

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



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REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

In the manuscript entitled “Nonlinear geometric phase coded ferroelectric nematic fluids for nonlinear soft-matter photonics”, the authors use photoaligned ferroelectric nematic liquid crystal q-plates to realize second harmonic optical vortices with pin-locked topological charges. Notably, the authors for the first time propose the concept of nonlinear geometric phase to describe the nonlinear optical response of the FNLC samples. Temperature and E-field tuning is demonstrated, as well as the spin-selective response of a q-plate realized with the help of weakly chiral material. The experiments are in all cases accompanied by a theoretical explanation. The manuscript is well-written, the methodology is sound and the presented theory is rather well-supported by experimental evidence. However, I do feel that additional control experiments could be performed to make an even more convincing case. Additionally, some other questions or remarks should be addressed before justifying publication in Nature Communications.

*Proper alignment of the ferroelectric nematic LC RM734 into q-plates has enabled several nonlinear optical experiments, in which the second harmonic generation has been studied. However, it is a bit striking that photo-alignment with SD1 indeed enables high-quality alignment of the RM734 material, operated at very high temperatures. The high temperatures (even above 180°C during the filling process) can be expected to destroy or at least weaken the alignment pattern imprinted into the SD1 layer. Please comment on the stability of the photoalignment over time (at elevated temperatures) and comment on possible pitfalls that might prevent reproduction of the presented results.

*Please also comment on the importance of the slow cooling rate (line 140 and 480-486) and show results of textures that are obtained upon faster cooling (in the supporting information).

*Please explain how the surface anchoring controls the polar anchoring orientation: it is clear how the SD1 alignment can be linked to a preferred azimuthal orientation of the LC director, but please comment on what defines the orientation of the polarization vector. Are

both orientations of the polarization vector equally likely to occur? For example in line 186-187: what defines whether the orientation is radially outward or inward?

*The lens-shaped defect that are clearly visible in figure S2, seem to be absent in Fig. 3(c). Please explain this and also comment on the observation of large defect lines in Fig. 3(c). How is this defect formation minimized and can it be done in a systematic and reliable way? Please also comment on the director alignment in the lens-shaped polar domains and add a reference to previous studies on this.

*line 249,250: please clarify the mentioned ratio of 46:54 for the co and cross-CP components. Is this measured and should this be compared to the theoretically predicted 50:50?

*line 254-257: a general remark that comes to mind when reading the manuscript, is related to the fact that only a rather limited amount of experiments are performed, limited to cells with one particular thickness. To better support the theory and show the scalability of the proposed concept, I believe it is required to repeat the experiments at least in one more cell with a substantially different thickness.

Typos/ language errors:

-line 76, 77: “such fluids was first predicted....” please correct this sentence.

-line 80: “polarization that coupling” please correct this sentence (replace (coupling(by (couples”)

-line 86-87: it is unclear what is exactly meant with “the lack of principle for nonlinear light field control”. Please rephrase.

-line 117: “...plane. The x and y...” The two sentences should be merged (“.. plane, the x and y...”)

Reviewer #2 (Remarks to the Author):

The manuscript deals with the use of ferroelectric nematic liquid crystals as q-plates able to transform the angular momentum of the second harmonic beam, generated in these materials because of the broken mirror symmetry $n \rightarrow -n$ of the molecular director and the inherent optical anisotropy.

Although the topic promises to be very interesting and with a relevant degree of novelty, the manuscript is hard to follow for those who are not familiar with geometric phase and spin-orbit interaction. As such, in its current form the manuscript is not suitable for Nature Communications, at least not before major revisions are made. Apart from the use of too many acronyms, which makes the reading not easy and should be avoided, too many concepts are given without definition and too many explanations necessary for a proper understanding of the main text are left as Supplementary Information (most of the time without even writing it).

The following is a detailed list of things it would be necessary to fix.

In the abstract authors write “investigations on lithography-free, tunable, locally engineered nonlinear fluids have been scarce. Corresponding principle is unclear”. This is not totally true. Literature reports several examples of soft materials used for structuring light, for example liquid crystals with their nonlinear optical properties.

The sentence “The second-order nonlinear susceptibility of a nanoscale LC molecule is found carrying spin-dependent nonlinear geometric phases” is unclear. The concept of nonlinear geometric phase should be at least briefly defined even if this is an abstract. Moreover, the response of liquid crystals is of collective kind, and it seems to me not correct speaking about nanoscale molecules.

Several sentences are not correctly written, some have missing parts, some other do not make sense. See for example:

For instance, the growing and poling of thin-film lithium niobate usually requiring thousands of degrees Celsius temperature, kilovolt voltages, or femtosecond laser writing with high power density, leading to inconvenience, limited processing scale, high costs and energy consumption.

second harmonic generation properties have been scarcely made until recently, thanks to the emergence of ferroelectric nematic liquid crystals (FNLCs).

Such fluids was first predicted by M. Born in 1916 that if LC molecules were properly designed, their molecules would spontaneously arrange into polar structures

If the FNLC molecule is rotated by an orientation angle of α in the x-y plane.

For the FW with both spin and orbital angular momentum states $(\sigma, \ell)0\omega$, it will experience an optical SOI process in the LC q-plate with the topological charge of q .

In line 73, after the citations [23-26] I suggest mentioning also some applications of liquid crystals for light manipulation in the frame of nonlinear optics (as self-phase modulation, tunable lenses, self-diffraction, just to cite three examples)

Line 88: What do author mean with the term “nonlinear spontaneous polarization”? The nonlinearity is usually defined in relation to an external field, so I don’t understand how the spontaneous polarization could be nonlinear. This is also mentioned in the caption of Fig.1 (line 133) where authors seems to confuse the spontaneous polarization of the ferroelectric nematic with the nonlinear polarization arising from the optical nonlinear interaction with the light beam (see also the sentence “Here, we find that the nonlinear spontaneous polarization of ferroelectric nematic fluids will carry distinct nonlinear geometric phases for co- and cross-circularly polarized (CP) components”).

Line 93: The geometric-phase coded nonlinear FNLC q-plate should be described somehow. How did authors build the cell? Which kind of alignment they made? Why did they choose that specific kind of alignment? Citing Figure 1 without any further explanation is definitely not enough.

Overall, the paragraph from line 88 to line103 is not clear. There are too many claims and very few explanations.

Line 107: The “nonlinear dipole moment” is another unclear concept.

Line 119: Reading the main text it is not clear how equations (1) are derived. Authors should mention that the calculations are in the SI.

Line 125: What do author mean with “spatially variant LC molecules”? If the Authors were referencing a pattern of alignment, for sake of clarity, it is better to state explicitly that the pattern is azimuthal with $q=1$.

As a general comment on the section “Nonlinear polarization of the ferroelectric nematic LC molecule”, a compact form of the nonlinear response could help in reading the rest of the theoretical details in the manuscript. Moreover, clearly stating that only χ_{aaa} is considered can improve the clarity of the rest of the text.

Because of the absence of a proper discussion in the text, I found Figure 1 not clear, although very beautiful. Does it show an idea? Or does it show experimental results?

Line 143: the nonlinear LC device should be described at least briefly. Is it a simple planar cell?

Caption of Figure 2: “Polarization dependent SHG intensity from the FNLC molecules aligned along $\alpha = 0^\circ$ direction”. What does $\alpha = 0^\circ$ indicates? This clear from Figure 1b, but it should be indicated in the text also, otherwise one doesn’t know if α is the angle that n (and P) forms with the x or with the y axis. Additionally, the spectral response can be fully appreciated if the Authors provide also the normalize SHG spectra.

Lines 170-172: The sentence does not make sense. Ferroelectric nematic LC have not been widely used for generating optical spin-orbit angular momentum states in linear optics.

Lines 173-174: Again, the concept of vectorial LC q -plate should be introduced properly.

Lines 174-175: the way that specific pattern of n is obtained should be described, at least briefly. Then, the details can be left for the SI.

Lines 185-187: I don't understand what authors mean. The sentence is too short and the way they deduce the polarization arrangement based on the flexoelectric effect should be discussed in some more details.

Line 187: Authors mention a linearly polarized femtosecond laser. Did they use it for all their experiments or only for some of them?

Line 193 – 194. I'm not sure I understand the sentence. How can authors measure the vectorial polarization property of the structured second harmonic beam with an analyzer? I know this is discussed in section S3, however I think the term vectorial property is here inappropriate.

Figure 3: It is not clear to me how author determine the incident wave polarization directions in Fig. 3d and the vector distribution of the second harmonic in Fig. 3e. Also providing the intensity profile of the second harmonic might help in the understanding of the result.

Line 210: there are several claims in the text about the nonlinear geometric phase, but no explanations about what it is. I think authors should define it in detail, being this the main focus of the manuscript.

Line 240: “spin-locked nonlinear geometric phases” see the previous comment.

Line 250: the supporting information in section 4 does not explain the discrepancies in the power distribution of the circular components of the second harmonic waves. One can notice that supporting information in the Supplementary Section does not consider the interplay between the second harmonic generation and the coupling between the spin and the orbital angular momenta that occurs during propagation. Shouldn't result (S18) be valid

for weak anisotropy i.e. quasi scalar propagation? Moreover, in S19-24 there are few things that deserve some additional consideration:

In S19, shouldn't α depend on the position of the considered infinitesimal slab?

In S20 the Jones matrix should link input and output fields of the slab

S22-23 are not clear. The process should be a distributed one.

Line 257: The Authors should clearly explain the meaning of the half-wave condition in this context.

Lines 281-282 and Fig. S9: Authors observe a ferroelectric nematic phase down to 40°C in a material that is expected to be in the liquid crystal phase only until $T = 80^\circ\text{C}$. I think this observation deserves a comment. Also, with reference to Figure 5-a and Fig. S9, does the second harmonic intensity changes when heating or cooling the sample?

Lines 313-315: Authors write that doping the ferroelectric nematic with a chiral agent “drives the self-assembly of a three-dimensional space-variant chiral q-superstructure with right-handedness, as depicted in the left panel of Fig. 6a”. How do they know this is the exact pattern?

Lines 335-336: Dynamic phase should not be used in the stead of average phase factor.

Equation (1) should be fixed since the Authors are referring to the components of the nonlinear material polarization. Same goes for eq. S (6).

Minor issues on terminology:

“dimensions of light” should be reworded as “degrees of freedom of light”

Reviewer #3 (Remarks to the Author):

"I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts."

Re: manuscript NCOMMS-24-20086-T

We greatly appreciate all the reviewers for accessing our manuscript of NCOMMS-24-20086-T, and providing valuable comments. All reviewers' comments and suggestions are quite helpful for us to substantially improve the quality and readability of this manuscript.

We have carefully revised our manuscript and Supplementary Information by taking the respected reviewers' comments as appropriate into account. Our point-by-point response to the reviewers' comments and the changes made in our revised manuscript with yellow color highlighted track change as follows. We hope the new version would be found more convincing and meet the journal's criteria for publication.

Response to reviewers

Reviewer #1

In the manuscript entitled “Nonlinear geometric phase coded ferroelectric nematic fluids for nonlinear soft-matter photonics”, the authors use photoaligned ferroelectric nematic liquid crystal *q*-plates to realize second harmonic optical vortices with spin-locked topological charges. Notably, **the authors for the first time propose the concept of nonlinear geometric phase to describe the nonlinear optical response of the FNLC samples. Temperature and E-field tuning is demonstrated, as well as the spin-selective response of a *q*-plate realized with the help of weakly chiral material. The experiments are in all cases accompanied by a theoretical explanation. The manuscript is well-written, the methodology is sound and the presented theory is rather well-supported by experimental evidence.** However, I do feel that additional control experiments could be performed to make an even more convincing case. Additionally, some other questions or remarks should be addressed before justifying publication in Nature Communications.

Response:

We really appreciate the reviewer's encouraging comments and recommendation on our work. We have added correlated experimental results and carefully revised our manuscript accordingly, which are quite helpful for substantially improving the quality of our manuscript.

Q1.1 *Proper alignment of the ferroelectric nematic LC RM734 into *q*-plates has enabled several nonlinear optical experiments, in which the second harmonic generation has been studied. However, it is a bit striking that photo-alignment with SD1 indeed enables high-quality alignment of the RM734 material, operated at very high temperatures. The high temperatures (even above 180°C during the filling process) can be expected to destroy or at least weaken the alignment pattern imprinted into the SD1 layer. Please comment on the stability of the photoalignment over time (at elevated temperatures) and comment on possible pitfalls that might prevent reproduction of the presented results.

A1.1 Response:

We thank the reviewer for the valuable comment.

Fortunately, the photoalignment layer did keep stable throughout our tests. The stability of the SD1 photoalignment layer is also documented in established literature, indicating that high temperatures (for instance, 200 °C) do not damage or weaken the SD1-based alignment pattern over a span of 2 hours (as stated as “The thermal stability of the photoaligned azo dye layers is sufficiently high” by Akiyama et al. *Liq. Cryst.* 29, 1321 (2002)). Furthermore, both Takada et al. *SID Int Symp Digest Tech Papers* 34, 620 (2003) and Chigrinov et al. *Liq. Cryst. Today* 14, 1 (2005) have noted the resilience of the photoalignment layer when exposed to high temperatures (e.g., 230 °C). We have commented on the stability of the photoalignment accordingly.

We list some pitfalls that might prevent reproduction of the presented results:

- The total exposure dose of photoalignment layer should be $\geq 5 \text{ J}\cdot\text{cm}^{-2}$ to achieve the desired orientations of SD1.
- The quality of photoalignment pattern would be affected or destroyed if it is illuminated by UV light (e.g., $25 \text{ mW}\cdot\text{cm}^{-2}$ @ 365 nm), which should be avoided.
- The moisture is another detrimental factor that can induce the rotational reorientation due to the polar nature of SD1 molecules. The photopatterning is most efficient at 25 °C and < 40 % humidity. After photoalignment, the substrates and cells should be kept in the dry atmosphere.
- After the filling of the photoaligned cell with ferroelectric nematic liquid crystal (FNLC) at the temperature of 190 °C, an appropriate annealing process is required to prevent defects.

To ensure the replicability of our findings, we have comprehensively outlined our fabrication procedures in the Methods Section.

Original Text in Manuscript:

Lines 480-483, page 16: “Following the filling of the photoaligned cell with RM734 at a temperature of 190 °C, a slow thermal annealing treatment ($0.5 \text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$) is carried out which drives thermotropic alignment of LC molecules atop the SD1 layer.”

Revised Text in Revised Manuscript:

Lines 557-572, page 18: “After a total exposure dose of $\sim 5 \text{ J}\cdot\text{cm}^{-2}$, the desired alignment pattern of SD1 would be achieved. The photoalignment layer demonstrated exceptional thermal stability⁴⁶, which remained stable throughout our tests. While ultraviolet light illumination and high humidity environment should be avoided to prevent potential degradation of the photoalignment quality. Following the infiltration of RM734 material into the cell at 190 °C, a three-step thermal annealing treatment was performed to facilitate the self-assembly of FNLC atop the SD1 layer. In step (1), the device was rapidly cooled down from 190 °C to 132 °C at a rate of $5 \text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$; in step (2), the device was gradually cooled from 132 °C to 126 °C at a rate of $0.5 \text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ to facilitate the flexoelectric coupling relaxation during the nematic to ferroelectric

nematic phase transition; step (3), the device was cooled from 126 °C to 120 °C at a rate of 1 °C·min⁻¹ to prevent crystallization. It is important to set a slow cooling rate during the phase transition to prevent the formation of defective ferroelectric nematic structures. Polarized optical microscopic textures resulting from rapid cooling rates of 5 °C·min⁻¹ and 20 °C·min⁻¹ were provided in Supplementary Fig. 13 for contrast.”

