value  $(1/3, 0, 0)_{\text{tri}}$  at a lock-in transition  $T_L = 129$  K [Figs. 3(a), 3(d)]. Lock-in effects are an established consequence of lattice pinning in other CDW materials, suggesting pinning plays an important role in stabilization of this CDW phase [19–23].

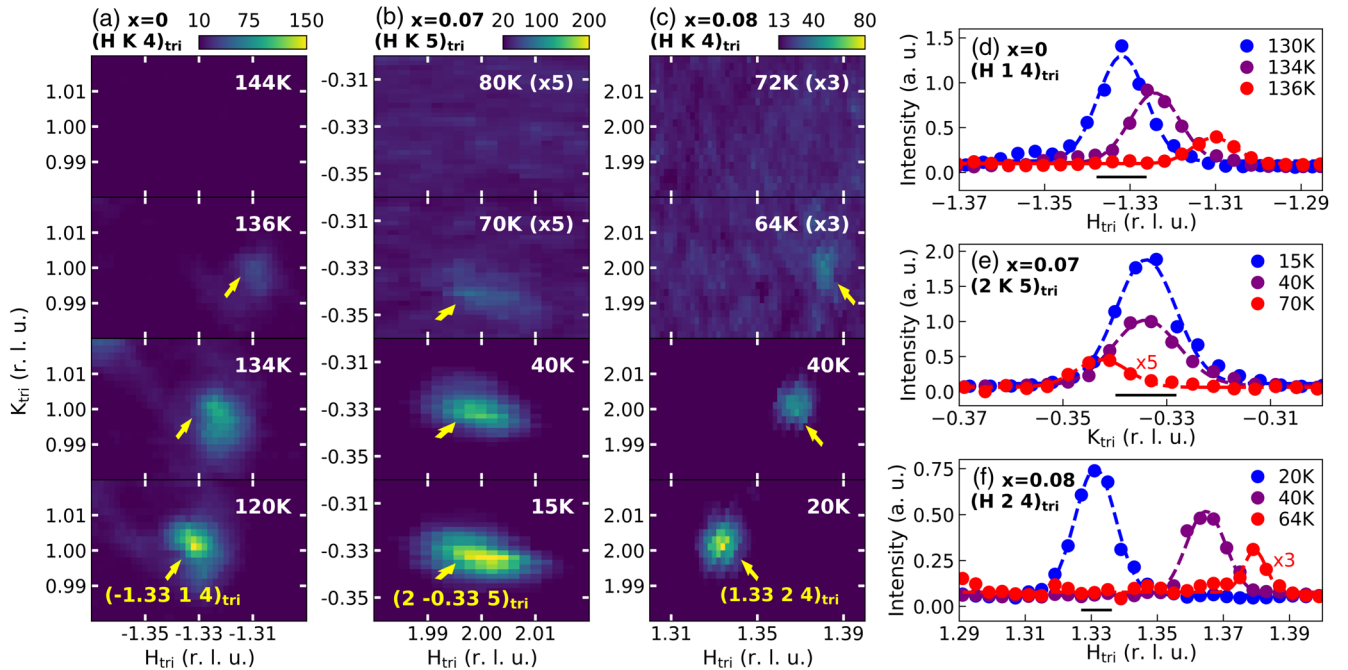


FIG. 3. CDW in the triclinic phase of  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  and lock-in transition. (a), (b), (c) Narrow ( $H, K$ ) maps of reciprocal space of the CDW in the triclinic phase of  $x = 0, 0.07, 0.08$  crystals, respectively, for a selection of temperatures. (d), (e), (f) Line momentum scans on the CDW reflections of  $x = 0, 0.07, 0.08$  crystals along modulation direction ( $H$  for  $x = 0, 0.08$  and  $K$  for  $x = 0.07$ ) showing lock-in behavior. The horizontal scale bars in panels (d)–(f) indicate the momentum resolution [10].

The CDW distortion is of the same order of magnitude as that in Peierls materials. An estimate of its magnitude can be obtained by comparing the intensities of a few CDW satellites with their associated primary Bragg reflections [10]. From the integrated intensities of the  $(-1.33, 1, 5)_{\text{tri}}$ ,  $(-1, 1, 5)_{\text{tri}}$ ,  $(-1.66, 1, 5)_{\text{tri}}$ , and  $(-2, 1, 5)_{\text{tri}}$  reflections, we estimate the lattice distortion  $\Delta \sim 0.14 \text{ \AA}$  (see Supplemental Material, Sec. VI and Fig. S7 [10]). This distortion is of the same order as in Peierls materials  $\text{TaS}_2$  and  $\text{TaSe}_2$ , which are  $0.23$  and  $0.052 \text{ \AA}$ , respectively [20]. We emphasize that this is only an order-of-magnitude estimate, and should not be considered a quantitative determination of the size of the lattice distortion. However, it suggests the CDW in  $\text{BaNi}_2\text{As}_2$  is likely driven at least in part by the electron-lattice interaction.

