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Yu He (✉ yu.he@yale.edu)

Yale University <https://orcid.org/0000-0003-0425-4529>

Cheng Chen

University of Oxford

Xiang Chen

University of California, Berkeley <https://orcid.org/0000-0003-3997-8148>

Weichen Tang

University of California, Berkeley

Zhenglu Li

University of California, Berkeley <https://orcid.org/0000-0002-3851-9241>

Siqi Wang

Yale University

Suhan Ding

Clemson University

Chris Jozwiak

Advanced Light Source, Lawrence Berkeley National Laboratory <https://orcid.org/0000-0002-0980-3753>

Aaron Bostwick

Lawrence Berkeley National Laboratory <https://orcid.org/0000-0002-9008-2980>

Eli Rotenberg

E. O. Lawrence Berkeley National Laboratory <https://orcid.org/0000-0002-3979-8844>

Makoto Hashimoto

SLAC National Accelerator Laboratory <https://orcid.org/0000-0003-1689-8997>

Donghui Lu

SLAC National Accelerator Laboratory <https://orcid.org/0000-0002-9708-0443>

Jacob Ruff

Cornell University

Steven Louie

University of California, Berkeley <https://orcid.org/0000-0003-0622-0170>

Robert Birgeneau

University of California, Berkeley <https://orcid.org/0000-0003-1192-8333>

Yulin Chen

University of Oxford <https://orcid.org/0000-0001-8960-9725>

Yao Wang

Clemson University <https://orcid.org/0000-0003-1736-0187>

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Lattice fluctuation induced pseudogap in quasi-one-dimensional Ta_2NiSe_5

Cheng Chen^{1,2*}, Xiang Chen^{3,4*}, Weichen Tang³, Zhenglu Li^{3,4}, Siqi Wang², Shuhan Ding⁵, Chris Jozwiak⁶, Aaron Bostwick⁶, Eli Rotenberg⁶, Makoto Hashimoto⁷, Donghui Lu⁷, Jacob P. C. Ruff⁸, Steven G. Louie^{3,4}, Robert Birgeneau^{3,4}, Yulin Chen¹, Yao Wang^{5†}, Yu He^{2,3,4†}

¹*Department of Physics, University of Oxford, Oxford, OX1 3PU, United Kingdom*

²*Department of Applied Physics, Yale University, New Haven, Connecticut, 06511, USA*

³*Physics Department, University of California, Berkeley, California 94720, USA*

⁴*Materials Science Division, Lawrence Berkeley National Lab, Berkeley, California 94720, USA*

⁵*Department of Physics and Astronomy, Clemson University, Clemson, South Carolina 29631, USA*

⁶*Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*

⁷*Stanford Synchrotron Radiation Lightsources, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA*

⁸*CHESS, Cornell University, Ithaca, NY 14853, USA*

*These authors contributed equally to this work

Correspondence to: ‡yaowang@g.clemson.edu †yu.he@yale.edu

Abstract:

In conventional solid-state systems, the development of an energy gap is often associated with a broken symmetry. However, strongly correlated materials can exhibit energy gaps without any global symmetry breaking -- the so-called pseudogap, most notably in the Mott insulating state¹ and the fluctuating superconducting or charge density wave states²⁻³. To date, lattice induced pseudogap remains elusive. With angle-resolved photoemission spectroscopy (ARPES) and single crystal x-ray diffraction, we identify a pseudogap in the quasi-1D excitonic insulator candidate Ta_2NiSe_5 . Strong lattice contribution is revealed by the pervasive diffuse scattering well above the transition temperature and the negative electronic compressibility in the pseudogap state. Combining first-principles and microscopic model calculations, we show that inter-band electron-phonon coupling can create fluctuating phonon-mediated electron-hole pairing or hybridization. This suppresses the spectral weight on the Fermi surface, causing a metal-to-insulator-like transition without breaking the global symmetry. Our work establishes the precedence of a pseudogap with a lattice origin, highlighting Ta_2NiSe_5 as a room-temperature platform to study lattice-induced charge localization and low dimensional fluctuations.

