Nature of symmetry breaking at the excitonic insulator transition: Ta$_2$NiSe$_5$

Giacomo Mazza,1,2,∗ Malte Rösner,3,∗ Lukas Windgätter,4 Simone Latini,4 Hannes Hübener,4 Andrew J. Millis,5,6 Angel Rubio,4,5,7 and Antoine Georges2,5,1,8,†

1 CPHT, CNRS, Ecole Polytechnique, IP Paris, F-91128 Palaiseau, France
2 Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France
3 Radboud University, Institute for Molecules and Materials, Heijendaalseweg 135, 6525 AJ Nijmegen, Netherlands
4 Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany
5 Center for Computational Quantum Physics, Flatiron Institute, New York, NY 10010 USA
6 Department of Physics, Columbia University, New York, NY, 10027 USA
7 Nano-Bio Spectroscopy Group, Departamento de Física de Materiales, Universidad del País Vasco, 20018 San Sebastian, Spain
8 DQMP, Université de Genève, 2 quai Ernest Ansermet, CH-1211 Genève, Suisse

Ta$_2$NiSe$_5$ is one of the most promising materials for hosting an excitonic insulator ground state. While a number of experimental observations have been interpreted in this way, the precise nature of the symmetry breaking occurring in Ta$_2$NiSe$_5$, the electronic order parameter, and a realistic microscopic description of the transition mechanism are, however, missing. By a symmetry analysis based on first-principles calculations, we uncover the discrete lattice symmetries which are broken at the transition. We identify a purely electronic order parameter of excitonic nature that breaks these discrete crystal symmetries and also leads to the experimentally observed lattice distortion into a monoclinic phase. Our results provide a theoretical framework to understand and analyze the excitonic transition in Ta$_2$NiSe$_5$ and settle the fundamental questions about symmetry breaking governing the spontaneous formation of excitonic insulating phases in solid-state materials.

Introduction. Spontaneous symmetry breaking is a fundamental organizing principle for understanding the emergence of long-range order. Identifying the underlying symmetry breaking is thus a key step in the characterization of the ordered phase. This can be an elusive task when the symmetry breaking field cannot be directly tuned experimentally or when different types of ordering are coupled. The so-called excitonic insulator is a prominent example of such an elusive state of matter. This phase [1–4] has been identified with the spontaneous condensation of excitons (bound electron-hole pairs) stemming from the Coulomb attraction between electrons and holes in the conduction and valence bands. Excitonic condensation has been observed and intensively investigated in specially designed devices such as bilayer quantum Hall systems [5–9] or by photo stimulation of electron-hole pairs [10–12]. In contrast, spontaneous excitonic condensation in bulk materials still remains an open question and its detection a major challenge.

Ta$_2$NiSe$_5$ (TNSe) has been proposed as a candidate material hosting a homogeneous excitonic condensate [13–16], i.e. without charge or other non-zero momentum order [17–19]. TNSe undergoes a structural transition from a high-temperature orthorhombic to a low-temperature monoclinic phase at $T_s \approx 328$ K [13, 20, 21]. Proposed evidence for excitonic condensation occurring simultaneously with the structural transition includes a characteristic flattening of the valence band close to the Γ point [14, 15, 22], the opening of a gap in the electronic spectrum [16, 23, 24], and coherent oscillations reminiscent of the excitation of an amplitude mode of the condensate [25]. Due to its characteristic chain structure TNSe has so far been interpreted as a quasi one-dimensional excitonic insulator [26, 27] and Kaneko et al. [27] proposed a scenario in which the coupling of the excitonic condensate with phonons gives rise to a combined excitonic and structural instability.

Symmetry considerations, however, call into question the very notion of excitonic “condensation” in the solid-state context. In an excitonic condensate the charge is assumed to be independently conserved for both particles and holes and the spontaneous formation of a particle-hole hybridization is understood as the breaking of the corresponding continuous $U(1)$ symmetry. In contrast, conservation laws in solids are determined by internal symmetries, such as crystal symmetries or topological invariants, which in general lack such a $U(1)$ symmetry. Nonetheless, internal discrete symmetries of the solid can result in an effective realization of the particle-hole conservation which occurs only at particular points in the Brillouin zone, but might be also approximately satisfied in regions nearby. Such an effective particle-hole conservation can be broken by a spontaneous hybridization at an excitonic transition. We propose that in the solid state context an excitonic instability is a general mechanism for breaking discrete internal symmetries rather than a condensation in a $U(1)$ theory.

