4D Chiral Photonic Actuators with Switchable Hyper-Reflectivity

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Cholesteric liquid crystals (CLCs) are chiral photonic materials reflecting only circularly polarized light with the same handedness as the helical polymer structure. Concurrent shape and color changes can be achieved using CLCs, but the fabrication of CLCs with switchable 3D shape, structural color, and hyper-reflectivity, that is, reflecting both left- and right-handed circularly polarized light simultaneously, has not yet been achieved. Here, CLC elastomer (CLCE) actuators are reported to reflect equal amounts of left- and right-handed circularly polarized light. Hyper-reflectivity is achieved by uniaxially stretching the partially crosslinked film to induce helix deformation which is then fully crosslinked to fix the deformed helical structure. The shape, structural color, and hyper-reflectivity of the polymer film are switchable with temperature. At high temperatures, only right-handed circularly polarized light is reflected and the color is redshifted. The film can be shaped in three dimensions: a structural colored 3D shaped beetle is fabricated using molding, which reflects both left- and right-handed circularly polarized light and shows reversible, temperature responsive structural color and 3D shape changes. Hence, 4D engineered bioinspired multifunctional materials are fabricated, which are interesting for applications ranging from sensing actuators to switchable hyper-reflective films and objects.

1. Introduction

In nature, organisms have astonishing control over their shape, color, and reflectivity. For example, cephalopods are masters of camouflage by adapting their color and shape to their surroundings.\(^1\),\(^2\) The iridescent metallic green beetle *Chrysina gloriosa* selectively reflects left-handed circularly polarized light due to the helicoidally arranged nested arcs,\(^3\) and the tortoise beetle *Charidotella egregia* is able to change the structural color of its cuticle reversibly when disturbed by stressful external events.\(^4\) Such organisms have inspired scientists to integrate adaptive shape and optical changes into synthetic multifunctional materials and objects. For example, structural color actuators have been fabricated using photonic crystals,\(^5\),\(^6\) cellulose nanocrystals\(^7\),\(^8\) and cholesteric liquid crystals.\(^9\)–\(^15\) In spite of the control over color and shape being achieved at the same time, the structural color shape changes are confined in-plane and macroscopic 3D deformations remain challenging.

Cholesteric liquid crystals (CLCs) form helical photonic structures, resulting in reflection of circularly polarized light with the same handedness as the helical structure, where the wavelength of light reflected by the structure is determined by the pitch and the average refractive index.\(^16\)–\(^23\) By engineering the helix, the photonic properties can be altered leading to reflection of both left- and right-handed circularly polarized light. In this fixed, static, hyper-reflective state the 50% reflection limit is exceeded.\(^24\)–\(^28\) When cholesteric liquid crystal elastomer (CLCE) films are stretched, the thickness decreases, which causes a change in reflected color.\(^13\),\(^29\),\(^30\) The perceived color and shape of such flat films can be programmed and erased.\(^14\) In addition, the helical structure can be deformed by applying uniaxial strain in the plane perpendicular to the helix axis, resulting in a reflection of both left- and right-handed circularly polarized light. This phenomenon has been both theoretically predicted and experimentally investigated.\(^12\),\(^31\)–\(^35\) When the strain is released, the film will recover to the normal helical state, where the reflection is again limited to 50%. Because of this, the strain must be continuously applied to maintain the hyper-reflective state. Consequently, environmentally responsive, hyper-reflective photonic actuators have not yet been reported.

Here, we present a simple method to prepare 3D shaped CLCE structural color actuators with switchable hyper-reflectivity. The temperature responsive CLCE is fabricated using a two-step crosslinking procedure that has been previously used to prepare 3D shaped actuators.\(^36\) The polarization of the structural color is programmed by uniaxially stretching the
partially crosslinked film, which deforms the helical structure and gives rise to a distorted helical structure that reflects both left- and right-handed circularly polarized light. The deformed helical structure is then fixed by fully crosslinking the film. Upon heating, the polarization selectivity of the film reversibly changes from reflection of equal amounts of left- and right-handed circularly polarized light to only right-handed circularly polarized light. The programmed CLCE films can reversibly undergo transition between the programmed shapes in response to temperature changes, with simultaneous changes in the structural color and polarization of the film. A 3D model shaped as a beetle with polarization independent structural color is created by molding. The artificial beetle changes its shape, color, and hyper-reflectivity upon altering the temperature. Our method provides a tool to fabricate bioinspired stimuli-responsive multifunctional materials with arbitrary 3D shapes and programmed reflectivity, color, and shape changes. Such materials are interesting for applications ranging from sensing actuators to switchable hyper-reflective films and objects.