Main:

In mean-field theory, a second-order phase transition is accompanied by spontaneous symmetry breaking. Informed by systems' symmetry, the phase transition of a macroscopic system with $\sim 10^{23}$ particles can be described by only a few parameters, namely the order parameters⁴. In many electronic transitions, an energy gap can be used as a liaison to the order parameter (Fig. 1(a), grey line)⁵⁻⁶. However, energy gaps can also occur without breaking any global symmetry^{2-3,7-8}, most notably the enigmatic “pseudogap” in the high- T_c cuprate superconductors due to strong Coulomb interactions or superconducting fluctuations⁹⁻¹¹. These non-symmetry-breaking gaps are believed to originate from strong electronic correlations and the lattice contribution is considered at most indirect¹²⁻¹⁴. Non-symmetry-breaking gaps are also observed in low-dimensional charge density wave (CDW) systems, where fluctuating electron density modulations arise at a given spatial frequency¹⁵⁻¹⁸. However, it remains unclear if lattice fluctuation itself is sufficient to form localized electron-hole pairs and drive a metal-to-insulator transition (MIT) without breaking the global symmetry.

In the pursuit of a potential room-temperature Bose-Einstein condensate, recent studies revealed evidence for exciton formation, strong lattice instability and electron-phonon coupling in quasi-1D ternary chalcogenide Ta_2NiSe_5 ¹⁹⁻²². Upon warming, a $q = 0$ monoclinic-to-orthorhombic structural transition happens at $T_s \sim 329\text{K}$, above which a semi-metallic electronic structure is supposedly restored and protected by the mirror symmetry of Ta chains about the Ni chain²³. However, an insulating behaviour persists up to 550K ¹⁹. This is in striking contrast to archetypal MIT systems such as the perovskite nickelates²⁴ and chain compound TTF-TCNQ²⁵, where the structural normal state often occurs in sync with the metallic electronic normal state. To date, the nature of the high-temperature electronic state in Ta_2NiSe_5 remains controversial²⁶⁻³¹. In addition, contentions remain regarding whether the system's ground state hosts an exciton condensate³²⁻³⁷. To examine possible lattice fluctuation induced insulating states and resolve the controversy, we systematically investigate the system's electronic and lattice degrees of freedom, tuned across the transition via both thermal and nonthermal methods.

High-temperature lattice instability:

Ta_2NiSe_5 crystallizes in a layered structure stacked with van der Waals interactions. Within each layer, the Ta and Ni atoms form a chain structure along the a -axis of the crystal (Fig. 1(c), inset). The system undergoes a structural phase transition from a high-temperature orthorhombic $Cmcm$ phase to a low-temperature monoclinic $C2/c$ phase at $T_s \sim 329\text{K}$ ¹⁹, indicated in both resistivity

and specific heat measurements (Fig. 1(b)), consistent with previous reports³⁴. Crossing the transition, the Ta atoms slightly shear along the chain direction, resulting in an increase of the β angle from 90° to 90.53° (as exaggerated in Fig 1(c))¹⁹. The second order nature of the transition is indicated by the gradual separation of $|Q(1\ 3\ 4)|$ and $|Q(-1\ 3\ 4)|$ peaks measured by the high-resolution synchrotron X-ray diffraction (Fig.1(c) and Extended Data Fig.1(a)). In contrast, a dramatically enhanced thermal diffuse scattering signal is observed at and above T_s (Fig. 1(d)), indicating the presence of extensive phonon softening. Strong lattice fluctuation is also reflected by a pronounced decrease of the $(2\ 0\ 0)$ peak intensity (Extended Data Fig. 1(b)) in a broad temperature range above T_s , which is mostly affected by the squared average of the atomic displacement along the chain direction. The observed strong lattice fluctuation above T_s aligns with the presence of soft transverse acoustic³⁸ and anharmonic optical modes²⁰⁻²².

