Dimensionality crossover to a two-dimensional vestigial nematic state from a three-dimensional antiferromagnet in a honeycomb van der Waals magnet

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The effects of fluctuations and disorder, which are substantially enhanced in reduced dimensionalities, can play a crucial role in producing nontrivial phases of matter such as vestigial orders characterized by a composite order parameter. However, fluctuation-driven magnetic phases in low dimensions have remained relatively unexplored. Here we demonstrate a phase transition from the zigzag antiferromagnetic order in the three-dimensional bulk to a Z_3 vestigial Potts-nematicity in two-dimensional few-layer samples of van der Waals magnet $NiPS_3$. Our spin relaxometry and optical spectroscopy measurements reveal that the spin fluctuations are enhanced over the gigahertz to terahertz range as the layer number of $NiPS_3$ reduces. Monte Carlo simulations corroborate the experimental finding of 3-fold rotational symmetry breaking, but show that translational symmetry is restored in thin layers of $NiPS_3$. Therefore, our results show that strong quantum fluctuations can stabilize an unconventional magnetic phase after destroying a more conventional one.

A vestigial order describes the partial melting of the primary order (η) of a spontaneous symmetry breaking phase[1,2]. In contrast to conventional short-range orders that do not break symmetries of their hosting crystalline lattices, a vestigial order corresponds to a composite order ($\eta^\dagger\tau\eta$, a quadratic form of η with a matrix τ) and breaks a subset of symmetries broken by the primary order[1,2]. Fluctuations and disorders are the typical causes for destroying primary orders, and potentially, introduce vestigial orders. Illustrated in Fig. 1a, the increasing temperature provides thermal fluctuations, and the reduced dimensionality can independently contribute to enhanced fluctuations. In between the long-range primary ordered region I ($\langle\eta\rangle \neq 0$ and $\langle\eta^\dagger\tau\eta\rangle \neq 0$) and the completely disordered region III ($\langle\eta\rangle = 0$ and $\langle\eta^\dagger\tau\eta\rangle = 0$), there could exist the region II for the vestigial ordered case[1,2] ($\langle\eta\rangle = 0$ but $\langle\eta^\dagger\tau\eta\rangle \neq 0$). The impact of introducing vestigial order has been profound, as it provides the intertwined nature of proximate phases within the rich phase diagrams for many quantum material systems including Cu-based high-T_c superconductors[1,3-7], Fe-based superconductors[8-11], charge density wave systems[12] and others[13]. Theoretical examples of primary versus vestigial orders are ample in many systems: charge or spin density wave v.s. nematicity[2,14,15] , pair density wave v.s. charge-4e superconductivity or nematicity or charge density wave[16-19], superconductivity v.s. charge-4e superconductivity or nematicity[20-22], and even the anyon condensation from $Z_2\times Z_2$ topological order quantum spin liquid v.s. Z_2 topological order quantum spin liquid[23], *etc*.

Experimental realizations of vestigial order have been primarily focused on the nematicity that breaks the rotational symmetry but preserves the translational symmetries of underlying crystal lattices[2,15]. This symmetry requirement allows the tetragonal and hexagonal systems to support the emergence of nematicity. Indeed, in tetragonal systems such as cuprates[6,7] and pnictides[9,15], Z_2 Ising-nematicity is observed to develop from the charge and spin fluctuations, which breaks the tetragonal 4-fold into the orthorhombic 2-fold rotational symmetry and retains the translation symmetries. In contrast, in hexagonal systems, Z_3 Potts-nematicity is anticipated, which breaks the hexagonal 6-fold (or 3-fold) rotational symmetry and preserves the translational symmetries[24-27]. Very recently, in systems such as bulk $Fe_{1/3}NbS_2$[28] and bulk $FePS_3$[29], characters of Potts-nematicity have been seen in their long-range ordered magnetic phases, i.e., region I in Fig. 1a. However, the intrinsic Potts-nematic order without the primary order, i.e., region II in Fig. 1a, has been much less explored. Comparing Z_2 Ising-nematicity and Z_3 Potts-nematicity, they are also expected to show distinct phase transitions upon the application of external strain[30].

Reducing dimensionality is one promising, but less explored route to enhance fluctuations. In fact, pioneering theoretical studies on nematicity often used two-dimensional (2D) models[2]. In this regard, we choose a 2D honeycomb magnetic system with the XY-type spin anisotropy, $NiPS_3$[31-35], to harvest stronger spin fluctuations for realizing Z_3 Potts-nematicity. This study makes a distinct effort from the mainstream research on 2D magnetism that works on maintaining long-range magnetic orders against fluctuations[36-40]. The monolayer $NiPS_3$ has the trigonal crystallographic point group D_{3d} with an out-of-plane 3-fold

rotational axis, and the bulk and few-layer $NiPS_3$ obey the monoclinic structural point group C_{2h} due to the lateral shift between adjacent layers along the *a* axis. The bulk $NiPS_3$ is considered undergoes an antiferromagnetic (AFM) transition at $T_{N,3D}$ = 155K. In the long-range ordered AFM state, the Ni^{2+} spins align along the *a* axis ferromagnetically (FM) within the zigzag chains and are coupled AFM between neighboring chains[41-44] (Fig. 1b). Such a long-range zigzag AFM order in $NiPS_3$ spontaneously breaks the 3-fold rotational symmetry of individual layers, and at the same time, breaks the translational symmetry with a wavevector $\mathbf{Q} = \mathbf{k_M}$ where $\mathbf{k_M}$ is the momentum at the M point in the Brillouin zone (Fig. 1b inset). Three degenerate zigzag AFM states are expected, with three $\mathbf{Q}$ rotated from each other by 120°, i.e., $\mathbf{Q}_j$ (j = 1, 2, 3). Such a broken rotational symmetry (BRS) and broken translational symmetry (BTS) AFM phase in bulk $NiPS_3$ has a non-zero primary order parameter $\eta_j(\mathbf{r}) = \left(\mathbf{S}(\mathbf{r}) - \mathbf{S}(\mathbf{r}+\mathbf{e}_j)\right) e^{i\mathbf{Q}_j\cdot\mathbf{r}}$ with $\mathbf{e}_j$ being the vector between neighboring spin sites and parallel to $\mathbf{Q}_j$, and also supports a finite secondary order parameter $\eta^\dagger\tau\eta = \left(\frac{\sqrt{3}}{2}(\eta_3^2 - \eta_2^2),\ \frac{1}{2}(2\eta_1^2 - \eta_2^2 - \eta_3^2)\right)$, belonging to region I in Fig. 1a. Due to the weak anisotropy in the XY-model, strong spin fluctuations in the 2D, and FM-AFM exchange competitions, few-layer $NiPS_3$ is expected to realize the scenario in region II where the nematic phase has BRS but no BTS (Fig. 1c), before going into the completely spin disordered, paramagnetic phase of region III (Fig. 1d).

