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# Magneto-nematic coupled dynamics in ferromagnetic nematic liquid crystals under magnetic field

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*ABSTRACT:* Dynamic properties of ferromagnetic nematic liquid crystals under an external magnetic field have investigated. The coupled dynamics of the magnetization, $\boldsymbol{M}$, and the director field, $\boldsymbol{n}$, associated with the liquid crystalline orientation order, have studied. In the framework of the hydrodynamic and and thermodynamic approach, the role of the dynamic cross-coupling in macroscopic dynamic behavior both the nematic liquid crystal and its magneto-optic properties is considered. The dependence of the temporal evolution of a magneto-induced response on the dissipative and reversible parts of the dynamic cross-coupling is considered.

*KEYWORDS* Ferromagnetic nematic liquid crystals; magnetization; director field; coupled dynamics; magneto-induced response; magneto-optic effect.

## 1. INTRODUCTION

Ferromagnetic nematic liquid crystals (NLC) are characterized by combination of the orientation structural ordering of the anisotropic (nematic) molecules with the magnetic ordering of magnetic nanoparticles, arched by interphase interaction with the nematic molecules. The strong effect of an external magnetic field on an orientation ordering molecular structure (describing by the nematic vector, $\boldsymbol{n}$) occurs indirectly via its interaction with magnetic moments of nanoparticles. The direct diamagnetic effect of the magnetic field on the molecular orientation is several orders of magnitude smaller than the indirect effect [1-4].

The magnetic field-induced change of the nematic director is accompanied by a corresponding change of the optical axis parallel to it. This is manifested in the magneto-optic effect of the polarization twisting of light passing through the ferromagnetic NLC. The electric field-induced distortion of the orientation ordering molecular structure is accompanied by the corresponding changes of the magnetization. This means existence of the electro-magnetic effect in the ferromagnetic phase of the ferromagnetic NLC.

Static and dynamic properties of the ferromagnetic NLCs are substantially determined by the coupling of the order parameters of the magnetic and the nematic molecular subsystems taking into account the boundary conditions and features of an intermolecular interaction. While the static properties are related to the equilibrium states of the ferromagnetic NLC, the dynamic properties are associated with the evolution of their transitions between the equilibrium states.

Description of the dynamics order parameters in the ferromagnetic NLCs is based on a hydrodynamics and an irreversible thermodynamics together with their symmetry [5-7]. This corresponds to the longwave and lowfrequency character of the temporal evolution of the magnetization and the molecular orientation ordering under the external magnetic field.

## 2. MODEL OF THE FREE ENERGY AND DYNAMIC EQUATIONS

The dynamic properties of the ferromagnetic NLCs is described in the framework of the model cell in the form of the plane-parallel capillary filled by the NLC with the suspense of ferromagnetic nanoparticle. In the given Cartesian coordinate system, the axis $\boldsymbol{z}$ is perpendicular to the cell surfaces and another two axes, $x$ and $y$ lie in the cell plane. Interaction between magnetic moments is enough to onset of the spontaneous magnetization, $\boldsymbol{M}$. Alignment of the nematic director, $\boldsymbol{n}$, relative to the axis $x$ is given by the interphase interaction between the nematic molecules and the plane-parallel cell surface. Collinearity of the magnetic moment of each of the ferromagnetic nanoparticles to the nematic director, $\boldsymbol{n}$, is given by its magnetic anisotropy. The external magnetic field, $\boldsymbol{H}$, applied perpendicular to the cell surface, causes a distorsion of the nematic director and the optical axis parallel to it. This is manifested in the light passing through the ferromagnetic NLC cell.

The static behavior of the magnetization, $\boldsymbol{M}$, and the director field, $\boldsymbol{n}$, are described by the equations of the variation problem for the functional of the free energy density. The latter can be represented in the form, $f = f_m + f_d + f_s$, where the first magnetic component,

$$f_m = -\mu_0 \boldsymbol{M} \cdot \boldsymbol{H} - \frac{1}{2} A_1 \left( \boldsymbol{M} \cdot \boldsymbol{Q} \cdot \boldsymbol{M} \right) + \frac{1}{2} A_2 \left( |\boldsymbol{M}| - M_0 \right)^2, \qquad (1)$$

