



Three-dimensional quasi-periodic ferroelectric domain structure and its second harmonic generation effect in potassium tantalate niobate crystal

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Nonlinear photonic crystals, ferroelectric domains, quasi-phase-matching, nonlinear optical properties

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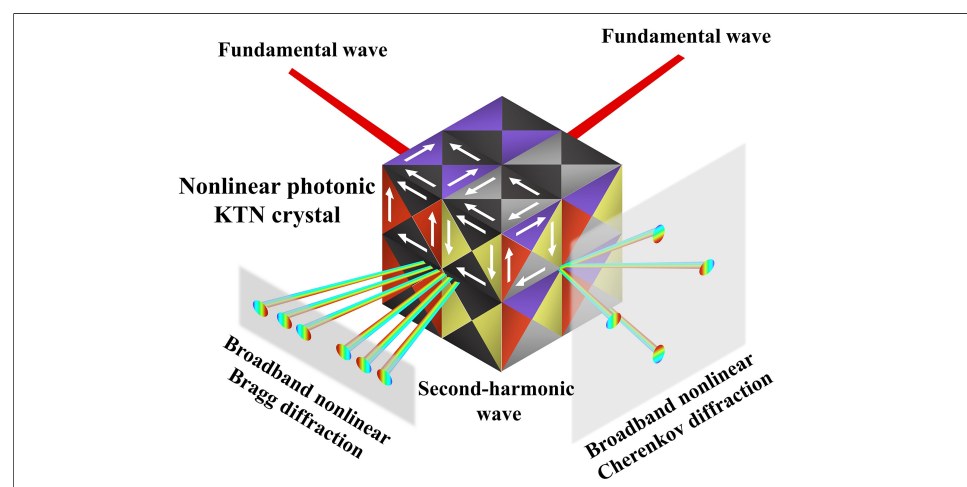
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Abstract

Nonlinear photonic crystals (NPCs) have garnered significant attention due to their capability to manipulate and enhance optical interactions via quasi-phase-matching. The periodic domain inversion structures inside NPCs, corresponding to periodically modulated second-order nonlinear coefficients $\chi^{(2)}$, enable the generation of light at new frequencies. However, due to the internal stress and defects, forming a structurally perfect superlattice in naturally grown crystals is challenging, and artificial engineering often entails high costs and complex procedures. In this study, we successfully fabricated a unique three-dimensional (3D) potassium tantalate niobate (KTN) NPC using a direct current poling treatment. Nonlinear Cherenkov diffraction occurs when the fundamental light propagates parallel to the poling direction, whereas nonlinear Bragg diffraction is observed when the light is incident perpendicular to the poling direction. By analyzing both the linear and nonlinear optical responses of the stably poled KTN crystal, we reveal the 3D

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distribution of ferroelectric domains, which advances the understanding of ferroelectric domain dynamics during the poling process and opens new avenues for research in anisotropic nonlinear optics and optoelectronic applications.

INTRODUCTION

The intrinsic properties of most crystals stem from the periodic arrangement of atoms and molecules. Leveraging this structure-property relationship, periodic ferroelectric domain structures with inverse polarization orientations are fundamental in defining nonlinear optical properties and have been extensively explored and engineered in nonlinear photonic crystals (NPCs)^[1–5]. In ferroelectric crystals, periodic domain inversion corresponds to the modulation of the second-order nonlinear coefficient $\chi^{(2)}$, thereby enabling quasi-phase-matching (QPM) in one-, two-, and even three-dimensional (3D) configurations^[6–10]. Notable examples include periodically poled lithium niobate (PPLN)^[11–13], barium calcium titanate fabricated via femtosecond laser direct writing^[14], and naturally grown potassium tantalite niobite^[15], all of which have generated various types of second-harmonic (SH) light, expanding the application prospects in the structural analysis of matter, nonlinear beam shaping, and nonlinear imaging^[16–20]. However, the presence of internal stress and defects poses significant challenges to the realization of a perfect superlattice in naturally grown ferroelectric crystals^[21]. Moreover, achieving second harmonic generation (SHG) with a structurally simple pattern necessitates stringent conditions and incurs high costs^[14,22], limiting practical applications. Thus, exploring innovative strategies for the design and fabrication of nonlinear photonic crystals holds significant potential for broadening their application avenues to diverse optical and photonic technologies.

Potassium tantalate niobate ($\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$) crystals have attracted considerable attention owing to their giant broadband refractive index, wide transmission range, and exceptional nonlinear optical responses^[23–26]. The spontaneous polarization (P_s), whose directions are parallel to three crystallographic axes in tetragonal-phase KTN crystals^[27], enables the formation of complex rotational domains. These domain structures enrich the 3D reciprocal lattice vectors (RLVs), thereby effectively relaxing the stringent constraints on crystal cutting direction and incident light polarization in QPM processes^[28,29]. Additionally, near the Curie temperature (T_C), a phase transition occurs from the ferroelectric tetragonal phase to the paraelectric cubic phase. By precisely controlling the tantalum-to-niobium ratio during crystal growth, the T_C can be tailored^[30], enabling the fabrication of nonlinear photonic crystals adaptable to diverse optical applications^[31,32]. A high conversion efficiency of up to 39% at 1,030 nm has been achieved in periodically poled KTN (PPKTN)^[24], and nonlinear Cherenkov radiation free from diffraction and chromatic walk-off has been realized in KTN:Li crystal^[33]. Wen *et al.* elucidated the relationship between SHG properties and ferroelectric domain structures through broadband nonlinear Bragg diffraction in KTN crystals^[34]. Given the structural flexibility and diversity of KTN crystals, they are considered a highly promising nonlinear optical platform for the development of novel photonic crystals.

