

Maier-Saupe model of liquid crystals: Isotropic-nematic phase transitions and second-order statistics studied by Shiino's perturbation theory and strongly nonlinear Smoluchowski equations

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(Received 1 March 2005; published 7 October 2005)

We study the first- and second-order statistical properties of a dynamical Maier-Saupe model for liquid crystals that is given in terms of a nonlinear Smoluchowski equation. Using Shiino's perturbation theory, we analyze the first-order statistics and give a rigorous proof of the emergence of a phase transition from a uniform distribution to a nonuniform distribution, reflecting phase transitions from isotropic to nematic phases, as observed in nematic liquid crystals. Using the concept of strongly nonlinear Fokker-Planck equations, the second-order statistics of the dynamical Maier-Saupe model is studied and an analytical expression for the short-time autocorrelation function of the orientation of the crystal molecules is derived.

DOI: [10.1103/PhysRevE.72.041703](https://doi.org/10.1103/PhysRevE.72.041703)

PACS number(s): 61.30.Gd, 64.70.Md

I. INTRODUCTION

Nematic liquid crystals composed of anisotropic, elongated molecules with cylindrical symmetry typically exhibit a phase transition from an isotropic phase to a nematic phase [1–4]. In the isotropic phase there is a random orientation of the symmetry axes, whereas in the nematic phase there is orientational order of the symmetry axes. According to the Maier-Saupe theory this order-disorder phase transition results from the competition of a fluctuating force that tends to destroy any orientational order and a mean-field force that is produced by all molecules and tends to align the molecule axes [5–7]. At present, many researcher have focused on dynamical aspects of nematic liquid crystals. In this context, a dynamical mean-field model in terms of a nonlinear self-consistent Smoluchowski equations has been proposed by Hess [8] and Doi and Edwards [9] and, since then, has found many applications and generalizations (see, e.g., [10–18]). To this end, one studies the single-particle density $P(\mathbf{u}, t; w)$ of the director \mathbf{u} describing the molecules' symmetry axes, where P is normalized to unity like $\oint P(\mathbf{u}, t; w) dO = 1$ and $w(\mathbf{u})$ denotes the initial distribution at time $t=0$. The evolution of $P(\mathbf{u}, t; w)$ for $t > 0$ is given by [8,9]

$$\frac{\partial}{\partial t} P(\mathbf{u}, t; w) = D_r \mathbf{L} \cdot \left\{ \mathbf{L} P + \frac{1}{kT} [\mathbf{L} e(\mathbf{u}, P)] P \right\}, \quad (1)$$

with $\mathbf{L} = \mathbf{u} \times \partial / \partial \mathbf{u}$. Here, D_r denotes the rotational diffusion constant and $e(\mathbf{u}, P)$ describes the self-consistent potential of the Maier-Saupe mean-field force (see below). Using spherical coordinates with the polar angle $\theta \in [0, \pi]$ and the azimuthal angle $\varphi \in [0, 2\pi]$, the director is given by $\mathbf{u} = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$. In what follows, we assume a cylindrical symmetry of the director field with respect to the z axis. In this case, the order parameter of the liquid crystal is then given by $q = \langle A(\cos \theta) \rangle$ with $A(\cos \theta) = [3 \cos^2(\theta) - 1]/2$ [1,2,4,19]. Alternatively, we may put $A(x) = P_2(x) = [3x^2 - 1]/2$, where P_2 is second Legendre polynomial. The Maier-Saupe energy functional reads $E[P] = -U_0 kT q^2 / 2$. The corresponding potential function $e(\theta)$ oc-

curing in Eq. (1) is given by the variational derivative $e = \delta E / \delta P$ and reads explicitly

$$e(\theta, P) = -U_0 kT A(\cos \theta) \langle A(\cos \theta) \rangle. \quad (2)$$

Here, U_0 is a parameter characterizing the interaction strength. In terms of the single-particle density $P(\mathbf{u}(\theta), t; w)$ normalized like $2\pi \int_0^\pi P(\mathbf{u}(\theta), t; w) \sin \theta d\theta = 1$, Eq. (1) becomes

$$\frac{\partial}{\partial t} P(\mathbf{u}(\theta), t; w) = \frac{D_r}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \left\{ 3U_0 \sin \theta \cos \theta q [P] P + \frac{\partial}{\partial \theta} P \right\}. \quad (3)$$

Following the study by Felderhof and Jones [20], we put now $x = \cos \theta$ with $x \in [-1, 1]$ and subjected to periodic boundary conditions. Then, the single-particle density $P(x, t; w)$ is given by $P(x(\theta), t; w) = P(\mathbf{u}(\theta), t; w) / 2\pi$ with $\int_{-1}^1 P(x, t; w) dx = 1$. Recall that we have $\mathbf{u} = (\sqrt{M(x)} \cos \varphi, \sqrt{M(x)} \sin \varphi, x)$ with $M(x) = 1 - x^2$. Therefore, $P(x, t; w)$ describes the first-order statistics of the z component of the director field \mathbf{u} . The order parameter now reads $q = [3 \langle x^2 \rangle - 1] / 2$, and Eq. (3) becomes

$$\frac{\partial}{\partial t} P(x, t; w) = \frac{\partial}{\partial x} M(x) \left\{ -\frac{9}{2} \kappa x \left(\langle x^2 \rangle - \frac{1}{3} \right) P + D_r \frac{\partial}{\partial x} P \right\} \quad (4)$$

for $\kappa = U_0 D_r$.

Two important issues in the context of the dynamic Maier-Saupe model (1) are (i) to give a rigorous proof that the model can describe a nematic-isotropic phase transition as observed in liquid crystals and (ii) to derive analytical expressions for the evolution of expectation values.