We start by verifying the enhancement of spin fluctuations in thinner $NiPS_3$ samples, using both the nitrogen-vacancy (NV) spin relaxometry and the quasi-elastic scattering (QES) in Raman spectroscopy that cover distinct frequency regimes from GHz to THz. NV spin relaxometry utilizes the excellent magnetic field sensitivity of NV spin centers to probe fluctuating magnetic fields at the NV electron spin resonance frequencies (~ GHz) generated by proximal magnetic materials[45-47]. The measured NV spin relaxation rate is proportional to the spectral density of the magnetic field noise, which scales with the spin fluctuation strength[46]. Here, we spatially visualize the spin fluctuations in exfoliated $NiPS_3$ flakes using the wide-field imaging mode of NV spin relaxometry[48]. A representative NV spin relaxation rate map is shown in Fig. 2a inset, for a 4-layer (4L) and a 10nm-thick $NiPS_3$ flake at 170K. Figure 2a shows the temperature dependence of the area-averaged NV spin relaxation rate for the 4L and 10nm $NiPS_3$ flakes, displaying a decreasing trend for spin fluctuations at lower temperatures for both samples. To account for the thickness difference of spin fluctuation, we extract the magnetic susceptibility χ of 4L and 10nm $NiPS_3$ samples, from the NV spin relaxation rate (see Supplementary Information Note 1). Figure 2b plots χ, which is proportional to the spin fluctuations[49], as a function of temperature for the 4L and 10nm $NiPS_3$ samples. It is evident that χ for 4L $NiPS_3$ is significantly enhanced in comparison with that for 10nm $NiPS_3$ below ~ 150K, whereas it is only slightly larger in 4L than in 10nm sample above ~ 150K.

Complementarily, Raman QES in $NiPS_3$ arises from spin fluctuations[50] and manifests as a slow delay profile in the Raman spectra for up to ~ 40cm^{-1} (in the order of THz), with two exemplary Raman

spectra of QES for a 4L and a 13.1nm $NiPS_3$ flake taken at 170K shown in Fig 2c. Note that both spectra are normalized to their corresponding thicknesses for a fair comparison of spin fluctuations between the two samples. It is evident that the spin fluctuations are stronger in 4L than in 13.1nm $NiPS_3$ over a spectral range up to ~40cm^{-1}, despite the overall decay at higher frequencies for both samples. Moreover, we can integrate the QES spectral weight (SW_{QES}) over 8 – 40cm^{-1}, with that from the phonon breathing mode excluded, to quantify the spin fluctuations, and track its temperature dependence (see Supplementary Information Note 2 for the characteristic behavior of the breathing mode). Figure 2d shows the temperature dependence of SW_{QES} for bilayer (2L), trilayer (3L), 4L, 13.1nm, and bulk $NiPS_3$, with each trace normalized to its corresponding SW_{QES} at 200K. There is a similarity amongst the SW_{QES} (T) plots that QES decreases as temperature reduces with a small kink at a critical temperature around 155K for bulk ($T_{N,3D}$) and a broad bump around 120K for 2L ($T_{N,2D}$) $NiPS_3$. But what is more striking and more important is the contrasting behaviors of QES at low temperatures among $NiPS_3$ of distinct thicknesses: bulk $NiPS_3$ shows a fully suppressed QES at temperatures below a characteristic temperature T_{sQES} ~90K that is lower than $T_{N,3D}$, 13.1nm $NiPS_3$ reveals a decreased T_{sQES} of ~40K, whereas 2L, 3L, and 4L $NiPS_3$ all feature finite QES down to our lowest temperature 10K, i.e., T_{sQES}, if present, is lower than 10K. Such a stronger contrast at lower temperatures of spin fluctuations between bulk and few layers is suggestive of different magnetic phases between 3D and 2D $NiPS_3$.

Up to now, it is convincingly shown that as $NiPS_3$ thickness reduces from 3D bulk to 2D flake, spin fluctuations are clearly enhanced, fulfilling the key requirement for realizing vestigial nematicity in 2D $NiPS_3$. A natural next step is to examine the magnetic phases upon thinning 3D bulk $NiPS_3$ down to 2D films, with a proper experimental tool to simultaneously detect BRS and BTS, as these two together are required to distinguish zigzag AFM, Potts-nematicity, and paramagnetism illustrated in Figs. 1b-1d. Here, we select Raman spectroscopy with which BRS can be detected through the splitting of doubly degenerate E_g phonons and BTS can be probed by zone-folded phonon modes. Figure 3a shows Raman spectra for bulk $NiPS_3$ taken in linearly parallel and crossed channels at 10K with a 633nm excitation laser, over selected frequency ranges of interest (see full-range Raman spectra in Supplementary Information Note 3). Figures 3b and 3c show the temperature dependent Raman spectra in the linearly parallel channel of Fig. 3a. First, the two Raman modes around 180cm^{-1} are degenerate above $T_{N,3D}$ = 155K and split below $T_{N,3D}$, whose frequency separation was assigned as the signature for zigzag AFM previously[34]. Despite the monoclinic stacking between honeycomb layers, the phonon modes obey the selection rules of D_{3d} above $T_{N,3D}$, and the two phonon modes at ~180cm^{-1} belong to the $E_g(D_{3d})$ symmetry. It is BRS, rather than BTS, from the zigzag AFM order that lifts the $E_g(D_{3d})$ doublet into one $A_g(C_{2h})$ and one $B_g(C_{2h})$ mode (thus named as P_{BRS}). As a result, the frequency separation here shall be proportional to the composite nematic order parameter $\eta^{\dagger}\tau\eta$, instead of η. Second, the Raman mode at ~30cm^{-1}, emerging below ~90K, has not been reported previously where the 515nm excitation laser was used [34,51]. Interestingly, this $T_{AFM,3D}$ = ~90K onset

