Magneto-optic dynamics in a ferromagnetic nematic liquid crystal

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We investigate dynamic magneto-optic effects in a ferromagnetic nematic liquid crystal experimentally and theoretically. Experimentally we measure the magnetization and the phase difference of the transmitted light when an external magnetic field is applied. As a model we study the coupled dynamics of the magnetization, \( M \), and the director field, \( \mathbf{n} \), associated with the liquid crystalline orientational order. We demonstrate that in the usual experimental setup used for measuring the relaxation rates of the splay-bend or twist-bend eigenmodes of a nematic liquid crystal one expects for a ferromagnetic nematic liquid crystal a mixture of at least two eigenmodes.

I. INTRODUCTION

In Ref. [1] Brochard and de Gennes suggested and discussed a ferromagnetic nematic phase combining the long-range nematic orientational order with long-range ferromagnetic order in a fluid system. The synthesis and experimental characterization of ferrofluids and ferrohydrostereoscleres, a combination of low-molecular-weight nematic liquid crystals (NLCs) with magnetic liquids leading to a superparamagnetic phase, started immediately [2] and continued thereafter [3–9]. These studies made use of ferrofluids or magnetorheological fluids (colloidal suspensions of magnetic particles) [10]; their experimental properties [10,11] have been studied extensively in modeling [12–17] using predominantly macroscopic descriptions [12–14,16].

On the modeling side, the macroscopic dynamics of ferromagnetics was given first for a relaxed magnetization [18] followed by taking into account the magnetization as a dynamic degree of freedom [19] as well as incorporating chirality effects leading to ferrohydrostereoscleres [20]. In parallel a Landau description including nematic as well as ferromagnetic order has been presented [21].

Truly ferromagnetic NLCs have been generated [22] in 2013 followed by reports of further ferromagnetic NLCs in Refs. [23,24], and their macroscopic static properties were characterized in detail [25]. Quite recently ferromagnetic cholesteric liquid crystals have been synthesized and investigated [26–28]. For a review on ferromagnetic NLCs, see Ref. [29].

In the present paper we describe in detail experimentally and theoretically the static and dynamic properties of ferromagnetic NLCs [30]. We analyze the coupled dynamics of the magnetization and the director, initiated and controlled by an external magnetic field. We show experimentally and theoretically that dissipative dynamic coupling terms influence qualitatively the dynamics. Experimentally, this is done by measuring the temporal evolution of the normalized phase difference associated with the dynamics of the director. Quantitative agreement between the experimental results and the model is reached and a dissipative cross-coupling coefficient between the magnetization and the director is accurately evaluated. It is demonstrated that this cross-coupling is crucial to account for the experimental results thus underscoring the importance of such off-diagonal effects in this first multiferroic fluid system. We also make concrete theoretical predictions of how the reversible dynamic cross-coupling terms between magnetization and director influence the macroscopic dynamics and how these effects can be detected experimentally. The experimental and theoretical dynamic results discussed in some detail in this paper for low magnetic fields in ferromagnetic NLCs demonstrate the potential for applications of these materials in displays and magneto-optic devices as well as in the field of smart fluids.

The paper is organized as follows. In Sec. II we describe the experimental setup followed in Sec. III by the macroscopic model. The connection between the measurements and the model is established in Sec. IV. In Sec. V we analyze the

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statics and in Secs. VI and VII we analyze in detail the coupled macroscopic dynamics of the magnetization and the director field when switching the external magnetic field on and off, respectively. Section VIII is dedicated to a theoretical analysis of fluctuations and light scattering and in the conclusions we give a summary of the main results and a perspective.

II. EXPERIMENTS

The experimental samples have been prepared along the lines described in detail in Refs. [22,25]. In brief, the BaSc₂Fe₁₂₋ₓO₁₉ nanoplatelets were suspended in the liquid crystal mixture E7 (Merck, nematic-isotropic transition temperature \( T_{NI} = 58^\circ C \)). The suspension was filled in liquid crystal cells with rubbed surfaces (thickness \( d = 20 \mu m \), Instec Inc.), which induced homogeneous in-plane orientation of the NLC. The volume concentration of the magnetic platelets in the nematic low-molecular-weight liquid crystal E7 (Merck) has been estimated to be \( \sim 1.3 \times 10^{-3} \) from the measurements of the magnetization magnitude [25] which was \( M_0 \sim 200 \, A/m. \) E7 suspensions show long-term stability, with a homogeneous response to magnetic fields and no aggregates for a period of several months. A surfactant (dodecylbenzene sulfonic acid) was used for the treatment of the nanoplatelets, which favors a perpendicular orientation of the NLC molecules with respect to the nanoplatelets. Quantitative values for the Frank coefficients for E7 are available in the literature [31].

Dynamics of the director was measured by inducing director reorientation in planarly treated 20-\( \mu m \) cells (pretilt in the range \( 1^\circ - 3^\circ \)) when applying a magnetic field perpendicularly to the cell plates, Fig. 1 (top). Experiments were performed on monodomain samples (see Ref. [29] for a description of monodomain sample preparation) so that the director is initially at \( 45^\circ \) with respect to the crossed polarizers, Fig. 1 (bottom). Using polarizing microscopy, the monochromatic light intensity transmitted through the sample was recorded with a complementary metal-oxide-semiconductor (CMOS) camera (IDS Imaging U1-3370CP, 997 fps) as a function of time on switching the magnetic field on and off. An interference filter (623.8 nm) was used to filter the light from the halogen lamp used in the microscope. The transmitted light intensity is related to the phase difference between the ordinary and the extraordinary light as will be explained below. The advantage of using polarisation microscopy is that the measurements are performed in the homogeneous region of the sample without spacers or other impurities. Recording the image of the sample during the measurements also allows us to simultaneously monitor the homogeneity of the response.

With the use of a vibrating sample magnetometer [25] (LakeShore 7400 Series VSM) also the equilibrium \( z \) component of the magnetic moment of the sample is measured. We note that this technique is not suitable for measuring the magnetization dynamically, as several seconds per measurement are required for ambient magnetic noise averaging.

III. MACROSCOPIC MODEL

Throughout the present paper we take into account the magnetization \( \mathbf{M} \) and the director field \( \mathbf{n} \) as macroscopic variables; in the following we focus on the essential ingredients of their dynamics necessary to capture the experimental results we will discuss. That is we assume isothermal conditions and discard flow effects. For a complete set of macroscopic dynamic equations for ferronematics we refer to Ref. [19].

The static behavior is described by the free energy density \( f(M, n, \nabla n) \),

\[
f = -\mu_0 \mathbf{M} \cdot \mathbf{H} - \frac{1}{2} A_1 (\mathbf{M} \cdot \mathbf{n})^2 + \frac{1}{2} A_2 (|\mathbf{M}| - M_0)^2 + f^F ,
\]

(1)

where \( \mu_0 \) is the magnetic constant, \( \mathbf{H} = H \hat{e}_z \) is the applied magnetic field, and \( A_{1,2} > 0 \) will be assumed constant. The first term represents the coupling of the magnetization and the external magnetic field. Since \( H \gg M_0 \), the local magnetic field is equal to \( \mathbf{H} \), which is fixed externally and is thus independent of the \( \mathbf{M}(\mathbf{r}) \) configuration. The second term describes the static coupling between the director field and the magnetization (originating from the magnetic particles). The third term describes the energy connected with the deviation of the modulus of the magnetization from \( M_0 \). The last term
is the Frank elastic energy associated with director distortions [32]

\[ f^F = \frac{1}{2} K_1 (\nabla \cdot \mathbf{n})^2 + \frac{1}{2} K_2 [\mathbf{n} \cdot (\nabla \times \mathbf{n})]^2 \]

\[ + \frac{1}{2} K_3 [\mathbf{n} \times (\nabla \times \mathbf{n})]^2, \tag{2} \]

with positive elastic constants for splay \((K_1)\), twist \((K_2)\), and bend \((K_3)\). The saddle-splay elastic energy [32] is zero in the considered geometry. While it is a good approximation to assume that \(|\mathbf{M}| = M_0\), we will take into account small variations of \(|\mathbf{M}|\) (corresponding to large values of \(\Delta z\)).

The anchoring of the director at the plates is taken into account using a finite surface anchoring energy [33],

\[ f^S = -\frac{1}{2} W (\mathbf{n}_S \cdot \mathbf{n})^2, \tag{3} \]

where \(W\) is the anchoring strength and \(\mathbf{n}_S = \hat{\mathbf{e}}_z \sin \phi_s + \hat{\mathbf{e}}_x \cos \phi_s\) is the preferred direction specified by the director prettil angle \(\phi_s\).

For the total free energy we have \(F = \int f \, dV + \int f^S \, dS\) and the equilibrium condition requires \(\delta F = 0\).