So far, a rigorous proof has not been given for the fundamental Maier-Saupe model (1). The reason for this is that although for nonlinear Fokker-Planck equations of the form (1) a bifurcation theory was developed by Shiino for order parameters that involve the first moment of P [21,22], a bifurcation theory for equations of the form (1) that involve

order parameters in terms of second- and higher-order moments—as in Eq. (4)—was not available for a long time. This situation, however, has changed recently. In a series of recent studies [23–27] a bifurcation theory for nonlinear Fokker-Planck equations involving second- and higher-order moments has been developed. Therefore, our first objective (see Sec. II A) is to apply this theory to the dynamical Maier-Saupe model given by Eq. (4).

Up to now, theoretical studies of dynamical mean-field models such as Eq. (1) have primarily been focused on the evolution of expectation values related to first-order statistics (i.e., moments and the order parameter); see, e.g., [9,10,12,13,20,28]. Only little is known about how to derive analytical expressions for expectation values related to second-order statistics (e.g., correlation functions). However, quantities of second-order statistics, in general, and correlation functions, in particular, provide very useful information. Especially, they have been exploited in the context of fluctuation-dissipation theorems in order to determine system parameters; see, e.g., [29,30]. In principle, second-order statistics can be determined numerically from the multivariate Langevin equation corresponding to Eq. (1). Only recently has it been shown how to determine them in an analytical fashion using the concept of so-called strongly nonlinear Fokker-Planck equations [31–36]. Therefore, our second objective (see Sec. II B) is to exploit this concept in order to get insights into the evolution of correlation functions as defined by the Maier-Saupe model (4) for liquid crystals with cylindrical symmetry.

II. BIFURCATION THEORY AND SECOND-ORDER STATISTICS

A. Bifurcation theory

We will carry out the bifurcation theory of the model (4) using several key concepts such as linear nonequilibrium thermodynamics, H theorems, Lyapunov's functionals, and Shiino's decomposition of perturbations as reviewed in [37].

1. Linear nonequilibrium thermodynamics

In order to interpret Eq. (4) in terms of linear nonequilibrium thermodynamics, we assume that the distribution $P(x, t; w)$ of the z component of the director field satisfies the continuity equation

$$\frac{\partial}{\partial t} P = - \frac{\partial}{\partial x} j, \quad (5)$$

where j denotes a probability current. We assume that there is a linear relationship between the current j and a thermodynamical force X^{th} given by $j(x, t) = M^*(x) P X^{\text{th}}$ [38–41], where M^* is a generalized mobility function that may depend on x and will be determined below. We assume that the thermodynamical force can be derived from a generalized chemical potential μ like $X^{\text{th}}(x, t) = -\partial\mu/\partial x$ and assume that μ is given by the variational derivative of a free energy functional $F: \mu(x, t) = \delta F / \delta P$. Then, the continuity equation (5) becomes the free-energy Fokker-Planck equation

$$\frac{\partial}{\partial t} P = \frac{\partial}{\partial x} M^*(x) P \frac{\partial}{\partial x} \frac{\delta F}{\delta P}. \quad (6)$$

We would like to emphasize that this approach is consistent with the GENERIC approach developed in [42–45]. For the Maier-Saupe model we use the free-energy functional $F[P] = U - D_r S$ involving the Maier-Saupe energy functional $U = -\kappa q^2/2$ and the Boltzmann entropy $S = -\int P \ln P dx$ (note that we put here the Boltzmann factor equal to unity). We can show that

$$\frac{\delta F}{\delta P} = - \frac{3}{2} \kappa x^2 q + D_r (1 + \ln P) \quad (7)$$

holds. Substituting Eq. (7) into Eq. (6), we see that the free-energy Fokker-Planck equation (6) is equivalent to the nonlinear Smoluchowski equation (4) if we put $M^*(x) = M(x)$.

2. Stationary distributions

In general, stationary distributions P_{st} of Eq. (6) are given by $j = \text{const}$. In particular, we find that the relation $j=0$ holds because we have $M = M^* = 0$ at $x = \pm 1$. From $j=0$ it follows that stationary distributions are given by $\delta F / \delta P = \mu$, where μ is constant and serves as a normalization factor. From Eq. (7) and $\delta F / \delta P = \mu$ it follows that the distributions P_{st} satisfy the implicit equation

$$P_{\text{st}}(x) = \frac{1}{Z(q)} \exp \left\{ \frac{3\kappa q x^2}{2D_r} \right\}, \quad (8)$$

where Z is a normalization constant depending on q and q is given by $q = [3\langle x^2 \rangle_{\text{st}} - 1]/2$. It is clear that the uniform distribution $P_{\text{st}}(x) = 1/2$ is a stationary distribution of Eq. (4) and satisfies the implicit equation (8) with $q=0$. In order to determine order parameter values different from zero, we need to solve the self-consistency equation

$$q = R(q), \quad (9)$$

with

$$R(m) = \int_{-1}^1 \frac{3x^2 - 1}{2} P(x, m) dx \quad (10)$$

and

$$P(x, m) = \frac{1}{Z(m)} \exp \left\{ \frac{3\kappa m x^2}{2D_r} \right\}. \quad (11)$$

For the sake of convenience, we introduce the function $R'(m) = m - R(m)$. Then, q is determined by $R'(q) = 0$. Figure 1 shows $R'(m)$ for several control parameters D_r/κ . We read off from Fig. 1 that for $D_r/\kappa > C$ with $C \approx 0.223$ there is only one solution of $R'(q) = 0$ which is given by $q=0$ and is related to the uniform distribution describing the isotropic phase. For $D_r/\kappa \in [1/5, C]$ there are three solutions of $R'(q) = 0$ given by $q_0 = 0$ and $q_{1,2} > 0$ related to the uniform distribution and two nonuniform distributions. The latter describe nematic phases. For $D_r/\kappa < 1/5$ the self-consistency equation $R'(q) = 0$ has two solutions given by $q_0 = 0$ and $q_1 > 0$. In sum, for $D_r/\kappa < C$ the Maier-Saupe model (4) de-

