

Phase-Transition-Induced Dynamic Surface Wrinkle Pattern on Gradient Photo-Crosslinking Liquid Crystal Elastomer

Corresponding Author: Professor Xuesong Jiang

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

[Comments]

This paper deals with the reversible deformation behavior of a gradient photo-crosslinking LCE film (AnLCE) with changing cross-link density in the direction of thickness. In particular, the use of photodimerizing An(SH)₂ is described as a novel matter. Reviewer thinks that the presented phenomena contains some interesting results, but nanoscopic higher-ordered structural analysis data containing XRD (especially in SAXS) and its explanation and consideration, description of demand properties of technology applied field is not sufficient.

[Questions]:

- (1) Actuating and the mechanical properties of a LCE film (RM257/ EDDT/ PETMP/ DPA) is described in a reference [24] in detail. The comparing results of AnE without LCE were shown in this paper, but could similar properties not be expressed by adjusting the UV-light intensity from above the film with a composition of only LCE (without An(SH)₂)?
- (2) If the property is due to photo-dimerization of anthracene, then XRD (especially in SAXS) should confirm the analysis of higher-order structure within the layer at the μm -order within the film cross-section. It would be better to examine the ordered direction of the surface and inner layers and the orientation direction before and after the dimerization of anthracene by the SAXS.
- (3) The film has a very low T_g and is very soft. Why can it be used as a display platform if it is so soft and stretchable? Isn't the display a fixed screen size and not allowed to change dimensions?

Reviewer #2

(Remarks to the Author)

In this manuscript, the authors present a novel strategy for fabricating and regulating micro-wrinkle patterns on the surface of gradient photo-crosslinking anthracene-containing liquid crystal elastomer (AnLCE) films, utilizing liquid crystal phase transition. The attractive multi-mode surface, which incorporates light scattering, structural color, fluorescence and birefringence patterns, can be simultaneously controlled by the thermal-induced erasure and mechanical recovery of surface micro wrinkles with high reversibility, achieving their functions in soft optics, information storage and anti-counterfeiting. Overall, this is an outstanding piece of work. I recommend accepting this manuscript after addressing the following suggestions/comments:

1. In comparison with the existing literature on the fabrication and regulation of wrinkled patterns via LCEs (e.g. Soft Matter 8, 7138; Soft Matter 10, 1411; Adv. Funct. Mater. 25, 1360; J. Mater. Res. 33, 4092; Adv. Mater. 35, 2206764), what are the principal advantages presented in this manuscript?
2. The polarized images in wrinkle-patterned AnLCE films were interesting. How can we understand the regulation of polarized light via surface wrinkling more deeply?
3. The wrinkles generated on the surface of a single-layer elastomer film are very small in size (with a wavelength of about 1 μm), how can this be explained?
4. In Fig. 2b, the annotation "Recovered" was incorrect. Should it be "Wrinkled"? There is a typo error "UV the intensity" in line 143.
5. In Fig. 3e, the title of the abscissa axis, "Erased-Recover Cycles", should be changed to "Erasure-Recovery" to correspond with the main text,

Reviewer #3

(Remarks to the Author)

This work describes a switchable wrinkling layer based on an liquid crystal elastomer. While there are some new elements, I have seen much of this work before and I feel overall it misses the novelty that might be expected for publication in this journal, and may be better suited for a more directed journal related to optics or materials. Because of similarity to many other works, I think the ultimate impact will be very limited, although the addition of the emitting molecule seems new.

Some general comments:

There are a lot of language errors, too many for me to list completely here. I will confine it to a few examples:

-pg 3, ln43: delete word 'while'

-pg 3, ln61: 'mult-imode'

-p5, ln95: "keep impervious" is awkward

-p11, ln216: 'shorter' wavelengths

-p11, ln225: "and then decreased 225 slightly to 0.34 μm in final" needs rewriting

-p11, ln 226: "In another word,..." should be "In other words..."

-p11, ln 227: 'increasing' should be 'increased'

-p11, ln 235: 'were corresponding' should be 'corresponded' or 'correspond'

-p13, ln280: 'this suppose' should be 'this supposition'

-p21, ln 437: 'operat'

*p11: "The residual strain may be owing to the limited mobility of liquid crystal domains

caused by high-density crosslinking points, making it difficult to keep up with the rapid changes in polymer chain segments during the stretching recovery cycle." I don't really follow. Over a longer time period, should this strain be relieved?

*<https://doi.org/10.1016/j.matt.2022.08.019> performed very similar effects although they looked at physical rather than optical effects, but deserve mention. Another relevant work: <https://doi.org/10.1016/j.ijssolstr.2017.10.018>

*P3, ln49: In what way is the LC phase transition 'extraordinary'?

*p3, ln61: "dynamic tunability with better flexibility and durability..." Better than what?

*Ref 26 essentially is what is being demonstrated in this work?

*p10, ln200 and 206: The work described in <https://doi.org/10.1021/acs.langmuir.8b01934> showed local application of UV light through masks to generate patterns invisible to the eye until exposed to heat, and likely should be referenced.

*The persistence of the wrinkles was studied after 15 days at room temp. How about at higher temps that are still environmentally relevant, say 40 or 50 degrees?

*p12, ln 252: "a viewing enhancement of surface information by varying the viewing angle" What enhancement?

*I do not understand what is driving the reformation of the identical wrinkling pattern of the sample after heating to erase the structure. How many repeats and what loss of fidelity? Figure 3e is very difficult to read.

*p15, ln310: The claim of 'long term stability' has not undergone any real rigorous test. Start with elevated temperatures.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

[2nd Comments]:

I accept the answers 1 and 3 as stated in the author's response. However, an answer 3 should be clearly limited to "soft" displays. This is because this technology can only be used in fields where soft materials are acceptable.

Unfortunately, I cannot accept an answer 2 regarding SAXS. In the structural drawings of Figures 1 and 4, authors show a polydomain and a nematic-like monodomain in the surface and inner layers. If the higher-order structure is formed as shown in the structural drawings, some SAXS peak should be shown around at $d = 2$ to 4 nm. Broad azimuthal data alone, as shown in Supplementary Figure 26, cannot be discussed without data on the 2θ or q spectra. Also, there is no description of the measurement conditions for the additional data, which is inappropriate for a technical paper. So reviewer think that it is necessary to add both out-of-plane and in-plane film data to show the validity of the structural drawings as shown in Figures 1 and 4. If XRD data cannot explain the structure, then the polydomain liquid crystal-like structure is no longer the basis for the functional expression of this Liquid Crystal Elastomer, it is simply the aggregation and stretching phenomena of the crosslinking chains with a difference in linear expansion coefficients.

Reviewer #2

(Remarks to the Author)

The authors have adequately addressed my questions and comments. I recommend accepting this manuscript.

Reviewer #3

(Remarks to the Author)

The comments of the other reviewers and the author have assuaged my concerns of novelty, and I am happy with the author's replies. I believe the article is suitable for publication.

I note the following textual errors:

*p11, ln 241: Should be "These results corresponded to..." Delete 'were'

*p12, ln 249: should be 'evolution of wrinkles...'

*p12, ln 258: should be 'short wavelength' rather than 'small'. Wavelength is a length, thus, shorter and longer.

Version 2:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

[3rd Comments]:

Reviewer accepts the authors' 3rd revised responses. I also thank the authors for the additional SAXS measurements. I supposed that some smectic structure might present in this LCE, but understood that only nematic structures of low order structure existed. Therefore, it should be described as "nematic polydomain" so that it is not misunderstood as "smectic polydomain" in illustrations in the paper.

[Comments on SAXS data analysis] Reviewer considered the following relationship between SAXS and higher-order structure of this material.

In Fig. R1, (2) under 200% elongation, the SAXS scattering is isotropic and unrelated to the nematic monodomain structure. However, (3) when the film is restored to its original length after releasing, a tail of diffraction scattering related to the long-periodic structure is observed in the stretching direction. This suggests that this SAXS result is a scattering tail due to the physical shape of the sheet wrinkle structure rather than to the higher-ordered structure of the LCE.

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Response to Reviewers:

Reviewer #1 (Remarks to Author): *This paper deals with the reversible deformation behavior of a gradient photo-crosslinking LCE film (AnLCE) with changing cross-link density in the direction of thickness. In particular, the use of photodimerizing An(SH)₂ is described as a novel matter. Reviewer thinks that the presented phenomena contains some interesting results, but nanoscopic higher-ordered structural analysis data containing XRD (especially in SAXS) and its explanation and consideration, description of demand properties of technology applied field is not sufficient.*

Response: We thank the referee for the very insightful and precious comments. According to the suggestion, we have performed additional experiments, including synchrotron SAXS and added corresponding discussions into the manuscript.