where $\mu_0$ is the magnetic constant, $\boldsymbol{H} = \boldsymbol{H}\hat{\boldsymbol{e}}_z$ is the applied magnetic field, $A_{1,2} > 0$, $\boldsymbol{Q} = S\left( \boldsymbol{n} \otimes \boldsymbol{n} - \frac{1}{3} \boldsymbol{I} \right)$ is the quadrupole tensor of the nematic orientation ordering [8]. The first term in (1) represent the coupling of the magnetization and external magnetic field. Since, $H >> M_0$, the local magnetic field is equal to $\boldsymbol{H}$, which is fixed externally and is thus independent of the $\boldsymbol{M}(\boldsymbol{r})$ configuration. The second term describes the static coupling between the director and the magnetization (originated from magnetic nanoparticles). The third term in (1) describes the energy connected with the deviation of the modulus from $M_0$.

The second distortion component, $f_d$, associated with the elastic distortion of the director field, is expressed via the gradient components of the quadrupole tensor, $\partial Q_{ij} / \partial x_k$, [8, 9] as,

$$f_d = \frac{1}{2} L_1 \frac{\partial Q_{ij}}{\partial x_k} \frac{\partial Q_{ij}}{\partial x_k} + \frac{1}{2} L_2 \frac{\partial Q_{ij}}{\partial x_j} \frac{\partial Q_{ik}}{\partial x_k} + \frac{1}{2} L_3 Q_{ij} \frac{\partial Q_{kl}}{\partial x_i} \frac{\partial Q_{ik}}{\partial x_j} \quad , \qquad (2)$$

where $L_i$, $i = 1,2,3$ is the elastic constant. In the vector representation [2],

$$f_d = \frac{1}{2} K_1 (\nabla \cdot \boldsymbol{n})^2 + \frac{1}{2} K_2 \left[ \boldsymbol{n} \cdot (\nabla \times \boldsymbol{n}) \right]^2 + \frac{1}{3} K_3 \left[ \boldsymbol{n} \times (\nabla \times \boldsymbol{n}) \right], \qquad (2a)$$

where $L_1 = (K_3 + 2K_2 - K_1) / 9S^2$, $L_2 = 4(K_1 - K_2) / 9S^2$ and $L_3 = 2(K_3 - K_1) / 9S^2$ with positive elastic constants for splay ($K_1$), twist ($K_2$) and bend ($K_3$).

The third component, $f_s$, is the a finite surface energy associated with the anchoring of the director at the plates, $f_s = -W \left( \boldsymbol{n}_s Q \cdot \boldsymbol{n}_s \right) / 2$, where $W$ is the anchoring strength and $\boldsymbol{n}_s = \hat{\mathbf{e}}_z \sin \varphi_s + \hat{\mathbf{e}}_x \sin \varphi_s$ is the preferred direction specified by the director prelit angle $\varphi_s$. For the total free energy, $F = \int dV \left( f_m + f_d \right) + \int dS \; f_s$ and equilibrium condition requires $\delta F = 0$.

The macroscopic dynamic equations for the magnetization and the director can be represented in the form [5, 6],

$$\dot{M}_i + X_i^R + X_i^D = 0, \quad \dot{n}_i + Y_i^R + Y_i^D = 0 \quad , \qquad (3)$$

where the quasicurrents have been split into reversible $(X_i^R, Y_i^R)$ and irreversible, dissipative $(X_i^D, Y_i^D)$ parts. The reversible (dissipative) parts have the same (opposite) behavior under time reversal as the time derivatives of the corresponding variables, i.e., the first and second equations in (3) are invariant under time reversal only if the dissipative quasicurrents vanish.

The quasicurrents are expressed as linear combinations of conjugate quantities (thermodynamic forces); they take the form,

$$h_i^M \equiv \frac{\partial f}{\partial M_i}, \quad h_i^n \equiv \delta_{ik}^{\perp} \frac{\partial f}{\partial n_k} = \delta_{ik}^{\perp} \left( \frac{\partial f}{\partial n_k} - \partial_j \Phi_{kj} \right), \qquad (4)$$

with $\Phi_{kj} = \partial f / \partial(\partial_j n_k)$ and the transverse Kronecker delta, $\delta_{ik}^{\perp} = \delta_{ik} - n_i n_k$ projects onto the plane perpendicular to the director due to the constraint $\boldsymbol{n}^2 = 1$.