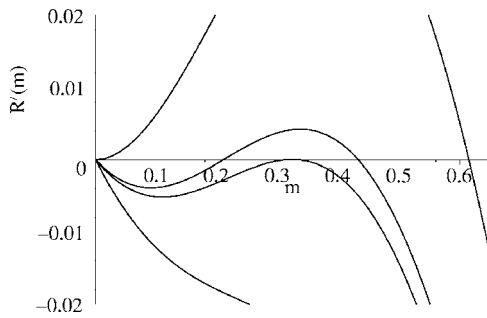


FIG. 1. Illustration of the self-consistency equation $R'(m) = R(m) - m = 0$. Solutions of $R'(m) = 0$ describe order parameters of stationary distributions. If $dR'(m)/dm < 0$ (> 0) at $R'(m) = 0$, the corresponding stationary distribution is stable (unstable). From bottom to top: $D_r/\kappa = 0.24$, $D_r/\kappa \approx 0.223$ (saddle node bifurcation point, emergence of the nematic phase), $D_r/\kappa = 0.22$, and $D_r/\kappa = 1/5$ (the isotropic phase becomes unstable).

scribes liquid crystals that can exhibit two different stationary phases: an isotropic and a nematic one. In view of these findings, the question arises as to whether or not both phases are stable in the sense that small perturbations will vanish. We will address this question next using Lyapunov's direct method in combination with an H theorem for the model (4). In doing so, we will also derive the aforementioned boundary value of $1/5$.

3. H theorem

For solutions of Eq. (6) the free energy F evolves like

$$\frac{d}{dt}F = - \int_{-1}^1 M(x)P \left[\frac{\partial}{\partial x} \frac{\delta F}{\delta P} \right]^2 dx \leq 0. \quad (12)$$

That is, we have a monotonically decreasing function. Note that in deriving Eq. (12) we have exploited the fact that the expression $j\delta F/\delta P|_{-1}^1$ arising due to partial integration vanishes. Next, we realize that the implication $\partial P/\partial t = 0 \Rightarrow dF/dt = 0$ holds. In addition, from $dF/dt = 0$ it follows that $\delta F/\delta P = \text{const}$ [see Eq. (12)] which, in turn, implies that $\partial P/\partial t = 0$ [see Eq. (6)]. In sum, the implication

$$\frac{d}{dt}F = 0 \Leftrightarrow \frac{\partial}{\partial t}P = 0 \quad (13)$$

holds. Furthermore, note that F is bounded from below because we have $q \leq 1$ and S is maximal for the uniform distribution $P = 1/2$ with $S[P = 1/2] = \ln 2$. That is, we have $F \geq -\kappa/2 + D_r \ln 2$ (see also [37,46]). From the boundedness of F and Eqs. (12) and (13) we conclude that any transient solution converges to a stationary one:

$$\lim_{t \rightarrow \infty} \frac{\partial}{\partial t}P = 0. \quad (14)$$

That is, we have an H theorem for the nonlinear Smoluchowski equation (4) similar to a variety of H theorems that have been derived recently in the context of nonlinear Fokker-Planck equations [20,22,41,47].

4. Stability analysis by means of Shiino's perturbation theory

As shown above, stationary distributions correspond to extrema of the free-energy functional F (Sec. II A 2) and F is a monotonically decreasing function of t (Sec. II A 3). Consequently, stable stationary distributions correspond to minima of F , whereas unstable stationary distributions correspond to maxima or saddle points of F (Lyapunov's direct method). Using Shiino's decomposition of perturbations [22–24,26,27], we determine next the character of the extrema of stationary distributions (8) for given order parameters q . To this end, we first put $P(x) = P_{\text{st}}(x) + \epsilon(x)$, where ϵ describes a small perturbation with $\int \epsilon(x) dx = 0$. From Eq. (7) it follows that the second variation of F is given by

$$\delta^2 F[P_{\text{st}}] = - \frac{9\kappa}{4} \left[\int x^2 \epsilon(x) dx \right]^2 + D_r \int \frac{\epsilon^2(x)}{P_{\text{st}}(x)} dx. \quad (15)$$

Now, let us expression the perturbation in terms of $\epsilon(x) = \sqrt{P_{\text{st}}(x)} \epsilon'(x)$ with $\epsilon'(x) = \chi_0(x) + \chi_{\perp}(x)$ and $\int \chi_0(x) \chi_{\perp}(x) dx = 0$. That is, $\chi_0(x)$ and $\chi_{\perp}(x)$ are orthogonal functions. We further require that the first expansion function χ_0 involves the order parameter q : $\chi_0 = \beta[A(x) - q] \sqrt{P_{\text{st}}} = \beta[A(x) - \langle A(x) \rangle_{\text{st}}] \sqrt{P_{\text{st}}}$. Note that from $\int \epsilon(x) dx = 0$ it then follows that $\int \chi_{\perp}(x) \sqrt{P_{\text{st}}(x)} dx = 0$ and $\int \chi_{\perp}(x) A(x) \sqrt{P_{\text{st}}(x)} dx = 0$. The second expansion function χ_{\perp} accounts for all contributions to $\epsilon'(x)$ orthogonal to χ_0 such that $\epsilon'(x)$ indeed reflects all possible perturbations satisfying $\int \sqrt{P_{\text{st}}(x)} \epsilon'(x) dx = 0$ [22,37]. In sum, the original perturbation function $\epsilon(x)$ reads

$$\epsilon(x) = \beta[A(x) - \langle A(x) \rangle_{\text{st}}] P_{\text{st}}(x) + \chi_{\perp}(x) \sqrt{P_{\text{st}}(x)}. \quad (16)$$

Substituting, Eq. (16) into Eq. (15) and exploiting the aforementioned orthogonality properties of χ_0 and χ_{\perp} , we obtain

$$\delta^2 F[P_{\text{st}}] = \beta^2 K_{A,\text{st}} (D_r - \kappa K_{A,\text{st}}) + D_r \int \chi_{\perp}^2(x) dx, \quad (17)$$

where $K_{A,\text{st}}$ denotes that the generalized stationary variance:

$$K_{A,\text{st}} = \langle [A(x) - \langle A \rangle_{\text{st}}]^2 \rangle_{\text{st}}. \quad (18)$$

