

Uniaxial-Pressure Control of Excitonic Fluctuations and Monoclinic Distortions in Ta₂NiSe₅

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Ta₂NiSe₅ undergoes a phase transition characterized by the opening of an energy gap and a reduction in its crystal symmetry below $T_c = 326$ K. The primary cause of this transition is debated to be either electron-hole correlations, in line with the excitonic insulator hypothesis, or structural instability. The chain structure of Ta₂NiSe₅ enables effective control over its electronic and lattice properties by applying uniaxial strain. In this study, we utilize polarization-resolved Raman spectroscopy to demonstrate that the electronic and structural order parameters respond differently to uniaxial pressure applied along the Ta-Ni chains. Compressive strain reduces monoclinic distortions while enhancing excitonic fluctuations. Such disparate behavior suggests that many-body effects may significantly amplify exciton fluctuations even when monoclinic distortions are minimized, thereby supporting the excitonic nature of the phase transition.

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Introduction—The quest for the long-proposed spontaneous condensation of excitons in solids [1,2] has driven researchers to study Ta₂NiSe₅ extensively as a potential candidate to achieve the ground state of an excitonic insulator [3]. The direct-gap semimetal-to-insulator transition of the quasi-one-dimensional electron system in Ta₂NiSe₅ is accompanied by symmetry breaking, which cooperatively induces a structural phase transition at $T_c = 326$ K. While emphasizing the likelihood of exciton condensation [4], the associated $q = 0$ monoclinic distortions introduce uncertainty as to whether the phase transition in Ta₂NiSe₅ is primarily due to electronic [5–9] or structural instability [10–14]. The ongoing debate focuses on the relative strengths of the electronic and structural contributions, under the assumption of a linear coupling of the lattice to the excitonic order parameter [4,11,13]. However, a more intricate interplay of electron-phonon coupling and many-body effects can significantly affect the quantitative comparison of these contributions [15–21]. The observation of giant exciton Fano resonances in the infrared spectra of Ta₂NiSe₅ [15] suggests that exciton states weakly bound to phonons can effectively gain

strength from the extensive area of exciton-polaron complexes [22], which overlap significantly along the Ta-Ni chain direction throughout the entire crystal [16]. Based on this hypothesis, exciton fluctuations can be critically influenced and controlled by strain applied along the Ta-Ni chains, affecting the overlap of the exciton-polaron complexes.

Recent experiments have demonstrated the efficacy of Raman scattering as a tool for selectively probing the electronic and lattice degrees of freedom in Ta₂NiSe₅, achieved by observing critical charge fluctuations and phonon anomalies near its phase transition at $T_c = 326$ K [8,20,23–25]. Raman scattering specifically reveals quasielastic scattering of B_{2g} symmetry, which is attributed to critical fluctuations of the excitonic order parameter. The B_{2g} excitonic susceptibility follows the Curie-Weiss behavior, indicating the electronic origin of the phase transition. Additionally, the B_{2g} phonon mode that corresponds to a monoclinic shearing motion of the Ta atoms undergoes significant dampening and nearly disappears in the crossed polarized spectra above T_c , in line with expectations for a B_{2g} phonon in the orthorhombic phase.

In this Letter, we employed the capability of Raman scattering to discern symmetry-selective information regarding electronic and lattice degrees of freedom to study the impact of uniaxial elastic strain on exciton fluctuations and monoclinic structural distortions at temperatures near T_c . We report that compressive strain suppresses the monoclinic distortion while enhancing excitonic fluctuations, indicating that these fluctuations are not cooperatively coupled with lattice instability. This contrasting

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behavior demonstrates that collective many-body effects significantly enhance exciton fluctuations in Ta_2NiSe_5 , even when the monoclinic distortions are suppressed by uniaxial pressure, and further supports the excitonic origin of the phase transition.