The dissipative quasicurrents take the form [6],

$$X_i^D = b_{ij}^D h_j^M + \chi_{ij}^D h_j^n, \quad Y_i^D = \frac{1}{\gamma_1} h_i^n + \chi \chi_{ij}^D h_j^M \qquad (5)$$

with

$$\chi_{ij}^D = \chi_1^D \delta_{ik}^{\perp} M_k n_j + \chi_2^D \delta_{ik}^{\perp} M_k n_k, \quad b_{ij}^D = b_{\parallel}^D n_i n_j + b_{\perp}^D \delta_{ik}^{\perp} \qquad (5a)$$

The reversible quasicurrents are obtained by requiring that the entropy production, $Y_i h_i^n + X_i h_i^m$ is zero [6]:

$$X_i^R = b_{ij}^R h_j^M + \chi^R \varepsilon_{ijk} n_j h_k^n, \quad Y_i^R = (\gamma_1^{-1})_{ij}^R h_j^n + \chi^R \varepsilon_{ijk} n_j h_k^M \ , \qquad (6)$$

where

$$b_{ij}^R = b_1^R \varepsilon_{ijk} M_k + b_2^R \varepsilon_{ijk} n_k n_p M_p + b_3^R \left( \varepsilon_{ipq} M_p n_q n_j - \varepsilon_{jpq} M_p n_q n_i \right), \qquad (6a)$$

$$(\gamma_1^{-1})_{ij}^R = (\gamma_1^{-1})_1^R \varepsilon_{ijk} n_k n_p M_p + (\gamma_1^{-1})_2^R \left( \varepsilon_{ijp} \varepsilon_{ipk} n_k n_j - \varepsilon_{jpk} n_k n_i \right) M_p \ . \qquad (6b)$$

The system (3) and (4)-(6) describe the dynamics of the magnetization and the director field of the ferromagnetic NLC. This is accompanied by the polarization twisting of the light passing through the NLC cell.

## 2. MAGNETO-OPTIC EFFECT IN THE FERROMAGNETIC NLC

In the framework of the given model, in equilibrium the magnetic-field-distorted director and magnetization fields are lying in the $xz$ plane, $\boldsymbol{n} = (\sin\theta(z), 0, \cos\theta(z))$ and $\boldsymbol{M} = M(\sin\psi(z), 0, \cos\psi(z))$, where $\theta$ and $\psi$ are the tilt angles of the director and the magnetization, respectively, from the axis, $x$. In the absence of the magnetic field, the director is tilted from the $x$ axis by the pretilt, $\varphi_s$. The average $z$ component of the magnetization is described by the expression,

$$M_z = \frac{1}{d} \int d\, zM \cos\psi(z) \, . \qquad (7)$$

The electric component of the linear polarized light along the axis $z$ has the form, $\boldsymbol{E} = E_0 \boldsymbol{j} \exp i(\boldsymbol{k}_i \cdot \boldsymbol{r} - \omega t)$, where $E_0$ is the electric field amplitude, $\boldsymbol{j}$ the initial polarization, $\boldsymbol{k}_i$ the wave vector, and $\omega$ the frequency of the incident light. In the given model the wave vector points in the $z$ direction, $\boldsymbol{k}_i = k_0 \hat{\mathbf{e}}_z$, with $k_0 = 2\pi / \lambda$ being the wave number. The polarization of the light therefore lies in the $xy$ plane and is described by the two-component complex vector, $\boldsymbol{j} = j_x(z)\hat{\mathbf{e}}_x + j_y(z)\hat{\mathbf{e}}_y$. As the light passes through the sample also the components of this (Jones) polarization vector change and we analyze these changes using the Jones matrix formalism (assuming perfectly polarized light) [10].

The incident light first goes through the polarizer oriented at $\pi/4$ with respect to the $x$ axis, and is linearly polarized with the initial Jones vector being, $\boldsymbol{j} = (1/\sqrt{2})(1,1)^{\mathrm{T}}$. The optical axis is parallel to the director and generally varies through the cell. For any ray direction the polarization is decomposed into a polarization perpendicular to the optical axis (ordinary ray) and a polarization which is partly in the direction of the optical axis (extraordinary ray). The ordinary ray experiences an ordinary refractive index $n_0$ and the extraordinary ray experiences a refractive index $n_e$ [7],

$$n_e^{-2} = n_{e0}^{-2}\sin\theta(z) + n_0^{-2}\cos\theta(z)\ , \qquad (8)$$

where $n_{e0} = n_e(z)_{z=0}$.

