

Supplementary Information

Nonlinear geometric phase coded ferroelectric nematic fluids for nonlinear soft-matter photonics

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Section 1: Nonlinear polarization of the ferroelectric nematic LCs

Ferroelectric nematic liquid crystals (FNLCs) exhibit strong nonlinear optical responses with non-zero second-order nonlinear optical susceptibility $\chi_{aaa}^{(2)} = 11.2 \text{ pm}\cdot\text{V}^{-1}$ and $\chi_{abb}^{(2)} = \chi_{bab}^{(2)} = \chi_{bba}^{(2)} = 1.2 \text{ pm}\cdot\text{V}^{-1}$ at 1064 nm (usually neglected), where a - and b -axis are parallel and perpendicular to the polar direction, respectively. When the a -axis aligns along the x -axis, i.e., $\alpha = 0^\circ$, $\chi_{xxx}^{(2)}$ equals to $\chi_{aaa}^{(2)}$. The expression for the induced transverse nonlinear polarization ($P_i^{2\omega}$) can be written as $P_i^{2\omega} = \varepsilon_0 \sum_{jk} \chi_{ijk}^{(2)} E_j^\omega E_k^\omega$, where $\chi_{ijk}^{(2)}$ represents the element of the

nonlinear susceptibility tensor $\chi^{(2)}$, E_j^ω is the component of the electric field with respect to the laboratory coordinates ($i, j, k = \{x, y, z\}$), and ε_0 is the vacuum permittivity¹. In the case of an FNLC director with an in-plane orientation angle α , the rotation matrices are given by

$$\Omega_{i'i} = \begin{pmatrix} \cos \alpha & \sin \alpha & 0 \\ -\sin \alpha & \cos \alpha & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad [\Omega^{-1}]_{ii'} = \begin{pmatrix} \cos \alpha & -\sin \alpha & 0 \\ \sin \alpha & \cos \alpha & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (\text{S1})$$

where $i', j', k' = \{x', y', z'\}$. In the local (primed) coordinate system, the nonlinear polarization yields

$$P_{i'}^{2\omega} = \sum_i \Omega_{i'i} P_i^{2\omega} = \varepsilon_0 \sum_{ijk} \Omega_{i'i} \chi_{ijk}^{(2)} E_j^\omega E_k^\omega. \quad (\text{S2})$$

When substituting $E_i^\omega = \sum_{i'} [\Omega^{-1}]_{ii'} E_{i'}^\omega$ into Eq. (S2), we obtain

$$P_{i'}^{2\omega} = \varepsilon_0 \sum_{j'k'} \sum_{ijk} \Omega_{i'i} \chi_{ijk}^{(2)} [\Omega^{-1}]_{jj'} [\Omega^{-1}]_{kk'} E_{j'}^\omega E_{k'}^\omega. \quad (\text{S3})$$

We can define the nonlinear susceptibility tensor $\chi_{lmn}^{(2)}$ in the local coordinate system as

$$\chi_{i'j'k'}^{(2)} = \sum_{ijk} \Omega_{i'i} \chi_{ijk}^{(2)} [\Omega^{-1}]_{jj'} [\Omega^{-1}]_{kk'}, \quad (\text{S4})$$

where

$$\begin{aligned} \chi_{x'x'x'}^{(2)} &= \chi_{xxx}^{(2)} \cos^3 \alpha & \chi_{y'y'y'}^{(2)} &= \chi_{xxx}^{(2)} \sin^2 \alpha \\ \chi_{x'y'y'}^{(2)} &= \chi_{xxx}^{(2)} \cos \alpha \sin^2 \alpha & \chi_{y'x'x'}^{(2)} &= \chi_{xxx}^{(2)} \cos^2 \alpha \sin \alpha \\ \chi_{x'x'y'}^{(2)} &= \chi_{xxx}^{(2)} \cos^2 \alpha \sin \alpha & \chi_{y'x'y'}^{(2)} &= \chi_{xxx}^{(2)} \cos \alpha \sin^2 \alpha \end{aligned} \quad (\text{S5})$$

Consequently, when the linearly polarized fundamental wave (FW) $\mathbf{E}^\omega = A_0 (\cos \phi \hat{\mathbf{e}}_x + \sin \phi \hat{\mathbf{e}}_y)$ is incident on an FNLC director with an in-plane orientation angle α , the nonlinear polarization of the generated second harmonic wave (SHW) can be calculated as

$$\begin{aligned} \mathbf{P}^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2 (\phi - \alpha) (\cos \alpha \hat{\mathbf{e}}_x + \sin \alpha \hat{\mathbf{e}}_y) \\ &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2 (\phi - \alpha) \hat{\mathbf{e}}_a \end{aligned} \quad (\text{S6})$$

where $\hat{\mathbf{e}}_a$ represents the unit vector of the a -axis.

Section 2: Photopatterning of the ferroelectric nematic LCs

Detailed photopatterning processes are provided as follows. The empty liquid crystal (LC) cell is placed at the image plane of the digital micromirror device-based microlithography system to record the patterns of LC director distribution via a multistep partly overlapping exposure process with a synchronous polarization control. For the case of the q -plate, the calculated director distribution varying from 0 to π is replaced by 72 subregions on average, and each subregion is assigned a uniform director value from $\pi/72$ to π in the interval $\pi/72$. This determines the exposure location. The sum-region of five adjacent subregions is exposed simultaneously with the exposure intensity of $0.8 \text{ mW}\cdot\text{cm}^{-2}$, exposure wavelength of 405 nm, and exposure time of ~ 3.0 s, which is insufficient to induce a stable reorientation of SD1. The subsequent exposure of the sum-region shifts one subregion with the polarizer rotating 5° synchronously. Finally, each subregion is exposed five times with a total exposure time of ~ 15.0 s, which is enough to reorient the SD1 molecules. Based on the above steps, we optimized the exposure time and the position of the image plane, and finally obtained the photopatterned FNLC cell with continuously distributed polarization directions. The FNLC RM734 was capillary-filled into the photo-patterned cell at 185°C , ultimately forming the nonlinear q -plate device. The photoalignment function is most efficient at 25°C and $<40\%$ humidity.