Newly Added Reference:

46. Akiyama, H. *et al.* Synthesis and properties of azo dye aligning layers for liquid crystal cells. *Liq. Cryst.* **29**, 1321–1327 (2002).

Q1.2 *Please also comment on the importance of the slow cooling rate (line 140 and 480-486) and show results of textures that are obtained upon faster cooling (in the supporting information).

A1.2 Response:

We thank the reviewer for the valuable suggestion.

The slow cooling rate is indeed important for high-quality ferroelectric nematic structure formation. This is because (1) according to our experimental results, the fast cooling treatment (e.g., 10 °C·min⁻¹) leads to more defects after phase transition from nematic phase to ferroelectric nematic phase; (2) Other studies, such as Mertelj *et al.* *Phys. Rev. X* **8**, 041025 (2018), also discussed the slow cooling rate, as “Fast cooling (the heater was switched off) resulted in a more inhomogeneous transition and the creation of defects.”. During the nematic to ferroelectric nematic phase transition, the splay and twist relaxation rates of orientation fluctuations of FNLCs are suddenly slowed down compared to that of the nematic phase. Thus, a slow cooling rate is to provide a long relaxation time for the orientational ferroelastic transition thus to achieve high-quality ferroelectric nematic structures. We have added comment on the importance of the slow cooling rate.

The textures that are obtained upon faster cooling are provided in Supplementary Fig. 13.

Original Text in Manuscript:

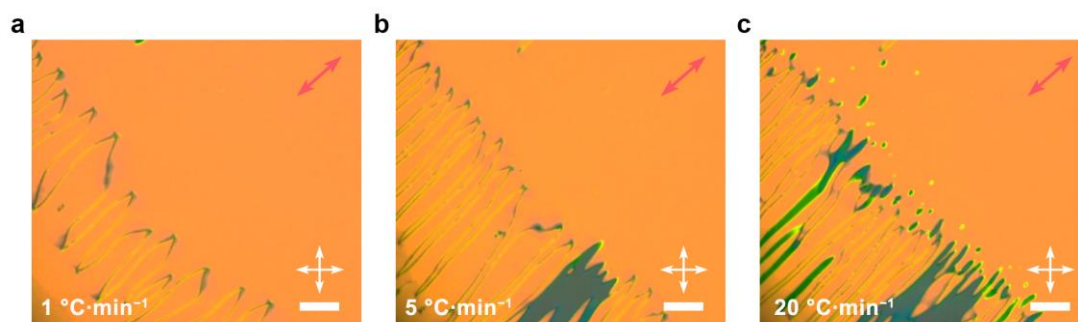
Lines 480-483, page 16: “Following the filling of the photoaligned cell with RM734 at a temperature of 190 °C, a slow thermal annealing treatment (0.5 °C·min⁻¹) is carried out which drives thermotropic alignment of LC molecules atop the SD1 layer.”

Revised Text in Revised Manuscript:

Lines 562-572, page 18: “Following the infiltration of RM734 material into the cell at 190 °C, a three-step thermal annealing treatment was performed to facilitate the self-assembly of FNLC atop the SD1 layer. In step (1), the device was rapidly cooled down from 190 °C to 132 °C at a rate of 5 °C·min⁻¹; in step (2), the device was gradually cooled from 132 °C to 126 °C at a rate of 0.5 °C·min⁻¹ to facilitate the flexoelectric coupling relaxation during the nematic to ferroelectric nematic phase transition; step

(3), the device was cooled from 126 °C to 120 °C at a rate of 1 °C·min⁻¹ to prevent crystallization. It is important to set a slow cooling rate during the phase transition to prevent the formation of defective ferroelectric nematic structures. Polarized optical microscopic textures resulting from rapid cooling rates of 5 °C·min⁻¹ and 20 °C·min⁻¹ were provided in Supplementary Fig. 13 for contrast.”

Newly added Supplementary Fig. 13:



Supplementary Fig. 13 | Polarized optical microscopic textures of the homogeneously photoaligned FNLC at different cooling rates of (a) 1 °C·min⁻¹, (b) 5 °C·min⁻¹, and (c) 20 °C·min⁻¹. The magenta arrows denote the direction of photoalignment. Scale bar: 100 μm.

Q1.3 *Please explain how the surface anchoring controls the polar anchoring orientation: it is clear how the SD1 alignment can be linked to a preferred azimuthal orientation of the LC director, but please comment on what defines the orientation of the polarization vector. Are both orientations of the polarization vector equally likely to occur? For example in line 186-187: what defines whether the orientation is radially outward or inward?

A1.3 Response:

We thank the reviewer for the valuable comment.

We did not deliberately create polar alignment surfaces. Thus, both orientations of the spontaneous polarization are equally likely to occur if providing a unidirectional photoalignment. The situation will be different if the alignment layer is photopatterned, especially with a splay alignment pattern. Our experimental results demonstrate the successful achievement of controllable *q*-plate patterns with a radial polarization distribution of FNLCs. The mechanism can be described as follows. In the ferroelectric nematic phase, where the polar ordering couples with the LC director orientation, the flexoelectric coupling, as well as the excluded volume of “wedge”-shaped LC molecules, also affects the orientation distribution of spontaneous polarization of FNLC [Ref. 39], which favors a splay deformation with respect to the polarization direction. This mean, when a splay *q*-plate pattern is photoaligned, a radially outward orientation of spontaneous polarization is more favorable, which satisfies the flexoelectric coupling with minimized excluded volume of “wedge”-shaped LC molecules. Following the

reviewer's suggestion, we have commented on what defines the arrangement of ferroelectric nematic q -plate orientation.

We further added the experiment to characterize the outward radial polarization of FNLC by applying an in-plane electric field (DC, $\pm 0.2 \text{ mV} \cdot \mu\text{m}^{-1}$). The polarized optical microscopic textures of the FNLC q -plate with a typical Maltese cross are presented in Figs. 3c-e. When a negative electric field is applied, the spontaneous polarization that does not align with the electric field undergoes reorientation, leading to the deformation of the Maltese cross pattern. When the electric field is inverted, we observe another deformed Maltese cross, of which the horizontal extinction brushes are deformed to the opposite direction. This result validates the radially outward polarization vector distribution in FNLC q -plate.

Original Text in Original Manuscript:

1. **Lines 177-187, page 6:** “Combined with the SH image of the FNLC q -plate in the near field (Supplementary Fig. 2), three typical features can be identified. Firstly, the Maltese cross, where the vectorial LC orientation is either parallel or perpendicular to the incident polarization, is clearly observed, and typical lens-shaped polar domains orient their elongated directions radially. Secondly, no obvious defect lines are found near the singularity point, indicating a continuously changed director distribution. In addition, a slightly whirl-like director distribution is observed very close to the singularity point, which serves to prevent the large accumulation of charges at the singularity point, thereby minimizing the free energy of the entire system. Thus, based on the flexoelectric effect³⁵, we can conclude that the polarizations are arranged radially with the same sign everywhere (Fig. 3b).”
2. **Lines 485-486, page 16:** “The polar LC molecules overall align themselves according to the SD1 orientations, forming the nonlinear LC q -plate.”

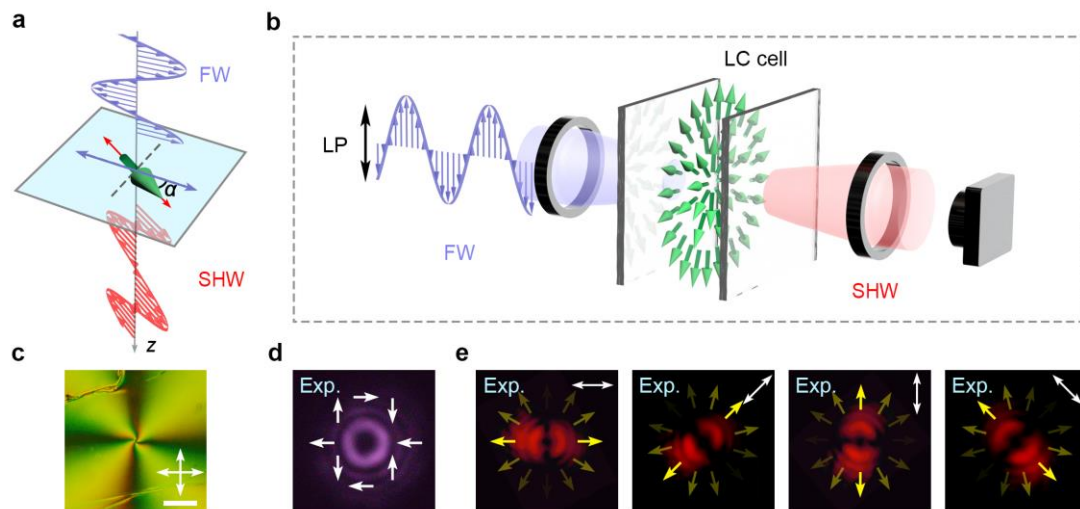
Revised Text in Revised Manuscript:

1. **Lines 208-222, page 7:** “A Maltese cross, where the polar LC orientation is either parallel or perpendicular to the crossed polarizers, is clearly observed. To deduce the inherent spontaneous polarization distribution, we apply an in-plane electric field (DC, $\pm 0.2 \text{ mV} \cdot \mu\text{m}^{-1}$) to the device. When a negative electric field is applied, the spontaneous polarization that does not align with the electric field undergoes reorientation, leading to the deformation of the Maltese cross pattern (Fig. 3d). When the electric field direction is inverted, the Maltese cross deforms in the opposite direction, with horizontal extinction brushes shifting contrary to the direction of the electric field (Fig. 3e). These phenomena indicate a radially outward polarization distribution in FNLC q -plate (Fig. 3b). Furthermore, such polarization distribution satisfies the flexoelectric coupling³⁹ with minimized excluded volume of wedge-shaped LC molecules, which facilitates the stable polar ordering formation. A whirl-like director distribution is also observed close to the singularity point, which prevents the large accumulation of charges at the

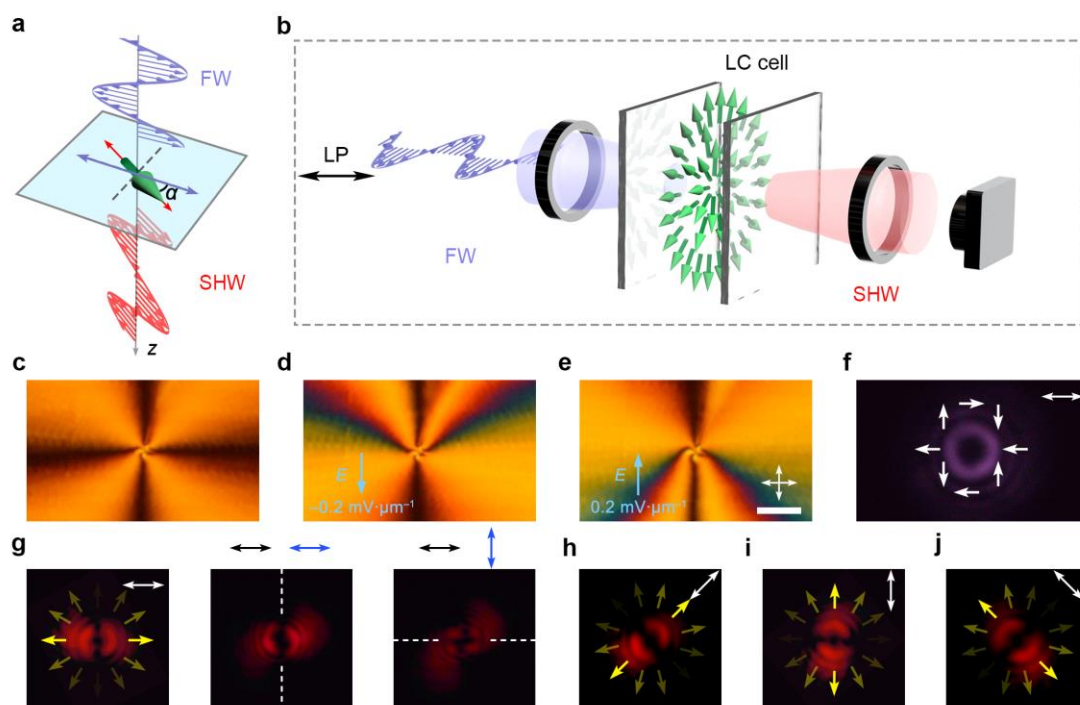
singularity point, thereby reducing the free energy of the entire system.”

2. **Lines 572-574, pages 18-19:** “Finally, the arrangement of ferroelectric nematic q -plate was guided by the photopatterned alignment layer combined with the flexoelectric coupling of FNLC.”

Original Fig. 3:



Revised Fig. 3:



Q1.4 *The lens-shaped defect that are clearly visible in figure S2, seem to be absent in Fig. 3(c). Please explain this and also comment on the observation of large defect lines in Fig. 3(c). How is this defect formation minimized and can it be done in a systematic

and reliable way? Please also comment on the director alignment in the lens-shaped polar domains and add a reference to previous studies on this.

A1.4 Response:

This is a good question. Thank you.

Polarized optical microscopic textures in Supplementary Fig. 2 and Fig. 3(c) are captured from two FNLC *q*-plate devices but with the same condition. The large defect line may be caused by some imperfections on the substrates. To minimize the occurrence of defects, it is imperative that the substrates are meticulously cleaned and that the sample undergoes a precise thermal annealing process (responded in A1.1). Thus, in revision, we further detailed a three-step thermal annealing treatment to provide a more systematic and reliable fabrication process for the FNLC *q*-plate.

To avoid confusion, a full plate of Fig. 3c is further provided in Supplementary Fig. 2a, which also exhibits lens-shaped domains.

The alignment conditions in and outside the lens-shaped polar domains are the same. The spontaneous polarization within and outside the lens-shaped polar domains are oppositely oriented (also see [Ref. 34]). Related descriptions have been added to the revised manuscript.

