Spontaneous light-induced Turing patterns in a dye-doped twisted nematic layer

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Optical pattern formation is usually due either to the combination of diffraction and nonlinearity in a Kerr medium or to the temporal modulation of light in a photosensitive chemical reaction. Here, we show a different mechanism by which light spontaneously induces stripe domains between nematic states in a twisted nematic liquid crystal layer doped with azo-dyes. Thanks to the photoisomerization process of the dopants, light in the absorption band of the dopants creates spontaneous patterns without the need of temporal modulation, diffraction, Kerr or other optical nonlinearity, but based on the different scales for dopant transport processes and nematic order parameter, which identifies a genuine Turing mechanism for this instability. Theoretically, the emergence of the stripe patterns is described on the basis of a model for the dopant concentration coupled with the nematic order parameter.

Non-equilibrium processes often lead to the formation of spatial periodic structures developed from a homogeneous state through the spontaneous breaking of symmetries¹⁴²⁴. This self-organization usually is a consequence of the force imbalance or transport optimization of energy, momenta and/or particles. Initially, these patterns were understood as the saturation of linear normal modes using nonlinear effects⁵⁴⁴. Hence, the characteristic length of these patterns is determined by the geometrical dimensions of the system under study. Classic examples are the Benard and Taylor-Couette patterns⁴. Another mechanism proposed to understand the pattern formation is based on the difference in transport or coupling processes in chemical reactions, known as Turing instability⁵. These patterns, Turing patterns, are characterised by having an intrinsic characteristic length, which is not determined by external factors or geometrical dimensions but by the diffusive coefficients and temporal scales of the system under study. This mechanism has been applied from biology to optics, passing through chemistry and physics⁵⁶⁶³⁴. An example of pattern formation in liquid crystals is the electroconvection⁸, which results in charge transport and convection effects. Similar patterns are observed in thin liquid crystal layers close to nematic-smectic transition⁹–¹¹, thin hybrid nematic layer¹² and polymer nematic liquid crystal¹³, where one of the elastic constants is much larger than the other ones. In the case of light and matter interactions, the spontaneous transverse optical grating formation has been observed in slightly asymmetric single-feedback mirror experiments using nematic liquid crystals as nonlinear optical media¹⁴. These patterns come from the interference between counter-propagating light in a nonlinear medium, that is, diffraction transforms phase variations into amplitude modulations while the optical nonlinearity converts amplitude into phase modulations. Likewise, patterns have also been observed by temporal modulation of light in photosensitive chemical reactions¹⁵. Light-induced effects in absorbing liquid crystals attracted a considerable interest for their potential applications in optical switching and image storage¹⁶. There are a number of phenomena connected with excitations of liquid crystal molecules or of dye-dopants added to a liquid crystal matrix host. Photoinduced conformational transformations, such as trans-cis isomerization of azo compounds, can change the orientational order parameter (see textbook¹⁶ and reference therein), influence the chiral properties¹⁷, or induce phase transitions¹⁸¹⁹. In this letter, we show that stripe domain patterns between different nematic states (i.e. molecules that are locally alternated between regions of higher and lower orientational order) can spontaneously arise in a dye-doped twisted nematic liquid crystal layer when illuminated under appropriate conditions. In this case, the pattern formation