High-temperature pseudogap state:

Given the persistent insulating behaviour above T_s , we investigate the electronic structure with high-resolution ARPES along the Γ -X high symmetry direction (geometry in Extended Data Fig. 2) across the transition temperature. Here, the high statistics and energy resolution enable spectra restoration up to ~ 150 meV above the Fermi level (E_F) after resolution-convolved Fermi-function division¹¹. As illustrated in Fig. 2(b), the low temperature spectra ($T < T_s$) show a pronounced single-particle gap, where the dispersion and orbital composition are consistent with density functional theory (DFT) calculations (Fig. 2(a)). In contrast, the high-temperature spectra ($T > T_s$) deviate from the calculated band structure in that a pronounced spectral weight depletion is observed around E_F (± 100 meV) despite the disappearance of the low temperature flat valence band top (Fig. 2(b) and Extended Data Fig. 3(a-b)). Such a strong intensity depletion cannot be addressed in the generic band theory: drastic orbital character change alone cannot account for the missing spectral weight in both Ta and Ni states, evidenced in both photon polarization channels. The optical matrix-element effect is also unlikely given the abruptness of the intensity drop in momentum space, as well as robust spectral weight depletion seen across a broad range of photon energies and Brillouin zones (Extended Data Fig. 3(c-d)). Such persistent spectral depletion at E_F naturally accounts for the insulating behaviour in resistivity and optical conductivity above T_s .

Figure 2(c) and 2(d) plot the energy distribution curves (EDCs) at the Fermi momentum (k_F), along with the peak energy positions and the zero-energy spectral weight. On the one hand, the structure-related band reconstruction at non-zero binding energy tracks the order parameter within a relatively small temperature range below T_s (Fig. 2(d))¹⁹. On the other hand, the zero-energy spectral weight evolves continuously from 120K to 380K with little influence from the

structural transition. Clearly, a much higher temperature is needed to fully restore the expected spectral weight of a gapless electronic normal state, consistent with resistivity data where metallicity is only recovered above 550K¹⁹. This single-particle “gapped” state does not require symmetry reduction of the orthorhombic lattice structure, and is analogous to the pseudogap found in cuprates. Its occurrence correlates with the pronounced lattice fluctuations observed in the same temperature range.

Low-temperature carrier doping dependence:

While the link between lattice fluctuations and the high-temperature pseudogap is established above T_s , the role of the lattice in the broken-symmetry ground state requires nonthermal tuning to clarify. Pump-probe experiments have achieved higher electronic or lattice temperatures, but contentions remain as to which channel determines the eventual restoration of metallicity²⁶⁻²⁹. Recently, surface potassium dosing is shown to be an effective nonthermal method to restore the gapless state at low temperatures³⁹⁻⁴⁰. Through the combined use of a micron-spot synchrotron ARPES and *in-situ* potassium dosing in the low temperature broken-symmetry state, we achieved an unprecedented level of quantification that is necessary to examine the thermodynamic stability of the ground-state electronic subsystem. Here, the relative potassium dosage is accessed through the core-level X-ray photoelectron spectroscopy (XPS) of K $3p$ orbital, and the electron doping per unit cell, is determined by the Luttinger volume at different dosing levels (Figs. 3(b)(c), Extended Data Fig. 5). Enabled by these improvements, a three-stage isothermal spectral evolution, which takes striking resemblance to the thermal phase transition, is revealed across the dosing-tuned MIT (Fig. 3(a) and Extended Data Fig. 4). First, the valence band rapidly downshifts upon dosing (shaded in blue). A pseudogap concurrently appears at E_F , similar to the spectra right above T_s . At the second stage (shaded in green), the energy position of the electron bands remains almost unchanged while the missing spectrum weight is gradually replenished, resembling the pseudogap filling at higher temperatures. Finally, a semi-metallic state is restored with the full conduction and valence bands intersecting each other (shaded in orange), which corresponds to the electronic normal state at sufficiently high temperatures achieved in the high-fluence pump-probe study²⁸.

Furthermore, the low-energy electronic structure shows a striking correspondence to an anomalous chemical potential evolution. Here, the chemical potential change is evaluated from the energy shift of the fully filled Ta $4f$ core-level (Fig. 3(b-c)), and cross referenced with the Se valence band shift (dashed line in Fig. 3(a)) to rule out external contributions⁴¹. Negative electronic compressibility (NEC), where the energy of the electronic subsystem decreases despite the addition

of electrons, appears when the pseudogap is being replenished in the second stage (Fig. 3(d), green region). Considering that purely electronic systems only exhibit repulsive interactions to added electrons, such an NEC behaviour requires the involvement of additional degrees of freedom, such as the fluctuating lattice, in order to maintain the thermodynamic stability of the full system.