Section 3: Nonlinear geometric phase of the ferroelectric nematic LCs

To unravel the spin-orbit interaction processes in the framework of nonlinear optics, the SHWs in the circular polarization (CP) basis with nonlinear geometric phases are studied. The coordinate transformation is performed between the linear polarization basis and the CP basis:

$$\Lambda_{li} = \begin{pmatrix} 1 & -i & 0 \\ 1 & i & 0 \\ 0 & 0 & \sqrt{2} \end{pmatrix} / \sqrt{2}, \quad [\Lambda^{-1}]_{il} = \begin{pmatrix} 1 & 1 & 0 \\ i & -i & 0 \\ 0 & 0 & \sqrt{2} \end{pmatrix} / \sqrt{2}, \quad (\text{S7})$$

where $l, m, n = \{L, R, z\}$. By coordinate transformation, the second-order susceptibility tensor in the CP basis yields

$$\chi_{lmn}^{(2)} = \sum_{ijk} \Lambda_{li} \chi_{ijk}^{(2)} [\Lambda^{-1}]_{jm} [\Lambda^{-1}]_{kn}, \quad (\text{S8})$$

$$\text{where } \chi_{RRR}^{(2)} = \chi_{RLL}^{(2)} = \chi_{RLR}^{(2)} = \chi_{LLL}^{(2)} = \chi_{LRR}^{(2)} = \chi_{LLR}^{(2)} = \frac{1}{2\sqrt{2}} \chi_{xxx}^{(2)}.$$

When the in-plane orientation of the LC director is rotated, the nonlinear polarization will carry the nonlinear geometric phase. As shown in Fig. 4a, when the LC director is counterclockwise rotated by an angle α , the rotation matrices in the CP basis are given by

$$R_{l'l}(\alpha) = \begin{pmatrix} e^{-i\alpha} & 0 & 0 \\ 0 & e^{i\alpha} & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad [R^{-1}(\alpha)]_{l'l'} = \begin{pmatrix} e^{i\alpha} & 0 & 0 \\ 0 & e^{-i\alpha} & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (\text{S9})$$

Then, the nonlinear polarization yields

$$P_{l'}^{2\omega}(z) = \sum_l R_{l'l}(\alpha) P_l^{2\omega}(z) = \varepsilon_0 \sum_{lmn} R_{l'l}(\alpha) \chi_{lmn}^{(2)} E_m(z) E_n(z). \quad (\text{S10})$$

By substituting $E_l^\omega = \sum_{l'} [R^{-1}(\alpha)]_{ll'} E_{l'}^\omega$ into Eq. (S10), we obtain

$$P_{l'}^{2\omega}(z) = \varepsilon_0 \sum_{m'n'} \sum_{lmn} R_{l'l}(\alpha) \chi_{lmn}^{(2)} [R^{-1}(\alpha)]_{nn'} [R^{-1}(\alpha)]_{mm'} E_{m'}(z) E_{n'}(z). \quad (\text{S11})$$

Consequently, the second-order susceptibility tensor of the FNLC molecule in the CP basis can be written as

$$\chi_{l'm'n'}^{(2)} = \sum_{lmn} R_{l'l}(\alpha) \chi_{lmn}^{(2)} [R^{-1}(\alpha)]_{nn'} [R^{-1}(\alpha)]_{mm'}. \quad (\text{S12})$$

Therefore, the nonlinear geometric phase contributed by the local and laboratory frame transformation is

$$R_{l'l}(\alpha) [R^{-1}(\alpha)]_{nn'} [R^{-1}(\alpha)]_{mm'}. \quad (\text{S13})$$

From Eq. (S12), we have the following components

$$\begin{aligned} \chi_{R'R'R'}^{(2)} &= \chi_{RRR}^{(2)} e^{i\alpha} & \chi_{R'L'L'}^{(2)} &= \chi_{RLL}^{(2)} e^{-3i\alpha} & \chi_{R'L'R'}^{(2)} &= \chi_{RLR}^{(2)} e^{-i\alpha} \\ \chi_{L'L'L'}^{(2)} &= \chi_{LLL}^{(2)} e^{-i\alpha} & \chi_{L'R'R'}^{(2)} &= \chi_{LRR}^{(2)} e^{3i\alpha} & \chi_{L'L'R'}^{(2)} &= \chi_{LLR}^{(2)} e^{i\alpha} \end{aligned} \quad (\text{S14})$$

For incident FW with arbitrary polarization state $\mathbf{E}^\omega(z=0) = (E_\sigma^\omega(z=0)\hat{\mathbf{e}}_\sigma + E_{-\sigma}^\omega(z=0)\hat{\mathbf{e}}_{-\sigma})$, where $\sigma = \pm 1$ stands for the right- and left-handed circular polarization (RCP and LCP) state, we analytically derive the second-order nonlinear polarization of co- and cross-CP second harmonic (SH) signals at the z plane as

$$\begin{aligned} P_\sigma^{2\omega}(z) &= \frac{1}{2\sqrt{2}} \varepsilon_0 \chi_{xxx}^{(2)} \left[e^{i\sigma\alpha} (E_\sigma^\omega(z))^2 + e^{-3i\sigma\alpha} (E_{-\sigma}^\omega(z))^2 + 2e^{-i\sigma\alpha} (E_\sigma^\omega(z)E_{-\sigma}^\omega(z)) \right], \\ P_{-\sigma}^{2\omega}(z) &= \frac{1}{2\sqrt{2}} \varepsilon_0 \chi_{xxx}^{(2)} \left[e^{3i\sigma\alpha} (E_\sigma^\omega(z))^2 + e^{-i\sigma\alpha} (E_{-\sigma}^\omega(z))^2 + 2e^{i\sigma\alpha} (E_\sigma^\omega(z)E_{-\sigma}^\omega(z)) \right] \end{aligned} \quad (\text{S15})$$

where $E_\sigma^\omega(z)$ and $E_{-\sigma}^\omega(z)$ refer to the co- and cross-CP components of propagating FW in the z plane, respectively. The acquired nonlinear geometric phases with respect to six different spin-orbit angular momentum states can be seen from Eq. (S15).