Comment 1: *Actuating and the mechanical properties of a LCE film (RM257/ EDDET/ PETMP/ DPA) is described in a reference [24] in detail. The comparing results of AnE without LCE were shown in this paper, but could similar properties not be expressed by adjusting the UV-light intensity from above the film with a composition of only LCE (without An(SH)₂)?*

Response to 1: .

We have firstly designed a normal LCE film without An(SH)₂ by one-step thiol-acrylate Michael addition polymerization. The LCE film was prepared by RM257/ EDDET/ PETMP/ DPA (without photoinitiator). No wrinkle formed on the surface after UV irradiation and subsequent stretching-releasing because no gradient crosslinking formed on the surface to create the mismatch of mechanical properties between the top layer and the inner layer.

To further illustrate the issue, we designed a LCE film composed of RM257/ EDDET/ PETMP/ DPA/ **HHMP** ([2-hydroxyethoxy)-2-methylpropiophenone]) (see detail in *RSC Adv.* **2015**, 5, 18997.). HHMP is a photoinitiator for the secondary crosslinking to fix the shape and LC orientation of the stretching LCE film. Upon low

UV intensity (5 mW/cm^2), no wrinkle pattern could be formed on the surface of normal LCE film via either UV-SR or S-UV-R.

The possible reason is that UV-triggered crosslinking via the initiation of HHMP is a typical radical polymerization of excess acrylate groups. The character of chain-growth reaction made it difficult to control the thickness of crosslinking and generate a gradient layer system. On the contrary, in AnLCE system, it is easy to confine the photo-crosslinking to the thin surface layer because of the high UV absorbance and high content of anthracene, and the character of bimolecular reaction.

Comment 2: *If the property is due to photo-dimerization of anthracene, then XRD (especially in SAXS) should confirm the analysis of higher-order structure within the layer at the μm -order within the film cross-section. It would be better to examine the ordered direction of the surface and inner layers and the orientation direction before and after the dimerization of anthracene by the SAXS.*

Response to 2:

We conducted SAXS experiments to study the influences of anthracene's dimerization. Since the crosslinking top layer is very thin and difficult to be separated from the surface-UV-crosslinked AnLCE film, we prepared a thin film fully exposed to UV, and another untreated AnLCE film to demonstrate the top layer and inner, respectively. The UV-crosslinked film was acquired before stretching and after releasing. The untreated AnLCE film simulating the uncrosslinked inner layer was subjected to the same test.

As shown in **Fig. R1a i), ii)**, the SAXS results showed that the untreated AnLCE films did not exhibit obvious orientation before and after stretching-releasing, the non-oriented state was further confirmed by scattering intensity-azimuthal angle curves (**Fig. R1b**). The UV-crosslinked film showed evident orientation after stretching-releasing (**Fig. R1a iii, iv and Fig. R1c**).

According to the fitting Gaussian curve of the 1D curve of scattering intensity versus azimuthal angle of UV-crosslinked AnLCE film after stretching-releasing (**Fig. R1c**), the degree of orientation (Π) was calculated as 41.4% by the following equation.

The full width at half maximum of the peak curve (FWHM = 105) was derived from the fitted Gaussian curve.

$$\Pi = \frac{180 - FWHM}{180}$$

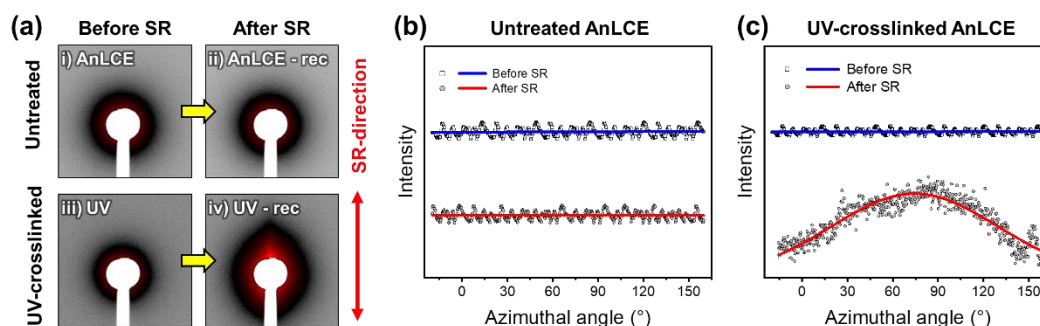


Fig. R1. The SAXS results of UV-crosslinked/untreated AnLCE films before and after stretching-releasing. **a** 2D-SAXS spectrum of i) untreated AnLCE, AnLCE film after stretching-releasing, UV-crosslinked film and iv) UV-crosslinked AnLCE film after stretching-releasing. **b** The 1D curve of scattering intensity versus azimuthal angle of the untreated AnLCE film before (blue) and after (red) stretching-releasing. **c** The 1D curve of scattering intensity versus azimuthal angle of the UV-crosslinked AnLCE film before (blue) and after (red) stretching-releasing.

The SAXS results have been added to Supplementary Information (Supplementary Fig. 26), and the relative discussion have been added to the part of mechanism study in the manuscript (Page 18, Line 372)

Comment 3: *The film has a very low Tg and is very soft. Why can it be used as a display platform if it is so soft and stretchable? Isn't the display a fixed screen size and not allowed to change dimensions?*

Response to 3:

A significant portion of flexible displays or sensors require soft materials as substrate to achieve large deformation such as stretching and bending (*J Electron Packaging* **2016**, 138, 020801.; *Light. Sci. Appl.* **2018**, 7, 35.). In addition, some of the stimulus-responsive materials used for anti-counterfeiting and information storage were

also applied in the form of applied strains. (*Adv. Funct. Mater.* **2023**, 33, 2305364.; *Adv. Mater.* **2024**, 2314201.)

In our manuscript, the AnLCE films was applied as a soft display platform. Information was write-in AnLCE films by photomasked UV exposure. By regulating the surface wrinkling via mechanical strain and heating, the encoded information could be on-demand decrypted and encrypted. During these process, AnLCE films were allowed to be suffering from large deformation and heating to controlled the surface micro wrinkles. With high usability and stability, AnLCE film is expected to be used for information encryption, anti-counterfeiting labels, and soft sensors.

Reviewer #2 (Remarks to the Author): *In this manuscript, the authors present a novel strategy for fabricating and regulating micro-wrinkle patterns on the surface of gradient photo-crosslinking anthracene-containing liquid crystal elastomer (AnLCE) films, utilizing liquid crystal phase transition. The attractive multi-mode surface, which incorporates light scattering, structural color, fluorescence and birefringence patterns, can be simultaneously controlled by the thermal-induced erasure and mechanical recovery of surface micro wrinkles with high reversibility, achieving their functions in soft optics, information storage and anti-counterfeiting. Overall, this is an outstanding piece of work. I recommend accepting this manuscript after addressing the following suggestions/comments:*

Response: The authors thank the referee for these positive comments. The manuscript has been carefully revised according to the suggestions.

Comment 1: *In comparison with the existing literature on the fabrication and regulation of wrinkled patterns via LCEs (e.g. *Soft Matter* 8, 7138; *Soft Matter* 10, 1411; *Adv. Funct. Mater.* 25, 1360; *J. Mater. Res.* 33, 4092; *Adv. Mater.* 35, 2206764), what are the principal advantages presented in this manuscript?*

Response to 1:

Thanks for your insightful comment. The recommended literatures are all pertinent to the wrinkle patterns based on liquid crystal elastomers, such as the reversible thermal-deformable substrate for dynamic wrinkle patterns, localized wrinkling via photo-oriented or mechanical-rubbing LCE layers, and the combination of chiral LCE and wrinkle surface. However, the following problems remain in these existing literatures: wrinkles fabricated by film transfer were not easy to localize wrinkles, making them difficult to use as information displays (*Soft Matter* **2012**, 8, 7138; *Soft Matter* **2014**, 10, 1411; *Adv. Mater.* **2023**, 35, 2206764); while the methods for localized wrinkling on the LCE surfaces exhibited a lack of regulation following the formation of wrinkles, thereby limiting the tunability of the surface information (*Adv. Funct. Mater.* **2014**, 25, 1360; *J. Mater. Res.* **2018**, 33, 4092).

