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Spectral Evidence for Unidirectional Charge Density Wave in Detwinned BaNi₂As₂

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In the iron-based superconductors, unconventional superconductivity emerges in proximity to intertwined electronic orders consisting of an electronic nematic order and a spin density wave (SDW). Recently, BaNi₂As₂, like its well-known iron-based analogue BaFe₂As₂, has been discovered to host a symmetry-breaking structural transition but coupled to a unidirectional charge density wave (CDW) instead of SDW, providing a novel platform to study intertwined orders. Here, through a systematic angle-resolved photoemission spectroscopy study combined with a detwinning B_{1g} uniaxial strain, we identify distinct spectral evidence of band evolution due to the structural transition as well as CDW-induced band folding. In contrast to the nematicity and spin density wave in BaFe₂As₂, the structural and CDW order parameters in BaNi₂As₂ are observed to be strongly coupled and do not separate in the presence of uniaxial strain. Furthermore, no nematic band splitting is resolved above the structural transition. Our measurements point to a likely lattice origin of the CDW order in BaNi₂As₂.

Quantum materials hosting unconventional superconductivity tend to develop complex phase diagrams where multiple electronic orders interact. In the Fe-based superconductors (FeSCs), the ubiquitous intertwined order takes form in a C_4 -rotational symmetry-breaking nematic phase and a spin density wave (SDW) [6, 31, 32]. The nematic order manifests in a tetragonal to orthorhombic structural transition, identified as electronically driven by a divergent nematic susceptibility from elastoresistance measurements [4]. Additionally, rotational symmetry-breaking is observed in the electronic, magnetic and optical properties [2, 5, 7, 11, 19, 30, 34, 35]. Superconductivity emerges when these competing orders are suppressed by either doping or pressure, resulting in T_c as high as 40 K [18, 28].

BaNi₂As₂—a nickel-based analog of the well-studied FeSC BaFe₂As₂—shares the same high-temperature tetragonal phase [23]. Distinct from BaFe₂As₂, BaNi₂As₂ is nonmagnetic and superconducts with a T_c of

0.6K [22]. Instead of the coupled nematicity and SDW found in BaFe₂As₂, BaNi₂As₂ exhibits a symmetry-lowering structural transition into a triclinic phase at $T_S = 136$ K accompanied by unconventional charge density waves (CDWs). The progression from the tetragonal state to the triclinic state is still under debate. From x-ray diffraction, Lee. *et al.* discovered that the CDW first appears in the tetragonal state as an incommensurate order (IC-CDW), then transits to a unidirectional IC-CDW at T_S and then locks into a commensurate order (C-CDW) below T_S [13, 14]. The crystal structure in this picture changes from tetragonal directly to triclinic at T_S in a first-order transition. Recent dilatometry work suggests that the crystal structure explicitly breaks C_4 rotational symmetry in the form of an orthorhombic phase in a second-order transition before additional symmetry-lowering into the triclinic phase at T_S in a first-order fashion [17]. Substitution of Co on the Ni site, or Sr on the Ba site, or P on the As site can completely