Microscopic explanation via inter-band electron-phonon coupling:

Exciton condensate formation was proposed as a viable route toward MIT soon after the success of the BCS theory of superconductivity⁷. In Ta₂NiSe₅, one leading contention is whether it is electron-phonon coupling or direct electron-hole Coulomb attraction that dominates the energy gap in the insulating state. We found that both the thermal and nonthermal evolution of the spectral function can be captured using a minimal model (Method and Extended Data Fig. 6) with symmetry-informed electron-phonon coupling. Following the symmetry of the monoclinic instability of the high-temperature orthorhombic lattice, a long-wavelength optical phonon, whose displacement breaks the local orthorhombic symmetry, couples the conduction band bottom (blue) to the valence band top (red) (Fig. 4(a), left)⁴². Enhanced by the low dimensionality, strong lattice fluctuation above T_s readily enables the band hybridization, and results in the electronic pseudogap without causing a global symmetry breaking (Fig.4(a), middle). This reproduces the pseudogap spectra above T_s (Fig. 2(b)). Further reducing temperature results in a divergence of the phonon number (Extended Data Fig. 7), which signifies the phonon condensation and drives the transition to the monoclinic phase. Subsequently, a hard hybridization gap forms below T_s (Fig.4(a), right). On the other hand, increasing electron carrier density lifts the Fermi level from the charge-neutral point and reduces the inter-band phonon dressing. Thus, doping ultimately drives the monoclinic phase into the orthorhombic phase even at low temperatures, which is quantitatively confirmed by both the many-body lattice model and first-principles calculation (Extended Data Fig. 8-9). The observed NEC also reflects strong electron-phonon coupling⁴³⁻⁴⁴ (Extended Data Fig. 9), since the additional electronic energy from the added carriers is absorbed into the lattice degree of freedom. Therefore, the phase diagram of Ta₂NiSe₅ along both the temperature and electron-doping axes can be sketched in Fig. 4(b), where the region shaded in green represents the lattice fluctuation induced pseudogap state above T_s . Clearly, both thermal and quantum fluctuations effects are strong.

On the other hand, without the phonon breaking the lattice mirror symmetry, direct prototypical inter-band Coulomb interaction alone can only repel the conduction and valence bands in their entirety, and is unable to create a hybridization gap or excite electrons from Ta to Ni orbitals²³ (Extended Data Fig.10(a)). Even in the presence of lattice-induced gap, Coulomb interaction up to 200 meV can only marginally increase the gap size, and severely weakens the

conduction band bottom spectral intensity due to much reduced band overlap (Extended Data Fig. 10(b)).

Discussion and outlook:

While electron-phonon coupling is a symmetry prerequisite and a dominant component in the low temperature energy gap, the strength of direct electron-hole Coulomb attraction V may also be estimated from the valence-conduction band overlap in the semi-metallic state. Such band overlap E_g (Fig. 4(a)) will be linearly reduced with increasing V and total electron-hole population $n_e + n_p$ (Extended Data Fig. 10(a)). By comparing the high temperature spectra ($E_g \cong 240$ meV, Extended Fig.3(b)), the ultrafast photodoped spectra ($E_g \cong 260$ meV, photodoping enhanced $n_e + n_p$)²⁸, and heavily K-doped spectra ($E_g \cong 260$ meV, heavily screened V , Extended Data Fig. 4, cycle 19), an upper bound of ~ 70 meV can be placed for V (see Method). This electronic Coulomb interaction would incur less than 5% enhancement to the low temperature gap (Extended Data Fig. 11), consistent with the earlier identification of a mostly lattice-induced insulating state in Ta₂NiSe₅.