The macroscopic dynamic equations for the magnetization and the director read [19,34]

\[ M_i + X^R_i + X^D_i = 0, \tag{4} \]

\[ \dot{n}_i + Y^R_i + Y^D_i = 0, \tag{5} \]

where the quasicurrents have been split into reversible \((X^R, Y^R)\) and irreversible, dissipative \((X^D, Y^D)\) parts. The reversible (dissipative) parts have the same (opposite) behavior under time reversal as the time derivatives of the corresponding variables, i.e., Eqs. \(4\) and \(5\) 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

\[ h_i^R = \frac{\delta f}{\delta M_i}, \]

\[ h_i^D = \frac{\delta f}{\delta n_i}, \]

\[ \dot{h}_i^R = \frac{\delta f}{\delta \dot{M}_i} = \frac{\delta f}{\delta M_i} - \frac{\delta f}{\delta \dot{n}_i} - \frac{\partial f}{\partial \Phi_{ki}}, \tag{6} \]

where \(\Phi_{ki} = \delta f / (\partial \dot{n}_i / \partial m_k)\) and where the transverse Kronecker delta \(\delta_{ik} = \delta_{ik} - n_i n_k\) projects onto the plane perpendicular to the director due to the constraint \(\mathbf{n}^2 = 1\).

In Ref. [30] we focused on the dissipative quasicurrents as they had a direct relevance for the explanation of the experimental results discussed there. In the present paper we also include the reversible quasicurrents, which give rise to transient excursions of \(\mathbf{M}\) and \(\mathbf{n}\) out of the switching plane.

The dissipative quasicurrents take the form [19]

\[ X_i^D = b_i^D \delta_{ik} M_k n_j + \chi_i^D \delta_{ij} M_k n_k, \tag{8} \]

\[ Y_i^D = \frac{1}{\gamma_1} h_i^D + \chi_i^D h_j^D, \tag{9} \]

\[ X_i^D = \frac{1}{\gamma_1} h_i^D + \chi_i^D h_j^D, \tag{10} \]

\[ b_i^D = b_i^D n_i n_j + b_i^D \delta_{ij}, \tag{11} \]

where \(\gamma_1\) is the preferred direction specified by the director.

Throughout the present paper we will discard the biaxiality of the material which arises for \(\mathbf{n} \parallel \mathbf{M}\).

The reversible quasicurrents are obtained by requiring that the entropy production \(Y_i^R = \eta_j^R h_j + \chi_i^R \epsilon_{ij} n_j h_k^R\) is zero [19]:

\[ X_i^R = \eta_j^R h_j^R + \chi_i^R \epsilon_{ij} n_j h_k^R, \tag{12} \]

\[ Y_i^R = \gamma_i^R \gamma_j^R \epsilon_{ij} n_j h_k^R, \tag{13} \]

where [18]

\[ b_i^R = b_i^R \epsilon_{ij} k_n k_p M_p + \gamma_i^R \gamma_j^R \epsilon_{ij} n_j h_k^R. \tag{14} \]

The coordinate system used here is shown in Fig. 1. As explained earlier, the average \(z\) component of the magnetization, \(M_z\), is measured by the vibrating sample magnetometer. In modeling, it is obtained by averaging the \(z\) component of the magnetization field,

\[ M_z = \frac{1}{d} \int_0^d M \cos \psi(z) \, dz. \tag{15} \]

In equilibrium the magnetic-field-distorted director and magnetization fields are lying in the \(xz\) plane, \(\mathbf{n} = (\sin \theta, 0, \cos \theta)\) and \(\mathbf{M} = M (\sin \psi, 0, \cos \psi)\). In the absence of the magnetic field, the director is tilted from the \(x\) axis by the prettil angle \(\phi_s\), Eq. (3). The coordinate system used here is shown in Fig. 1. As explained earlier, the average \(z\) component of the magnetization, \(M_z\), is measured by the vibrating sample magnetometer. In modeling, it is obtained by averaging the \(z\) component of the magnetization field,
To derive the expression for the phase difference we start with an electric field $\mathbf{E}$, which is linearly polarized after the light passes through the polarizer,
\[
\mathbf{E} = E_0 \mathbf{j} e^{i(\mathbf{k}_0 \cdot \mathbf{r} - \omega t)},
\]
where $E_0$ is the electric field amplitude, $\mathbf{j}$ the initial polarization, $\mathbf{k}_0$ the wave vector, and $\omega$ the frequency of the incident light. In our case the wave vector points in the $z$ direction, $\mathbf{k}_0 = k_0 \mathbf{e}_z$, with $k_0 = \frac{2\pi}{\lambda}$ being the wave number. The polarization of the light therefore lies in the $xy$ plane and is described by the two-component complex vector $\mathbf{j} = j_x(z) \mathbf{e}_x + j_y(z) \mathbf{e}_y$. As the light passes through the sample the components of this (Jones) polarization vector change and we analyze these changes using the Jones matrix formalism (assuming perfectly polarized light) [35].

The incident light first goes through the polarizer oriented at $45^\circ$ with respect to the $x$ axis, Fig. 1, and is linearly polarized with the initial Jones vector being $\mathbf{j} = \frac{1}{\sqrt{2}}(1,1)^T$. The optical axis is parallel to the director and generally varies through the cell. For any ray direction we can decompose the polarization 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 $n_o$ and the extraordinary ray experiences a refractive index $n_e$,
\[
n_e^{-2}(z) = n_o^{-2} \sin^2 \theta(z) + n_o^{-2} \cos^2 \theta(z),
\]
where $n_o$ is the extraordinary refractive index.

To calculate the intensity of the transmitted light, one first divides the liquid crystal cell into $N$ thin slices of width $h = d/N$ and describes the effect of each slice on the polarization by the phase matrix
\[
W(z) = \begin{pmatrix}
e^{i k_0 [n_e(z) - n_o] h/2} & 0 \\
0 & e^{-i k_0 [n_e(z) - n_o] h/2}
\end{pmatrix}.
\]
In the limit $N \to \infty$ we can express the transmission matrix of the liquid crystal cell as
\[
T = \begin{pmatrix}
e^{i \phi/2} & 0 \\
0 & e^{-i \phi/2}
\end{pmatrix},
\]
where we have introduced the phase difference
\[
\phi = k_0 \int_0^d [n_e(z) - n_o] dz.
\]

In general, as we will see, the director can have also a nonzero component in the $y$ direction. In this case the simple expression for the transmission matrix Eq. (22) does not hold anymore and must be generalized.

We start the derivation of the general transmission matrix by assuming a general orientation of the director,
\[
\mathbf{n} = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta).
\]
The azimuthal angle of the director $\varphi$ can vary through the cell and the transformation matrix at point $z$ is
\[
T(z) = R(-\varphi(z)) W(z) R(\varphi(z)),
\]
where $R$ is the rotation matrix
\[
R(\varphi) = \begin{pmatrix}
cos(\varphi) & \sin(\varphi) \\
-\sin(\varphi) & \cos(\varphi)
\end{pmatrix}.
\]

Our goal is to find the transfer matrix for the whole cell,
\[
T = \prod_{z \in [0,d]} T(z),
\]
where the arrow denotes the ordered product starting from $T(0)$ at the right side. We first notice that
\[
T(z) \approx I + i \frac{k_0 [n_e(z) - n_o] h}{2} \begin{pmatrix}
\cos[2\varphi(z)] & \sin[2\varphi(z)] \\
\sin[2\varphi(z)] & -\cos[2\varphi(z)]
\end{pmatrix},
\]
where $I$ is the identity matrix. Consequently, we can write $T(z)$ as an exponential,
\[
T(z) = \lim_{h \to 0} \exp[i A(z) h],
\]
where
\[
A(z) = \frac{k_0 [n_e(z) - n_o]}{2} \begin{pmatrix}
\cos[2\varphi(z)] & \sin[2\varphi(z)] \\
\sin[2\varphi(z)] & -\cos[2\varphi(z)]
\end{pmatrix}.
\]

We can now rewrite Eq. (27) as
\[
T = \lim_{h \to 0} \exp \left[ i \sum_{z \in [0,d]} A(z) h \right] = \exp \left[ i \int_0^d A(z) dz \right],
\]
where we used
\[
e^{Ah} e^{Bh} = e^{(A+B)h} + \frac{1}{2} [A,B] h^2 + O(h^3).
\]
The exponential of the $2 \times 2$ matrix from Eq. (31) reads
\[
T = \begin{pmatrix}
\cos(c) + i \frac{b}{2} \sin(c) & i \frac{b}{2} \sin(c) \\
i \frac{b}{2} \sin(c) & \cos(c) - i \frac{b}{2} \sin(c)
\end{pmatrix},
\]
where
\[
c = \sqrt{a^2 + b^2} \\
a = \frac{k_0}{2} \int_0^d [n_e(z) - n_o] \cos[2\varphi(z)] dz, \\
b = \frac{k_0}{2} \int_0^d [n_e(z) - n_o] \sin[2\varphi(z)] dz.
\]