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is not mediated by the light diffraction or the temporal light modulation, neither by the Kerr or other optical
nonlinearity, but originates from the different scales for dopant concentration and order parameter transport
process, hence, identifying a novel mechanism of light-induced Turing instability. Experimentally, we consider a
twisted nematic liquid crystal cell, namely, the liquid crystal molecules have mutually orthogonal planar anchor-
ing onto the two glass substrates that constitute the confining walls of the cell. When the liquid crystal sample
is illuminated with a linearly polarized Gaussian beam at a wavelength inside the absorption band of the dopants,
for a certain critical input power value, a transition from the homogeneous nematic state to a spatially modu-
lated one with a striped structure is observed (cf. Fig. 1). This structure accounts for a spatial modulation of the
nematic liquid crystal molecular orientation. Noticeably, the orientation of the stripes is perpendicular to the
linear polarization of the illuminating light. Theoretically, we are able to describe the emergence of the stripe
patterns on the basis of a model for the concentration of azo-dye dopants in the excited state (cis-state) coupled
with the order parameter of the twisted nematic layer. This model allows us to identify the mechanism of pattern
emergence, which is due to the different scales for transport processes of dopants and order parameter, i.e. it cor-
responds to a Turing instability. Adiabatically, by eliminating the dopant concentration, the order parameter ful-
fills a Turing-Swift-Hohenberg type equation, which allows us to determine analytically the instability criterion.
The Turing-Swift-Hohenberg equation is a paradigmatic model for pattern formation in several contexts, such as
hydrodynamics, chemistry, plant ecology, nonlinear optics, and elastic materials4.

Results

Light-induced pattern formation. Applying a light beam with wavelength in the absorption band of the
methyl red20 creates the gradual emergence of stripe domains between nematic states, first in the central region of
the illuminated area, then, the domains invade the whole illuminated zone. This process takes about three hours
to invade an area of 3 cm². After a while the different domains are merged generating several defects; mainly dis-
locations are identified, which are characterized by joints, that is, locally, regions with different wavenumbers3.
Figure 1 shows the typical stripe patterns with several dislocations observed. It is also observed that if the sample
is illuminated with a wavelength which is not in the absorption band of the dye, the pattern does not emerge. In
particular, samples of dye-doped liquid crystal with identical configurations illuminated with red light (He-Ne
Laser @ \( \lambda = 633 \) nm and \( I = 35 \) mW) at similar power do not exhibit any effect.

Noticeably, the patterns are mostly oriented orthogonally to the direction of the light electric field. Indeed,
when the electric field is rotated the patterns are reoriented in the direction orthogonal to it. Notice that the
pattern wavelength (around 35 \( \mu \)m) does not correspond to the cell thickness, but it is on the same order of
magnitude.

The light intensity emerging from the dye-doped nematic liquid crystal sample accounts for the molecular
orientation, which is characterized by transversal spatial oscillations. Fig. 2(c) schematizes the molecular rear-
nangement in an intermediate plane of the sample.

As the sample is not illuminated, the dye molecules are in the trans state and oriented along the liquid crystal
nematic director21. When the sample is illuminated by a light beam in the absorption band of the dopants, the
scenario changes because the light induces a photo-isomerization process and the dopants undergo a transition.
from the trans to the cis state, corresponding to different molecular configurations of the azo-dyes\textsuperscript{16,21}. Likewise, the dyes in the cis state produce the reorientation of the liquid crystal molecules. However, the liquid crystal molecules are oriented in different directions. Hence, this transition is characterised by a decrement of the liquid crystal molecular order—which corresponds to an entropic effect—and, correspondingly, by the modification of the average refractive index \( n \) of the sample\textsuperscript{21}, which is averaged along the longitudinal direction \( z \), so that the average refractive index \( \bar{n} \) remains a function of the cross-sectional coordinates \((x, y)\). This index has the form\textsuperscript{16,22}

\[
\bar{n}(x, y, z) = \frac{1}{d} \int_{0}^{d} \frac{n_{o}n_{e}}{\sqrt{n_{o}^{2}\cos^{2}\theta(x, y, z) + n_{e}^{2}\sin^{2}\theta(x, y, z)}} \, dz,
\]

where \( d \) is the thickness of the sample, \( n_{o} \) and \( n_{e} \) are the ordinary and extraordinary refractive index of the liquid crystal respectively, and \( \theta(x, y, z) \) is the average angle between the molecules and the horizontal axis of the sample. For example, if the molecules are oriented in the plane of the sample \( \theta = 0 \) and \( \bar{n} = n_{o} \). Indeed, modifications of molecular order produce a change in \( \bar{n} \). Note that clearer stripes (darker stripes) in Figs 1 and 2(b) account for regions where molecules are more ordered (disordered).