Introducing the stability coefficient $\tilde{\lambda} = D_r - \kappa K_{A,\text{st}}$, we write Eq. (18) as

$$\delta^2 F[P_{\text{st}}] = \beta^2 K_{A,\text{st}} \tilde{\lambda} + D_r \int \chi_{\perp}^2(x) dx. \quad (19)$$

Consequently, for $\tilde{\lambda} > 0$ we have $\delta^2 F > 0$. Stationary distributions P_{st} that yield $\tilde{\lambda} > 0$ describe free-energy minima and correspond to stable distributions. In contrast, for $\tilde{\lambda} < 0$ we have $\delta^2 F < 0$ for $\epsilon(x)$ with $\chi_{\perp} = 0$. Therefore, stationary distributions P_{st} that yield $\tilde{\lambda} < 0$ describe free-energy maxima or saddle points and correspond to unstable distributions.

Finally, we need to determine $K_{A,\text{st}}$. For the isotropic phase given by $P_{\text{st}} = 1/2$ with $q = 0$ we find $K_{A,\text{st}} = 1/5$, which gives us $\tilde{\lambda}(q = 0) = D_r - \kappa/5$. We conclude that for $D_r/\kappa > 1/5$ the isotropic phase is stable. In contrast, for $D_r/\kappa < 1/5$ the isotropic phase is unstable. In general (i.e.,

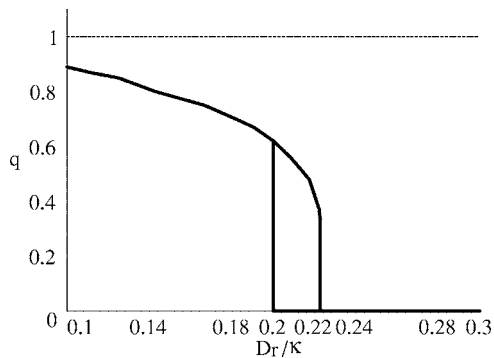


FIG. 2. Bifurcation diagram of the order parameter q of the Maier-Saupe model computed from the self-consistency equation $R'(q)=R(q)-q=0$. Only solutions related to stable stationary phases are shown.

for $q \neq 0$), $K_{A,st}$ can be determined from Fig. 2 [37]. In order to see this we differentiate Eq. (10) with respect to m and evaluate the result at $m=q$. Thus, we get

$$\left. \frac{dR}{dm} \right|_{m=q} = \frac{\kappa}{D_r} K_{A,st}. \quad (20)$$

Therefore, the stability parameter $\tilde{\lambda}$ as a function of q is given by

$$\tilde{\lambda}(q) = D_r \left(1 - \left. \frac{dR(m)}{dm} \right|_{m=q} \right). \quad (21)$$

This expression can be written like $\tilde{\lambda}(q) = -D_r dR'(m)/dm|_{m=q}$. Since q is defined by the relation $R(q)-q=0$ —that is, by $R'(q)=0$ —we obtain

$$\tilde{\lambda}(q=m) = -D_r \left. \frac{dR'(m)}{dm} \right|_{R'(m)=0}. \quad (22)$$

In other words, if the slope of $R'(m)$ at the intersection with the m axis is negative, then $\tilde{\lambda}(q)$ is positive and P_{st} is stable. If the slope of $R'(m)$ at the intersection with the m axis is positive, then $\tilde{\lambda}(q)$ is negative and P_{st} is unstable. From Fig. 1 we see that for $D_r/\kappa \in [1/5, C]$ there are two nonvanishing solutions of $R'(q)=0$ given by $q_1 < q_2$. The solution with q_1 exhibits a positive slope of $R'(m)$ at q_1 and corresponds to an unstable distribution and an unstable nematic phase. For q_2 the slope of $R'(m)$ at $m=q_2$ is negative and, consequently, the solution is related to a stable nonuniform distribution describing a stable nematic phase. For control parameters $D_r/\kappa < 1/5$ the nonvanishing solution q_1 of $R'(q)=0$ yields a negative slope of $R'(m)$ at $m=q_1$ and reflects a nonuniform stationary distribution P_{st} describing a stable nematic phase.

In sum, the stability analysis for the Maier-Saupe model (4) yields the following result: For $D_r/\kappa > C$ there exists only the isotropic phase with $q=0$ and the isotropic phase is stable. For $D_r/\kappa \in [1/5, C]$ an isotropic phase with $q=0$ and two nematic phases with $q \neq 0$ exist. The isotropic phase and the nematic phase with the larger order parameter are stable. The nematic phase with the smaller order parameter is unstable. For $D_r/\kappa < 1/5$ an isotropic phase and a nematic

phase exist. The nematic phase is stable, whereas the isotropic phase is unstable. That is, given small perturbations, there will be a phase transition from the isotropic phase to the nematic phase. Figure 2 shows the order parameter q as a function of the control parameter D_r/κ and summarizes the bifurcation diagram of the Maier-Saupe model (4). Note that in Fig. 2 only q values related to stable phases are shown.