temperature for the 30cm^{-1} mode coincides with the temperature for the suppression of QES (T_{sQES}) and the temperature for the emergence of the zone-center magnon at ~43cm^{-1} (Supplementary Information Note 4). We assign this mode to be a phonon mode folded from the Brillouin zone boundary M point to the center Γ point due to the zigzag AFM order, due to this onset temperature coincidence above and after the following examinations. We first observed the in-plane and out-of-plane magnetic field independence for this mode and rule out the possibility of magnon[42,43] (see Supplementary Information Note 5). We then compared to the calculated phonon dispersion spectra of bulk $NiPS_3$ and found no intralayer or interlayer Γ point phonons around 30cm^{-1} [52,53]. Considering that the zigzag AFM order has a wavevector $\mathbf{Q} = \mathbf{k_M}$, we checked the phonon spectra at the M point, and indeed, found a computed M point phonon mode at ~30cm^{-1} [52]. Furthermore, we examined the polarization dependence of this Raman mode and observed its clear anisotropy, confirming it is a single-Q zone-folding process that breaks the 3-fold rotational symmetry (Supplementary Information Note 6). We note that the blue shift of this 30cm^{-1} mode upon increasing temperature possibly results from thermal expansion of lattice at higher temperatures (Supplementary Information Note 7). Thus, this zone-folded ~30cm^{-1} mode is a direct consequence of BTS (labeled as P_{BTS}), and its strength should scale with the primary zigzag AFM order parameter η. From our result, we find that in bulk $NiPS_3$, the spins remain highly dynamic between 155K and ~90K with BRS but no BTS ($T_{N,\,3D} > T > T_{AFM,\,3D}$), and the long-range, static AFM order with both BRS and BTS forms below ~90K ($T < T_{AFM,\,3D}$), which is consistent with the magnetic susceptibility measurements (Supplementary Information Note 8).

Having established signatures of both BRS and BTS, as well as their relationship to the primary and composite order parameters, we proceed to examine their evolutions upon the dimensionality reduction. Figure 3d shows Raman spectra of P_{BRS} and P_{BTS} at 10K for a range of thickness from bulk down to 2L. The signature of BTS, P_{BTS}, vanishes when the thickness reduces below ~10.6nm, whereas the footprint for BRS, P_{BRS}, remains observable from bulk down to 2L $NiPS_3$. While no observable BRS feature has been seen in 1L $NiPS_3$, the linewidth of the ~180cm^{-1} E_g mode in 1L is ~10cm^{-1}, which is much greater than both the linewidth and the splitting of P_{BRS} in 2L and thicker $NiPS_3$ (Supplementary Information Note 9). Moreover, the zone-center magnon at ~43cm^{-1} disappears below ~10.6nm, same as the BTS does (Supplementary Information Note 10). This contrast between BTS and BRS features reveals the fact that the zigzag AFM order in 3D bulk $NiPS_3$ transitions into the nematic order in 2D few-layer $NiPS_3$ across a critical thickness of t_C ~10.6nm. We rule out the built-in strain in few-layer $NiPS_3$ as a potential concern (Supplementary Information Note 11), and we comment that the survival of the BRS signature down to 2L is consistent with the presence of exciton PL down to 2L[31-33] as they both concern the nematic order parameter. We further quantify the thickness dependence of both signatures, via four important parameters, the frequency separation of P_{BRS} ($\Delta\omega_{BRS}$), the line width of P_{BRS} (Γ_{BRS}), the linewidth of P_{BTS} (Γ_{BTS}), and the intensity ratio between P_{BTS} and P_{BRS} (I_{BTS}/I_{BRS}). It has been discussed above that $\Delta\omega_{BRS}$ scales with the nematic order parameter, and its thickness independence in Fig. 3e confirms the robust presence of nematicity down to

the bilayer. Furthermore, the nematicity coherence determines Γ_{BRS}, and the zigzag AFM coherence sets Γ_{BTS}. Figure 3f shows that Γ_{BRS} remains nearly constant across t_C ~10.6nm and only starts to increase slightly in 3L and more significantly in 2L. In contrast, Figure 3g illustrates a clearly divergent behavior of Γ_{BTS} approaching t_C from above. The distinct trends between Γ_{BRS} and Γ_{BTS} across t_C clearly demonstrate the separation between the primary order (i.e., the zigzag AFM order of region I) and the vestigial order (i.e., the nematic order of region II) in the phase diagram illustrated in Fig. 1a. The suppression of $I_{\mathrm{BTS}}/I_{\mathrm{BRS}}$ down to zero when thinning $NiPS_3$ across t_C (Fig. 3h) further corroborates with the melting of the zigzag AFM order but the survival of the vestigial nematicity below t_C. To experimentally construct a thickness (dimensionality) v.s. temperature phase diagram for $NiPS_3$, we performed temperature dependent measurements for selected thickness including bulk, 74.0nm, 25.4nm, 13.1nm, and few-layer. Figures 3i and 3j shows the temperature dependence of BTS and BRS features, respectively. The characteristic temperature for BTS (T_{AFM}) rapidly decreases, evolving from ~90K for bulk to ~40K for 13.1nm $NiPS_3$ and showing remarkable consistency with T_{sQES}. In contrast, the temperature dependence of BRS shows a clearly order-parameter-like onset (T_N) that remains nearly constant from bulk to 13.1nm $NiPS_3$ (see Supplementary Information Note 12 for data of 2L-5L $NiPS_3$). We conclude the temperature and thickness dependence in the phase diagram in Fig. 3k, where region I, region II, and region III are found.

Having confirmed the presence of intrinsic vestigial nematicity in few-layer (thickness less than t_C) $NiPS_3$ belong to region II of Fig. 1a, we move forward to check the symmetry class of this nematicity. We select a large size 4L $NiPS_3$ (lateral dimension of ~40μm) to image the nematic domains using the scanning optical linear dichroism (LD) microscopy. The LD technique is a static probe known to be sensitive to BRS and has been used to study few-layer $NiPS_3$[32]. While the BRS signature in Raman process around 180cm^{-1} can potentially be caused by a dynamic nematic liquid[54], the finite LD signal confirms the static BRS of this nematic state in $NiPS_3$. For the same reason as P_{BRS} above, the LD signal scales with the vestigial nematic order parameter, rather than the zigzag AFM order parameter, and therefore onsets at $T_{N,3D}$ ~ 155K for bulk $NiPS_3$ (Supplementary Information Note 13). Figure 4a shows a LD map for the 4L $NiPS_3$, where domains of different LD signals can be seen with clear boundaries between different domains. Surveying the angular dependence of the LD-induced polarization rotation across the 4L sample, we find that there are three, and only three, distinct patterns that are shown in Fig. 4b-d. These three patterns are related by the 3-fold rotational operation, corresponding to the three nematic domain states with the nematic order parameter rotated by 120° from one another i.e., Z_3 Potts-nematicity (schematic shown in the inset of Fig.4e-g). The *a* axis direction, defined as the nematic order parameter orientation, is calibrated by the polarization resolved photoluminescence (PL) measurement (see Supplementary Information Note 14). Such three Potts-nematic domain states are further confirmed by the angular dependence of the lower frequency P_{BRS} mode (Figs. 4e-4g) whose anisotropy originates from the rotational symmetry reduction by the vestigial nematicity in the few-layer case. Thermal cycling does not change the nematic domains and nematic order

parameter orientations within domains (see Supplementary Information Note 15), indicating that the nematic domain states are likely pinned by the structural monoclinic interlayer stacking[55]. We note that the weak coupling between the structural monoclinicity and the vestigial nematicity should not impact the physics of interest here that enhanced fluctuations in 2D melt the zigzag AFM order and introduce the Z_3 Potts-nematicity.