For the incident FW with the spin state of σ , it will evolve into two components $E_\sigma^\omega(z) \propto e^{-ikz} e^{i\ell_0\varphi} E_\sigma^\omega(z=0)$ and $E_{-\sigma}^\omega(z) \propto e^{-ikz} e^{2i\sigma\alpha} e^{i\ell_0\varphi} E_\sigma^\omega(z=0)$ due to the linear geometric phase effect. By substituting them separately into Eq. (S15), the nonlinear polarization consistently exhibits this form:

$$\begin{aligned} P_\sigma^{2\omega}(z) &= \frac{1}{2\sqrt{2}} \varepsilon_0 \chi_{xxx}^{(2)} e^{-ik\eta_e z} e^{i(2\ell_0\varphi+\sigma\alpha)} (E_\sigma^\omega(z=0))^2 \\ P_{-\sigma}^{2\omega}(z) &= \frac{1}{2\sqrt{2}} \varepsilon_0 \chi_{xxx}^{(2)} e^{-ik\eta_e z} e^{i(2\ell_0\varphi+3\sigma\alpha)} (E_\sigma^\omega(z=0))^2. \end{aligned} \quad (\text{S16})$$

Here, σ merely indicates whether the outgoing spin is the same as or opposite to the incident spin, without specifically denoting left- or right-handed circular polarization. This result shows spin-locked geometric phases originating from the cascaded linear and nonlinear spin-orbit interactions (SOIs) in the process of nonlinear frequency conversion. Thus, the co- and cross-CP components of the SH signal will acquire different geometric phases $2\ell_0\varphi+\sigma\alpha$ and $2\ell_0\varphi+3\sigma\alpha$.

For the case of an FNLC q -plate, of which the director distribution follows the equation of $\alpha(\varphi) = q\varphi$, the angular momentum states of the SHWs are $(\sigma, 2\ell_0+\sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0+3\sigma q)_{2\omega}$. The intensity of each component of the SHW is calculated by the equation of $I_\sigma^{2\omega}(z) \propto |P_\sigma^{2\omega}(z)|^2$. Due to the same amplitude coefficients (related to $\chi_{xxx}^{(2)}$), the intensities of the two components $P_\sigma^{2\omega}(z)$ and $P_{-\sigma}^{2\omega}(z)$ are calculated to be equivalent. Subsequently, these two components of the SHWs propagate towards the exit plane. The linear spin-orbit interaction during the propagation of the SHWs results in spin-locked angular momentum states, $(\sigma, 2\ell_0+\sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0+3\sigma q)_{2\omega}$, without any redistribution of light intensities. That is, the conversion efficiencies are identical. Consequently, the intensities of the co-circularly and cross-circularly polarized SH optical vortices are equal. Additionally, we accumulate all the coherent SHWs propagating towards the exit plane at $z = L$ (L is the thickness of the FNLC film), which are generated from all the LC layers, to obtain the total nonlinear polarizations for both circular polarization components. As a result, these two components are equivalent as well.

Section 4: Measurement of the SHG efficiency

The equation for the conversion efficiency of the second harmonic generation (SHG) process in the FNLC device can be expressed as:

$$\eta = \frac{\langle POW^{2\omega} \rangle}{\langle POW^{\omega} \rangle}, \quad (S17)$$

where $\langle POW^{\omega} \rangle$ and $\langle POW^{2\omega} \rangle$ represent the averaged power of FW and SHW. To determine the maximal efficiency, the temperature of the nonlinear LC q -plate is rapidly decreased to 27 °C in order to improve the damage threshold while maintaining it in the ferroelectric nematic phase. The power of the incident FW is measured as 124 mW by using a thermal power meter (model S401C, Thorlabs). The FW then passes through a quarter-wave plate, a long-pass filter with a cut-off wavelength of 700 nm, a nonlinear LC q -plate, and a short-pass filter with a cut-off wavelength of 700 nm. The power of the generated SHW is measured as 20 μ W by using the same power meter. Therefore, the nonlinear conversion efficiency is calculated to be $\eta = 1.6 \times 10^{-4}$ using Eq. (S17). In addition, we also confirmed the existence of the room-temperature ferroelectric nematic phase and verified the non-damaging nature of the nonlinear LC q -plate through cross-polarized optical microscopy observation.

Section 5: Electrical switching of the SH optical vortices

When an electric field of a triangular wave (0.5 Hz, $0.12 \text{ V} \cdot \mu\text{m}^{-1}$, Fig. S10) is applied vertically to the LC device, the nonlinear LC molecular dipoles are realigned, resulting in the shifted SH susceptibility from the transverse direction ($\chi_{aaa}^{(2)}$) to the longitudinal direction ($\chi_{bbb}^{(2)}$). Within one electric field cycle, the nonlinear susceptibility will be switched twice from $|\chi_{aaa}^{(2)}|$ (at 0 V) to $|\chi_{bbb}^{(2)}|$ (at -0.5 V and 0.5 V). Thus, the time-domain waveform of the SH intensity switching operates at a frequency of 1 Hz, which is twice that of the applied electric field (Fig. S11).

Section 6: Theory of the spin-selective SH vortices from a chiral LC q -superstructure

The nonlinear chiral LC q -superstructure is partitioned into layers of uniform thickness Δz along z -axis to calculate the spin-selective SH optical vortices. Firstly, we calculate the evolution of the FW propagating in the three-dimensional chiral LC q -superstructure. Assuming that the electric field of the FW is $\mathbf{E}_{\text{in}}^{\omega}(z=0) = (E_{\sigma}^{\omega}(z=0)\hat{\mathbf{e}}_{\sigma} + 0\hat{\mathbf{e}}_{-\sigma})$, we use Jones matrix $\mathbf{M}$ to describe the linear propagation process in the N^{th} layer FNLC,

$$\mathbf{M}_N^{\omega} = e^{-ik_{\omega} n_{\text{eff},\omega} N\Delta z} \begin{pmatrix} \cos(\Delta\Gamma_{\omega}/2) & -i\sin(\Delta\Gamma_{\omega}/2)e^{-2i\sigma\alpha(\varphi, N\Delta z)} \\ -i\sin(\Delta\Gamma_{\omega}/2)e^{2i\sigma\alpha(\varphi, N\Delta z)} & \cos(\Delta\Gamma_{\omega}/2) \end{pmatrix}, \quad (\text{S18})$$

where $\alpha(\varphi, N\Delta z) = q\varphi + 2\pi N\Delta z/p$ represents the LC director distribution of the chiral FNLC q -plate, $\Delta\Gamma = 2\pi(n_e - n_o)\Delta z/\lambda$ is the phase delay of the FNLC, λ is the wavelength, $n_{\text{eff}} = (n_o + n_e)/2$ is the effective refractive index, n_o is the ordinary refractive index of the FNLCs, and p denotes the helical pitch. α in Eq. (S18) depends on the position of the considered infinitesimal slab. The electric field of FW when it propagates from the first layer to the N th layer can be calculated by

$$\mathbf{E}^{\omega}(z = N\Delta z) = \mathbf{M}_N^{\omega} \cdots \mathbf{M}_2^{\omega} \mathbf{M}_1^{\omega} \mathbf{E}_{\text{in}}^{\omega}(z = 0). \quad (\text{S19})$$