B. Second order statistics

1. Strongly nonlinear Smoluchowski equation and Langevin equations

In general, the second-order statistics of the z component of the director \mathbf{u} is described by the joint probability density $P(x, t; x', t')$. The function $P(x, t; x', t')$ can be computed from the transition probability density $P(x, t|x', t')$ and the transient probability density $P(x, t')$ like $P(x, t; x', t') = P(x, t|x', t')P(x, t')$. Since $P(x, t')$ is defined by Eq. (4), we are left with the problem of determining the evolution of $P(x, t|x', t')$. In order to solve this problem, we first note that the nonlinear Smoluchowski equation (4) involves the drift coefficient $D_1(x, P) = 3M(x)\kappa x q(t)$ with $q(t) = \langle [3x^2 - 1]/2 \rangle$. The explicit evolution of $q(t)$ depends on the initial distribution $w(x)$. For every initial distribution $w(x)$, the drift term $D_1(x, P)$ can be regarded as a time-dependent drift coefficient $D'_1(x, t) = D_1(x, P)$. Therefore, for every w we can assign to the nonlinear Smoluchowski equation a linear Smoluchowski equation with a drift term that depends explicitly on t . Due to this property, we say that the nonlinear Smoluchowski equation belongs to the class of strongly nonlinear Fokker-Planck equations [31,37]. In line with the theory of strongly nonlinear Fokker-Planck equations, we can show that the second-order statistics $P(x, t|x', t')$ is determined by

$$\begin{aligned} \frac{\partial}{\partial t} P(x, t|x', t') = & \frac{\partial}{\partial x} M(x) \left\{ -\frac{9}{2} \kappa x \left(\int x^2 P(x, t; w) dx \right) \right. \\ & \left. - \frac{1}{3} \right\} P(x, t|x', t') + D_r \frac{\partial}{\partial x} P(x, t|x', t'). \end{aligned} \quad (23)$$

Note again that (as mentioned in the Introduction) the variable x describes the z component of the director \mathbf{u} . Consequently, Eq. (23) describes the second-order statistics of the z component of \mathbf{u} .

Let us dwell next on the Langevin equation associated with Eqs. (4) and (23), which will be used below for numerical simulations. Let $X(t) \in [-1, 1]$ denote the random variable of the stochastic process described by Eqs. (4) and (23) with $P(x, t) = \langle \delta(x - X(t)) \rangle$ and $P(x, t; x', t') = \langle \delta(x - X(t)) \delta(x' - X(t')) \rangle$, where $\delta(\cdot)$ denotes the delta function. First, we realize that the diffusion term in Eq. (4) has the so-called Klimontovich form [48,49]. The Klimontovich form can be transformed into the Stratonovich form, which gives us

$$\begin{aligned} \frac{\partial}{\partial t} P(x,t|x',t') = & -\frac{\partial}{\partial x} \left\{ \frac{9}{2} \kappa x M(x) \left(\int x^2 P(x,t;w) dx - \frac{1}{3} \right) \right. \\ & \left. + \frac{D_r dM}{2 dX} \right\} P(x,t|x',t') \\ & + D_r \frac{\partial}{\partial x} \sqrt{M} \frac{\partial}{\partial x} \sqrt{M} P(x,t|x',t'). \end{aligned} \quad (24)$$

From Eq. (24) we read off that $X(t)$ satisfies the self-consistent Stratonovich Langevin equation

$$\begin{aligned} \frac{d}{dt} X(t) = & \frac{9\kappa}{2} M(X) X \left(\langle [X(t)]^2 \rangle - \frac{1}{3} \right) + \frac{D_r dM}{2 dX} \\ & + \underbrace{\sqrt{D_r M(X)} \Gamma(t)}_{\text{Stratonovich}}, \end{aligned} \quad (25)$$

where $\Gamma(t)$ is a Langevin force [50,51] normalized like $\langle \Gamma(t)\Gamma(t') \rangle = 2\delta(t-t')$. Instead of the Stratonovich calculus we will use in the numerics the Ito calculus. Using the identity [52]

$$\underbrace{\sqrt{D_r M(X)} \Gamma(t)}_{\text{Stratonovich}} = \frac{D_r dM}{2 dX} + \underbrace{\sqrt{D_r M(X)} \Gamma(t)}_{\text{Ito}}, \quad (26)$$

we can transform Eq. (25) into

$$\begin{aligned} \frac{d}{dt} X(t) = & \frac{9\kappa}{2} M(X) X \left(\langle [X(t)]^2 \rangle - \frac{1}{3} \right) + D_r \frac{dM}{dX} \\ & + \underbrace{\sqrt{D_r M(X)} \Gamma(t)}_{\text{Ito}}, \end{aligned} \quad (27)$$

see also [37](Sec. 7.2.3). Note that the Ito-Langevin equation can also be derived in an alternative way by writing the Klimontovich form (24) into Ito form

$$\begin{aligned} \frac{\partial}{\partial t} P(x,t|x',t') = & -\frac{\partial}{\partial x} \left\{ \frac{9}{2} \kappa x M(x) \left(\int x^2 P(x,t;w) dx - \frac{1}{3} \right) \right. \\ & \left. + D_r \frac{dM}{dX} \right\} P(x,t|x',t') + D_r \frac{\partial^2}{\partial x^2} M P(x,t|x',t') \end{aligned} \quad (28)$$

and using the Ito calculus to derive Eq. (27) from Eq. (28). For more sophisticated methods to deal with self-consistent Stratonovich- and Ito-Langevin equations the reader is also referred to [16,53]. In closing this section, we would like to point out that the Stratonovich and Ito Langevin equations given by Eqs. (25) and (27), respectively, can also be derived directly as the rotational Brownian walk of a director \mathbf{u} that evolves in a self-consistent potential $e(\mathbf{u}, P)$ (see Sec. II C).

2. Short-time correlations

The objective is now to derive analytical expressions of the autocorrelation function $C(\Delta t) = \langle X(t)X(t+\Delta t) \rangle_{\text{st}}$ and the