In this manuscript, we present a strategy for the fabrication and regulation of micro wrinkles on the surface of a single-layer and freestanding liquid crystal elastomer film. The localized wrinkling and for surface image display was achieved by direct UV irradiation of the AnLCE surface with photomasks, and the phase-transition-induced surface dynamic wrinkle enabled the switchable information on the surface.

In comparison with these recommended literatures, this strategy featured facile processing, localized patterning, flexible regulation, and multi-mode display, especially the regulation of polarized light by surface wrinkling was expected to be used as soft sensors and displays.

Comment 2: *The polarized images in wrinkle-patterned AnLCE films were interesting. How can we understand the regulation of polarized light via surface wrinkling more deeply?*

Response to 2:

The key factor for manipulating surface polarized light is to controlled the macroscopic orientation of the films. In this manuscript, we presented the regulation of polarized light via surface wrinkling. When the AnLCE films were prepared, it exhibited a polydomain nematic phase with microscopic orientation but macroscopic disorder so that the films showed bright birefringence at all angles relative to the crossed polarizer. In order to change the orientation of a specific area on the AnLCE's surface, we utilized the high spatio-temporal resolution and the characteristic of photo-crosslinking to construct the photo-chemical boundary: the photomasked UV exposure enabled the localized wrinkling. After stretching and releasing, 1D-ordered wrinkles formed in the exposed area, the mesogens reserved their orientation parallel to the wrinkle wave, while the unexposed areas kept flat and return to the polydomain state. It generated a surface with 2 orientation states: the polydomain phase in the unexposed area (flat) and monodomain phase area (wrinkled). The difference between the wrinkled and flat areas can be observed by rotating the sample: the wrinkle area altered the brightness between bright and dark, while the non-wrinkle area exhibited constantly bright.

Comment 3: *The wrinkles generated on the surface of a single-layer elastomer film are very small in size (with a wavelength of about 1 μm), how can this be explained?*

Response to 3:

We obtained wrinkles in an average wavelength of 1 μm and amplitude of 0.4 μm in this study. This micron-submicron size is not easy to be achieved by general polymeric film unless by metal coating or plasma treated PDMS elastomer surface (*Adv. Sci.* **2023**, 10, 2207210.). The extreme small-size should be caused by the intrinsic hindrance of liquid crystal groups, which strongly inhibited intermolecular photodimerization of anthracene, and consequently resulted ultrathin photo-crosslinked layer and small modulus variation. The UV crosslinking on surface was verified by cross-section AFM. The thickness of crosslinked top layer and gradient layer were both below 1 μm , giving rise to small-sized wrinkles reasonably.

Comment 4: *In Fig. 2b, the annotation “Recovered” was incorrect. Should it be “Wrinkled”? There is a typo error “UV the intensity” in line 143.*

Response to 4:

We have revised the manuscript and corrected the error in Fig. 2b and its annotation (Page 8, Line 166)

Comment 5: *In Fig. 3e, the title of the abscissa axis, “Erased-Recover Cycles”, should be changed to “Erasure-Recovery” to correspond with the main text.*

Response to 5:

We have revised the manuscript and modified the description for Fig. 3e.

Reviewer #3 (Remarks to the Author): *This work describes a switchable wrinkling layer based on a liquid crystal elastomer. While there are some new elements, I have seen much of this work before and I feel overall it misses the novelty that might be expected for publication in this journal, and may be better suited for a more directed journal related to optics or materials. Because of similarity to many other works, I think the ultimate impact will be very limited, although the addition of the emitting molecule seems new.*

Response: The authors thank the referee for the insightful comments.

Liquid crystal elastomers (LCEs), with diversity of deformation abilities based on their phase transition, have shown a potential to fabricate and regulate dynamic surface, especially surface wrinkle with tunable morphology in response to external stimuli, as well as their application in actuators. Existing studies on “LCE + wrinkle” systems are mainly based on “Film/LCP” bilayer systems, and they did achieve the formation of wrinkle patterns by liquid crystal phase transition, but these wrinkle patterns are still limited in terms of both difficulties in preparation and regulation. For instance, the film/LCE bilayer system prepared by transferring a hard film on an LCE substrate with uniaxial alignment, which reversibly converts the wrinkle-planar state but it is difficult to achieve localized wrinkling, and there is only one steady state of either wrinkle or planar state at room temperature (*Soft Matter* **2012**, *8*, 7138; *Soft Matter* **2014**, *10*, 1411). In some film/LCP systems using the photo-aligned substrate, although the localization of wrinkles has been achieved, the further regulation after wrinkling has not been reported in detail (*Adv. Funct. Mater.* **2014**, *25*, 1360; *Langmuir* **2018**, *34*, 10543). Moreover, problems such as cracking and delamination of the top surface still limit the application of these bilayer wrinkle systems. Therefore, it is highly desirable to produce a wrinkle on a freestanding film with the characteristics of good tunability, and localized patterning.

In this work, we provided a new method for fabricating and dynamically regulating surface wrinkles via liquid crystal phase transition on an anthracene-containing liquid crystal elastomer (AnLCE) film. Switchable surface wrinkle in response to thermal-

mechanical stimuli on a single-layer AnLCE film and light-induced localized patterning was achieved in our work. The combination of anthracene (An) and LCE enabled the on/off switchable and multi-mode surface pattern, which was on-demand controlled by the generation and erasure of surface micro wrinkles. In this AnLCE system, the anthracene moiety not only serves as the fluorescence-emission unit, but also plays a key role in formation of a gradient crosslinking layer under the irradiation of 365 nm UV light. After UV exposure, 1D oriented wrinkles are generated via mechanical-induced surface instability, resulting from the mechanical mismatch of surface UV-crosslinked layer and the inner layer. The AnLCE film is expected to be used for information encryption, anti-counterfeiting labels, and soft sensors.

Comment 1: *There are a lot of language errors, too many for me to list completely here. I will confine it to a few examples:*

-pg 3, ln43: delete word ‘while’

-pg 3, ln61: ‘mult-imode’

-p5, ln95: “keep impervious” is awkward

-p11, ln216: ‘shorter’ wavelengths

-p11, ln225: “and then decreased 225 slightly to 0.34 μm in final” needs rewriting

-p11, ln 226: “In another word,..” should be “In other words...”

-p11, ln 227: ‘increasing’ should be ‘increased’

-p11, ln 235: ‘were corresponding’ should be ‘corresponded’ or ‘correspond’

-p13, ln280: ‘this suppose’ should be ‘this supposition’

-p21, ln 437: ‘operat’

Response to 1: The authors thank the referee for pointing out the errors. We have corrected them according to the comments and checked the manuscript carefully.

Comment 2: *p11: “The residual strain may be owing to the limited mobility of liquid crystal domains caused by high-density crosslinking points, making it difficult to keep up with the rapid changes in polymer chain segments during the stretching recovery cycle.” I don’t really follow. Over a longer time period, should this strain be relieved?*

Response to 2:

To verify the length contraction during storage, we prepared a UV-crosslinked strip with approx. 30% residual strain after releasing from 100% stretching. The strip was stored at room temperature (25 °C), and its length was measured once a day for 7 days. The length did not show observable recovery (**Fig. R2**). The stability of the UV-crosslinked film could also demonstrate the sustaining of the surface wrinkles.

The reason of the “locking” strain may be the restriction of inner stress relaxation in the crosslinked film. Although the process should be automatically run (finally lead to the wrinkle erase and length retraction), there should be a barrier to inhibit the relaxation rate that we could not observe it for a limited period. The data obtained from the dynamic thermomechanical analysis (DMA) indicated a continued decrease in the storage modulus of the AnLCE films as the temperature increased. This can be considered leading to an acceleration of stress relaxation. Therefore, we supposed that the high-density crosslinking restricted the mobility of the mesogens to return to the polydomain phase as the substrate retracted.

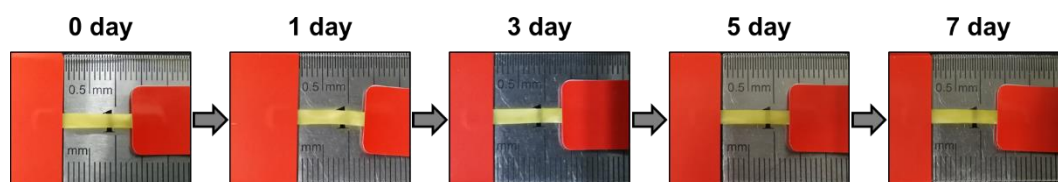


Fig. R2. Photograph of UV-crosslinked AnLCE film during 7-day storage at room temperature.