In this work, we induced a novel 3D KTN NPC solely through direct current (DC) poling. Broadband Nonlinear Cherenkov diffraction and nonlinear Bragg diffraction were observed when the fundamental light was incident along different crystal axes. According to QPM theory and domain width distribution obtained from the polarizing light microscope (PLM) image, the mechanism of broadband nonlinear Bragg diffraction in the fundamental wavelength (FW) from 850 to 1,375 nm was explained. Furthermore, based on the multi-polarization SHG effects, we demonstrated the feasibility of nonlinear optical imaging in the stably poled KTN. By correlating the 3D SHG patterns with the real-space imaging of visible light, the ferroelectric domain structures were clearly revealed. This unique 3D nonlinear KTN photonic crystal holds great promise for advancing research in anisotropic nonlinear optics and the development of tunable photonic devices.

MATERIALS AND METHODS

Materials preparation

First, high-quality KTN single crystals were grown using the top-seeded solution growth (TSSG) method. The KTN sample, measuring $0.30 \times 1.05 \times 1.96 \text{ mm}^3$ ($x \times y \times z$), was characterized for its dielectric properties by an LCR meter (E4980A, Keysight, USA). All six planes of KTN were polished, followed by annealing from 400°C to room temperature ($T_{\text{room}} = 25^\circ\text{C}$) at a cooling rate of $0.5^\circ\text{C}/\text{min}$ to eliminate stress induced by polishing. Subsequently, the two x - y planes of KTN were coated with golden electrodes via magnetron sputtering (VTC-16-D, Ke Jing, China) and DC poling treatment ($1 \text{ kV}/\text{mm}$ for 8 h, at T_{room}) was conducted on the crystals along the z -axis. After removing the electric field, the sample was left undisturbed for several days to achieve a stably poled state.

Optical property measurements

The transmission spectrum was measured using a grating spectrometer (Omni- λ , Zhuo Li, China). The SHG response and nonlinear image propagation experiments were conducted using a mode-locked Nd: YAG nanosecond laser (EO-1064-N, CNI, China, $\lambda = 1,064 \text{ nm}$, pulse width = 8 ns, repetition frequency = 20 kHz). Broadband nonlinear properties were investigated by applying an optical parametric oscillator (OPO, LS-2145, LOTIS TII, Belarus, pulse width = 10 ns, repetition frequency = 10 Hz). Far-field diffraction imaging was performed using a continuous-wave laser (04-01 series, Cobolt CalypsoTM, Sweden, $\lambda = 491 \text{ nm}$). Light intensity distribution was recorded by a charge-coupled device (CCD, GS3-U3-41C6M-C, FLIR, USA), while images of nonlinear optical spots were captured with a camera (EOS M, Canon, Japan). In SHG experiments, the processed crystal was positioned at the focal point of a convex lens to enhance the SHG intensity and the distance l between the sample and the screen was minimized to reduce the divergence effect of the lens. The polarization state of the fundamental light was modulated using linear polarizers and a half-wave plate. The optical filter was employed to remove the fundamental wavelength. The observation screen was made of quartz glass with one frosted surface (for diffuse scattering) and one polished surface. The Galilean-type beam expander was utilized to ensure the fundamental wavelength beam size was sufficiently large for carrying object image information of templates. The “X” and “Y” templates were produced with stainless steel by laser machining techniques. The polarization beam splitter (PBS) was used to split an incident light beam into two orthogonally polarized beams: One beam is P -polarized (parallel to the plane of incidence), the other beam is S -polarized (perpendicular to the plane of incidence). The beam splitter (BS) was introduced to combine two light beams into the same optical path without introducing polarization-dependent effects.

