

Intertwined Polar, Chiral, and Ferro-Rotational Orders in a Homo-Ferro-Rotational Insulator

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Intertwined orders refer to strongly coupled and mutually dependent orders that coexist in correlated electron systems, often underpinning key physical properties of the host materials. Among them, polar, chiral, and ferro-rotational orders have been theoretically known to form a closed set of intertwined orders. However, experimental investigation into their mutual coupling and physical consequences has remained elusive. In this work, we employ the polar-chiral insulator Ni_3TeO_6 as a platform and utilize a multimodal optical approach to directly probe and reveal the intertwining among polarity, chirality, and ferro-rotational order. We demonstrate how their coupling governs the formation of domains and dictates the nature of domain walls. Within the domains, we identify spatial inversion symmetry as the operation connecting two domain states of opposite polarity and chirality, with a homo-ferro-rotational state serving as the prerequisite for these interlocked configurations. At the domain walls, we observe a pronounced enhancement of in-plane polarization accompanied by a suppression of chirality. By combining with Ginzburg-Landau theory within the framework of a preexisting homo-ferro-rotational background, we uncover the emergence of mixed Néel- and Bloch-type domain walls. Our findings highlight the critical role of intertwined orders in defining domain and domain-wall characteristics and open pathways for domain switching and domain-wall control via intertwined order parameters.

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I. INTRODUCTION

Polarity refers to directionality, while chirality denotes handedness [1–3]. Polar and/or chiral structures are ubiquitous in nature [4–8], spanning from fundamental particles to complex minerals and across physical, chemical, and biological systems. Remarkably, when polarity and chirality coexist within a single system, a third type of order, ferro-rotational order [9,10], naturally arises as a composite of the two. This ferro-rotational order, together with polar and chiral orders, form a closed set of intertwined orders: The presence of any two of these three orders necessitates

the existence of the third. Unlike polarity and chirality, ferro-rotational order is a relatively recent theoretical concept [10,11] and has only recently been realized experimentally [12–20]. It is pictorially represented by a closed-loop, head-to-tail arrangement of electric dipole moments, resulting in zero net polarization and preserved inversion symmetry but with a defined handedness. The order parameter characterizing this state is an axial vector moment $\vec{A}$, defined as the sum over cross products between the local dipole moments $\vec{P}_i$ and their respective position vectors $\vec{r}_i$, i.e., $\vec{A} = \sum_{i=1}^N \vec{r}_i \times \vec{P}_i$. Despite the conceptual link among polar, chiral, and ferro-rotational orders, direct experimental investigation into their interplay remains largely unexplored to date.

In this study, we use Ni_3TeO_6 as the material platform to investigate the intertwined relationship among polar, chiral, and ferro-rotational orders within structural domains and at domain walls. The crystal structure of Ni_3TeO_6 belongs to the polar-chiral point group 3, characterized by a single out-of-plane threefold rotational symmetry (C_3). It adopts the

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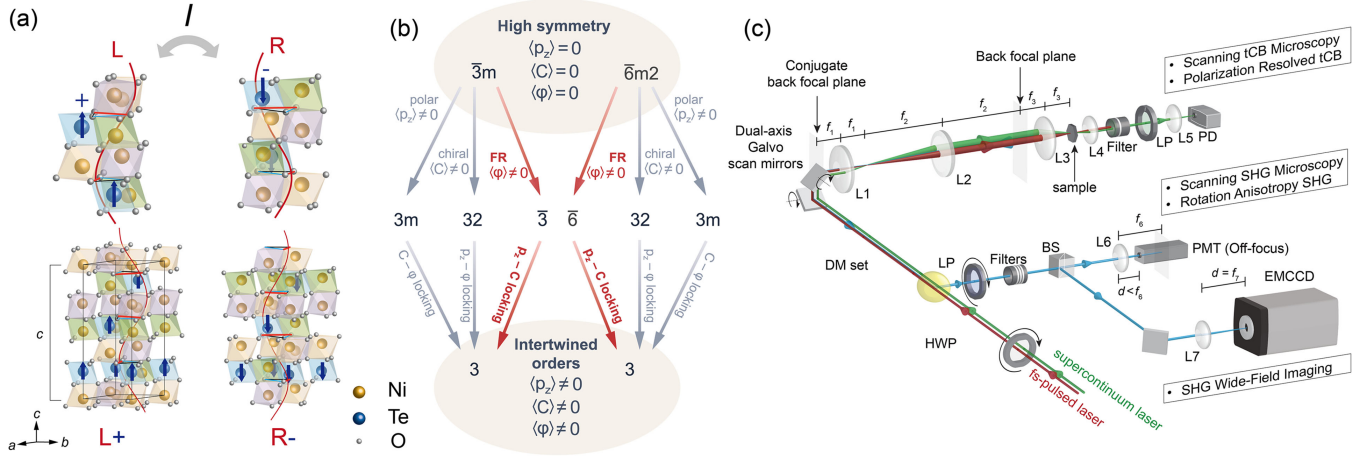