Here, we demonstrate this concept for the case of TNSe by uncovering the symmetries that are broken by an excitonic instability. To this end, we construct a minimal yet realistic model for TNSe including its electronic band structure and electron-electron interactions from
first principles. We show the existence of an electronic instability of excitonic origin leading to an electronic phase that breaks a set of discrete symmetries of the high-temperature orthorhombic phase, and is compatible with the low-temperature monoclinic structure. This analysis settles the fundamental question of identifying which symmetries are broken at the excitonic transition, and is therefore of general relevance to the understanding and the eventual control of such transitions in TNSe and solid-state materials in general.

Crystal symmetries in Ta$_2$NiSe$_5$. We consider the relation between crystal symmetries and electronic structure by performing DFT calculations in the high-temperature orthorhombic phase, with $a \simeq 3.51$ Å and $b \simeq 15.79$ Å being the lattice constants in $x$- and $y$-directions of the Bravais lattice. This unit-cell is composed of two formula units with atoms arranged in two parallel Ta-Ni-Ta chains (A/B) along the $z$-direction. The chains are shifted by half a lattice constant along $x$ and displaced along $z$, which results in four reflection symmetries with planes parallel and perpendicular to the Ta-Ni-Ta chains ($\sigma_{A/B}^{||L}$) and one inversion symmetry point $I$, as depicted in Fig. 1. Based on these ab initio calculations, we construct six $d_{xz}$-like maximally localized Wannier functions (MLWF) centered at the Ta and Ni positions, which are shown in Fig. 1 [28]. In each Ta-Ni-Ta chain the Ta-centered $d_{xz}$ orbitals, $\varphi_{Ta}(\vec{R})$, are aligned along the chains and tilted around the $x$-axis following the Ta-Se bonds [see Fig. 1(b)]. The Ni-centered $d_{xz}$ MLWFs, $\varphi_{Ni}(\vec{R})$, are also parallel to the Ta-Ni-Ta chains, but rotated by 45° around the $y$-axis. The Se contributions are thus indirectly accounted for by deforming and rotating the $d_{xz}$ orbitals.

The reflection symmetries act differently on the Ta and Ni-centered MLWFs. While the $\varphi_{Ta}(\vec{R})$ MLWFs are unaffected by all reflections, $\varphi_{Ni}(\vec{R})$ change sign under $\sigma_z$. It follows that the intra-chain Ta-Ta and Ni-Ni hoppings $t_{TaTa/NiNi}(\vec{R}) = \langle \varphi_{Ta/Ni}(\vec{R})|\hat{H}|\varphi_{Ta/Ni}(0) \rangle$ have opposite signs. We find $t(\vec{0})_{TaTa} \approx -640$ meV and $t(\vec{0})_{NiNi} \approx 250$ meV, which are respectively mainly responsible for the conduction and valence bands dispersions in the $M\bar{Z}$ and $\Gamma X$ directions of the Brillouin zone. These bands are about 2.5 eV and 1.5 eV wide with predominant Ta (blue) and Ni (red) character as visible in the Wannier-interpolated band structure in Fig. 2. Conduction and valence bands overlap along $\bar{Z}\Gamma$ where the bands become much less dispersive and are mainly characterized by bonding/anti-bonding splittings of the Ta and Ni states between the two Ta-Ni-Ta chains.

Importantly, hopping matrix elements between Ta- and Ni-like states within the chains are not forbidden by symmetry. The corresponding hybridization has so far been interpreted as the order parameter of a quasi one-dimensional excitonic phase [26, 27, 29]. However, even in the orthorhombic phase and without any excitonic amplitudes, we obtain non-zero matrix elements between Ta- and Ni-MLWFs within the same chain $t_{TaNi}(\vec{R})$. Specifically, $t_{TaNi}(\vec{0}) \approx 36$ meV which decreases with the
distance \( R_x \) along the chains. This result is universal to all tested DFT exchange-correlation functionals [28] and shows that a spontaneous hybridization of this kind is not a valid order parameter for the excitonic transition.