Experimental details—Thin single crystals of Ta_2NiSe_5 were grown and characterized as described elsewhere [7]. To apply strain to the samples, Razorbill CS100 and CS130 compact cryogenic strain cells were used [26,27]. Before mounting in the strain device, the crystals were shaped with typical lateral dimensions of $1.5 \times 0.25 \text{ mm}^2$ along the a and c axes, respectively, as illustrated in Figs. 1(a) and 1(b). A rigid-bridge method was introduced to prevent the soft and fragile crystals from breaking and bending, maintaining the material's crystalline structure during strain cycling [28,29]. The samples were cleaved to thicknesses of less than $50 \mu\text{m}$ and attached to a titanium bridge using Loctite™ Stycast epoxy. The compressive or tensile strain applied to the bridge was transferred to the crystal along the a axis, parallel to the Ta-Ni chains. The efficiency was estimated by a resistance-based strain meter, achieving 90% efficiency up to 332 K, with the highest strain

reaching $\epsilon_{xx} \approx \pm 0.3\%$ [28]. The rigid-bridge method facilitates the application of elastic tensile and compressive strains. Our study of strain-dependent transport and optical properties revealed that these properties are entirely reversible and reproducible when the applied strain is cycled. Electric resistivity along the a axis, aligned with the strain direction, was measured using a four-probe configuration with a quantum design physical property measurement system (PPMS). Raman scattering measurements were performed over the 7 cm^{-1} to 340 cm^{-1} energy range using a Horiba LabRam HR800 spectrometer. A continuous wave 632.8-nm laser at a power of $460 \mu\text{W}$ was utilized, featuring a light spot size of $5 \mu\text{m}$ that keeps local heating within 10 K. The incident light was polarized along the a axis of the crystal. The analyzer set the polarization of the measured Raman spectra to either the a axis or c axis for parallel aa or crossed ac polarization geometry, respectively.

Results and discussions—We first studied the effect of uniaxial strain on the transition temperature T_c . The transition in Ta_2NiSe_5 is most readily seen in transport [7]. The resistivity ρ_a parallel to the Ta and Ni chains is measured as a function of temperature across T_c , under a -axis strains from -0.3% (compressive) to 0.3% (tensile). A well-defined kink is observed in $\rho_a(T)$ at T_c , which corresponds to a step in the temperature dependence of the activation energy defined as $E_p = -k_B T^2 (\partial \ln \rho_a / \partial T)$ [28], in agreement with previous reports [7,30]. Figure 1(c) shows the strain dependence of the phase transition temperature and the transition width, determined by analyzing the derivative of the resistance temperature dependence, as illustrated in Fig. 1(d). T_c systematically decreases with increasing compressive and tensile strain in the heating and cooling phases, exhibiting a broad maximum at zero strain. Notably, the compressive strain has a more pronounced impact on the phase transition, making it broader and reducing T_c by approximately 20 K at -0.3% strain. This pronounced asymmetry between compressive and tensile phases suggests that the optimal lattice parameters at zero strain are possibly achieved as a result of different concurring lattice effects on the electron band properties, which suppress T_c in the opposite strain phases. We used polarization-resolved Raman scattering under uniaxial strain to selectively probe exciton fluctuations and lattice distortions to address this issue.

The polarization dependencies and temperature anomalies observed in the Raman scattering of Ta_2NiSe_5 at zero strain are consistent with previous studies [8,20,23–25]. As shown in Fig. S4 and Table S1 of Supplemental Material [28], all eleven Raman-active phonons—three B_{2g} and eight A_g modes, as defined in the high-temperature $Cmcm$ orthorhombic phase—are resolved within the measured temperature range of 295 K to 350 K. The phonons are superimposed on an electronic background consisting of a quasielastic peak (QEP) and a higher energy