In the pseudogap phase without any global structural symmetry lowering over an extensive temperature range above T_s , phonons cannot be treated under the Born-Oppenheimer approximation, where the lattice's impact on the electronic structure is only reflected by a nonzero lattice distortion $\langle x \rangle$. Instead, phonons act on the electronic structure through a fluctuating state where $\langle x \rangle = 0$ but $\langle x^2 \rangle \neq 0$ (Fig.1(a) middle inset and Fig. 4(b)). In this regard, the pseudogap state may be conceptually likened to the 'preformed excitons' proposed in recent studies³¹, except that they are facilitated by lattice fluctuations. In comparison, fluctuating CDW states in cuprates, K_{0.3}MoO₃, ZrTe₃, NbSe₃, and NbSe₂ are usually intertwined with electronic instabilities induced by strong Coulomb interaction¹⁵⁻¹⁸. In Ta₂NiSe₅, however, the major distinction lies in the low-energy band crossings protected by a lattice mirror symmetry, prohibiting Coulomb interaction induced charge transfer to the leading order. Therefore, these distinct lattice-symmetry-protected low-energy band crossings can be used to engineer accentuated lattice fluctuation effects in correlated materials.

While we qualitatively explain the observed spectral behaviours using a single phonon mode, further experiments have indicated multiple phonons involved in this transition: the diffuse scattering and inelastic x-ray scattering results point to extensive transverse acoustic phonon softening³⁸; Raman scattering also suggest strong involvement of optical phonons²⁰⁻²². Distinct from the acoustic-phonon-driven structural transition in 3D perovskite PrAlO₃⁴⁵, the strong fluctuations in Ta₂NiSe₅ seem to suggest additional contributions from the lower system dimension

and optical phonons. Critical exponents can provide further insight into the nature of this transition, which may be accessed through *in-situ* gated diffraction or elastic constant measurements. The exceptional multitude of lattice fluctuations offers great potential to realize optically, electrically and mechanically controlled MIT, and a selective collapse of a many-body entangled state into a long-range ordered symmetry-breaking state.

Methods:

Sample synthesis

Single crystals of Ta₂NiSe₅ were grown by the chemical vapor transfer method with iodine (I₂) as the transport agent. Starting materials composed of Ta powder (99.99%), Ni powder (99.99%), Se powder (99.99%) with the nominal molar ratio 2:1:5 were fully ground and mixed together inside the glovebox. About 50 mg of iodine was added to the mixture of the starting powder. The mixture was then vacuumed, backfilled with 1/3 Argon and sealed inside a quartz tube with an inner diameter of 8 mm, an outer diameter of 12 mm and a length of about 120 mm. The sealed quartz tube was placed horizontally inside a muffle furnace during the growth. The hot end reaction temperature was set to 950 K and the cold end was left in the air with the temperature stabilized at 850K. Long and thin single crystals were harvested by quenching the furnace in air after one week. Excess iodine was removed from the surfaces of the crystals with ethanol.

Physical properties measurement

Electric resistivity and heat capacity measurements were carried out by using a commercial PPMS (Quantum Design). The electric resistivity was measured by the four-probe method with the current applied in the ac-plane of a Ta₂NiSe₅ single crystal. The specific heat measurement was performed in the temperature range from 200K to 400K where the background signal was recorded in the same temperature range.

Angle-resolved photoemission spectroscopy (ARPES)

Synchrotron-based ARPES measurements were performed at beamline BL5-2 of Stanford Synchrotron Radiation Laboratory (SSRL), SLAC, USA, and BL 7.0.2 of Advanced Light Source (ALS), USA. The samples were cleaved *in situ* and measured under ultra-high vacuum below 3×10^{-11} Torr. Data was collected by R4000 and DA30L analyzers. The total energy and angle resolutions were 10 meV and 0.2°, respectively.

Thermal diffuse scattering

Hard X-ray single-crystal diffraction is carried out at the energy of 44 keV at beamline QM2 of the Cornell High Energy Synchrotron Source (CHESS). The needle-like sample is chosen with a typical lateral dimension of 100 microns, then mounted with GE Varnish on a rotating pin before being placed in the beam. A Pilatus 300M 2D detector is used to collect the diffraction pattern with the sample rotated 360 degrees around three different axes at 0.1° step and 0.1s/frame data rate at each temperature. The full 3D intensity cube is stacked and indexed with beamline software.

