Oscillating Chiral-Nematic Fingerprint Wipe Away Dust

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This work presents an approach to create mechanical undulations at a solid organic coating surface under the influence of an electric field. The coating is fabricated through polymerization of chiral reactive mesogens aligned in their fingerprint mode on top of interdigitated electrodes. The fingerprint mode gives a corrugation of the surface perpendicular to the helix axes. When a lateral alternating electric field is applied, the order parameter of the helicoidally packed mesogens is reduced. This simultaneously leads to an inversion of the fingerprint heights, an overall thickness increase, and a chaotic and fast surface oscillation. These three effects work in concert stimulating wavy deformation figures at the coating surface. The process is fast and reversible; the dynamics of the topographic textures stop immediately when the electric field is switched off. The continuous generation of surface undulations sustains transport of species at the coating surface. It removes dust and debris providing an active dust control.

Artificial self-cleaning materials and surfaces are widely studied and applied in commercial coatings,[1,2] often they are inspired by nature,[3,4] such as the self-cleaning qualities of plant leaves[4,5] or the dirt-rejecting properties of animal skins.[6] In general, water wettability, either (super-)hydrophobic[7,8] or (super-)hydrophilic,[9] attributes to the debris rejecting behavior. Learning from nature, man-made self-cleaning surfaces combine delicate and complex topographic architectures with special chemistry.[10] With the assistance of water, for instance coming from rain, the surfaces clean themselves efficiently by rolling droplets taking up the dirt.[2,11] However, in dry and dusty environments the of the surface topography. The induced vibrations mechanically transport dusts and debris to the edges of the surface, eventually supported by gravity when the surface is tilted. In a previous publication, we reported on homeotropic nematic liquid crystal network (LCN) which we could bring into a volumetric vibration by a high-frequency AC electric field.[12] The dynamic topographies were irregular and rather small, in the order of 100 nm. In order to create larger deformations, we focus here on a chiral-nematic LCN that is brought in the so-called fingerprint mode with periodically alternating planar and homeotropic molecular alignment. The electrically induced change of the order parameter, with periodic alternating contraction and expansion, is anticipated to enhance the surface deformation to dimensions capable to transport sand grains. In addition, the topographic periodicity will be determined by the pitch of the molecular helix rather than by electrode spacing.

The particle-rejecting coating is made by in situ photopolymerization of a mixture of chiral reactive mesogens that forms a crosslinked LCN on top of interdigitated electrodes (IDE). We chose for the IDE as it provides an in-plane electrical field while the LC coating will form an electrical isolating shield protecting potential users. By controlling the surface conditions of this IDE substrate prior to polymerization as described in Experimental Section the rod-like mesogens in the LCN adapted a helicoidal organization with the helix axes parallel to the surface as schematically shown in Figure 1a. The helix axes take a worm-like configuration (Figure 1b) also known as fingerprint texture. The black area in Figure 1b corresponds to the molecules that are homeotropically aligned while the planar orientation is optically birefringent showing blue color. More fingerprints patterns are anticipated to enhance the surface deformation to dimensions capable to transport sand grains. In addition, the topographic periodicity will be determined by the pitch of the molecular helix rather than by electrode spacing.

The underlying deformation mechanism is to decrease the LCN order parameter and bringing the LCN into oscillation by a lateral access to water is often limited and water-based self-cleaning approaches are not applicable. We report here a new concept of a dry self-cleaning method without the aid of water. This will solve a problem for solar farms placed in deserts where accumulated dust and sand at the surfaces of the photovoltaic cells block the sun light, which might cut power production as a result of a sand storm by 40%.[12] For reference, contaminated solar panels in solar farms are still removed from deposited dust by manually brushing. In addition, dust mitigating technologies may be used to protect any mechanical/optical/electric systems that carry out Lunar/Mars missions against dust hazards.[13] The water-free self-cleaning philosophy we propose here is based on an orchestrated change...
alternating electric field (AC). Realignment of the LCN is prohibited by the high crosslink density of the polymer network. Subsequently, it creates microscopic molecular voids and related geometrical deformations in the polymer film as shown in Figure 1c. To enhance the interaction with the field, we designed a mixture that consists of mono- and diacrylates exhibiting a net positive dielectric anisotropy (Figure 1d). Monomers 1 and 2 possess a nematic phase in which the molecules tend to align uniaxially along the common director. Their ratio is optimized to obtain the preferred responsive behavior of the solid polymer network with respect to the deformation amplitude, speed, and reversibility. Monomer 3 is added to induce large positive dipole moment in the polymer network by its polar cyano end group. Chiral monomer 4 induces the chiral-nematic phase in the mixture that forms the molecular helices. Photoinitiator 5 initiates the polymerization process upon UV illumination. The sample preparation details are illustrated in Experimental Section.