To characterize the molecular orientational configurational structure of the observed patterns, we have analyzed the sample by rotating \( P_{2} \) polarizer (analyzer) concerning to \( P_{1} \) polarizer, which is a standard technique for characterizing liquid crystal textures\textsuperscript{23}. When the dye-doped twisted liquid crystal sample is between two parallel polarizers, that is, the angle between polarizers is 0\(^{\circ}\), the light that crosses the sample exhibits a striped pattern with a different tenuous intensity. Fig. 3(a) shows the typical picture observed in this configuration of the polarizers. The stripes and between stripes have different colors. Hence, one infers that these regions have a different average refractive index. When the analyzer is rotated with respect to the first polarizer, the image darkens slightly and the patterns continue to be observed (see Fig. 3(b)). When the polarizers are orthogonal, the angle between the analyzer and the first polarizer is 90\(^{\circ}\); one continues to observe the pattern with darker browns (cf. Fig. 3(c)). This analysis is a rigorous proof that the material is birefringent. Note that the stripes are brown and not black. Indeed, the light crosses these regions. Therefore, both the stripes and between stripes the material are liquid crystal phase with a different average refractive index. This index is spatially modulated.

**Theoretical description.** To describe the dynamics of the twisted nematic liquid crystal layer, one can introduce a scalar order parameter \( S(\mathbf{r}, t) \) that accounts for the alignment of the molecules along the director

**Figure 2.** Stripe domain induced by photo-isomerization in a dye-doped twisted nematic liquid crystal cell. (a) Schematic representation of the twisted molecular configuration for a small portion of the unenlightened cell. (b) Experimental stripe domain induced by photo-isomerization of the dyes. (c) Schematic representation of an intermediate plane of the cell of the molecular configuration for a small portion of the enlightened cell.
Eqs (2) and (3) is rewritten as
\[
\partial_t S(\vec{r}, t) = -\bar{A}S + BS^2 - ES^3 + d \nabla^2 S - DS (\nabla \phi)^2,
\]
and
\[
S(\partial_t \phi(\vec{r}, t) = DS \nabla^2 \phi + 2D \nabla S \cdot \nabla \phi,
\]
where \(\bar{A}, B\) and \(E\) are parameters that characterize this transition and \(D\) stands for the elastic coupling (see the textbook\(^{23}\) for details of the parameters). A twisted cell of thickness \(h\) is characterised by the boundary conditions \(\phi(x, y, z = 0, t) = 0\), \(\phi(x, y, z = d, t) = \phi_0\) and \(d\) is the thickness of the liquid crystal layer (see Fig. 3). The cell that we have considered in our experiment is characterised by having \(\phi_0 = \pi/2\). To describe our system, we consider that the liquid crystal cell is sufficiently thin and that the director rotates uniformly from one plate to the other. Under these assumptions, we get \(S(\vec{r}, t) = S(x, y, t)\) and \(\phi(\vec{r}, t) = \pi z/2h\). Note that this solution trivially satisfies Eqs (2) and (3) is rewritten as
\[
\partial_t S(x, y, t) = -AS + BS^2 - ES^3 + D \nabla^2 S,
\]
where \(A = \bar{A} + D(\pi/2h)^2\) and \(\nabla^2\) stands for the laplacian in transversal coordinate. This model predicts that the nematic and the isotropic liquid transition in a twisted cell is of subcritical nature. It is important to mention that the previous model Eq. (4), renormalizing the linear term, also describes planar \((\phi(\vec{r}, t) = \phi_0\) constant) and homeotropic \((\mathbf{n} = \hat{z})\) cells\(^8\). On the other hand, the concentration of molecules in the \textit{cis}-state \(C(\vec{r}, t)\) in a thin layer satisfies a relaxation and diffusion equation of the form\(^{21}\)
\[
\partial_t C = -\lambda [C - C_0(I)] + \delta \nabla^2 C,
\]
where \(\lambda\) is the decay rate related to the transition from \textit{cis} to \textit{trans} state by thermal relaxation, \(C_0\) is the equilibrium concentration of molecules in the \textit{cis} state that is proportional to the total intensity of the incident light \(I\). Precisely, \(C_0(I) \equiv \gamma I/(1 + \eta I)\) where \(\gamma\) and \(\eta\) are dimensional parameters\(^{21}\). \(\delta\) is the diffusion coefficient of the concentration of \textit{cis} state.