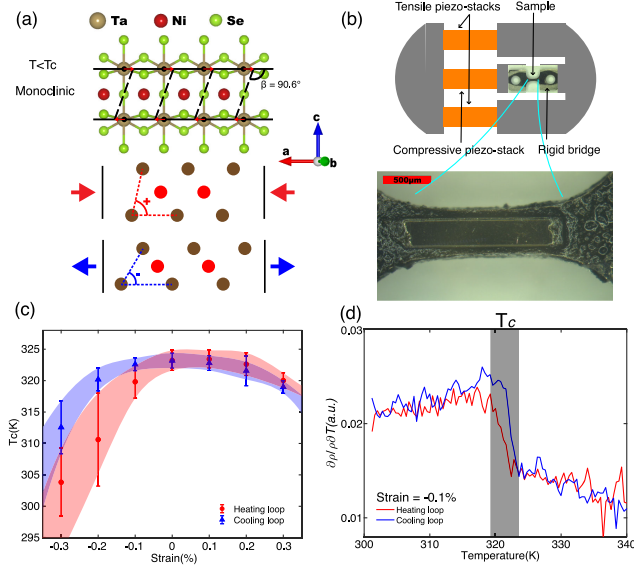


FIG. 1. (a) In-plane schematic illustrating the lattice distortion of the Ta-Ni chains in Ta_2NiSe_5 , associated with the orthorhombic-to-monoclinic phase transition, and the effects of uniaxial strain through compression and extension along the chain direction. (b) Schematic view of the piezoelectric-based uniaxial strain device and the Ta_2NiSe_5 sample mounted on the titanium rigid bridge illustrating the experimental geometry used for strain measurements. (c) The dependence of the phase transition temperature and transition width on strain, as determined from a -axis electrical transport measurements with increasing (red) and decreasing (blue) the temperature within the 300 K to 350 K range [28]. (d) The temperature-dependent activation energy of the resistivity data measured at -0.1% strain, demonstrating how the transition parameters were defined for each strain value.

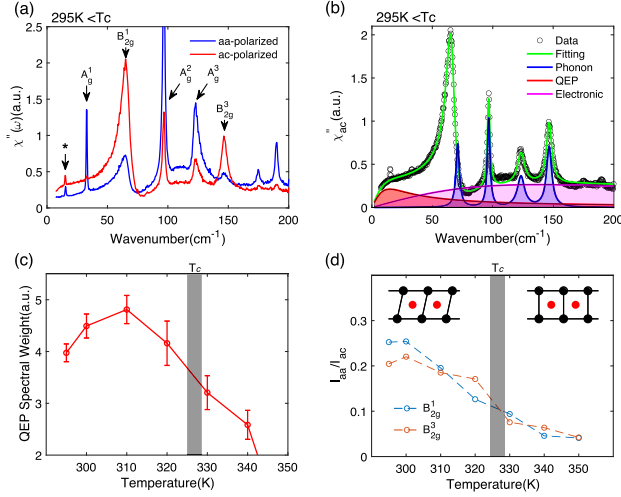


FIG. 2. (a) The polarization dependence of Raman susceptibility under zero strain, derived from Raman scattering intensity using Bose correction [28]. In parallel aa and cross ac polarization geometries, the A_g and B_{2g} symmetries are probed, respectively, according to the Raman selection rules above T_c . Below the orthorhombic-to-monoclinic phase transition temperature T_c , the B_{2g} symmetry breaks into A_g . The asterisk marks a peak due to the notch filter's Rayleigh leakage. (b) The cross-polarization Raman susceptibility is modeled using a generalized Fano approach, incorporating four phonons coupled to a quasi-elastic peak (QEP) and a higher energy electronic continuum [28]. The decomposed bare phonons and electronic continuum contributions are shown in blue and pink, respectively, with the red shading indicating the QEP contribution, which reflects the excitonic fluctuation. (c) The temperature dependence of the integrated spectrum weight of the QEP. (d) The phonon intensity ratios, I_{aa}/I_{ac} , between the aa and ac polarization geometries are plotted as a function of temperature across T_c , serving as fingerprints of the structural transition.