However, reflection symmetries \( \sigma_{\perp}^{A/B} \) constrain the Ta-Ni hoppings which change sign under \( \sigma_{\perp}^{A/B} \). For example, for the A chain, we get \( \sigma_{\perp}^{A}(t_{\text{Ta-Ni}}(\vec{R}_x, R_y)) = -t_{\text{Ta-Ni}}(-R_x + a, R_y) \). These constraints imply that the Wannier Hamiltonian averaged along the \( x \)-direction is block-diagonal with respect to the Ta- and Ni- states

\[
\hat{H}(k_x = 0, R_y) = \sum_{R_x} \hat{H}(R_x, R_y) = \begin{pmatrix} \hat{H}_{\text{Ta}}(R_y) & 0 \\ 0 & \hat{H}_{\text{Ni}}(R_y) \end{pmatrix}.
\]

In momentum space, this means that \( t_{\text{Ta-Ni}}(k_x = 0, k_y = 0) \) so that the bands along the \( ZT \) path have purely Ta or Ni character, as shown in Fig. 2. Therefore, any excitonic instability resulting from a spontaneous Ta-Ni hybridization must lower the above symmetry and show-up along the \( ZT \) direction. We provide evidence of such an instability by considering the effect of the electron interactions in a minimal model derived from the above analysis.

**Minimal Model.** We consider a two-dimensional lattice with six atoms per unit cell. The electronic Hamiltonian

\[
\hat{H} = \hat{H}_{\text{hop}} + \hat{H}_U + \hat{H}_V
\]

includes a hopping term \( \hat{H}_{\text{hop}} \), a local \( \hat{H}_U \) Coulomb interaction term, and nearest-neighbor \( \hat{H}_V \) one. We define \( \psi_{\vec{R}\sigma}^\dagger \equiv (c_{1\sigma}(\vec{R}) \ldots c_{6\sigma}(\vec{R})) \), where \( c_{j\sigma}(\vec{R}) \) creates an electron with spin \( \sigma \) in a localized orbital on the \( j \)-th atom (labels in Fig. 1) of the unit cell \( \vec{R} \).

\[
\hat{H}_{\text{hop}} = \sum_{\vec{R}\sigma} \sum_{j} \psi_{\vec{R}\sigma}^\dagger \mathbf{T}(\vec{d}) \psi_{\vec{R}\sigma}
\]

contains intra-cell, \( \mathbf{T}(\vec{0}) \), as well as inter-cell, \( \mathbf{T}(\pm a, \pm b) \), terms. The matrix elements are chosen consistently with the above symmetry requirement and in order to reproduce the main features of the Wannier band structure [28].

The electrons interact through a local Hubbard-like term

\[
\hat{H}_U = U \sum_{\vec{R}} \sum_{j} \hat{n}_{j+}(\vec{R}) \hat{n}_{j-}(\vec{R}),
\]

where we assumed the same \( U \) for the six atoms, a choice supported by a constrained RPA [30] analysis of the Coulomb matrix elements \( (U_{\text{Ta}} \approx 2.1 \text{ eV}, U_{\text{Ni}} \approx 2.4 \text{ eV}) \). The next leading terms are intra-chain density-density interactions between neighbouring Ta and Ni atoms \( (V \approx 0.9 \text{ eV}) \)

\[
\hat{H}_V = V \sum_{j=1,2,3,4} \sum_{\vec{R}\sigma\sigma'} \left[ \hat{n}_{j\sigma}(\vec{R}) + \hat{n}_{j\sigma}(\vec{R} + \vec{d}_x) \right] \hat{n}_{5\sigma'}(\vec{R}) + V \sum_{j=1,2,3,4} \sum_{\vec{R}\sigma\sigma'} \left[ \hat{n}_{j\sigma}(\vec{R}) + \hat{n}_{j\sigma}(\vec{R} + \vec{d}_y) \right] \hat{n}_{6\sigma'}(\vec{R}).
\]

