## Stochastic envelope equations for nonequilibrium transitions and application to thermal fluctuations in electroconvection in nematic liquid crystals

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Near the threshold of continuous nonequilibrium transitions in spatially extended pattern-forming systems thermal fluctuations are enhanced in analogy to equilibrium phase transitions. These fluctuations anticipate the deterministic pattern above threshold. A Langevin-equation approach based on Landau's method to determine the stochastic terms in hydrodynamic systems is presented and applied to fluctuations in electrohydrodynamic convection in nematic liquid crystals. The resulting set of equations is then transformed into a universal stochastic envelope (or amplitude) equation of the Ginzburg-Landau type, valid near threshold. The resulting fluctuations are compared with recent experiments and with an estimate based on equilibrium theory. The methods are formulated in a general way so that application to other pattern-forming systems is readily possible.

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## I. INTRODUCTION

The influence of thermal fluctuations on patternforming instabilities in extended nonequilibrium systems near the primary bifurcation has recently received renewed attention, both experimentally and theoretically.

Fluctuations were studied experimentally in the context of Rayleigh-Bénard convection (RBC) in simple liquids [1], in binary mixtures [2], and in gases [3]. Furthermore, fluctuations in Taylor-Couette flow (TCF) [4, 5] and director fluctuations in electrohydrodynamic convection (EHC) [6-8] were measured. Theoretical estimates show that EHC and RBC in gases are particularly suited to measure thermal fluctuations directly. In fact, in the other experiments some kind of noise amplification mechanism was used [9]: In RBC the response to a time-dependent temperature difference, changing from a subcritical to a supercritical value, was measured; in TCF throughflow was added in order to allow for convective amplification of fluctuations in the convectively unstable region [4, 10]; in RBC in binary mixtures near the Hopf bifurcation one naturally has a convectively unstable region [2].

Theoretical predictions for the effect of thermal fluctuations in pattern-forming systems were first given for RBC in simple liquids [11-14]. There is a large and as yet unexplained discrepancy with the experiments of Ref. [1]. More recently fluctuations in RBC in binary mixtures [15], in TCF with throughflow [4, 16-18], in RBC with throughflow [19], and in EHC [8, 20] have been considered.

In the most general theoretical approach the macroscopic dynamic equations are supplemented with noise terms which account for the microscopic degrees of freedom and whose stochastic properties are given by the fluctuation-dissipation theorem (FDT) [21]. This method, which was introduced by Landau and Lifshitz [22, 23] to calculate fluctuations in bulk samples of simple liquid, was used for RBC and also for the description of transient patterns in the magnetic Fréedericksz transition in nematic liquid crystals [20]. Near threshold, the resulting system can then often be reduced to a stochastic generalization of the usual envelope (or amplitude) equations, and one obtains the macroscopic (or mesoscopic) fluctuations by solving these equations. Sometimes the stochastic forces are replaced by stochastic initial conditions determined from the equilibrium fluctuations without driving force. This method, which is useful in systems with an intrinsic amplification mechanism for fluctuations, was applied to the TCF system with throughflow [4, 10]. In addition, reasonable (but *ad hoc*) assumptions were used for estimates. Thus an estimate for director fluctuations in EHC was given by Rehberg et al. [8] by extrapolating the equilibrium fluctuations (without electric field), which are given by the equipartition theorem, to nonzero external electrical fields. For TCF the fluctuation strength of the stochastic envelope equation was assumed to be similar to that for RBC [4].

In the general method the main assumption is that the stochastic forces have the same intensity outside of equilibrium as in equilibrium and therefore can be calculated using the Langevin formulation of the FDT [21, 24, 25]. This generalization should be valid as long as the external forces (shear, temperature gradient, electrical fields, etc.), which drive the system out of equilibrium, are small compared to the internal fields effective on a molecular scale, i.e., the system is everywhere near local equilibrium. The method consists of two steps: derivation of the stochastic terms of the macroscopic equations and reduction to stochastic envelope equations near threshold. Solutions of the latter equations giving the fluctuations of the amplitudes have been presented numerically or analytically for the most relevant cases [26], so the derivation of the fluctuating forces of the envelope equations can be considered as the crucial step.