First-principles DFT calculation

Ab initio calculations are performed using the Quantum ESPRESSO package⁴⁶. The structural relaxation is calculated using the r²SCAN⁴⁷ functional with a semiempirical Grimme's DFT-D2 Van-der-Waals correction⁴⁸. A $30 \times 30 \times 15$ **k**-mesh was used with a 100 Ry wavefunction energy cut-off.

Model calculation

Full many-body model simulations allowing fluctuations

We perform many-body model simulations to address the impact of electron-phonon coupling and lattice fluctuations. According to the fitting with experimental results, we map the electronic structure Ta₂NiSe₅ into a two-band model (Extended data Fig. 6), in a similar form as Ref. 42:

$$H = \sum_{k,\sigma} \varepsilon_k^c c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k,\sigma} \varepsilon_k^v f_{k\sigma}^\dagger f_{k\sigma} + V \sum_{i,\sigma,\sigma'} (n_{i\sigma}^c + n_{i+1\sigma}^c) n_{i\sigma'}^f \\ + \sum_{k,q,\sigma} \frac{g_q}{\sqrt{N}} [(a_q + a_{-q}^\dagger) c_{k+q,\sigma}^\dagger f_{k\sigma} + H.c.] + \sum_q \omega_q a_q^\dagger a_q$$

where $c_{k\sigma}^\dagger$ ($c_{k\sigma}$) creates (annihilates) an electron at the conduction band (primarily Ta 5*d*) for momentum k and spin σ , with dispersion given by ε_k^c , while the $f_{k\sigma}^\dagger$ ($f_{k\sigma}$) creates (annihilates) an electron at the valence band (primarily Ni 3*d*), with dispersion given by ε_k^v . Accordingly, the $n_{i\sigma}^c$ and $n_{i\sigma}^f$ are the density operators for the conduction and valence band, respectively. Ideally, the conduction band should contain two sub-bands due to the two Ta chains. However, they form bonding and anti-bonding bands with opposite (x-axis) mirror symmetries, and only one of them hybridizes with the valence band⁴⁸. To simplify the many-body simulation, we pick one of them representing the conduction band.

The direct hybridization between the conduction and valence bands is forbidden in the orthorhombic phase by the inversion symmetry²³. However, this hybridization is enabled by an anti-

symmetric lattice distortion, parametrized as a momentum-dependent displacement x_q and quantized as the phonon mode $x_q = a_q + a_{-q}^\dagger$. As the Fermi momentum k_F is much smaller than $2\pi/a_0$, we further restrict the e-ph coupling to the zone centre $g_q = g\delta_q$. Therefore, other phonon modes are decoupled from electrons and can be ignored. The phonon frequency is chosen to be 2THz²⁰⁻²². To generate the simulations in Fig.4 of the main text, we set the coupling strength $g = 70$ meV and Coulomb interaction $V = 0$.

We perform an exact diagonalization (ED) simulation on an 8-site cluster (effectively 16 sites due to the two bands). The diagonalization of the Hamiltonian leads to the ground-state wavefunction $|G\rangle$ and all excited states (denoted as $|m\rangle$, with $|m=0\rangle = |G\rangle$). The finite-temperature spectral function, for each band, is calculated through a canonical ensemble average

$$A^c(k, \omega) = -\frac{1}{\pi N Z} \sum_{m, \sigma} e^{-\frac{E_m}{k_B T}} \text{Im} \langle m | c_{k\sigma}^\dagger \frac{1}{\omega + H - E_m + i\delta} c_{k\sigma} | m \rangle$$

$$A^f(k, \omega) = -\frac{1}{\pi N Z} \sum_{m, \sigma} e^{-\frac{E_m}{k_B T}} \text{Im} \langle m | f_{k\sigma}^\dagger \frac{1}{\omega + H - E_m + i\delta} f_{k\sigma} | m \rangle$$

where Z is the partition function. The ensemble-averaged equal-time observables, e.g., the average phonon occupation, are defined in a similar manner

$$\langle O \rangle = \frac{1}{Z} \sum_m e^{-\frac{E_m}{k_B T}} \langle m | O | m \rangle$$