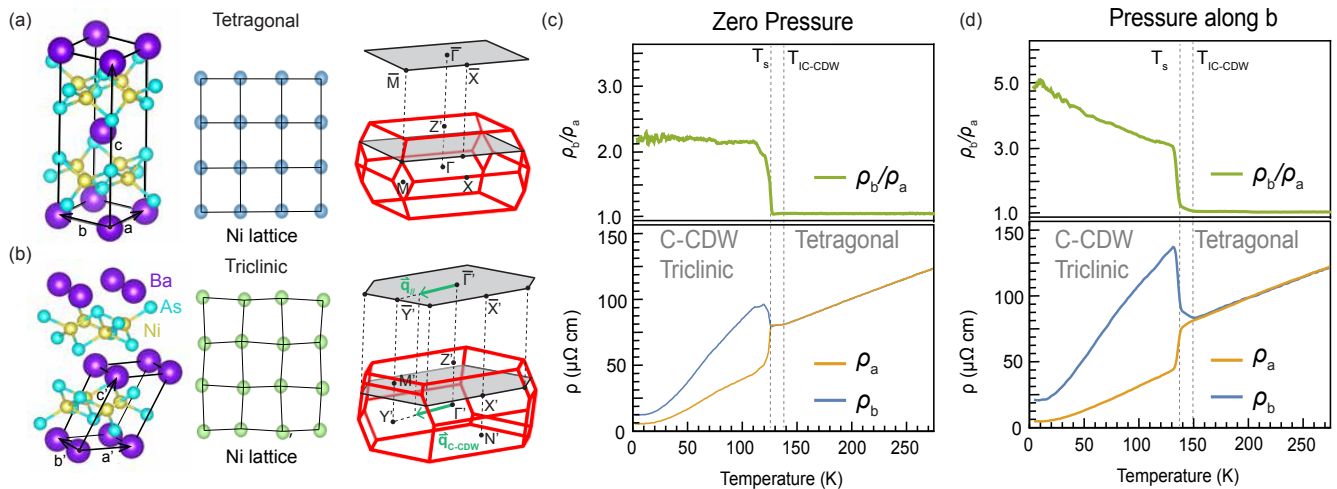


FIG. 1. (a)-(b) Crystal structure and BZ for the tetragonal and triclinic phases. The red (gray) BZ represents the 3D (2D projected) BZ. The green arrows in (b) denote the q -vectors for C-CDW and its in-plane component. (c)-(d) Temperature-dependent in-plane resistivity ρ_a (yellow), ρ_b (blue) and ρ_b/ρ_a (green) with and without uniaxial compressive strain along b . Transition temperatures T_S and T_{IC-CDW} are denoted by the gray dashed lines.

suppress the triclinic phase and the associated CDW orders, reaching a maximum superconducting T_c of 3.5 K.

Although the hierarchy of the symmetry-breaking orders in BaNi_2As_2 appears to be similar to BaFe_2As_2 , it is still unclear whether there exists electronically-driven nematicity. Elastoresistance measurements show that $\text{Ba}_{1-x}\text{Sr}_x\text{Ni}_2\text{As}_2$ and $\text{BaNi}_2(\text{As}_{1-x}\text{P}_x)_2$ exhibit diverging nematic susceptibilities in the B_{1g} channel. A Ginzburg–Landau analysis suggests that the divergence of nematic susceptibility in $\text{Ba}_{1-x}\text{Sr}_x\text{Ni}_2\text{As}_2$ could be driven by either the lattice or electronic degrees of freedom [8]. In contrast, in BaFe_2As_2 , the divergence in the B_{2g} channel is electronically-driven. Reports present conflicting results on which phase transition corresponds to these divergence peaks and whether the associated fluctuations are responsible for the enhancement of T_c at the optimal doping [8, 10]. In addition, nematic/IC-CDW fluctuations have been reported for a large temperature window above T_S by studies including Raman scattering [33], inelastic X-ray scattering [26, 27], near-edge x-ray absorption fine structure [17], and thermodynamics measurements [16]. Therefore BaNi_2As_2 and its doping series offer a rich platform to investigate intertwined orders where the electronic structure could provide important insights into the nature of nematicity, CDWs, and their connection to unconventional superconductivity.

Here, using angle-resolved photoemission spectroscopy (ARPES) under uniaxial strain, we reveal the evolution of the electronic structure of BaNi_2As_2 across its phase transitions. We find evidence of unidirectional band folding consistent with the reported C-CDW wavevector in the C-CDW/triclinic state, and rotational symmetry breaking that onsets abruptly at the triclinic tran-

sition. Furthermore, the extracted temperature evolution of the spectral features identifies a distinct order parameter for the C-CDW from that of the triclinic structural transition. However, the two order parameters are observed to be strongly coupled even in the presence of uniaxial strain, distinct from the case of BaFe_2As_2 where strain lifts the onset of the nematic band shift to above that of the SDW order. Our results taken together suggest a strongly lattice-driven intertwined order in parent BaNi_2As_2 and much weaker nematic fluctuations in a large temperature window compared to that of BaFe_2As_2 .

