Spectral Evidence for Unidirectional Charge Density Wave in Detwinned BaNi$_2$As$_2$

Yucheng Guo, Mason Klemm, Ji Seop Oh, Yaofeng Xie, Bing-Hua Lei, Sergey Gorovikov, Tor Pedersen, Matteo Michiardi, Sergey Zhidanovich, Andrea Damascelli, Jonathan Denlinger, Makoto Hashimoto, Donghui Lu, Sung-Kwan Mo, Rob G. Moore, Robert J. Birgeneau, David J. Singh, Pengcheng Dai, and Ming Yi

1 Department of Physics and Astronomy, Rice University, Houston, Texas, USA
2 Department of Physics, University of California, Berkeley, California, USA
3 Department of Physics and Astronomy, University of Missouri, Columbia, Missouri, USA
4 Canadian Light Source Inc., University of Saskatchewan, Saskatoon, SK S7N 2V3, Canada
5 Department of Physics and Astronomy, University of British Columbia, Vancouver, BC V6T 1Z1, Canada
6 Quantum Matter Institute, University of British Columbia, Vancouver BC V6T 1Z4, Canada
7 Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany
8 Lawrence Berkeley National Laboratory, Berkeley, California, USA
9 Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory, Menlo Park, California, USA
10 Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA
11 Department of Chemistry, University of Missouri, Columbia, Missouri, USA

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The emergence of unconventional superconductivity in proximity to intertwined electronic orders is especially relevant in the case of iron-based superconductors. Such order consists of an electronic nematic order and a spin density wave in these systems. BaNi$_2$As$_2$, like its well-known iron-based analog BaFe$_2$As$_2$, also hosts a symmetry-breaking structural transition that is coupled to a unidirectional charge density wave (CDW), providing a novel platform to study intertwined orders. Here, through a systematic angle-resolved photoemission spectroscopy study combined with a detwinnning $B_1g$ 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 BaFe$_2$As$_2$, the structural and CDW order parameters in BaNi$_2$As$_2$ are observed to be strongly coupled and do not separate in the presence of uniaxial strain. Our measurements point to a likely lattice origin of the CDW in BaNi$_2$As$_2$.

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) [1,2]. The nematic order manifests in a tetragonal to orthorhombic structural transition, identified as electronically driven by a divergent nematic susceptibility from elastoresistance measurements [3]. Additionally, rotational symmetry-breaking is observed in the electronic, magnetic and optical properties [5,12]. Superconductivity emerges when these competing orders are suppressed by either doping or pressure, resulting in $T_c$ as high as 40 K [13,14].

BaNi$_2$As$_2$—a nickel-based analog of the well-studied FeSC BaFe$_2$As$_2$—shares the same high temperature tetragonal phase [15]. Distinct from BaFe$_2$As$_2$, BaNi$_2$As$_2$ is nonmagnetic and superconducts with a $T_c$ of 0.6K [16]. Instead of the coupled nematicity and SDW found in BaFe$_2$As$_2$, BaNi$_2$As$_2$ exhibits a symmetry-breaking 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 different unidirectional IC-CDW at $T_S$ and then locks into a commensurate order (C-CDW) below $T_S$ [17,18]. 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 a orthorhombic phase in a second order transition before additional symmetry-lowering into the triclinic phase at $T_S$ in a first-order fashion [19]. Substitution of either Co on the Ni site or Sr on the Ba site can completely suppress the triclinic phase and the associated CDW orders, reaching a maximum $T_c$ of 3.5 K. Ela
toresistance measurements show that Ba$_{1-x}$Sr$_x$Ni$_2$As$_2$ exhibits a diverging nematic susceptibility in the $B_{1g}$ channel, which may be the cause for the enhancement of $T_c$ near optimal doping [20]. A Ginzburg–Landau analysis suggests that the divergence of nematic susceptibility in Ba$_{1-x}$Sr$_x$Ni$_2$As$_2$ could be driven by either lattice or electronic degrees of freedom, in contrast to BaFe$_2$As$_2$, where the divergence in the $B_{2g}$ channel is electronically-driven. BaNi$_2$As$_2$ therefore offers 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 probe the electronic structure evolution of BaNi$_2$As$_2$. 