We then let the light pass through an analyzer $P_\alpha$ at an angle $\alpha$,
\[
P_\alpha = \begin{pmatrix}
\cos^2 \alpha & \sin \alpha \cos \alpha \\
\sin \alpha \cos \alpha & \sin^2 \alpha
\end{pmatrix},
\]
which gives for the final Jones vector ($\alpha = -45^\circ$)
\[
\mathbf{j}' = \frac{ia \sin(c)}{\sqrt{2}c} \begin{pmatrix}
1 \\
-1
\end{pmatrix}.
\]
This yields the measured normalized intensity
\[
\frac{I}{I_0} = j'^* j = \frac{a^2}{c^2} \sin^2(c).
\]

Next we evaluate the relation between the phase difference and the measured intensity. Let $\mathbf{j}$ be the Jones vector after the liquid crystal cell,
\[
\mathbf{j} = \begin{pmatrix}
z_1 e^{i \varphi} \\
z_2
\end{pmatrix}.
\]
where \( z_1 \) and \( z_2 \) are real and \( z_1^2 + z_2^2 = 1 \). Generally \( |z_1| \neq |z_2| \). After an analyzer with \( \alpha = -45^\circ \) we have a Jones vector
\[
j' = \frac{1}{2} \left( z_1 e^{i\phi} - z_2 \right) \begin{pmatrix} 1 \\ -1 \end{pmatrix}
\]
(39)
and the intensity is related to the phase difference as
\[
\frac{I}{I_0} = \frac{1}{2} \left[ 1 - 2z_1z_2 \cos(\phi) \right].
\]
(40)
Only if the director is restricted to the \( xz \) plane, \( z_1 = z_2 \) and we have
\[
\frac{I}{I_0} = \frac{1}{2} \left[ 1 - \cos(\phi) \right] = \sin^2 \left( \frac{\phi}{2} \right).
\]
(41)
such that the relation between the intensity and the phase difference is
\[
\phi = m\pi \pm 2 \arcsin \left( \frac{I}{I_0} \right),
\]
(42)
where \( m \in \mathbb{Z} \) and the sign \( \pm \) is determined by demanding that \( \phi \) is sufficiently smooth. Generally, however, the quantity obtained from the measured intensity by Eq. (42) is not the phase difference. It is the phase difference only when the director field is in the \( xz \) plane. For the analysis of the dynamics not confined to the \( xz \) plane, Sec. VI C, we will therefore use the normalized intensity Eq. (37).

In the case when the dynamics is in the \( xz \) plane, to compare the numerical results with the experiments and also to compare the dynamics with the director of the dynamics of the magnetization, it is convenient to introduce the normalized phase difference
\[
r(H) = 1 - \frac{\phi(H)}{\phi_0},
\]
(43)
where \( \phi_0 \) is the phase difference at zero magnetic field. The normalized phase difference is zero at \( t = 0 \) and is always smaller or equal to 1. It can also assume negative values as we will see.

V. STATICS

In this section we present experimental and numerical results of statics and derive analytic formulas for the equilibrium configurations in the low and large external magnetic field limits.

In Fig. 2 we compare the numerical results of the equilibrium normalized phase difference to the experimental data. Below we will show in Eqs. (52) and (53) that the equilibrium normalized phase difference is quadratically dependent on the applied magnetic field at small magnetic fields. The normalized phase difference saturates quickly above \( \mu_0 H = 10 \) mT at a value which is less than 1, which means there is a limit to how much the director field deforms. We also observe that the dependence of the equilibrium normalized phase difference is not symmetric with respect to the \( \mu_0 H = 0 \) axis, which is seen in experiments as well. The reason for this is the nonzero pretilt at both glass plates.

From the fits to the model we extract values for the anchoring strength \( W \), the pretilt angle \( \varphi_s \), the Frank elastic constant \( K \equiv K_1 = K_3 \) in the one constant approximation, and the static coupling coefficient \( A_1 \):
\[
W \sim 4 \times 10^{-5} \text{ J/m}^2,
\]
(44)
\[
\varphi_s \sim -0.05,
\]
(45)
\[
K \sim 17 \text{ pN},
\]
(46)
\[
A_1 \sim 140 \mu_0.
\]
(47)
The extracted parameters Eqs. (44)–(47) correspond to the (local) minimum of the sum of squares of residuals between the numerical and experimental values of the normalized phase difference. This minimum was sought in sensible parameter ranges (for example, the Frank elastic constant was sought in the range between 5 and 25 pN). There are several indications that this minimum is at least very close to the global one. First, the extracted value of the Frank elastic constant is close to the value of \( K_3 \) in the pure E7 NLC. Second, the extracted pretilt is within the range specified by the cell provider. Moreover, the value of the static coupling is similar to that estimated for the ferromagnetic NLC based on pentylicyanobiphenyl (5CB) [25].

The limiting behaviors of the normalized phase difference and the normalized \( z \) component of the magnetization as the magnetic field goes to zero or infinity can be calculated analytically. In all cases the boundary condition is
\[
K \frac{\partial \tilde{\alpha}}{\partial z} + \frac{\partial \tilde{f}^S}{\partial \theta} = 0,
\]
(48)
where $\nu_z$ is the $z$ component of the surface normal pointing upwards at $z = d$ and downwards at $z = 0$.

### A. Low magnetic fields

The free energy density in lowest order in deviations of magnetization and director field from the equilibrium is

$$f = \frac{1}{2}K \left( \frac{\partial \theta}{\partial z} \right)^2 + \frac{1}{2}A_1M_0^4(\theta - \psi)^2 + \mu_0HM_0\psi. \quad (49)$$

The equilibrium solutions for the angles are

$$\theta(z) = \frac{1}{2} \frac{\mu_0HM_0d^2}{K} z(z - d) - \frac{\mu_0HM_0d^2}{2W} + \frac{\pi}{2} - \nu_z, \quad (50)$$

$$\psi(z) = \theta(z) - \frac{\mu_0HM_0}{A_1M_0^2}. \quad (51)$$

After inserting the solutions Eqs. (50) in equations for the normalized phase difference and magnetization, one gets

$$r(H) = \frac{\nu_z}{6K} \frac{\mu_0HM_0d^2}{2} \left[ \frac{\mu_0HM_0d^2}{20K} \left( 1 + 10\frac{\xi}{d} + 30\frac{\xi^2}{d^2} + \left( 1 + 6\frac{\xi}{d} \right) \nu_z \right) \right], \quad (52)$$

where $\xi = K/W$ is the so-called anchoring extrapolation length and $r_0 = n_{eo}(n_{eo} + n_{ho})/(2n_{eo}^2)$. In the limit of infinite anchoring the normalized phase difference reads

$$r(H) = \frac{n_{eo}^2 n_{ho}^2}{6K} \left( \frac{\mu_0HM_0d^2}{20K} \nu_z \right) + \nu_z. \quad (53)$$

One can also observe that the location of the minimum of the normalized phase difference is shifted to a value $\nu_z \min$ determined by the pretilt:

$$-\frac{10K\nu_z (1 + 6\frac{\xi}{d})}{M_0d^2(1 + 10\frac{\xi}{d} + 30\frac{\xi^2}{d^2})} \xrightarrow{w \to \infty} -\frac{10K\nu_z}{M_0d^2}. \quad (54)$$

Equations (52) and (54) are useful for determining the anchoring strength $W$ and the pretilt $\psi^\circ$.

From the behavior of the normalized phase difference at low fields [Eqs. (52) and (53)] one cannot determine the value of the static coupling $A_1$. It can, on the other hand, be determined from the low-field behavior of the magnetization. In Fig. 2 we see that the behavior is linear for low magnetic fields as can be shown analytically:

$$\frac{M_z}{M_0} = \frac{\nu_z}{\frac{1}{A_1M_0^2} + \frac{1}{12K} + \frac{d}{2W}} + \mu_0HM_0. \quad (55)$$

### B. Large magnetic fields

In the large-magnetic-field limit we assume that both the polar angle of the director and the magnetization are either close to 0 if the applied magnetic field is positive (+) or close to $\pi$ if the applied magnetic field is negative (−). The corresponding solutions will be denoted as $\theta^+(z), \theta^-(z), \psi^+(z), \psi^-(z), M_z^+, M_z^-, r^+, r^-$. The free energy in the case of a positive magnetic field is

$$f \approx \frac{1}{2}K \left( \frac{\partial \theta}{\partial z} \right)^2 + \frac{1}{2}A_1M_0^4(\theta - \psi)^2 + \frac{1}{2}\mu_0HM_0\psi^2. \quad (56)$$

The equilibrium solutions for the angles $\theta^+(z)$ and $\psi^+(z)$ are

$$\theta^+(z) = \frac{\xi}{\xi} - \nu_z \cosh \left[ qd \left( \frac{\xi}{\xi} - \nu_z \right) \right], \quad (57)$$

$$\psi^+(z) = \frac{\xi}{\xi} - \nu_z \cosh \left[ qd \left( \frac{\xi}{\xi} - \nu_z \right) \right], \quad (58)$$

where

$$q^2 = \frac{q_0^2 - \mu_0|H|M_0}{A_1M_0^2} \quad (59)$$

with $q_0 = \sqrt{A_1M_0^2/K}$ (which is proportional to the inverse “magnetization coherence length” of the director).