To comprehend the experimental finding of the crossover from 3D zigzag AFM to 2D Potts-nematicity when the $NiPS_3$ thickness decreases across t_c, we further performed large-scale Monte Carlo simulations to visualize the magnetic phase for bilayer $NiPS_3$ and examine its primary (η) and composite ($\eta^\dagger\tau\eta$) order parameters. The magnetic state at finite temperatures is simulated under the spin Hamiltonian with intralayer Heisenberg exchange coupling up to the third nearest neighbor (J_1, J_2, J_3), a strong easy-plane anisotropy (D_z), a weak easy-axis anisotropy (D_x), interlayer nearest and next-nearest exchange coupling (J_{l-n} and J_{l-nn}) (see Methods). Note that the coordination for J_{l-n} obeys the 3-fold rotational symmetry whereas that J_{l-nn} breaks the 3-fold rotational symmetry and therefore encodes the monoclinic interlayer stacking. Figure 5a plots the temperature dependence of the nematicity order parameter $\eta^\dagger\tau\eta = \langle|\boldsymbol{m}_3|\rangle$ scaled with $L^{\beta/\nu}$ for four system sizes $L = 36, 60, 84, 108$, where temperature is in unit of $|J_1|$ and β and ν are critical exponents for 2D Z_3 Potts-nematicity. All four traces cross at a single temperature (inset of Fig. 5a), defining the nematicity critical temperature, $T_{\mathrm{N,2L}} = 1.285|J_1| = 67.1$ K, on the same order of the experimental value of 120K in Fig. 2d. This observation clearly confirms the emergence of Potts-nematicity in 2L $NiPS_3$ (see Supplementary Information Note 16 for the consistent results for 1L – 5L $NiPS_3$). We note that such a 2D Potts-nematic phase transition remains when the effect from the monoclinic stacking (J_{l-nn}/J_{l-n}) is small, which is the case of our experimental findings. But it can evolve into a crossover when J_{l-nn}/J_{l-n} gets larger (Supplementary Information Note 17). We further contrast this 2D Potts-nematic state against the zigzag AFM order by computing and comparing the nematicity and spin correlation, $C_{\mathrm{nematicity}}(\mathrm{r})$ and $C_{\mathrm{spin}}(\mathrm{r})$, at a temperature T=1.25$|J_1|$<$T_{\mathrm{N,2L}}$, as shown in Fig. 5b. After the initial decay in short distance for both curves, which are normalized to their corresponding values at r = 2, $C_{\mathrm{nematicity}}(\mathrm{r})$ reaches a saturation plateau that is consistent across all four system sizes, whereas in contrast, $C_{\mathrm{spin}}(\mathrm{r})$ keeps having further decay as the system size increases. Note that the increase at distance greater than half of the system size is due to the finite size effect. The size-independent plateau behavior for $C_{\mathrm{nematicity}}(\mathrm{r})$ confirms the long-range order of the composite nematic order parameter, but the size-dependent ever-decaying trend for $C_{\mathrm{spin}}(\mathrm{r})$ demonstrate the "disorder" nature of the zigzag AFM state, corroborating with our experimental finding and belonging to Region II in Fig. 1a. To directly visualize the real-space spin arrangements for this unique magnetic phase, we plot a snapshot of the simulated result in Fig. 5c for spin texture and in Fig. 5d for normalized nematicity director texture. The spins (red and blue arrows) in Fig. 5c are randomly canted away from the zigzag chain direction, losing the spin coherence at long distance as

revealed by Fig. 5b. As a comparison, the nematicity directors (green double arrows) in Fig. 5d show a much better homogeneity and therefore a long-range coherence as found in Fig. 5b.

Our work on the thickness dependence of spin fluctuations and magnetic phases in $NiPS_3$ clearly demonstrate that the enhanced fluctuations in 2D introduce the Z_3 Potts-nematic state by partially melting the conventional zigzag AFM order. Based on this result, we envision multiple emergent research opportunities as following. First, it is a promising venue to exploit enhanced fluctuations in the 2D, rather than suppressing them, to discover spin-fluctuation-driven magnetic phases, including and going beyond this Z_3 spin-induced Potts-nematicity. This direction distinguishes from the mainstream effort in 2D magnetism research thus far. Second, it remains as an open experimental question what the spin coherence length is in 2D $NiPS_3$, while the corresponding Potts-nematicity coherence length is much beyond optical wavelength (i.e., ~1μm). In fact, it is a more general question experimentally how partially melting the primary order parameter is in a vestigial state, to address which experimental techniques with high spatial resolution (i.e., ~ nanoscale) are required. Third, because the monoclinic interlayer stacking provides an effective field to couple with the Potts-nematicity in 2D $NiPS_3$, it would be interesting to explore what the magnetic state will be if twisting two $NiPS_3$ flakes to create a moiré superlattice whose supercell contains all three monoclinic stacking geometries. Finally, given that 3D $NiPS_3$ realizes the Mott insulating state with the strong electron correlations[56] and also hosts ultra-narrow exciton emission lines possibly from the many-body effect[31,57], 2D $NiPS_3$ offers a unique platform to explore the interplay between spin degree of freedom (DOF) with strong fluctuations and charge DOF of strong correlations, a parameter regime inaccessible previously in 3D systems.