FIG. 1. Crystal structure, symmetry-breaking pathways, and multimodal optical platform. (a) Crystal structure of Ni_3TeO_6 , showing two domain states related by the spatial inversion (I) operation, each with interlocked polar and chiral orders. The upper panel illustrates a zoom-in visualization of crystal structure, while the bottom panel shows the unit cell of the Ni_3TeO_6 . Polarity is shown by the blue arrows; Te atoms shifting upwards correspond to polarity +, and vice versa. Chirality is indicated by the rotation of the triangular interfaces between oxygen cages following the red helices; clockwise rotation indicates chirality L and counterclockwise for R. (b) Symmetry-breaking pathways from two high-symmetry point group candidates, where the polar (p_z), chiral (C), and ferro-rotational (φ) orders are zero, to the low-symmetry point group, where each of the three order parameters is nonzero. The intermediate point groups each only contain one of the three order parameters, while in the second symmetry-breaking step, the remaining two order parameters emerge, locked by the extant order parameter. (c) Schematic of the multimodal optical platform. Scanning optical SHG microscopy, RA SHG, wide-field SHG imaging, scanning tCB microscopy, and polarization resolved tCB measurements can be performed in the same setup by adjusting corresponding optical components to suitable wavelengths based on the laser source used for the measurements.

$\text{A}_2\text{BB}'\text{O}_6$ structural motif, where the A_2B sites correspond to three crystallographically distinct Ni positions and the B' site corresponds to Te locations [21–23]. In Ni_3TeO_6 , the Ni^{2+} and Te^{6+} ions reside within highly distorted oxygen octahedra, and based on the local octahedral environments, the Ni sites are categorized as Ni^{I} , Ni^{II} , and Ni^{III} . Vertical displacements of the Ni^{2+} and Te^{6+} ions away from the centers of their respective oxygen cages generate a net polarization along the c axis, which can be either upwards (+) or downwards (−) as illustrated in Fig. 1(a). The distortions of the oxygen octahedra further give rise to unequal O-O bond lengths in the triangular interfaces between the $\text{Ni}^{\text{I}}/\text{Ni}^{\text{II}}\text{O}_6$ and $\text{Ni}^{\text{III}}/\text{TeO}_6$ layers, which can be categorized as longest (red), intermediate (black), and shortest (blue) as marked in Fig. 1(a). The orientation of these triangles, viewed along the c axis, rotates either clockwise or counterclockwise, leading to a chiral structure with either left-handed (L) or right-handed (R) character. In an as-grown Ni_3TeO_6 crystal, polarity and chirality are reported to be interlocked [23,24], yielding only L+ and R− domain states (or alternatively, only L− and R+).

To see the origin of the interlocked relationship between polarity and chirality, we trace the spontaneous symmetry-breaking pathways from high-symmetry parent point groups to the polar-chiral point group 3. The parent groups that forbid both polar and chiral orders are either $\bar{3}m$ or $\bar{6}m2$. Each of these two point groups can evolve into

point group 3 via one of three distinct symmetry-breaking pathways, each involving a two-step process, as illustrated in Fig. 1(b). In the high-symmetry parent phase ($\bar{3}m$ or $\bar{6}m2$), all three order parameters vanish: The polar order $\langle p_z \rangle$, chiral order $\langle C \rangle$, and ferro-rotational order $\langle \varphi \rangle$ are all zero. Furthermore, the interlocked coupling is established when their axes are aligned (Supplemental Material [25]). In the first symmetry-breaking step, one of these three orders develops: A polar order ($\langle p_z \rangle \neq 0$) leads to point group $3m$, a chiral order ($\langle C \rangle \neq 0$) yields 32 , and a ferro-rotational order ($\langle \varphi \rangle \neq 0$) results in $\bar{3}$ (or $\bar{6}$) depending on the parent group. In the second step, the remaining two orders emerge in an interlocked fashion, yielding the final low-symmetry point group 3, characterized by coexisting, intertwined polar, chiral, ferro-rotational orders, $\langle p_z \rangle \neq 0$, $\langle C \rangle \neq 0$, and $\langle \varphi \rangle \neq 0$. For Ni_3TeO_6 , where polarity and chirality are locked, we identify two possible symmetry-lowering pathways: $\bar{3}m \rightarrow \bar{3} \rightarrow 3$ and $\bar{6}m2 \rightarrow \bar{6} \rightarrow 3$, both of which involve the ferro-rotational order arising first [highlighted in red in Fig. 1(b)]. Crucially, the distinction between these two routes lies in the symmetry operation that relates polar-chiral domain states: spatial inversion for the first path versus a horizontal mirror plane for the second, as both can simultaneously reverse polarity and chirality in Ni_3TeO_6 . At present, open questions remain as to which of these symmetry operations governs domain switching in Ni_3TeO_6 and what structural features characterize the corresponding domain walls.