The normalized $z$ component of the magnetization for large fields is

$$\frac{M_z}{M_0} = 1 - \frac{(\xi - \nu_z)^2(qd + \sinh(qd))A_1^2M_0^6}{4qd[1 + q\xi \tanh(qd)]^2 \cosh(qd)^2} \quad (60)$$

and the normalized phase difference is

$$r^+(H) = 1 - \frac{n_0\nu_zKd}{2\phi_0} \left[ \frac{\xi}{\xi} - \nu_z \right]^2 \frac{qd + \sinh(qd)}{1 + q\xi \tanh(qd)} \cosh(qd)^2 + \frac{8qd + 8 \sinh(qd) + \sinh(2qd)}{8qd \cosh(qd)^4}, \quad (61)$$

where $r_\infty = (n_{eh}^2 - n_{eh}^2)/n_{eh}^2$.

It follows from symmetry that $\theta^-(\nu_z) = \pi - \theta^+(\nu_z), \psi^-(\nu_z) = \pi - \psi^+(\nu_z), M_z^-(\nu_z) = -M_z^+(\nu_z), \hbox{ and } r^-(\nu_z) = r^+(\nu_z)$.

Since the magnetization is not anchored at the boundary, in Eq. (60) it was sufficient to consider terms not higher than $(\psi^+)$. On the other hand, due to the anchoring of the director field, in Eq. (61) we expanded the phase difference to the order $(\psi^+)^4$. It should be noted that the approximation for the phase difference is better if the anchoring $W$ is low, i.e., $q\xi \gg 1$ or $W \ll \sqrt{A_1M_0^2/K}$.  


For low magnetic fields, the numerically calculated polar angle of the director is in agreement with Eq. (50). In the large-magnetic-field limit, where $qd \gg 1$, and if $q\xi \gg 1$ in addition, one can study asymptotic behavior of Eqs. (60) and (61):

$$r^+ (H) \approx r^+ (\infty) - \frac{f^+ (q_0)}{\mu_0 |H|},$$

$$\frac{M^+}{M_0} \approx 1 - \frac{h^+ (q_0)}{(|\mu_0 H|)^2},$$

where $f^+$ and $h^+$ are functions of static parameters for positive magnetic fields and $r^+ (\infty) = \lim_{\mu_0 H \to \infty} f^+ (H)$. The behavior of the magnetization $M_z$, Fig. 2, may at a first glance look like the Langevin function, often observed in magnetic systems. Equation (63) tells us that this is not the case, since the Langevin function saturates with the first power in magnetic field, whereas here the saturation Eq. (63) is of second order in $H$.

### C. Comparison of analytic approximations with numerics

A comparison of analytic and numeric results for the director polar angle $\theta (z)$ is made in Figs. 3 and 4 for small and large magnetic fields, respectively. We find a good agreement for small magnetic fields up to 0.7 mT and for large magnetic fields above 4 mT. It should be emphasized that the values of the magnetic fields at which the approximations become valid depend on the values of the static parameters. We use the values Eqs. (44)–(47) extracted from the fits to the macroscopic model.

In Fig. 5 we compare analytic and numeric results for the $z$ component of the magnetization and the normalized phase difference. Again we find a good agreement between the results at similar ranges of the magnetic field. From the insets of Fig. 5 one can conclude that for our system a magnetic field as small as 1 mT can be considered as large already. The notable discrepancy of the numeric and analytic normalized phase difference at large magnetic fields is due to the fact that one has expanded the expression for the phase difference, Eq. (23), up to the order $\theta^4$. Since $\theta$ does not saturate to zero, this means that the constant term of Eq. (61) is slightly different from the actual value determined numerically.

The agreement between experimental data and the model for two key static properties underscores that we have solid ground for the analysis of the dynamic results which now follows.
VI. SWITCH-ON DYNAMICS

In this section we present the experimental and theoretical results of the dynamics that takes place when the magnetic field is switched on.

In Fig. 6 we plot the comparison of experimental and theoretical data for the dynamics of the normalized phase difference (top) as well as the theoretical results for the normalized \(z\) component of the magnetization (bottom) for two values of the applied magnetic field. As an inset we show that for small times the magnetization grows linearly, which is also obtained analytically in Sec. VI A. As expected the rise time for the magnetization is reduced as the applied magnetic field is increased. The inset for the top graph shows that the initial phase difference is quadratic in time, which is again obtained also analytically, see Sec. VIA.

The fits for the comparison of the experimental and theoretical normalized phase difference are performed by varying the dynamic parameters taking into account the fundamental restrictions \[30\] on their values, at fixed values of the static parameters Eqs. (44)–(47). The model captures the dynamics very well for all times from the onset to the saturation. The extracted values of the dynamic parameters are

\[ \gamma_1 \sim 0.13 \text{ Pa s}, \]

\[ b_D^\perp \sim 1.5 \times 10^5 \text{ Am/V s}^2, \]

\[ \chi_D^2 \sim 4 (\text{Pa s})^{-1}. \]

The dissipative cross-coupling coefficient \( \chi_D^2 \) is within the allowed interval determined by the restriction \[30\]

\[ \left| \chi_D^2 \right| < \frac{b_D^\perp}{\gamma_1 M_0^2} \approx 5.4 (\text{Pa s})^{-1}. \]

The remaining two dynamic parameters do not affect the dynamics significantly and are set to \( b_\parallel = b_\perp \) and \( \chi_1^D = 0 \).

To extract from the time evolution of the normalized phase difference, Fig. 6 (top), a switching time \( \tau \) as a measure of an overall relaxation rate of the dynamics, we use a squared sigmoidal model function,

\[ f(t) = C' \left[ 1 - \frac{1 + C}{1 + C \exp(-2t/\tau)} \right]^2. \]

Remarkably, the relaxation rate, \( 1/\tau(H) \), shows a linear dependence on \( H \), Fig. 7. We were first interested in the effect of the dissipative cross-coupling on \( 1/\tau(H) \). We find that a reasonably strong dynamic cross-coupling \( \chi_D^2 \) is needed in order to obtain the observed linear magnetic field dependence of the relaxation rate. In the absence of this dynamic cross-coupling, Fig. 7, the relaxation rate levels off already at low fields as expected since the transient angle between \( \mathbf{M} \) and \( \mathbf{n} \) gets larger and starts to decrease for even higher magnetic fields.

The best match of the relaxation rates \( 1/\tau(H) \) extracted from the experimental data and the model, Fig. 7, allows for a robust evaluation of the dissipative cross-coupling between the magnetization and the director:

\[ \chi_2^D = (4.0 \pm 0.7) (\text{Pa s})^{-1}. \]

A. Initial dynamics

We investigate the initial dynamics of the normalized phase difference and magnetization on application of the magnetic
field. Up to linear order we also take into account the pretilt. Initially, \( \mathbf{n} \) and \( \mathbf{M} \) are parallel to \( \mathbf{n}_3 \). Keeping the modulus of the magnetization exactly fixed, the initial thermodynamic forces Eqs. (6) and (7) are

\[
h^2 = 0, \quad h^M = \mu_0 H(\varphi_s, 0, -1),
\]

where \( h^M \) is the projection of \( h^M \) perpendicular to \( \mathbf{M} \). With that, the initial quasicurrents are

\[
Y_i = \chi_{ij}^D h_{i}^{L-M} + \chi^R \varepsilon_{ijkl} h_{k}^{L-M} \Rightarrow \quad Y = \mu_0 H(\chi_{ij}^D M_0 \varphi_s, \chi^R, -\chi_{ij}^D M_0),
\]

\[
X_i = b_i^D h_{i}^{L-M} + b_i^R h_{i}^{L-M} \Rightarrow \quad X = \mu_0 H(0, b_i^D \varphi_s, -b_i^R + b_i^R M_0, -b_i^D). \tag{72}
\]

At finite \( \chi_{ij}^D \) and zero \( \chi^R \) it follows from Eq. (71) that the \( z \) component of the director field responds linearly in time as well as linearly in the magnetic field for small times:

\[
n_z(t) \approx \varphi_s + \frac{A_1 M_0 b_0^D \mu_0 H t}{2 \gamma_1}. \tag{73}
\]

As a contrast, if \( \chi_{ij}^D \) is zero, then the director responds through the nonzero molecular field \( h^o \) due to the static coupling \( A_1 \),

\[
h_o^z = -A_1 M_0 M_z(t) = -A_1 M_0 b_0^D \mu_0 H t, \tag{74}
\]

where \( M_z(t) = b_0^D \mu_0 H t \) is the initial response of the \( z \) component of the magnetization, Eq. (72). The \( z \) component of the director field thus responds quadratically in time rather than linearly,

\[
n_z(t) \approx \varphi_s + \frac{A_1 M_0 b_0^D \mu_0 H t}{2 \gamma_1} t^2. \tag{75}
\]

For small times \( t \) we can express the refractive index Eq. (20) as

\[
n_o(t) \approx n_o \left[ 1 - \frac{n_o^2}{2 n_o^2} (\varphi_s + \chi_{ij}^D M_0 \mu_0 H t)^2 \right]. \tag{76}
\]