The inclusion of dye-dopants increases the nonlinear response of liquid crystals under the excitation of external fields\(^{16-19,21,25}\). Indeed, the behavior of liquid crystals changes drastically when dopants are considered. To describe the pattern formation induced by the photo-isomerization process in a dye-doped twisted nematic layer, let us consider the concentration of molecules in the \textit{cis}-state \(C(\vec{r}, t)\) and the scalar order parameter \(S(\vec{r}, t)\), which satisfy the rate equations

\[\text{Figure 3. Stripe domain induced by photo-isomerization in a dye-doped twisted nematic liquid crystal cell between polarizers } P_1 \text{ and } P_2 \text{ with different relative angles. The upper and bottom panels, respectively, account for the observed snapshots and schematic configuration of the polarizers and liquid crystal sample. (a) Both polarizer are parallel, the relative angle between polarizers is } 0^\circ, \text{ (b) the relative angle between polarizers is } 45^\circ, \text{ and (c) both polarizers are crossed, that is, the relative angle between polarizers is } 90^\circ.\]
For a weak anisotropy, the bistable states are 

\[ S, \ C = (S_{00}, C_0) \equiv (0, C_0) \quad \text{and} \quad (S, C) = (S_{11}, C_1) \equiv (1, C_1) \begin{pmatrix} \alpha \beta + B + \sqrt{(\alpha \beta + B)^2 - 4(A + \beta C_0)} \end{pmatrix}/2, C_0 - \alpha S_0 \]

which account, respectively, for an isotropic liquid and a dye-doped nematic phase. For small coupling \( \alpha \) between the order parameter and the \( \text{cis} \) concentration, the stable nematic phase corresponds to \((S_{11}, C_1)\). By increasing the coupling parameter, the homogeneous nematic phase becomes unstable, giving rise to the emergence of striped domains. Namely, the order parameter \( S \) exhibits spatial modulation. Figure 4 shows the typical observed pattern when a weak anisotropy is considered (\( \delta_2 \approx \delta_1 \)) and the associated bifurcation diagram. Note that the texture observed experimentally shows a good agreement with that observed from model Eq. (6). Hence, the order parameter \( S \) and concentration of dopants \( C \) exhibit spatial oscillations along the direction of the light electric field. This physically means that molecules locally alternate between regions of higher and lower orientational order, as observed experimentally shows quite a good agreement with that observed from model Eq. (6). Hence, the order parameter \( S \) and concentration of dopants \( C \) exhibit spatial oscillations along the direction of the light electric field. This physically means that molecules locally alternate between regions of higher and lower orientational order, as seen in Figs 1, 2(b) and 4.

Figure 4 shows the real part of the growth rate, \( \text{Re}(\sigma) \), as a function of the wavelength modulus \( k = k_0 \) for fixed parameters at the spatial bifurcation (\( \alpha = \alpha_c \)) and below the spatial instability (\( \alpha < \alpha_c \)). Note that the spatial...
instability of the homogeneous nematic phase occurs while increasing $\alpha$ above a critical value $\alpha_c$. We then consider the anisotropic case ($\delta_i \neq \delta_j$) of model (6). In this case, the instability occurs in the most unstable direction on wavenumber space, which depends on the relative values of $\delta_i$ and $\delta_j$, so that, beyond the instability the stripes will be along the direction of the smallest diffusion coefficient, which corresponds to the direction orthogonal to the light electric field ($\hat{\epsilon}_i > \hat{\epsilon}_j$). Hence, we can conclude that the differences of scales of transport and relaxation processes for the order parameter $S(r^i, t)$ and the cis state concentration $C(r^i, t)$ are responsible for the emergence of patterns.