High-quality single crystals of BaNi_2As_2 were synthesized using the self-flux method [23]. Resistivity measurements were carried out in a Quantum Design Physical Properties Measurement System using a clamp to supply an in-plane, uniaxial pressure [3]. ARPES measurements were performed at the QMSC beamline at the Canadian Light Source, beamlines 4.0.3, 7.0.2 and 10.0.1 of the Advanced Light Source, and beamline 5-2 of the Stanford Synchrotron Radiation Lightsource with electron analyzers at an energy and angular resolution of 12 meV and 0.1° , respectively. The samples were cleaved *in-situ* with base pressures below 5×10^{-11} Torr. The polarization used was circular left (CL) unless otherwise noted. To avoid the domain mixing effect, we used a mechanical clamp to detwin the BaNi_2As_2 crystals along the [010] direction, with a typical pressure of 5-10 MPa [9].

At room temperature, tetragonal BaNi_2As_2 belongs to the space group $I4/mmm$ ($a=4.142\text{\AA}$, $c=11.65\text{\AA}$) (Fig. 1a). It undergoes a first-order phase transition to the triclinic structure ($a=4.21\text{\AA}$, $b=3.99\text{\AA}$, $c=6.31\text{\AA}$, $\alpha=105.2^\circ$, $\beta=108.6^\circ$, $\gamma=89.3^\circ$, space group $P\bar{1}$) at $T_S =$

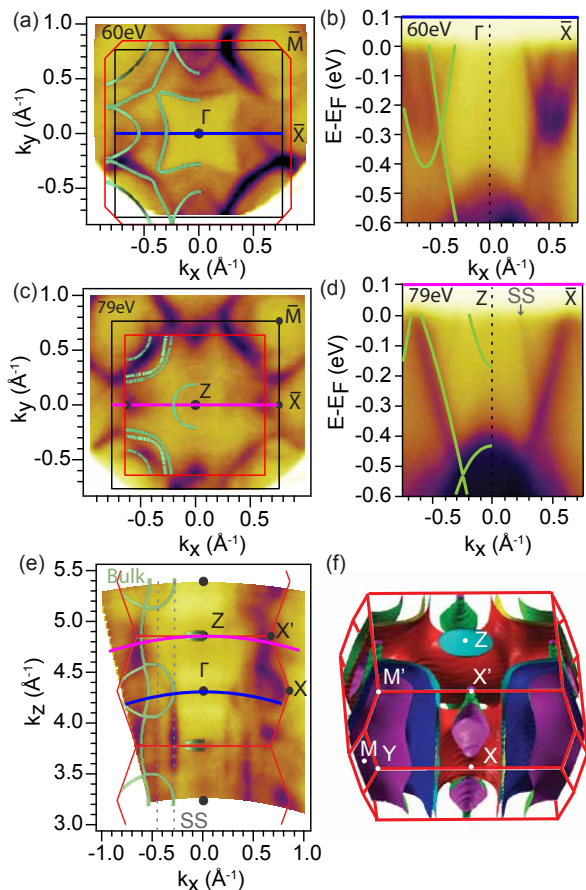


FIG. 2. Electronic structure in the tetragonal state (170 K). (a) Fermi surface (FS) measured with 60 eV photons ($k_z = 0$). (b) Band dispersions measured along Γ -X as marked by the blue lines in (a) and (e). (c)-(d) Same as (a)-(b) but measured with 79 eV photons ($k_z = \pi$), where band dispersions are measured along Γ -Z as marked by the pink lines in (c) and (e). (e) Photon energy-dependent measurement in the k_x - k_z plane, with an inner potential of 15 eV. (f) DFT calculation of the 3D FSs. In addition, DFT-calculated band dispersions and FSs are overlaid on all data in green solid lines. Grey arrows and dashed lines mark the surface states. All data were taken with circular right (CR) polarization.