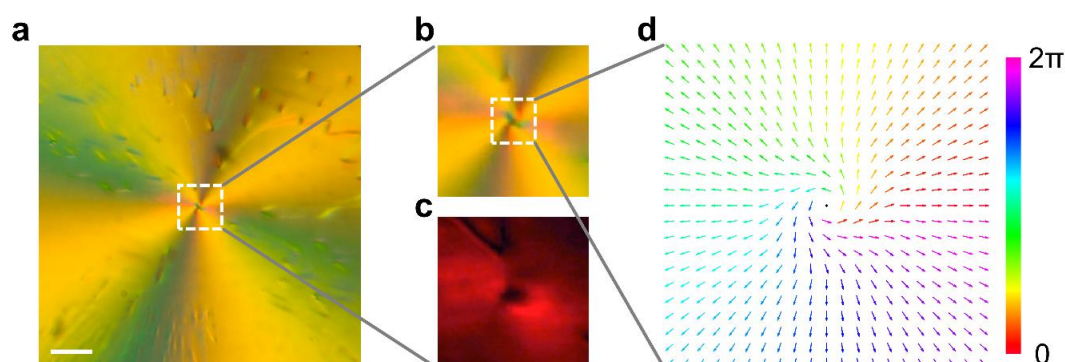
Original Text in Manuscript:

1. **Lines 143-144, page 4:** “...polar FN phase (120 °C, with distinctive lens-shaped polar domains).”
2. **Lines 480-483, page 16:** “Following the filling of the photoaligned cell with RM734 at a temperature of 190 °C, a slow thermal annealing treatment (0.5 °C·min⁻¹) is carried out which drives thermotropic alignment of LC molecules atop the SD1 layer.”

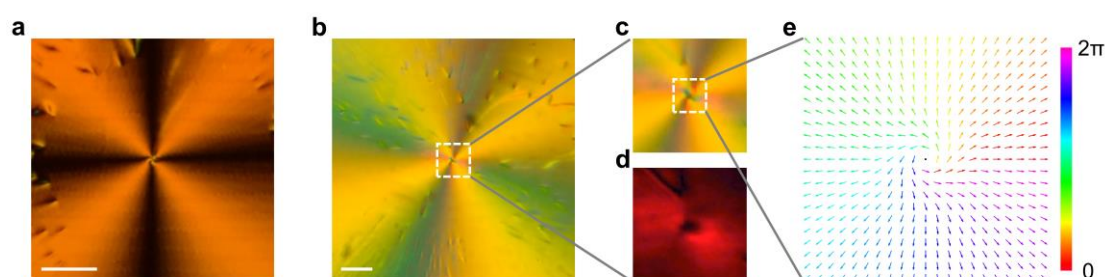
Revised Text in Revised Manuscript:

1. **Lines 151-154, page 5:** “...polar ferroelectric nematic phase (120 °C, with distinctive lens-shaped polar domains). The polar orientation within the lens-shaped domain demonstrates an inverse direction relative to the surrounding region³⁴.”
2. **Lines 562-569, page 18:** “Following the infiltration of RM734 material into the cell at 190 °C, a three-step thermal annealing treatment was performed to facilitate the self-assembly of FNLC atop the SD1 layer. In step (1), the device was rapidly cooled down from 190 °C to 132 °C at a rate of 5 °C·min⁻¹; in step (2), the device was gradually cooled from 132 °C to 126 °C at a rate of 0.5 °C·min⁻¹ to facilitate the flexoelectric coupling relaxation during the nematic to ferroelectric nematic phase transition; step (3), the device was cooled from 126 °C to 120 °C at a rate of 1 °C·min⁻¹ to prevent crystallization.”

Original Supplementary Fig. 2:



Revised Supplementary Fig. 2:



Q1.5 *line 249,250: please clarify the mentioned ratio of 46:54 for the co and cross-CP components. Is this measured and should this be compared to the theoretically predicted 50:50?

A1.5 Response:

Thank you for the reviewer's specialized question.

The intensities of the second harmonic waves (SHWs) with the co- and cross-circular polarization components were measured as 46:54. Theoretically, we calculated the intensities of the co- and cross-circularly polarized second harmonic (SH) optical vortices in Supplementary Section 3, that are 50:50.

Thus, our experimental result (46:54) is close to the theoretical calculation (50:50). The slight discrepancy might be attributed to the experimental error during the circular components filtered through a combined sheet polarizer and quarter waveplate.

We have revised these descriptions accordingly.

Original Text in Manuscript:

Lines 248-251, page 8: “Furthermore, the intensities of the SHWs with the co- and cross-CP components are almost evenly distributed, with a ratio of approximately 46:54, which is insensitive to the thickness of the LC film (see Supplementary Section 4).”

Revised Text in Revised Manuscript:

Lines 305-309, page 10: “The intensities of the co- and cross-circular polarization

components of the SHW are almost evenly distributed. The measured intensity ratio is approximately 46:54, which is very close to the theoretical prediction (see Supplementary Section 3). The minor discrepancy may be attributed to the imperfections of the polarizer and quarter-wave plate.”

Q1.6 *line 254-257: a general remark that comes to mind when reading the manuscript, is related to the fact that only a rather limited amount of experiments are preformed, limited to cells with one particular thickness. To better support the theory and show the scalability of the proposed concept, I believe it is required to repeat the experiments at least in one more cell with a substantially different thickness.

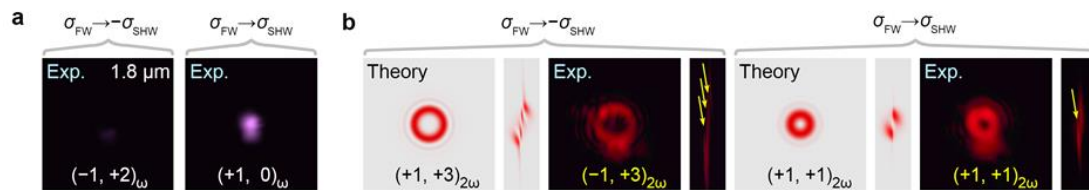
A1.6 Response:

Following the reviewer’s great suggestion, we repeated the experiments in the cell with a different thickness of 1.8 μm , to better support the theory and show the scalability of the proposed concept. The results are provided in Supplementary Fig. 6. For the fundamental wave (FW) with angular momentum state of $(+1, 0)_\omega$ incidence, the theoretical and experimental states of the SHWs include $(+1, 0)_\omega$, $(+1, +1)_{2\omega}$, and $(-1, +3)_{2\omega}$. The missing 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-circular polarization 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).

Added Text in Revised Manuscript:

Lines 317-321, page 10: “Furthermore, we conducted the experiments on an alternative device featuring a LC film thickness of 1.8 μm . The results are provided in Supplementary Fig. 6, which further validate our theory and show the scalability of the spin-locked nonlinear geometric phases with cascaded spin-orbit interactions.”

Newly added Supplementary Fig. 6:



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).

Q1.7 Typos/ language errors:

- line 76, 77: “such fluids was first predicted...” please correct this sentence.
- line 80: “polarization that coupling” please correct this sentence (replace (coupling(by (couples”))
- line 86-87: it is unclear what is exactly meant with “the lack of principle for nonlinear light field control”. Please rephrase.
- line 117: “...plane. The x and y...” The two sentences should be merged (“...plane, the x and y...”)

A1.7 Response:

We have corrected these typos/language errors accordingly.

Original Text in Manuscript:

1. **Line 76, page 2:** “Such fluids was first predicted...”
2. **Line 80, page 2:** “...polarization that coupling...”
3. **Lines 86-87, page 3:** “...mainly due to the difficulties in polarization manipulation and the lack of principle for nonlinear light field control.”
4. **Line 117, page 3:** “...plane. The x and y...”

Revised Text in Revised Manuscript:

1. **Line 79, page 2:** “Such fluids **were** first predicted...”
2. **Line 83, page 2:** “...polarization that **couples**...”
3. **Lines 88-89, page 3:** “...the light-matter interactions in LCs within the framework of second-order nonlinear optics have not yet been explored”
4. **Lines 123-125, page 4:** “If the polar direction of FNLC is rotated by an orientation angle of α relative to the x-axis, the second-order nonlinear polarization...”

Reviewer #2

The manuscript deals with the use of ferroelectric nematic liquid crystals as q -plates able to transform the angular momentum of the second harmonic beam, generated in these materials because of the broken mirror symmetry $\mathbf{n} \rightarrow -\mathbf{n}$ of the molecular director and the inherent optical anisotropy.

Although **the topic promises to be very interesting and with a relevant degree of novelty**, the manuscript is hard to follow for those who are not familiar with geometric phase and spin-orbit interaction. As such, in its current form the manuscript is not suitable for Nature Communications, at least not before major revisions are made. Apart from the use of too many acronyms, which makes the reading not easy and should be avoided, too many concepts are given without definition and too many explanations necessary for a proper understanding of the main text are left as Supplementary Information (most of the time without even writing it).

Response:

We greatly appreciate the respected reviewer for his or her positive comments on the interest and novelty of our work. According to the valuable suggestions, we have made changes throughout the revised manuscript (including those specifically listed in the point below) to make the manuscript more readable and easier to follow.

Some abbreviations (e.g., SOI, CP and CD) are now mentioned in full term following the reviewer's suggestion. We have also added definitions of important concepts (e.g., spin-locked nonlinear geometric phases) and necessary explanations (e.g., polarization distribution of the SH vector vortex beam) to the revised manuscript. We hope the new version would be found more convincing and suitable for publication now.

Q2.1 In the abstract authors write “investigations on lithography-free, tunable, locally engineered nonlinear fluids have been scarce. Corresponding principle is unclear”. This is not totally true. Literature reports several examples of soft materials used for structuring light, for example liquid crystals with their nonlinear optical properties.

A2.1 Response:

We agree with the reviewer that this sentence in the abstract is not totally true. This helps us to improve the rigor of the manuscript. Thank you.

Liquid crystals with nonlinear optical properties for structuring light can be broadly categorized into two types: conventional apolar liquid crystals and inversion-symmetry-breaking liquid crystals with spontaneous polarization.

- Conventional apolar liquid crystals: Previous literature leverages the optically induced birefringence, third-order processes (four-wave mixing), and photon absorption of these materials to modulate the transmitted light. In this way, nonlinear optical effects, such as self-phase modulation [Khoo et al. *Appl. Phys. Lett.* 52, 2108–2110 (1988)], self-focusing and defocusing [Braun et al. *Phys. Rev. A* 48, 611 (1993)], stimulated orientation scattering [Zeldovich et al. *JETP Lett. (Engl. Transl.)* 41, 515 (1985)], and nonlinear absorbers [Ehrlich et al. *Opt. Lett.* 22, 1843–1845 (1997)], were achieved.
- Polar liquid crystals: Second order processes such as second harmonic generation are conducted in symmetry-breaking smectic-C* LCs [Liu et al. *Opt. Lett.* 15, 267–269 (1990)], electrically poled nematic LCs [Saha et al. *Appl. Phys. Lett.* 34, 423–425 (1979)], or LC surfaces [Zhuang et al. *Phys. Rev. B* 59, 12632 (1999)]. However, these works did not encompass the steering of light beams or the shaping of their spatial modes within the context of second-order nonlinear optical processes.

To avoid confusion, we have revised the abstract to better reflect the current state of research and to clarify the unique aspects of our work. Specifically, we have focused on the second-order nonlinear wavefront engineering based on a spatially patterned polar liquid crystal with a large spontaneous electric polarization. This approach allows for simultaneous manipulation of multiple degrees of freedom of light, including

spatially varied phase, amplitude, and angular momenta, as well as frequency conversion. More importantly, our proposed theory introduces the concept of the geometric phase in nonlinear frequency conversion, which is distinct from the mechanisms reported in previous literature.

Original Text in Manuscript:

Lines 32-34, page 1: “...investigations on lithography-free, tunable, locally engineered nonlinear fluids have been scarce. Corresponding principle is unclear.”

Revised Text in Revised Manuscript:

Lines 32-33, page 1: “...investigation of programmable soft materials for second-order nonlinear wavefront engineering has never been performed previously.”

Q2.2 The sentence “The second-order nonlinear susceptibility of a nanoscale LC molecule is found carrying spin-dependent nonlinear geometric phases” is unclear. The concept of nonlinear geometric phase should be at least briefly defined even if this is an abstract. Moreover, the response of liquid crystals is of collective kind, and it seems to me not correct speaking about nanoscale molecules.

A2.2 Response:

We sincerely thank the reviewer for the great suggestions.

We have rewritten this sentence and added descriptions of the nonlinear geometric phase in the revised manuscript.

We agree with the reviewer that the response of liquid crystals is of collective kind in linear optics. However, in this work, we find the nonlinear geometric phases for the co- and cross-circular polarization components of SHW relative to the incident polarization are spin locked. Their intensities are equal. Both the nonlinear geometric phases and intensities are insensitive to the thickness of the LC film. Thus, for even a nanoscale LC molecule, the nonlinear response is still valid. Considering the SHG efficiency, we have changed the word “molecule” to “director” or deleted. We also added an experiment and discussions about the thickness tolerance of the nonlinear geometric phases.

Original Text in Manuscript:

1. **Lines 35-36, page 1:** “The second-order nonlinear susceptibility of a nanoscale LC molecule is found carrying spin-dependent nonlinear geometric phases.”
2. **Line 104, page 3:** “Nonlinear polarization of the ferroelectric nematic LC molecule”
3. **Lines 122-123, page 4:** “...the polar direction of the FNLC molecule...”
4. **Lines 133-134, page 4:** “The nonlinear polarization direction is along the long axis of the molecule.”
5. **Lines 166-167, page 5:** “Polarization dependent SHG intensity from the FNLC molecules aligned along $\alpha = 0^\circ$ direction.”
6. **Line 199, page 6:** “Schematic illustration of the SH response of an FNLC

molecule...”

7. **Lines 210-211, page 7:** “...to describe the nonlinear optical response of an FNLC molecule.”
8. **Lines 215-216, page 7:** “In the local coordinate of the FNLC molecule...”
9. **Lines 251-252, page 8:** “In order to experimentally verify the concept of nonlinear geometric phase in the FNLC molecule...”
10. **Lines 295-296, page 10:** “As depicted in Fig. 5b, the nonlinear dipoles of the LC molecules undergo an electrical realignment...”

Revised Text in Revised Manuscript:

1. **Lines 35-38, page 1:** “The second-order nonlinear susceptibility of a nanoscale LC director is found carrying spin-dependent nonlinear geometric phases, which describe imprinted nonlinearity phases originating from the spin coupling effect of light during the nonlinear frequency conversion process.”
2. **Line 110, page 3:** “Nonlinear polarization of the ferroelectric nematic LC”
3. **Line 131, page 4:** “...the polar direction of FNLC.”
4. **Lines 139-140, page 4:** “The nonlinear polarization is along the polar direction of LC.”
5. **Lines 184-185, page 6:** “Polarization-dependent SHG signals from the homogeneously photoaligned FNLC device. $\alpha = 0^\circ$, i.e., the orientation angle of FNLC with respect to x-axis is 0° .”
6. **Lines 237-238, page 8:** “Schematic illustration of the SH optical response of an FNLC director...”
7. **Lines 253-254, page 8:** “...to describe the nonlinear optical response of an FNLC director,...”
8. **Line 259, page 8:** “In the local coordinate of the unidirectionally aligned FNLC,...”
9. **Line 296, page 10:** “In order to experimentally verify the nonlinear geometric phases,...”
10. **Lines 361-362, page 12:** “As depicted in Fig. 5b, the spontaneous polarization of FNLC undergoes electrical realignment,...”