Next, by substituting Eq. (S19) into Eq. (S15), we obtain the nonlinear polarization of the N^{th} layer is calculated as follows,

$$\mathbf{P}^{2\omega}(z = N\Delta z) = \frac{1}{2\sqrt{2}} \varepsilon_0 \chi_{\text{xxx}}^{(2)} \begin{pmatrix} e^{i\sigma\alpha(\varphi, N\Delta z)} (E_{\sigma}^{\omega}(N\Delta z))^2 + e^{-3i\sigma\alpha(\varphi, N\Delta z)} (E_{-\sigma}^{\omega}(N\Delta z))^2 + 2e^{-i\sigma\alpha(\varphi, N\Delta z)} (E_{-\sigma}^{\omega}(N\Delta z) E_{-\sigma}^{\omega}(N\Delta z)) \\ e^{3i\sigma\alpha(\varphi, N\Delta z)} (E_{\sigma}^{\omega}(N\Delta z))^2 + e^{-i\sigma\alpha(\varphi, N\Delta z)} (E_{-\sigma}^{\omega}(N\Delta z))^2 + 2e^{i\sigma\alpha(\varphi, N\Delta z)} (E_{-\sigma}^{\omega}(N\Delta z) E_{-\sigma}^{\omega}(N\Delta z)) \end{pmatrix}. \quad (\text{S20})$$

Once we have the nonlinear polarization $\mathbf{P}^{2\omega}(z = N\Delta z)$ generated from each infinitesimal layer, we can calculate the SHG signal output from the three-dimensional chiral LC q -superstructure as follows.

The SHWs (including σ and $-\sigma$ components) of each layer linearly propagate to the exit plane and they are separately accumulated to obtain the output co- and cross-CP components of SH signal. Considering the linear propagation of the generated SHW from $z = N\Delta z$ to $z = L$ (exit plane), we obtain the output nonlinear polarization of N th layer as

$$\mathbf{P}_N^{2\omega}(z = L) = \mathbf{M}_{L/\Delta z}^{2\omega} \cdots \mathbf{M}_{N+2}^{2\omega} \mathbf{M}_{N+1}^{2\omega} \mathbf{P}^{2\omega}(z = N\Delta z). \quad (\text{S21})$$

The output nonlinear polarizations are summed separately for each CP component, and the result is

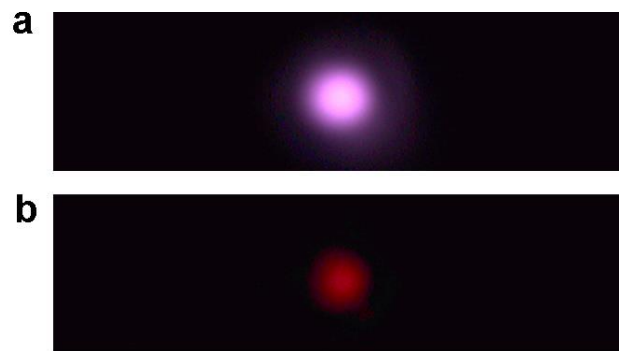
$$\mathbf{P}_{\text{sum}}^{2\omega}(z = L) = \sum_N \mathbf{P}_N^{2\omega}(z = L) = \begin{pmatrix} \sum_N P_{\sigma, N}^{2\omega}(z = L) \\ \sum_N P_{-\sigma, N}^{2\omega}(z = L) \end{pmatrix}. \quad (\text{S22})$$

Thus, the total intensity of SHW is

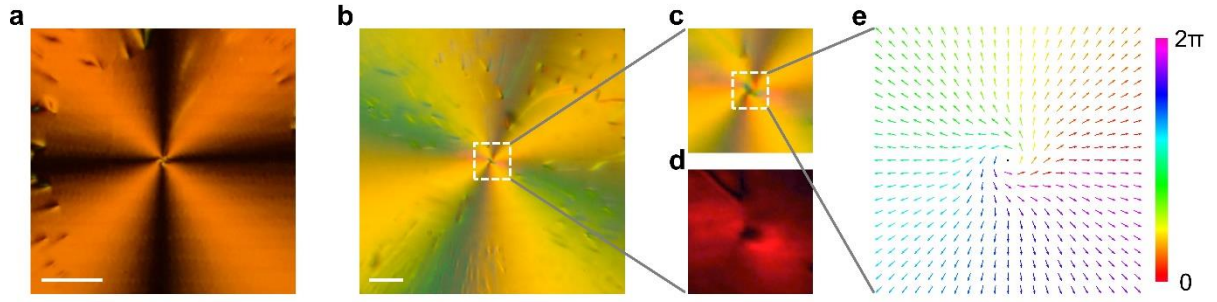
$$I_{\text{total}}^{2\omega} \propto \left| P_{\sigma, \text{sum}}^{2\omega}(z = L) \right|^2 + \left| P_{-\sigma, \text{sum}}^{2\omega}(z = L) \right|^2. \quad (\text{S23})$$

According to the above derivation, the nonlinear circular dichroism of the far-field SHWs is calculated to be ~ 0.96 with self-developed codes by the commercial software MATLAB R2022a.