136K (Fig. 1b). The corresponding 3D Brillouin Zones (BZ) of the tetragonal and triclinic phases are shown accordingly, where the rotational C_4 symmetry is broken for the triclinic phase. The C-CDW q -vector in the triclinic state is denoted by q_{C-CDW} , which does not lie in the projected BZ plane while its in-plane component ($q_{//}$) is very close to one-third of the projected tetragonal BZ (see Supplemental Material [1]). We carried out temperature-dependent in-plane electrical resistivity measurements along the two orthogonal directions with and without uniaxial pressure. In contrast to BaFe_2As_2 [3], even in the absence of detwinning stress, we observe resistivity anisotropy (ρ_b / ρ_a) below T_S , signaling a strong structural distortion and associated unequal

domain populations. In the presence of stress, the resistivity anisotropy is enhanced, demonstrating that the applied uniaxial stress redistributes the domain populations. In addition, we observe a kink above T_S in the zero-stress sample, which corresponds to the reported IC-CDW or orthorhombic transition [10, 13]. In the measurement under stress, the kink feature is replaced by the onset of the resistivity anisotropy, which can be interpreted as either a detwinning effect of the crystal orthorhombic domains [34] or strain-induced rotational symmetry-breaking [25]. It is interesting to point out that, in contrast to BaFe_2As_2 , the uniaxial pressure does not broaden the structural transition [3, 29]. To visualize the electronic structure of BaNi_2As_2 , we present ARPES measurements taken in the tetragonal phase. The Fermi surface maps (FSMs) and band dispersions along high symmetry directions corresponding to the $k_z = 0$ (Fig. 2ab and $k_z = \pi$ (Fig. 2cd) planes are taken with 60 eV and 79 eV photons, respectively (Fig. 2e). The C_4 symmetry of the tetragonal state is observed, with good overall agreement with Density Functional Theory (DFT) calculations without a renormalization factor. The weak correlation effect is consistent with previous ARPES reports [20, 36]. The most intense Fermi surfaces are large pockets around the M points. As Ni has two more electrons than Fe, BaNi_2As_2 is effectively a heavily electron-doped analog of BaFe_2As_2 [12]. This can be seen in the dispersions, where the hole band maxima typically seen near E_F in BaFe_2As_2 are ~ 0.5 eV below E_F in BaNi_2As_2 (Fig. 2d). From a photon energy dependence study (Fig. 2e), we observe k_z -dispersive bulk bands, as well as non-dispersive bands that have no correspondence in the bulk band calculations and must be therefore surface states (denoted by SS).

Having identified the electronic structure in the tetragonal state, we now examine the low-temperature triclinic phase. To probe the intrinsic single-domain electronic structure, we apply uniaxial compressive strain along the crystal axis [010], which aligns all the domains along the shorter b -axis. The main features of the low-temperature FSM of the strained crystal (Fig. 3a) resemble their tetragonal counterparts except for the appearance of new bands marked in cyan, which can be understood as folded copies of the M pocket that only appear along the b -direction, hence breaking the rotational C_4 symmetry. This anisotropy can be demonstrated by a comparison of the momentum distribution curve (MDC) taken at equivalent momenta along the k_x and k_y directions, where only a peak is seen across the folded feature (Fig. 3e). Such band folding is a signature of translational symmetry breaking. We note that the folding vector is approximately $\frac{1}{3}$ of the BZ boundary, which is consistent with the in-plane projection of the q_{C-CDW} observed by x-ray diffraction [13, 20]. Therefore, this band folding is a signature of the unidirectional CDW, which was not observed in previous ARPES studies of BaNi_2As_2 .