2. Results and Discussion

2.1. The Preparation of the CLCE Film

The CLCE is made using a two-step crosslinking involving a base-catalyzed thiol-acrylate reaction and a light-induced free radical polymerization reaction reported previously. The CLC mixture is composed of 6 components (Figure 1A). Monomer 1 is a liquid crystal diacrylate monomer and monomer 2 is a chiral dopant that induces the formation of

**Figure 1.** A) Chemical structures of molecules used in the CLC mixture. B) The two-step crosslinking procedure to prepare the temperature responsive cholesteric liquid crystal elastomer film. The thiol-acrylate polymerization is carried out between two glass plates. After removal of the glass plates, the first thiol-acrylate network film is obtained. After stretching, the remaining acrylate groups are photo-polymerized to fully crosslink the film and fix the strain. The transmittance spectra of the film, C) before and D) after being stretched to a strain of 47.5% and photo-polymerized. The insets in (C) and (D) show the photographs of the film before and after being stretched, respectively.
the cholesteric liquid crystal phase. Dithiol 3 and tetrathiol 4 act as a chain extender and a crosslinker for the first network, respectively. Triethylamine 5 is the catalyst for the thiol-acrylate Michael addition to form the first network. Photoinitiator 6 is added to initiate the second polymerization upon UV irradiation. The CLCE is prepared using a composition with 10 mol% excess acrylate and 25 mol% of the thiols coming from crosslinker molecule 4. It is found that the cholesteric-to-isotropic transition temperature of the mixture prior to the thiol-acrylate polymerization is around 36 °C, verified using polarized optical microscopy (POM) (Figure S1, Supporting Information).

The CLC mixture was melted and vortexed to ensure good mixing, and then cooled to room temperature (~22 °C). A drop of the mixture was put on a polyvinyl alcohol (PVA)-coated glass substrate with pieces of tape glued onto the edges to serve as spacers and another slide of PVA-coated glass was placed directly on top to form a cell. The glass substrates were sheared along one direction to align the mixture. As the cholesteric-to-isotropic transition temperature increased during the thiol-acrylate polymerization, the reaction was first carried out at 22 °C for 2 h and then at 55 °C for 5 h (Figure 1B) to speed up the crosslinking process. After the first crosslinking, the sample was immersed into deionized water at 50 °C for a few hours to dissolve PVA and detach the film from the glass substrates to obtain a freestanding film. The thickness of the film was measured using POM and found to be 130 μm (Figure S2, Supporting Information). The completion of the thiol-acrylate polymerization was verified using Fourier transform infrared spectroscopy (FT-IR) (Figure S3, Supporting Information). The absorption band at 2550 cm⁻¹ corresponding to the S–H stretching vibration disappeared completely, demonstrating that the thiol groups have reacted with the acrylate groups. Differential scanning calorimetry (DSC) revealed a glass transition temperature around –16 °C (Figure S4A, Supporting Information) and a cholesteric-to-isotropic transition temperature at 82 °C. After thiol-acrylate polymerization, the film showed a weak reflection between 500 and 600 nm (Figure 1C) and a low transparency, indicating that the alignment was not perfectly planar but focal conic instead (see the inset photograph in Figure 1C and the POM image in Figure S5A, Supporting Information). The film was then stretched at room temperature to a strain of 47.5% and then UV irradiated at room temperature to induce the radical photo-polymerization and fix the deformation of the film. After this polymerization step, at room temperature the film has a fixed shape and a greenish blue color with a reflection band centered at 500 nm (Figure 1D; Figure S8, Supporting Information). Hyper-reflectivity with a 90% reflection of unpolarized light is observed. The film is also more transparent and fewer defects are observed (see the POM image, Figure S5B, Supporting Information), which means the alignment is improved by stretching as expected. The glass transition temperature of the film after photo-polymerization is around –10 °C as determined by DSC (Figure S4B, Supporting Information).