In Co-substituted samples, a CDW is no longer observed in the tetragonal phase. However, at  $x = 0.07$  and  $x = 0.08$  an incommensurate CDW still appears at the (reduced) triclinic transition [Figs. 3(b)–3(c)]. Both materials still exhibit lock-in transitions, at  $T_L = 47.5 \text{ K}$  for  $x = 0.07$  and  $T_L = 25 \text{ K}$  for  $x = 0.08$  [Figs. 3(e)–3(f)]. The commensurate CDW in both crystals has the same wave vector as the  $x = 0$  compound. However, strangely, the CDW in the  $x = 0.07$  sample is oriented in the  $K$  direction, with wave vector  $(0, 1/3, 0)_{\text{tri}}$ , while in  $x = 0$  and  $0.08$  it is along  $H$ . We conclude that, although  $H$  and  $K$  directions are not equivalent in the triclinic phase, the anisotropy is too small to pin the direction of the CDW modulation, which is nevertheless unidirectional in all samples.

No CDW was observed in the  $x = 0.12$  sample, which also exhibits no triclinic transition (Fig. S4(d) [10]). The CDW intensity and degree of commensurability for all samples are summarized in Figs. 4(c)–4(d).

The overall picture that emerges is as follows (see the phase diagram in Fig. 5). The  $x = 0$  compound develops weak, precursor CDW fluctuations with wave vector  $(0.28, 0, 0)_{\text{tet}}$  in the tetragonal phase upon cooling. These fluctuations are preempted at  $T_{\text{tri}} = 136 \text{ K}$  by the first order, tetragonal-to-triclinic transition and the appearance of a strong, primary CDW with wave vector  $(0.31, 0, 0)_{\text{tri}}$ . Note that these two wave vectors are indexed in different unit cells and correspond to very different locations in momentum space [Fig. 2(c)]. Upon further cooling, the CDW shifts and pins to the commensurate value  $(1/3, 0, 0)_{\text{tri}}$  at  $T_L = 129 \text{ K}$ . Magnetic measurements show a drop in the susceptibility at  $T_{\text{tri}}$  [Fig. 4(a)], suggesting the spin configuration changes when the CDW forms, though neutron measurements have not detected antiferromagnetic order [9].

The CDW coincides with unusual changes in the electronic structure. Angle-resolved photoemission spectroscopy (ARPES) studies of  $\text{BaNi}_2\text{As}_2$  ( $x = 0$ ) found that its  $\alpha$  band shifts significantly with temperature and opens Fermi surface pockets below  $T_{\text{tri}}$  [11]. The CDW wave vector,  $(1/3, 0, 0)_{\text{tri}}$ , nests these holelike  $\alpha$  pockets