The LCN coating is fabricated through spin-coating of the monomer mixture (Figure 1d) from solution on the glass substrate with the IDE electrode pattern (Figure 1e; Figure S3, Supporting Information) and subsequent polymerization by UV light. The in-plane electric field to address the oscillation of LCN coating is provided by the indium tin oxide (ITO)-based IDE. Each electrode is 3 µm wide and adjacent electrodes are separated by a gap of 5 µm.
To gain insight into the electric field distribution over the LCN, we carried out a finite element simulation which shows that at a film thickness of 4 µm, the electric field strength is still 50% of its value near the substrate (Figure 1f; Figure S4, Supporting Information).

The surface topographies and their corresponding deformation dynamics are characterized by digital holographic microscopy (DHM). Looking to the surface of the monomer mixture in its fingerprint state, it exhibits a worm-like undulation with a periodicity equal to half of the pitch of the cholesteric helix. In literature, this is described to be caused by a difference in surface tension between the planar and the homeotropic area known as the Marangoni effect.[23] In fact, in our LC phase also additional effects play a role related to the intrinsic elastic behavior of the monomeric liquid crystal that discourage splay and bend deformation of the twisted molecules. During polymerization, the surface undulation is frozen-in with the remark that also here other effects will play a role. For example, polymerization shrinkage is anisotropic and higher in the homeotropic regions promoting the planar area to protrude. This effect is competing with the Marangoni effect, however the results as shown in Figure 2a suggest that the Marangoni effect observed in the monomer is the most dominant with the

![Figure 2](image-url)

**Figure 2.** Actuation of the surface topographies. 3D images measured by DHM showing the surface topographies a) before and b) during electric actuation. Position 1 points the planar molecular orientation and position 2 is homeotropic orientation. c) Corresponding 2D cross-section profiles of (a) and (b). The zero height corresponds to the initial average of corrugations. d) Thickness increase upon actuation measured by white light interferometer. e) Time-dependent kinetics of the height changes in both planar and homeotropic regions as the AC field is switched “on” and “off” for three cycles. The arrows indicate the moment that the electric field is turned “on” and “off.” f) A close-up view on the chaotic oscillation at those two locations.
highest regions found at the location where the molecules are oriented perpendicular to the substrate. Upon applying an electric field, we observed that the hills contract into a valley, and the initial valleys expand forming hills. Consequently, the surface structure is completely inverting as presented in Figure 2b. The dynamics of this process is shown in Movie S1 of the Supporting Information.

We explain this observation as follows. Placing the LCN coating in an alternating electric field, the dielectric interaction between the field and the LCN stimulates the oscillating vibration of the network. As demonstrated in uniaxially aligned LCN,[24] the molecules start to deviate from their initial highly ordered and compact state. This leads to order parameter reduction[25] and, simultaneously, excessive volume formation. The decrease in order parameter induces anisotropic stresses and potentially leading to deformations.[26–30] However, in a flat uniaxial-oriented coating, the lateral geometric changes are prohibited by its substrate. It only manifests itself as a uniform height increase of the materials between the electrodes. But in the helicoidally ordered chiral system we can observe the surface corrugations. Following the conventional deformations in oriented liquid crystal polymer films, shrinkage occurs parallel to the director and expansion perpendicular to it. Translated to our coating, the perpendicularly aligned volume will shrink in the z-direction while expanding in lateral direction, building up stresses in the adjacent planar area. The adjacent planar domains simultaneously absorb the stresses by expanding in the z-direction on top of their order parameter related expansion. This leads to the inversion phenomena shown in Figure 2a,b. Typically, the coating is actuated at room temperature under an alternating root mean square (rms) voltage of 80.6 V (electric field \( E = 16.1 \text{ V}_\text{rms} \text{ mm}^{-1} \)) at the frequency of 900 kHz. The corrugation inversion clearly seen in Figure 2c reaches between 250 and 300 nm in a 4 \( \mu \text{m} \) thick coating, which is around 6% of the initial coating thickness. We have investigated the actuations in detail by characterizing several parameters including temperature, frequency, electric field strength, and mechanical properties. Related results are given in Figures S5–S9 of the Supporting Information.