The effect of each elementary layer of the model NLC cell on the light polarization in the direction $\boldsymbol{z}$ is described by the phase matrix,

$$W(z) = \begin{pmatrix} e^{ik_0[n_e(z)-n_0]\Delta z/2} & 0 \\ 0 & e^{-ik_0[n_e(z)-n_0]\Delta z/2} \end{pmatrix}, \qquad (9)$$

where $\Delta z$ is the width of the elementary layer. In the limit, $\Delta z \to 0$, the transmission matrix of the liquid crystal cell is expressed as

$$T = \begin{pmatrix} e^{i\phi/2} & 0 \\ 0 & e^{-i\phi/2} \end{pmatrix}, \quad \phi = k_0\int_o^d dz\left[n_e(z) - n_0\right]\ , \qquad (10)$$

where $d$ is the width of the cell, $\phi$ is the phase difference.

In general, the director can have also a nonzero component in the $y$ direction. In this case the simple expression for the transmission matrix (10) does not hold anymore and must be generalized. In this case, a general orientation of the director is described as $\boldsymbol{n} = (\sin\theta(z)\cos\varphi(z), \sin\theta(z)\sin\varphi(z), \cos\theta(z))$, where the azimuthal angle of the director $\varphi$ can vary through the cell and the transformation matrix at point $z$ is

$$\mathrm{T}(z) = \mathrm{R}[-\varphi(z)]\mathrm{W}(z)\mathrm{R}[\varphi(z)],\ \ \mathrm{R}(\varphi) = \begin{pmatrix} \cos[\varphi(z)] & \sin[\varphi(z)] \\ -\sin[\varphi(z)] & \cos[\varphi(z)] \end{pmatrix}. \qquad (11)$$

Consequently, the transfer matrix for the whole cell is described as the product, $\mathrm{T} = \lim_{\Delta z\to 0}\prod_{z\in[0,d]}\mathrm{T}(z)$, which is transformed to the form [7],

$$\mathrm{T} = \exp\left[i\int_0^d \mathrm{A}(z)dz\right],\quad \mathrm{A}(z) = \frac{k_0[n_e(z)-n_o]}{2}\begin{pmatrix} \cos[2\varphi(z)] & \sin[\varphi(z)] \\ \sin[2\varphi(z)] & -\cos[2\varphi(z)] \end{pmatrix}, \qquad (12)$$

The exponential of the $2\times 2$ matrix in (12) takes the form,

$$\mathrm{T}=\begin{pmatrix} \cos(a_3)+i\dfrac{a_1}{a_3}\sin(a_3) & i\dfrac{a_2}{a_3}\sin(a_3) \\ i\dfrac{a_2}{a_3}\sin(a_3) & \cos(a_3)-i\dfrac{a_1}{a_3}\sin(a_3) \end{pmatrix}, \quad a_3=\sqrt{a_1^2+a_2^2}\ , \qquad (13)$$

where

$$a_1=\frac{k_0}{2}\int_0^d[n_e(z)-n_0]\cos[2\varphi(\mathrm{z})]dz,\quad a_2=\frac{k_0}{2}\int_0^d[n_e(z)-n_0]\sin[2\varphi(\mathrm{z})]dz\ . \quad (13a)$$

The subsequent passage of the light through an analyzer,

$$\mathrm{P}_\alpha=\begin{pmatrix}\cos^2\alpha & \sin\alpha\cos\alpha \\ \sin\alpha\cos\alpha & \sin^2\alpha\end{pmatrix},$$

which gives the Jones vector ($\alpha=\pi/4$), $\boldsymbol{j}'=i\left(a\sin(\mathrm{c})/\sqrt{2}c\right)(1,-1)^{\mathrm{T}}$. This yields the normalized intensity, $I/I_0=\boldsymbol{j}'^{*\mathrm{T}}\boldsymbol{j}'=(a/c)^2\sin^2 c$.