The symmetries of the Hamiltonian are revealed by an investigation of the intra-chain Ta-Ni hybridization as a function of the distance along \( x \) between the Ta and Ni atoms

\[
\Delta_{ij}(x) = \langle c_i^\dagger(\vec{R}_x, 0) c_j(0, 0) \rangle,
\]

where \( i = 1, 2, 3, 4 \) and \( j = 5, 6 \) for the A(B) chain. For each chain, \( x \) is defined by taking the Ni atom in that chain as origin, so that \( x = R_x + a/2 \) (for A and + for B). We have dropped the spin index as we focus on the spin-singlet case. For the A-chain, \( \Delta_{15}(x) \) and \( \Delta_{25}(x) \) transform as

\[
\sigma_{\perp}^{A} \Delta_{15}(x) = -\Delta_{15}(-x) \quad \sigma_{\perp}^{A} \Delta_{25}(x) = \Delta_{25}(x)
\]

so that \( \Delta_{15}(x) = -\Delta_{15}(-x) \) and \( \Delta_{15}(x) = \Delta_{25}(x) \), as depicted in Fig. 3(a). Similarly, reflection symmetries for the B-chains imply \( \Delta_{36}(x) = -\Delta_{36}(-x) \) and \( \Delta_{46}(x) = -\Delta_{46}(-x) \).

We investigate the possible breaking of the above symmetries due to electronic interactions, by utilizing a Hartree-Fock (HF) variational wave-function allowing for a non-zero homogeneous order parameter of the form:

\[
\phi = \begin{pmatrix} \phi_{15} \\ \phi_{25} \\ \phi_{36} \\ \phi_{46} \end{pmatrix}, \quad \phi_{ij} \equiv \Delta_{ij}(a/2) + \Delta_{ij}(-a/2).
\]

The four \( \phi_{ij} \) are in general independent, allowing in principle for 16 different phases corresponding to the different
breaking patterns of the reflections and inversion symmetries. Here, we focus on the symmetry breaking channel consistent with the low-temperature monoclinic phase of TNSe. In the monoclinic phase all reflections are broken, while their products $I_{A/B} = \sigma_{\perp}^{A/B} \sigma_{\parallel}^{A/B}$ and the inversion $I = I_{A/B} T$ ($T$ being the translation between the two Ni atoms) are preserved. This constrains the components $\phi_{ij}$ as $\phi_{15} = -\phi_{25}$ and $\phi_{36} = -\phi_{46}$, due to preservation of $I_{A/B} T$ and for $I_{A/B} T$ being translated by $\phi_{15} = \phi_{46}$ and $\phi_{25} = \phi_{36}$ due to $I$, leading to an order parameter of the form $\vec{\phi} = \phi_{0}(+1, -1, -1, +1)$.

The obtained zero-temperature phase diagram in the $U$-$V$ plane, Fig. 4(a), shows three distinct regions. At fixed value of $U$, the order parameter $\phi_{0}$ vanishes for $V$ smaller than a lower critical value [$V < V_{c}^{*}(U)$] and for $V$ larger than an upper critical value [$V > V_{c}^{*}(U)$]. In these regions the Ta-Ni hybridizations transform in accordance with Eq. (7), as shown in Fig. 4(b) and (d). These two symmetric ground states are characterized by different electronic properties. For $V < V_{c}^{*}(U)$ [Fig. 4(b) and (g)] the valence and conduction bands overlap, while for $V > V_{c}^{*}(U)$ [Fig. 4(d) and (e)] the bands are separated by an energy gap.

In the intermediate region $V_{c}^{*}(U) < V < V_{c}^{*}(U)$ [Fig. 4(c) and (f)] a solution with $\phi_{0} \neq 0$ is stabilized. This is the hallmark of the excitonic instability as witnessed in Fig. 4(f) by the emergence of a sizable hybridization between valence and conduction bands all along the $\overline{ZT}$ path. Valence and conduction bands acquire a strong Ta and Ni character respectively (which is absent in the symmetric phase) and the degeneracy of the Ta-like conduction bands along $\overline{ZT}$ is lifted by hybridization with Ni-like valence bands. In real space this translates into the emergence of otherwise symmetry forbidden Ta-Ni hybridizations, $\Delta_{35}$ and $\Delta_{26}$, which couple the two chains, Fig. 3(b).