The coefficients \( a \) and \( b \) from Eq. (34) are then

\[
a \approx \frac{k_o d}{2} [n_z(t) - n_o][1 - 2(\chi^R \mu_0 H)^2 t^2], \tag{77}
\]

\[
b \approx \frac{k_o d}{2} [n_z(t) - n_o][-2 \chi^R \mu_0 H t]
\]

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_{ij}^D \mu_0 H M_0\phi_0 \sin(\phi_0) t
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Therefrom we extract the value of the dissipative cross-coupling parameter $\chi^D_2$ between director and magnetization,

$$\chi^D_2 \sim (4.0 \pm 0.5) \text{(Pa s)}^{-1},$$

and from the parameter $p$ of Eq. (79) we extract the pretilt,

$$\varphi_s \sim -0.065 \pm 0.01.$$

The normalized $z$ component of the magnetization Eq. (18) is linear in $t$:

$$\frac{M_z}{M_0} = \varphi_s + \frac{b^D}{M_0} \mu_0 H t,$$

which is in accord with Fig. 6 (bottom). From the initial behavior one can therefore directly determine the dissipative coefficient $b^D$. Let us define the initial rate of the director reorientation as the time derivative of the director $z$ component at $t = 0$,

$$\frac{1}{\tau_d} = \frac{\partial n_z}{\partial t} \bigg|_{t=0}.$$

For a nonzero dissipative cross-coupling coefficient $\chi^D_2$ the initial rate, Eq. (73), is

$$\frac{1}{\tau_d} = \chi^D_2 M_0 \mu_0 |H|.$$

However, if $\chi^D_2 = 0$, then the initial rate of the director reorientation is proportional to the $z$ component of the magnetization, Eq. (74),

$$\frac{1}{\tau_s} = A_1 M_0 \gamma_1 |M_z(t)|.$$

The relaxation rates Eqs. (87) and (88) 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.

## B. Dissipative cross-coupling

We have demonstrated that the dissipative cross-coupling of the director and the magnetization, i.e., the $\chi^D_2$ terms of Eqs. (8) and (9), affects the dynamics decisively and is crucial to explain the experimental results. It is described by the parameters $\chi^D_1$ and $\chi^D_2$ of Eq. (10). Here we check the sensitivity of the dynamics to the values of these two parameters. Varying $\chi^D_1$ while keeping $\chi^D_2 = 0$, Fig. 12, we see that the influence of $\chi^D_1$ is rather small and is not substantial. Moreover, the initial dynamics is not affected, Fig. 12 (inset).

On the other hand, increasing $\chi^D_2$ strongly reduces the rise time of the normalized phase difference, Fig. 13, and also strongly affects the initial behavior (inset). For large values of $\chi^D_2$ one also observes an overshoot in the normalized phase difference.

By inspecting Eq. (10) one sees that the influence of $\chi^D_1$ is largest when $\mathbf{M} \perp \mathbf{n}$, $\mathbf{h}^n \parallel |\mathbf{M}|$, and $\mathbf{h}^M \parallel \mathbf{n}$. On the other hand, the influence of $\chi^D_2$ is largest when $\mathbf{M} \parallel \mathbf{n}$. Since $\mathbf{M}$ and $\mathbf{n}$ 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 $\chi^D_2$ affects the dynamics more than $\chi^D_1$. 
C. Reversible cross-coupling

The reversible cross-coupling of the director and the magnetization, described by the $\chi^R$ terms of Eqs. (12) and (13), has not been considered up to this point. We focus on the reversible cross-coupling coefficient $\chi^R$ and put both reversible tensors $b^R_{ij}$ and $(\gamma - 1)^R_{ij}$ of Eqs. (14) and (15) to zero.

If the reversible currents are included, then both variables wander out of the $xz$ plane dynamically, which will be described by the azimuthal angles $\delta$ and $\phi$ of the magnetization and the director, respectively, defined by $M = M_0(\cos \delta \sin \psi, \sin \delta \sin \psi, \cos \psi)$, $n = (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta)$. The dynamic behavior of both azimuthal angles is shown in Fig. 14.

Contrary to the polar angles we find that the response of the azimuthal angle of the director is faster than that of the magnetization. From Fig. 14, we read off that the maximum azimuthal angles increase with $\chi^R$, being higher for the magnetization than for the director.

We note again that here we only included the reversible cross-coupling $\chi^R$. From the initial quasichorrents Eqs. (71) and (72) one can see that the initial azimuthal response of the magnetization can be faster than that of the director if the coefficients of the tensor $b^R_{ij}$ are sufficiently large,

$$|b^R_{ij} + \chi^R_{ij}| > |\chi^R|/M_0.$$  \hfill (89)

There exists a direct way of detecting the possible dynamics in the $xy$ plane. The intensity of the transmitted light in the experiments with crossed polarizers at $45^\circ$ and $-45^\circ$ is given by Eq. (37),

$$I/I_0 = \frac{a^2 c^2 \sin^2(c)}{\sin^2(c)}.$$  \hfill (90)

It is this quantity that is typically measured. On the other hand, crossed polarizers at $0^\circ$ and $90^\circ$ give us the intensity

$$I/I_0 = \frac{b^2 c^2 \sin^2(c)}{\sin^2(c)},$$  \hfill (91)

with $a$ and $b$ given by Eq. (34). This method is better suited for detecting the $xy$ dynamics, since $b$ is more sensitive to the deviation of the director field from the $xz$ plane.

Our numerical calculations have revealed that, due to the reversible dynamics, the magnetization and the director are not confined to the $xz$ plane. As a consequence, the maxima of the time-dependent intensity of transmitted light are lower than unity, Fig. 15, in contrast to the case of a purely in-plane (dissipative) dynamics. Observation of the lower maxima could thus be an indication of the azimuthal dynamics. This effect is more prominent at higher magnetic fields and at higher values of the reversible cross-coupling coefficients.

In recent experiments no clear-cut consequences of the azimuthal dynamics have been found using crossed polarizers at $0^\circ$ and $90^\circ$. In the following we will therefore discard the reversible dynamics.

VII. SWITCH-OFF DYNAMICS

Dynamics of the normalized phase difference after switching off the magnetic field has been also measured. In experiments, the initial state is obtained by switching on the desired magnetic field and waiting for a couple of seconds. Contrary to the previous experiments, here the initial state is not homogeneous.
FIG. 15. Time dependence of the normalized intensity of transmitted light for zero and nonzero values of the reversible cross-coupling coefficient $\chi^R$; $\mu_0 H = 5\, \text{mT}$.

In Fig. 16 we compare the experimental and numerical normalized phase difference at two different fields. We observe, similarly to the switch-on case, that the normalized phase difference goes through a minimum. This is again explained by the fact that the director field goes through a state, which is approximately aligned with the surfaces of the glass plates. Numerical calculations reveal that a strong dissipative cross-coupling causes the initial behavior of the normalized phase difference to be a linear function in time, Fig. 17, as found experimentally, Fig. 16.

To extract a relaxation time $\tau$ of the normalized phase difference, we use an exponential function

$$ f(t) = f(0)e^{-t/\tau}. $$

(92)

FIG. 16. Experimental and numerical normalized phase difference as a function of time at different values of the applied magnetic field.

The relaxation rate $1/\tau$ for the experimental data is shown in Fig. 18. It saturates at a finite value as one increases the magnetic field. This is expected since the initial director and magnetization fields do not change much with magnetic field any more when the field is large. In Fig. 19 the relaxation rate of both the computed phase difference and the magnetization is shown. One can see that the relaxation rate of the magnetization is smaller than that of the normalized phase difference, due to the fact that it is the director that is driven by the nonzero elastic force, while the magnetization only follows. This is true for all allowed values of the dynamic cross-coupling parameters.

One can derive analytic formulas for the relaxation rate in the limit of low magnetic fields. With the assumption that the relaxation follows a simple exponential function, it is possible to extract the relaxation rate $1/\tau_{\text{eff}}$ from the initial time derivative of the normalized phase difference,

$$ r(H,t) \approx r(H,t = 0) \left( 1 - \frac{t}{\tau_{\text{eff}}} \right). $$

(93)

Note that Eq. (93) is defined only when $r(H,t = 0) \neq 0$.

One starts with the director quasicurrent $Y$, Eq. (9).

The response of the $z$ component of the director field is

FIG. 17. Normalized phase difference as a function of time at $5\, \text{mT}$, calculated with $\chi^D = 0$ and $\chi^D = 4.0\, (\text{Pa s})^{-1}$.