RESULTS AND DISCUSSION

Figure 1A shows the temperature-dependent dielectric constant of the KTN crystal. The crystal exhibits a T_C of 101°C , in a tetragonal phase ($P4mm$) at T_{room} . The composition ($\text{KTa}_{0.51}\text{Nb}_{0.49}\text{O}_3$) can be concluded according to the relation of Nb concentrations x vs. T_C [$T_C (^\circ\text{C}) = 676x - 241.15$] [30]. The SHG image of an annealed KTN (T -phase) exhibits a cross pattern due to the disordered distribution of ferroelectric domains [34,35], while a fully poled state with a single domain structure results in a single SHG spot [36]. In the stably poled state, however, three pairs of non-collinear SHG spots emerge on the plane parallel to the poling direction (z -axis) [Figure 1B]. These spots exhibit a strong polarization dependence. We investigated the polarization relationship between SHG and FW through the relative intensity of SHG. The linear polarized light along the z -axis was obtained using a linear polarizer (P_1) and the half-wave plate (HWP) was introduced to control the polarization angle θ of fundamental light (from 0° to 180° , with an interval of 30°) by changing the angle between z -axis and optical axis of HWP without a power change of FW [Figure 1C]. We found that the four SHG spots (Type-0 and Type-I) near the center spot exhibited only a horizontal polarization component (extraordinary light), whereas the two spots (Type-II) farthest from the center

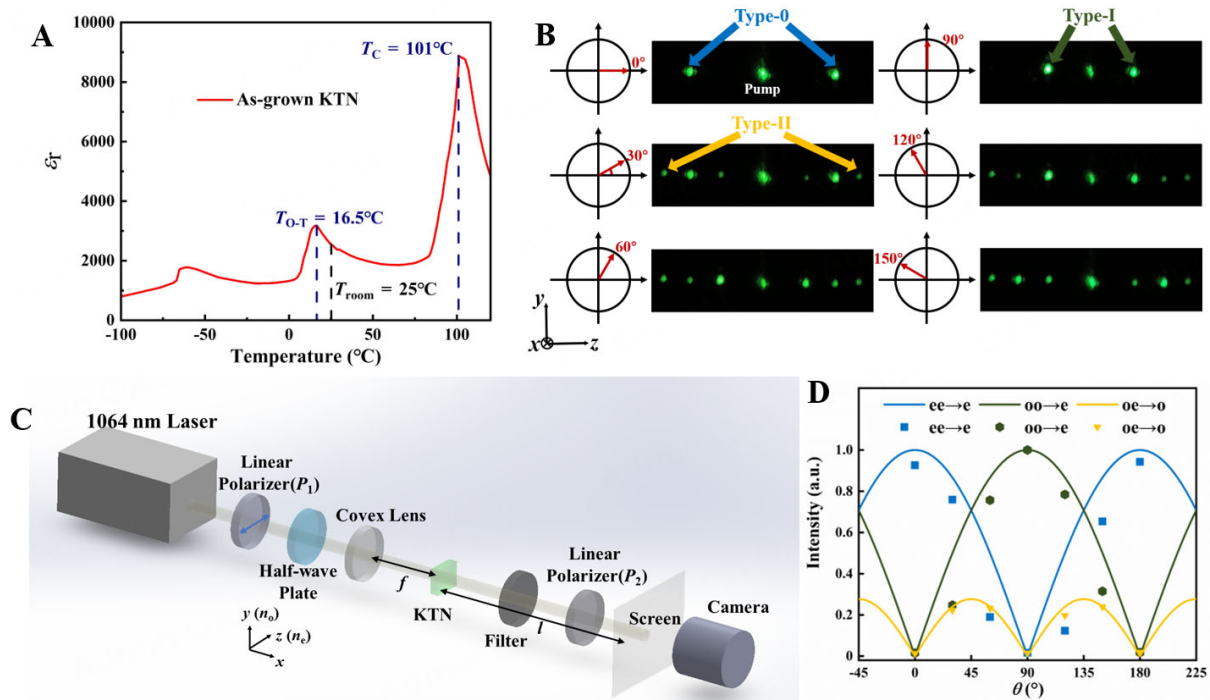


Figure 1. (A) The relative dielectric permittivity (ϵ_r) vs. temperature at 1 kHz. T_{O-T} : the orthorhombic-to-tetragonal phase transition temperature; T_{room} : the room temperature; T_C : Curie temperature, the tetragonal-to-cubic phase transition temperature. (B) Different polarization states (left) of the fundamental light incident along the x-axis and the corresponding SHG spots (right). (C) Optical path diagram for investigating the polarization dependence of SHG. (D) Relative intensity of SHG spots in different polarization states.

exhibited only a vertical polarization component (ordinary light) when another linear polarizer (P_2) was introduced into the optical path. The relative intensity of the SHG spots was obtained from pixels of the same size in the grayscale-processed RGB image without saturation. The Type-0 intensity is proportional to the square of the horizontal polarization component (E_z) of the fundamental light, the Type-I is proportional to the square of the vertical component (E_y), and the Type-II is proportional to the square of the smaller of the horizontal and vertical components [Figure 1D]. Therefore, there are three QPM processes in the y - z plane of stably poled KTN: $ee \rightarrow e$ (Type-0), $oo \rightarrow e$ (Type-I), and $oe \rightarrow o$ (Type-II) [Figure 2A, left part].