The upper valence band develops a mostly flat dispersion around $\Gamma$. While this has usually been interpreted as a distinctive signature of an homogeneous excitonic condensate, we show here that the interpretation is not unique. In fact this feature is a result of a direct-to-indirect gap insulator transition, driven by the splitting between the hybridized bands along $\overline{ZT}$, that can occur inside the broken-symmetry phase [Fig. 4(h)]. By decreasing $V$ the splitting increases, while the bottom of the conduction band moves closer to the Fermi level. At $V = V_{c}^{*}(U)$ the conduction band crosses the Fermi level and the system undergoes a Lifshitz transition accompanied by the formation of a Fermi surface in the metallic phase. We find that this is a first-order transition which restores the symmetry for $V < V_{c}^{*}(U)$. The symmetry-broken phase is found only in a small region of the phase space close to the Lifshitz transition. In this regime the symmetric phase is characterized by a very small gap, reinforcing the relevance of the above phase transition for TNSe which in the high-temperature phase

FIG. 4. (a) Order parameter at zero-temperature as function of $V$ for $U = 2.50$ eV. Dots represent $V = 0.73$, $V = 0.785$ and $V = 0.83$ corresponding to panels (b)-(d). Inset: phase diagram in $U$–$V$ plane. Shaded region corresponds to the symmetry-broken phase. Dashed line indicates a metal-insulator Lifshitz transition. (b)-(d): Ta-Ni hybridization along Ta-Ni-Ta A chain in the symmetric (b) and (d) and symmetry-broken (c) phases. Crosses/diamonds correspond to upper/lower part of the Ta-Ni-Ta chain. (e)-(g): Bands along $M$–$Z$–$\Gamma$–$X$ corresponding to panels (b)-(d). Red/blue corresponds to Ni/Ta character. (h): Gap evolution inside the broken-symmetry phase. $U = 2.50$ eV and $V = 0.79, 0.77, 0.75$ eV from light grey to black lines. Full/dashed green arrows highlight the direct/indirect gap.
has been reported to be a zero-gap semiconductor [16].
The symmetry-broken region shrinks as the Hubbard $U$
is decreased until it disappears for $U \lesssim 1.25$ eV for which
$V^\ast_s(U)$ and $V^\ast_s(U)$ merge into the Lifshitz transition line.
We highlight that our constrained RPA values for $U$ and $V$
in close vicinity of the symmetry-broken region.

**Structural phase transition.** The electronic configuration
associated with the excitonic phase is not compatible with the symmetries of the lattice. This implies
that the electronic order parameter must have a linear coupling to lattice modes breaking the crystal symmetry [27, 31]. Hence, the excitonic transition will co-exist with a structural transition, as indeed observed experimentally. From the pattern of the Ta-Ni hybridizations in the broken-symmetry phase [Fig. 3(b)], one anticipates a distortion of the unit cell in which Ta atoms from the same Ta-Ni-Ta chain are tilted in opposite directions [arrows in Fig. 3 (b)]. This corresponds to a structural transition from the orthorhombic to the monoclinic structure, which we confirm to be present by performing a full structural relaxation within DFT [28]. The interplay between the electronic and lattice instability is an interesting question for future investigations.

**Conclusions.** We have performed a symmetry analysis backed up by first-principle calculations of Ta$_2$NiSe$_5$, which has general implications for the excitonic transitions in solids. While the conventional excitonic-insulator model has a spontaneously broken continuous $U(1)$ symmetry, we show that in realistic solids there is no such symmetry: the electronic order parameter corresponds to the breaking of discrete symmetries only. Important consequences include that all collective modes are gapped and that there is no dissipationless transport or excitonic superfluidity. We identify explicitly all discrete symmetries relevant to TNSe, including the corresponding electronic order parameters and provide clear evidence for a transition into an excitonic insulator phase. This transition breaks these symmetries in a manner consistent with the experimentally observed lattice distortion into a monoclinic phase, and the order parameter couples linearly to lattice modes.

Because we find a spontaneous electronic instability for realistic values of the interactions, our results strongly suggest that the driving force behind the coupled transition is electronic in nature. However, a definitive confirmation of this point calls for experimental probes which can selectively address the electronic and lattice degrees of freedom. In the context of iron-based superconductors, where a similar question arises in relation to nematicity, it has proven possible to probe the electronic component of the susceptibility associated with the nematic instability [32, 33]. Ultra-fast spectroscopies offer another possible route [22, 25, 34, 35], by exploiting the very different time scales associated with electronic and lattice degrees of freedom.