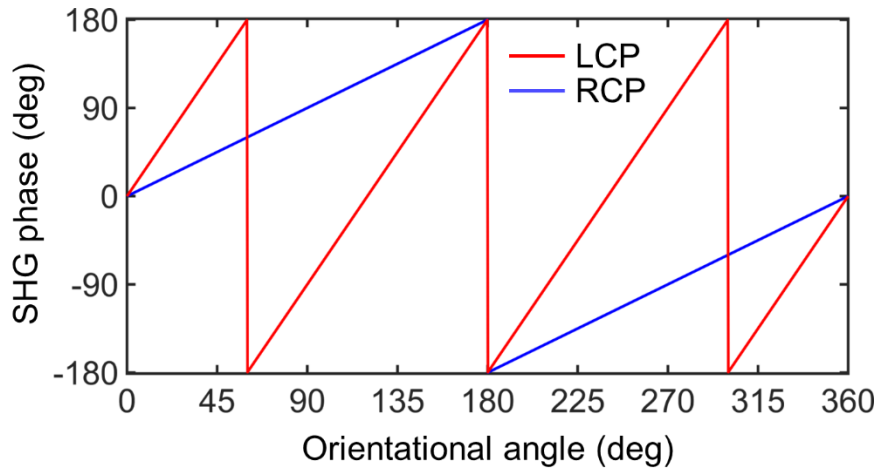
Supplementary Figures:



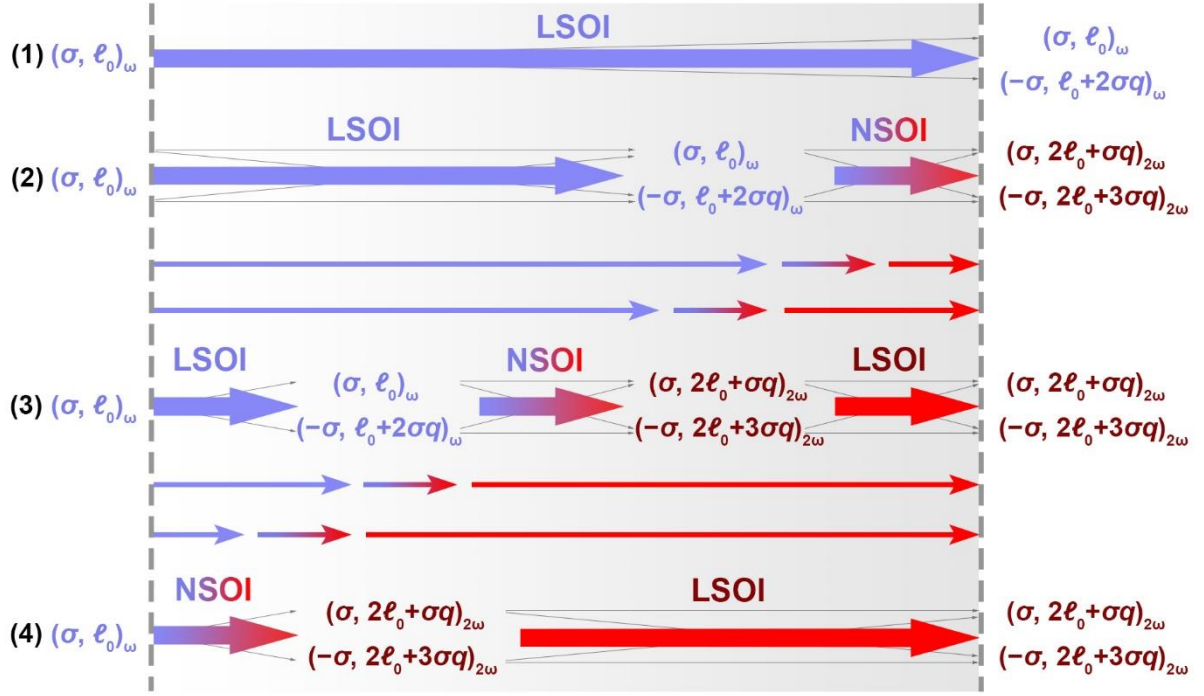
Supplementary Fig. 1 | Intensity profiles of fundamental and SH Gaussian beams. Intensity profiles of **a**, FW and **b**, SHW. The radius of the FW is slightly bigger than that of the SHW due to the quadratic power dependence relationship in the SHG process.



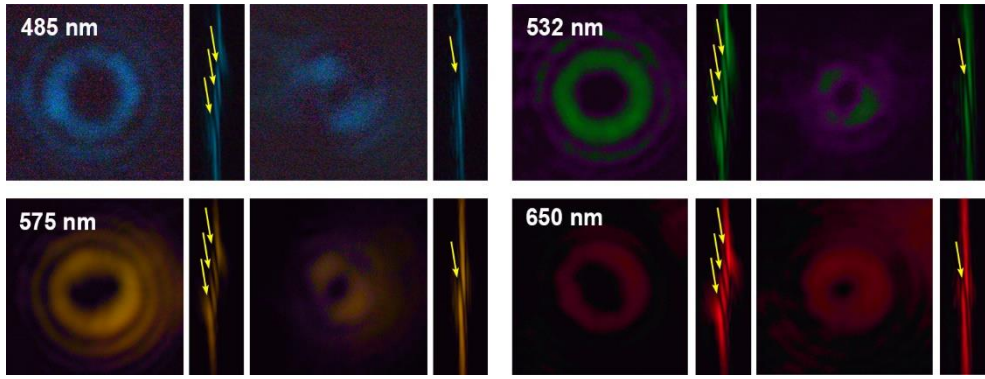
Supplementary Fig. 2 | Typical textures of FNLC q -plate. **a**, Full plate of Fig. 3c in the manuscript. The texture is captured under a crossed polarized optical microscope. Scale bar: 100 μm . **b**, Texture of the polar LC q -plate showing elongated lens-shape patterns indicative of the local director orientation. The image is obtained in the optical path during SHG characterization. The extinction is not totally black where the polar LC orientation is either parallel or perpendicular to the crossed polarizers. This phenomenon might be attributed to the experimental error with a series of polarizing optical elements that correspond to a monochromatic laser wavelength, while the SH femtosecond laser is not monochromatic with the full width at half-maxima of about 20 nm. Scale bar: 100 μm . **c**, Enlarged view of **(b)**. **d**, SH image of **(b)**. **e**, Illustration of the q -plate's center with a whirl-like texture.



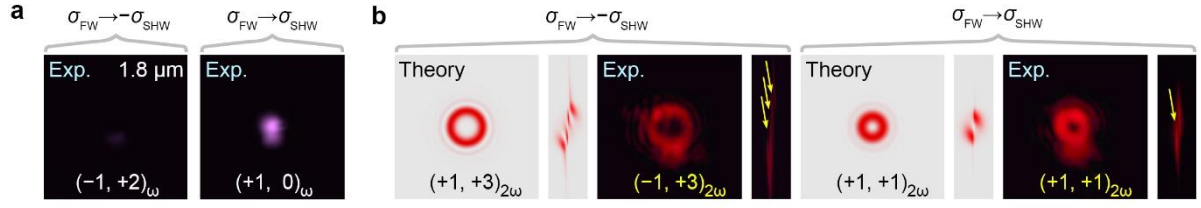
Supplementary Fig. 3 | Numerical calculation of the nonlinear geometric phase from a rotated FNLC molecule for FW with angular momentum state of $(+1, 0)_\omega$. The simulated nonlinear geometric phase for the LCP-SHW is three times that of the RCP-SHW. Source data are provided as a Source Data file.