The P_s of the tetragonal samples is oriented along six possible directions ($[100]_C$, $[010]_C$, $[001]_C$, and their opposites) and periodically distributed up-down P_s can provide RLVs to satisfy the QPM conditions^[27]. The RLVs (G_o , G_p , G_{II}) are all parallel to the poling direction (z -axis), corresponding to the periodic alternating arrangement of P_s along the y -axis^[37,38] [Figure 2B, left part]. The absence of G along the y -axis means that P_s toward $+z$ and $-z$ originally have been reoriented to the same direction by the electric field E ^[34]. The real-space imaging, obtained from direct-field transmission^[39,40] [Supplementary Figure 1], shows that the domain regions (bright stripes) and domain walls (black dashed lines) appear alternately in the y - z plane of Figure 2C. The domain walls are vertical and in a quasi-periodic arrangement. Considering the uniformity of P_s along the z -direction, the formation of domain walls parallel to the crystallographic orientation (y -axis) originates from opposite P_s along the y -axis in tetragonal KTN crystals^[41]. Given the high consistency between the linear and nonlinear responses, the domain structures in the y - z plane shown in Figure 2B are considered reliable.

The QPM processes (Type-0, Type-I, and Type-II) were achieved in a range from 850 to 1,375 nm of FW. Using the length l and the distance d , between the side spots and the central spot, the SHG emission angles θ_{out} are calculated ($\tan \theta_{out} = d/l$). The theoretical output angles θ_{out}^{theo} for the three pairs of SHG spots are

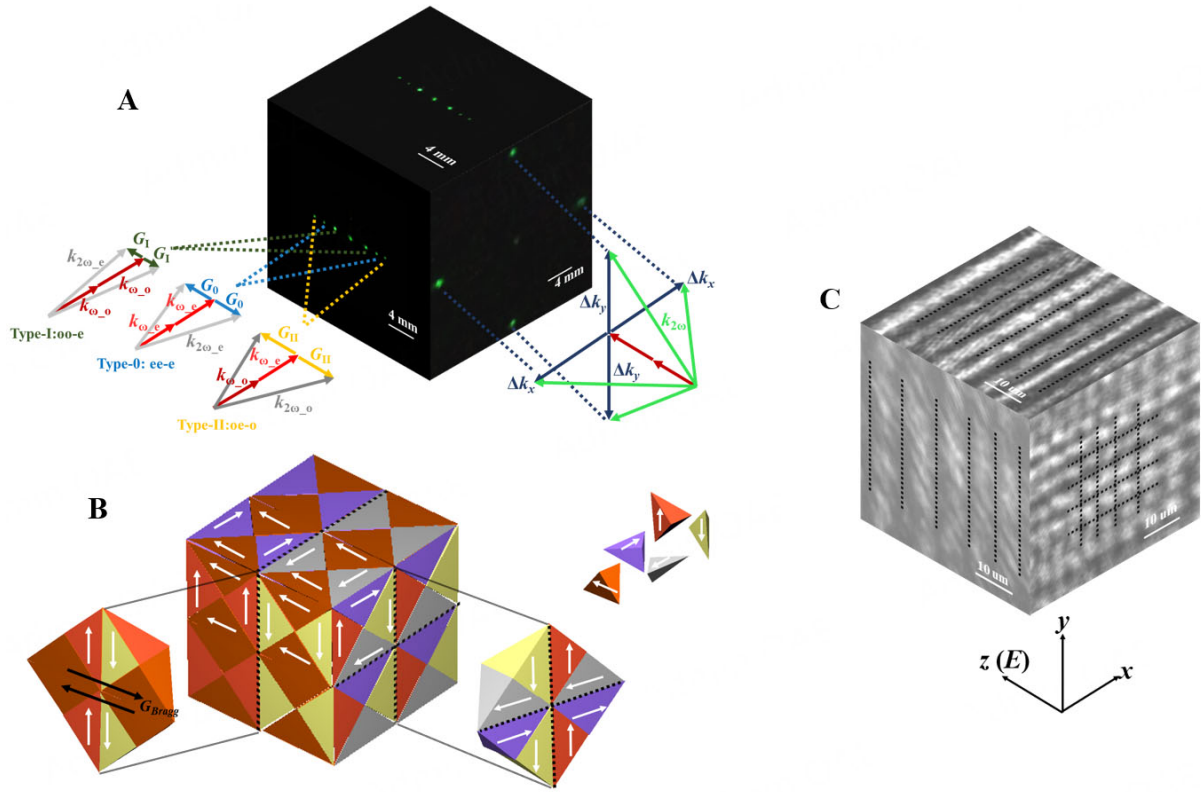


Figure 2. Stably poled KTN. (A) SHG patterns when the FW is incident along the x, y, and z directions. The left part shows three QPM processes of nonlinear Bragg diffraction, and the right part shows the phase-matching condition of nonlinear Cherenkov diffraction. $l = 45$ mm. Scale bars = 4 mm. (B) Self-assembled domain structure. The left part shows ferroelectric domains flipped along the y-axis, producing in-plane G_{Bragg} parallel to the z-axis, and the right part shows ferroelectric domains flipped along the x- and y-axes, with 180° domain walls along the x- and y-axes. White arrows represent the orientation of P_s and the tetrahedra in the insert are the building blocks of this structure. (C) Real-space imaging. The black dashed lines are in-plane domain walls in (B and C).