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Supplemental information for:
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Giacomo Mazza,1,2,* Malte Rösnner,3,* Lukas Windgätter,4 Simone Latini,4 Hannes Hübener,4 Andrew J. Millis,5,6 Angel Rubio,4,5,7 and Antoine Georges2,5,1,8,†

1 CPHT, CNRS, Ecole Polytechnique, IP Paris, F-91128 Palaiseau, France
2 Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France
3 Radboud University, Institute for Molecules and Materials, Heijendaalseweg 135, 6525 AJ Nijmegen, Netherlands
4 Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany
5 Center for Computational Quantum Physics, Flatiron Institute, New York, NY 10010 USA
6 Department of Physics, Columbia University, New York, NY, 10027 USA
7 Nano-Bio Spectroscopy Group, Departamento de Física de Materiales, Universidad del País Vasco, 20018 San Sebastian, Spain
8 DQMP, Université de Genève, 24 quai Ernest Ansermet, CH-1211 Genève, Suisse

I. AB-INITIO CALCULATIONS

Our ab-initio calculations are performed using density functional theory (DFT) initially applying the generalized gradient approximation (GGA / PBE)\textsuperscript{1} within the PAW formalism\textsuperscript{2} as implemented in the Vienna Ab initio Simulation Package (VASP)\textsuperscript{3,4}. We start with fully relaxing the internal atomic coordinates of an orthorhombic unit cell with $a \approx 3.51$ Å, $b \approx 14.07$ Å, and $c \approx 15.79$ Å as lattice constants. To this end we use a $12 \times 12 \times 3$ K-grid and an energy cut-off of 368 eV. The positions are optimized until all forces are smaller than 0.005 eV/Å.

Due to the layered structure, screening is reduced so that enhanced Coulomb interactions are expected. To take the resulting correlation effects into account, we use the modified Becke-Johnson exchange potential\textsuperscript{5}, which has been shown to have a similar accuracy as hybrid functional or $GW$ approaches\textsuperscript{6}. The involved $c\text{mbj}$ parameter is self-consistently find to be $c\text{mbj} = 1.26$ on a $20 \times 20 \times 5$ K-grid.

The resulting Kohn-Sham states are subsequently projected onto six $d_{xz}$-like Wannier orbitals centered at the Ta and Ni sites, which are maximally localized using the Wannier90 package\textsuperscript{7} applying an inner (frozen) window of about $\pm 0.3$ eV around the Fermi level. Thereby, the overlap between the original Kohn-Sham states and the reconstructed ones is maximized throughout the low-energy window.

These six maximally localized Wannier functions are also used as the basis for the evaluation of the Coulomb matrix elements calculated within the constrained Random Phase Approximation (cRPA)\textsuperscript{8} as recently implemented by M. Kaltak within VASP\textsuperscript{9}. We use in total 120 bands (about 50 unoccupied) and apply the weighted disentanglement procedure from Ref.\textsuperscript{10}.

II. STRUCTURAL PHASE TRANSITION

Starting from the relaxed orthorhombic geometry we introduce a small distortion to $\beta = 90^\circ + \delta$ (see figure 1) to seed the monoclinic phase and perform a full relaxation allowing for an optimization of the cell shape, cell-volume and atomic coordinates afterwards. To this end we use a $24 \times 16 \times 8$ K-grid and the PBE (GGA) functional. As a result we find distorted angles of $\alpha = 90.150^\circ$ and $\beta = 90.571^\circ$, yielding a triclinic structure (changes to the lattice constants are negligible). This corresponds to an in-plane monoclinic distortion combined with a tilting in the direction perpendicular to the planes. While the inter-layer geometry might suffer from neglected van-der-Waals forces, the in-plane structure is mostly governed by electron-lattice couplings which are sufficiently well captured by DFT. The in-plane monoclinic distortion is thus reliable and intrinsically driven already on the level of DFT.