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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 $\rho_a$ (yellow), $\rho_b$ (blue) and $\rho_b/\rho_a$ (green) with and without uniaxial compressive strain along b.

across the phase transitions. We find no evidence of Fermi surface nesting in the tetragonal state, unidirectional band folding consistent with the reported C-CDW q-vector in the C-CDW/triclinic state, and rotational symmetry-breaking that onsets abruptly at the triclinic transition. Furthermore, the extracted temperature evolution of the spectral features identifies a distinct order parameter for the C-CDW from 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$_2$As$_2$ where strain lifts the nematic band splitting to onset well above that of the SDW phase. In contrast to BaFe$_2$As$_2$ [21], even in the absence of a detwinning stress, we observe resistivity anisotropy ($\rho_b - \rho_a$) 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 is likely the reported IC-CDW transition [17]. 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 [3], or strain-induced rotational symmetry-breaking [23]. It is interesting to point out that, in contrast to BaFe$_2$As$_2$, the uniaxial pressure does not broaden the structural transition [21] [24].

To visualize the electronic structure of BaNi$_2$As$_2$, we present ARPES measurements taken in the tetragonal phase. The Fermi surface maps (FSMs) and band dispersions along high symmetry directions correspond to the triclinic structure ($a=4.21\,\text{Å}$, $b=3.99\,\text{Å}$, $c=6.31\,\text{Å}$, $\alpha=105.2^\circ$, $\beta=108.6^\circ$, $\gamma=89.3^\circ$, space group P$\bar{1}$) at $T_S = 136$K (Fig. 1(b)). 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 $q_{\text{C-CDW}}$, 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 SM). We carried out temperature dependent in-plane electrical resistivity measurements along with two orthogonal directions with and without uniaxial pressure. In contrast to BaFe$_2$As$_2$ [21], even in the absence of a detwinning stress, we observe resistivity anisotropy ($\rho_b - \rho_a$) 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 is likely the reported IC-CDW transition [17]. 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 [3], or strain-induced rotational symmetry-breaking [23]. It is interesting to point out that, in contrast to BaFe$_2$As$_2$, the uniaxial pressure does not broaden the structural transition [21] [24].

At room temperature, tetragonal BaNi$_2$As$_2$ belongs to the space group I4/mmm ($a=4.142\,\text{Å}$, $c=11.65\,\text{Å}$) (Fig. 1(a)). It undergoes a first-order phase transition to the triclinic structure ($a=4.21\,\text{Å}$, $b=3.99\,\text{Å}$, $c=6.31\,\text{Å}$, $\alpha=105.2^\circ$, $\beta=108.6^\circ$, $\gamma=89.3^\circ$, space group P$\bar{1}$) at $T_S = 136$K (Fig. 1(b)). 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 $q_{\text{C-CDW}}$, 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 SM). We carried out temperature dependent in-plane electrical resistivity measurements along with two orthogonal directions with and without uniaxial pressure. In contrast to BaFe$_2$As$_2$ [21], even in the absence of a detwinning stress, we observe resistivity anisotropy ($\rho_b - \rho_a$) 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 is likely the reported IC-CDW transition [17]. 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 [3], or strain-induced rotational symmetry-breaking [23]. It is interesting to point out that, in contrast to BaFe$_2$As$_2$, the uniaxial pressure does not broaden the structural transition [21] [24].

To visualize the electronic structure of BaNi$_2$As$_2$, we present ARPES measurements taken in the tetragonal phase. The Fermi surface maps (FSMs) and band dispersions along high symmetry directions correspond to the k$_z=0$ (Fig. 2(a)-(b)) and k$_z = \pi/2$ (Fig. 2(c)-(d)) planes are taken with 60 eV and 79 eV photons, respectively (Fig. 2(e)). The C$_4$ symmetry of the tetragonal state...
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. 3(e)). Such band folding is a signature of translational symmetry breaking. We note that the folding vector is approximately $\frac{1}{3}$, which is consistent with the in-plane projection of the $q_{C-CDW}$ observed by x-ray diffraction [17, 20]. Therefore, this band folding is a signature of the unidirectional CDW, which was not observed in previous ARPES studies of BaNi$_2$As$_2$ [25].