Comment 3: <https://doi.org/10.1016/j.matt.2022.08.019> performed very similar effects although they looked at physical rather than optical effects, but deserve mention. Another relevant work: <https://doi.org/10.1016/j.ijsostr.2017.10.018>

Response to 3:

We have added the citation of the recommended literatures according to the referee’s suggestion. (Page 3, Line 55 and Page 10, Line 207)

Li et.al. demonstrated the programming 3D shape by harnessing the molecular-material-structural interactions of liquid crystal elastomers to create diverse stiffness-

heterogeneous, reversibly morphing structures via locally controlled photopolymerization and mechanical training (*Matter* **2022**, 5, 4332). By utilizing this method, it is expected to realize complex deformation of LCEs. However, what we demonstrate is the photo-induced localized surface patterning, rather than localized deformation.

Fu et.al. demonstrated a theoretical and numerical study on the surface wrinkling of a bilayer LCE/elastomer system (*Int J Solids Struct*, **2018**, 132-133, 264). The initial LC was vertically aligned to the film surface. And the surface wrinkle was generated via the UV-induced isomerization of azobenzene molecule in the LC layer. Localized surface wrinkling was investigated by masked UV exposure. However, this work focused on theoretical and numerical study did not show additional experimental data to fit the calculation results. In contrast, we demonstrated a new strategy to regulate surface wrinkles via liquid crystal phase transition and realized the regulation multi-mode information on the surface.

Comment 4: *P3, ln49: In what way is the LC phase transition 'extraordinary'?*

Response to 4:

In this manuscript, we describe that LCEs have a unique phase transition behavior. They can undergo reversible deformation or optical patterning by phase transition. The phase transition induced actuations of LCEs have been widely explored (*Adv. Mater.* **2023**, 35, 2209566; *Adv. Funct. Mater.* **2023**, 33, 2304769). The transition among different LC phases also give rise to the change of optical or polarized optical information (*Adv. Optical Mater.* **2020**, 8, 1902098; *Nat. Mater.* **2024**, 23, 834).

In our study, the LC phase transition was utilized to fabricating and regulating the micro wrinkles on the LCE surface. The surface information was simultaneously controlled by the wrinkle evolution. Therefore, the LC phase transition was considered as one of the most important factors in this study. In order to make it more rigorous in its presentation, we have modified the description to “**unique**” (Page 3, Line 53).

Comment 5: *p3, ln61: “dynamic tunability with better flexibility and durability...”*
Better than what?

Response to 5:

The term “better” is used in contrast to the bilayer wrinkle system based on above liquid crystal polymers. In general, bilayer wrinkle systems usually consist of rigid top layer (high modulus metals or rigid polymers) and soft substrate (low modulus LCEs). When the bilayer systems suffer from external stimuli, wrinkles form on the surface due to the differences of mechanical properties between top layer and substrate. However, in some extreme condition, such as large deformation, delamination or cracking of top layer usually occurred and leads to bilayer structural failure. Whereas there is no obvious interface in the single or gradient systems. It allows them to maintain structural stability under complex mechanical or thermal condition. The description has been modified the description as “dynamic tunability with better flexibility and durability **in comparison to a traditional laminated wrinkle system**” (Page 3, Line 66).

Comment 6: *Ref 26 essentially is what is being demonstrated in this work?*

Response to 6:

Although both the existing literature and our own work demonstrate the fabrication and the regulation of surface wrinkles on a monolayer LCE, there remain several significant differences.

In the recommended literature, authors demonstrated a method for fabricating 2D disordered wrinkle on the LCE surface. A gradient crosslinking with a low-modulus top layer was prepared by surface oxygen inhibition during photo-polymerization. Wrinkles were generated on the LCE surface via subsequent solvent evaporation. The resulting wrinkle pattern can be modulated by thermal phase change and mechanical stretching, but wrinkles recovered automatically after withdrawal of these stimuli. It means there is only one steady state of wrinkles. Moreover, the wrinkle size and wrinkling area is hard to be precisely controlled due to the characteristic of solvent evaporation. Therefore, the information storage can only be achieved by combining multiple samples

or stacking them in a laminated system. To some extent, this limits the applicability of this study to information display.

In our study, we demonstrate a method for fabricating 1D ordered wrinkles on the LCE surface. A gradient crosslinking LCE film with a high-modulus top layer was prepared by surface crosslinking via UV-dimerization of anthracene. Wrinkles were generated on the LCE surface via subsequent mechanical strain. The wrinkles' dimension and orientation could be controlled by the UV exposure and stretching parameter. The flat/wrinkle states were dynamic regulated via heating and mechanical strain. By utilizing the high spatio-temporal resolution of light and the dynamic properties of surface wrinkles, localized patterning via masked UV exposure enabled the one-step encoding of information on a single LCE film and the on-off switchable surface pattern enabled the decryption/encryption of the encoded multi-mode information.

We have summarized the principal difference between literature and manuscript work in **Table R1**:

Table R1. Principal difference between the provided reference and this work

	[Reference]	This work
Structure of elastomer film	Gradient crosslinking with low-modulus top wrinkle layer (surface oxygen inhibition)	Gradient crosslinking with high-modulus top wrinkle layer (surface photo-dimerization)
Wrinkling mechanism and wrinkle structure	solvent evaporation, 2D disordered wrinkle	Mechanical strain, 1D ordered wrinkle
Wrinkling control	Not reported	Size control by changing UV and stretching strain; orientation controlled by stretching direction. Localized patterning via masks
Methods for information display	<ul style="list-style-type: none"> ● Arrangement of samples: Multi-level display through different degrees of heating and stretching ● Stacking layered system combining with additional photo-thermal layer: direct laser writing and self-erasure 	Masked UV exposure encoding, mechanical-induced decryption and thermal-induced encryption on a freestanding LCE film.

Information display mode(s)	<ul style="list-style-type: none"> ● Transparency of films 	<ul style="list-style-type: none"> ● Transparency of films ● Structural color ● Emission of fluorescence, ● Polarized images
Potential impacts	<ul style="list-style-type: none"> ● Fabrication and regulation of wrinkles on LCE films. (both) ● Dual-state of surface under room temperature (in this work) ● Regulating polarized light by surface wrinkling and multi-mode information display on a single film (in this work) 	

[Reference]: *Angew. Chem. Int. Ed.* **2023**, 62, 202313728. (as [Ref. 27] in the revised manuscript.)

Comment 7: *p10, ln200 and 206: The work described in <https://doi.org/10.1021/acs.langmuir.8b01934> showed local application of UV light through masks to generate patterns invisible to the eye until exposed to heat, and likely should be referenced.*

Response to 7:

In this recommended literature, a method for localized wrinkling based on photo-aligned LC substrate was described. However, the principal difference of recommended literature and our work is the method for generating and regulating the wrinkle patterns: in the literature's description, the wrinkles formed when exposed to heat; in our work, the wrinkles formed by mechanical stimulation and tuned by temperature. We have cited this recommended literature and its related work (*Adv. Funct. Mater.* **2014**, 25, 1360) in a specific location. (Page 10, Line 207)

Comment 8: *The persistence of the wrinkles was studied after 15 days at room temp. How about at higher temps that are still environmentally relevant, say 40 or 50 degrees?*

Response to 8:

In our study, the fabricated wrinkle pattern exhibited acceptable stability to maintain its microstructure at room temperature. The wrinkle was erased while suffering from heating. During this continuous heating process, surface morphologies of wrinkled film were acquired after heating to defined temperature for 2 min.

According to the suggestion, we carried out the stability tests on the wrinkle pattern of the temperature at 40 and 50 °C. As shown in **Fig. R3**, we observed the surface morphology of an AnLCE film with alternate wrinkle-flat areas (the black area was wrinkled, while the white area was flat). When the film was heated to 40 °C and kept for 12 h, the wrinkled area kept black. It confirmed the stability of wrinkle pattern upon slightly heating. While heating up to 50 °C, the wrinkles were stable in the first 4 hours. After that, the wrinkles slightly faded with the decrease in amplitude (about 50% decrease). It further supported the acceleration of stress relaxation by increasing temperature.