Added Text in Revised Manuscript:

Lines 309-321, page 10: “It is well known that the linear geometric phase effect relies on the conversion of the incident circular polarization light into the opposite helicity with imprinted geometric phases $e^{\pm i2\alpha}$. In this case, the energy allocation of co- and cross-circularly polarized light components is determined by the thickness of the FNLC film. In contrast, the nonlinear geometric phase encoded co- and cross-circular polarization SH vortices have an equal energy distribution regardless of the thickness of the LC film. This feature enables a remarkable wavelength tolerance of LC device in the SHG process, as evidenced by the generation of paired nonlinear optical vortices with equivalent energies at various wavelengths (see Supplementary Fig. 5). Furthermore, we conducted the experiments on an alternative device featuring a LC film thickness of 1.8 μm . The results are provided in Supplementary Fig. 6, which further validate our theory and show the scalability of the spin-locked nonlinear

geometric phases with cascaded spin-orbit interactions.”

Q2.3 Several sentences are not correctly written, some have missing parts, some other do not make sense. See for example:

- i) For instance, the growing and poling of thin-film lithium niobate usually requiring thousands of degrees Celsius temperature, kilovolt voltages, or femtosecond laser writing with high power density, leading to inconvenience, limited processing scale, high costs and energy consumption.
- ii) second harmonic generation properties have been scarcely made until recently, thanks to the emergence of ferroelectric nematic liquid crystals (FNLCs).
- iii) Such fluids was first predicted by M. Born in 1916 that if LC molecules were properly designed, their molecules would spontaneously arrange into polar structures
- iv) If the FNLC molecule is rotated by an orientation angle of α in the x-y plane.
- v) For the FW with both spin and orbital angular momentum states $(\sigma, \ell_0)_\omega$, it will experience an optical SOI process in the LC q -plate with the topological charge of q .

A2.3 Response:

We thank the reviewer for pointing out these mistakes. They have all been amended in our revised manuscript.

Original Text in Manuscript:

1. **Lines 61-64, page 2:** “For instance, the growing and poling of thin-film lithium niobate usually requiring thousands of degrees Celsius temperature, kilovolt voltages, or femtosecond laser writing with high power density, leading to inconvenience, limited processing scale, high costs and energy consumption.”
2. **Lines 75-76, page 2:** “...second harmonic generation properties have been scarcely made until recently, thanks to the emergence of ferroelectric nematic liquid crystals (FNLCs).”
3. **Lines 77-78, page 2:** “Such fluids was first predicted by M. Born in 1916 that if LC molecules were properly designed, their molecules would spontaneously arrange into polar structures.”
4. **Lines 116-118, page 3:** “If the FNLC molecule is rotated by an orientation angle of α in the x-y plane. The x and y components of the second-order nonlinear polarization can be analytically expressed as...”
5. **Lines 227-228, page 7:** “For the FW with both spin and orbital angular momentum states $(\sigma, \ell_0)_\omega$, it will experience an optical SOI process in the LC q -plate with the topological charge of q .”

Revised Text in Revised Manuscript:

1. **Lines 62-65, page 2:** “For instance, the growing and poling of lithium niobate usually **require** thousands of degrees Celsius temperature, kilovolt voltages or femtosecond laser writing with high power density, leading to inconvenient fabrication, limited scale, high cost, and large energy consumption.”

2. **Lines 76-78, page 2:** "...second-order nonlinear optical response in soft materials has garnered limited attention, owing to the intrinsic high symmetry of these materials that usually precludes the emergence of polar ordering."
3. **Lines 79-81, page 2:** "Such fluids were first predicted by M. Born in 1916 that if LC molecules were properly designed, their molecules would self-assemble into polar arrangements³¹."
4. **Lines 123-125, page 4:** "If the polar direction of FNLC is rotated by an orientation angle of α relative to the x -axis, the second-order nonlinear polarization can be analytically expressed as..."
5. **Lines 271-273, page 9:** "For the incident FW with an arbitrary spin and orbital angular momentum state $(\sigma, \ell_0)_\omega$, the light will experience a linear spin-orbit interaction in the ferroelectric nematic q -plate."

Q2.4 In line 73, after the citations [23-26] I suggest mentioning also some applications of liquid crystals for light manipulation in the frame of nonlinear optics (as self-phase modulation, tunable lenses, self-diffraction, just to cite three examples)

A2.4 Response:

We sincerely thank the reviewer for the constructive comment. We have added these applications in the introduction accordingly.

Original Text in Manuscript:

Lines 69-73, page 2: "A possible solution to these challenges would be soft materials^{21,22}, especially liquid crystals (LCs), due to their unique anisotropic optical properties, self-assembly characteristics, flexible domain engineering, as well as dynamic tunability, which have made them ideal for structured light field manipulation in the framework of linear optics²³⁻²⁶."

Revised Text in Revised Manuscript:

Lines 70-75, page 2: "A possible solution to these challenges would be soft materials^{21,22}, especially liquid crystals (LCs), due to their unique anisotropic optical properties, self-assembly characteristics, flexible domain tailoring, as well as dynamic tunability. These features have made them ideal for light manipulation in the regimes of both linear optics (e.g., optical vortex generator, holography, and topological beam steering)²³⁻²⁷ and nonlinear optics (e.g., self-phase modulation, self-focusing, and self-diffraction)²⁸⁻³⁰."

Newly Added References:

28. Khoo, I. C., Michael, R. R. & Finn, G. M. Self-phase modulation and optical limiting of a low-power CO₂ laser with a nematic liquid-crystal film. *Appl. Phys. Lett.* **52**, 2108–2110 (1988).
29. Braun, E., Faucheux, L. P. & Libchaber, A. Strong self-focusing in nematic liquid crystals. *Phys. Rev. A* **48**, 611–622 (1993).
30. Durbin, S. D., Arakelian, S. M. & Shen, Y. R. Laser-induced diffraction rings from

a nematic-liquid-crystal film. *Opt. Lett.* **6**, 411–413 (1981).

Q2.5 Line 88: What do author mean with the term “nonlinear spontaneous polarization”? The nonlinearity is usually defined in relation to an external field, so I don’t understand how the spontaneous polarization could be nonlinear. This is also mentioned in the caption of Fig.1 (line 133) where authors seems to confuse the spontaneous polarization of the ferroelectric nematic with the nonlinear polarization arising from the optical nonlinear interaction with the light beam (see also the sentence “Here, we find that the nonlinear spontaneous polarization of ferroelectric nematic fluids will carry distinct nonlinear geometric phases for co- and cross-circularly polarized (CP) components”).

A2.5 Response:

We agree with the reviewer on the difference between “spontaneous polarization” and “nonlinear polarization”, and we apologize for the wrong phrase used in Line 88 in the original manuscript. It has been amended accordingly. In addition, we have carefully gone through the whole manuscript and fixed them.

Original Text in Manuscript:

1. **Lines 88-91, page 3:** “Here, we find that the nonlinear spontaneous polarization of ferroelectric nematic fluids will carry distinct nonlinear geometric phases for co- and cross-circularly polarized (CP) components...”
2. **Lines 307-310, page 11:** “Periodic switching of the intensity of the SH vortices by electrical realignment of the nonlinear polarization of ferroelectric nematic q -plate from the transverse to the longitudinal directions.”

Revised Text in Revised Manuscript:

1. **Lines 90-92, page 3:** “Here, we find that the second-order nonlinear polarization of ferroelectric nematic LC (Figs. 1a and 1b) will carry distinct nonlinear geometric phases for the left- and right-handed circular polarization components of the SH signal.”
2. **Lines 374-376, page 13:** “Periodic switching of the intensity of the SH vortices by electrically tuning the spontaneous polarization of ferroelectric nematic q -plate from the transverse direction to the longitudinal direction.”

Q2.6 Line 93: The geometric-phase coded nonlinear FNLC q -plate should be described somehow. How did authors build the cell? Which kind of alignment they made? Why did they choose that specific kind of alignment? Citing Figure 1 without any further explanation is definitely not enough.

A2.6 Response:

We agree with the reviewer that more descriptions about geometric-phase coded nonlinear FNLC q -plate and Fig. 1 should be added. Thus, we have revised this paragraph accordingly.

The alignment pattern that is azimuthally continuously varied with the topological charge of $q = 1$ has been added.

We choose this alignment pattern in order to demonstrate the continuous control of the nonlinear geometric phase gradients for nonlinear wavefront shaping. The reason why we choose this specific kind of alignment has also been added.

We also detailed the description of the output optical states from the FNLC q -plate (Fig. 1c).

Original Text in Manuscript:

Lines 91-97, page 3: “In the proof-of-concept experiment, we study the nonlinear optical spin-orbit interaction (SOI) process in a geometric-phase coded nonlinear FNLC q -plate (Fig. 1). If the fundamental wave (FW) carries an angular momentum state of $(\sigma, \ell_0)_\omega$, where σ and ℓ_0 represent the spin and orbital angular momentum states, the cascaded linear and nonlinear optical SOI processes occur in the LCs. This will lead to the emergence of spin-locked second harmonic (SH) optical vortices with different orbital angular momentums (denoted as ℓ).”

Revised Text in Revised Manuscript:

Lines 92-104, page 3: “Continuous control of these nonlinearity phases for nonlinear wavefront shaping can be further achieved through spatially-variant engineering of the ferroelectric nematic fluid. In the proof-of-concept experiment, we study the nonlinear optical spin-orbit interaction process in a nonlinear geometric-phase coded topological FNLC q -plate. As illustrated in Fig. 1c, the polar orientations of FNLCs exhibit a radial distribution, which are achieved via photopatterning a continuously varied alignment configuration with a topological charge of $q = 1$. When a fundamental wave (FW) with angular momentum state of $(\sigma, \ell_0)_\omega$ is incident on the ferroelectric nematic q -structure, where σ ($\sigma = \pm 1$) and ℓ denote the spin and orbital angular momentum states, respectively, the cascaded spin-orbit interaction enables two spin-orbit locked SH optical vortices, specifically $(\sigma, 2\ell_0 + \sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}$, in addition to the two fundamental optical vortices, namely $(-\sigma, \ell_0 + 2\sigma q)_\omega$ and $(\sigma, \ell_0)_\omega$.”

Q2.7 Overall, the paragraph from line 88 to line 103 is not clear. There are too many claims and very few explanations.

A2.7 Response:

Thank you for the valuable comment. We have rewritten this paragraph to make it clearer.

Original Text in Manuscript:

Lines 88-103, page 3: “Here, we find that the nonlinear spontaneous polarization of ferroelectric nematic fluids will carry distinct nonlinear geometric phases for co- and cross-circularly polarized components, which will bring significant opportunities in the

active and arbitrary nonlinear waveform control. In the proof-of-concept experiment, we study the nonlinear optical spin-orbit interaction (SOI) process in a geometric-phase coded nonlinear FNLC q -plate (Fig. 1). If the fundamental wave (FW) carries an angular momentum state of $(\sigma, \ell_0)_\omega$, where σ and ℓ_0 represent the spin and orbital angular momentum states, the cascaded linear and nonlinear optical SOI processes occur in the LCs. This will lead to the emergence of spin-locked second harmonic (SH) optical vortices with different orbital angular momentums (denoted as ℓ). Moreover, we demonstrate the thermally and electrically switchable as well as chiral elastic force controlled nonlinear LC superstructures, in which both the generation and propagation of the SH vortices can be effectively manipulated. The geometric phase coded FNLC device represents a soft-matter photonic platform for nonlinear optics, which may have important applications in optical communications, high-resolution imaging, high-density data storage, and high-dimensional optical information processing.”

Revised Text in Revised Manuscript:

Lines 90-109, page 3: “Here, we find that the second-order nonlinear polarization of ferroelectric nematic LC (Figs. 1a and 1b) will carry distinct nonlinear geometric phases for the left- and right-handed circular polarization components of the SH signal. Continuous control of these nonlinearity phases for nonlinear wavefront shaping can be further achieved through spatially-variant engineering of the ferroelectric nematic fluid. In the proof-of-concept experiment, we study the nonlinear optical spin-orbit interaction process in a nonlinear geometric-phase coded topological FNLC q -plate. As illustrated in Fig. 1c, the polar orientations of FNLCs exhibit a radial distribution, which are achieved via photopatterning a continuously varied alignment configuration with a topological charge of $q = 1$. When a fundamental wave (FW) with angular momentum state of $(\sigma, \ell_0)_\omega$ is incident on the ferroelectric nematic q -structure, where σ ($\sigma = \pm 1$) and ℓ denote the spin and orbital angular momentum states, respectively, the cascaded spin-orbit interaction enables two spin-orbit locked SH optical vortices, specifically $(\sigma, 2\ell_0 + \sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}$, in addition to the two fundamental optical vortices, namely $(-\sigma, \ell_0 + 2\sigma q)_\omega$ and $(\sigma, \ell_0)_\omega$. Moreover, we present thermally and electrically switchable, as well as twist-elastic force controlled, FNLC q -superstructures, which enable the generating and tuning of SH optical vortices. This study introduces a soft-matter platform for nonlinear manipulation of light, offering a great promise of applications in optical communications, high-resolution imaging, high-density data storage, and high-dimensional optical information processing.”

Q2.8 Line 107: The “nonlinear dipole moment” is another unclear concept.

A2.8 Response:

We thank the reviewer for pointing out this. According to our previous responses to Q2.5, we have changed it to a more accurate description.

Original Text in Manuscript:

Lines 107-108, page 3: “The nonlinear dipole moment of the LC molecule is along the

polarization axis due to the integration of extended π -bridged donor-acceptor groups³⁶.”

Revised Text in Revised Manuscript:

Lines 113-114, page 3: “The long π -bridged donor-acceptor groups are along the spontaneous polarization of the RM734 molecule...”

Q2.9 Line 119: Reading the main text it is not clear how equations (1) are derived. Authors should mention that the calculations are in the SI.

A2.9 Response:

Following the reviewer's suggestion, we have added the description that the calculations are provided in the revised supplementary information.

Original Text in Manuscript:

Lines 120-121, pages 3-4: “...where ϵ_0 is the vacuum permittivity, 2ω is the angular frequency of the SH wave (SHW).”

Revised Text in Revised Manuscript:

Lines 127-129, page 4: “...where $\hat{e}_a$ represents the unit vector of the a -axis, ϵ_0 is the vacuum permittivity, 2ω is the angular frequency of the SH wave (SHW). Detailed formula derivation is provided in Supplementary Section 1.”

Q2.10 Line 125: What do author mean with “spatially variant LC molecules”? If the Authors were referencing a pattern of alignment, for sake of clarity, it is better to state explicitly that the pattern is azimuthal with $q=1$.

A2.10 Response:

We thank the reviewer for the valuable suggestion. We use “spatially variant LC molecules” to provide a more general situation where the FNLCs are photopatterned, rather than just homogeneously photoaligned. To make it clearer, we have revised this sentence.

Original Text in Manuscript:

Lines 123-125, page 4: “Assuming that the FW has an angular momentum state of $(\sigma, \ell_0)_\omega$ ($\sigma = \pm 1$) and the LC device consists of spatially variant LC molecules...”