Besides the anisotropic deformations, the network vibration also induces dynamic free volume between the rod-like molecules and creates an overall density reduction by nonoccupied volume.[31] As long-range expansion is restricted by the strongly adhering substrate the excessive volume can only escape at the coating surface resulting in an overall thickness increase. Figure 2d shows an average thickness increase of 100 nm measured in the vicinity of a scratch in the film, which is around 2.5% of the initial coating thickness. A value which is of the same order as published earlier for light induced vibrational effects and adds to corrugation inversion.[24]

We further investigated the time-dependent electromechanical response of the LCNs. We explicitly selected two distinguished positions in both a homeotropic and a planar region, as indicated in Figure 2a. The response time of activation and relaxation in both regions are in the order of 10 s, which is much slower than the frequency of the applied alternating electric field (900 kHz) and the on/off switch of the field, which takes place instantaneously. This implies that the deformation kinetics is mainly governed by the polymer viscoelasticity, rather than by the dielectric properties of the LCN. Figure 2e presents three reversible switching cycles.

The kinetic measurements from DHM reveal a tertiary chaotic oscillation superimposed on the electric activated area. This was earlier observed for a homeotropically aligned LCN where volume effects were dominating.[14] To demonstrate that they also occur in this specific case, we zoom in at one arbitrary selected profile for both a planar and a homeotropic region and followed their height changes evolving in time (Figure 2f). Without applying the electric field, the surface deforms and starts to oscillate around the formed profiles. We use the standard deviation (SD) value to quantify the amplitude of the oscillation. Generally, a low value indicates that the data is tightly clustered around the mean, while a large standard deviation indicates the data spreads away from the mean value. In our sample, the SD value as derived from Figure 2f is 3 nm for the inactivated surface. Upon actuation, it increases to 16 nm. A further closer view on the oscillations superimposed on the activated protrusions/indentations suggests that they vibrate in a chaotic manner. Both the amplitudes and the frequencies develop irregularly under the continuous AC field. The chaotic oscillation continues until the electric field is switched off after which they immediately stop, even before the formed topographies relax to the initial states. We postulate that the electrically induced chaotic oscillations are related to some natural frequency occurring in the system. We previously found that volume expansion is enhanced when resonance occurs between the driving frequency and the system eigenmodes.[24] Upon deformation conditions change and the system tries to find a new equilibrium, which in term changes the resonance conditions.[32] This continuous loop leads to chaotic fluctuations until the electric field is stopped.

Next, we analyze the interaction between the electric field and the molecular orientation into more detail. When a low molecular weight nematic LC is subjected to an electrical field \( E \), the LC molecules will re-orient. This is the basis for many electro-optical applications. In the case of our LCNs full reorientation is prohibited by the network. Nevertheless, a dielectric torque is exerted on the mesogens, which is proportional to the dielectric anisotropy \( \Delta \varepsilon = \varepsilon_\parallel - \varepsilon_\perp \) (\( \varepsilon_\parallel \) and \( \varepsilon_\perp \) are the principal dielectric permittivities referred to the director \( n \)) and quadratic in \( E \). The orientation (polarization) of permanent dipoles, as for instance in the cyano groups of the dangling side groups, involves cooperative motions of molecular segments in our viscoelastic medium with timescales of the order of the field frequencies. Not only this will result in an overall small decrease of the order parameter but it is also anticipated to exert mechanically coupled forces on the acrylate main chains of the LCN. In the particular system under investigation there are a few complexation factors. The dielectric permittivity is frequency dependent, which alters the dielectric torques when the frequency changes. Moreover, the director of the fingerprint cholesteric is complex, not only by the twist in the molecular helix but also by the random figures of the helix axes as shown in Figure 1b. The maximum torque occurs when the electric dipole direction is perpendicular to the AC field while it becomes zero when the dipole is parallel to the electric field. Figure 3a,b illustrates this effect on a molecule with its dipole moment parallel to its long
axis. However, in a fingerprint texture that is made without additional directional forces, the directions of the helix axes distribute randomly as shown in Figure 1b. To analyze the interaction between the electric field and the LCN, we need to control the formation of fingerprint in a preferred direction with respect to the electric field lines. This is achieved by exerting a unidirectional low-pressure buffing on the polyimide alignment layer prior to processing the monomer mixture. Consequently, regular linear patterns appear as shown in Figure 3c,d. In Figure 3c the helix axes are parallel to the AC field line and consequently all mesogenic entities are perpendicular to the field. Under the polarized optical microscope, we observe this as line structures orthogonally to the electric field. While in Figure 3d molecules rotate along the helix axes perpendicular to the field lines and the line patterns are parallel to the field. The bright blue lines correspond to molecules planar aligned and the black lines are molecules homeotropically orientated. Activating under an AC electric field, line structures in Figure 3c exhibit large deformations as nearly all the positive dipole molecules are perpendicular to the electric field line (Figure 3e,g), which couple a net maximum torque to the LCN. Whereas, the line textures in Figure 3d show minor surface changes (Figure 3f,h).