continuum. Figure 2(a) illustrates the Raman susceptibility of Ta_2NiSe_5 in the representative range of 7 cm^{-1} to 200 cm^{-1} , measured at 295 K in the parallel aa (blue) and crossed ac (red) polarization geometries. The B_{2g}^1 phonon, which softens above the monoclinic-to-orthorhombic phase transition, along with the QEP scattering, provides crucial insights into the transition mechanism, indicating both symmetry breaking and exciton fluctuations [8,23]. To quantitatively parametrize the observed features, we model the ac polarized Raman susceptibility of a coupled system of two electronic contributions and four phonon oscillators ($B_{2g}^{1,3}$ and $A_g^{1,2}$) using a phenomenological approach based on a Green's-function formalism [28]. The negligible contribution of the heavily damped weak B_{2g}^2 phonon does not affect the quality of the fit. Figure 2(b) presents the cross-polarized Raman susceptibility under zero strain at 295 K decomposed into bare phonons (blue) coupled to the QEP (red) and higher energy electronic continuum (pink). The low-energy QEP is attributed to

excitonic fluctuation scattering, theoretically predicted to appear in the B_{2g} channel [11]. As observed in previous Raman studies [8,23], the QEP emerges in cross polarization as the temperature rises above 250 K, reaches its maximum intensity around T_c , and subsequently diminishes in its spectral weight. The temperature dependence of the QEP ac Raman susceptibility, extrapolated to zero frequency, follows Curie-Weiss behavior above T_c , with a Weiss temperature of approximately 241 K [8]. Figure 2(c) demonstrates that the QEP contribution, measured at zero strain in a limited temperature range around T_c , replicates this temperature behavior well.

The QEP spectral weight indicates excitonic fluctuations, while polarization- and temperature-dependent phonon intensities can reveal monoclinic distortions. The B_{2g} phonon modes exhibit distinct temperature-dependent intensity distribution between the cross-polarized ac and parallel-polarized aa scattering geometries. The Raman susceptibility shown in Fig. 2(a) at 295 K is consistent with the minor Raman tensor elements due to the small lattice distortion angle [8,23–25]. In the high-temperature orthorhombic $Cmcm$ phase, the B_{2g}^1 phonon at 65 cm^{-1} dominates the ac spectra, with slight leakage into the aa polarization scattering around T_c . This phonon mode is attributed to the c -axis shear vibration of Ta and Se atoms, as illustrated in Fig. S5(a) of Supplemental Material [28]. The intensity ratio's temperature dependence between the aa and ac polarization geometries is shown to be proportional to $\sqrt{1 - T/T_c}$, indicating it as a marker of monoclinic lattice distortion below T_c [20]. It exhibits strong damping, resulting in a highly asymmetric Fano line shape due to its interaction with the electronic continuum. This mode, sharing symmetry with the excitonic fluctuations, becomes frozen at the transition [8]. The B_{2g}^1 and B_{2g}^3 phonon aa/ac intensity ratio, plotted against temperature in Fig. 2(d), demonstrates a gradual reduction in monoclinic distortions with temperature during the monoclinic-to-orthorhombic phase transition.

After identifying the parameters and features in the Raman spectra to monitor, we proceeded to the primary objective of the study: to examine the impact of uniaxial strain on exciton fluctuations and monoclinic distortions in the crystal structure of Ta_2NiSe_5 selectively. Measurements were performed in cross- and parallel-polarization configurations at temperatures ranging from 295 K to 350 K, and under strain applied from -0.3% (compression) to 0.3% (tension), as shown in Figs. S7 and S8 of Supplemental Material [28]. Figure 3 shows the strain dependencies of the Raman susceptibility obtained for two representative temperatures, 295 K and 320 K. Unlike hydrostatic pressure, which causes all phonons to harden due to compression of the crystal lattice, uniaxial strain has varying effects on the resonance frequencies of phonon modes. These effects depend on the specific patterns of