FIG. 18. Experimental switch-off relaxation rate of the normalized phase difference as a function of the applied magnetic field.
Figure 20 shows the relaxation rate of the normalized phase component of the magnetization after switching off the magnetic field of strength $\mu_0 H$ at $\chi_2^D = 4$ (Pa s)$^{-1}$ and $\psi_s = 0$.

$$n_z \approx n_z(z,t = 0) - Y_z(z,t = 0)\,t,$$

which one uses in Eq. (23) for the phase difference,

$$\frac{1}{\tau_{\text{eff}}} = \frac{2k_0r_0(n_e - n_o)}{\phi_0r(H,t = 0)} \int_0^d dz\, n_z(z)Y_z(z) \left[1 + \frac{n_e - n_o}{n_2} n_2(z)\right]^{3/2},$$

(94)

where all $z$-dependent quantities are evaluated at $t = 0$. In the last step the integrand is expanded up to linear order in time and the relaxation rate in the low-magnetic-field limit is finally expressed as

$$\frac{1}{\tau_{\text{eff}}} = \frac{(1 + r_0\psi_s)^2}{20K_1} \left[(1 + 6\frac{\xi}{\lambda} + 12\frac{K_1\psi_s}{\lambda^2})\chi_2^D\right],$$

(95)

which is linear in the dissipative cross-coupling coefficient $\chi_2^D$.

Not only does the dissipative cross-coupling make the switching process faster when switching on the field, this can also be true for switching off the field, Figs. 17 and 20. Figure 20 shows the relaxation rate of the normalized phase difference at a high magnetic field as a function of the dissipative cross coupling coefficient $\chi_2^D$. As expected, the relaxation rate decreases with increasing rotational viscosity $\gamma_1$. The relaxation rate at first increases with increasing values of $\chi_2^D$, which seems also to be the case for small magnetic fields described by Eq. (95). For values above approximately $\chi_2^D = 3.5$ (Pa s)$^{-1}$, the relaxation rate starts to decrease rather rapidly. This is in contrast with the field switch-on case, where the response is faster for increasing values of $\chi_2^D$.

The increasing part of the dependence $\tau^{-1}(\chi_2^D)$ in Fig. 20 is due to the director elastic forces, which drive the switch-off dynamics and also enter Eq. (4) through the dissipative cross-coupling governed by $\chi_2^D$. At higher values of $\chi_2^D$ one must, however, also consider the part of the thermodynamic forces corresponding to the static $(A_1)$ coupling between the director and the magnetization. Focusing only on the director equation Eq. (5), one sees that the director relaxes towards the magnetization with a characteristic time set by the rotational viscosity and the static coupling $(A_1)$. On the other hand, the positive value of $\chi_2^D$ has the opposite effect. While both fields eventually relax to the ground state parallel to $x$, the angle between them is decreasing slower and slower as the dynamic cross-coupling $(\chi_2^D)$ gets larger. For small magnetic fields one can study the relaxation rate of the dynamic eigenmodes [Eq. (122)] of the next section. The value of $\chi_2^D$ above which the relaxation rate starts to decrease then reads

$$\chi_2^D = \begin{cases} \frac{A_1b_D^2}{A_1M_0^2 + K(\pi/2)^2} & \text{if } \frac{1}{\gamma_1} > \frac{b_D^2}{M_0^2}, \\ \frac{1}{\gamma_1} & \text{if } \frac{1}{\gamma_1} < \frac{b_D^2}{M_0^2}. \end{cases}$$

(96)

In our case $\frac{1}{\gamma_1} > \frac{b_D^2}{M_0^2}$ holds and the maximum is at $\chi_2^D \approx 3.5$ (Pa s)$^{-1}$.

The switch-on case is different in that the dynamics is driven by the external magnetic field. If the external field is sufficiently high (large compared to $A_1M_0$), the static cross-coupling effects, which decrease the relaxation rate in the switch-off case through the increasing dynamic cross-coupling $\chi_2^D$, can be neglected and hence the relaxation rate is monotonically increasing with $\chi_2^D$.

VIII. FLUCTUATIONS AND LIGHT SCATTERING

Nematic liquid crystals appear turbid in sufficiently thick layers [32]. The scattering of light is caused by strong director fluctuations which cause fluctuations in the dielectric tensor

$$\varepsilon_{ij} = \varepsilon_{\perp} \delta_{ij} + \varepsilon_{\parallel} n_i n_j,$$

(97)

where $\varepsilon_{\perp}$ and $\varepsilon_{\parallel}$ are dielectric susceptibilities for the electric field perpendicular and parallel to the director, respectively. Fluctuations are easy to observe experimentally and are used to determine the viscoelastic properties of liquid crystals [36].

In this paper we derive the relaxation rates of the fluctuations without taking into account the effects of flow. Since the director is coupled to the magnetization, we now have two fluctuation modes for each director fluctuation mode of the usual nematic [22].

The fluctuating director and magnetization fields are linearized as

$$\mathbf{n} = \mathbf{n}_0 + \delta \mathbf{n}, \quad \mathbf{M} = \mathbf{M}_0 + M_0 \delta \mathbf{m},$$

(98)
where the equilibrium director $\mathbf{n}_0$ and magnetization $\mathbf{M}_0$ fields point in $x$ direction in which a magnetic field is applied, whereas fluctuations $\delta \mathbf{n}$ and $\delta \mathbf{m}$ are perpendicular, $\mathbf{n}_0 \cdot \delta \mathbf{n} = \mathbf{M}_0 \cdot \delta \mathbf{m} = 0$. The ansatz for the director fluctuations is

$$
\delta n(r) = \frac{1}{V} \sum_q \delta n(q) e^{iqr},
$$

where $q = q_x \hat{e}_x + q_y \hat{e}_y + q_z \hat{e}_z$ is the wave vector of the fluctuation. A similar ansatz is used for the fluctuations of the magnetization. In a confined system, the fluctuation spectrum generally depends on the interaction of the nematic with the surface [36]. For simplicity we will use the infinite anchoring limit, so that $q_z = n\pi/d, n \in \mathbb{N}$, while $q_x$ and $q_y$ are in principle arbitrary. For details regarding the anchoring effect we refer to Ref. [36].

To understand the static light-scattering experiments one must determine thermal averages of the fluctuations. This is done by finding linear combinations of the variables in terms of which the free energy functional Eq. (1) is expressed as a sum of quadratic terms and making use of equipartition. Such linear combinations are uncorrelated (statistically independent). A systematic way to perform this decomposition is to write the free energy of a fluctuation $\mathbf{q}$-mode as a quadratic form and find the corresponding eigenvalues and eigenvectors,

$$
F(\mathbf{q}) = \frac{1}{2} \delta \mathbf{x}(\mathbf{q})^T \mathbf{E}(\mathbf{q}) \delta \mathbf{x}(\mathbf{q}),
$$

where $\delta \mathbf{x}(\mathbf{q}) = [\delta n_x(\mathbf{q}), \delta n_y(\mathbf{q}), \delta n_z(\mathbf{q})]$, in short $\delta \mathbf{x}(\mathbf{q}) \equiv [n_x, m_y, m_z]$, is the vector of the fluctuation amplitudes, $\mathbf{E}(\mathbf{q})$ is a self-adjoint matrix, and superscript $H$ is the conjugate transpose.

In lowest order of fluctuations, the contributions Eq. (100) of the free energy Eq. (1) are [32]

$$
F(\mathbf{q}) = \frac{1}{2V} \left( (K_1 q_x^2 + K_2 q_y^2 + K_3 q_z^2 + A_1 M_0^2) n_x^2 \right. \\
+ (K_1 q_x^2 + K_2 q_y^2 + K_3 q_z^2 + A_1 M_0^2) n_y^2 \left. \\
+ (K_1 q_x^2 + K_2 q_y^2 + K_3 q_z^2 + A_1 M_0^2) n_z^2 \\
+ (K_1 - K_2) q_x q_y (n_x n_y + n_z m_z) \\
+ (K_1 - K_2) q_x q_z (n_x n_z + n_y m_y) \\
+ (K_1 - K_2) q_y q_z (n_y n_z + n_x m_x) \\
+ (\mu \mu M_0 + A_1 M_0^2) |m_x|^2 + |m_y|^2 \\
+ (\mu \mu M_0 + A_1 M_0^2) |m_y|^2 + |m_z|^2 \\
- A_1 M_0^2 (n_x m_y + n_y m_x + n_z m_z + n_z m_z) \right),
$$

For completeness (not needed here), the volume-integrated free energy is $F = \sum_{\mathbf{q}} F(\mathbf{q})$.

Before giving the eigenvectors of the quadratic form $\mathbf{E}$, we perform a rotation in the $yz$ plane, $(n_x, n_y) \rightarrow (n_1, n_2)$ and $(m_x, m_y) \rightarrow (m_1, m_2)$, where new bases in this plane are $[\hat{e}_1, \hat{e}_2]$ and $[\hat{e}_M^1, \hat{e}_M^2]$. Vectors $\hat{e}_1$ and $\hat{e}_2$ are normal to the $(\mathbf{q}, \mathbf{n}_0)$ and $(\mathbf{q}, \mathbf{m}_0)$ plane, respectively, and vectors $\hat{e}_M^1$ and $\hat{e}_M^2$ are normal to $\hat{e}_1$ and $\hat{e}_2$, respectively. It should be emphasized that we are studying the case $\mathbf{n}_0 \parallel \mathbf{m}_0$, so the planes $(\mathbf{q}, \mathbf{n}_0)$ and $(\mathbf{q}, \mathbf{m}_0)$ are identical. In the confined system, this would not be the case if the external magnetic field were applied in any direction other than parallel to the initial homogeneous state.