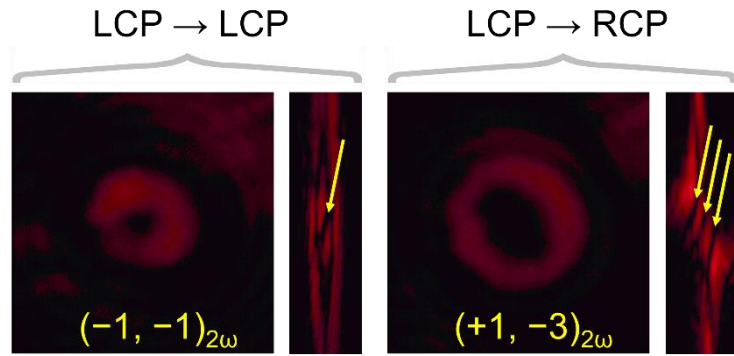
Supplementary Fig. 4 | Cascaded SOI processes simultaneously occurring in the ferroelectric nematic q -plate. LSOI: linear SOI; NSOI: nonlinear SOI. These cascaded SOI processes can be categorized into four classes, including: (1) LSOI-only (LSOI; spin-orbit angular momentum states: $(\sigma, \ell_0)_\omega \rightarrow \{(\sigma, \ell_0)_\omega, (-\sigma, \ell_0 + 2\sigma q)_\omega\}$); (2) cascaded LSOI and NSOI (LSOI $\rightarrow$ NSOI; spin-orbit angular momentum states: $(\sigma, \ell_0)_\omega \rightarrow \{(\sigma, \ell_0)_\omega, (-\sigma, \ell_0 + 2\sigma q)_\omega\} \rightarrow \{(\sigma, 2\ell_0 + \sigma q)_{2\omega}, (-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}\}$); (3) cascaded LSOI, NSOI, and LSOI (LSOI $\rightarrow$ NSOI $\rightarrow$ LSOI; spin-orbit angular momentum states: $(\sigma, \ell_0)_\omega \rightarrow \{(\sigma, \ell_0)_\omega, (-\sigma, \ell_0 + 2\sigma q)_\omega\} \rightarrow \{(\sigma, 2\ell_0 + \sigma q)_{2\omega}, (-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}\} \rightarrow \{(\sigma, 2\ell_0 + \sigma q)_{2\omega}, (-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}\}$). This is the most common situation in the ferroelectric nematic q -plate; (4) cascaded NSOI and LSOI (NSOI $\rightarrow$ LSOI; spin-orbit angular momentum states: $(\sigma, \ell_0)_\omega \rightarrow \{(\sigma, 2\ell_0 + \sigma q)_{2\omega}, (-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}\} \rightarrow \{(\sigma, 2\ell_0 + \sigma q)_{2\omega}, (-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}\}$).



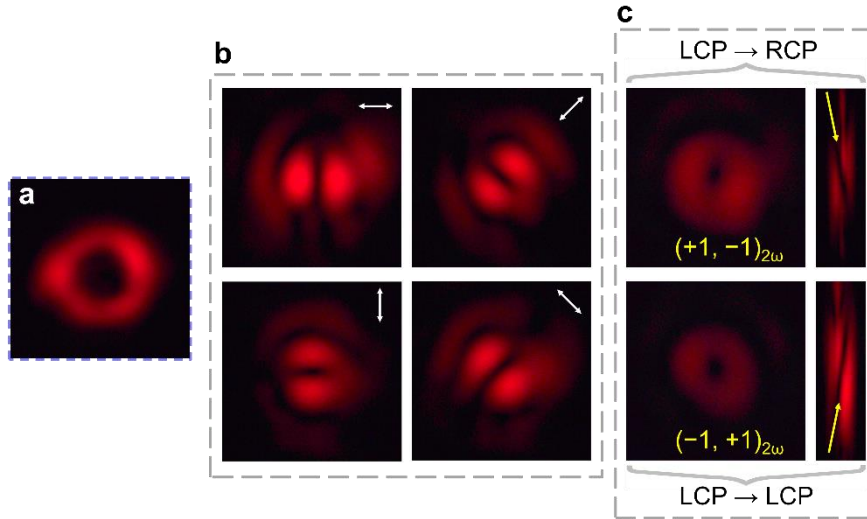
Supplementary Fig. 5 | Broadband wavelength tolerance of the FNLc q -plate in the SHG process. The incident FW has the angular momentum state of $(+1, 0)_\omega$. The wavelengths of the SHWs are 485 nm, 532 nm, 575 nm, and 650 nm, respectively.



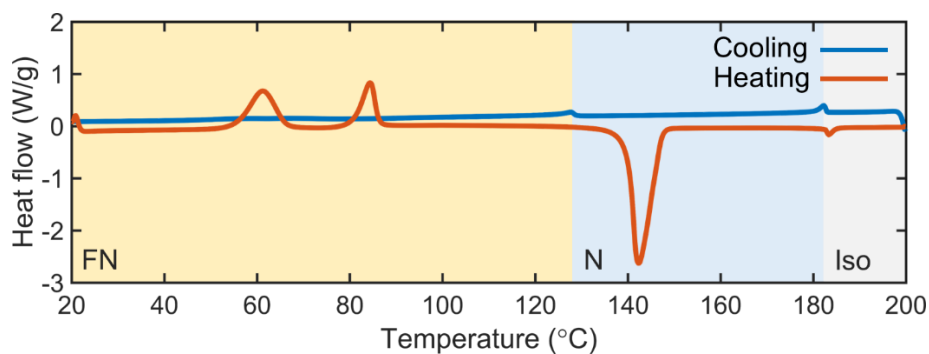
Supplementary Fig. 6 | Angular momentum states generated from another FNLC device with the LC film thickness of 1.8 μm. a, Linear and **b**, nonlinear geometric phase associated optical vortex generation in the FNLC q -plate. The angular momentum state of the incident FW is $(+1, 0)_\omega$. Four output angular momentum states are $(+1, 0)_\omega$, $(-1, +2)_\omega$, $(+1, +1)_{2\omega}$, and $(-1, +3)_{2\omega}$. The abbreviation “Exp.” represents the experimental results. The low-intensity state of $(-1, +2)_\omega$ is due to the low conversion efficiency induced by the mismatch of half-wave condition. The generated co- and cross-CP SH vortices $(+1, +1)_{2\omega}$ and $(-1, +3)_{2\omega}$ are consistent with the results shown in Fig. 4d, with a nearly equal intensity distribution (48:52).