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Author Contributions Statement

Z.S., R.H., and L.Z. conceived the idea and initiated this project. Z.S. exfoliated $NiPS_3$ thin flakes with different layer numbers. G.Y., Z.Y., C. N. and Z.S. carried out the Raman experiments under the supervision of L.Z. and R.H.. M.H. performed the NV spin relaxometry under the supervision of C.D.. C.Z. carried out the Monte Carlo simulations under the supervision of K.S. and Z.Y.M.. Q. L. and Z. S. carried out the AFM measurements and the photoluminescence measurements guided by H.D. and L.Z.. N.H. grew the high-quality $NiPS_3$ bulk single crystals under the supervision of D.M.. G.Z. performed the susceptibility measurement under the supervision of L.L.. X.X. performed the phonon calculations under the supervision of L.Y.. Z.S., R.H., and L.Z. analyzed data and wrote the manuscript. All the authors participated in the discussion of the results.

Competing Interests Statement

The authors declare no competing interest.

Figure Captions

Figure 1| Schematic phase diagrams of vestigial order and magnetic states in $NiPS_3$. a, Schematic phase diagram to show the primary order (η) and the vestigial order ($\eta^\dagger\tau\eta$) as functions of temperature (vertical axis) and other independent factors, such as enhanced fluctuations at reduced dimensionality or disorder effect (horizontal axis). Three distinct regions are shown: (I) long-range primary order ($\langle\eta\rangle \neq 0$ and $\langle\eta^\dagger\tau\eta\rangle \neq 0$, shaded with blue color), (II) long-range vestigial order with short-range primary order ($\langle\eta\rangle = 0$ but $\langle\eta^\dagger\tau\eta\rangle \neq 0$, shaded with orange color), and (III) disorder ($\langle\eta\rangle = 0$ and $\langle\eta^\dagger\tau\eta\rangle = 0$). Examples of primary order *v.s.* vestigial order are listed in the phase diagram, including charge or spin density

wave v.s. nematicity[2,14,15] , pair density wave v.s. charge-4*e* superconductivity or nematicity or charge density wave[16-19], superconductivity v.s. charge-4*e* superconductivity or nematicity[20-22], *etc*. **b,** Schematic of the long-range zigzag AFM order in 3D $NiPS_3$ with both BRS and BTS, corresponding to the region I in **a**. The Ni^{2+} spins are aligned along the zigzag chain. The green parallelogram represents the in-plane unit cell of the crystal structure without the AFM order, and the blue rectangle shows the doubled in-plane unit cell for the zigzag AFM state. Inset: Brillouin zone (BZ) folding in the momentum space. The *M* point at BZ boundary without the AFM order folds to the Γ point of BZ center by the zigzag AFM order. **c,** Schematic of the spin-induced nematic state in 2D $NiPS_3$ with BRS but without BTS, corresponding to the region II in **a**. Inset: Corresponding BZ without folding due to the lack of BTS. **d,** Schematic of the spin disordered state without BRS and BTS in $NiPS_3$, corresponding to the region III in **a**. Inset: Corresponding BZ. In **b-d**, only the Ni atoms are shown for simplicity.

Figure 2| Thickness-dependent spin fluctuations in few-layer $NiPS_3$ measured by NV relaxometry and Raman spectroscopy. a, Temperature dependence of NV spin relaxation rate Γ_M measured at NV centers underneath the 10 nm- and 4L-$NiPS_3$. Data points are presented as $\Gamma_M \pm \delta\Gamma_M$, where Γ_M and $\delta\Gamma_M$ denote the optimal fitting parameter and one standard error, respectively. Inset: NV spin relaxation map for the few-layer $NiPS_3$ samples at 170 K. The black dashed lines outline the boundary of the 10 nm- and 4L-$NiPS_3$ samples. The scale bar is 4 μm. **b,** Fitted magnetic susceptibility χ of 10 nm- and 4L-$NiPS_3$ as a function of temperature. Data points are presented as $\chi \pm \delta\chi$, where χ and $\delta\chi$ denote the optimal fitting parameter and one standard error, respectively. **c,** Normalized QES Raman spectra measured with λ=532 nm in 13.1nm and 4L-$NiPS_3$ at 170 K, respectively. The shaded area denotes the breathing mode in 4L-$NiPS_3$ that is excluded in calculating the QES spectral integral in **d**. **d,** Temperature-dependent integrated QES intensity over 8 – 40 cm^{-1} in $NiPS_3$ with different thickness. The dark blue shading highlights the peak/bump near the nematic transition, and the light blue gradient shading highlights the temperature dependent evolution of QES. The arrows indicate the temperatures at which QES of bulk and 13.1nm samples vanishes. Data points are presented as SW ± δSW, where SW and δSW denote the optimal fitting parameter and one standard error, respectively.

Figure 3| Thickness dependence of BTS and BRS signatures in Raman spectroscopy. a, Linearly polarized Raman spectra measured with λ=633 nm in both parallel (black curves) and crossed (gray curves) channels at 10 K on bulk $NiPS_3$. The blue and red areas denote the P_{BTS} mode and P_{BRS} mode, respectively. **b,** Temperature-dependent Raman spectra of P_{BTS} mode in the parallel channel of bulk $NiPS_3$. Here, to have a clearer presentation of the temperature dependence of P_{BTS} mode, we subtracted the QES signal from the raw data for temperatures above 90 K. **c,** Temperature-dependent Raman spectra of P_{BRS} mode in the parallel channel of bulk $NiPS_3$. **d,** Layer-dependent Raman spectra in both parallel (filled circles) and crossed (empty circles) channels at T=10 K. The dark (light) solid line represents the fitted curves in the parallel

(crossed) channel by using the Lorentz function. **e,** Thickness dependence of fitted frequency separation of P_{BRS}, $\Delta\omega_{BRS} = \omega_{BRS-h} - \omega_{BRS-l}$, at T=10 K. Here, ω_{BRS-h} was fitted from cross channel and ω_{BRS-l} was fitted from parallel channel. **f,** Thickness dependence of fitted linewidth Γ_{BRS} of P_{BRS} at T=10 K. Here, Γ_{BRS} was fitted from the low-frequency mode of split P_{BRS} mode in parallel channel. **g,** Thickness dependence of fitted linewidth Γ_{BTS} of P_{BTS} at T=10 K. The linewidth shows a divergent behavior as the thickness decreasing to the critical thickness ~ 10 nm. **h,** Thickness dependence of the fitted intensity ratio of I_{BTS}/I_{BRS} at T=10 K. Here we choose the intensity ratio of I_{BTS}/I_{BRS}, which is defined as $(I_{BTS\text{-}parallel}+I_{BTS\text{-}crossed})/(I_{BRS\text{-}l\text{-}parallel}+I_{BRS\text{-}l\text{-}crossed})$, to focus on the relative strength between P_{BTS} and P_{BRS} and exclude the possible impacts of the absolute intensity of P_{BTS} and P_{BRS} from different dielectric environment. The color shading in **e**-**h** corresponding to region II (yellow) and I (blue), following that in Fig. 1a, and t_c denotes the crossover critical thickness. **i,** Temperature dependence of P_{BTS} mode in different thickness $NiPS_3$. **j,** Temperature dependence of P_{BRS} mode in different thickness $NiPS_3$. Data points in **e**-**j** are presented as the optimal fitting parameters with one standard error from the fittings of Raman spectra. **k,** thickness v.s. temperature phase diagram of $NiPS_3$.