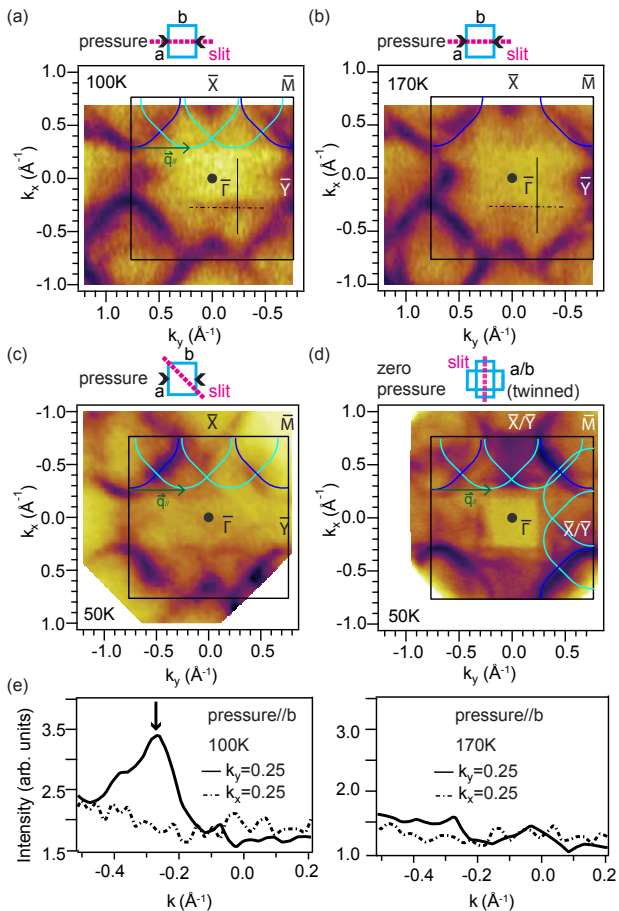


FIG. 3. FSs of detwinned BaNi_2As_2 . (a) FS taken at 79 eV in the triclinic phase (100K) with horizontal compressive uniaxial pressure. (b) Same as (a) but taken in the tetragonal phase (170K). (c) Same as (a) but taken at 50K with a 45° analyzer slit direction. (d) FS taken with CR 60eV photons at 50K without uniaxial pressure. Blue lines mark the Fermi pockets around \bar{M} , while cyan lines indicate bands folded from the \bar{M} points due to the C-CDW. (e)(f) MDCs along $k_y=0.25\text{\AA}^{-1}$ (solid line) and $k_x=0.25\text{\AA}^{-1}$ (dashed line) for the strained sample at 100K and 170K respectively, as marked in (a)-(b). The black arrow denotes the folded feature.

To exclude that the rotational symmetry-breaking effect is due to extrinsic photoemission matrix elements, we measured the strained sample in a geometry where the a and b directions are symmetric with respect to the analyzer slit (Fig. 3c). In this geometry, the photoemission matrix elements are equivalent along the k_x and k_y directions therefore any observed difference must be intrinsic to the band structure. We note that the folded bands still only appear along the strained direction, reflecting a true C_2 symmetry. In addition, above the CDW ordering temperature (Fig. 3b), the folded bands in the strained sample disappear, restoring the C_4 symmetry, as also confirmed by the disappearance of the peak in the MDC (Fig. 3e). For comparison, the FSM of an unstrained twinned crystal (Fig. 3d) shows folded bands

in both directions, consistent with the understanding of unidirectional CDW folding under mixed domains. The comparison between the FSMs of the strained and unstrained crystals clearly establishes that uniaxial strain is effective at detwining the crystal and important for resolving the observed CDW band folding.

Next, we examine in detail the evolution of the rotational and translational symmetry breaking in the electronic structure. Specifically, we traced the dispersions along the $\bar{\Gamma}$ - \bar{X} and $\bar{\Gamma}$ - \bar{Y} directions measured on detwinned crystals as a function of temperature (Fig. 4). While dispersions along the two orthogonal directions are identical in the tetragonal phase (170 K), bands are strongly modified in the low-temperature phase (100 K), with band crossing only along $\bar{\Gamma}$ - \bar{X} but not along $\bar{\Gamma}$ - \bar{Y} (Fig. 4b-g). To understand the observed dispersions, we illustrate with a schematic in Fig. 4a. In the normal state above T_{IC-CDW} , the Fermi surface consists of large pockets around the \bar{M} points. Two types of transitions occur that modify the bands. First, the structural transition from tetragonal to triclinic is reflected in a broken rotational symmetry, where the blue dispersion bends down along $\bar{\Gamma}$ - \bar{Y} . Second, the unidirectional CDW folds the \bar{M} pockets along k_y , resulting in a folded parabolic band (cyan) that only appears along $\bar{\Gamma}$ - \bar{X} .