To capture the high momentum resolution comparable to experiments, we employ the twisted average boundary condition (TABC) in the simulation, with 50 equal-spacing phases for the spectral simulation and 30 phases for the equal-time observables. In addition to achieving the momentum resolution, the TABC is known to reduce the finite-size effects, particularly for the model with $q = 0$ interactions⁴⁹. The ED simulation provides the solution of the full many-body state in 1D and preserves reflection symmetry, i.e. $\langle x \rangle \equiv 0$; however, the fluctuation of displacements $\langle x^2 \rangle \neq 0$, reflecting the phonon squeezed state.

Mean-field simulations for the symmetry breaking state at $T < T_s$

To reflect the spectral properties with the presence of symmetry breaking, we simulate the spectral function by projecting the phonon fluctuations to a classical displacement. Specifically, we assume that the ground state collapses to a symmetry-breaking state characterized by a finite X . Treating this X as a classical variable, we obtain the mean-field equation for electrons

$$H_{MFT}(X) = \sum_{k, \sigma} \varepsilon_k^c c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k, \sigma} \varepsilon_k^v f_{k\sigma}^\dagger f_{k\sigma} + g_0 X \sum_{k, q, \sigma} [c_{k+q, \sigma}^\dagger f_{k\sigma} + H.c.].$$

Using the $\langle x^2 \rangle$ obtained by ED simulations for various temperatures, we estimate the mean-field by $X = \sqrt{\langle x^2 \rangle}$ and evaluate the spectral functions for the corresponding symmetry-broken states.

Estimation of the upper bound of the Coulomb interaction

The strength of direct electron-hole Coulomb attraction V is estimated from the valence-conduction band overlap in the semi-metallic state.

If the system has a finite inter-band Coulomb interaction in the form of the Hamiltonian

$$H_{\text{int}} = V \sum_{i,\sigma,\alpha,\sigma'} (n_{i\alpha\sigma}^c + n_{i+1\alpha\sigma}^c) n_{i\sigma'}^f,$$

the doped carriers will result in a shift of site energy for the conduction and valence band, respectively. Here, we consider the presence of two degenerate conduction bands (Ta 5d) as they are important for the sum rule of dopants. Ignoring the impact of electron-phonon coupling, the system is resistant to the excitonic condensation, and each band has conserved electron numbers (for specific global doping). We denote their total electron numbers (considering the conduction-band degeneracy) as N_c and N_f , respectively. Thus, the effective Hamiltonian can be mapped into

$$H_{\text{int}}^c \approx V \sum_{i,\sigma,\alpha,\sigma'} (n_{i\alpha\sigma}^c + n_{i+1\alpha\sigma}^c) \langle n_{i\sigma'}^f \rangle = \frac{2VN_f}{N} \sum_{i,\alpha,\sigma} n_{i\alpha\sigma}^c$$

for the conduction band and

$$H_{\text{int}}^f \approx V \sum_{i,\sigma,\alpha,\sigma'} \langle n_{i\alpha\sigma}^c + n_{i+1\alpha\sigma}^c \rangle n_{i\sigma'}^f = \frac{2VN_c}{N} \sum_{i,\sigma} n_{i\sigma}^f$$

for the valence band. Therefore, the doping-induced change of the band overlap corresponds to

$$\Delta\mu = \frac{2V(\Delta N_c - \Delta N_f)}{N}$$

According to the Luttinger volume, the doped electrons for the 15th-doping (Fig. 3(c)) reaches ~ 0.4 per unit cell, indicating $(\Delta N_c + \Delta N_f)/N = 0.46$. As the Fermi velocity is comparable in two bands, it leads to $(\Delta N_c - \Delta N_f)/N \approx 0.152$. Therefore, with an upper bound of $\Delta\mu \sim 20\text{meV}$, we obtain $V < 67\text{meV}$. Such a Coulomb interaction is too small to address the spectral gap (over 200meV) at the low temperature gapped state. Therefore, from a quantitative perspective independent from the symmetry analysis, we conclude that the Coulomb interaction does not play a major role in the observed insulating state.