Figure 4| Polarization-dependent LD and Raman data in 4L-$NiPS_3$. a, Spatially resolved LD map for a 4L-$NiPS_3$. The black dashed lines outline the boundary of 4L-$NiPS_3$ samples (optical image shown in Supplementary Fig. S15j). The three dots denote the three Potts-nematic domains with the nematic order parameter rotated 120° from one another. **b-d,** Angular-dependent polarization rotation $\Delta\theta$ in the corresponding nematic domains in the 4L-$NiPS_3$ as shown in **a**, in which red area is positive sign and gray area is negative sign. The arrows indicate the *a* axis in the corresponding domain, which is determined by the polarization-dependent PL measurement (Supplementary Information Note 14). **e-g,** Polar plots of the polarization dependence of fitted $P_{BRS\text{-}l}$ intensity at T=10 K in the corresponding Potts-nematic domain in the 4L-$NiPS_3$ shown in **a**, in which the arrows denote the *a* axis in the corresponding domain. Inset: Schematic of the nematic order directors for the three different Potts-nematic domain states.

Figure 5| Monte Carlo calculations of the magnetic state in 2L $NiPS_3$. a, Temperature dependence of the scaled Potts-nematic order parameter ($\langle|m_3|\rangle$) for four different system sizes, L = 36, 60, 84, and 108 with $J_{l\text{-}n}$ = -0.07 and $J_{l\text{-}nn}$ = 0. Inset: Zoom-in around nematic phase transition temperature $T_{N,\,2L}$=1.285$|J_1|$. The crossing point of scaled nematic order parameter for different system size determines the nematic transition point with the 2D Z_3 Potts exponents $\beta = 1/9$, $\nu = 5/6$. **b,** Correlation of Potts-nematicity ($C_{nematicity}$) and spin (C_{spin}) at a temperature T=1.25$|J_1|$<$T_{N,2L}$, as a function of distance for four different system sizes, respectively. The plot is in log-log scale and we normalize the two correlation functions at r = 2. Each data point in **a** and **b** is the average of 32 Monte Carlo bins with the same computational parameters and conditions, and each bin is measured with 10000 Monte Carlo steps. The error bars are defined as their standard deviations. **c,** Snapshot of simulated spin configuration below $T_{N,\,2L}$, which shows the lack of spin coherence.

d, Image of simulated nematic director (normalized $\boldsymbol{m}_{3,i}$ configuration) for the spin state in **c**, showing homogeneity of nematicity.

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Methods

Sample fabrication. High-quality $NiPS_3$ single crystals were grown by the chemical vapor transport (CVT) method. Few-layer $NiPS_3$ samples were prepared on Si/SiO_2 by mechanical exfoliation from high-quality bulk $NiPS_3$ single crystals in the nitrogen-filled glovebox with oxygen level below 0.1 ppm and water level below 0.5 ppm. For the measurement of NV magnetometry, one hBN thin flake was first picked up by the poly(bisphenol A carbonate) stamp, and then the selected few-layer $NiPS_3$ samples on Si/SiO_2 were picked up by the hBN flake on the poly(bisphenol A carbonate) stamp. The hBN/few-layer $NiPS_3$ was transferred onto diamond membrane for NV magnetometry measurements. The thickness of the few-layer samples were first determined based on the optical contrast and then confirmed by the atomic force microscopy (AFM) after measurement.

Raman measurement. Raman spectroscopy measurements were performed by using the λ=532 nm and 633 nm excitation lasers. The laser beam on the sample was focused down to ~2–3 μm in diameter, and the laser power was kept below 1 mW to minimize the local heating effect during measurements.

Backscattering geometry was used. The scattered light was detected by a thermoelectrically cooled charge-coupled device (CCD) camera from Horiba Scientific, with a spectral resolution of 0.4 cm^{-1}. The temperature dependent Raman measurement was carried out in a commercial variable-temperature, closed-cycle cryostat (Cryo Industries of America). For the magnetic field dependent Raman measurement, a commercial superconducting magnet (Cryo Industries of America) was used to achieve magnetic field from 0 to 7 T.

Raman spectra fitting procedure. We used multiple Lorentzian functions, $\sum_N \frac{A_N(\frac{\Gamma_N}{2})^2}{(\omega-\omega_N)^2+(\frac{\Gamma_N}{2})^2} + C$, to fit the P_{BTS} and P_{BRS} modes, where A_N is the peak intensity, ω_N is the peak position, Γ_N is the peak linewidth, C is background, N=1 for the single peak and N=2 for the double peaks. For the QES signal, we first remove the breathing mode signal if present and then fitted the Raman data in the Raman shift range 8-40 cm^{-1} by using the Lorentzian function centered at 0 cm^{-1}, $\frac{A_N(\frac{\Gamma_N}{2})^2}{\omega^2+(\frac{\Gamma_N}{2})^2} + C$ where C is a constant background. Finally, we integrated the fitted curves without background in range 8-40 cm^{-1} as the QES intensity.