Figure 3. Torque exerted on the molecules with respect to the electric field and dipole. Schematic representation of molecule with positive dipole moment experiences a) largest torque when it is perpendicular to the applied electric field, and b) smallest torque when it is parallel to the AC field. Optical microscope image of a regular grating texture observed between crossed polarizers, line patterns are c) perpendicular to the electric field, and d) parallel to the electric field. Insets present the rotating direction of the helix and the IDE substrate. e,f) The corresponding surface profiles of the initial nonactuated state (blue line) and actuated (red line) of (c) and (d), respectively. g,h) 3D images of the activated topographies of (c) and (d), respectively.
This is due to the angle between positive dipole molecules and the electric field varying from 0 to $\pi/2$, resulting in the torque ranging from zero to maximum.

For potential applications, it is of interest whether the dynamics of the surface topographies can be utilized to transport particles placed on top of the film. Figure S10 (Movie S3) of the Supporting Information shows that an electrically activated surface is capable to shift few isolated sand grains over the entire coating surface. Based on this, we studied the coating further toward a practical application by investigating self-cleaning properties under dry conditions, i.e., without the help of water. Currently, most self-cleaning solutions are based on lotus flower effects utilizing superhydrophobic surface properties, which need water to clean. However, there are very limited self-cleaning strategies developed for dust contamination in a dry environment,[33] for example, solar panels placed in deserts or, somewhat more futuristic, light-driven vehicles operating on other planets. Our LCN coatings enable the cleaning under dry conditions where particles are removed mechanically by surface vibrations. For this purpose, we take the advantage of the gravity by tilting the coating somewhat from its horizontal position. In this specific experiment, the tilt angle is kept at $37^\circ$. In Figure 4a(1), the sands cover the entire active area of the coating. The sand grains have typical dimension 0.25–0.55 mm (microscopic images of the sand particles are given in Figure S11, Supporting Information), much larger than the periodicities of the corrugations or the electrodes. Directly upon switching on the electric field (Figure 4a; Movies S4, Supporting Information), nearly 100% of the sands in the activated area are removed within the first cleaning sequence in 1 s. The fast cleaning speed is attributed to the chaotic oscillatory surface fluctuations when coating is actuated. The electric energy

Figure 4. Snapshots of sand being removed from the surface through the dynamic dry self-cleaning approach. The sizes of sand particles are around 0.25–0.55 mm in diameter. a) (1) Tilt angle of the sample. (2)–(4) Dry sand is removed within 1 s upon switching on the electric field. b) (1) Tilt angle of the sample. (2)–(4) 10 wt% humid sand sticks to the coating surface, also when the field is on. c) (1) Sample is placed at $42^\circ$. (2)–(4) Removing of the 10 wt% humid sand on the fluorinated coating within 3 s after switching the field on.
consumed for this cleaning cycle is small and calculated as 28.6 mJ cm\(^{-2}\) (Figure S12, Supporting Information).

Humid dust has proved extremely difficult to remove due to the presence of capillary forces between particles mutually and between the particles and the surface.\(^{[34]}\) Figure 4b(2)–(4) shows humid sand grains obtained by adding 10 wt% water. The wet sands adhered firmly to the coating surface after switching on the electric field, even when the surface was tilted further toward vertical 42° (Movie S5, Supporting Information). We now enhance the cleaning efficiency of our approach further by lowering the surface energy of the coating by modification with a fluorinated top layer. For this purpose, we functionalize the coating surface by 1H,1H,2H,2H-perfluorodecyltriethoxysilane through chemical vapor process (details in Experimental Section). The contact angle is increased from 73° to 116° (Figure S13, Supporting Information). This hydrophobic surface reduces the adhesion of the sand, especially for the wet/humid grains. Figure 4c(2)–(4) shows that on the hydrophobic coating, the 10 wt% humid sands are immediately removed upon switching on the electric field within 3 s (Movie S6, Supporting Information).