Figure 2. A) Photographs of the film photopolymerized with a strain of 47.5% exposed to LCP and RCP light at room temperature. B) The reflectance spectra of the film measured with LCP and RCP at room temperature. C) Photographs of the film photo-polymerized with a strain of 47.5% between crossed linear polarizers at different angles.
2.2. Hyper-Reflectivity

The reflected greenish blue color of the stretched film is visible through both left- (LCP) and right-handed circular polarizers (RCP) at room temperature (Figure 2A), indicating the film reflects both left- and right-handed circularly polarized light. Reflectance spectra of the film were measured on a spectrophotometer, using the combination of a linear polarizer and a quarter-wave plate to generate circularly polarized light. Measurements were performed with left- and right-handed circularly polarized light; the baselines were corrected using the corresponding type of light. Remarkably, reflectance spectra show reflection bands with the same reflection at the peak center, regardless of whether they were exposed to right- or left-handed circularly polarized light (Figure 2B), indicating full reflection of light of both circular polarization types.

The same degree of reflection of both left- and right-handed circularly polarized light indicates that the stretched CLCE film acts as a 1D photonic material: when the film is uniaxially stretched in the plane, the director predominantly aligns along the stress axis, leading to a deformed helix structure (see Figure 3E) that approaches a 1D Bragg reflector state reflecting both left- and right-handed circularly polarized light to the same degree. The deformed helix gives rise to in-plane birefringence and thus optical retardation for light transmitted through the film. We propose that in the first few micrometers of the stretched CLCE film, right-handed circularly polarized light is reflected, and the transmitted left-handed circularly polarized light is converted to elliptically polarized light by the birefringence, distorted helical structure (vide infra), and eventually turns into right-handed circularly polarized light through the thick film (130 µm). This right-handed circularly polarized light is reflected and converted into left-handed circularly polarized light again. Hence equal amounts of left- and right-handed circularly polarized light are reflected and hyper-reflectivity is observed.

When the film is stretched, the realignment of the LC molecules should lead to the formation of a new alignment director positioned along the stretching direction. To verify this hypothesis, the stretched CLCE film was placed between crossed linear polarizers.
and rotated; the film changed from a dark state (0°) to a bright state (45°), and when further rotated to 90°, the film became dark again (Figure 2C). This change indicates birefringence in the plane of the film, revealing the presence of a 1D photonic structure. For the film that was not stretched, before photo-polymerization there is no change when it is rotated from 0° to 45° to 90° (Figure S6, Supporting Information), meaning there is no birefringence in the plane. This is expected, as the alignment of the LC molecules uniformly rotates along the helical axis (see Figure 3E). These results reveal that the LC molecules in the stretched film have a preferential alignment along the stretch direction.

2.3. Temperature Responsive Color, Shape, and Polarization Changes

We first studied the temperature-responsive color changes of the stretched film by measuring the transmittance spectra at different temperatures using an Ocean Optics spectrometer attached to the POM (Figure S7, Supporting Information). Upon heating from 22 to 171 °C, the reflection band displayed a red shift from 500 to 670 nm (Figure 3A). In addition, the transmittance at 430 nm increases within this temperature range, indicating less scattering at higher temperatures. The reflection band measured at high temperature is much stronger than the reflection band of the first crosslinked network (Figure 3A versus Figure 1C). The change of reflection band is small; between 22 and 55 °C, after which the band shift is larger until 118 °C, and then the reflection band shift is again small up till 171 °C (Figure 3B). The larger shift between 55 and 118 °C is likely related to the cholesteric-to-isotropic transition temperature of the first crosslinked network, which is around 82 °C (Figure S4A, Supporting Information). When the intensity of the reflection band is plotted as a function of the temperature (see the black dots in Figure 3B) the intensity gradually dropped from 90% to 55% when heating from 22 to 171 °C. Heating-cooling cycles were carried out for ten times, showing that the color changes are fully reversible (Figure S9A, Supporting Information). The film is greenish blue at 22 °C and red at 171 °C (Figure 3C), corresponding well with the reflection bands measured at these temperatures (Figure 3A).