Derivation of angular dependence of Raman modes. We derived the polarization dependence of Raman intensity in the parallel channel as a function of polarization rotation angle θ. The measured Raman intensity (I) is proportional to the square of the product of incident light polarization ($\hat{e}_i$), Raman tensor (R) and the scattered light polarization ($\hat{e}_f$): $I \propto |\langle \hat{e}_i | R | \hat{e}_f \rangle|^2$. In the parallel channel, the incident light $|\hat{e}_i\rangle = \binom{cos\theta}{sin\theta}$, and the scattered light $|\hat{e}_f\rangle = \binom{cos\theta}{sin\theta}$. The Raman tensor of P_{BTS} mode is $\begin{pmatrix} a & 0 \\ 0 & b \end{pmatrix}$. So, the intensity $I_{BTS} \propto |(cos\theta, sin\theta)\begin{pmatrix} a & 0 \\ 0 & b \end{pmatrix}\binom{cos\theta}{sin\theta}|^2 = \frac{1}{8}(a-b)^2 cos4\theta + \frac{1}{2}(a^2-b^2)cos2\theta + \frac{1}{8}(3a^2+2ab+3b^2)$ (Supplementary Fig. S6a). The Raman tensors of $P_{BRS\text{-}l}$ and $P_{BRS\text{-}h}$ are $\begin{pmatrix} e & 0 \\ 0 & -e \end{pmatrix}$ and $\begin{pmatrix} 0 & f \\ f & 0 \end{pmatrix}$, respectively. So the intensity $I_{BRS-l} \propto |(cos\theta, sin\theta)\begin{pmatrix} e & 0 \\ 0 & -e \end{pmatrix}\binom{cos\theta}{sin\theta}|^2 = \frac{e^2}{2}(\cos 4\theta + 1)$ and $I_{BRS-l} \propto |(cos\theta, sin\theta)\begin{pmatrix} 0 & f \\ f & 0 \end{pmatrix}\binom{cos\theta}{sin\theta}|^2 = \frac{f^2}{2}(1-\cos 4\theta) = \frac{f^2}{2}\left(\cos 4\left(\theta + \frac{\pi}{4}\right) + 1\right)$. Both patterns of $P_{BRS\text{-}l}$ and $P_{BRS\text{-}h}$ show four-fold symmetry and have a 45° phase shift from one another (Supplementary Fig. S6b).

NV spin relaxometry measurement. The widefield NV spin relaxation rate measurements were performed in a closed-cycle optical cryostat, and the measurement temperature ranges from 4.5 K to 350 K. A microsecond long green laser pulse generated by an electrical driven 515 nm laser is used to initialize NV centers

to $m_s = 0$ state and read out the spin-state dependent photoluminescence. The laser beam spot focused on the diamond surface is around 20 μm ×20 μm. A CMOS camera is used to collect NV fluorescence images, and both pulses to trigger the camera exposures and drive the laser are controlled by a programmable pulse generator. Nanosecond-long microwave pulses were generated by sending the continuous microwave currents to a microwave switch electrically controlled by a programmable pulse generator, and then were delivered to Au stripline patterned on diamond samples. An external field ~ 70 G is generated by a cylindrical NdFeB permanent magnet attached to a scanning stage inside the optical cryostat. Further details of NV relaxometry measurement protocol can be found in Supplementary Information Note 1.

Linear dichroism measurement. LD measurement was performed by using laser diode with wavelength 635 nm (Thorlabs PL202) and 20 μW power. A photo-elastic modulator (PEM; PEM-100, Hinds Instruments) on the incident path was used for the measurement of LD. The incident light with linear polarization with 45° angle with respect to the PEM fast axis was modulated by a mechanical copper and PEM with retardance of λ/2 by sequence. The light was then passed through a half-wave plate and focused onto the sample in normal incidence with a 50× objective lens. The reflected light passed the half-wave plate again and then went into the detector through a plate beamsplitter. The reflected light was measured by a balanced amplified photodetector (ThorLabs PDB210A). The signal was demodulated by two lock-in amplifiers (Zurich Instruments, MFLI 500kHz/5MHz) separately, of which one lock-in amplifier is referenced to the second harmonic of the PEM frequency 2f = 84.183 kHz and the other lock-in amplifier is referenced to the chopping frequency f = 511 Hz to get the total reflectance as a normalization. For the polarization-dependent LD measurement, the polarization direction of incident light was rotated by the half-wave plate. Thus, the polarization rotation $\Delta\theta$ induced by the LD of few-layer $NiPS_3$ can be written as $\Delta\theta = LDsin(2\alpha + \varphi)$, in which α is the polarization direction of incident light, and φ is phase dependent on the relative angle between the crystal axis and the polarization direction of incident light at zero. $\Delta\theta$ is zero when the polarization direction of incident light is along the crystal axes direction. The angular dependent signal was fitted by the equation $\Delta\theta = LDsin(2\alpha + \varphi)$ and a small nonzero background from inevitable birefringence of the optical components in the whole setup was subtracted during the fitting process. For the LD scanning measurement, a two-axis Galvo scanning mirror paired with a confocal imaging system was used to move the light spot on the sample in normal incidence with a constant fluence and the half-wave plate was fixed at a certain angle during scanning.

Polarization-dependent PL measurement. PL measurement was performed by using a continuous-wave solid-state laser with 532 nm (Thorlabs) to excite the few-layer $NiPS_3$ sample. The beam size focused on sample was ~2.5 μm in diameter. The collected signals are detected by a Princeton Instruments spectrometer

with a CCD camera. The sample is kept at 5 K using a Montana CA100 system. For the polarization-dependent PL measurements, a polarizer was put in the detection beam line to change the polarization direction of collected light into the CCD camera.

Monte Carlo simulations. The magnetic Hamiltonian of $NiPS_3$ can be described by 2D XY type spin Hamiltonian with an easy-plane single-ion anisotropy ($D_z > 0$) and an easy-axis single-ion anisotropy ($D_x < 0$)[43]:

$$H = \sum_{\langle ij \rangle} J_{ij}(S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z) + D_z \sum_i (S_i^z)^2 + D_x \sum_i (S_i^x)^2$$

in which J_{ij} is exchange coupling parameters, D_z is easy-plane single-ion anisotropy and D_x is easy-axis single-ion anisotropy. Here, we considered the nearest exchange coupling interaction J_1, next-nearest exchange coupling interaction J_2, third-nearest exchange coupling interaction J_3 and nearest exchange coupling interaction between neighboring layers $J_{l\text{-}n}$. We performed large-scale Monte Carlo simulations on bilayer $NiPS_3$ systems. In our simulations, we employed various techniques including local updates, Woff updates, and the over-relaxation technique. The spins are treated as classical O(3) vectors, and the simulation is warmed up with 10^4 Monte Carlo steps. In our simulation, we set the parameters $\{J_1, J_2, J_3, J_{l\text{-}n}, D_x, D_z\}$ to be $\{-1, 0, 2.22, -0.07, -0.002, 0.047\}$, with $|J_1|$ as the unit[35,43]. We simulated the bilayer case ($L_z = 2$) with periodic boundary conditions along the x and y directions, but with open boundary conditions along the z direction, where the linear system size ranges from $L = 36$ to $L = 108$. We note that the opposite signs between J_1 and J_3 introduces geometric frustrations, which can cause the sign problem in quantum Monte Carlo simulations. Therefore, we decided to choose the classical Monte Carlo simulations of S = 1 for this current study. Our simulated results well capture the nematic phase transition of this 2D XY magnetic system in turn support the validity and sufficiency of classical Monte Carlo simulations here.