III. MINIMAL MODEL

We consider a two-dimensional minimal model with six atoms per unit cell (with one $d_{xz}$-like orbital each) reproducing the double chain structure of a TNSe layer. We take into account i) single particle hoppings, ii) intra-atomic density-density interactions, and iii) nearest-neighbor density-density interactions. For simplicity we recall here the definition
of the Hamiltonian which we already introduced in the main text:

$$\hat{H} = \hat{H}_{hop} + \hat{H}_{U} + \hat{H}_{V}$$

$$= \sum_{\vec{R}_{\sigma}} \sum_{\delta} \Psi_{\vec{R}_{\sigma}}^{\dagger} \tilde{T}(\delta) \Psi_{\vec{R}_{\sigma}} + U \sum_{\vec{R}} \sum_{j=1,...,6} \hat{n}_{j\uparrow}(\vec{R}) \hat{n}_{j\downarrow}(\vec{R}) +$$

$$+ V \sum_{j=1,2} \sum_{\vec{R}_{\sigma}\sigma'} \left[ \hat{n}_{j\sigma}(\vec{R}) + \hat{n}_{j\sigma}(\vec{R} + \delta_{x}) \right] \hat{n}_{5\sigma'}(\vec{R}) + V \sum_{j=3,4} \sum_{\vec{R}_{\sigma}\sigma'} \left[ \hat{n}_{j\sigma}(\vec{R}) + \hat{n}_{j\sigma}(\vec{R} - \delta_{x}) \right] \hat{n}_{6\sigma'}(\vec{R}),$$

with $\Psi_{\vec{R}_{\sigma}}$ a spinor defined as $\Psi_{\vec{R}_{\sigma}} = \left( c_{1\sigma}^{\dagger}(\vec{R}) \ c_{2\sigma}(\vec{R}) \ c_{3\sigma}(\vec{R}) \ c_{4\sigma}(\vec{R}) \ c_{5\sigma}(\vec{R}) \ c_{6\sigma}(\vec{R}) \right)$. The hopping matrix $\tilde{T}(\delta)$ contains intra-cell $[\tilde{T}(0)]$ as well as nearest-cells terms $[\tilde{T}(\pm a_{x}, \pm a_{y})]$ corresponding to the main contributions of the Wannier Hamiltonian derived above. These matrix elements are summarized in the scheme of Fig. 2(A), which includes Ta-Ta (a) and Ni-Ni (b) intra-chain, Ta-Ni intra- (c)-(d) and inter-chain (e)-(f) hoppings as well as inter-chain Ni-Ni (g) and Ta-Ta (j)-(h) hoppings. Dashed/full pairs of arrows indicate that in order to preserve the symmetry, these matrix elements must be anti-symmetric under a reflection with respect to a plane perpendicular to the chains. We have also indicated symmetry-forbidden Ta-Ni hybridization, that become non-zero upon symmetry breaking. The matrix elements are summarized in the Table I. Fig. 2(B) shows the comparison between the band structure of the minimal and the Wannier model.

### A. Hartree-Fock

We consider a single-particle variational wavefunction $|\Psi_{0}\rangle$ that allows for the breaking of the crystal symmetries. The variational energy is computed by decoupling the interaction terms in the standard way:

$$\langle \Psi_{0}|\hat{n}_{j\uparrow}(\vec{R})\hat{n}_{j\downarrow}(\vec{R})|\Psi_{0}\rangle \approx \langle \Psi_{0}|\hat{n}_{j\uparrow}(\vec{R})|\Psi_{0}\rangle \langle \Psi_{0}|\hat{n}_{j\downarrow}(\vec{R})|\Psi_{0}\rangle$$

(2)

and for $i \neq j$

$$\langle \Psi_{0}|\hat{n}_{i\sigma}(\vec{R})\hat{n}_{i\sigma'}(\vec{R}')|\Psi_{0}\rangle \approx \langle \Psi_{0}|\hat{n}_{i\sigma}(\vec{R})|\Psi_{0}\rangle \langle \Psi_{0}|\hat{n}_{i\sigma'}(\vec{R}')|\Psi_{0}\rangle - \delta_{\sigma\sigma'} \langle \Psi_{0}|c_{j\sigma}(\vec{R})c_{i\sigma'}(\vec{R}')|\Psi_{0}\rangle \langle \Psi_{0}|c_{j\sigma}(\vec{R})c_{i\sigma'}(\vec{R}')|\Psi_{0}\rangle.$$  

(3)
Intra-chain Ta-Ta hopping (a)-(b) $T_{ii}(a_x, 0) = T_{ii}(-a_x, 0) = -0.72 \text{ eV}$ \quad i = 1, \ldots, 4
\quad T_{ii}(0, 0) = 1.35 \text{ eV}

Intra-chain Ni-Ni hopping (a)-(b) $T_{ii}(a_x, 0) = T_{ii}(-a_x, 0) = 0.30 \text{ eV}$ \quad i = 5, 6
\quad T_{ii}(0, 0) = -0.36 \text{ eV}