In this paper we give a general description of the

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scheme leading from macroscopic stochastic equations to stochastic amplitude equations and we derive the stochastic equations for EHC as important example. In Sec. II the derivation of stochastic envelope equations from given dynamical equations and boundary conditions is presented for a large class of systems including all the above examples: Starting from the autonomous case (time-independent external driving) with only one critical mode at threshold [27, 28] the derivation is generalized to systems with a rapid periodic time dependence such as the usual ac-driven EHC [29] and also to systems with more than one degenerated mode at threshold such as left- and right-traveling waves, oblique rolls, or twodimensional isotropic systems. Finally we discuss briefly fluctuations above threshold. In Sec. III we derive the stochastic terms of the standard set of EHC equations for a planarly aligned nematic slab, [29] and specialize the resulting expressions to planar boundary conditions and normal rolls in the conductive (low external ac frequency) regime. In Sec. IV the fluctuating forces of the amplitude equation in the normal-roll regime are calculated and the resulting equal-time director fluctuations are compared with recent measurements of the fluctuation intensity and with the estimate of Ref. [8]. We also investigate the continuous transition from normal rolls to oblique rolls, occurring in EHC at the Lifshitz point [29], and predict an interesting crossover of the exponents of the fluctuation intensity and correlation length near the Lifshitz frequency. Section V gives a summary and a discussion.

## II. GENERAL REDUCTION SCHEME TO DETERMINE THE NOISE TERM OF THE STOCHASTIC ENVELOPE EQUATIONS

#### A. Starting point

The dynamics of large classes of one- and twodimensionally extended pattern-forming systems, including all systems mentioned in the introduction, can be cast into the following set of symbolic equations for the relevant macroscopic variables  $\boldsymbol{u}(\boldsymbol{r},t)$ , defined for real x, yand  $z \in [-d/2, d/2]$ :

$$[\underline{S}(\nabla, R, t)\partial_t + \underline{L}(\nabla, R, t)] u(r, t)$$
$$= N(u, \nabla, R, t) + \xi(r, t), \quad (1)$$

. . . .

$$\langle \xi_{\alpha}(\boldsymbol{r},t) \rangle = 0,$$
  
 $\langle \xi_{\alpha}(\boldsymbol{r},t)\xi_{\beta}(\boldsymbol{r}',t') \rangle = O_{\alpha\beta}(\boldsymbol{\nabla},\boldsymbol{u})\delta(\boldsymbol{r}-\boldsymbol{r}')\delta(t-t').$  (2)

The components  $u_{\alpha}$  of u represent, e.g., velocities, temperature, director components, etc. They are formulated as deviations from the basic unstructured state, i.e., N(u = 0) = 0, so that u = 0 is always a solution of the deterministic part of the system. The matrix-differential operators  $\underline{S}$  and  $\underline{L}$  contain the linear and N the nonlinear parts of the dynamics. They depend on external control parameters R. The components of  $\boldsymbol{\xi}$ 

represent stochastic forces. Since they arise from coupling to the microscopic degrees of freedom, their second moments can be assumed to be  $\delta$  correlated in space and time [31], and by virtue of the central limit theorem the distribution function itself is Gaussian. In the case of thermal noise, the Hermitian correlation matrix  $\boldsymbol{O}$  is determined by the fluctuation-dissipation theorem and can be written as  $\underline{O} = \underline{D}(\nabla)\underline{K}(u)\underline{D}^{T}(-\nabla)$ , where  $\underline{K}$  denotes a symmetric matrix and  $\underline{D}$  is a matrix-differential operator (see Sec. III). The system is considered to be translational invariant in the (x, y) plane (large aspect ratio limit), i.e,  $\underline{S}, \underline{L}, N$ , and  $\underline{O}$  do not depend explicitly on  $\boldsymbol{x} = (x, y)$ . In the z direction appropriate boundary conditions at  $z = \pm d/2$  are assumed. In general  $\underline{S}$ ,  $\underline{L}$ , and N are allowed to depend periodically on time. Adaptation of our treatment to a quasi-one-dimensional situation is straightforward and will be used freely. The system is assumed to depend on control parameters  $\boldsymbol{R}$  so that there are regions in parameter space where the unstructured state u=0 is stable and other regions where the system is linearly unstable with respect to certain classes m of modes with wave numbers  $\boldsymbol{k}^{(m)}$  in the (x, y)plane. The index m characterizes the z dependence. For each class the condition of neutral stability, where the mode becomes unstable for the first time, defines a threshold surface in parameter space and a critical wave number  $k_c^{(m)}$ , not necessarily unique. The nonlinearities are supposed to act in a stabilizing way (forward bifurcation). This allows the description of the near-threshold dynamics by envelope equations involving only the amplitudes of the critical modes [32]. In the following we present general expressions for the noise strength of the stochastic envelope equations for several relevant equivalence classes of critical solutions of (1). The derivation proceeds from the simplest case to the more complicated ones: (i) autonomous systems below threshold, with no degeneracy at the absolute threshold, except for translational invariance (in Fourier space this case was treated previously [27, 28]); (ii) generalization to nonautonomous systems ("rapid" time-periodic driving); (iii) inclusion of degeneracies of the critical modes, e.g., systems exhibiting right- and left-traveling waves or isotropic systems with  $k_c \neq 0$ ; and (iv) fluctuations above threshold.