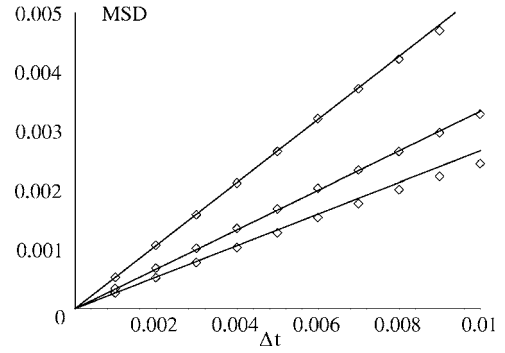


FIG. 3. Mean-square displacement (MSD) $\langle \delta X(\Delta t)^2 \rangle_{\text{st}}$ of the dynamical Maier-Saupe model given by Eqs. (4) and (23) as a function of Δt . Solid line: analytical expression computed from $\langle \delta X(\Delta t)^2 \rangle_{\text{st}} = 4D_r(1-q)\Delta t/3$ [see Eq. (31)] valid for short-time differences Δt . Diamonds: exact result obtained by solving the Maier-Saupe model numerically by means of the corresponding self-consistent Ito-Langevin equation (27) using an Euler forward scheme [50] (single time step 10^{-5} , number of realizations 10 000, random numbers via Box-Muller). Parameters (from bottom to top): $\kappa=5$ ($q=0.60$), $\kappa=6$ ($q=0.75$), and $\kappa=7$ ($q=0.80$). Other parameters: $D_r=1$. Initial distribution $w(x) = [\delta(x-0.8) + \delta(x+0.8)]/2$. Before computing $\langle \delta X(\Delta t)^2 \rangle_{\text{st}}$, the Euler forward scheme was iterated repeatedly until the system settled down in the stationary regime (single time step 10^{-3} , 1000 iterations; the order parameter q was checked to be equivalent to q derived from the self-consistency equation).

mean-square displacement $\langle [\delta X(\Delta t)]^2 \rangle_{\text{st}}$ for small time differences Δt . To this end, we exploit the strongly nonlinear Smoluchowski equation (23). As can be shown by a detailed calculation (see the Appendix), from Eq. (23) it follows that

$$\lim_{\Delta t \downarrow 0} \frac{d}{d\Delta t} C(\Delta t) = -D_r \langle M(X) \rangle_{\text{st}}. \quad (29)$$

Consequently, we have $C(\Delta t) = C(0) - D_r \langle M(X) \rangle_{\text{st}} \Delta t + O(\Delta t^2)$. Using $C(0) = \langle X^2 \rangle_{\text{st}} = (2q+1)/3$ and $\langle M(X) \rangle_{\text{st}} = 1 - \langle X^2 \rangle_{\text{st}} = 2(1-q)/3$, we get

$$C(\Delta t) = \frac{2q+1}{3} - \frac{2D_r(1-q)}{3} \Delta t + O(\Delta t^2). \quad (30)$$

Let $\langle [\delta X(\Delta t)]^2 \rangle_{\text{st}}$ denote the mean-square displacement with $\delta X(\Delta t)$ defined by $\delta X(\Delta t) = X(t+\Delta t) - X(t)$. Then, in general, we have $\langle [\delta X(\Delta t)]^2 \rangle_{\text{st}} = 2[C(0) - C(\Delta t)]$. In the context of the Maier-Saupe model, from Eq. (30) it follows that $\langle [\delta X(\Delta t)]^2 \rangle_{\text{st}} = 2D_r \langle M(X) \rangle_{\text{st}} \Delta t + O(\Delta t^2)$. That is, the mean-square displacement evolves like

$$\langle [\delta X(\Delta t)]^2 \rangle_{\text{st}} = \frac{4D_r(1-q)}{3} \Delta t + O(\Delta t^2). \quad (31)$$

Figure 3 shows $\langle [\delta X(\Delta t)]^2 \rangle_{\text{st}}$ as a function of Δt for several parameters q as obtained from Eq. (31) and as obtained by solving numerically the Ito-Langevin equation (27).

C. Microscopic derivation of Eqs. (25) and (27) from Brownian rotational motion

In what follows, we will exploit the concept of Brownian rotational motion as reviewed, for example, in Doi and Edwards [9] (Sec. 8.2) and Coffey *et al.* [54] (Secs. 1.15 and 7.2). Accordingly, our departure point is the rotational evolution equation of the director \mathbf{u} :

$$\frac{d}{dt}\mathbf{u}(t) = \mathbf{\Omega}(t) \times \mathbf{u}(t), \quad (32)$$

where $\mathbf{\Omega}(t)$ is the angular velocity of the director \mathbf{u} . Let \mathbf{a} describe the angles corresponding to $\mathbf{\Omega}$ like $d\mathbf{a}/dt = \mathbf{\Omega}$. The angles satisfy the second-order Langevin equation

$$I \frac{d^2}{dt^2} \mathbf{a} + \gamma \frac{d}{dt} \mathbf{a} = \mathbf{N} + \sqrt{\gamma k T} \mathbf{\Gamma}, \quad (33)$$

where I is the inertia of the molecule under consideration, γ is a damping coefficient, \mathbf{N} is the torque, and $\mathbf{\Gamma}(t)$ is a Langevin force with $\mathbf{\Gamma}(t) = (\Gamma_1, \Gamma_2, \Gamma_3)$ and $\langle \Gamma_i(t) \Gamma_k(t') \rangle = 2 \delta_{ik} \delta(t - t')$. Note that the noise amplitude is given by $\gamma k T$ [54]—just as in the case of the Kramers equation [50]. As usual, we will consider the overdamped motion. That is, at issue now is to reduce the second-order Langevin equation (33) to a first-order Langevin equation by neglecting the inertia term $I d^2 \mathbf{a} / dt^2$. Roughly speaking, we simply put $I = 0$. A more sophisticated transformation procedure is the Smoluchowski limit [50]. Putting $I = 0$ or using the Smoluchowski limit, from Eq. (33) we obtain

$$\frac{d}{dt} \mathbf{a} = \gamma^{-1} \mathbf{N} + \sqrt{\frac{k T}{\gamma}} \mathbf{\Gamma}. \quad (34)$$

We can rewrite Eq. (34) as

$$\mathbf{\Omega} = \gamma^{-1} \mathbf{N} + \sqrt{\frac{k T}{\gamma}} \mathbf{\Gamma}. \quad (35)$$