Apart from the color change, the dimensions of the stretched film also change (Figure 3C). The length in the stretching direction of the film decreased, and the width, perpendicular to the stretching direction, increased. When the length and width of the film are plotted as a function of temperature from 22 to 171 °C (Figure 3D), the length decreased from 29.5 to 20 mm and the width increased accordingly from 7 to 8.9 mm, revealing temperature responsive shape changes of roughly 30%. The dimension change is also fully reversible (Figure S9B, Supporting Information). Similar to the wavelength change, the length change is small, from 22 to 58 °C, and then larger between 58 and 119 °C, after which length change is small again. The width showed a similar but opposite shape change.

Figure 4. A) Photographs of the film photo-polymerized with a strain of 47.5% taken with no polarizer and through LCP and RCP at different temperatures. B) The transmittance spectra of the film photo-polymerized with a strain of 20% at different temperatures measured with LCP and C) RCP.
To study the temperature responsive polarization change of the reflected light, the same stretched film was heated up on a hot plate from 22 to 197 °C and photographs were taken with no polarizers, LCP, and RCP to verify the reflection (Figure 4A). When heated from 22 to 142 °C, the color of the film changed from greenish-blue to red, and the colors were visible with both LCP and RCP, indicating the film reflected both left- and right-handed circularly polarized light in this temperature range. When further heated to 174 °C, the film was still red, but the color could only be seen with no polarizer or with RCP and was no longer visible through the LCP, which means the film did not reflect left-handed circularly polarized light at this temperature. When further heated to 197 °C, the color was still only visible with no polarizer or with RCP. This means the reflection of left-handed circularly polarized light vanished when heating the film to a high temperature. This behavior is supported by the temperature dependent transmittance spectra with LCP and RCP. The reflection band measured with LCP gradually decreases and eventually disappears while the one measured with RCP remains at high temperature (Figure 4B,C; Figure S10, Supporting Information). The intensity of the overall reflection band also diminishes upon heating the film (Figure 3A,B) as the contribution of LCP reflection decreases.

Based on this data, we propose the following mechanism for the temperature responsive shape, color, and reflectivity changes. The observed shape change in the stretched CLCE film is similar to that in planar aligned nematic liquid crystal elastomer films. The thermotropic decrease in order causes contraction along the nematic director, and expansion perpendicular to the director.[10,41] This behavior supports the presence of a stretched, deformed helical 1D photonic structure (Figure 3E). The temperature induced decrease in order also results in a corresponding increase in the thickness of the films, leading to a redshift of the reflection band. Most likely the mechanism of temperature responsive reflectivity changes is similar to the reversible shape memory in a nematic liquid crystal elastomer actuator.[18] When heated up, the alignment and shape of the stretched CLCE film prepared here undergoes transition from the programmed state where the helix is deformed to the initial state where the helix is not deformed, resulting in a change from hyper-reflectivity to reflection of only right-handed circularly polarized light.

### 2.4. Effect of Different Strains on Color, Shape, and Polarization Changes

To study the effect of different strains on the reversible color, shape, and polarization change behavior, after thiol-acrylate polymerization, the films were uniaxially stretched at varying strains, that are, 0%, 20%, 47.5%, and 62.5%, and photo-polymerized. Films with reflection bands of 538, 516, 500, and 491 nm were obtained for the strains of 0%, 20%, 47.5%, and 62.5%, respectively (Figure S11, Supporting Information). There is a linear relation between the reflection band shift and strain: the more the film is stretched, the larger the reflection band shift. This behavior can be explained by the compression of the pitch in the direction perpendicular to the plane of the film. The transmittance spectra of the films with different strains at room temperature were measured with unpolarized light and with LCP and RCP, respectively (Figure S12, Supporting Information). The film that was not stretched (0% strain) before photo-polymerization only reflects right-handed circularly polarized light (Figure S12A, Supporting Information). The 20% stretched film reflects left- and right-handed circularly polarized light to the same degree (Figure S12B, Supporting Information). When the film is stretched to 62.5%, the transparency is improved further, and the film still reflects both left- and right-handed circularly polarized light in a comparable way (Figure S12C, Supporting Information).

The temperature-responsive color change of the different stretched films was studied (Figure 3A; Figure S13, Supporting Information). The reflection band of all the films redshifts upon heating, as expected (Figure 5A). When heating from room temperature (22 °C) to 171 °C, the degree of stretching only affects the start positions of the reflection bands and there is no big difference in the wavelength shift at high temperature (171 °C) for different films.