obtained, based on the QPM conditions [Equation (1)], Sellmeier equations^[42] [Supplementary Equation (1)], and Snell's law ($n_{2\omega} \sin \theta_i = \sin \theta_{\text{out}}^{\text{theo}}$, $i = 0, \text{I}, \text{II}$), in which n_{ω} , $n_{2\omega}$ are the refractive indices of fundamental light and SHG, respectively, and θ_i are the QPM angles inside the stably poled KTN.

$$\begin{aligned} \tan \theta_0 &= \frac{|G_0|}{|2k_{\omega-e}|} = \frac{n_{2\omega-e}}{n_{\omega-e}} - 1 \\ \tan \theta_{\text{I}} &= \frac{|G_{\text{I}}|}{|2k_{\omega-o}|} = \frac{n_{2\omega-o}}{n_{\omega-o}} - 1 \\ \tan \theta_{\text{II}} &= \frac{|G_{\text{II}}|}{|k_{\omega-o} + k_{\omega-e}|} = \frac{n_{2\omega-o}}{n_{\omega-o} + n_{\omega-e}} - 1 \end{aligned} \quad (1)$$

Figure 3A shows that the θ_{out} points match the $\theta_{\text{out}}^{\text{theo}}$ fitting curve, suggesting that the spots on the y-z originate from nonlinear Bragg diffraction^[34,43]. Notably, since the poling direction aligns with the fourfold axis of the primitive cell, the self-assembled structures of ferroelectric domains formed in stably poled KTN are similar in the planes parallel to the z-axis [Figure 2B]. Therefore, the SHG properties and linear response of domains in the x-z plane are like those in the y-z plane, as shown in Figure 2A and C.

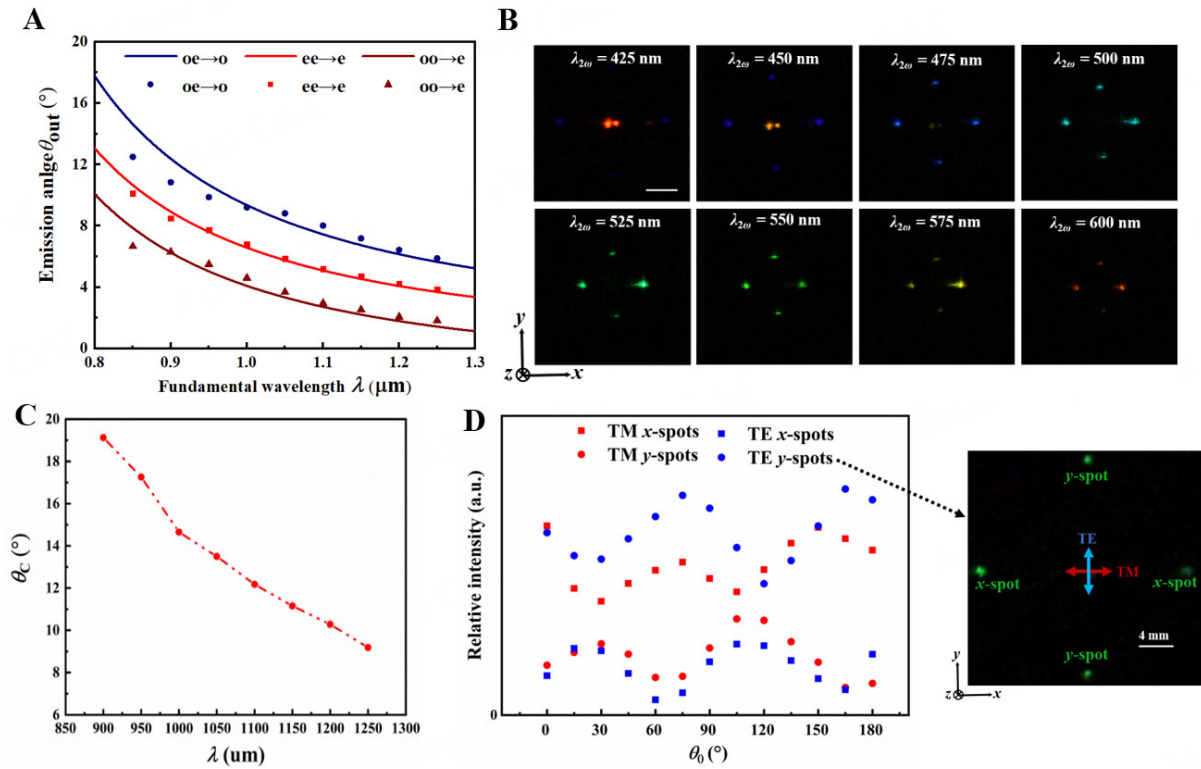


Figure 3. (A) Emission angles of Type-0, Type-I and Type-II vs. fundamental wavelength. (B) Nonlinear Cherenkov diffraction at different SHG wavelengths (425 to 600 nm, with an interval of 25 nm). $l = 82.3$ mm. Scale bar = 20 mm. (C) Cherenkov angle vs. fundamental wavelength. (D) Relative SHG intensity vs. the fundamental polarization angle θ_0 , measured with respect to the x- and y-axes. The insert shows the analyzed points in the x-y plane.