Due to the complex and long expression of the growth rate $\sigma$ as a function of the parameters, the former study is only accessible through numerical analysis. To understand more deeply and analytically the origin of the spatial instability, we consider the extreme limit in which one variable, the cis state concentration, follows adiabatically the isotropic phase ($\delta_i = \delta_j$) of model (6). In this case, the instability occurs in the most unstable direction $\hat{\epsilon}_i$ of dye-doped nematics phase ($S$, $C_\text{cis}$) as a function of the wavenumber in $y$-direction $k_y$ for different values of coupling $\alpha$ parameter and the same other previous parameters. The blue and purple curves correspond to different components of the real part of the growth rate $\sigma$. The solid and dashed curves correspond, respectively, to the growth rate at critical $\alpha \equiv \alpha_c = 4.1$ (spatial instability) and at $\alpha = 1 < \alpha_c$ (below the instability).

\[
\frac{\partial S}{\partial t} = -[A + \beta C_\text{cis}(I)]S + (B + \alpha \beta)S^2 - S^3 + \frac{D^2}{\lambda} \nabla^4 S + (1 - D\alpha)\nabla^2 S - \frac{\alpha}{\lambda} (\delta_1 \partial_{x^1} + \delta_y \partial_{y^1}) \nabla^2 S.
\]

This equation corresponds to a Turing-Swift-Hohenberg type equation $^{4,28}$. This type of model generically describes pattern formation in several contexts, ranging from biology, ecology, chemistry to physics. A necessary condition for the observation of patterns is that the effective diffusion coefficient is negative (anti-diffusion), i.e. $\alpha D < 1$, which, indeed, imposes that the system needs two different transport scales in order to observe the emergence of patterns. By increasing $\alpha$ the isotropic phase exhibits a spatial instability for $4(A + \beta C_\text{cis}) + 5(1 + \beta\alpha)S_\text{cis} = (D^2 - \alpha\delta_i k_y^2)\lambda - (1 - D\alpha)k_y^2$, where the critical wavenumber is $k_y = \sqrt{(1 - D\alpha)/2(D^2 - \alpha\delta_i)}$.

Figure 5 shows a typical stripe domain obtained from numerical simulation of Eq. (7). All numerical simulations presented are obtained by considering finite differences code with Runge-Kutta order-4 algorithm. Therefore, the simple Turing-Swift-Hohenberg type Eq. (7) qualitatively well describes the dynamics of stripe domains observed in the experiments. The theoretical description presented also contains the case of dye-doped planar nematic liquid crystal cells, considering constant $\phi(r^i, t) = \phi_\text{cis}$. However, in this case, the effective linear parameter $A$ in $S(r^i, t)$ is modified ($A = A_\text{cis}$). Besides, one expects that because the dopants are oriented in a single direction the coupling with the light is weaker, that is, the $C_\text{cis}(I)$ parameter must be smaller. Therefore, one requires higher light intensity to induce patterns. Figure 6 shows experimental pattern induced by photo-isomerization in a dye-doped laminar nematic liquid crystal cell. The wavelength observed is smaller than that found in the twisted configuration. However, these observations allow us to conclude that the phenomenon of spontaneous light-induced patterns in a dye-doped nematic liquid crystal layer is robust and does not depend on the particular choice of the liquid crystal alignment.

**Discussion**

In conclusion, we have experimentally shown that linearly polarized light induces stripe domains between nematic states in a twisted dye-doped nematic liquid crystal cell when the intensity is above a critical value. Indeed, light creates spontaneous patterns without the need of temporal modulation, diffraction, Kerr or other optical nonlinearity, but just based on the different scales for dopant transport processes and nematic order parameter, which
identifies a genuine Turing mechanism for this instability. Theoretically, we are able to describe the emergence of stripe patterns and to show that the different scales for dopant transport processes and the order parameter are responsible for their emergence. In the limit where there is a significant timescale separation between variables, a simple Turing-Swift-Hohenberg type model is derived, which allows performing an analytical analysis. Experimentally, we observed rich front dynamics between stripe domains and homogenous nematic phase, showing the system’s capability of responding to the optical addressing and opening novel perspectives in the field of optical control of micro-structured soft matter materials and spontaneously self-organized optical media.