We then use the spectral signatures of these two distinct symmetry breaking to trace out their order parameters. First, we trace the spectral signature associated with the rotational symmetry breaking in the form of the band shift along $\bar{\Gamma}$ - \bar{Y} . As captured by MDC3 in Fig. 4eh, an abrupt change occurs at T_S in a strongly first-order fashion. This is also reflected in the apparent jump in the temperature-dependent spectral weight integrated across the gray momentum range (Fig. 4hi). Second, we trace the band folding by comparing the MDC across the cyan band for $\bar{\Gamma}$ - \bar{X} and $\bar{\Gamma}$ - \bar{Y} (Fig. 4h). The presence of the folded band is seen in a hump in the MDC along $\bar{\Gamma}$ - \bar{X} (MDC1) but not $\bar{\Gamma}$ - \bar{Y} (MDC2). The spectral weight of the hump gradually decreases as the temperature is raised, confirmed by the integrated spectral weight in the marked momentum range plotted against temperature, which mimics the behavior of a second-order phase transition (Fig. 4j). This is in contrast to the abrupt jump of the rotational symmetry-breaking in Fig. 4i, suggesting that the CDW and triclinic structural transition have distinct order parameters.

Comparing the two extracted order parameters, we note that both appear to onset simultaneously in a single phase transition within our experimental uncertainty, although the CDW band folding order parameter evolves much more smoothly than that of the band shift. This contrast is also apparent in the collapsed MDC curves in Fig. 4h. The abrupt nature of the onset suggests that the two order parameters correspond to the first-order structural transition at T_S into the triclinic phase and the C-CDW order, respectively. Since a small kink