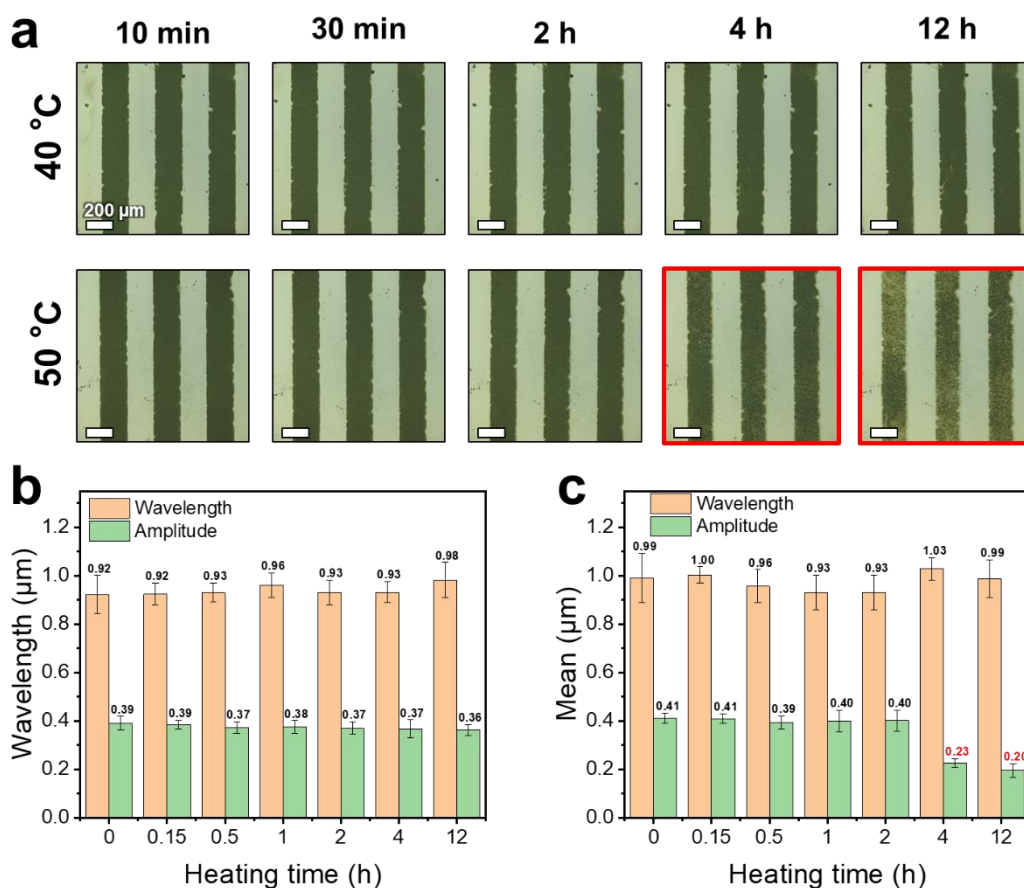


Fig. R3. Stability test of surface wrinkles upon heating. **a** Optical images of wrinkle surface under 40 °C and 50 °C heating. Scale bar: 200 μm. **b** and **c** The statistical data of characteristic wavelength and amplitude of surface wrinkles under 40 °C (**b**) and 50 °C (**c**) heating. Results are shown as mean ± SD, n = 6.

Comment 9: *p12, ln 252: “a viewing enhancement of surface information by varying the viewing angle” What enhancement?*

Response to 9:

“Enhancement” comes from the display of structural colors in natural light, which provides an additional display mode for surface information. When a wrinkle pattern is formed on the surface of the AnLCE, a strong contrast between the macroscopically wrinkled and flat areas is created due to the scattering of the wrinkle morphology on the light, whereas the flat area region presents transmission or specular reflection. At the same time, since the wavelength of the wrinkles is small ($\sim 1 \mu\text{m}$), the structural colors generated by the 1D oriented micro-wrinkles can be observed when varying the observing angle, thus enhancing the readability of the encoded information, which is shown in **Fig. R4**.

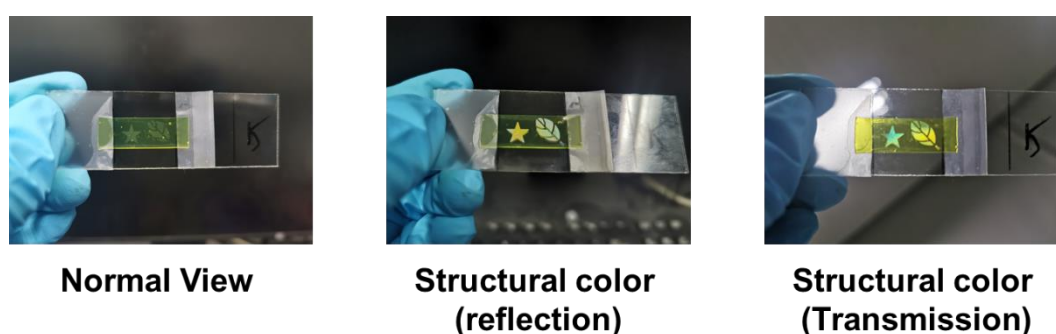


Fig. R4. Photograph illustration of structural-color-enhanced information readability.

Comment 10: *I do not understand what is driving the reformation of the identical wrinkling pattern of the sample after heating to erase the structure. How many repeats and what loss of fidelity? Figure 3e is very difficult to read.*

Response to 10:

The “identical” should be attribute to the similarity of sinusoid wrinkle waves on the surface of AnLCE films. As shown in **Fig. R5** and **Fig. R6**, in the erasure-recovery of wrinkles, the wavelength remains constant as the amplitude of the wrinkles decreases with heating, and the film is considered to be “erased”. After subsequent stretching-

releasing, the wrinkle reformed, and its amplitude is recovered to the value before erasure.

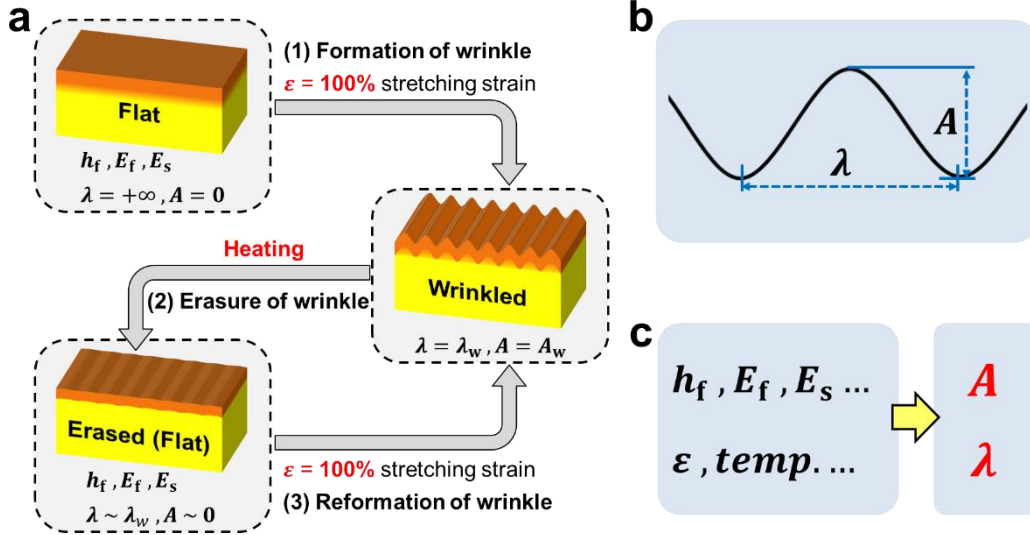


Fig. R5. Schematic illustration of the fabricating and regulating of surface wrinkles. **a** Schematic illustration of the formation, erasure and reformation of surface wrinkles. **b** Characteristic dimension: wavelength (λ) and amplitude (A) of wrinkles. **c** Principal parameters for determining the wrinkles' dimension.

The resulting wrinkle dimension (amplitude and wavelength) was determined by the characteristics of the wrinkle system (including the thickness of the rigid top layer, mechanical properties of the top layer and the inner layer) and the external stimuli (applied strain, temperature, et.al.) (*Adv. Sci.* **2023**, 10, 2207210). These are described in **Fig. R5** and the following equations:

$$\lambda = 2\pi h_f \left(\frac{E_{\text{eff}}}{3\bar{E}_s} \right)^{1/3}, E_{\text{eff}} = \bar{E}_f + \frac{\bar{E}_s - \bar{E}_f}{4(h_f/h_g + 1)^3} \quad (1)$$

$$A = (h_f + h_g) \left(\frac{\varepsilon_0}{\varepsilon_c} - 1 \right)^{1/3}, \varepsilon_c = -\frac{1}{4} \left(\frac{3\bar{E}_s}{E_{\text{eff}}} \right)^{2/3} \quad (2)$$

Where the subscript f, g and s represent the top layer toward UV irradiation, gradient crosslinking layer and substrate backward UV irradiation, respectively. While

h_f and h_g represents the thickness of top layer and gradient layer. $\bar{E}_f = E_f/(1 - \nu_f^2)$ and $\bar{E}_s = E_s/(1 - \nu_s^2)$ are plane strain modulus of the top layer and substrate. The ε_0 and ε_c represent compressive strain and critical threshold strain, respectively.