Making the fundamental light propagate along the z-axis results in a diamond-like distribution of SHG [Figure 2A, x-y facet], formed by two spots in the x direction and two spots in the y direction. As reported in Figure 3B and C, SHG output has a wide range from 425 to 600 nm, and the measured values of emission angle $\theta_c(\lambda)$ are significantly larger than those of nonlinear Bragg diffraction at the same FW wavelengths [Supplementary Table 1], corresponding to nonlinear Cherenkov diffraction^[44]. It is noted that the Cherenkov mechanism is unrelated to QPM^[33]. Its phase-matching condition is fulfilled when the longitudinal phase mismatch is zero and the transverse phase mismatch ($\Delta k_x, \Delta k_y$) can automatically be compensated by ferroelectric domain walls^[44]. In the x-y plane, the phase-matching condition for the SHG spots distributed along the x-/y-axis is satisfied by y-/x-aligned 180° domain walls^[45]. The real-space imaging of the x-y plane in Figure 2C exhibits a quasi-square lattice arrangement of bright spots and two sets of orthogonally arranged 180° domain walls. Based on the relationship between domain wall formation and domain orientations, there are quasi-periodically opposite-oriented ferroelectric domain structures along both the x and y directions, indicating the existence of a quasi-periodic vortex structure of ferroelectric domains^[15,33,40]. For light incident along the optical axis, its polarization states are characterized using transverse electric (TE) and transverse magnetic (TM) modes. The SHG polarization intensity is $P_i(2\omega) = d_{ijk}E_j(\omega)E_k(\omega)$, where $E(\omega)$ is the pump field and d_{ijk} is the nonlinear optical susceptibility tensor. As reported in Figure 3D, the nonlinear polarization intensity of the TE component of the x-spots follows: $P_y(2\omega) \propto (E_y(\omega))^2$, which reveals the periodically distributed P_s with $\pm y$ orientations in x-y plane. The TM component of the x-spots follows: $P_x(2\omega) \propto (E_x(\omega))^2$, corresponding to the periodically distributed P_s with $\pm x$ orientations^[33]. Since the z-axis is the fourfold axis of the primitive cell in KTN crystals, the polarization response characteristics of the y-spots are similar to those of the x-spots. The polarization response characteristics of the nonlinear Cherenkov diffraction further confirm the existence of vortex domain structures [Figure 2B, x-y facet].

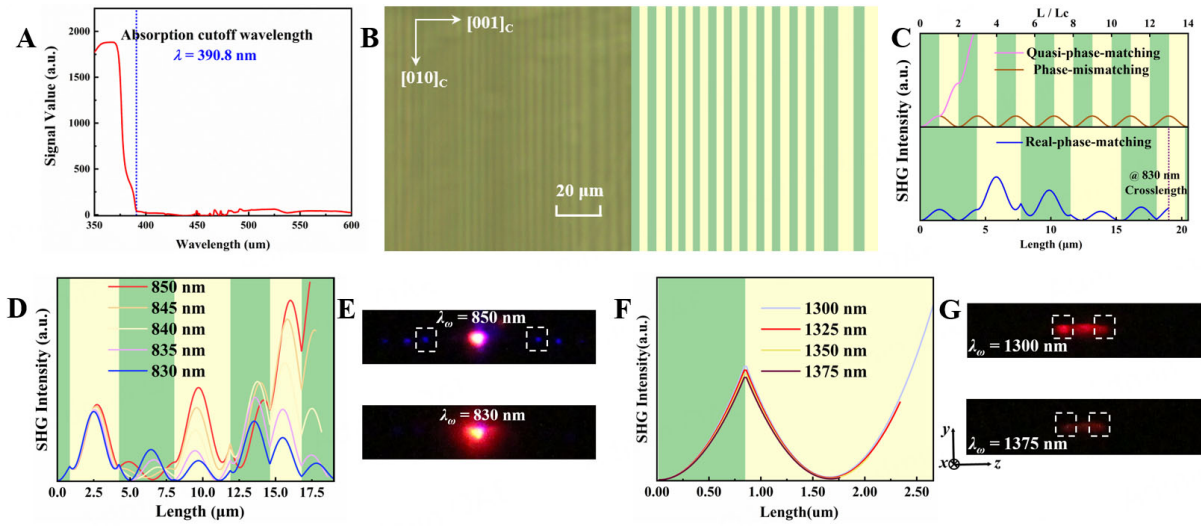


Figure 4. (A) Transmission spectrum of the stably poled KTN. (B) Polarization microscope image and corresponding d_{eff} distribution diagram (dark color represents positive and light color represents negative) of stably poled KTN in the y - z plane. (C) Schematic of the relationship between SHG (Type-I, at 830 nm FW) intensity and propagation distance under quasi-phase-matching, phase-mismatching, and real-space-matching. The intensity range is the same in both the upper and lower parts. Schematic of SHG change vs. fundamental wavelength (D) from 830 to 850 nm, with an interval of 5 nm, and (F) from 1,300 to 1,375 nm, with an interval of 25 nm. SHG spot patterns at (E) $\lambda_{\omega} = 830$ and 850 nm, (G) $\lambda_{\omega} = 1,300$ and 1,375 nm. The white dashed box encloses the SHG spots generated by the Type-I QPM process.