To approach these open questions, we employ a multimodal optical platform, shown in Fig. 1(c), to investigate as-grown, single-crystal homo-ferro-rotational Ni_3TeO_6 , with emphasis on selecting a suite of targeted optical techniques to probe specific order parameters and their characteristics. Rotation anisotropy (RA) electric dipole and electric quadrupole second harmonic generation (ED and EQ SHG) is used to resolve the full symmetry of the polar and ferro-rotational order, respectively [12,14,16,28–32], while polarization-resolved transmission circular birefringence (tCB) is applied to detect the presence of chiral order [33,34]. In addition, wide-field ED and EQ SHG imaging distinguishes domains with opposite polarities and ferro-rotationalities, respectively [16,28,35,36], and scanning ED and EQ SHG and tCB microscopies are used to spatially resolve the polar and ferro-rotational and chiral orders, respectively, both within domains and across domain walls. Importantly, all optical modalities are integrated within a single experimental setup, enabling measurements to be performed on the same sample spot (up to a diffraction-limited resolution accuracy of $2\ \mu\text{m}$) under identical conditions (e.g., during the same thermal cycle or under the same magnetic field sweep). This integrated, in-situ, multi-messenger optical approach is expected to provide a comprehensive characterization of materials with intertwined

symmetry-breaking orders, for example, the intertwined polar, chiral, and ferro-rotational orders in Ni_3TeO_6 .

II. EXPERIMENT AND DISCUSSION

A. Spatial inversion relationship

We began our investigation with RA SHG measurements on a single crystal Ni_3TeO_6 to probe its full symmetry properties. Polar plots of RA SHG taken in the linearly parallel polarization channel, shown in Fig. 2(a), correspond to three representative locations: two polar-chiral domains (labeled D1 and D2) and a domain wall (DW) between them. The RA SHG patterns from D1 and D2 appear nearly identical, exhibiting a clear C_3 rotational symmetry with a slight angular offset of approximately 7° from the crystallographic a and b axes. The rotation of the RA SHG patterns towards the identical direction over the multiple domains indicates the uniform ferro-rotational state across the entire sample (Supplemental Material [25]). This homo-ferro-rotational state likely originates from the fact that the ferro-rotational transition temperature is much higher than the crystal growth temperature [37]. This observation is consistent with the polar-chiral point group 3 assigned to Ni_3TeO_6 , as the lack of vertical mirrors and in-plane rotational axes in point group 3 naturally allows a