In our study, the processing parameters for wrinkle preparation were determined as 365 nm UV exposure time of 600s at 35 mW/cm² and stretching ratio of 100%. The UV exposure determined the thickness and the modulus of top layer (h_f , h_g , \bar{E}_f , \bar{E}_s), while the stretching ratio determined the applied strain (ε_0). Therefore, the wrinkle size, both amplitude and wavelength were determined in the initial wrinkle fabrication.

After heating and cooling to room temperature, wrinkle pattern on the surface was erased, but the principal properties of the gradient-crosslinked AnLCE film, especially the crosslinking density of top layer, were not changed, because the phase-transition-induced wrinkle erasure was a physical process. The surface after erasure could be considered as same to the initially flat state. The reformation of wrinkles with similar dimensions (average wavelength and amplitude) was finally achieved following the application of stretching-releasing cycles in the same processing parameter (100% stretching strain) to the erased film (**Fig. R6a**).

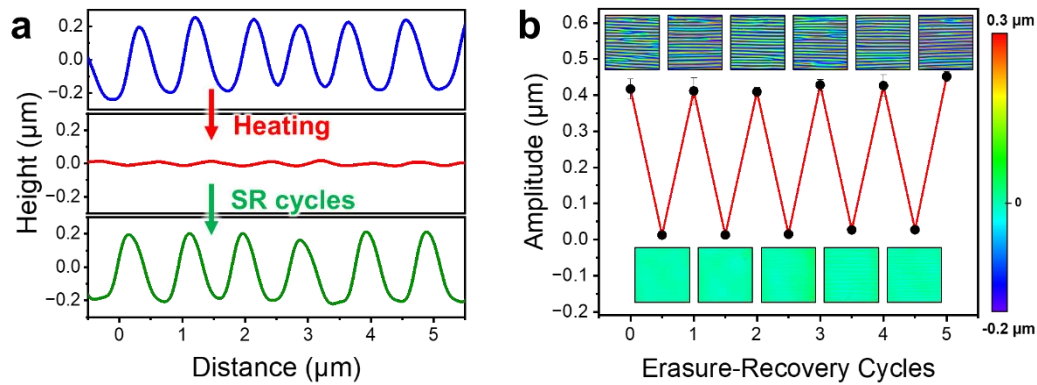


Fig. R6. The erasure and recovery of wrinkles. **a** Cross-section profiles of surface wrinkles before erasure (blue line), after thermal erasure (red line), and after recovery (green line). **b** Variation of amplitude values under erasure-recovery cycles. The insets are the 2D pattern of the surface morphology. These 2 figures are the updated version of Fig. 3d and 3e in the revised manuscript.

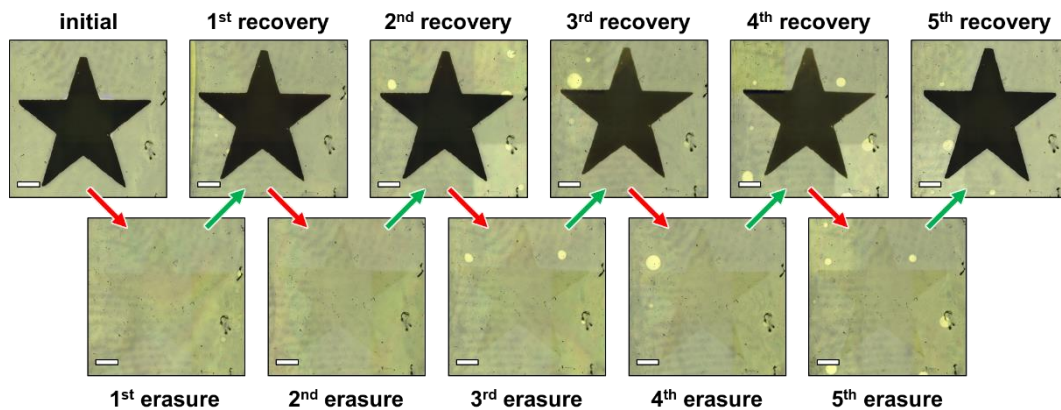


Fig. R7. Optical images of patterned AnLCE film during erasure-recovery. Scale bar: 1 mm.

We performed at least 5 erasure-recovery cycles to demonstrated its good reversibility, as shown in **Fig. R6b** and Fig. 3e in the manuscript, the amplitude of microwrinkles decreased and recovered to initial values. When the wrinkle pattern is erased, the macroscopic surface pattern (determined by masked exposure) cannot be recognized by the naked eye. After the recovery of wrinkle pattern, no obvious distortion or loss of fidelity was found in the macroscopic scale, which is shown in **Fig. R7**. (It should be noted that the macroscopic pattern (optical images) emerged while surface buckling occurred. The repeating stretching-releasing serves only to transform the buckling microstructure, such as folding, into uniform wrinkles.). Fig. 3e illustrates the variation in wrinkle amplitude value during erasure-recovery cycles. While the insets represent the 2D pattern of the wrinkle surface. We have increased the size of the insets in Fig. 3e to make them easier to read (**Fig. R6b**).

Comment 11: *p15, ln310: The claim of ‘long term stability’ has not undergone any real rigorous test. Start with elevated temperatures.*

Response to 11:

The claim of “long term stability” was described as the stability of wrinkle under room temperature. The stability of the wrinkle pattern has been shown in the Supplementary Fig. 16 and discussed in the part of wrinkle fabrication (Page 11, Line 225). The wavelength and amplitude of wrinkles remained stable over the 15-day

storage period at room temperature ($< 30\text{ }^{\circ}\text{C}$). Therefore, after the thermal and mechanical regulation of the prepared wrinkles, we concluded the wrinkle pattern “long-term stability, rapid response and well-reversibility”. We have now changed the description to “**acceptable stability**”. (Page 15, Line 318)

Reviewer #1 (Remarks to the Author):

Comment 1: *I accept the answers 1 and 3 as stated in the author's response. However, an answer 3 should be clearly limited to "soft" displays. This is because this technology can only be used in fields where soft materials are acceptable.*

Response to 1:

Thank you for your professional comments and help related to this manuscript. We have revised the manuscript carefully according to your comments.

Comment 2: *Unfortunately, I cannot accept an answer 2 regarding SAXS. In the structural drawings of Figures 1 and 4, authors show a polydomain and a nematic-like monodomain in the surface and inner layers. If the higher-order structure is formed as shown in the structural drawings, some SAXS peak should be shown around at $d = 2$ to 4 nm. Broad azimuthal data alone, as shown in Supplementary Figure 26, cannot be discussed without data on the 2θ or q spectra.*

Response to 2:

Thank you for your constructive comment. We repeated the small-angle X-ray scattering (SAXS) experiments and added the q spectra derived from the scattering images. However, the existing SAXS results did not show the expected scattering peaks, which is corresponding to previous studies by many other people (e.g., *Adv. Mater.* **2023**, *35*, 2209853; *Adv. Mater.* **2021**, *33*, 2103755).

The detailed SAXS experiment and discussion are shown below:

Three AnLCE samples were selected for small-angle X-ray scattering (SAXS) experiment: **(1)** The raw AnLCE films, without any treatment, was used to demonstrate the substrate. **(2)** The surface UV-exposed AnLCE films was exposed to UV light at an intensity of 35 mW/cm^2 for a period of 10 minutes. **(3)** The fully UV-crosslinked AnLCE film, which was exposed to a higher intensity (200 mW/cm^2) UV light for a period of 30 minutes on both sides, was used to demonstrate the UV-crosslinked top layer. The thickness of the sample films was controlled as approximately $130 \text{ }\mu\text{m}$.

SAXS experiments were conducted on an Xeuss 2.0 WAXS/SAXS system (Xenocs, France). The wavelength of X-ray was 0.154 nm. And the X-ray detector was Pilatus 3R 300K, with single pixel size was 172 μm . During experiment, the distance between sample and detector was set to be 538.1 mm, and samples underwent X-ray exposure for 300 s. The scattering data of AnLCE films were acquired **(1)** before stretching, **(2)** under 200% stretching ($3L_0$) and **(3)** after releasing to their original length, respectively.