Data availability

The data will be deposited to an open-access Yale repository and released upon final publication.

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AUTHOR CONTRIBUTIONS

Y.H. and X.C. conceived the project. C.C. performed the ARPES experiment and analysis with the help of S.W., Y.H., and Y.C.. X.C. synthesized and characterized the samples with EDX. X.C. carried out transport and specific heat measurements with guidance from R.B., J.R., Y.H., and X.C. carried out the high-energy X-ray diffraction experiment. Y.W. and S.D. performed the model calculation. W.T. performed DFT calculation with guidance from Z.L. and S.G.L.. D.H.L., M.H., C.J., A.B., and E.R. maintain the ARPES beamlines. C.C., Y.W. and Y.H. wrote the manuscript with input from all authors.

DECLARE OF INTEREST

The authors declare no competing interests.

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FIGURES AND LEGENDS

FIG 1| Evidence of strong lattice fluctuation in Ta_2NiSe_5 above T_s . (a) Schematic of a second-order phase transition. The grey line marks the evolution of the order parameter $\langle M \rangle$, while the red line marks the fluctuation term $\sqrt{\langle M^2 \rangle}$. The region shaded in red indicates the fluctuating region around the transition temperature. (b) Temperature-dependent resistivity of Ta_2NiSe_5 . $T_s \sim 329\text{K}$ is marked by the purple triangle. Inset: phonon background-subtracted specific heat near T_s . (c) Temperature dependence of $|Q(1\ 3\ 4)|$ and $|Q(-1\ 3\ 4)|$ peaks from single crystal X-ray diffraction. Inset: top view of the lattice structures across the phase transition. (d) High energy x-ray diffuse scattering in the $H0L$ plane. Thermal diffuse scattering signal between the $(2\ 0\ 6)$ and $(2\ 0\ 8)$ Bragg peaks at (i) 305 K, (ii) 329 K, and (iii) 360 K, respectively. All figures are plotted under the same log color scale to highlight the evolution of the diffuse signal (green arrows).

FIG 2| Temperature dependence of the single-particle gap in Ta_2NiSe_5 . (a) DFT calculations of the single-particle bands below and above T_s . States from Ta $d_{x^2-y^2}$ and Ni d_{xy} orbitals are emphasized, corresponding to the linear horizontal (LH, blue) and linear vertical (LV, red) channels in the experiments respectively. (b) Photoemission spectra along $X - \Gamma - X$ direction in both photon polarization channels at select temperatures. (c) Temperature dependent Energy Distribution Curves (EDCs) at k_F ($0.075\text{\AA}^{-1}$, orange dashed line in (b)) for both photon polarization channels. (d) Temperature dependence of the (i) EDC peak position and (ii) integrated spectral intensity around $E_F \pm 30\text{meV}$. Vertical purple dashed lines mark the structural transition at T_s .

FIG 3| Evidence of pseudogap and negative electronic compressibility (NEC) with potassium dosing. **(a)** Photoemission spectra along $X - \Gamma - X$ directions at discrete potassium dosing cycles (lower-left indices) at 80K. Distinct regions are shaded in different colors, marking the evolution stages of the band structure. The lower dashed line indicates the evolution of high energy bands from Se orbitals. **(b)** Evolution of K $3p$ and Ta $4f$ corelevel peaks with potassium dosing. **(c)** Carrier density and chemical potential estimated from the peak area increment of K $3p$ corelevel and the peak position shift of Ta $4f$ corelevel, respectively. **(d)** Electronic compressibility calculated from the parameters deduced in (c). Red line is calculated from the polynomial fit of the scattered data points in (c).

FIG 4| Lattice fluctuation induced pseudogap and the temperature-doping phase diagram of Ta_2NiSe_5 . (a) Simulated band structure from the model with only inter-band electron-phonon coupling: the semi-metallic normal state (left), the pseudogap state caused by lattice fluctuation (middle), and the broken-symmetry state caused by structural band hybridization (right). (b) Doping-temperature phase diagram of Ta_2NiSe_5 . The crystal structures within different regions are illustrated on the side.

Supplementary Files

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