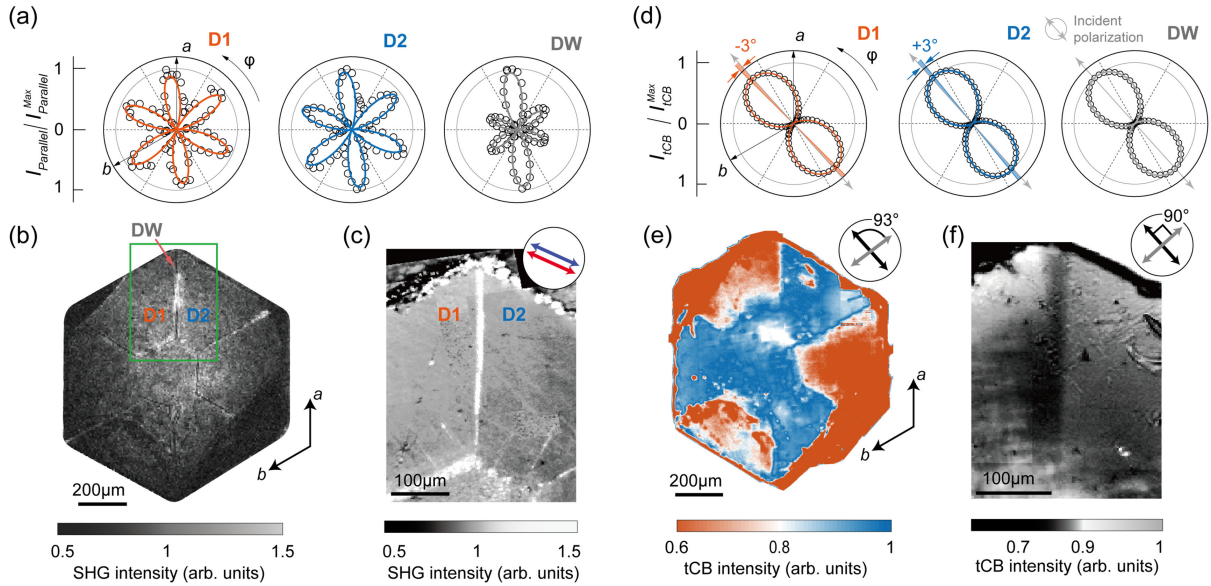


FIG. 2. Full symmetry properties of homo-ferro-rotational Ni_3TeO_6 probed by the combination of SHG and tCB. (a) Polar plots of RA SHG taken in the parallel channel under the normal incidence at domain 1 (D1), domain 2 (D2), and the DW, as shown in panel (b). The analyzer for the reflected SHG (400-nm) beam is corotated with the polarization of the incident fundamental (800-nm) beam. Black open circles are experimental data, and colored curves are their best fit with the derived expression for ED SHG (see Supplemental Material [25]). (b) Wide-field SHG image and (c) scanning SHG microscopy image captured with a fixed incident fundamental polarization and reflected SHG polarization as shown in the inset of panel (c). In panel (c), the polarization is chosen to maximize the measured SHG intensity. (d) Polar plots of angle-resolved tCB at D1, D2, and DW, with the incident polarization marked as a double-sided arrow. The colored curves are the best fit with polarization rotation, which gives the polarization rotations as marked in the figure by the red and blue shaded areas. Scanning tCB microscopy images captured with the analyzer angle set (e) at 93° and (f) 90° from the incident polarization. The angle for panel (e) is chosen to selectively suppress the transmission at D1, while panel (f) is chosen to allow similar transmission between both domains.

rotation of the RA SHG lobes away from the in-plane crystal axes. Because of either the spatial inversion or horizontal mirror relationship between D1 and D2, their ED SHG susceptibility tensors are related by a π - or 0-phase shift, respectively, $\chi_{ijk}^{\text{ED}}(\text{D1}) = -\chi_{ijk}^{\text{ED}}(\text{D2})$ or $\chi_{ijk}^{\text{ED}}(\text{D1}) = +\chi_{ijk}^{\text{ED}}(\text{D2})$, with $i, j, k = x$ and y for the normal incidence geometry; thereby, their resulting SHG electric fields differ by a sign or remain the same at each polarization angle of the RA SHG measurement. However, because RA SHG detects the optical intensity, which is quadratic in the electric field and insensitive to the overall optical phase shift, the RA SHG patterns of D1 and D2 remain indistinguishable. In contrast, the RA SHG pattern measured at a domain wall, which is approximately parallel to the a axis, shows a marked departure from the C_3 rotational symmetry. It features pronounced lobes near the wall direction, potentially suggesting the emergence of an in-plane polarization at domain walls to account for the broken C_3 symmetry (Supplemental Material [25]).

To map the polar-chiral domain distribution across this Ni_3TeO_6 single crystal, we performed the wide-field SHG imaging measurement, which is sensitive to the optical phase difference between adjacent domains [28]. As shown in Fig. 2(b), the wide-field SHG image reveals prominent dark lines of suppressed SHG intensity separating neighboring domains. These features arise from destructive interference between the SHG electric fields of domains D1 and D2, indicating a π -phase difference between their electric fields. This observation provides direct evidence that D1 and D2 are related by the spatial inversion operation, rather than by the horizontal mirror operation, pinning down the symmetry-breaking pathway of $\bar{3}m \rightarrow \bar{3} \rightarrow 3$. We further identified an A_u optical phonon mode of the point group $\bar{3}$ and the associated lattice displacement that breaks the inversion symmetry, providing a microscopic pathway for the emergence of polarity and chirality and their interlocking relationship (Supplemental Material [25]). Therefore, this wide-field SHG imaging not only shows the spatial distribution of structural domains in Ni_3TeO_6 single crystals but also unambiguously establishes the spatial-inversion relationship between the two domain states with interlocked polarity and chirality. Although the orientation of polarity cannot be directly determined from our SHG data, reversals in its direction across domain walls are clearly indicated by the destructive interference-induced dark lines observed in the wide-field imaging results.

B. Polarity and chirality at domain walls

To investigate the domain walls between polar-chiral domains in Ni_3TeO_6 , we performed scanning optical SHG microscopy, a technique specifically designed to minimize the destructive interference effect observed in wide-field

SHG imaging. As shown in Fig. 2(c), the scanning SHG image reveals a bright line with enhanced SHG intensities at the domain wall separating domains D1 and D2, in stark contrast to the corresponding dark line seen in Fig. 2(b). This enhanced SHG signal at the domain wall typically suggests a local increase in the inversion-symmetry breaking. Additionally, linear optical microscopy images taken at both the fundamental and SHG wavelengths show a uniform appearance across the single crystal, with no observable contrast between domains and domain walls (Supplemental Material [25]). This finding rules out variations in linear optical contrasts as the origin of the enhanced SHG signal. We therefore confidently attribute it to the presence of a stronger inversion-symmetry-breaking source, such as increased polarity or chirality localized at domain walls.