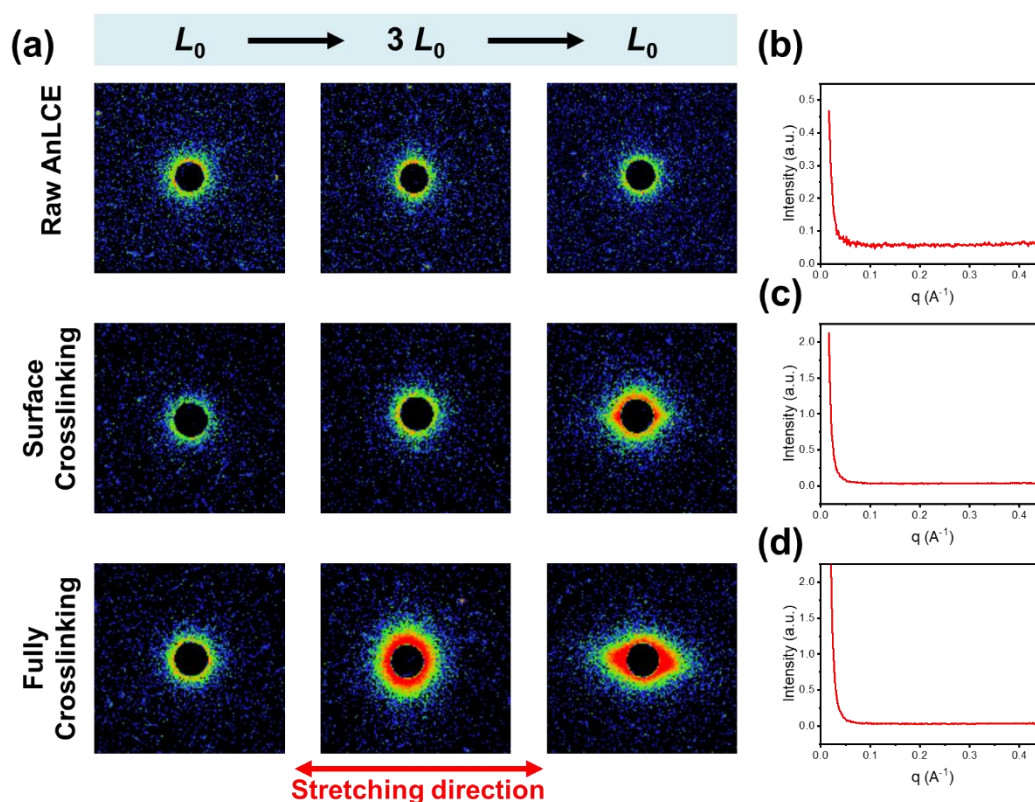


Fig. R1 SAXS results of AnLCE films. **a** The scattering images of AnLCE films before stretching, under 200% stretching and after releasing, respectively. **b-d** the q spectra derived from the scattering images after releasing. Spectra designated **b**, **c**, **d** corresponded to the raw AnLCE, surface crosslinked AnLCE and fully crosslinked AnLCE film, respectively.

The SAXS results of (1) raw AnLCE, (2) surface crosslinked AnLCE and (3) fully crosslinked AnLCE film were shown in **Fig. R1a**. The 3 AnLCE films did not display apparently anisotropic character before stretching. After releasing from 200% stretching, evident orientation was found in the scattering patterns of surface

crosslinking and fully crosslinking AnLCE film, and none obvious orientation was found in the raw AnLCE sample due to the recovery of the polydomain phase. These results indicated the liquid crystal orientation in the UV-crosslinked top layer and confirmed the UV crosslinking play a key role in the formation of anisotropic structure after mechanical stimulus.

While the **Fig. R1b-d** were the q spectra of AnLCE films derived from the scattering images after releasing. The recommended scattering peak did not found in these q spectra, which should be attribute to the LC alignment of nematic phase, that has no higher ordered alignment except the orientation of mesogens. In some relative works about the nematic LCE, the SAXS images should not have any diffraction dot. For example, in **Referenced Fig. 1**, a polydomain nematic LCE prepared via thiol-acrylate Michael addition showed an isotropic ring in the WAXS pattern. Only one scattering pot in with the scattering intensity radially decreased from the beam center in the SAXS image, therefore, there was no peak showed in the SAXS curve (**Ref. Adv. Mater. 2023, 35, 2209853**).

[REDACTED]

Referenced Fig. 1 [REDACTED]. Reference: *Adv. Mater.* **2023, 35, 2209853**.

If the nematic LC phase transfer to another phase with higher ordered structure in the LCEs, there will be some diffraction dots in their corresponded SAXS pattern. Another example reference could be used to further clarify the absence of diffraction dots in our SAXS results, as shown in **Referenced Fig. 2**, an ionic LCE fiber (IonoLCE) underwent the transition from polydomain nematic to monodomain nematic and

smectic phase during stretching (**Ref.:** *Adv. Mater.* **2021**, *33*, 2103755). The IonoLCE fibers was polydomain state at first, the corresponded SAXS images shows a uniformed scattering ring because of the macroscopic isotropic character of polydomain nematic phase. Under moderate stretching (100%), the LCE transferred to monodomain nematic phase, the anisotropic scattering ring was found. When the IonoLCE was subjected to higher stretching (>200%), it transferred to smectic phase featuring a longitudinal periodicity of ≈ 4 nm and shortened face-to-face mesogen distance of ≈ 4 Å. The 4-nm spacing was mainly determined by the length of chain spacer (EDDET and PETMP).

[REDACTED]

Referenced Fig. 2 [REDACTED]. Reference: *Adv. Mater.* **2021**, *33*, 2103755.

Nevertheless, in our AnLCE, the ~~wrinkled top layer following stretching - releasing~~ remained in the monodomain nematic phase along the “wave-like” wrinkle surface(**Fig. 1a** and **Fig. 4a** in the manuscript). The nematic phase should not display other diffraction dot in the SAXS images except the anisotropic in the center scattering dot in the SAXS image. Meanwhile, the introduction of dithiol-anthracene will replace some content of EDDET, it may interrupt the detection of the highly order structure in some extent. Therefore, we think it is reasonable of the absence of the diffraction dots in the SAXS images (no peak in the q spectrum) in our crosslinking AnLCE samples after mechanical stimulus.

Comment 3: *Also, there is no description of the measurement conditions for the additional data, which is inappropriate for a technical paper.*

Response to 3: Thank you for your professional comments. We have revised the manuscript carefully and added the description of the measurement conditions into the section of **Characterizations**.

Comment 4: *So reviewer think that it is necessary to add both out-of-plane and in-plane film data to show the validity of the structural drawings as shown in Figures 1 and 4. If XRD data cannot explain the structure, then the polydomain liquid crystal-like structure is no longer the basis for the functional expression of this Liquid Crystal Elastomer, it is simply the aggregation and stretching phenomena of the crosslinking chains with a difference in linear expansion coefficients.*

Response to 4:

Authors thanks you for your insightful comments. We repeated the small-angle X-ray scattering (SAXS) experiments and added the q spectra derived from the scattering images (**Supplementary Fig. 26** in the Supplementary Information). Although the existing SAXS results did not show the expected scattering peaks, we still verified the supposal (**Fig. 1** and **Fig. 4** in the manuscript) of the functional expression of liquid crystal through supplemental controlled experiment and discussion.

In this work, the properties of surface wrinkling are the combination of the anthracene and liquid crystal molecules: the anthracene plays the role in the generation of the crosslinking top layer via UV irradiation and enables the surface wrinkling from subsequent stretching-releasing, while the stabilization the surface wrinkle and the thermal regulation was the important function expression of the LC phase transition.

In order to ascertain whether the aggregation and stretching phenomena of the crosslinking chains with a difference in linear expansion coefficients play the key role in the surface wrinkling in AnLCE films, another non-liquid crystalline elastomer (AnBPE) film was designed and its wrinkling behavior was investigated. In this AnBPE film, the liquid crystalline monomer (RM 257) was replaced by Bisphenol A ethoxylate

diacrylate (BPAEDA, $M_n = 449$), which is a non-liquid crystalline monomer. The incorporation of the bisphenol unit will result in a greater degree of stiffness along the main chain in comparison to the PEGDA chain. The chemical compounds utilized in the synthesis of AnBPE films are illustrated in **Fig. R2a**.