Since, the Jones vector after the NLC cell is expressed as $\boldsymbol{j}=(z_1e^{i\phi},z_2)^{\mathrm{T}}$, where $z_1$ and $z_2$ are real and $z_1^2+z_2^2=1$, and after an analyzer with $\alpha=\pi/4$ the Jones vector $\boldsymbol{j}'=(1/2)(z_1e^{i\phi}-z_2)(1,-1)^{\mathrm{T}}$, then the intensity is related to the phase difference as

$$\frac{I}{I_0}=\frac{1}{2}(1\text{-}2z_1z_2\cos\phi)\,. \qquad (14)$$

Only if the director is restricted to the $xz$ plane, $z_1=z_2$ and $I/I_0=\sin^2\phi/2$ whence the phase difference is

$$\phi=m\pi\pm 2\arcsin\left[\frac{I}{I_0}\right],\quad m\in Z\ . \qquad (15)$$

This is phase difference only when the nematic director field is in the $xz$ plane. In the case of the dynamics not confined to the $xz$ plane and it is used the above mentioned general relation (14) in the form, $r(H)=1-\varphi(H)/\varphi_0$,where $\varphi_0$ is the normalized phase difference at zeromagnetic field.

## 3. MAGNETIC FIELD INDUCED RESPONSE OF THE NLC CELL

The initial dynamics of the normalized phase difference and magnetization on application of the magnetic field is investigated. Up to linear order the pretilt is taken into account. Initially, $\boldsymbol{n}$ and

$\boldsymbol{M}$ are parallel to $\boldsymbol{n}_s$. Keeping the modulus of the magnetization exactly fixed, the initial thermodynamic forces (4) are

$$\boldsymbol{h}^n = 0, \quad \boldsymbol{h}^{\perp M} = \mu_0 H(\varphi_s, 0, -1) \ , \qquad (16)$$

where $\boldsymbol{h}^{\perp M}$ is the projection of $\boldsymbol{h}^M$ perpendicular to $\boldsymbol{M}$. With that, the initial quasicurrent are described by the equations,

$$Y_i = \chi_{ij}^D h_j^{\perp M} + \chi^R \varepsilon_{ijk} n_j h_k^{\perp M} \Rightarrow \boldsymbol{Y} = \mu_0 H(\chi_2^D M_0 \varphi_s, \chi^R, -\chi_2^D M_0) \, , \qquad (17)$$

$$X_i = b_{ij}^D h_j^{\perp M} + b_{ij}^R h_j^{\perp M} \Rightarrow \boldsymbol{X} = \mu_0 H(b_\perp^D \varphi_s, -(b_1^R + b_2^R) M_0, -b_\perp^D) \, . \qquad (18)$$

At finite $\chi_2^D$ and zero $\chi^R$ it follows from (17) that the $z$ component of the director field responds linearly in time as well as linearly in the magnetic field for small times,

$$n_z(t) \approx \varphi_s + \chi_2^D M_0 \mu_0 H t \, . \qquad (19)$$

As a contrast, if $\chi_2^D$ is zero, then the director responds through the nonzero molecular field $h_z^n$ due to the static coupling $A_1$,

$$h_z^n = -A_1 M_0 M_z(t) = -A_1 M_0 b_\perp^D \mu_0 H t \ , \qquad (20)$$

where $M_z(t) = b_\perp^D \mu_0 H t$ is the initial response of the $z$ component of the magnetization, (18). The $z$ component of the director field thus responds quadratically in time rather than linearly,

$$n_z(t) = \varphi_s + \frac{A_1 M_0 b_\perp^D \mu_0 H}{2\gamma_1} t^2 \qquad (21)$$

For small times, $t$, the refractive index in (8) can be expressed as

$$n_e(t) \approx n_{e0} \left[ 1 - \frac{n_{e0}^2 - n_o^2}{2n_o^2} \right] (\varphi_s + \chi_2^D M_0 \mu_0 H t)^2 \ . \qquad (22)$$

The coefficients $a_1$ and $a_2$ in (13a) are then

$$a_1 \approx \frac{k_0}{2}[n_e(z) - n_0][1 - 2(\chi^R \mu_0 H)^2 t^2, \quad a_2 = \frac{k_0}{2}\frac{k_0}{2}[n_e(z) - n_0](-2\chi^R \mu_0 H)t \quad (23)$$

and the normalized intensity of the transmitted light for small times is

$$\frac{I}{I_0} \approx \sin^2\left(\frac{\phi_0}{2}\right) - r_0 \varphi_s \chi_2^D \mu_0 H M_0 \phi_0 \sin(\phi_0) t$$

$$- \left[ (\chi_2^D \mu_0 H M_0)^2 \phi_0 \sin(\phi_0) + 4(\chi^R \mu_0 H)^2 \sin^2\left(\frac{\phi_0}{2}\right) \right] t^2 \, . \qquad (24)$$