Generally, broadband SHG capability is believed to originate from the broad range of ferroelectric domains. However, the period Λ required to satisfy broadband QPM conditions varies continuously. For the 415 to 687.5 nm SHG output, which is not absorbed by the sample [Figure 4A], Λ ranges from 2.924 to 53.074 μm in the $oo \rightarrow e$ process. Such a multitude of periodic structures nested into each other is impractical in real situations. The nonlinear Bragg diffraction arises from perfect quasi-phase-matching (QPM), that is, the reciprocal lattice vector exactly compensates the phase mismatch between the fundamental and second-harmonic wavevectors ($\Delta k = k_{2\omega} - 2k_{\omega} = G$)^[15]. The RLVs (G_o , G_p , G_{II}) are aligned along the z -axis, which dictates that only y -polarized ferroelectric domains contribute to the nonlinear optical interaction. We explain the upper and lower limits of the SHG wavelength for Type-I, based on the domain distribution obtained from PLM. Given that P_s along the y -axis on both sides of the domain walls are opposite, the effective nonlinear optical susceptibility d_{eff} in the y - z plane exhibits an alternating positive and negative distribution^[46] [Figure 4B]. The optical interaction length L_{int} along the z -axis in the y - z plane can be calculated based on the crystal thickness t and θ_i ($L_{int} = t/\tan\theta_i$) [Supplementary Figure 2]. According to the relationship between the electric field of fundamental light and SHG (E_{ω} and $E_{2\omega}$) [Equation (2)], SHG variations in QPM, phase-mismatching (PMM), and real-phase-matching (RPM) at 830 nm FW are shown in Figure 4C. The situations in both PMM and RPM are similar, as neither can achieve an effective output of 415 nm light.

$$\frac{dE_{2\omega}}{dz} = i \frac{2\omega d_{eff}}{cn_{2\omega}} E_{\omega}^2 e^{-i|G_1|z} \quad (2)$$

The coherence length L_c of the 830 nm fundamental wave is 1.562 μm , while the minimum domain width in statistical analysis is 1.977 μm [Supplementary Figure 3]. As the wavelength rises, L_c increases correspondingly [Supplementary Figure 4 and Supplementary Equation (2)], leading to better matching with the domain distribution, which enables SHG signal output, as shown in Figure 4D and E. Regarding the upper limit of SHG, the intensity gradually decreases due to the reduction in interaction length with

increasing wavelength [Figure 4F and G].

Notably, the quasi-periodic structure is supposed to originate from growth striations caused by a built-in spatially periodic oscillation in composition^[39,47]. The oscillation in composition can fabricate the flexoelectric field E_{flexo} in KTN crystals, which can control domain structures by inducing local polarization orientation^[21,48]. However, the non-uniform growth temperature of cross-section and convective instabilities during the crystal pulling process introduce thermal stress that affects the domain configuration underlying the growth striations^[32,49]. The application of a direct current electric field facilitates ferroelectric domain reconfiguration through domain wall migration, which relaxes the thermomechanical stresses^[50]. The domains are oriented uniformly with the direction of the applied electric field (fully poled state). With the electric field removed, some domains return to their original positions due to the E_{flexo} (stably poled state), which can minimize the global free energy of the system^[34]. Therefore, the approximately periodic striation grating is supposed to emerge with its primal morphological characteristics.

Nonlinear imaging has long been a focal point in the applications of nonlinear photonic crystals^[51–53]. Building on the multi-polarization SHG effect shown in Figure 1B, we explored the nonlinear imaging potential of stably poled KTN crystals. A beam-expanded 45°-polarized 1,064 nm laser is split into the *P* (extraordinary) and *S* (ordinary) light by a PBS, to carry the object image information of the “X” and “Y” templates, respectively [Figure 5A]. Then, these two beams are combined into the same optical path along the *x*-axis by a BS and are focused into the stably poled KTN crystal. The Fast Fourier Transform (FFT) of the Collins formula was introduced to fit the SHG distribution, and the initial FW is set as a plane wave with a Gaussian distribution. Simulation results shown in Figure 5B present harmonic patterns at the CCD receiving plane, specifically, the e-channel (ee → e) carrying “X” information, the o-channel (oo → e) carrying “Y” information, and the mix-channel (oe → o) formed by the smaller value points of the “X” and “Y” distribution. Figure 5C–E reports the CCD images of the three channels. The discrepancies between real SHG images and simulation patterns are primarily due to the FW beam expansion, which increases the laser transmission distance, causing the fundamental light to more closely resemble spherical light. However, the “X” and “Y” CCD images still retain Gaussian distributions, with their morphologies exclusively determined by the image information encoded in the extraordinary and ordinary light components, respectively. The intensity output of the mix-channel corresponds to the overlapping regions of SHG patterns from the o-channel and e-channel [Figure 5C–E, red box regions], and there is no output outside the region, which is consistent with Type-II QPM conditions: the simultaneous presence of ordinary and extraordinary light. Based on this relationship and multi-wavelength SHG effects, designing different intensity distributions in the o- and e-channels can achieve rich and diverse nonlinear information transmission in the mix-channel.