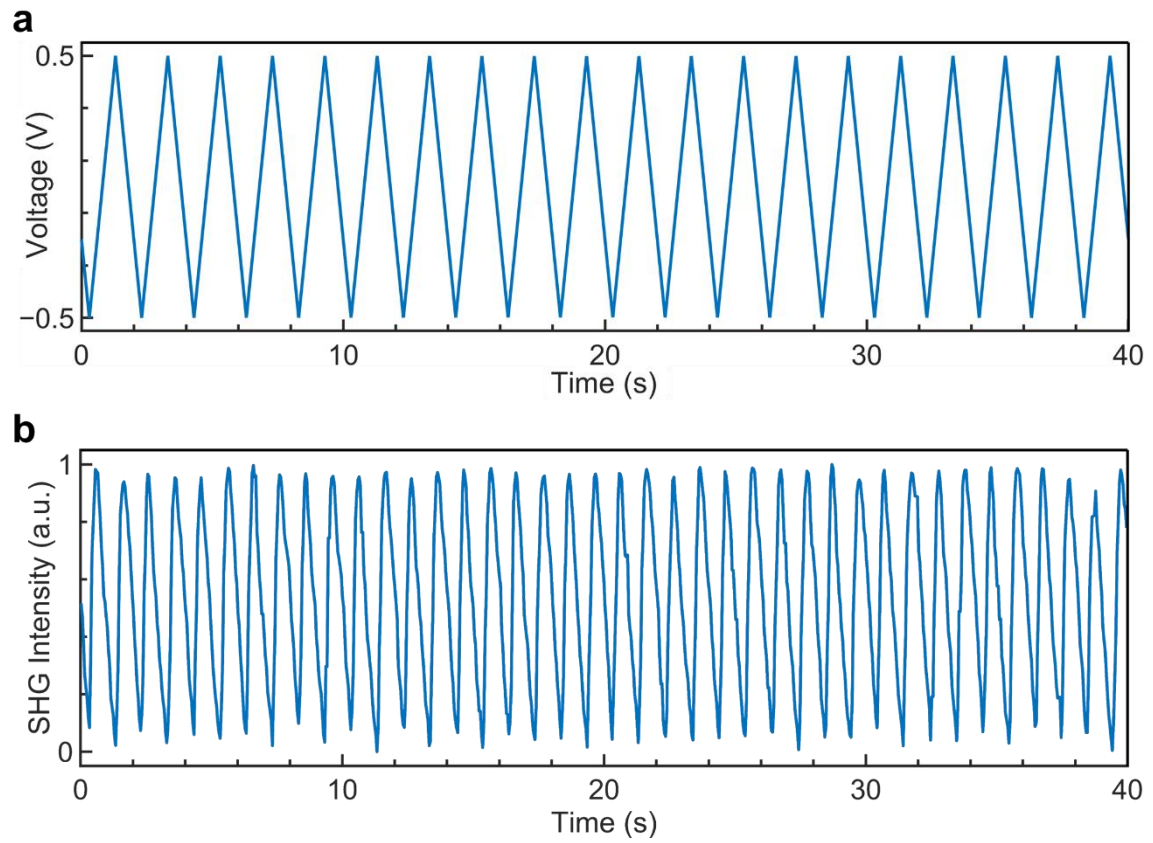
Supplementary Fig. 7 | Intensity profiles of SH vortices for incident FW with angular momentum state of $(-1, 0)_\omega$. LCP $\rightarrow$ LCP (LCP $\rightarrow$ RCP) represent incident and output circular polarization, respectively.



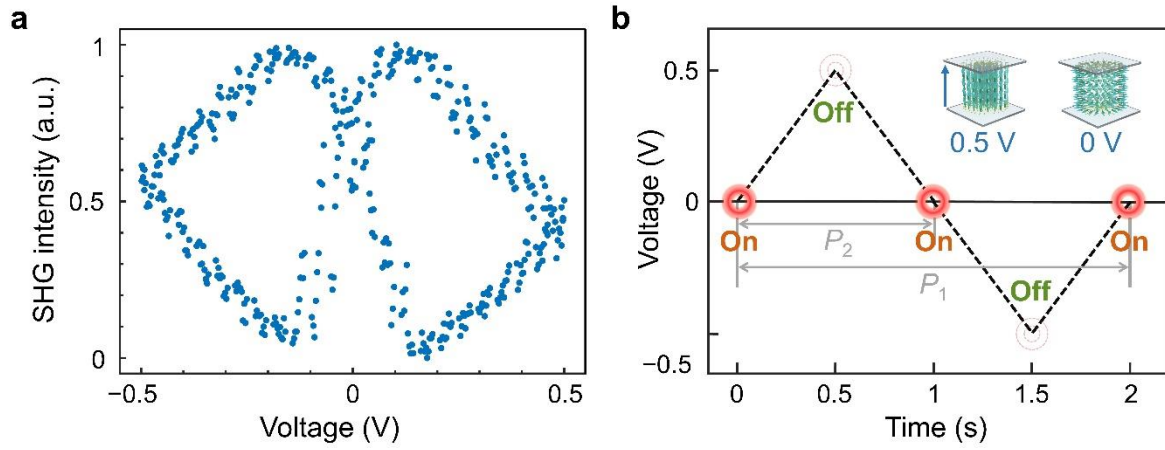
Supplementary Fig. 8 | SH vortex beam generation and characterization for the incidence of CP vortex beams with $(-1, +1)_\omega$. **a**, The generated SH beam. **b**, Characterization of the vectorial polarization distribution of the SH wave. White arrows show the polarization direction of the analyzer. **c**, Intensity profiles and orbital angular momentum characterizations of the SH vortices. LCP → LCP (LCP → RCP) represent incident and output circular polarization, respectively.