In the lowest order of $t$, for the phase difference, one gets a linear term that is also linear in pretilt and a quadratic term which does not vanish if the pretilt is zero,

$$r(H) \approx r_0\left[(\chi_2^D M_0\mu_0 H)^2 t^2 + 2(\chi_2^D M_0\mu_0 H)t\right] \equiv k^2t^2 + pt \ . \qquad (25)$$

Equation (25) will be used to extract the dissipative cross-coupling coefficient $\chi_2^D$ and the pretilt $\varphi_s$ from the magneto-optic effect. Furthermore, from (25) one can see that in the case of positive (negative) pretilt the normalized phase difference has a minimum at negative (positive) magnetic fields. From the time of this minimum, $t_{\min} = -\varphi_s/(\chi_2^D\mu_0 HM_0)$, one can calculate the ratio of the pretilt and the dissipative cross-coupling. If $\chi_2^D = 0$, then the time of the minimum decreases more slowly with increasing magnetic field, $t_{\min} = \left(-(2\gamma_1\varphi_s)/(A_1 b_\perp^D\mu_0 HM_0)\right)^{1/2}$.

The normalized $z$ component of the magnetization (7) is linear in $t$:

$$\frac{M_z}{M_0} = \varphi_s + \frac{b_\perp^D}{M_0}\mu_0 H\, t \ . \qquad (26)$$

For a nonzero dissipative cross-coupling coefficient $\chi_2^D$ the initial rate, (19), is $1/\tau_d = \chi_2^D M_0\,|H|$. However, if $\chi_2^D = 0$, then initial rate of the director reorientation is proportional to the magnetization, (20), $1/\tau_{s|} = |M_z(t)|\,A_1 M_0/\gamma_1$.

The two latter relaxation rates describe two different mechanisms of the director reorientation. The former is associated with the dynamic coupling of the director and the magnetization, whereas the latter is governed by the static coupling $A_1$ of the director and the magnetization. Here a deviation of the magnetization from the director is needed to exert a torque on the director.

The dissipative cross-coupling of the director and the magnetization, i.e., the $\chi_{ij}^D$ terms of (5), affects the dynamics decisively and is crucial to explain the experimental results. It is described by the parameters $\chi_1^D$ and $\chi_2^D$ of (5a). Varying $\chi_1^D$ while keeping $\chi_2^D = 0$ leads to the small influence of $\chi_1^D$ that is not substantial. Moreover, the initial dynamics is not affected. On the other hand, increasing $\chi_2^D$ strongly reduces the rise time of the normalized phase difference and strongly affects the initial behavior. For large values of $\chi_2^D$ one also observes an overshoot in the normalized phase difference. From (5a) one sees that the influence of $\chi_1^D$ is largest when $\boldsymbol{M} \perp \boldsymbol{n}$, $\boldsymbol{h}'' \perp \boldsymbol{n}$ and $\boldsymbol{h}^M \parallel \boldsymbol{n}$. On the other hand, the influence of $\chi_2^D$ is largest when $\boldsymbol{M} \parallel \boldsymbol{n}$. Since **M** and **n** are initially parallel and, moreover, the transient angle between them never gets large due to the strong static coupling compared to the magnetic fields applied, it is understandable that $\chi_2^D$ affects the dynamics more than $\chi_1^D$.

## 4. CONCLUSIONS

The dynamics properties of the ferromagnetic NLCs under the external magnetic field have studied. In the framework of the hydrodynamic and irreversible thermodynamic approach on the base of the dynamic equations, the nonequilibrium temporal evolution of the coupled magnetic and the orientation ordering molecular subsystems is considered. The considerable effect of the dynamic cross-coupling on the magnetic field-induced dynamics of the magnetization and the nematic director field is shown. The interconnection between the dynamics of magneto-optic effect and the coupled system of the magnetization and the orientation ordering molecular structure has been analyzed.

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