CONCLUSIONS

In summary, we obtain a novel 3D KNT NPC just by poling treatment. The self-assembled ferroelectric domain structures enable broadband QPM, facilitating both nonlinear Cherenkov and Bragg diffraction. The mechanisms behind the broadband Bragg diffraction effect are well explained by the domain distribution in planes parallel to the poling direction. This 3D domain configuration advances the understanding of the self-assembled dynamics of ferroelectric domains in stably poled KTN and highlights the potential of KTN-based NPCs in anisotropic nonlinear optics and tunable photonic devices. Additionally, by leveraging the polarization-dependent SHG effects, we successfully demonstrated multi-channel nonlinear imaging, which opens new avenues for advanced optical information processing and multiplexed imaging applications.

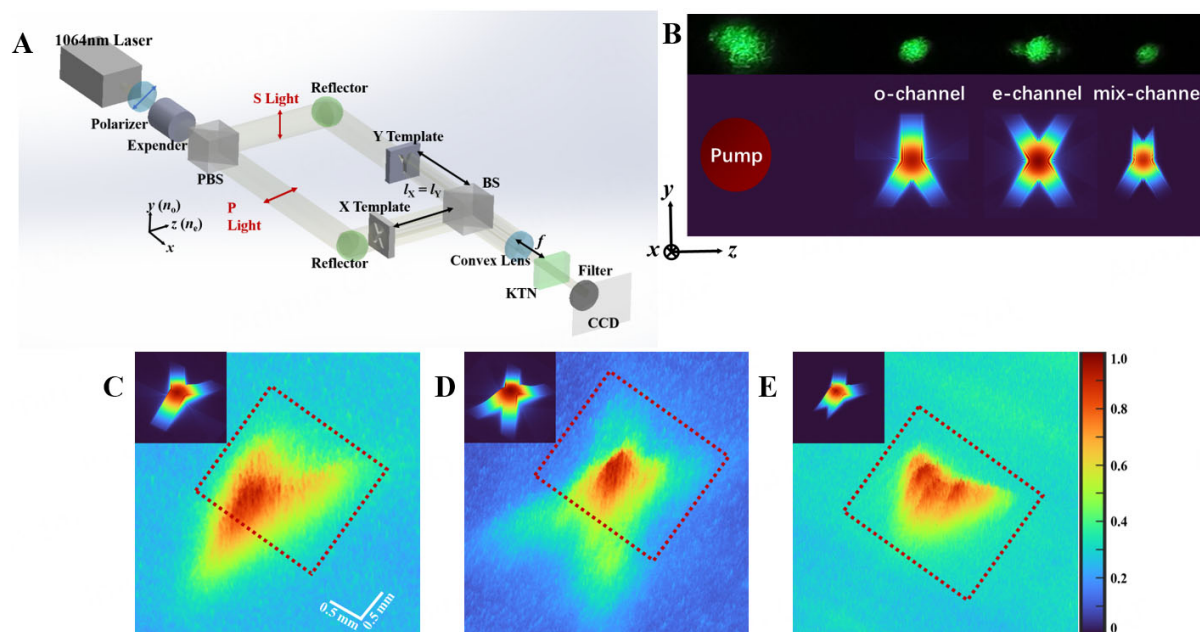


Figure 5. (A) A diagram of multi-channel nonlinear image propagation. (B) Simulated two-dimensional SHG distribution of the o-channel ($oo \rightarrow e$), e-channel ($ee \rightarrow e$), and mix-channel ($oe \rightarrow o$) at the CCD receiving plane; the inversion of the image is due to the effect of the lens. The insert shows an experimentally measured SHG distribution pattern (one side), generated by a 45° -polarized 1,064 nm laser propagating through the optical setup shown in Figure 1C. Real distribution of (C) o-channel, (D) e-channel, and (E) mix-channel. The inserts show the corresponding simulated results, and the area within the red box represents the overlapping region of “X” and “Y” images. (B–E) shares a common color bar.

DECLARATIONS

Authors' contributions

Materials growth and characterization: Meng, X.; Hu, C.; Wang, Y.

Optical path design and construction: Li, Z.; Zhang, Z.

Software and simulations: Wen, X.; Zhang, Z.

Data analysis and interpretation: Zhang, Z.; Tan, P.

Conception, design, writing and editing: Zhang, Z.; Tan, P.; Tian, H.; Song, Q.

All authors contributed to the manuscript and were involved in the discussion.

Availability of data and materials

The data that support the findings of this study are available within the article and its [Supplementary Materials](#).

AI and AI-assisted tools statement

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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Supplementary Materials

[Supplementary Materials](#)

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