The torque is given by $\mathbf{N} = \mathbf{u} \times \mathbf{F}(\mathbf{u}, P)$, where \mathbf{F} is a self-consistent force that may depend on the probability density P . We derive \mathbf{F} from a potential $e(\mathbf{u}, P)$ like $\mathbf{F} = -\partial e(\mathbf{u}, P) / \partial \mathbf{u}$. Substituting these definitions into Eqs. (32) and (35) together with $D_r = k T / \gamma$, we get the self-consistent Stratonovich Langevin equation

$$\frac{d}{dt} \mathbf{u}(t) = -\frac{D_r}{k T} \left[\mathbf{u} \times \frac{\partial}{\partial \mathbf{u}} e(\mathbf{u}, P) \right] \times \mathbf{u} + \sqrt{D_r} \mathbf{\Gamma} \times \mathbf{u}. \quad (36)$$

Now, let us turn to the special case addressed in Sec. I. We have $\mathbf{u} = (u_1, u_2, u_3) = (\sqrt{M(X)} \cos \varphi, \sqrt{M(X)} \sin \varphi, X)$ and $e = e(X, P)$. Our next objective is to evaluate the third component of Eq. (36) describing the evolution of $u_3 = X$. From the equivalence $-\left[\mathbf{u} \times \frac{\partial e}{\partial \mathbf{u}} \right] \times \mathbf{u} = -|\mathbf{u}|^2 \frac{\partial e}{\partial \mathbf{u}} + (\mathbf{u} \cdot \frac{\partial e}{\partial \mathbf{u}}) \mathbf{u}$ and $\frac{\partial e}{\partial \mathbf{u}} \times \mathbf{u} = \frac{\partial e}{\partial X} \mathbf{u}$, we obtain

$$-\left[\mathbf{u} \times \frac{\partial}{\partial \mathbf{u}} e(\mathbf{u}, P) \right] \times \mathbf{u} \Big|_3 = -M(X) \frac{de}{dX}, \quad (37)$$

whereas the term $\mathbf{\Gamma} \times \mathbf{u}$ gives us $\mathbf{\Gamma} \times \mathbf{u} \Big|_3 = \sqrt{M(X)} (\sin \varphi \Gamma_1 - \cos \varphi \Gamma_2)$. In sum, from Eq. (36) we obtain for the component $u_3 = X$ the Stratonovich Langevin equation

$$\frac{d}{dt} X = -D_r \frac{M(X)}{k T} \frac{de}{dX} + \sqrt{D_r M(X)} (\sin \varphi \Gamma_1 - \cos \varphi \Gamma_2). \quad (38)$$

Likewise, we can derive an evolution equation for φ . From $\tan \varphi = u_2 / u_1$ it follows that $M(x) d\varphi / dt = u_1 du_2 / dt - u_2 du_1 / dt$. Exploiting the first and second components of Eq. (36) we obtain

$$\frac{d}{dt} \varphi = -X \sqrt{\frac{D_r}{M(X)}} (\cos \varphi \Gamma_1 + \sin \varphi \Gamma_2) + \sqrt{D_r} \Gamma_3. \quad (39)$$

The main point to make here is that Eqs. (38) and (39) can be cast into the form $dX / dt = h_x + g_{11} \Gamma_1 + g_{12} \Gamma_2$ and $d\varphi / dt = g_{21} \Gamma_1 + g_{22} \Gamma_2 + g_{23} \Gamma_3$. Just as in [50] (Secs. 3.4.1 and 12.1.2), we can determine then from the set of noise amplitudes g_{ik} the drift coefficients D_x and D_φ and the diffusion coefficients D_{xx} and $D_{\varphi\varphi}$ of the X and φ dynamics, respectively. A detailed calculation shows

$$D_x(x, P) = -D_r \frac{M(x)}{k T} \frac{de}{dx} + D_r \frac{dM}{dx}, \quad (40)$$

$$D_{xx}(x) = D_r M(x), \quad (41)$$

$$D_\varphi = 0, \quad (42)$$

$$D_{\varphi\varphi} = D_r, \quad (43)$$

where the drift D_x involves the spurious drift term $D_r dM / dx$. Finally, we exploit the concept of stochastic equivalence, which states that two Langevin equations that look formally different can nevertheless describe the same stochastic process provided that they exhibit the same drift and diffusion coefficients [50]. From Eqs. (40)–(43) it follows that the Stratonovich Langevin equations

$$\frac{d}{dt} X = -D_r \frac{M(X)}{k T} \frac{de}{dX} + \frac{D_r}{2} \frac{dM}{dx} + \underbrace{\sqrt{D_r M(X)} \Gamma_x}_{\text{Stratonovich}}, \quad (44)$$

$$\frac{d}{dt} \varphi = D_r \Gamma_\varphi \quad (45)$$

and the Ito Langevin equations

$$\frac{d}{dt} X = -D_r \frac{M(X)}{k T} \frac{de}{dX} + D_r \frac{dM}{dx} + \underbrace{\sqrt{D_r M(X)} \Gamma_x}_{\text{Ito}}, \quad (46)$$

$$\frac{d}{dt} \varphi = D_r \Gamma_\varphi \quad (47)$$

are stochastically equivalent to Eqs. (38) and (39) for Langevin forces with $\langle \Gamma_i(t) \Gamma_k(t') \rangle = 2 \delta_{ik} \delta(t - t')$ with $i, k \in \{x, \varphi\}$. For $e(x, P)$ given by Eq. (2) the stochastic evolution equations (44) and (46) are equivalent to Eqs. (25) and (27) derived in Sec. II B. In this sense, we have shown that there is a microscopic derivation of the Langevin equations (25) and

(27) based on the director Brownian motion given by Eqs. (32) and (33).