A general fluctuation $\delta \mathbf{x}(\mathbf{q})$ can be written as

$$
\delta \mathbf{x} = t_1 \mathbf{t}_1 + p_1 \mathbf{p}_1 + t_2 \mathbf{t}_2 + p_2 \mathbf{p}_2,
$$

where $\mathbf{t}_1, \mathbf{t}_2, \mathbf{p}_1, \mathbf{p}_2$ are the eigenvectors of the quadratic form $\mathbf{E}$ and $t_1, t_2, p_1, p_2$ are the amplitudes of these uncorrelated fluctuations.

![FIG. 21. The normalized coefficients Eq. (103) of the eigenvectors $t_1$ and $t_2$ as a function of the applied magnetic field with $q_x = 0$ and $q_\perp = \pi/2; K_1 = K_2$.](image)

The excitation modes $t_1$ and $p_1$ are the analogues of the splay-bend mode in the usual NLCs, whereas $t_2$ and $p_2$ are the analogues of the twist-bend mode.

It is found that in the limit of large magnetic fields these excitations become decoupled, i.e., one eigenvector only contains the fluctuation of the director field and the other the fluctuation of the magnetization field, Figs. 21 and 22.

The thermal averages of the squared amplitudes of the independent excitations read

$$
\langle |t_\alpha(q)|^2 \rangle = \frac{k_B T V}{2 (2 A_1 M_0^2 + \mu \mu H M_0 + K_0 q_\perp^2 + K_3 q_z^2 - s_\alpha)},
$$

$$
\langle |p_\alpha(q)|^2 \rangle = \frac{k_B T V}{2 (2 A_1 M_0^2 + \mu \mu H M_0 + K_0 q_\perp^2 + K_3 q_z^2 + s_\alpha)},
$$

with $k_B$ the Boltzmann constant and $T$ the temperature, whereas their thermal cross-correlations are zero.

If $K_1 = K_2$, then the splay-bend ($\alpha = 1$) and the twist-bend ($\alpha = 2$) excitation modes have the same structure [Eqs. (105) and (106)] (Figs. 21 and 22), as well as the same energy.
and thermal amplitude [Eqs. (107) and (108)]. The same is
true in the degenerate case when \( q = q_\parallel \mathbf{e}_z \), i.e., for a pure
bend excitation (in an unconfined system), where there is no
difference between the modes \( \alpha = 1,2 \) and the bases \( \{ \mathbf{e}_1^B, \mathbf{e}_2^B \} \) and
\( \{ \mathbf{e}_1^M, \mathbf{e}_2^M \} \) are chosen arbitrarily in the
\( yz \) plane.

The space correlations are expressed as
\[
\langle t_\alpha(r)t_\alpha'(r') \rangle = \frac{1}{V^2} \sum_{q,q'} \langle t_\alpha(q) t_\alpha(-q') \rangle e^{-i(q \cdot r + q' \cdot r')},
\]
and similarly for \( \langle p_\alpha(r)p_\alpha'(r') \rangle \), whereas \( \langle t_\alpha(r) p_\alpha'(r') \rangle = 0 \). In the
large magnetic field limit these correlations are
\[
\langle t_\alpha(r)t_\alpha'(r') \rangle \approx \frac{k_BT}{4\pi K} \delta(r),
\]
and similarly for \( \langle p_\alpha(r)p_\alpha'(r') \rangle \).

In experiments one measures the intensity of the scattered
light. To calculate this intensity, we need an expression for the
amplitude of the outgoing electric field. We start with an
incident electric field \( \mathbf{E}_0 \), described by a plane wave:
\( \mathbf{E} = E_0 e^{i(k_0 \cdot r - \omega t)} \), where \( k_0 \) is the wave vector, \( E_0 \) the amplitude,
and \( \omega \) the frequency of the incident light. We then proceed with a
summation of the electric field contributions of the scattered
light through the whole cell, treating every point \( r \) as a radiating
dipole. Last, we project the electric field on the axis \( \hat{f} \) of the
analyzer. The electric field amplitude of the scattered light is
\[ E_f(q,t) = \frac{E_0 \omega}{c^2 R} e^{i(k_0 \cdot r - \omega t)} \int_V d^3 r \ e^{-iq \cdot r} \hat{f}_i \left[ \delta_{ij} - \delta_{ij} \right] \hat{f}_j, \]
where \( k_f \) is the wave vector of the scattered light, \( R \) is the
distance from the sample to the detector at \( r' \), and \( q = k_f - k_0 \)
is the fluctuation wave vector. In the last line of Eq. (112) we
discarded the Fourier contribution of \( \delta_{ij} \), since it is nonzero
only if \( q = 0 \). We have assumed that \( R \) is large compared to
the size of the scattering region, which in turn is much larger
than the wave length of the light, and that we are in the limit of
small dielectric anisotropy.

In our calculations below, we will be using details of an
experimental setup usually used for measuring splay-bend
fluctuations in a NLC, which in our geometry have \( \hat{a} = \delta n \mathbf{e}_x \),
\( q_\parallel = 0 \), \( \mathbf{e}_1^{h,M} = \mathbf{e}_y \), and \( \mathbf{e}_1^{v,M} = \mathbf{e}_z \). In this case we have a
polarizer and an analyzer that are both in the \( xz \) plane. The
polarizer \( i \) is parallel to the \( x \) axis, whereas the analyzer \( \hat{f} \) is at
an angle \( \varphi \) from the \( x \) axis. In Eq. (112), the projection of the
fluctuating part of the dielectric tensor Eq. (97) reads
\[
\hat{f}_i \left[ \delta_{ij} - \delta_{ij} \right] \hat{f}_j = \delta_{ij} f_z \delta_{ij},
\]
where \( f_z = \hat{f} \cdot \mathbf{e}_z \). Using the expansion
\[
\delta_{ij} \approx (t_1(t_1 + p_1 p_1) \cdot \mathbf{e}_n, 0)
\]
the scattering cross section \( \sigma = \langle V_f^2 | q(t) E_f(q,t) \rangle \) with \( q \cdot \mathbf{e}_y = 0 \) is
\[
\sigma = \frac{\varepsilon_2^2 \omega^4}{c^4} \langle |\delta n_z(q)|^2 \rangle f_z^2,
\]
with the coefficient
\[
C^\pm = \frac{(Z_1^\pm)^2}{1 + (Z_1^\pm)^2}.
\]
In the usual experimental setup one observes two splay-bend
modes, \( t_1 \) and \( p_1 \), as opposed to the usual NLC, where one
observes only one splay-bend mode.

Asymptotic behaviors of the coefficients \( C^+_1 \) and \( C^-_1 \) at large
magnetic fields,
\[
C^+_1 \approx 1 - \frac{A_1^2 M_0^2}{(\mu_0 H M_0)^2},
\]
\[
C^-_1 \approx 2 \left( K_1 q_z^2 + K_3 g_z^2 \right) - 3 A_1^2 M_0^2
\]
\[
\left( \mu_0 H M_0 \right)^2,
\]
reveal that in the large-magnetic-field limit only the eigenmode
\( t_1 \) contributes to the scattering cross section [Eq. (115)].

The dynamics of the fluctuations is probed by dynamic light
scattering, where one measures the time correlation of the light
intensity \( I(t) \),
\[
g^{(2)}(t) = \frac{\langle I(0) I(t) \rangle}{\langle I(0) \rangle^2}.
\]
Assuming Gaussian fluctuations it follows that
\[
g^{(2)}(t) = 1 + |g^{(1)}(t)|^2,
\]
where
\[
g^{(1)}(t) = \frac{\langle E_f(q,0) E_f(q,t) \rangle}{\langle |E_f(q,0)|^2 \rangle}
\]
is the time correlation of the scattered light electric field.