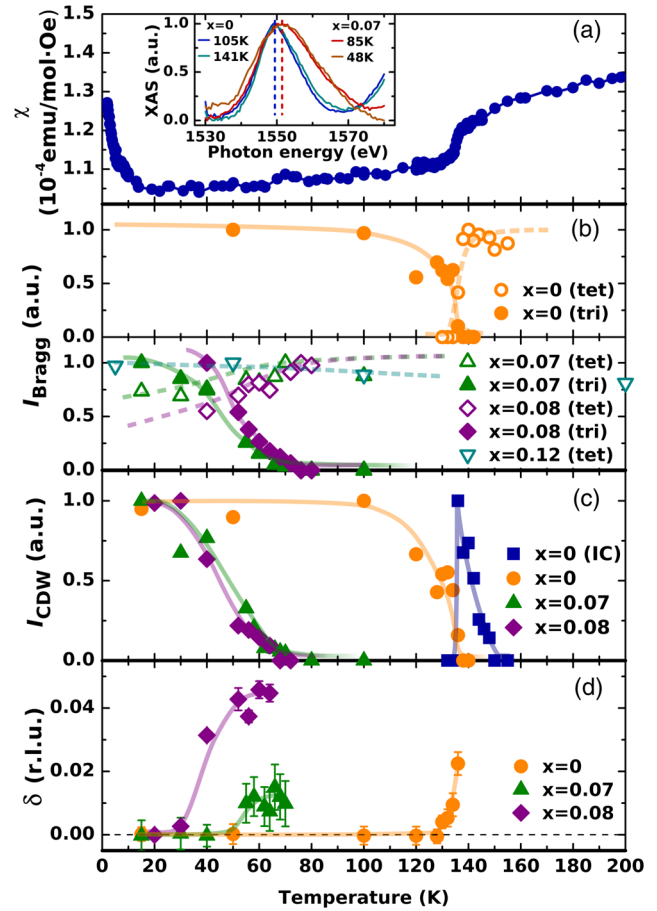


FIG. 4. Summary of the temperature dependence of various properties of  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$ . (a) Magnetic susceptibility of  $\text{BaNi}_2\text{As}_2$  ( $x = 0$ ) measured at  $10 \text{ kOe}$ . The anomaly at  $136 \text{ K}$  suggests a change in magnetic configuration at the triclinic transition, and the rise at low temperature is a Curie-Weiss effect. (inset) XAS spectra at the As  $L_1$  edge for two temperatures and two compositions. Dashed lines represent the center of each spectrum. (b) Integrated intensities of the tetragonal and triclinic reflections (Fig. 1) for all four compositions. All curves are scaled to the maximum observed intensity. (c) Integrated intensities of the CDW reflections (Fig. 2) for all four compositions, again scaled to the maximum intensity. (d) Incommensuration parameter,  $\delta$ , defined as the distance in momentum space to the closest commensurate point showing the lock-in transition at low temperature.

(Fig. S9 [10]), suggesting they may have some connection to the CDW formation. However, no energy gap is observed to open at  $T_{\text{tri}}$ , and no evidence for band folding, which would be expected when translational symmetry is broken, is observed. We conclude that the observed CDW is unconventional in that it is connected to electronic structure changes but does not follow a traditional Fermi surface nesting paradigm [19].

Co doping suppresses the precursor fluctuations, reduces  $T_{\text{tri}}$ , and broadens the triclinic transition, leading to an extended heterogeneous coexistence region of tetragonal and triclinic phases [Figs. 1(b)–1(c) and 4(b)]. This

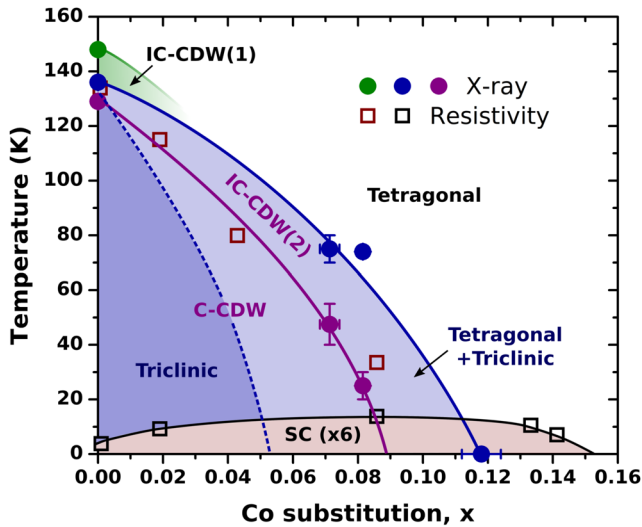


FIG. 5. Phase diagram of  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$ . (green) Incommensurate CDW in the tetragonal phase. (dark purple) Triclinic phase exhibiting a commensurate CDW. (light purple) Heterogeneous region exhibiting coexisting tetragonal and triclinic phases as well as either an incommensurate CDW (IC-CDW) or a commensurate CDW (C-CDW). (beige) Superconducting phase, whose maximum  $T_c$  arises in a region of heterogeneous coexistence (data points taken from Ref. [7]).

broadening suggests that disorder, perhaps from the Co dopants, plays an important role despite the high crystallographic quality of the crystals [10]. Surprisingly, disorder has less effect on the CDW itself. The lock-in temperature  $T_L$  is reduced by Co doping (Fig. 5), but the CDW remains resolution limited in all materials [Figs. 3(d)–3(f)].

The observation of nonuniversal CDW orientation that the modulation runs along the  $H$  direction at  $x = 0$  and  $0.08$ , but along  $K$  at  $x = 0.07$  [Figs. 3(a)–3(c)] could indicate a large nematic susceptibility. While the triclinic distortion explicitly breaks rotational symmetry of the system below  $T_{\text{tri}}$ , recent elastoresistance measurements on  $\text{BaNi}_2\text{As}_2$  show evidence for electronic nematic order which breaks rotational symmetry of the tetragonal phase above the triclinic transition [24]. This suggests the direction of the CDW may be determined by tiny extrinsic influences such as strains due to sample mounting.

In summary, we showed using x-ray scattering that the pnictide superconductor  $\text{Ba}(\text{Ni}_{1-x}\text{Co}_x)_2\text{As}_2$  exhibits a unidirectional and incommensurate CDW. The CDW is accompanied by the appearance of  $\alpha$  Fermi surface pockets in photoemission, suggesting it forms by an unconventional mechanism. Nevertheless, the size of the lattice distortion,  $\Delta \sim 0.14 \text{ \AA}$  [10], is of the same order as many well-known Peierls materials, suggesting the electron-lattice interaction is involved. Co doping suppresses the CDW, which plays a role analogous to antiferromagnetism in iron-based superconductors. Our study demonstrates that pnictide superconductors can exhibit CDW order, which may be the origin of  $T_c$  suppression in this material.

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