Methods
The nematic liquid crystals are characterised by having a rod-like molecular structure, that is, these molecules are distinguished by having a uniaxial structure. In a temperature range, these molecules are locally aligned forming the nematic phase (thermotropic liquid crystal). To substantially increase the coupling between the light and the nematic liquid crystal dye-dopants are added to a liquid crystal matrix host. Then a requirement is that the dye-dopant molecules have a uniaxial rod-like structure, which is not necessarily a liquid crystal. Likewise, the concentration in weight of the dye-dopant in the liquid crystal must be low in order to not degrade the properties of the liquid crystal and ensure the solubility of the mixture. In the case of E7 liquid crystal and methyl-red dye, the experiments were performed in mixtures in the range of 0.25% up to 1% concentration by weight.

The experimental setup is depicted in Fig. 1. A dye-doped nematic liquid crystal (DDLC) cell subjected to an orthogonal Gaussian laser beam is studied. The cell was filled with an E7 nematic liquid crystal doped with the azo-dye Methyl-Red at a concentration of 0.75% in weight. The elastic constants of the liquid crystal under consideration are, respectively, \( K_1 = 11.2 \), \( K_2 = 6.8 \), and \( K_3 = 18.6 \) (\( N/m^2 \)) and the relative parallel and perpendicular dielectric constants are \( \varepsilon_\parallel = 18.96 \) and \( \varepsilon_\perp = 5.16 \). The cell consists of two glass plates coated with Poly-Vinyl-Alcohol (PVA) and rubbed to favour the planar alignment of the liquid crystal molecules, nematic director parallel to the substrates. The cell is a sandwich type with \( d = 25 \mu m \) thick spacers. The gap is filled with the dye-doped nematics liquid crystal. The transversal region covered by the liquid crystal is a square of the order of 4 cm². The rubbing directions on the glass plates were such to impose twisted anchoring conditions of the liquid crystal molecules, namely, parallel anchoring directions for the molecules on the confining plates, (see Fig. 2).

This type of configuration favours the dopant molecules to be positioned with different orientations, which ensures a relevant coupling with the light that crosses the sample. Figure 2(a) illustrates schematically the molecules when the sample is not illuminated. To induce the patterns, the cell is irradiated with a frequency doubled Nd:YVO₄ laser, with wavelength \( \lambda_p = 532 \ nm \) in the absorption band of the dopants, and with vertical polarization (following \( y \)-axis, cf. Fig. 1). The cell was subjected to input powers between \( P = 100 \) mW and \( P = 200 \) mW. Two plano-convex lenses increase the laser beam diameter to 2 cm. Additionally, two linear crossed polarizers \( P_1 \) and \( P_2 \) are positioned at the input and output of the dye-doped nematic liquid crystal sample, respectively, to analyze the response of the light that crosses the cell. Likewise, the second polarizer \( P_1 \) (analyser) can be rotated with respect to the first polarizer \( P_1 \) to characterise the birefringence properties of the liquid crystal sample. A beam-splitter (BS) is placed in between the liquid crystal sample and polarizer \( P_1 \) to decrease the intensity of light, and thus, to achieve a better image. The transmitted beam is recorded with a CCD camera (Thorlabs DCU224M, 1280 × 1024 pixels).

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Author Contributions

M.C. and M.W. conceived the experiments with inputs from U.B. and S.R., I.A. and G.G. performed numerical simulations with inputs from M.C., I.A. and M.C. worked on theoretical description, G.G. and M.W. conducted the experiments and analysed the results. All the authors discussed the results. G.G., M.C. and M.W. wrote the manuscript with inputs from I.A., U.B., and S.R.

Additional Information

Competing Interests: The authors declare no competing interests.

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