To pinpoint the source of this enhanced inversion symmetry breaking at the domain walls, we performed tCB measurements, probing linear polarization rotation, which is sensitive to chirality but insensitive to polarity. As shown in Fig. 2(d), we conducted polarimetric measurements of the transmitted light through domains D1 and D2 and the domain wall, using the same incident linear polarization oriented 45° clockwise from the a axis. The measurements were performed at room temperature, which is higher than the Néel temperature T_N at 52K [38–40]. Hence, we ruled out the potential contribution of the magnetic moment to the tCB response. The transmitted light remains linearly polarized at all three sites. However, its polarization direction varies distinctly: In D1, the polarization rotates by $+3^\circ$ (counterclockwise); in D2, by -3° (clockwise); and crucially, at the domain wall, by 0° (no rotation). These results confirm the opposite chirality between D1 and D2 and, furthermore, the absence of chirality along the out-of-plane direction, given our normal incidence geometry of tCB.

We further investigated the chiral domains and domain walls using scanning tCB microscopy. To enhance the contrast between domains with opposite chirality, we set the analyzer for the transmitted light at 93° relative to the incident polarization, allowing the transmission for D2 but suppressing that for D1 and producing a tCB image of this Ni_3TeO_6 crystal shown in Fig. 2(e). The spatial distribution of D1 and D2 closely matches that observed in the wide-field SHG image in Fig. 2(b), providing direct visualization of the interlocked relationship between polarity and chirality in Ni_3TeO_6 . To confirm the achiral nature along the out-of-plane direction at domain walls, we adjust the analyzer to be 90° from the incident polarization, a configuration that highlights regions with no polarization rotation. The resulting tCB image in Fig. 2(f) shows a dark line with zero polarization rotation aligned along the domain wall, consistent with the absence of chirality at domain walls. The contrast between suppressed tCB and

enhanced SHG signals at domain walls strongly indicates the change of the relationship between polarity and chirality from that within domains.

C. Symmetry evolution across domain walls

To better quantify the characteristics of domain walls in Ni_3TeO_6 , we analyzed the intensity profiles of the SHG and tCB signals along the direction normal to the domain-wall orientation, as shown in Fig. 3(a). These line profiles were obtained by averaging six linecuts extracted from the images in Figs. 2(c) and 2(f), respectively. While approaching the center of the domain wall, the tCB intensity, proportional to the optical rotation, gradually decreases, while the SHG intensity in the parallel channel increases markedly. These two traces exhibit clear opposite trends. Given that the tCB signal originates exclusively from chirality and approaches zero at the domain walls, the enhanced SHG signal observed in the same region is attributed primarily to polarity. The contrasting behavior between tCB and SHG suggests a scenario that the polarization is suppressed along the out-of-plane direction, which leads to the suppression in chirality and tCB but is enhanced in the in-plane directions, contributing to the enhancement of SHG. Additionally, the SHG line profile

reveals a polar domain-wall width of $2.9 \pm 0.7 \mu\text{m}$ (Supplemental Material [25]), while the tCB trace shows an achiral domain-wall width of $45.2 \pm 4.2 \mu\text{m}$, nearly an order of magnitude larger. The wide chiral domain wall with suppressed optical rotation was also visualized by white-light cross-polarized microscopy, confirming that the vanishing of the tCB signal and broadened domain walls are intrinsic (Supplemental Material [25]). This pronounced disparity in domain-wall widths between the two order parameters both motivates and is corroborated by our theoretical calculations, in which we assume polar order as the primary order parameter and chiral order as secondary (see Supplemental Material [25]).

In addition to the SHG intensity enhancement near the domain walls, the RA SHG patterns exhibit distinct evolutions across the domain wall, as shown in Fig. 3(b). In the upper panel, the RA patterns are normalized to the maximum intensity at the center of the domain wall (indicated by the black arrow in the middle polar plot), highlighting the intensity evolution across the domain wall. The lower panel displays the same set of RA SHG patterns but with each normalized to its own maximum intensity to more clearly illustrate changes in symmetry. As one approaches the center of the domain wall, the RA SHG