Revised Text in Revised Manuscript:

Lines 191-193, pages 6-7: “Assuming that the FW possesses an angular momentum state of $(\sigma, \ell_0)_\omega$ ($\sigma = \pm 1$) and the LC device is photopatterned with a specific spatially-variant LC configuration...”

Q2.11 As a general comment on the section “Nonlinear polarization of the ferroelectric

nematic LC molecule”, a compact form of the nonlinear response could help in reading the rest of the theoretical details in the manuscript. Moreover, clearly stating that only $\chi_{aaa}^{(2)}$ is considered can improve the clarity of the rest of the text.

A2.11 Response:

We thank the reviewer for the valuable comment and we also agree with the reviewer that clearly stating that only $\chi_{aaa}^{(2)}$ is considered is necessary. To provide a more compact form of the nonlinear response, we have revised the Eq. (1) to make its physical implication clearer. We have elaborated on the physical implications of Eq. (1) in detail, and validated this with experimental results (Fig. 2e and newly added Fig. 2f) described in the next paragraph. The context of more complex situations with photopatterned FNLCs has been adjusted to the next section. We have also stated that only $\chi_{aaa}^{(2)}$ is considered for better understanding.

Original Text in Manuscript:

1. **Lines 116-123, pages 3-4:** “If the FNLC molecule is rotated by an orientation angle of α in the x - y plane. The x and y components of the second-order nonlinear polarization can be analytically expressed as

$$\begin{aligned} \mathbf{P}_x^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\alpha - \phi) \cos \alpha \hat{\mathbf{e}}_x \\ \mathbf{P}_y^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\alpha - \phi) \sin \alpha \hat{\mathbf{e}}_y, \end{aligned} \quad (1)$$

where ε_0 is the vacuum permittivity, 2ω is the angular frequency of the SH wave (SHW). Equation (1) reveals the unique anisotropy of the SH optical response (Fig. 1b), i.e., the excited nonlinear polarization aligns with the polar direction of the FNLC molecule (see more details in Supplementary Section 1).”

2. **Lines 153-156, page 5:** “By continuously rotating the incident polarization direction (ϕ) of the FW, the intensity of SHW generated from the FNLC ($\alpha = 0^\circ$) shows a dumbbell-shaped polar diagram in Fig. 2e, with the highest value observed at $\phi = 0^\circ$ and the lowest value at $\phi = 90^\circ$. These findings agree well with the theoretical prediction by Eq. (1).”
3. **Lines 169-172, page 5:** “To experimentally verify the capability of the light field manipulation for nonlinear soft-matter photonics, we focus on the optical SOI process in a nonlinear-geometric-phase coded FNLC q -plate, as a demonstration, which has been widely used for generating optical spin-orbit angular momentum states in linear optics.”

Revised Text in Revised Manuscript:

1. **Lines 122-135, pages 3-4:** “As the values of $\chi_{abb}^{(2)}$, $\chi_{bab}^{(2)}$, and $\chi_{bba}^{(2)}$ are about one tenth that of $\chi_{aaa}^{(2)}$, only $\chi_{aaa}^{(2)}$ is taken into account in our theoretical model. If the polar direction of FNLC is rotated by an orientation angle of α relative to the x -axis, the second-order nonlinear polarization can be analytically expressed as

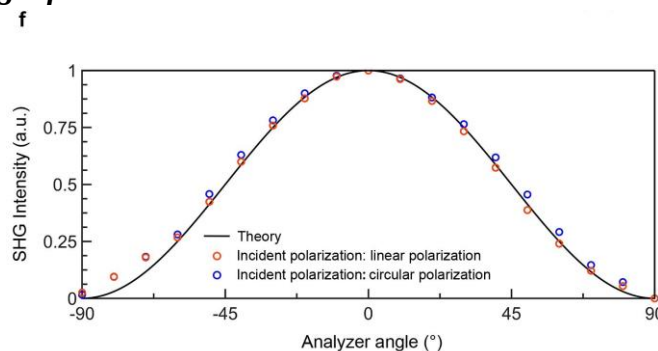
$$\begin{aligned}\mathbf{P}^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\phi - \alpha) (\cos \alpha \hat{\mathbf{e}}_x + \sin \alpha \hat{\mathbf{e}}_y) \\ &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\phi - \alpha) \hat{\mathbf{e}}_a\end{aligned}, \quad (1)$$

where $\hat{\mathbf{e}}_a$ represents the unit vector of the a -axis, ε_0 is the vacuum permittivity, 2ω is the angular frequency of the SH wave (SHW). Detailed formula derivation is provided in Supplementary Section 1. Equation (1) reveals the unique SH optical response (Fig. 1b). Specifically, the excited nonlinear polarization is linearly polarized along the a -axis, i.e., the polar direction of FNLC. It can be found that the magnitude of excited nonlinear polarization is primarily modulated by the $\cos^2$ term. The maximum and minimum SHG intensities occur at $\phi - \alpha = 0^\circ$ and $\phi - \alpha = 90^\circ$, respectively. This means the nonlinear optical response is polarization dependent and the largest SH intensity can be achieved when the polarization of FW is parallel to the FNLC.”

2. **Lines 162-173, page 5:** “When a linearly polarized FW passes through the device and the polarization (ϕ) of FW is continuously rotating, the intensity of SHW generated from the FNLC ($\alpha = 0^\circ$) shows a dumbbell-shaped polar diagram (Fig. 2e). This validates the polarization-dependent nonlinear optical response of the FNLC. The highest value is observed at $\phi = 0^\circ$, i.e., when the incident polarization is parallel to FNLC, and the lowest value is at $\phi = 90^\circ$. The polarization state of SHW is further characterized using an analyzer (Fig. 2f). Regardless of the incident polarization state (circular polarization or linear polarization), the analyzed SHG signals show a maximum intensity at 0° and a minimum intensity at $\pm 90^\circ$, which is consistent with Malus’s law. This behavior verifies the generated linearly polarization of the SHW, which is parallel to the polar orientation of FNLC and agrees well with the theoretical prediction by Eq. (1).”
3. **Lines 190-203, pages 6-7:** “By leveraging the programmable photopatterning of LCs, the nonlinear polarization can be further engineered. Assuming that the FW possesses an angular momentum state of $(\sigma, \ell_0)_\omega$ ($\sigma = \pm 1$) and the LC device is photopatterned with a specific spatially-variant LC configuration, the FW will undergo an optical spin-orbit interaction within the context of linear optics, resulting in transformation into a new angular momentum state of $(-\sigma, \ell_0 + 2\sigma)_\omega$. Concurrently, distinctive angular momentum states within the framework of nonlinear optics will be generated, which are determined by both the FW states and the geometric phase encoded FNLC structure (Fig. 1c).

A particular class of wavefront engineering that has garnered substantial attention over the past decade is the generation of beams carrying orbital angular momentum. Here, in the proof-of-concept experiment, we photopattern a topological q -plate structure with the polar FNLC material to demonstrate the intriguing spin-orbit interactions in the SHG process (for more details of photopatterning, see Methods and Supplementary Section 2).”

Newly added Fig. 2f:



f, Relationship between the SHG intensity and the angle of the analyzer. The SHG intensities are normalized for the linear polarization incidence and circular polarization incidence, respectively.

Q2.12 Because of the absence of a proper discussion in the text, I found Figure 1 not clear, although very beautiful. Does it show an idea? Or does it show experimental results?

A2.12 Response:

We thank the reviewer for the valuable comment. Figure 1 shows the main idea of our work. We also use Figure 1 to better understand the theoretical predictions. To better convey this, we have revised the correlated context accordingly, including the last paragraph in the introduction, the theoretical prediction of nonlinear polarization for linear polarization incidence (Fig. 1b) in the section “Nonlinear polarization of the ferroelectric nematic LC”, and the description of Fig. 1c in the section “Optical spin-orbit interaction in the nonlinear LC *q*-plate”.

Original Text in Manuscript:

- Lines 88-97, page 3:** “Here, we find that the nonlinear spontaneous polarization of ferroelectric nematic fluids will carry distinct nonlinear geometric phases for co- and cross-circularly polarized components, which will bring significant opportunities in the active and arbitrary nonlinear waveform control. In the proof-of-concept experiment, we study the nonlinear optical spin-orbit interaction (SOI) process in a geometric-phase coded nonlinear FNLC *q*-plate (Fig. 1). If the fundamental wave (FW) carries an angular momentum state of $(\sigma, \ell_0)_\omega$, where σ and ℓ_0 represent the spin and orbital angular momentum states, the cascaded linear and nonlinear optical SOI processes occur in the LCs. This will lead to the emergence of spin-locked second harmonic (SH) optical vortices with different orbital angular momentums (denoted as ℓ).”
- Lines 121-123, page 4:** “Equation (1) reveals the unique anisotropy of the SH optical response (Fig. 1b), i.e., the excited nonlinear polarization aligns with the polar direction of the FNLC molecule (see more details in Supplementary Section 1).”
- Lines 123-129, page 4:** “Assuming that the FW has an angular momentum state of

$(\sigma, \ell_0)_\omega$ ($\sigma = \pm 1$) and the LC device consists of spatially variant LC molecules, the FW will experience the optical SOI process (in the framework of linear optics) and is converted into a new angular momentum state $(-\sigma, \ell_0 + 2\sigma)_\omega$. In the meantime, the angular momentum states of the SHWs are generated, which are determined by both the FW states and the geometric phased coded FNLCs (Fig. 1c).”

Revised Text in Revised Manuscript:

1. **Lines 90-104, page 3:** “Here, we find that the second-order nonlinear polarization of ferroelectric nematic LC (Figs. 1a and 1b) will carry distinct nonlinear geometric phases for the left- and right-handed circular polarization components of the SH signal. Continuous control of these nonlinearity phases for nonlinear wavefront shaping can be further achieved through spatially-variant engineering of the ferroelectric nematic fluid. In the proof-of-concept experiment, we study the nonlinear optical spin-orbit interaction process in a nonlinear geometric-phase coded topological FNLC q -plate. As illustrated in Fig. 1c, the polar orientations of FNLCs exhibit a radial distribution, which are achieved via photopatterning a continuously varied alignment configuration with a topological charge of $q = 1$. When a fundamental wave (FW) with angular momentum state of $(\sigma, \ell_0)_\omega$ is incident on the ferroelectric nematic q -structure, where σ ($\sigma = \pm 1$) and ℓ denote the spin and orbital angular momentum states, respectively, the cascaded spin-orbit interaction enables two spin-orbit locked SH optical vortices, specifically $(\sigma, 2\ell_0 + \sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}$, in addition to the two fundamental optical vortices, namely $(-\sigma, \ell_0 + 2\sigma q)_\omega$ and $(\sigma, \ell_0)_\omega$.”
2. **Lines 129-135, page 4:** “Equation (1) reveals the unique SH optical response (Fig. 1b). Specifically, the excited nonlinear polarization is linearly polarized along the a -axis, i.e., the polar direction of FNLC. It can be found that the magnitude of excited nonlinear polarization is primarily modulated by the $\cos^2$ term. The maximum and minimum SHG intensities occur at $\phi - \alpha = 0^\circ$ and $\phi - \alpha = 90^\circ$, respectively. This means the nonlinear optical response is polarization dependent and the largest SH intensity can be achieved when the polarization of FW is parallel to the FNLC.”
3. **Lines 190-197, pages 6-7:** “By leveraging the programmable photopatterning of LCs, the nonlinear polarization can be further engineered. Assuming that the FW possesses an angular momentum state of $(\sigma, \ell_0)_\omega$ ($\sigma = \pm 1$) and the LC device is photopatterned with a specific spatially-variant LC configuration, the FW will undergo an optical spin-orbit interaction within the context of linear optics, resulting in transformation into a new angular momentum state of $(-\sigma, \ell_0 + 2\sigma)_\omega$. Concurrently, distinctive angular momentum states within the framework of nonlinear optics will be generated, which are determined by both the FW states and the geometric phase encoded FNLC structure (Fig. 1c).”

Q2.13 Line 143: the nonlinear LC device should be described at least briefly. Is it a simple planar cell?

A2.13 Response:

We thank the reviewer for the question. The nonlinear LC device is based on a planar cell. It comprises two glass substrates coated with ITO electrodes and photoalignment layers. After homogeneously photoaligned and infiltrated with FNLC, the device is encapsulated and undergoes a fine thermal annealing treatment to avoid large defects, thus achieving a uniform spontaneous polarization distribution. Then, we use this device to perform nonlinear optical experiments. The electrodes facilitate the application of electric fields to electrically tune the nonlinear optical properties.

Following the reviewer's suggestion, we have added this in the revised manuscript to provide a clearer understanding of the device structure.

Original Text in Manuscript:

Pages 139-142, page 4: “Then, we experimentally investigate the SHG response of the homogeneously photoaligned FNLCs. The sample undergoes fine thermal annealing treatment to achieve large domains with uniform spontaneous polarization before testing the SHG properties.”

Revised Text in Revised Manuscript:

Pages 145-150, pages 4-5: “Then, we experimentally demonstrate the SH optical response in a homogeneously photoaligned FNLC device. Such device comprises two glass substrates coated with ITO electrodes and photoalignment layers. After unidirectionally photoaligned and infiltrated with FNLC, the device is encapsulated and undergoes a thermal annealing treatment (see Methods for more details) to achieve a uniform spontaneous polarization distribution.”

Q2.14 Caption of Figure 2: “Polarization-dependent SHG intensity from the FNLC molecules aligned along $\alpha = 0^\circ$ direction”. What does $\alpha = 0^\circ$ indicates? This clear from Figure 1b, but it should be indicated in the text also, otherwise one doesn't know if α is the angle that **n** (and **P**) forms with the x or with the y axis. Additionally, the spectral response can be fully appreciated if the Authors provide also the normalize SHG spectra.

A2.14 Response:

We thank the reviewer for the valuable suggestion. Following the reviewer's suggestion, we have clarified the orientation angle of α in the revised manuscript. We have normalized the intensity profile and SHG spectra in Figs. 2b-d.