To exclude rotational symmetry-breaking effect 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. 3). 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. 3(b)), 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. 3(c)). For comparison, the FSM of an unstrained twinned crystal (Fig. 3(d)) 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 detwinning the crystal and important for resolving the observed CDW band folding. We also note that besides the bulk bands described here, we observed a set of surface states (SS) that strongly reflects a broken C$_4$ symmetry observed in strained crystals, demonstrating the effectiveness of the applied strain (See SM). These states have no correspondence in the bulk DFT calculated bands and do not disperse with $k_z$, consistent with a surface origin.

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 the $\Gamma-X$ and $\Gamma-Y$ 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 $\Gamma-X$ but not along $\Gamma-Y$ (Fig. 4(b)-(g)). To understand the observed dispersions, we illustrate with a schematic in Fig. 4(a). 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 bro-
The presence of the folded band is seen in a hump in the MDC across the cyan band for \(\bar{\Gamma} - \bar{X}\). Second, we trace the band folding by comparing the MDCs along \(\bar{\Gamma} - \bar{X}\) but not \(\bar{\Gamma} - \bar{Y}\) (Fig. 4(i)). The contrast is also apparent in the collapsed MDC curves in Fig. 4(h). Due to the abrupt onset, we ascribe the band shift transition to the first-order structural transition into the triclinic phase. While a small kink is observed above \(T_S\) in the resistivity indicating the onset of IC-CDW and potential orthorhombic transition and nematic order, we do not observe any apparent change in the band dispersions above \(T_S\) (see SM). This does not preclude the existence of a nematic phase above \(T_S\), but rather indicates that the energy scale of the symmetry-breaking in the electronic structure associated with IC-CDW and nematic order is much smaller than that observed for the nematic phase in BaFe\(_2\)As\(_2\).

Finally, we 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. 4(i)-(j)), showing very similar behavior as those obtained on strained samples. In particular, we observe no elevation in the onset temperature of either order parameter in the warming up measurement. This is again in contrast to BaFe\(_2\)As\(_2\) (Fig. 4(k)), where the uniaxial strain clearly elevates the onset temperature of the observed orbital anisotropy and separates it from the SDW ordering temperature [27]. 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\(_2\)As\(_2\) and that nematic fluctuations are much weaker than that in BaFe\(_2\)As\(_2\). The C-CDW, from a lack of Fermi surface nesting conditions, is likely dominated by lattice. The recent DFT results show that two distinct structures corresponding to complex As bonding patterns and drive distortions of the Ni layers which possibly explain the unconventional CDW behavior of the ground state [28].

In the broader context of FeSCs, the coupling of the nematic and magnetic order varies as well. In BaFe\(_2\)As\(_2\), the structural transition is second-order, followed by a first-order magnetic transition. In the structural homolog SrFe\(_2\)As\(_2\), the structural and magnetic transitions are strongly first-order and occur simultaneously. Uniaxial strain also does little in harvesting the nematic fluctuations above \(T_S\) [24]. The case of BaNi\(_2\)As\(_2\) as we demonstrate here, appears to be similar in spirit to that of SrFe\(_2\)As\(_2\), where very weak rotational symmetry-
FIG. 4. Temperature evolution through the CDW transitions on detwinned BaNi$_2$As$_2$. (a) Summary of the band evolution across T$_{C-CDW}$. (b)-(d) Temperature dependence of band dispersions along Γ-X for (b) T < T$_{C-CDW}$, (c) T$_{C-CDW}$ < T < T$_{IC-CDW}$, and (d) T > T$_{IC-CDW}$. (e)-(g) Same as (b)-(d) but taken along Γ-Y. Dashed line 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 unstained 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) Temperature evolution of d$_{xz}$ and d$_{yz}$ band positions in BaFe$_2$As$_2$ with and without uniaxial strain (reproduced from [5]).

breaking exists above the strongly first-order structural transition. However, with substitution of P on the As site or Sr on the Ba site, nematic fluctuations appear to grow both in the size of the anisotropic thermal expansion above $T_S$ [19], as well as a diverging nematic susceptibility, which have been proposed to be responsible for the enhancement of $T_c$ [20]. BaNi$_2$As$_2$ 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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*mingyi@rice.edu

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