In conclusion, we have developed a coating that forms dynamic surface undulations fuelled by an alternating electric field. This new approach is based on resonance enhanced microscopic (di)electric coupling of polar mesogens to the electric field. We illustrate three orders of dynamics occurring spontaneously. The first order is the formation of topographies as the external field is switched “on,” which reaches close to 6% of the coating thickness. Simultaneously, the coating thickness increases 2% as the results of volume expansion. Maintaining the electric field, a tertiary oscillation evolving in both height and lateral dimension starts to occur. Both the field-on and the field-off responses occur in seconds presumably determined by the viscoelastic properties of the polymer.

The coating provides a self-cleaning action that actively repels particles under both dry and humid conditions efficiently through its dynamic oscillating surface topographies. The mechanism is different from alternative techniques based on electrostatic repulsion which require longer times and higher voltages. The technology that we propose here can be easily integrated into applications through the existing fabrication processes that are already established in liquid crystal display (LCD) industries. The flexibility of combination of both dry and wet self-cleaning principles allows the coating to function under various conditions. The coating has several other advantages: low energy consumption, high transparency to the visible spectrum of light (Figure S14, Supporting Information), and they are based on reactive liquid crystal mesogens, which have a proven lifetime in the LCD industry. Of course, outdoor durability still needs to be optimized and the coating will require protection against abrasion of sand during a sand storm. This is part of our present research. We anticipate this coating can be readily implemented in the dry and dust area where water is pricy or not available for cleaning.

**Experimental Section**

**Materials:** IDE substrates were kindly provided by Merck. Monomers 1–3 were purchased from Merck Chemical Company. Chiral dopant 4 (Parliocolor LC756) was obtained from BASF and photoinitiator 5 (Irgacure 819) was obtained from Ciba company. For a typical sample, 20 pbw monomer 1, 45 pbw monomer 2, 33 pbw monomer 3, 2 pbw photoinitiator 5, and various amounts of chiral dopant monomer 4 ranging from 0.1 to 1 pbw were used. The amount of chiral dopant was varied to prepare samples with different pitches. 1H,1H,2H,2H-perfluorodecyltriethoxysilane was purchased from Sigma-Aldrich.

**Sample Preparation:** The glass substrates with patterned ITO were cleaned by ultrasonication for 20 min in acetone and isopropanol, respectively, and followed by drying with nitrogen flow. The cleaned substrates were then subjected to a UV-ozone treatment for 20 min. Perfluorodecyltriethoxysilane is used to obtain homeotropic alignment. In detail, the polyimide was spin-coated onto the substrate at a speed of 5000 rpm for 40 s. It was baked at 100 °C for 10 min to evaporate the solvent and then at 220 °C for 90 min. To fabricate liquid crystal polymer coating, the liquid crystal monomer mixture was spin-coated at 1000 rpm (acceleration rate 300 rpm s\(^{-1}\)) from tetrahydrofuran (25 wt%) solution onto IDE substrate and subsequently polymerized by UV illumination at 21 °C for 30 min in N\(_2\) environment using a mercury lamp (Omnicure EXFO S2000).

The fingerprint pattern is formed in the monomer state by a delicately balanced force between helical twisting power that rotates molecules along the helix axis (induced by chiral dopant 4) and the strong anchoring force exerted by the substrate (from homeotropic polyimide) that unwinds the helix to align homeotropically. Consequently, the helix axis is brought parallel to the substrate and a full \(2\pi\) molecular rotation describes the chiral pitch as shown in Figure 1a. Details on the helix pitch are elaborated in Figure S1 of the Supporting Information.

The hydrophobicity of the coating surface was introduced by using a vapor phase reaction by 1H,1H,2H,2H-perfluorodecyltriethoxysilane at 125° for 1 h. Prior to the silanization, the sample was treated with UV-ozone for 20 min.

**Characterization:** The fingerprint configuration was checked by optical polarized microscope (Leica DM2700). The alternating electric field with sinusous wave function was provided by a function generator (Tektronix AFG3252C). The electric signal from function generator was then amplified through an amplifier (Falco Systems WMA-300). The output voltage was measured by the oscilloscope (Keysight InfiniiVision DSO-X 3032T). The surface topographies were measured by Digital Holography Microscope (Lynée Tec.). Thickness of samples was measured by interferometer (Fogale Nanotech Zoomsurf) and profiler meter (Bruker Dektak XT). UV–vis spectra of the coating were characterized by Ocean Optics HR 2000+. The profile of the water droplet on the liquid crystal coatings was measured by a contact angle device (Dataphysics OCA-20). The mechanical properties (e.g., storage modulus and loss modulus) of the LCN film modulus were measured with a Dynamic Mechanical Thermal Analysis (Q800, TA instrument) with a ramp rate of 3 °C min\(^{-1}\).

**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.

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