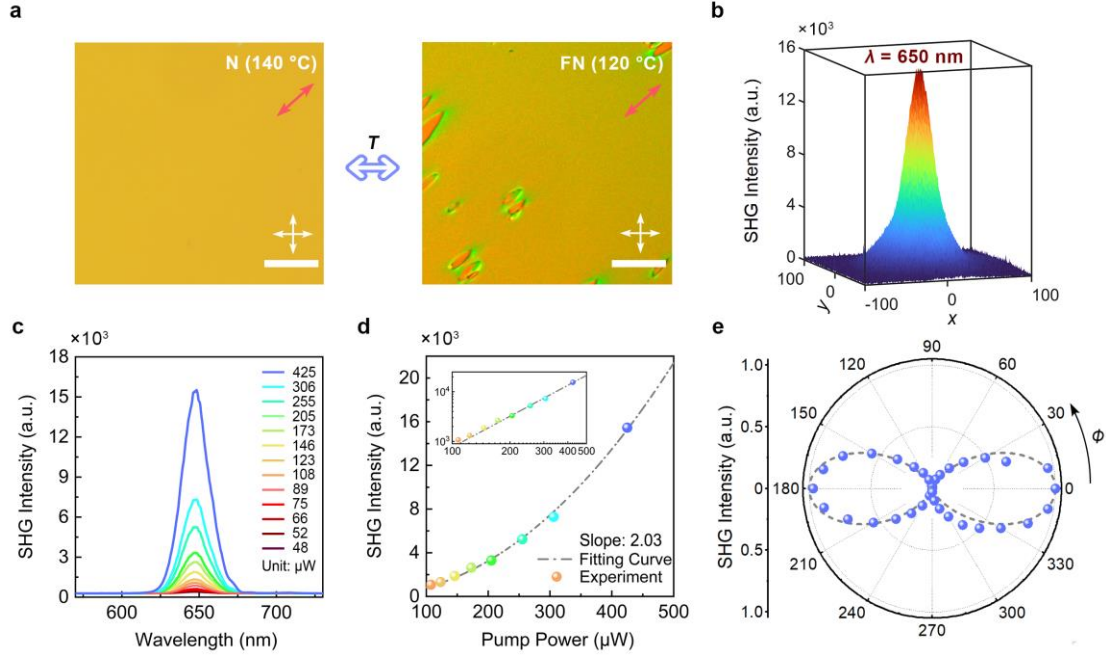
Original Text in Manuscript:

- Pages 116-117, page 3:** “If the FNLC molecule is rotated by an orientation angle of α in the x - y plane...”
- Pages 166-167, page 5:** “Polarization dependent SHG intensity from the FNLC molecules aligned along $\alpha = 0^\circ$ direction.”

Revised Text in Revised Manuscript:

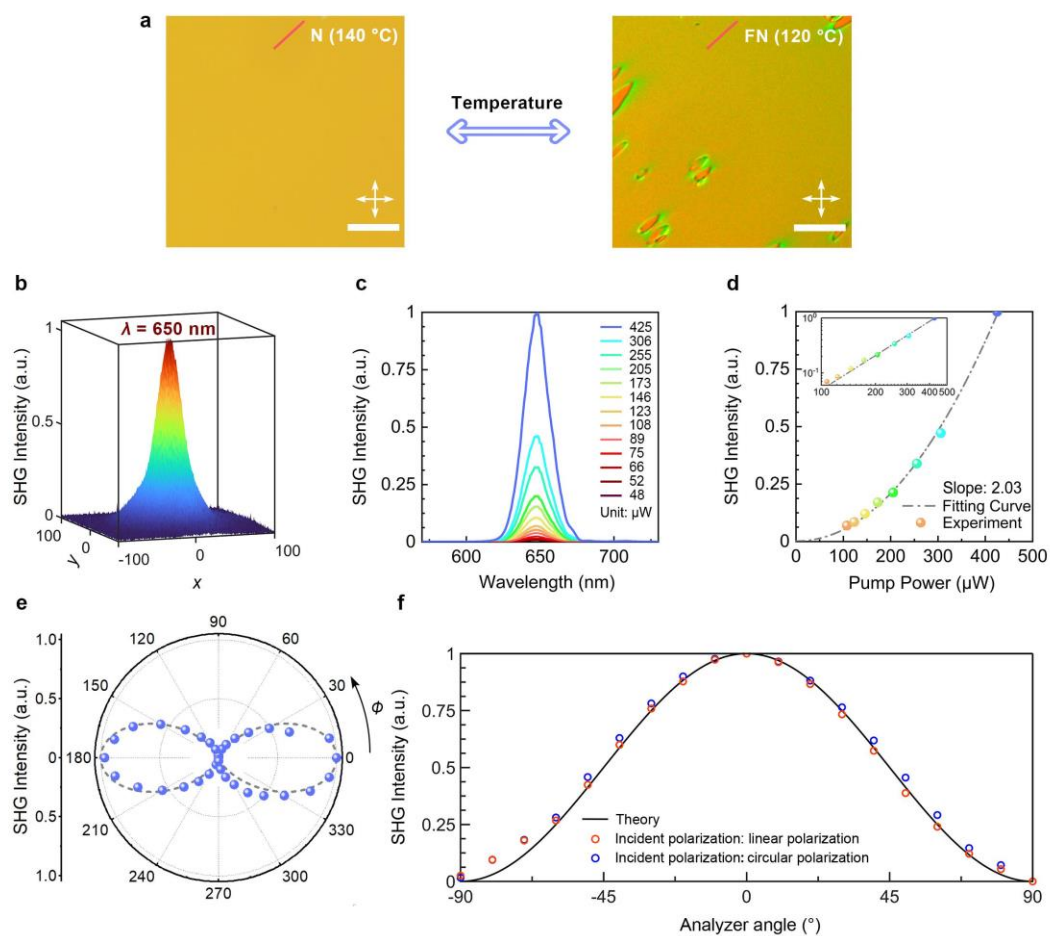
1. Pages 123-124, page 4: “If the polar direction of FNLC is rotated by an orientation angle of α relative to the x -axis,...
2. Pages 184-186, page 6: “Polarization-dependent SHG signals from the homogeneously photoaligned FNLC device. $\alpha = 0^\circ$, i.e., the orientation angle of FNLC with respect to the x -axis is 0° .”

Original Fig. 2:



b, Profile of the SHG beam, $\lambda_{SH} = 650$ nm. **c**, SHG spectra of the homogeneous FNLCs under different pumping powers. The central wavelength of the SHG spectra is 650 nm, corresponding to the fundamental wavelength at 1300 nm. **d**, Typical power-dependent SHG intensity. The linearly fitted slope value is 2.03 (inset), confirming the quadratic nature of the SHG process.

Revised Fig. 2:



b, **Normalized** profile of the SHG beam, $\lambda_{SH} = 650$ nm. **c**, **Normalized** SHG spectra of the homogeneous FNLCs under different pumping powers. The central wavelength of the SHG spectra is 650 nm, corresponding to the fundamental wavelength at 1300 nm. **d**, **Normalized** power-dependent SHG intensity.

Q2.15 Lines 170-172: The sentence does not make sense. Ferroelectric nematic LC have not been widely used for generating optical spin-orbit angular momentum states in linear optics.

A2.15 Response:

We agree with the reviewer that ferroelectric nematic LC have not been widely used for generating optical spin-orbit angular momentum states in linear optics. Accordingly, we have rewritten the sentence in the revised manuscript.

Original Text in Manuscript:

Lines 169-172, page 5: “we focus on the optical SOI process in a nonlinear-geometric-phase coded FNLC q -plate, as a demonstration, which has been widely used for generating optical spin-orbit angular momentum states in linear optics.”

Revised Text in Revised Manuscript:

Lines 198-203, page 7: “A particular class of wavefront engineering that has garnered substantial attention over the past decade is the generation of beams carrying orbital angular momentum. Here, in the proof-of-concept experiment, we photopattern a topological q -plate structure with the polar FNLC material to demonstrate the intriguing spin-orbit interactions in the SHG process (for more details of photopatterning, see Methods and Supplementary Section 2).”

Q2.16 Lines 173-174: Again, the concept of vectorial LC q -plate should be introduced properly.

A2.16 Response:

Thanks for the valuable suggestion. We use the word “vectorial” to distinguish conventional apolar q -plates and polar q -plates. In revision, to mitigate potential confusion arising from the use of the term “vectorial” for both the polar LC structure and the vector light field, we have revised the terminology from “vectorial LC” to “polar LC” or “ferroelectric nematic LC”.

Original Text in Manuscript:

1. **Line 37, page 1:** “By taking advantage of photopatterning, three-dimensional vectorial LC q -structures...”
2. **Lines 178-180, page 6:** “...Maltese cross, where the vectorial LC orientation is either parallel or perpendicular to the incident polarization, is clearly observed...”
3. **Lines 278-279, page 6:** “Here, we show the thermally and electrically switchable nonlinear optical responses of the vectorial LC q -plate.”

Revised Text in Revised Manuscript:

1. **Lines 38-39, page 1:** “By taking advantage of photopatterning, three-dimensional ferroelectric nematic q -structures...”
2. **Lines 208-209, page 7:** “...Maltese cross, where the polar LC orientation is either parallel or perpendicular to the crossed polarizers, is clearly observed...”
3. **Lines 340-342, page 11:** “Here, we show the thermally and electrically switchable nonlinear optical responses of the ferroelectric nematic LC q -plate.”

Q2.17 Lines 174-175: the way that specific pattern of $\mathbf{n}$ is obtained should be described, at least briefly. Then, the details can be left for the SI.

A2.17 Response:

We agree with the reviewer that the way that the specific pattern of $\mathbf{n}$ is obtained should be described to inform the readers in the main text. We have revised it accordingly.

Original Text in Manuscript:

Lines 170-175, pages 5-6: “...we focus on the optical SOI process in a nonlinear-

geometric-phase coded FNLC q -plate, as a demonstration, which has been widely used for generating optical spin-orbit angular momentum states in linear optics. Figures 3a and 3b schematically depict the vectorial optical property of the SHWs from a vectorial LC q -plate. The spatially variant LC directors follow the equation of $\alpha(\varphi) = q\varphi + \alpha_0$, where φ is the azimuthal angle, $q = 1$ is the topological charge, and α_0 is a constant.”

Revised Text in Revised Manuscript:

Lines 200-207, page 7: “...we photopattern a topological q -plate structure with the polar FNLC material to demonstrate the intriguing spin-orbit interactions in the SHG process (for more details of photopatterning, see Methods and Supplementary Section 2).

Firstly, we study the optical properties of the SHW when a linearly polarized femtosecond laser passes through the device (Figs. 3a and 3b). The spatially variant LC director distribution follows the equation of $\alpha(\varphi) = q\varphi$, where φ is the azimuthal angle and q is the topological charge set to 1.”

Q2.18 Lines 185-187: I don’t understand what authors mean. The sentence is too short and the way they deduce the polarization arrangement based on the flexoelectric effect should be discussed in some more details.

A2.18 Response:

The reviewer is correct; we did not express this clearly. Actually, we deduce the polarization arrangement mainly based on the experimental results combined with the flexoelectric coupling effect.

In the revised manuscript, we have rewritten the text and provided a more convincing experimental result to deduce the polarization arrangement by applying an in-plane electric field (DC, $\pm 0.2 \text{ mV} \cdot \mu\text{m}^{-1}$). The polarized optical microscopic texture of the FNLC q -plate with a typical Maltese cross is presented in Figs. 3c-e. When a negative electric field is applied, the spontaneous polarization that does not align with the electric field undergoes reorientation, leading to the deformation of the Maltese cross pattern. When the electric field is inversed, we observe another deformed Maltese cross, of which the horizontal extinction brushes are deformed to the opposite direction. This result validates the radially outward polarization vector distribution in FNLC q -plate.

Original Text in Manuscript:

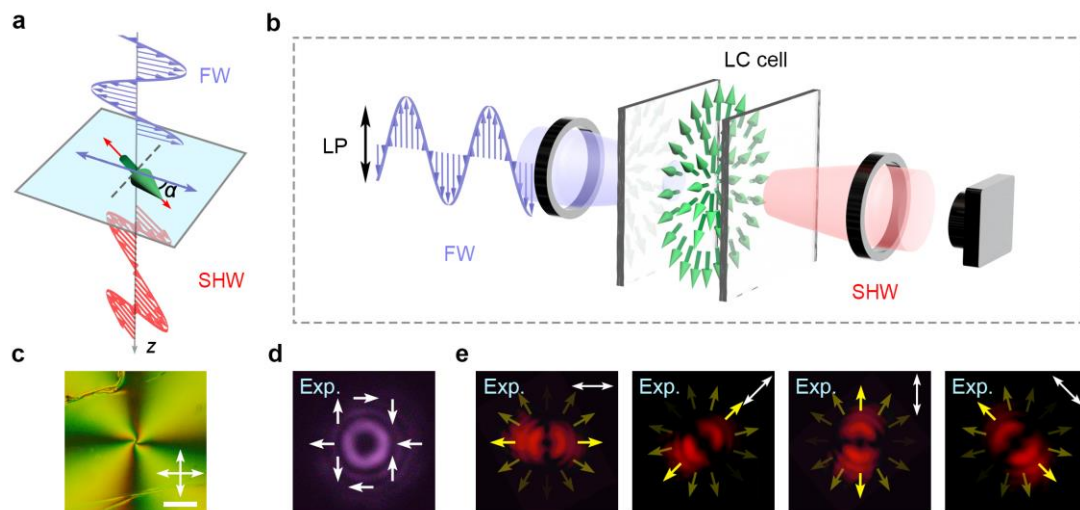
Lines 177-187, page 6: “Combined with the SH image of the FNLC q -plate in the near field (Supplementary Fig. 2), three typical features can be identified. Firstly, the Maltese cross, where the vectorial LC orientation is either parallel or perpendicular to the incident polarization, is clearly observed, and typical lens-shaped polar domains orient their elongated directions radially. Secondly, no obvious defect lines are found near the singularity point, indicating a continuously changed director distribution. In addition, a slightly whirl-like director distribution is observed very closed to the

singularity point, which serves to prevent the large accumulation of charges at the singularity point, thereby minimizing the free energy of the entire system. Thus, based on the flexoelectric effect³⁵, we can conclude that the polarizations are arranged radially with the same sign everywhere (Fig. 3b)."

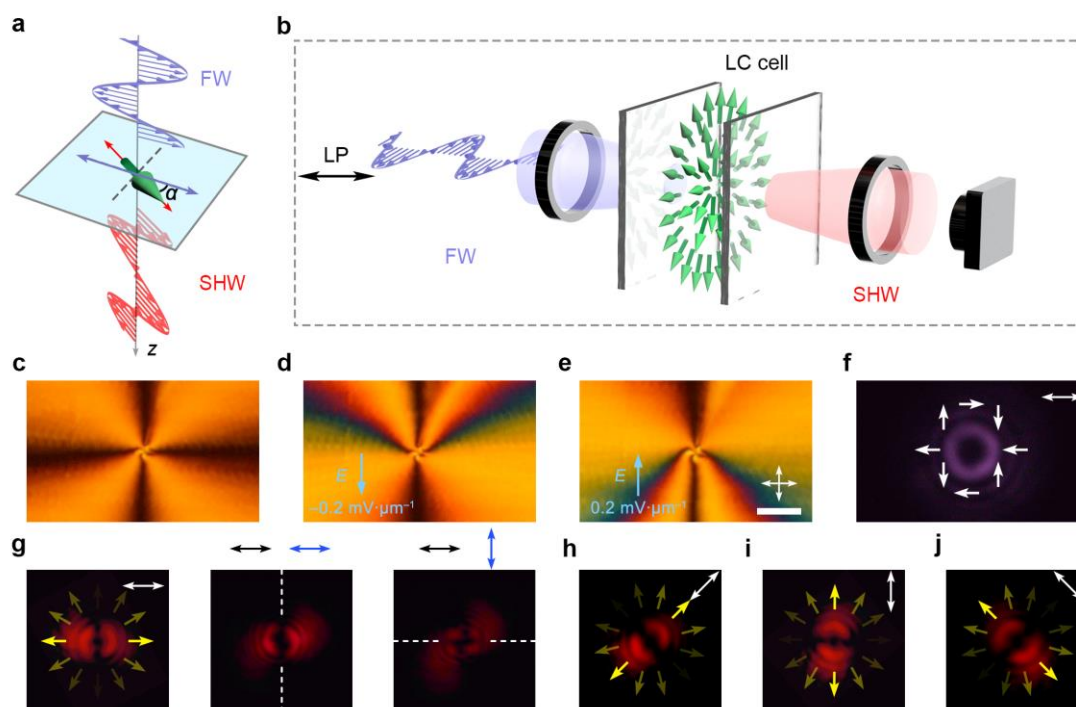
Revised Text in Revised Manuscript:

Lines 208-222, page 7: "A Maltese cross, where the polar LC orientation is either parallel or perpendicular to the crossed polarizers, is clearly observed. To deduce the inherent spontaneous polarization distribution, we apply an in-plane electric field (DC, $\pm 0.2 \text{ mV} \cdot \mu\text{m}^{-1}$) to the device. When a negative electric field is applied, the spontaneous polarization that does not align with the electric field undergoes reorientation, leading to the deformation of the Maltese cross pattern (Fig. 3d). When the electric field direction is inversed, the Maltese cross deforms in the opposite direction, with horizontal extinction brushes shifting contrary to the direction of the electric field (Fig. 3e). These phenomena indicate a radially outward polarization distribution in FNLC *q*-plate (Fig. 3b). Furthermore, such polarization distribution satisfies the flexoelectric coupling³⁹ with minimized excluded volume of wedge-shaped LC molecules, which facilitates the stable polar ordering formation. A whirl-like director distribution is also observed close to the singularity point, which prevents the large accumulation of charges at the singularity point, thereby reducing the free energy of the entire system."