The dimensions of the films with different programmed strains were also measured over this temperature range (Figure 5B). It is found that, upon heating, for strains of 20%, 47.5%, and 62.5%, the relative lengths of the films decreased from 1.2, 1.475, and 1.625 times the original length before stretching to their initial length when heated up to 159 °C. When further heated to 178 °C, the relative lengths of the films continued to decrease to around 0.96. The width of these

![Figure 5](https://www.afm-journal.de/)

Figure 5. The relative wavelength of the reflection band (A) and the relative dimension (B) of the films that were photo-polymerized with a strain of 0%, 20%, 47.5%, and 62.5% as a function of temperature. The relative wavelength (dimension) is defined as the dynamic wavelength (dimension) divided by the wavelength (dimension) before stretching.
films increased upon heating. The relative length contraction and width expansion are highly dependent on how much the film was stretched before photo-polymerization: the more the film is stretched, the larger the dimensional change of the film will be. This means that the dimension changes can be easily programmed using varied strains. In contrast to the stretched films, when the film was not stretched before photo-polymerization, that is, at a strain of 0%, both the length and width contract upon heating.

The polarization response to temperature changes was also studied for different films. For the film with a strain of 20% (Figure 4B,C; Figure S14, Supporting Information), both left- and right-handed circularly polarized light is reflected at room temperature. When heated, the reflection of left-handed circularly polarized light vanished around 150 °C as the film is not visible with LCP and the reflection band measured with LCP disappears (Figure 4B), while the right-handed circularly polarized light is still reflected as the film is still visible with RCP and the reflection band measured with RCP remains at high temperature (Figure 4C). For the film with a strain of 62.5% (Figure S15, Supporting Information), the reflection of left-handed circularly polarized light starts to vanish above 197 °C.

These temperature responsive polarization changes are reversible upon cooling.

The data presented show that the temperature responsive shape, color, and polarization changes can be programmed by the degree of stretching (strain) of the film after the first crosslinking step.

2.5. 4D Chiral Photonic Actuators with Switchable Hyper-Reflectivity

To demonstrate 4D chiral photonic actuators with switchable hyper-reflectivity, a structural colored beetle was created using molding (Figure 6A). After the thiol-acrylate polymerization, the film was first stretched with a strain of 30% and then molded between a positive and negative “beetle” mold. The film was then removed from the mold and photo-polymerized at room temperature to fix the 3D shape, color, and polarization. A green colored hollow 3D beetle with a length of 18 mm, a width of 11 mm and a height of 4 mm at room temperature was created (Figure 6B). While the C. gloriosa beetle (Figure 6C) only reflects left-handed circularly polarized light, the green color of the fabricated beetle...
is visible through both LCP and RCP at room temperature, indicating the reflection of both left- and right-handed circularly polarized light (Figure 6D). Upon heating to 197 °C, the color of the beetle became red and the 3D shape became a 2D flat film with a length of 14 mm, a width of 12 mm and a thickness around 130 µm. The color of the flat beetle is only visible through RCP, indicating the reflection of exclusively right-handed circularly polarized light at high temperature. When cooling down to room temperature, the color, shape, and polarization of the reflected light restored with no visible lag. Other 3D photonic actuators can also be created. Using a different mold, a 3D smiley face that can change its color, shape, and polarization was fabricated (Figure S16, Supporting Information).

3. Conclusion

Reversible color, shape, and polarization changes are achieved in 3D objects using a two-step crosslinking and molding process. After initial crosslinking, a CLCE film is mechanically deformed to program the color, shape, and reflectivity and then photo-polymerized to fix the engineered properties. The stretching results in the reflection of both left- and right-handed circularly polarized light. A 3D structural colored beetle is fabricated that can reversibly change its color, shape, and polarization dependence upon heating. Using different molds and by programming the colors using different strains, arbitrary shapes with a variety of colors can be achieved. The objects fabricated with programmable shape and color changing functions mimic living organisms and create new responsive hyper-reflectivity properties. Hence, an easy and versatile method has been developed to create stimuli responsive 3D materials with multiple switchable optical and actuating functions.