Intra-chain Ta-Ta hopping (c)-(d) $T_{15}(0) = -T_{15}(a_x, 0) = T_{25}(0) = -T_{25}(a_x, 0) = 0.035 \text{ eV}$
$T_{36}(0) = T_{46}(0) = -T_{15}(0)$

Inter-chain Ta-Ni hopping (e)-(f) $T_{15}(-a_x, a_y) = -T_{45}(a_x, a_y) = T_{35}(a_x, 0) = -T_{35}(-a_x, 0) = 0.04 \text{ eV}$
$T_{26}(a_x, 0) = T_{16}(a_x, -a_y) = T_{45}(a_x, a_y)$

Inter-chain Ni-Ni hopping (g) $T_{65}(a_x, a_y) = T_{65}(a_x, 0) = T_{65}(0, 0) = T_{65}(a_x, 0) = 0.030 \text{ eV}$

Inter-chain Ta-Ta hopping (h)-(i) $T_{61}(-a_x, a_y) = T_{61}(0, a_y) = T_{23}(0, 0) = T_{23}(a_x, 0) = 0.020 \text{ eV}$

Table I. Elements of the hopping matrix $T(\tilde{\delta})$. Matrix elements are grouped accordingly to the scheme in Fig. 2 with letters corresponding to the different hopping processes indicated by arrows.

Taking the variation with respect to $|\Psi_0\rangle$ the HF Hamiltonian reads

$$\hat{H}_{HF} = \hat{H}_{\text{hop}} + \sum_{k\sigma} \Psi_{k\sigma}^\dagger \left( \begin{array}{cc} \hat{h}_A(k) & 0 \\ 0 & \hat{h}_B(k) \end{array} \right) \Psi_{k\sigma}$$  \hspace{1cm} (4)

where $\hat{h}_A(k)$ and $\hat{h}_B(k)$ are the decoupled interaction Hamiltonian for the A and B chain respectively. Specifically, accordingly to the atom labeling of Fig. 2,

$$\hat{h}_A(k) = \begin{pmatrix} \delta \varepsilon_1 & 0 & w_{15}^*(k) \\ 0 & \delta \varepsilon_2 & w_{25}^*(k) \\ w_{15}(k) & w_{25}(k) & \delta \varepsilon_5 \end{pmatrix} \quad \hat{h}_B(k) = \begin{pmatrix} \delta \varepsilon_1 & 0 & w_{36}^*(k) \\ 0 & \delta \varepsilon_4 & w_{46}^*(k) \\ w_{36}(k) & w_{46}(k) & \delta \varepsilon_6 \end{pmatrix}$$  \hspace{1cm} (5)

with

$$\delta \varepsilon_{i=1,2} = \frac{U}{2} n_i + 2V n_5 \quad \delta \varepsilon_{i=3,4} = \frac{U}{2} n_i + 2V n_6 \quad \delta \varepsilon_5 = \frac{U}{2} n_5 + 2V (n_1 + n_2) \quad \delta \varepsilon_6 = \frac{U}{2} n_6 + 2V (n_3 + n_4)$$  \hspace{1cm} (6)

and

$$w_{15} = -V \Delta_{15}(\vec{0})(1 - e^{-i k_x a}) - V \phi_{15} e^{-i k_x a} \quad w_{36} = -V \Delta_{36}(\vec{0})(1 - e^{i k_x a}) - V \phi_{36} e^{i k_x a}.$$  \hspace{1cm} (7)

In these equations $n_i = \langle \Psi_0 | c_{i\sigma}^\dagger(\vec{0}) c_{i\sigma}(\vec{0}) | \Psi_0 \rangle$, $\Delta_{ij}(\vec{0}) = \langle \Psi_0 | c_{i\sigma}^\dagger(\vec{0}) c_{j\sigma}(\vec{0}) | \Psi_0 \rangle$ and $\phi_{ij}$ are the order parameters defined in the main text. All the above parameters are self-consistently determined by diagonalizing the HF Hamiltonian.
starting from an initial guess.

* These two authors equally contributed
giacomo.mazza@polytechnique.edu
m.roesner@science.ru.nl
† ageorges@flatironinstitute.org

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