Original Fig. 3:



Revised Fig. 3:



Q2.19 Line 187: Authors mention a linearly polarized femtosecond laser. Did they use it for all their experiments or only for some of them?

A2.19 Response:

We thank the reviewer's question.

The linearly polarized femtosecond laser is used in some of our experiments. To avoid confusion, we have reviewed the entire manuscript and clarified the incident polarization of FW for different experiments.

Original Text in Manuscript:

- Lines 145-147, page 4:** "The high-quality alignment technique enables the observation of a strong emission of the SHW with a Gaussian beam profile, as depicted in Fig. 2b and Supplementary Fig. 1."
- Lines 153-156, page 5:** "By continuously rotating the polarization direction (ϕ) of the FW, the intensity of the SHW shows a dumbbell-shaped polar diagram in Fig. 2e, with the highest value observed at $\phi = 0^\circ$ and the lowest value at $\phi = 90^\circ$."
- Lines 209-211, page 7:** "In order to better understand the nonlinear optical SOI processes occurring in the LC q -plate, we for the first time propose the concept of nonlinear geometric phase to describe the nonlinear optical response of an FNLC molecule^{5,37}."
- Lines 283-285, page 9:** "For the FW with Gaussian type distribution $(+1, 0)_\omega$, the SHG intensity as a function of the temperature is shown in Fig. 5a."

Revised Text in Revised Manuscript:

1. **Lines 155-158, page 5:** “The high-quality alignment of FNLC enables the observation of a strong emission of the SHW with a Gaussian beam profile when a circularly polarized femtosecond laser (FW) passes through the device, as depicted in Fig. 2b and Supplementary Fig. 1.”
2. **Lines 162-166, page 5:** “When a linearly polarized FW passes through the device and the polarization (ϕ) of FW is continuously rotating, the intensity of SHW generated from the FNLC ($\alpha = 0^\circ$) shows a dumbbell-shaped polar diagram (Fig. 2e). This validates the polarization-dependent nonlinear optical response of the FNLC.”
3. **Lines 251-252, page 8:** “Then, we use a circularly polarized FW for incidence to systematically investigate the nonlinear spin-orbit interaction of light occurring in the LC q -plate.”
4. **Lines 347-349, page 11:** “For a circularly polarized FW with a Gaussian type distribution $(+1, 0)_\omega$, the SHG intensity as a function of temperature is shown in Fig. 5a.”

Q2.20 Line 193-194. I’m not sure I understand the sentence. How can authors measure the vectorial polarization property of the structured second harmonic beam with an analyzer? I know this is discussed in section S3, however I think the term vectorial property is here inappropriate.

A2.20 Response:

We apologize for the confusion caused to the reviewer.

We measure the polarization distribution (vector nature) of the structured second harmonic beam by passing it through an analyzer. Extinction regions occur where the polarization is perpendicular to the analyzer. Thus, by analyzing the extinction pattern during the rotation of analyzer, we can deduce the polarization distribution of the SH light field. To make it clearer, we have detailed the measurement process and added the experiment results in Fig. 3g in the revised manuscript.

In addition, we changed the term “vectorial polarization property” to “vector nature”.

Original Text in Manuscript:

Lines 193-196, page 6: “Moreover, the vectorial polarization property of the structured SH beam is further experimentally measured by using an analyzer, and the SHWs with radial polarization distributions and singularity points are confirmed (Supplementary Section 3 and Supplementary Fig. 3).”

Revised Text in Revised Manuscript:

Lines 228-234, page 7: “Moreover, the vector nature of the SH beam for the linear polarization incidence ($\phi = 0^\circ$) is further characterized by using an analyzer behind the device. Extinction regions occur where the polarization is perpendicular to the analyzer. Thus, by analyzing the extinction pattern (the second and third panels in Fig. 3g), the SHW with a radial polarization distribution and a singularity point is confirmed. These

results agree well with the nonlinear polarization model described in the previous section.”

Q2.21 Figure 3: It is not clear to me how author determine the incident wave polarization directions in Fig. 3d and the vector distribution of the second harmonic in Fig. 3e. Also providing the intensity profile of the second harmonic might help in the understanding of the result.

A2.21 Response:

We thank the reviewer's valuable comment. Fig. 3f (Fig. 3d in the original manuscript) exhibits the polarization distribution of the vector vortex beam generated by the polar LC q -plate, which is not the incident polarization distribution. We characterize this polarization distribution with the same measurement method as the second harmonic vector beam (answered in A2.20).

To avoid confusion, we have revised the related content and added a white double-sided arrow in Fig. 3f (Fig. 3d in the original manuscript) to indicate its incident polarization. We have also detailed the characterization method for the polarization distribution of second harmonic light field and added the intensity profiles of analyzed SH light field in Fig. 3g in the revised manuscript to help understand the results. Revised Fig.3 has been provided in A2.18 for reference.

Original Text in Manuscript:

Lines 187-196, page 6: “When a linearly polarized femtosecond laser passes through the device, a vector vortex FW ($|\ell| = 2$) is generated due to the geometric phase effect²⁶ in the framework of linear optics (Fig. 3d). Under the pumping of FWs with different polarization directions, a series of structured SH beams with two-lobe intensity profiles are observed (Fig. 3e). The highest SH intensity consistently occurs in the direction which is parallel to that of pumping polarization, as observed in Fig. 2e. Moreover, the vectorial polarization property of the structured SH beam is further experimentally measured by using an analyzer, and the SHWs with radial polarization distributions and singularity points are confirmed (Supplementary Section 3 and Supplementary Fig. 3).”

Revised Text in Revised Manuscript:

Lines 223-234, page 7: “Upon a linearly polarized FW incidence, a vector vortex beam in the realm of linear optics ($|\ell| = 2$) is generated due to the imprinted linear geometric phase²⁶ (Fig. 3f). Simultaneously, an SH vector vortex beam with a two-lobe intensity profile is also observed (Fig. 3g), of which the strongest intensity occurs in the direction that is parallel to the incident polarization. When rotating the incident polarization, the SH light field follows its rotation, as shown in Figs. 3h-j. Moreover, the vector nature of the SH beam for the linear polarization incidence ($\phi = 0^\circ$) is further characterized by using an analyzer behind the device. Extinction regions occur where the polarization is perpendicular to the analyzer. Thus, by analyzing the extinction pattern (the second and third panels in Fig. 3g), the SHW with a radial polarization distribution and a singularity

point is confirmed. These results agree well with the nonlinear polarization model described in the previous section.”

Q2.22 Line 210: there are several claims in the text about the nonlinear geometric phase, but no explanations about what it is. I think authors should define it in detail, being this the main focus of the manuscript.

A2.22 Response:

Following reviewer's valuable suggestion, we have revised this sentence and added a definition of the nonlinear geometric phase.

Original Text in Manuscript:

1. **Lines 35-36, page 1:** “The second-order nonlinear susceptibility of a nanoscale LC molecule is found carrying spin-dependent nonlinear geometric phases.”
2. **Lines 209-211, page 7:** “In order to better understand the nonlinear optical SOI processes occurring in the LC q -plate, we for the first time propose the concept of nonlinear geometric phase to describe the nonlinear optical response of an FNLC molecule^{5,37}.”

Revised Text in Revised Manuscript:

1. **Lines 35-38, page 1:** “The second-order nonlinear susceptibility of a nanoscale LC director is found carrying spin-dependent nonlinear geometric phases, which describe imprinted nonlinearity phases originating from the spin coupling effect of light during the nonlinear frequency conversion process.”
2. **Lines 252-255, page 8:** “Nontrivial nonlinear geometric phases^{5,41} are disclosed to describe the nonlinear optical response of an FNLC director, of which the magnitudes are closely related to the geometric rotation of the LC director orientation and the spin states of the SHWs.”

Q2.23 Line 240: “spin-locked nonlinear geometric phases” see the previous comment.

A2.23 Response:

We thank the reviewer for the valuable suggestion.

“Spin-locked nonlinear geometric phases” refers to the two outcome nonlinearity phases of $\sigma q\varphi$ and $3\sigma q\varphi$ that are locked to specific spin states regardless of the propagation. The former nonlinear geometric phase of $\sigma q\varphi$ always corresponds to the same circular polarization as that of the incident FW; and the latter nonlinear geometric phase of $3\sigma q\varphi$ is for the opposite circular polarization compared to that of the incident FW. Accordingly, we have added related description about the spin-locked nonlinear geometric phases.

Original Text in Manuscript:

Lines 239-248, page 8: “...the second items $\sigma q\varphi$ and $3\sigma q\varphi$ come from the spin-locked nonlinear geometric phases from the FNLC molecules, respectively. Therefore, the SH

beams acquire additional orbital angular momentums of σq and $3\sigma q$, respectively. Finally, the angular momentum states of the SHWs become $(\sigma, 2\ell_0 + \sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}$.

Notably, the angular momentum states of the SHWs generated by the $\mathbf{E}_{\text{Lab}, -\sigma}^{\omega}$ component of the FWs are the same as that of the ones in Eq. (2) (details in Supplementary Section 4, Eq. (S16), Supplementary Fig. 5). It is also interesting to note that for both linear and nonlinear SOI processes, the orbital angular momentum states are spin-locked (Fig. 4b and Supplementary Fig. 5)."

Revised Text in Revised Manuscript:

Lines 284-295, pages 9-10: "The second items $\sigma q\varphi$ and $3\sigma q\varphi$ come from the nonlinear geometric phases generated from the FNLC molecules, which are locked to the co- and cross-spin states with the incident polarization, respectively. We thus call them spin-locked nonlinear geometric phases. Therefore, the SH beams acquire additional orbital angular momenta of σq and $3\sigma q$. Finally, the angular momentum states of the SHWs become $(\sigma, 2\ell_0 + \sigma q)_{2\omega}$ and $(-\sigma, 2\ell_0 + 3\sigma q)_{2\omega}$. Notably, the angular momentum states of the SHWs generated by the $\mathbf{E}_{\text{Lab}, -\sigma}^{\omega}$ component of the FW are the same as that in Eq. (3) (details in Supplementary Section 3, Eq. (S15), Supplementary Fig. 4), further demonstrating the spin locking effect. It is also interesting to note that for the linear spin-orbit interaction during the propagation of SHWs towards the exit plane, the angular momentum states are spin-locked as well (Fig. 4b and Supplementary Fig. 4)."

Q2.24 Line 250: the supporting information in section 4 does not explain the discrepancies in the power distribution of the circular components of the second harmonic waves. One can notice that supporting information in the Supplementary Section does not consider the interplay between the second harmonic generation and the coupling between the spin and the orbital angular momenta that occurs during propagation. Shouldn't result (S18) be valid for weak anisotropy i.e. quasi scalar propagation? Moreover, in S19-24 there are few things that deserve some additional consideration:

In S19, shouldn't α depend on the position of the considered infinitesimal slab?

In S20 the Jones matrix should link input and output fields of the slab

S22-23 are not clear. The process should be a distributed one.

A2.24 Response:

We sincerely thank the reviewer for the helpful comment.

We have added the explanation of discrepancies in the power distribution of the circular components of the second harmonic waves in the revised manuscript.

The interplay between the SHG and the SOI coupling have been considered in the Supplementary Section of revised supporting information (Eqs. S15 and S16). We agree with the reviewer that Eq. (S18) is valid for quasi scalar propagation. Although our

scenario satisfies quasi scalar propagation (focus lens, NA = 0.08), Eq. (S18) is not totally true. Thus, we have deleted Eq. S18 and added the calculations in the revised Supplementary Section 3.

In S19. Yes. The reviewer is right. α depends on the position of the considered infinitesimal slab. $\alpha(\varphi, z) = q\varphi + 2\pi z/p$. We use different values of α in the Jones matrixes $\mathbf{M}$ corresponding to the N^{th} layer ($N = 1, 2, 3, \dots, L/\Delta z$). To make it clearer, we have added related description that α depends on the position in the revised Supplementary Information.

In S20. Yes. The reviewer is right. Accordingly, we have revised this equation.

In S22-23. Following the reviewer's suggestion, we detailed the derivation procedures and rewritten these two equations. You are very rigorous and professional, thank you again.

Original Text in Manuscript:

Lines 248-251, page 8: “Furthermore, the intensities of the SHWs with the co- and cross-CP components are almost evenly distributed, with a ratio of approximately 46:54, which is insensitive to the thickness of the LC film (see Supplementary Section 4).”

Revised Text in Revised Manuscript:

Lines 305-309, page 10: “The intensities of the co- and cross-circular polarization components of the SHW are almost evenly distributed. The measured intensity ratio is approximately 46:54, which is very close to the theoretical prediction (see Supplementary Section 3). The minor discrepancy may be attributed to the imperfections of the polarizer and quarter-wave plate.”

Original Text in Supplementary Information:

1. **Lines 117-123, page 6:** “The transmitted SH intensity $I^{2\omega}$ is expressed as^{1,3}

$$I^{2\omega} \propto |\mathbf{P}^{2\omega}|^2 = \frac{|\chi_{\text{eff}}^{(2)}|^2 \omega^2 |\mathbf{E}^\omega|^4 L^2}{2\varepsilon_0 c^3 (n_e^\omega)^2 n_e^{2\omega}} \text{sinc}^2\left(\frac{\Delta k L}{2}\right), \quad (\text{S18})$$

where $\chi_{\text{eff}}^{(2)}$ is the effective second-order nonlinear susceptibility, $\Delta k = k^{2\omega} - 2k^\omega$ is the phase mismatch, L is the interaction length, c is the speed of light in vacuum, and n_e^ω and $n_e^{2\omega}$ are the refractive indices of the extraordinary light for the FW and SHW.