To calculate the time dependence of the fluctuations, we first
linearize the system of dynamic equations and determine
the dynamic eigenmodes. Considering only the dissipative
dynamics, Eqs. (8) and (9), and using δn = δn1ε1 + δn2ε2, δm = δm1εM1 + δm2εM2, we find a 2 × 2 homogeneous system for each α = 1,2,
\[
\frac{1}{\tau} \frac{\partial n_\alpha}{\partial t} = \left[ \frac{1}{\gamma_1} (K_\alpha q_\alpha^2 + K_{3q}^2 + A_1 M_0^2) - \chi_2^D A_1 M_0^2 \right] n_\alpha + A_1 M_0^2 \left( \frac{1}{\gamma_1} + \chi_2^D \mu_0 M_0 \right) m_\alpha,
\]
\[
\frac{1}{\tau} \frac{\partial m_\alpha}{\partial t} = [-b_\perp^D A_1 + \chi_2^D (K_\alpha q_\alpha^2 + K_{3q}^2 + A_1 M_0^2)] n_\alpha + \left[ b_\perp^D A_1 \left( 1 + \frac{\mu_0 H M_0}{A_1 M_0^2} \right) - \chi_2^D A_1 M_0^2 \right] m_\alpha,
\]
(122)
which can be rewritten as
\[
\begin{pmatrix} A - \frac{1}{\tau} \end{pmatrix} \begin{pmatrix} \frac{\partial n_\alpha}{\partial t} \\ \frac{\partial m_\alpha}{\partial t} \end{pmatrix} = 0
\]
(123)
and has nontrivial solutions if det(A - 1/τ) = 0. The dynamic eigenmodes are the eigenvectors of the matrix A,
\[
t_\alpha^h = c_\alpha^e e_m + d_\alpha^e e_M,
\]
(124)
\[
p_\alpha^h = c_\alpha^e e_m + d_\alpha^e e_M,
\]
(125)
where the components c_α^e, c_α^p, d_α^e, d_α^p are functions of the static and dynamic material parameters and will not be given explicitly. It is important to realize that the dynamic fluctuation modes (Eqs. (117) and (118)) includes the electric field time correlation Eq. (121) is a single exponential
\[
g^{(2)}(t) = 1 + e^{-2t/\tau}.\]
(130)
It is found that the dynamics of the eigenmodes t_α^h slows down (t_α^h → ∞) at a negative critical magnetic field, here given for q = q_α, ε_α:
\[
\mu_0 H_\alpha^{(s)} = -\frac{A_1 M_0 K_\alpha q_\alpha^2}{K_\alpha q_\alpha^2 + A_1 M_0^2}.
\]
(131)
The negative value of the critical magnetic field means that it is pointing in the direction opposite to the magnetization. If the applied magnetic field is more negative than the critical field, then the magnetization starts to reverse. In NLCs, K_2 < K_1 usually holds and it is the twist mode t_2^h that slows down at a less-negative magnetic field. With the smallest wave number q_1 = π/d we get μ_0 H_1^{(s)} = -2.5 mT.

In Fig. 23 we present the magnetic field dependence of the relaxation rate of almost pure bend (q_1 ≫ q_2) fluctuations. We also depict the corresponding eigenmodes at a small positive field and at large magnetic fields.

For a general fluctuation, in the limit of large magnetic fields the relaxation rate of the faster (magnetization-like) p_α^h mode is proportional to the applied magnetic field (Fig. 23 presents the bend fluctuation as an example),
\[
\frac{1}{\tau_\alpha^h} = A_1 \left( b_\perp^D - \chi_2^D M_0^2 \right)^2 \left( \chi_2^D M_0^2 + K_2 q_\alpha^2 + K_3 q_\alpha^2 \right) \frac{b_\perp^D}{b_\perp^D M_0 \mu_0 H}.
\]
(132)

The relaxation rate of the slower (director-like) t_α^h mode saturates at a finite value (Fig. 23 presents the bend fluctuation in terms of the dynamic eigenmodes [Eqs. (124) and (125)], where the components c_α^e, c_α^p, d_α^e, d_α^p are functions of the static and dynamic material parameters and will not be given explicitly. It is important to realize that the dynamic fluctuation modes (Eqs. (117) and (118)) include the electric field time correlation Eq. (121) is a single exponential
\[
\begin{align*}
g^{(2)}(t) &= 1 + e^{-2t/\tau}.
\end{align*}
\]
(130)
It is found that the dynamics of the eigenmodes t_α^h slows down (t_α^h → ∞) at a negative critical magnetic field, here given for q = q_α, ε_α:
\[
\mu_0 H_\alpha^{(s)} = -\frac{A_1 M_0 K_\alpha q_\alpha^2}{K_\alpha q_\alpha^2 + A_1 M_0^2}.
\]
(131)
The negative value of the critical magnetic field means that it is pointing in the direction opposite to the magnetization. If the applied magnetic field is more negative than the critical field, then the magnetization starts to reverse. In NLCs, K_2 < K_1 usually holds and it is the twist mode t_2^h that slows down at a less-negative magnetic field. With the smallest wave number q_1 = π/d we get μ_0 H_1^{(s)} = -2.5 mT.

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\[
\frac{1}{\tau_\alpha^h} = A_1 \left( b_\perp^D - \chi_2^D M_0^2 \right)^2 \left( \chi_2^D M_0^2 + K_2 q_\alpha^2 + K_3 q_\alpha^2 \right) \frac{b_\perp^D}{b_\perp^D M_0 \mu_0 H}.
\]
(132)

The relaxation rate of the slower (director-like) t_α^h mode saturates at a finite value (Fig. 23 presents the bend fluctuation...
as an example),

$$\frac{1}{\tau_a} = \frac{A_1 M_0^2}{\gamma_1} \left[ 1 - \frac{(\chi_D^g M_0)^2}{b_D^g} \right].$$

(133)

It is also illuminating to study the relaxation rates of general fluctuations at zero magnetic field, \( H = 0 \). Expanding the relaxation rates to second order in \( q \) and \( q_1 \) one gets

$$\frac{1}{\tau_a} = \frac{A_1 M_0^2}{\gamma_1} \left( 1 - 2\chi_D^g + b_D^g \gamma_1 \right) + \frac{(K_m q_1^2 + K_m q_1^2) \Xi_p}{\gamma_1},$$

(134)

$$\frac{1}{\tau_a} = \frac{(K_m q_1^2 + K_m q_1^2) \Xi_r}{\gamma_1},$$

(135)

where

$$\Xi_p = \frac{(\chi_D^g \gamma_1 - 1)^2 M_0^2}{b_D^g \gamma_1 + (1 - 2\chi_D^g \gamma_1) M_0^2},$$

(136)

$$\Xi_r = \frac{\gamma_1 (b_D^g - \chi_D^g M_0^2 \gamma_1)}{b_D^g \gamma_1 + (1 - 2\chi_D^g \gamma_1) M_0^2}.$$  

(137)

From Eqs. (134) and (135) one can see that the relaxation rate \( 1/\tau_a \) of the faster (optic) mode \( p^b \) stays finite in the limit \( q \to 0 \). The slower mode \( \tau_a' \) is, on the other hand, acoustic, i.e., \( 1/\tau_a' \to 0 \) as \( q \to 0 \).

**IX. SUMMARY AND PERSPECTIVE**

In the present extensive study we have presented detailed experimental and theoretical investigations of the dynamics of the magnetization and the director in a ferromagnetic liquid crystal in the absence of flow. We have shown that a dissipative cross-coupling between these two macroscopic variables, which has been determined quantitatively, is essential to account for the experimental results also for the compound E7 as a nematic solvent for the ferromagnetic nematic phase. Before, this was demonstrated for 5CB as a nematic solvent [30]. We also find that all the experimental results presented here for E7 complement well and are consistent with the previous ones using 5CB as the nematic component. Remarkably, the dissipative cross-coupling \((\chi_D^g)\) found for the E7-based ferromagnetic nematic liquid crystal is about a factor of 5 smaller than that of the 5CB based, while the dissipative coefficient of the magnetization \((b_D^g)\) is (only) twice as large. This leads to an interesting suggestion for future experimental work, namely to address the question of which molecular features determine the strength of this dissipative cross-coupling. The nematic phases of 5CB and E7, respectively, show one qualitatively different feature: The nematic phase of 5CB is well known to favor the formation of transient pairlike aggregates [37] because of its nitrile group, while such tendencies are reduced in E7 since it is mixture of four different compounds and also contains a terphenyl. A natural experiment to study these features in more detail would be to investigate the dependence of the dissipative cross-coupling on the magnetic particle concentration on one hand and to investigate mixtures of the nematic solvents 5CB and E7 on the other to learn more about the coupling mechanisms between the nematic order and the magnetic order.

We have also analyzed the consequences of an out-of-plane dynamics, i.e., out of the plane spanned by the magnetic field and the spontaneous magnetization. We give predictions for both the azimuthal angles of director and magnetization as well as for the intensity change related to the reversible dynamic cross-coupling terms between the two order parameters, the magnetization and the director. We find that from both measurements a value for the reversible cross-coupling terms can be extracted.

From the present analysis the next steps in this field appear to be quite well defined. First, the incorporation of flow effects appears to be highly desirable both from a theoretical as well as from an experimental point of view. Early experimental results in this direction have been described in Ref. [38], where it has been shown that viscous effects can be tuned by an external magnetic field of about \( 10^{-2} \) T by more than a factor of two. From a theoretical perspective, questions like the analogs of the Miesowicz viscosities and flow alignment are high on the priority list [39].

Moreover, it will be important to realize, although perhaps experimentally challenging, a nematic or cholesteric liquid crystalline version of uniaxial magnetic gels and rubbers [40,41]. Cross-linking a ferromagnetic nematic would give rise to the possibility to obtain a soft ferromagnetic gel, opening the door to a new class of magnetic complex fluids. This way, one could combine the macroscopic degrees of freedom of the first liquid multiferroic, namely the ferromagnetic nematic liquid crystal, with the strain field as well as with relative rotations. In a step towards this goal, we will derive macroscopic dynamic equations generalizing those for uniaxial magnetic gels and ferrofluids to obtain the macroscopic dynamics for ferromagnetic nematic and cholesteric gels [42].

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