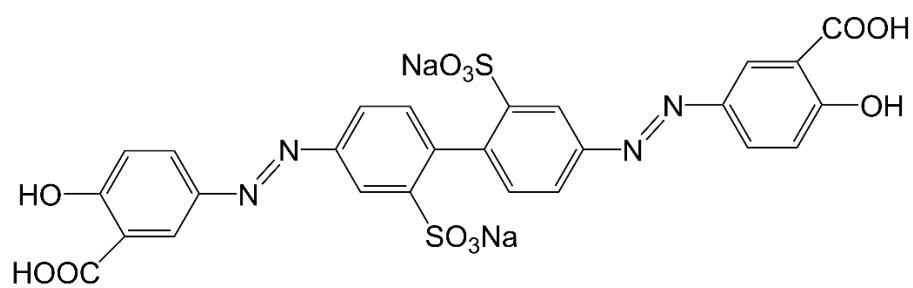
Supplementary Fig. 9 | Differential scanning calorimetry traces of RM734. The heating and cooling rates are $10\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$. This result is obtained by using a differential scanning calorimeter (Mettler-Toledo DSC1, STARe). According to the cooling process, the phase sequence of RM734 follows isotropic phase - $182.3\text{ }^{\circ}\text{C}$ - nematic phase - $127.7\text{ }^{\circ}\text{C}$ - ferroelectric nematic phase. FN: ferroelectric nematic phase. N: nematic phase. Iso: isotropic phase. Source data are provided as a Source Data file.



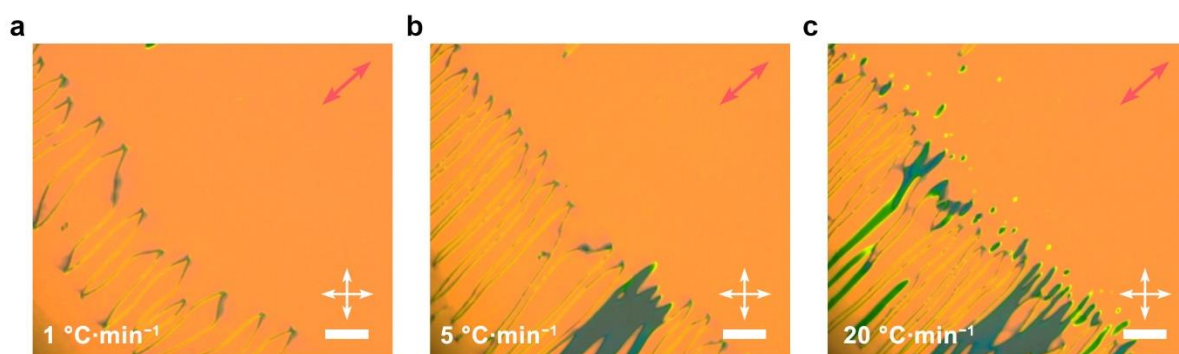
Supplementary Fig. 10 | Applied electric field and reversibly periodic modulation of the SH vortices. a, A triangular wave (0.5 Hz , $0.12\text{ V}\cdot\mu\text{m}^{-1}$) is applied vertically to the LC device for 40 s. **b**, Electrically switchable spin-orbit angular momentum states for 40 cycles. The total switching cycling number in our experiment is 438. Source data are provided as a Source Data file.



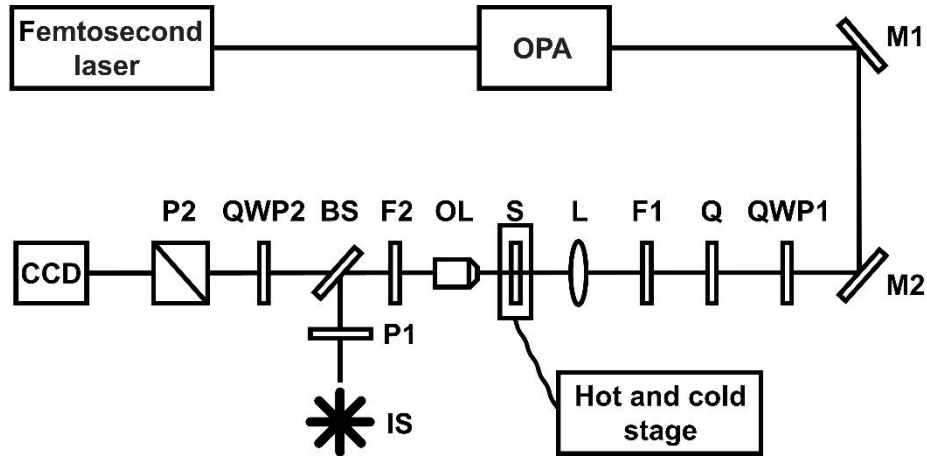
Supplementary Fig. 11 | Electrical switching of the SH vortices. **a**, Lissajous figure for the SHG intensity versus applied voltage (20 cycles). The modulation frequency of the SHG intensity is twice that of the applied electric field. **b**, Periodicity relationship of the SH vortex switching and the applied electric field. $P_1 = 2P_2$. The red circles denote the generated SHW. Source data are provided as a Source Data file.



Supplementary Fig. 12 | Molecule structure of photoalignment agent SD1.



Supplementary Fig. 13 | Polarized optical microscopic textures of the homogeneously photoaligned FNLC at different cooling rates of (a) $1\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$, (b) $5\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$, and (c) $20\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$. The magenta arrows denote the direction of photoalignment. Scale bar: $100\text{ }\mu\text{m}$.



Supplementary Fig. 14 | Optical setup for the measurement of the SHG signals from the LC device. OPA, Optical Parametric Amplifier; M, mirror; QWP, quarter-wave plate; Q, LC q -plate; F1, long-pass filter (cut-on wavelength: 700 nm); F2, short-pass filter (cut-off wavelength: 700 nm); L, spherical lens ($f = 150$ mm); OL, objective lens (numerical aperture $NA = 0.25$); S, the FNLC device; BS, beam splitter; P, linear polarizer; IS, illumination source. The Q works at the fundamental frequency and is used to generate the FW vortex beam.

Supplementary Reference

1. Boyd, R. W. *Nonlinear optics*, 3rd edn. (Academic Press, 2008).