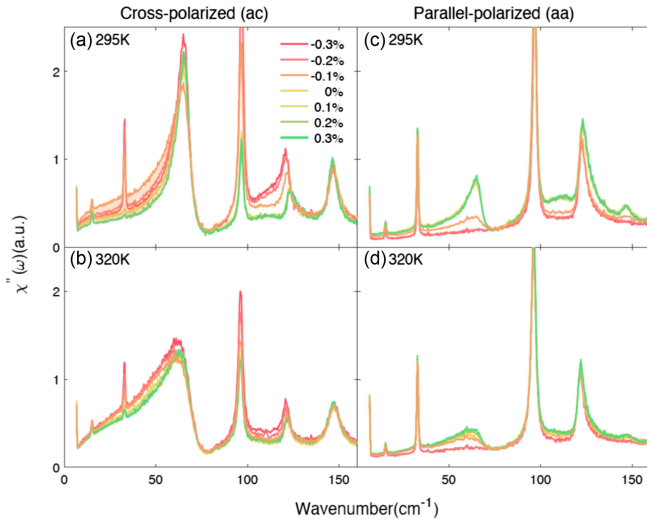


FIG. 3. Cross-polarized (a),(b) and parallel-polarized (c),(d) Raman spectra of Ta_2NiSe_5 under a -axis strain ranging from -0.3% to 0.3% measured at two representative temperatures around T_c . (a),(b) The QEP, evident in the ac spectra, exhibits significant strain dependence, reaching its maximum at -0.1% compressive strain. (c),(d) The 65 cm^{-1} and 158 cm^{-1} phonon modes belong to the B_{2g} irreducible representation in the orthorhombic phase and becomes activated in the aa spectra due to monoclinic distortions. Compressive strain effectively suppresses its intensity in the aa geometry. In contrast, tensile strain enhances its polarization leakage, indicating a monotonic increase in monoclinic distortions with increasing interatomic distances along the a axis.

vibrational eigenvectors associated with each phonon. For example, consider the behavior of the A_g^3 phonon at 125 cm^{-1} , which shifts toward higher energies under tensile strain and softens under compression along the a axis. This phonon mode involves Se atoms vibrating against neighboring atoms along the c axis, as illustrated in Fig. S5(b) of Supplemental Material [28]. It is particularly sensitive to c -axis strain transferred from the applied a -axis uniaxial strain, following the principles of Poisson's ratio under strain. The continuous hardening of the 125 cm^{-1} phonon under a -axis strain applied from -0.3% to 0.3% provides direct evidence that the uniaxial strain is effectively applied to the Ta_2NiSe_5 crystal, resulting in an increase in the lattice structural parameter a .

In contrast to the response of phonon eigenfrequencies to strain, the behavior of phonon intensities in polarized Raman spectra provides clear insights into the monoclinic distortions of the orthorhombic aristotype structure ($Cmcm$) under a -axis strain, as previously discussed. The most evident phenomenon is the behavior of the 65 cm^{-1} and 158 cm^{-1} phonons in the ac spectra with crossed polarizers. The appearance of these $B_{2g}^{1,3}$ modes, identified for the $Cmcm$ structure, in the ac Raman spectra around T_c is attributed to monoclinic distortions. Figures 3(c) and 3(d) demonstrate that tensile strain along

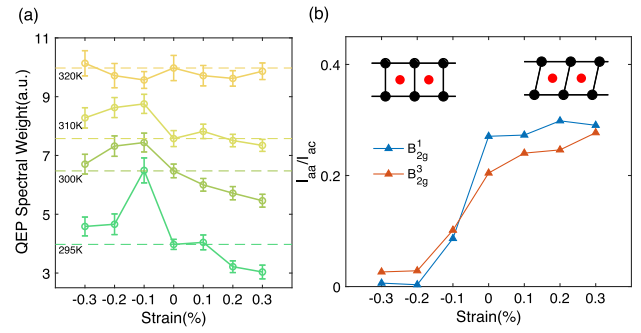


FIG. 4. (a) The dependence of the QEP spectral weight on strain at various temperatures, with each curve shifted vertically for clarity. The dashed lines correspond to the values at zero strain from Fig. 2(c). At 295 K , the QEP weight shows a maximum at -0.1% strain, which becomes less defined with increasing temperature and eventually disappears above T_c . (b) The phonon intensity ratio I_{aa}/I_{ac} as a function of strain for $B_{2g}^{1,3}$ modes, measured at 295 K . Tensile strain increases monoclinic structural distortions, while compressive strain tends to shift the crystal structure towards a less distorted orthorhombic phase.

the a axis enhances the phonon intensity, while compressive strain suppresses it, rendering it nearly undetectable and indicating a restoration of structural order to the highly symmetric state. The phonon intensity ratios between the aa and ac Raman geometries as a function of a -axis strain are summarized in Fig. 4(b). A comparison with the zero-strain temperature dependencies of the phonon intensity ratios shown in Fig. 2(d) reveals that uniaxial pressure applied along the Ta-Ni chains drives the system toward the high-temperature orthorhombic phase.