The controlled elastomer films, AnBPE, without LC gen were synthesis by two-step polymerization. The mole ratio of BPAEDA/An(SH)₂/EDDET/PETMP/Irgacure 784 is 115/40/40/10/2 with 15% excess of diacrylate groups. The monomer mixture containing 1.15 mmol of PEGDA, 0.4 mmol of An(SH)₂, 0.4 mmol of EDDET and 0.1 mmol of PETMP and 0.02 mmol of initiator, I 784, were dissolved in 0.4 g of toluene at 80 °C. When the mixture was totally melted to a clear liquid, 100 mg of DPA/toluene solution was added to the mixture to catalyze the TAMAP reaction. After overnight TAMAP reaction, the mixture was irradiated by green light for fully cure via radical photo polymerization initiated by the I 784. The AnBPE films were fully dried at 80 °C in a vacuum oven for 12 h.

The strategy “UV-SR” was then utilized to fabricate wrinkle pattern on the AnBPE film. The AnBPE film was subjected to UV irradiation at an intensity of 35 mW cm⁻² for a duration of 600 seconds, utilizing a stripe-pattern photomask with a width space of 150 μm between the exposed and non-exposed areas. After UV irradiation, the AnBPE film was subsequently stretched to $2L_0$ and released. Following the release, wrinkles formed on the UV-exposed area of the AnBPE surface (displayed in black), while the non-exposed area kept flat, detailed result was showed in **Fig. R2b**. This demonstrated the anthracene provided the surface photocrosslinking to generate the mismatch in the strain behavior of the surface layer and the inner layer and finally led to the surface wrinkling under the mechanical stretching-releasing. After the generation of surface of AnBPE, the amorphous state of the flat and wrinkled AnBPE was determined by polarized optical microscopy (POM), the polarized optical images showed that the wrinkle surface of AnBPE was dark at the angle of 45° to the polarizer, which indicated the isotropic structure in the wrinkled layer.

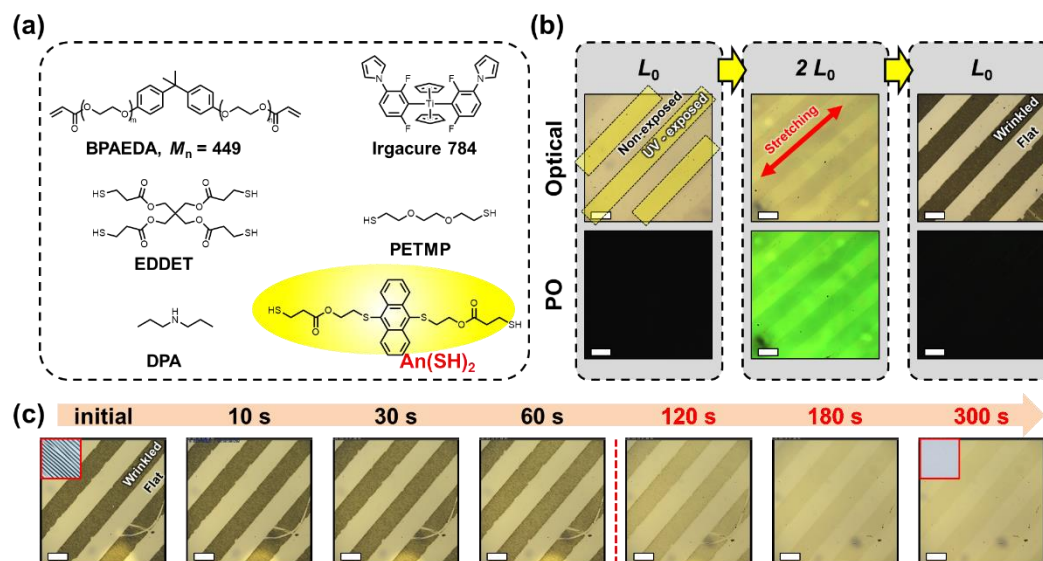


Fig. R2 The surface wrinkling and its evolution of normal elastomer AnBPE. **a** Chemicals for fabricating controlled AnBPE. **b** The optical and polarized optical images of AnBPE's surface during wrinkle fabrication. **c** The surface self-erasure of surface wrinkles during placement. Scale bar: 200 μm . The insets were the amplified images of surface morphologies in the wrinkle area, with a size of 32 $\mu\text{m} \times 32 \mu\text{m}$.

Although the surface wrinkles of AnBPE could be produced by UV-SR, the resulting wrinkle pattern could not maintain itself for long periods of time in the absence of any external stimulus at room temperature. As shown in **Fig. R2c**, the wrinkled area exhibited stability during the initial 60 seconds after generation. Subsequently, the color of the wrinkled area underwent a continuous fading due to the reduction in surface light scattering, which is caused by the evolution of the surface wrinkle. Ultimately, the color of the wrinkled area underwent a transformation, becoming identical to that of the initially flat area. This indicated the totally collapse of the wrinkle structure in the UV-exposed area.

The controlled experiments on the surface wrinkling of AnBPE film demonstrated that the generation of stable surface wrinkles and their thermal regulation were not solely dependent on the surface crosslinking via anthracene, but also on the incorporation of liquid crystal. If the functional expression of surface wrinkles is solely dependent on the aggregation and stretching phenomena of the crosslinking chains with a difference in linear expansion coefficients, the generated surface wrinkles tend to

recover its flat surface because the stress relaxation of the wrinkled top layer (**Ref.:** *Adv. Funct. Mater.* **2022**, *32*, 2207691; *Adv. Mater.* **2024**, 2314201). Even if some surface wrinkle can be generated via thermal expansion, the wrinkles can only temporarily be erased by heating and subsequently recover to a wrinkled surface upon the removal of the thermal stimulus (**Ref.:** *Nature* **1998**, *393*, 146; *Sci. Adv.* **2018**, *4*, eaar5762; *Sci China Mater* **2024**, *67*, 363).

In contrast, in our work on the AnLCE wrinkle system, the generated surface wrinkles (by both UV-SR and S-UV-R) exhibited well stability in the 15-days test (**Supplementary Fig. 16** in the Supplementary Information) and underwent unidirectional erasure by heating (**Fig. 3** in the manuscript), and the wrinkles did not recover without external mechanical stimuli. The results demonstrated that the self-maintaining properties and thermal regulation of the surface wrinkles of AnLCE was the functional expression of the phase transition of the liquid crystal.

Reviewer #2 (Remarks to the Author):

The authors have adequately addressed my questions and comments. I recommend accepting this manuscript.

Response:

We sincerely thank you for your efforts and help related to this manuscript.

Reviewer #3 (Remarks to the Author):

The comments of the other reviewers and the author have assuaged my concerns of novelty, and I am happy with the author's replies. I believe the article is suitable for publication.

I note the following textual errors:

**p11, ln 241: Should be "These results corresponded to..." Delete 'were'*

**p12, ln 249: should be 'evolution of wrinkles...'*

**p12, ln 258: should be 'short wavelength' rather than ;small'. Wavelength is a length, thus, shorter and longer.*

Response:

Thank you for your constructive feedback and careful checking. The textual errors have been carefully corrected.

Reviewer #1 (Remarks to the Author):

Comment 1: *Reviewer accepts the authors' 3rd revised responses. I also thank the authors for the additional SAXS measurements. I supposed that some smectic structure might present in this LCE, but understood that only nematic structures of low ordered structure existed. Therefore, it should be described as "nematic polydomain" so that it is not misunderstood as "smectic polydomain" in illustrations in the paper.*

Response:

Thank you for your professional comments and help related to this manuscript. To avoid misunderstanding, the relative description of “polydomain” has been changed to “polydomain nematic” in the main text according to your comments.

Comment 2: *[Comments on SAXS data analysis] Reviewer considered the following relationship between SAXS and higher-order structure of this material.*

In Fig. R1, (2) under 200% elongation, the SAXS scattering is isotropic and unrelated to the nematic monodomain structure. However, , a tail of diffraction scattering related to the long-periodic structure is observed in the stretching direction. This suggests that this SAXS result is a scattering tail due to the physical shape of the sheet wrinkle structure rather than to the higher-ordered structure of the LCE.

Response:

Authors thank you for your constructive comments and explanation on the results of our experiments. In the scattering images, the isotropic pattern of “(2) under 200% elongation” may be attributed to the relatively low resolution of the used SAXS equipment.

And the authors considered that the anisotropic scattering patterns after releasing (3) should be the cooperation of both the LC alignment and the physical shape of the sheet wrinkle. Because the alone wrinkle structures are in much larger size (typical wavelength > 500 nm), so that it is hard to be identified in the small-angle area if without the LC alignment.