4. Experimental Section

Materials: The diacrylate liquid crystal monomer 1 was purchased from Merck. Monomer 2 was received from BASF. 2,2′-(Ethylenedioxy) diethanethiol (3), pentaerythritol tetrakis (3-mercaptopropionate) (4), triethylamine (5), and PVA were obtained from Sigma-Aldrich. Photoinitiator 6 was obtained from CIBA.

Mixture Used to Determine the Cholesteric-to-Isotropic Transition Temperature of the Monomers Using POM: 70.48 wt% monomer 1, 6.47 wt% monomer 2, 13.86 wt% dithiol 3, and 6.19 wt% tetrahydro 4 were added into a vial and melted and then vortexed to ensure proper mixing. The mixture was then dropped on a glass slide and covered with a second slide, which was then placed on the THM5600 hot-stage and monitored with POM in transmission mode under crossed-polarizers.

PVA Functionalized Glass Substrate: To prepare the PVA functionalized glass substrate, 3 × 3 cm² glass plates were cleaned in acetone for 30 min using ultra-sonication, followed by 30 min in ethanol and subsequently treated by UV-Ozone (PR-100, Ultra Violet Products) for 20 min. 5 wt% PVA with a molecular weight of 9000 was dissolved in distilled water and spin-coated on a clean 3 × 3 cm² glass plate using a spin coater (Karl Suss CT 62) by rotating at 2500 rpm for 30 s. The PVA coated glass plates were then placed at 60 °C for 30 min and were then rubbed on a velvet cloth.

Film Preparation: 70.48 wt% monomer 1, 6.47 wt% monomer 2, 13.86 wt% dithiol 3, 6.19 wt% tetrahydro 4, 2 wt% catalyst 5, and 1 wt% photoinitiator 6 in Figure 1A were added into a vial and melted and vortexed to ensure proper mixing. Next, around 150 mg of the mixture was placed between two PVA-coated glass plates with pieces of Scotch tapes glued onto the edges to serve as spacer and then sheared to get the proper alignment. Both PVA alignment layers and shearing approaches were employed to align the viscous mixture. The cell was then placed on a 22 °C hot plate for 2 h and then 55 °C for 5 h to finish the thiol-acrylate polymerization. The cell was then immersed into water at 50 °C for around 5 h to open the cell and obtain a freestanding film. The film was then mechanically deformed to program the desired shape, color, and reflectivity, and photo-polymerized in a nitrogen box with UV light using an Omnicure S2000 UV lamp at an intensity of 15 mW cm⁻² for 10 min.

The Preparation of 3D Objects: The film after thiol-acrylate polymerization was first stretched and then pressed between a positive and negative mold of the same object. The film was then removed from the mold and photo-polymerized for 10 min under UV light in a nitrogen box. The positive “beetle” mold was 3D printed using polyactic acid (PLA) with an Ultimaker 3 3D printer. The positive “smiley face” mold was made in copper. The negative molds were created from pressing the positive molds into polydimethylsiloxane (PDMS) precursor, which was then cured at 80 °C for 2 h.

Characterization: The transmittance spectra with unpolarized light and circularly polarized light were measured using an Ocean Optics spectrometer attached to the POM, where the objective used is 20x with a numerical aperture (NA) of 0.40. A linear polarizer was combined with a quarter-wave plate to generate circularly polarized light. The temperature of the sample was controlled by a Linkam THM5600 hot stage. Reflectance spectra with circularly polarized light were measured on a Perkin Elmer Lambda 750 UV/Vis/NIR spectrophotometer equipped with a linear polarizer in combination with a quarter-wave plate. Photographs were taken with an iPhone 7 and a Sony Cyber-shot camera. FT-IR spectrum was measured on a Varian 670-IR FT-IR spectrometer used in transmission mode. DSC curves were measured with a DSC Q1000 from TA Instruments. A rate of 10 °C min⁻¹ was used for both heating and cooling ramps. POM images were taken using a Leica CTR6000 polarized optical microscope in transmission mode, equipped with a Leica DFC 420C camera. Temperature was controlled using a Linkam hot stage. Films were positioned on their side and the thicknesses of the films were monitored with the POM. Photographs of the films were taken through LCP and RCP at different temperatures to determine the temperature responsive polarization change of the films.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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