We now focus on a key observation in the strained Raman scattering measurements: the nonmonotonic behavior of the QEP under uniaxial pressure. The evolution of the Raman susceptibility spectra, shown in Fig. 3(a), reveals a distinct change in the low-energy electronic background when uniaxial strain is applied at 295 K . Notably, the electronic background decreases with tensile strain, whereas under compression, the behavior is nonmonotonic, displaying a pronounced maximum at -0.1% strain. The increase in peak intensity at -0.1% uniaxial compression is noticeable in both the original Raman intensity spectra and after correcting for the Bose factor in the susceptibility spectra at temperatures from 295 K to 310 K , below T_c , as shown in Figs. S7 and S8 of Supplemental Material [28]. As the temperature approaches T_c , this effect diminishes and becomes obscured by overlapping phonon changes, as illustrated at 320 K in Fig. 3(b). Figure 4(a) summarizes the dependence of the QEP spectral weight on a -axis strain ranging from -0.3% to 0.3% at various temperatures. To account for phonon contributions and quantitatively analyze the strain behavior of the QEP, we employed the generalized Fano model, similar to our previous analysis of temperature dependencies at zero strain, as illustrated in Fig. 2(b).

The observed increase in the B_{2g} QEP scattering at -0.1% strain suggests that the initial a -axial pressure significantly enhances the critical fluctuations of the exciton order parameter at temperatures below T_c , before these fluctuations are suppressed with further compression. Concurrently, as we previously discussed, intensity of the 65 cm^{-1} and 158 cm^{-1} phonons, of the same B_{2g} symmetry, are monotonically suppressed in the aa Raman spectra of Fig. 3(c), indicating a reduction in the monoclinic distortions responsible for the structural transition. This counteracting effect of uniaxial pressure on exciton fluctuations and symmetry-breaking structural distortions may reflect the shift of the Weiss temperature toward T_c .

This behavior suggests that electron correlations and structural instability do not contribute cooperatively to establishing the insulating ground state of Ta_2NiSe_5 . To consider exciton-phonon coupling in Ta_2NiSe_5 , it is necessary to account for more complex collective many-body effects, beyond a simple linear coupling of the lattice to the excitonic order parameter. The formation of exciton-polaron complexes is evidenced by the giant exciton Fano resonances in the absorption spectra of Ta_2NiSe_5 [15]. In this context, the wave function of a weakly bound exciton involves a large polaron region surrounding lattice vibrations, with coherent oscillations of the electronic polarization over the whole region [22]. Consequently, the collective excitation oscillator strength is significantly enhanced, by several orders of magnitude, proportional to the factor $(r_p/r_0)^3$, where r_p is the polaron radius and r_0 is the interatomic distance. In Ta_2NiSe_5 quasi-1D electron system, the increase in this factor as the interatomic distance decreases can explain the enhancement of exciton fluctuations induced by strain applied along the Ta–Ni chains, affecting the overlap of exciton-polaron complexes.

Conclusion—We utilized Raman scattering to discern symmetry-selective information regarding electronic and lattice degrees of freedom in Ta_2NiSe_5 and found that uniaxial pressure applied along the Ta–Ni chains has opposite effects on exciton fluctuations and symmetry-breaking structural distortions at temperatures near T_c . Our findings suggest that collective many-body effects may significantly amplify critical fluctuations of the exciton order parameter, even when monoclinic distortions are suppressed by uniaxial pressure. This further supports the excitonic origin of the phase transition in Ta_2NiSe_5 .

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Data availability—The data supporting the findings of this study are publicly available at [31].

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