We further checked the spin Hamiltonian with nearest and next-nearest neighbor interlayer exchange coupling for few-layer $NiPS_3$, where

$$H = \sum_{\langle i,j \rangle} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle i,j \rangle\rangle} J_2 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle\langle i,j \rangle\rangle\rangle} J_3 \vec{S}_i \cdot \vec{S}_j + \sum_{\{i,j\}} J_{l-n} \vec{S}_i \cdot \vec{S}_j + \sum_{\{\{i,j\}\}} J_{l-nn} \vec{S}_i \cdot \vec{S}_j + \sum_i D_z (S_i^z)^2 + \sum_i D_x (S_i^x)^2$$

J_{l-n} and J_{l-nn} are the nearest and next-nearest neighbor interlayer exchange coupling, respectively. It is the bond environment of J_{l-nn}, $\{\{i,j\}\}$, that accounts for the broken 3-fold rotational symmetry from the monoclinic stacking.

In our simulation, we define the order parameter of the Z_3 rotational symmetry $\boldsymbol{m_3}$ as[27]

$$\boldsymbol{m_3} = \boldsymbol{\sigma_1}\, \epsilon_1 + \boldsymbol{\sigma_2}\, \epsilon_2 + \boldsymbol{\sigma_3}\, \epsilon_3$$

Here, $\boldsymbol{\sigma_i}$ represents the bond direction, given by $\boldsymbol{\sigma_1} = (0,1)$, $\boldsymbol{\sigma_2} = (-\sqrt{3}/2, -1/2)$, and $\boldsymbol{\sigma_3} = (\sqrt{3}/2, -1/2)$. And ϵ_i refers to the average neighboring bond energy along the $\boldsymbol{\sigma_i}$ direction, i.e., $\epsilon_i = \boldsymbol{S_r} \cdot \boldsymbol{S_{r+e_i}}$. This definition of the nematic order parameter is consistent with $\eta^\dagger \tau \eta = \left(\frac{\sqrt{3}}{2}(\eta_3^2 - \eta_2^2),\ \frac{1}{2}(2\eta_1^2 - \eta_2^2 - \eta_3^2)\right)$ via writing $\eta^\dagger \tau \eta$ as $\eta_1^2\boldsymbol{\sigma_1} + \eta_2^2\boldsymbol{\sigma_2} + \eta_3^2\boldsymbol{\sigma_3}$ where $\boldsymbol{\sigma_j} = \left(\cos[\frac{2\pi}{3}(j-1) + \frac{\pi}{2}], \sin[\frac{2\pi}{3}(j-1) + \frac{\pi}{2}]\right)$. Noting that $\eta_j^2 = \eta_j \cdot \eta_j = \left(\boldsymbol{S_r} - \boldsymbol{S_{r+e_j}}\right) \cdot \left(\boldsymbol{S_r} - \boldsymbol{S_{r+e_j}}\right) = -2\boldsymbol{S_r} \cdot \boldsymbol{S_{r+e_j}} + \boldsymbol{S_r^2} + \boldsymbol{S_{r+e_j}^2}$ where the last two terms $\boldsymbol{S_r^2}$ and $\boldsymbol{S_{r+e_j}^2}$ are constants, $\eta_1^2\boldsymbol{\sigma_1} + \eta_2^2\boldsymbol{\sigma_2} + \eta_3^2\boldsymbol{\sigma_3} = -2\sum_j \left(\boldsymbol{S_r} \cdot \boldsymbol{S_{r+e_j}}\right)\boldsymbol{\sigma_j} + const\ \sum_j \boldsymbol{\sigma_j} = -2\sum_j \left(\boldsymbol{S_r} \cdot \boldsymbol{S_{r+e_j}}\right)\boldsymbol{\sigma_j}$ because of $\sum_j \boldsymbol{\sigma_j} = 0$. Therefore, $\eta^\dagger \tau \eta = \left(\frac{\sqrt{3}}{2}(\eta_3^2 - \eta_2^2),\ \frac{1}{2}(2\eta_1^2 - \eta_2^2 - \eta_3^2)\right)$ and $\boldsymbol{m_3}$ only differ from each other by a constant factor of 2.

The norm of $\boldsymbol{m_3}$, $\langle|\boldsymbol{m_3}|\rangle$, is the nematic order parameter. If Z_3 rotation symmetry is present, $\langle|\boldsymbol{m_3}|\rangle$ will vanish, however, $\langle|\boldsymbol{m_3}|\rangle$ will become non-zero in the case of Z_3 rotation symmetry breaking. Meanwhile, we measure the nematic correlation, $C_{m_3}(r) = \langle \boldsymbol{m}_{3,i}\boldsymbol{m}_{3,i+r} \rangle$, in which the nematic director $\boldsymbol{m}_{3,i} = \boldsymbol{\sigma_1}\,\epsilon_{i,1} + \boldsymbol{\sigma_2}\,\epsilon_{i,2} + \boldsymbol{\sigma_3}\,\epsilon_{i,3}$ is defined on each lattice site *i*. By examining the behavior of $\langle|\boldsymbol{m_3}|\rangle$ and $C_{m_3}(r)$, we can identify the occurrence of nematic order. We also measure the spin correlation, $C_s(r) = \langle \boldsymbol{S}_i\,\boldsymbol{S}_{i+r} \rangle$, to investigate whether long-range spin order occurs. We also performed the calculations via the Binder ratio to compare with our temperature dependence of the nematic order parameter (Supplementary Information Note 17).

Phonon calculations. The first-principles simulations are performed using the Vienna ab initio Simulation Package (VASP). The atomic structure optimization is performed within the generalized gradient approximation (GGA) and Perdew–Burke–Ernzerhof (PBE) exchange-correlation functional. The Monkhorst–Pack k-mesh is set to 7 x 7 x 1 with a 550-eV kinetic cutoff energy. All structures are fully relaxed until the force on each atom is less than 0.02 eV/Å. To avoid spurious interactions between periodic images, a vacuum space is set to be more than 15 Å. Phonon calculations were performed by the supercell approach, wherein supercells containing 2 x 2 x 1 unit cells were used. The real-space force constants were calculated in the density-functional perturbation theory (DFPT) implemented in the VASP code, and phonon frequencies were calculated from the force constants using the PHONOPY code.

Data availability

Source data are provided with this paper. All other data that support this study are available from the corresponding authors upon reasonable request.