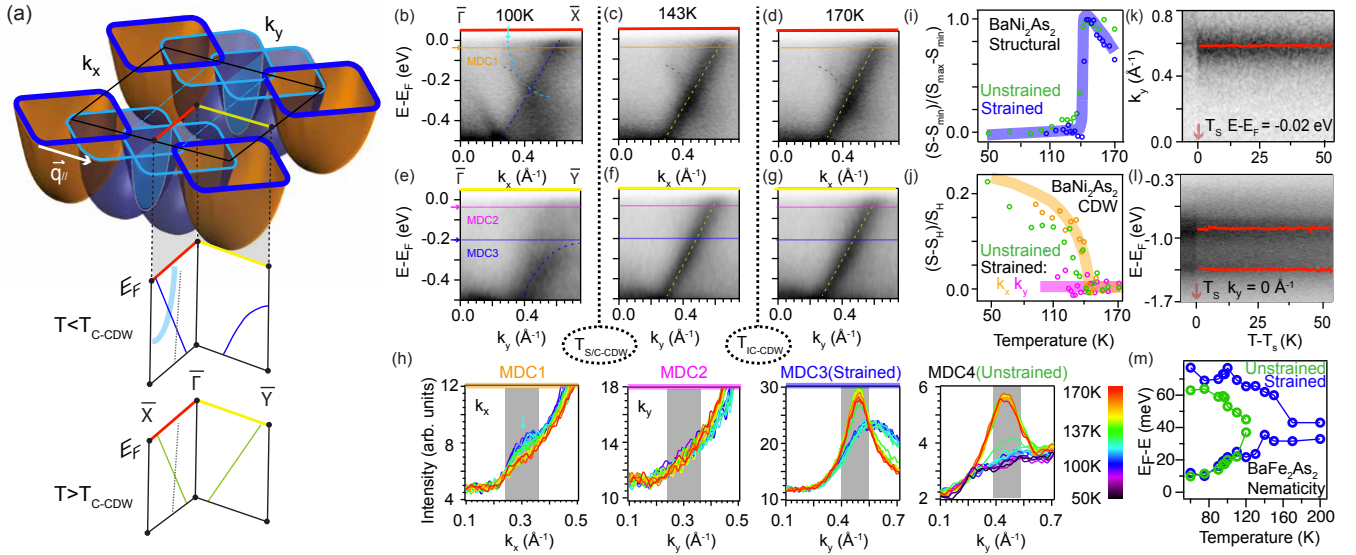


FIG. 4. Temperature evolution through the CDW transitions on detwinned BaNi_2As_2 . (a) Summary of the band evolution across $T_{S/IC-CDW}$. (b)-(d) Temperature dependence of band dispersions along $\bar{\Gamma}-\bar{X}$ for (b) $T < T_{S/IC-CDW}$, (c) $T_{S/IC-CDW} < T < T_{IC-CDW}$, and (d) $T > T_{IC-CDW}$. (e)-(g) Same as (b)-(d) but taken along $\bar{\Gamma}-\bar{Y}$. Dashed lines are guides to the eyes. (h) Temperature dependence of MDCs as marked in (b)-(g). The locations of MDCs are as marked. (i) Integrated spectral weight as a function of temperature within the gray window for MDC3 and MDC4. Each curve is normalized to the maximum spectral weight. (j) Integrated spectral weight within the gray window in MDC1 and MDC2 and the equivalent MDC taken on the unstrained crystal as a function of temperature. Each curve is normalized by calculating $(S-S_H)/S_H$, where S_H is the average spectral weight between 145K-170K. Solid lines are guides to the eyes. (k) (l) Continuous temperature evolution of the MDCs and EDCs starting slightly below $T_{S/IC-CDW}$. The red line denotes the peak position by fitting (see Supplemental Material [1]). (m) Temperature evolution of d_{xz} and d_{yz} band positions in BaFe_2As_2 with and without uniaxial strain (reproduced from [34]).

is also observed above T_S in the resistivity indicating the onset of IC-CDW and potential nematic order, we searched for evidence of nematic band shift above T_S via a measurement of the dispersions along $\bar{\Gamma}-\bar{Y}$ under a slow continuous temperature ramp. The EDC (energy distribution curve) and MDC evolutions (Fig. 4kl) show no resolvable shift or band folding. From a fitting of these bands, we obtain an upper bound of any potential nematic band shift to be 10 meV as shown in the Supplemental Material [1], much smaller than that observed for the nematic phase in BaFe_2As_2 (Fig. 4m), and on the scale of shift accountable by the lattice orthorhombicity in BaFe_2As_2 [34]. The lack of band folding in the IC-CDW region suggests that the associated order parameter is much smaller than that of the C-CDW order on the sample surface, consistent with a recent STM report that also did not find evidence of IC-CDW [21].

We then study the effect of uniaxial strain on the order parameters extracted from the spectral signatures. The spectral weights acquired from unstrained crystals by following the same procedures described above are plotted in (Fig. 4ij), showing very similar behavior as those obtained on strained samples. In particular, we observe no elevation in the onset temperature of either order parameter during the warming-up. This is again in contrast to BaFe_2As_2 (Fig. 4k), where the uniaxial strain clearly

elevates the onset temperature of the observed orbital anisotropy and separates it from the SDW ordering temperature [24]. The results taken together suggest that the translational symmetry-breaking of the C-CDW and the structural transition into the triclinic phase are strongly coupled in BaNi_2As_2 , and that nematic fluctuations are much weaker than that in BaFe_2As_2 . The C-CDW, from a lack of Fermi surface nesting conditions, is likely dominated by lattice. Recent DFT results show that two distinct structures compete due to complex As bonding patterns and drive distortions of the Ni layers which possibly explain the unconventional CDW behavior of the ground state [15].

In the broader context of FeSCs, the coupling of the nematic and magnetic order varies as well. In BaFe_2As_2 , the structural transition is second-order, followed by a first-order magnetic transition. In the structural homolog SrFe_2As_2 , the structural and magnetic transitions are strongly first-order and occur simultaneously. Uniaxial strain also does little in harvesting the nematic fluctuations above T_S [29]. The case of BaNi_2As_2 as we demonstrate here appears to be similar in spirit to that of SrFe_2As_2 , where very weak rotational symmetry-breaking exists above the strong first-order structural transition. However, with the substitution of P on the As site or Sr on the Ba site in BaNi_2As_2 , nematic fluctu-

ations appear to grow both in the size of the anisotropic thermal expansion above T_S [17], as well as a diverging nematic susceptibility, which has been proposed to be responsible for the enhancement of T_c [8]. BaNi_2As_2 family, therefore, offers a rich platform analogous to the magnetic FeSCs where intertwined order from the charge-nematic sector interacts with superconductivity.

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- [1] See Supplemental Material at [URL will be inserted by publisher] for the illustration of in-plane components of CDW q-vectors for the triclinic and tetragonal phases; temperature evolution of the high symmetry cuts taken on strained and unstrained samples; continuous temperature evolution of band dispersion of the high symmetry cuts; EDC analysis of CDW gap.
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