Due to the same value of $|\chi_{\text{eff}}^{(2)}|^2$ for $P_\sigma^{2\omega}$ and $P_{-\sigma}^{2\omega}$, the intensities of the co- and cross-CP SH optical vortices should be the same as each other.”

2. **Lines 139-140, page 8:**

$$“\mathbf{M}_\omega^N = e^{-ik_\omega n_{\text{eff},\omega} N \Delta z} \begin{pmatrix} \cos(N \Delta \Gamma_\omega / 2) & -i \sin(N \Delta \Gamma_\omega / 2) e^{-2i\sigma\alpha} \\ -i \sin(N \Delta \Gamma_\omega / 2) e^{2i\sigma\alpha} & \cos(N \Delta \Gamma_\omega / 2) \end{pmatrix}, \quad (\text{S19})$$

where $\alpha = q\varphi + 2\pi z/p$ represents the LC director distribution of the chiral FNLC q -plate,...”

3. **Line 144, page 8:** “ $\mathbf{E}^\omega(N\Delta z) = \mathbf{M}_\omega^N \mathbf{E}_{\text{in}}^\omega(0)$. (S20)”

4. **Line 149, page 8:** “ $\mathbf{P}_N^{2\omega}(L) = \mathbf{M}_{2\omega}^{\left(\frac{L}{\Delta z} - N\right)} \mathbf{P}^{2\omega}(N\Delta z)$. (S22)”

5. **Line 152, page 8:** “ $\mathbf{P}_{\text{sum}}^{2\omega}(L) = \sum_N \mathbf{P}_N^{2\omega}(L)$. (S23)”

Revised Text in Revised Supplementary Information:

1. **Lines 114-126, page 5:**

“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 $\chi_{\text{xxx}}^{(2)}$), 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.”

2. **Lines 157-162, page 8:**

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

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,... α in Eq. (S22) depends on the position of the considered infinitesimal slab.”

3. **Line 164, page 8:**

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

4. **Line 174, page 8:**

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

5. **Line 178, page 8:**

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

Q2.25 Line 257: The Authors should clearly explain the meaning of the half-wave condition in this context.

A2.25 Response:

We thank the reviewer for the valuable suggestion. We have added the meaning of the half-wave condition accordingly.

Original Text in Manuscript:

Lines 256-257, page 8: “The existence of the unconverted FW (+1, 0)_ω is mainly due to the LC film thickness which is not optimized to satisfy the half-wave condition²⁶.”

Revised Text in Revised Manuscript:

Lines 299-302, page 10: “The existence of the unconverted FW (+1, 0)_ω is mainly due to the LC film thickness, which is not optimized to make the phase retardation satisfy the half-wave condition $\Gamma = (2m + 1)\pi$ (m is a natural number)²⁶.”

Q2.26 Lines 281-282 and Fig. S9: Authors observe a ferroelectric nematic phase down to 40 °C in a material that is expected to be in the liquid crystal phase only until T = 80 °C. I think this observation deserves a comment. Also, with reference to Figure 5-a and Fig. S9, does the second harmonic intensity changes when heating or cooling the sample?

A2.26 Response:

We thank the reviewer for this comment. 80 °C may be a typical ferroelectric nematic-crystallization phase transition temperature for RM734. Nevertheless, this temperature can vary depending on the cooling rate, which is supported by previous research works, such as Ref. 39 and Mrukiewicz et al. *Phys. Chem. Chem. Phys.* 25, 13061 (2023). To avoid confusion, we have explained this in the revised manuscript.

The changes in second harmonic intensity when heating and cooling the sample will be different if the temperature range includes the crystallization temperature. In Supplementary Fig. 9, the differential scanning calorimetry trace in the heating (cooling) process is started from (ended at) 20 °C, which is below the crystallization temperature of 40 °C. Thus, these two differential scanning calorimetry traces exhibit difference. If the sample does not undergo crystallization, the second harmonic intensity profiles are expected to remain consistent when heating and cooling the FNLC material.

Original Text in Manuscript:

Lines 279-282, page 9: “The differential scanning calorimetry thermogram illustrates the sequential presence of different LC phases, including the isotropic phase (> 182 °C), nematic phase (128 °C ~ 182 °C), and FN phase (40 °C ~ 128 °C) (Supplementary Fig. 9).”

Revised Text in Revised Manuscript:

Lines 342-346, page 11: “The differential scanning calorimetry thermogram illustrates the sequential presence of different LC phases, including the isotropic phase ($> 182\text{ }^{\circ}\text{C}$), nematic phase ($128\text{ }^{\circ}\text{C} \sim 182\text{ }^{\circ}\text{C}$), and ferroelectric nematic phase ($40\text{ }^{\circ}\text{C} \sim 128\text{ }^{\circ}\text{C}$) (Supplementary Fig. 9). The FNLC material crystallizes around $40\text{ }^{\circ}\text{C}$, a temperature that can vary depending on the cooling rate³⁹.”

Q2.27 Lines 313-315: Authors write that doping the ferroelectric nematic with a chiral agent “drives the self-assembly of a three-dimensional space-variant chiral q -superstructure with right-handedness, as depicted in the left panel of Fig. 6a”. How do they know this is the exact pattern?

A2.27 Response:

This is a good question. It is indeed challenging to determine the exact configuration in the three-dimensional space-variant chiral q -superstructure. Nevertheless, we could reasonably infer its inherent structure in the left panel of Fig. 6a according to our experimental condition, for better understanding (see the left panel of Fig. 6a). First, the SD1 molecules in the photoalignment layer determine the orientations of FNLCs near the substrates, as shown in Fig. R1a. Next, due to the dopant of right-handed chiral agent (HTP: $\sim 12.2\text{ }\mu\text{m}^{-1}$ @ $120\text{ }^{\circ}\text{C}$, 1 wt%), we can calculate the helical pitch which is about two times of the cell cap. This means the rotation angle of the LC director is about 180° (Fig. R1b). Finally, by considering the photopatterned structure $\alpha(\varphi) = q\varphi$, we visualize the inherent spontaneous polarization distribution in the three-dimensional chiral q -superstructure (Fig. R1c).



Figure R1 | Illustration of the self-assembly of a three-dimensional right-handed q -superstructure. **a**, Spontaneous polarization near the substrates according to the photoalignment. **b**, Helical structure of the spontaneous polarization with the helical axis perpendicular to the substrates. **c**, Three-dimensional chiral LC q -superstructure.

To avoid confusion, we have changed related description in the revised manuscript as follows.

Original Text in Manuscript:

Lines 314-316 page 11: “...a three-dimensional space-variant chiral q -superstructure with right-handedness, as depicted in the left panel of Fig. 6a. The distribution of the LC director follows the equation $\alpha(\varphi, z) = q\varphi + 2\pi z/p \dots$ ”

Revised Text in Revised Manuscript:

Lines 382-385 pages 13-14: "...a three-dimensional, space-variant chiral q -superstructure with right-handedness is achieved, as illustrated in the left panel of Fig. 6a. For the sake of simplicity, one can assume that the variation of the LC director follows a linear distribution as described by the equation $\alpha(\varphi, z) = q\varphi + 2\pi z/p, \dots$ "

Q2.28 Lines 335-336: Dynamic phase should not be used in the stead of average phase factor.

A2.28 Response:

We respectfully disagree. The dynamic and geometric phases are generally treated as independent and together give rise to the total phase of a physical system [Leinonen et al. *Phys. Rev. Applied* 21, 014033 (2024)]. In Lines 335-336, we attribute two phase factors, A and B , as the dynamic phase that evolves with the propagation and geometric phase that is accrued when the system is made to trace out a (usually closed) path, respectively. Thus, for the correspondence of the geometric phase, we opt to keep the use of dynamic phase.

Q2.29 Equation (1) should be fixed since the Authors are referring to the components of the nonlinear material polarization. Same goes for eq. S(6).

A2.29 Response:

We thank the reviewer for the suggestion. We have revised Eq. (1) and Eq. (S6) to make their physical implications clearer by considering their correspondence to the spontaneous polarization of FNLC.

Original Text in Manuscript:

Lines 116-121, pages 3-4: "If the FNLC molecule is rotated by an orientation angle of α in the x - y plane. The x and y components of the second-order nonlinear polarization can be analytically expressed as

$$\begin{aligned} \mathbf{P}_x^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\alpha - \phi) \cos \alpha \hat{\mathbf{e}}_x \\ \mathbf{P}_y^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\alpha - \phi) \sin \alpha \hat{\mathbf{e}}_y \end{aligned} \quad (1)$$

where ε_0 is the vacuum permittivity, 2ω is the angular frequency of the SH wave (SHW)."

Revised Text in Revised Manuscript:

Lines 123-128 page 4: "If the polar direction of FNLC is rotated by an orientation angle of α relative to the x -axis, the second-order nonlinear polarization can be analytically expressed as

$$\begin{aligned}\mathbf{P}^{2\omega} &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\phi - \alpha) (\cos \alpha \hat{\mathbf{e}}_x + \sin \alpha \hat{\mathbf{e}}_y) \\ &= \varepsilon_0 A_0^2 \chi_{aaa}^{(2)} \cos^2(\phi - \alpha) \hat{\mathbf{e}}_a\end{aligned}\quad (1)$$

where $\hat{\mathbf{e}}_a$ represents the unit vector of the a -axis, ε_0 is the vacuum permittivity, 2ω is the angular frequency of the SH wave (SHW).”

Original Text in Supplementary Information:

Lines 49-51 page 2: “...the nonlinear polarization of the generated second harmonic wave (SHW) can be calculated as

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

Revised Text in Revised Supplementary Information:

Lines 49-52 page 2: “...the nonlinear polarization of the generated second harmonic wave (SHW) can be calculated as

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

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

Q2.30 Minor issues on terminology:

- “dimensions of light” should be reworded as “degrees of freedom of light”

A2.30 Response:

Thank you for your valuable comment. We have changed the “dimensions of light” to “degrees of freedom of light” in the revised manuscript.

Original Text in Manuscript:

Lines 27-28 page 1: “Simultaneous manipulation of multiple dimensions of light lies at the heart of photonics due to the myriad of applications it has fostered.”

Revised Text in Revised Manuscript:

Lines 27-28 page 1: “Simultaneous manipulation of multiple **degrees of freedom of light** lies at the heart of photonics due to the myriad of applications it has fostered.”

Reviewer #3

“I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.”

Response:

We greatly appreciate the respected reviewer for his or her precious time and effort in evaluating our work. We hope the new version would be found more convincing and suitable for publication now.

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

The authors thoroughly revised the manuscript and adequately addressed all the reviewer comments. I now recommend the manuscript for publication in Nature Communications.

Reviewer #2 (Remarks to the Author):

I found the manuscript largely improved. It is now clear, and both the novelty and relevance of the results can be properly appreciated. Authors fixed most of the issues I raised, there are only a few minor revisions left, that I list below.

. Abstract: I really don't like the expression "nanoscale LC director". Is it possible to change it? Maybe writing just "The second-order nonlinear susceptibility of the FNLC"?

. Lines 86-87: The polar order characterization of FNLC has been reported in several papers, besides those cited in 37-39. See for example Kumari et al., Nat Commun. 14, 748 (2023) and Marni et al., Soft Matter, 20, 4878-4885 (2024). I suggest adding at least these two references for a proper account of previous work.

. Line 324: How do authors obtain FW with those angular momentum states?

After these minor issues are addressed, I will be glad to recommend the manuscript for publication.

Reviewer #3 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Re: manuscript NCOMMS-24-20086-A

We greatly appreciate all the reviewers for accessing our manuscript of NCOMMS-24-20086-A, and providing valuable comments. All reviewers' comments and suggestions are quite helpful for us to substantially improve the quality and readability of this manuscript.

We have revised our manuscript following the reviewer 2' suggestions. Our point-by-point response to the reviewers' comments and the changes made in our revised manuscript with yellow color highlighted track change as follows. Thank you.

Response to reviewers**Reviewer #1**

The authors thoroughly revised the manuscript and adequately addressed all the reviewer comments. I now recommend the manuscript for publication in Nature Communications.

Response:

We really appreciate the reviewer's recommendation on our work.

Reviewer #2

I found the manuscript largely improved. It is now clear, and both the novelty and relevance of the results can be properly appreciated. Authors fixed most of the issues I raised, there are only a few minor revisions left, that I list below.

Response:

We greatly appreciate the reviewer for his or her valuable comments and recommendation on our work.

Q2.1 Abstract: I really don't like the expression "nanoscale LC director". Is it possible to change it? Maybe writing just "The second-order nonlinear susceptibility of the FNLC"?

A2.1 Response:

Following reviewer's suggestion, we have revised it in the abstract.

Original Text in Manuscript:

Lines 35-36, page 1: "The second-order nonlinear susceptibility of a nanoscale LC director is found carrying spin-dependent nonlinear geometric phases..."

Revised Text in Revised Manuscript:

Lines 32-34, page 1: "Here, we propose the concept of nonlinear geometric phase in an emerging ferroelectric nematic fluid, of which the second-order nonlinear susceptibility carries spin-dependent nonlinearity phases."

Q2.2 Lines 86-87: The polar order characterization of FNLC has been reported in several papers, besides those cited in 37-39. See for example Kumari et al., Nat

Commun. 14, 748 (2023) and Marni et al., Soft Matter, 20, 4878-4885 (2024). I suggest adding at least these two references for a proper account of previous work.

A2.2 Response:

We sincerely thank the reviewer for the comment. We have included these works for reference accordingly.

Original Text in Manuscript:

Lines 86-87, page 2: "...and achieving high-resolution polar ordering characterizations³⁷⁻³⁹..."

Revised Text in Revised Manuscript:

Lines 81-82, page 2: "...and achieving high-resolution polar ordering characterizations³⁷⁻⁴¹..."

Added References:

40. Marni, S. *et al.* Fluid jets and polar domains, on the relationship between electromechanical instability and topology in ferroelectric nematic liquid crystal droplets. *Soft Matter* **20**, 4878–4885 (2024).

41. Kumari, P., Basnet, B., Wang, H. & Lavrentovich, O. D. Ferroelectric nematic liquids with conics. *Nat. Commun.* **14**, 748 (2023).

Q2.3 Line 324: How do authors obtain FW with those angular momentum states?

A2.3 Response:

This is a good question. The FWs with those angular momentum states are obtained by using circularly polarized light normally impinging to two home-made nematic LC *q*-plates with the topological charges of -0.5 and $+0.5$. To make it clearer, we have added the description in this paragraph.

Added Text in Revised Manuscript:

Lines 320-322, page 10: "We obtain FWs with these angular momentum states by using home-made nematic LC *q*-plates with the topological charges of -0.5 and $+0.5$, respectively."

Reviewer #3

"I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts."

Response:

We greatly appreciate the reviewer for his or her precious time and effort in evaluating our work. Thank you.