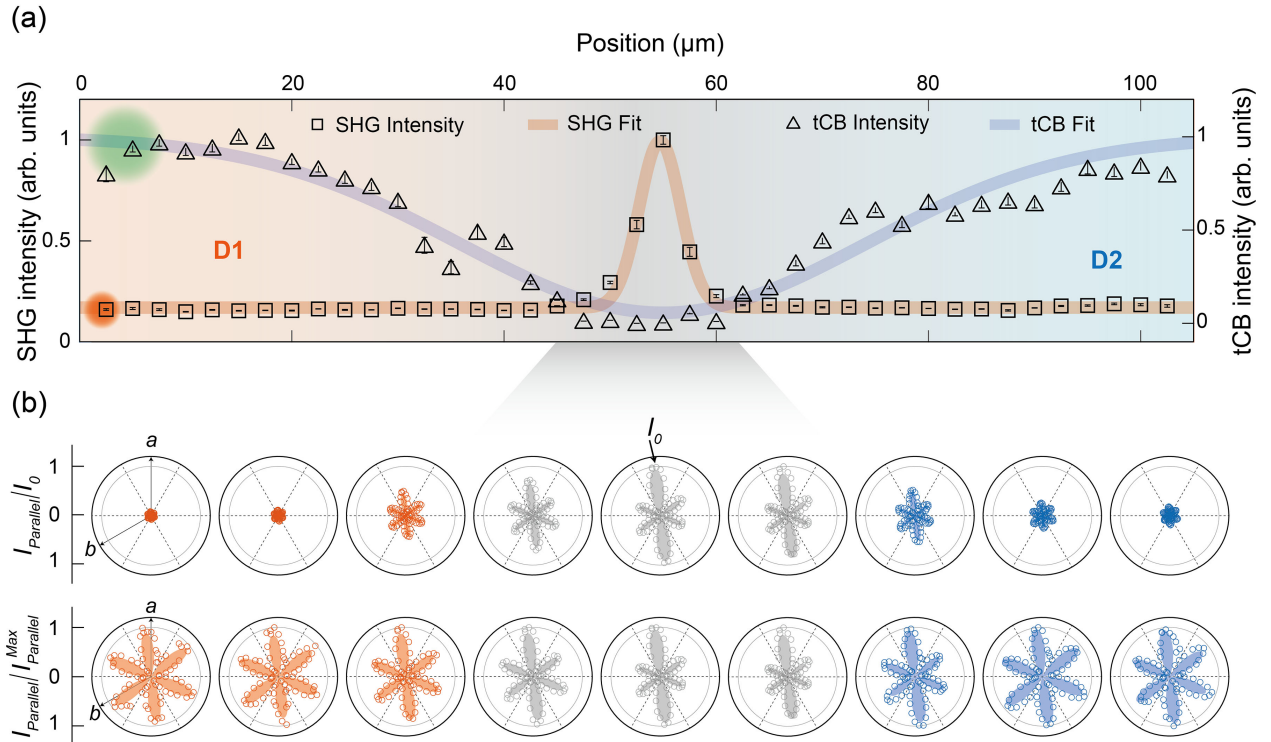


FIG. 3. Evolution of polar and chiral orders across the domain wall in Ni_3TeO_6 . (a) Spatial profile of SHG and tCB intensity across the domain wall as shown in Figs. 2(c) and 2(f). Open squares and triangles are experimental data, while the colored curves are the Gaussian fits for the normalized intensity. The red and green spots indicate the spot size of the laser beam with respect to the scale of the x axis. (b) Evolution of RA SHG across the domain wall. Open circles are experimental data, and shaded areas are their best fit with the derived expression for ED SHG based on symmetry. Top row is normalized by the maximal SHG intensity at the domain wall, and the polar plots on the bottom row are normalized with respect to the maximal SHG intensity at each position.

pattern evolves from six even lobes, indicative of the out-of-plane C_3 rotational symmetry within the domains, to three asymmetric lobe pairs of varying size, signifying the breaking of the C_3 symmetry at the domain wall. Since the out-of-plane polarization naturally preserves the C_3 symmetry, this C_3 symmetry breaking is therefore attributed to the emergence of in-plane polarization components at the domain wall (Supplemental Material [25]), echoing the simultaneous suppression of out-of-plane polarization but emergence of in-plane polarization indicated by the contrasting trend of SHG and tCB signals across the domain wall.

D. Mixed Bloch-Néel-type domain walls

Conventional polar domain walls are classified as either Bloch-type or Néel-type, depending on the orientation of the in-plane polarization relative to the domain wall: In-plane polarization is parallel in Bloch-type walls, while it is perpendicular in Néel-type walls. To determine the domain wall type in Ni_3TeO_6 , we employ Ginzburg-Landau theory informed by experimental observations.