III. CONCLUSIONS

A Smoluchowski equation that is nonlinear with respect to its probability density has been studied in order to describe the orientation of elongated molecules in nematic liquid crystals. The nonlinearity reflects the emergence of a mean-field force that acts on individual molecules and is produced by all molecules as predicted by the Maier-Saupe theory. For phases with cylindrical symmetries, we have studied both the first- and second-order statistical properties of the model. With regard to the first-order statistics, we have carried out a completely analytical bifurcation theory using concepts introduced in earlier studies by Shiino. In doing so, we have been able to determine the stability of both isotropic and nematic phases for several parameter regimes. We have determined three kinds of parameter regimes: a monostable one involving only the isotropic phase and a bistable parameter regime involving a stable isotropic and a stable nematic phase. Finally, we have determined another monostable parameter regime exhibiting only one stable phase: the nematic phase. The critical control parameter at which the isotropic-nematic phase transition from the isotropic phase (of the bistable regime) to the nematic phase (of the second monostable regime) occurs is given simply by $1/5$. With regard to the second-order statistics, the evolution equation for the transition probability density has been derived within the framework of so-called strongly nonlinear Fokker-Planck equations. In particular, analytical expressions for the auto-correlation function and the mean-square displacement have been determined. Following [55,56] one may define the running diffusion coefficient $D^{(z)}$ on the basis of the mean-square displacement $\langle \delta X(\Delta t)^2 \rangle$ like $\langle \delta X(\Delta t)^2 \rangle = \int_0^{\Delta t} D^{(z)} dz$. For the Maier-Saupe model, we then find that the short-time diffusion coefficient $D^{(0)} = \langle \delta X(\Delta t)^2 \rangle / \Delta t$ for $\Delta t \rightarrow 0$ is related to the rotational diffusion coefficient D_r and the order parameter q by $D^{(0)} = 4D_r(1-q)/3$. This implies that in the isotropic phase (i.e., for $q=1$) the short-time diffusion coefficient is given by $D^{(0)} = 4D_r/3$, whereas for a nematic phase with complete alignment of the molecules (i.e., for $q=1$) we have $D^{(0)} = 0$.

APPENDIX: DERIVATION OF EQ. (29)

We proceed similar to a previous study [36]. While in [36] the assumption has been made that surface terms arising due to partial integration vanish (which is usually the case in the context of natural boundary conditions), in this appendix we explicitly show that they vanish in the context of the Maier-Saupe model (which involves periodic boundary conditions). First, we write Eq. (23) as a continuity equation involving the transition probability current $j(x, t|x', t')$:

$$\frac{\partial}{\partial t} P(x, t|x', t') = - \frac{\partial}{\partial x} j. \quad (\text{A1})$$

Multiplying Eq. (A1) with x , integrating with respect to x , and integrating by parts gives us the relation

$$\frac{d}{dt} \langle X(t) \rangle |_{X(t')=x'} = -xj|_{-1}^1 + \int j(x, t|x', t') dx, \quad (\text{A2})$$

which involves the surface term $xj|_{-1}^1$. This surface term vanishes because we have $j(x, t|\cdot) \propto M(x)$ with $M(x=\pm 1)=0$, which implies that the product xj vanishes at $x=\pm 1$. Let us write $j(x, t|x', t') = M(x)h(x, \langle x^2 \rangle)P + D_r \partial[M(x)P]/\partial x$ with $h(x, \langle x^2 \rangle) = 9\kappa x(\langle x^2 \rangle - 1/3)/2$. Substituting this expression into Eq. (A2) and integrating by parts yields

$$\begin{aligned} \frac{d}{dt} \langle X(t) \rangle |_{X(t')=x'} &= \int MhP(x, t|x', t') dx \\ &+ D_r \int \frac{dM}{dx} P(x, t|x', t') dx \\ &- M(x)P(x, t|\cdot) |_{-1}^1. \end{aligned} \quad (\text{A3})$$

The surface term $M(x)P(x, t|\cdot)|_{-1}^1$ vanishes because of $M(x=\pm 1)=0$. Now, let us focus on the stationary case. In this case, we have

$$\begin{aligned} \frac{d}{d\Delta t} C(\Delta t) &= \frac{d}{d\Delta t} \int \langle X(t' + \Delta t) \rangle |_{X(t')=x'} P_{\text{st}}(x') dx' \\ &= \int \frac{d}{dt} \langle X(t) \rangle |_{X(t')=x', t=t'+\Delta t} P_{\text{st}}(x') dx'. \end{aligned} \quad (\text{A4})$$

Substituting Eq. (A3) into Eq. (A4), we obtain

$$\begin{aligned} \frac{d}{d\Delta t} C(\Delta t) &= \int M(x)h(x, \langle x^2 \rangle_{\text{st}})x' P_{\text{st}}(x, t' + \Delta t; x', t') dx dx' \\ &+ D_r \int \frac{dM(x)}{dx} x' P_{\text{st}}(x, t' + \Delta t; x', t') dx dx'. \end{aligned} \quad (\text{A5})$$

In the limit $\Delta t \rightarrow 0+$ we have $P_{\text{st}}(x, t' + \Delta t; x', t') = \delta(x - x')P_{\text{st}}(x)$, which implies that

$$\begin{aligned} \lim_{\Delta t \rightarrow 0+} \frac{d}{d\Delta t} C(\Delta t) &= \int M(x)h(x, \langle x^2 \rangle_{\text{st}})x P_{\text{st}}(x) dx \\ &+ D_r \int \frac{dM(x)}{dx} x P_{\text{st}}(x) dx. \end{aligned} \quad (\text{A6})$$

In the stationary case, from Eq. (4) it follows that $Mh(x, \langle x^2 \rangle_{\text{st}})P_{\text{st}} = D_r M dP_{\text{st}}/dx$. Multiplying this relation with x and integrating with respect to x , we obtain

$$\begin{aligned} \int M(x)h(x, \langle x^2 \rangle_{\text{st}})x P_{\text{st}}(x) dx &= -D_r \int \frac{d[xM(x)]}{dx} P_{\text{st}}(x) dx \\ &+ D_r xM(x)P_{\text{st}}|_{-1}^1. \end{aligned} \quad (\text{A7})$$

The surface term $D_r xM(x)P_{\text{st}}|_{-1}^1$ vanishes because of $M(x=\pm 1)=0$. Substituting Eq. (A7) into Eq. (A6), we obtain Eq. (29).

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