The intertwined nature of polar, chiral, and ferro-rotational orders in Ni_3TeO_6 mandates a symmetry-allowed trilinear coupling term in the free energy, $F = \dots + g p_z C \varphi + \dots$, where g is the coupling constant. Given that Ni_3TeO_6 follows the symmetry-breaking pathway $\bar{3}m \rightarrow \bar{3} \rightarrow 3$, the ferro-rotational order (φ), which emerges first and sets the stage for the interlocked polarity and chirality, is the rigid order parameter—its modulation across the crystal incurs the largest energy cost among the three. As a result, φ remains uniform even across domain walls, allowing the trilinear coupling to be reduced to an effective bilinear form, $F = \dots + g_0 p_z C + \dots$, where $g_0 = g\langle\varphi\rangle$, with $\langle\varphi\rangle$ representing the spatially uniform ferro-rotational background. This effective coupling locks the relative signs of p_z and C , leading to (R+, L−) domains for $g_0 < 0$ and (R−, L+) for $g_0 > 0$.

Experimental SHG measurements reveal strong signals across domain walls, implying that polar order persists at the wall. We therefore model the system with an instability toward a polar order and calculate it with the sine-Gordon theory, under the ferro-rotational point group $\bar{3}$ for a polarization vector $\vec{p} = (p_x, p_y, p_z)$. The resulting analytical soliton solutions (Supplemental Material [25]) describe the spatial evolution of p_x , p_y , and p_z across the domain wall. As shown in Fig. 4(a), p_z switches sign across the wall and vanishes at its center, while p_x , p_y , and their combined in-plane magnitude $p_{\parallel} = \sqrt{p_x^2 + p_y^2}$ all peak at the center. Here, p_x , perpendicular to the wall, corresponds to the Néel-type, while p_y , parallel to the wall, corresponds to the Bloch-type. Their coexistence reveals that the domain wall is of mixed Bloch-Néel character. Moreover, the ratio p_x/p_y remains constant across the domain wall, indicating that the in-plane polarization vector maintains a uniform

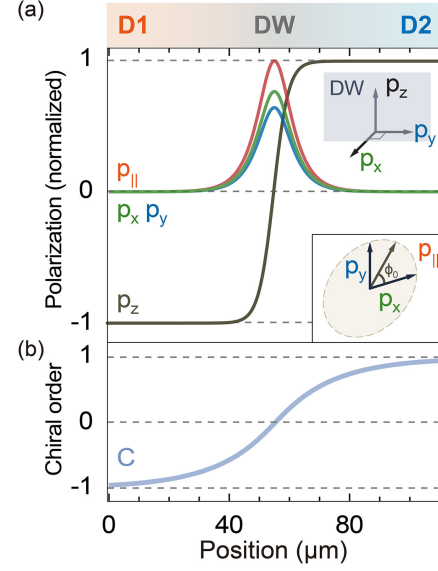


FIG. 4. Mixed Bloch-Néel-type domain wall shown by the Ginzburg-Landau theory. (a) Computed sine-Gordon solution (normalized) for the minimization of Ginzburg-Landau free energy for the polar order parameter components p_x , p_y , and p_z across the domain wall, as well as the in-plane component $p_{\parallel} = \sqrt{p_x^2 + p_y^2}$. As shown in the inset, p_x and p_y are related by a control parameter φ_0 which is not constrained by symmetry. Subsequently, the in-plane polar component is generally nonzero in both the x (Néel) and y (Bloch) directions, leading to a mixed Bloch-Néel character. (b) Chiral order (C_z) parameter profile over the domain wall, given by the sine-Gordon solution.

orientation, though not aligned with crystallographic axes [Fig. 4(a) inset]. This result is consistent with RA-SHG patterns, which exhibit intensity variation but preserve orientation when crossing the domain wall.

Using the spatial profile of p_z , we then calculate the chiral order parameter profile C by minimizing the free energy $F_C + F_{p_z-C}$, where F_C is the intrinsic chiral order energy and $F_{p_z-C} = g_0 p_z C$ for the bilinear coupling (Supplemental Material [25]). As shown in Fig. 4(b), the location dependence of C closely follows that of p_z but exhibits a broader domain-wall width. This finding confirms the interlocked nature of p_z and C even at domain walls, where both approach zero in contrast to their finite values inside domains. The broader width of the chiral domain wall further suggests that p_z acts as the leading order parameter while C forms the secondary order parameter.

III. CONCLUSIONS

In summary, we have revealed the uniform ferro-rotational background, evidenced by the identical rotational direction of RA SHG plots across the sample. Crucially, this rigid ferro-rotational background underpins the interlocked relationship between polarity and chirality within

domains, which plays a decisive role in shaping the domain structures and the mixed Néel and Bloch characters at domain walls. The intertwined relationship of polar, chiral, and ferro-rotational orders within domains and their decoupling characters at domain walls is elucidated by the combination of linear and nonlinear optics. Given the nascent stage of research into disentangling these intertwined orders, complementary experimental techniques are essential for developing a more in-depth and comprehensive understanding. Nonetheless, our findings provide key insights into strategies for domain and domain-wall control in systems exhibiting such intertwined orders. For instance, to alter the interlocking between polar and chiral orders, it is necessary to first switch the underlying ferro-rotational state. Likewise, tuning domain-wall properties or driving domain-wall motion requires access to and manipulation of this foundational ferro-rotational order. Recent demonstrations of THz phonon-excited control of ferro-rotational order point to a realistic pathway for switching the ferro-rotational background [20]. In Ni_3TeO_6 , such control should allow reconfiguration of the interlocked polar-chiral state and tuning of mixed Néel-Bloch walls. Furthermore, this framework of intertwined orders can be extended to the magnetic analog, where ferromagnetic, helimagnetic, and ferro-toroidal [35,41] orders similarly form a closed and interdependent set.

IV. METHODS

A. Ni_3TeO_6 single crystal growth

Ni_3TeO_6 single crystals were grown by the chemical vapor transfer method. The powder samples of Ni_3TeO_6 were synthesized first using the solid-state reaction method. High-purity powder of NiO (99.99%) and Te_2O_5 (99.99%) were mixed in stoichiometric ratios. The mixed powders were ground, pelletized, and then sintered in air at 700 °C for 72 hours in a box furnace, with two intermediate grinding steps. The resulting green powder samples were sealed in evacuated quartz tubes together with Cl_2 gas as a transport agent. The tubes were placed in a two-zone furnace for 7 days, where the hot and cold ends were maintained at 700 °C and 600 °C, respectively.

B. Multimodal optical characterization platform

Optical measurements, including SHG and tCB, were conducted within the multimodal optical platform shown in the schematic in Fig. 1(c).

The SHG measurements were conducted using a 200-kHz pulsed laser with the center wavelength at 800 nm and a pulse duration of 50 fs. For RA SHG and scanning SHG microscopy measurements, the laser was focused on the sample surface to a spot size of $5.0 \pm 0.4 \mu\text{m}$ full width at half maximum (FWHM), while a galvo mirror was used in conjunction with a 4f-imaging system to adjust the position of the laser within the focal plane while maintaining isotropic fluence. Since the SHG

signal scales with the square of the fundamental light intensity, the effective spot size of the SHG probe is estimated to be $3.5 \pm 0.3 \mu\text{m}$ FWHM. Wavelength-appropriate half waveplates and polarizers were used to define the incident and reflected polarizations. Reflected SHG light (400 nm) was collected by a photomultiplier tube after a set of lowpass and bandpass filters to reject the fundamental wavelength. The photomultiplier tube was intentionally placed before the lens focal plane. This defocused configuration prevented destructive interference between reflected signals from neighboring domains with opposite polarity and chirality and enabled the detection of an intrinsic response of the domain wall. For wide-field imaging, the incoming fundamental beam divergence was adjusted so that a large area of the sample was illuminated. The SHG light was mapped onto a two-dimensional electron-multiplying charge-coupled device (EMCCD) through the well-defined imaging system, where the SHG signal from adjacent domains within the diffraction limit will arrive at the same location on the EMCCD detector, leading to interference. In most SHG setups reported in the literature, detectors were placed right at the focal plane, which gave rise to a similar imaging condition as our wide-field imaging setup and enabled interference.

The tCB measurements were conducted using a super-continuum white light fiber laser equipped with an acousto-optic tunable filter set to a center wavelength of 517 nm. The incident laser was focused on the sample to a spot size of $7 \pm 0.6 \mu\text{m}$ FWHM, and the transmitted light was then focused onto an amplified Si photodetector after a set of bandpass filters to isolate the 517-nm wavelength.

C. X-ray diffraction measurements

X-ray diffraction was used to determine the crystal axes of the macroscopic specimen. Using a Rigaku SmartLab x-ray diffractometer, with a $\text{Cu-K}\alpha$ source (wavelength: 1.5406 Å) and a $\text{Ge}(220)$ monochromator, operated in the parallel-beam configuration, 2θ - ω scans (2θ : diffracted angle between the incident beam and the detector; ω : incident angle) were used to evaluate the lattice constant, diffraction angle, and orientation of the top facet. A ϕ scan (in-plane rotation) with in-plane stage rotation was used to determine the remaining crystal directions.

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W. Z., Y. A., and L. Z. conceived the idea and initiated the project. W. Z., Y. A., and X. G. performed the optical experiment, under the supervision of L. Z. J. Y. and S.-W. C. grew the Ni_3TeO_6 single crystals. T. C. performed the x-ray diffraction measurements to determine the crystal axes, under the supervision of J. T. H. N. A. performed the transmission electron microscopy measurements. K. S. performed the Ginzburg-Landau theory calculations. W. Z., J. H. Y., Y. A., and L. Z. analyzed the data and wrote the manuscript. All authors participated in discussions of the results.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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