# B. Autonomous nondegenerate systems below threshold

This case excludes isotropic systems that are extended in both x and y as well as Hopf bifurcations in cases with reflexion symmetry. Nevertheless one may have spontaneous periodic motion at threshold as a result of throughflow applied externally, see, e.g., [4, 5, 19]. Because of translational invariance, a Fourier transform

$$\boldsymbol{u}(\boldsymbol{r},t) = \int d^2 \boldsymbol{k} e^{i\boldsymbol{k}\cdot\boldsymbol{x}} \overline{\boldsymbol{u}}_{\boldsymbol{k}}(z,t),$$
$$\overline{\boldsymbol{u}}_{\boldsymbol{k}}(z,t) = \frac{1}{(2\pi)^2} \int d^2 \boldsymbol{x} e^{-i\boldsymbol{k}\cdot\boldsymbol{x}} \boldsymbol{u}(\boldsymbol{r},t), \qquad (3)$$

with  $\boldsymbol{x} = (x, y)$  and  $\boldsymbol{k} = (k_x, k_y)$ , diagonalizes the linear

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part of the problem with respect to the  $\boldsymbol{x}$  dependence

$$[\underline{S}(i\boldsymbol{k},\partial_{z},\boldsymbol{R})\partial_{t}+\underline{L}(i\boldsymbol{k},\partial_{z},\boldsymbol{R})]\,\overline{\boldsymbol{u}}_{\boldsymbol{k}}(z,t)=\overline{\boldsymbol{\xi}}_{\boldsymbol{k}}(z,t).$$
(4)

The expectation values of the Fourier-transformed fluctuation strengths are given by

$$\begin{split} \langle \bar{\boldsymbol{\xi}}_{\boldsymbol{k}\alpha}(z,t) \rangle &= \langle \bar{\boldsymbol{\xi}}_{\boldsymbol{k}\alpha}(z,t) \bar{\boldsymbol{\xi}}_{\boldsymbol{k}'\beta}(z',t') \rangle \\ &= \langle \bar{\boldsymbol{\xi}}_{\boldsymbol{k}\alpha}^{*}(z,t) \bar{\boldsymbol{\xi}}_{\boldsymbol{k}'\beta}^{*}(z',t') \rangle = 0, \\ \langle \bar{\boldsymbol{\xi}}_{\boldsymbol{k}\alpha}^{*}(z,t) \bar{\boldsymbol{\xi}}_{\boldsymbol{k}'\beta}(z',t') \rangle &= \frac{1}{(2\pi)^{2}} \left[ \underline{O}(i\boldsymbol{k},\partial_{z},\boldsymbol{u}=0) \right]_{\alpha\beta} \\ &\times \delta(\boldsymbol{k}-\boldsymbol{k}') \delta(z-z') \delta(t-t'). \end{split}$$

$$\end{split}$$

$$(5)$$

With the ansatz  $\overline{\boldsymbol{u}}_{k}(z,t) = e^{\sigma t} \boldsymbol{f}_{k}(z)$ , the deterministic part of (4) represents an eigenvalue problem for the (in general complex) growth rate  $\sigma$ . The corresponding eigenfunctions  $\boldsymbol{f}_{k}^{(m)}(z,\boldsymbol{R})$  depend on the wave vector  $\boldsymbol{k}$ , the branch index m which characterizes the z dependence, and the control parameters. Since the system is supposed to have no degeneracies, only modes from one branch become critical [ $\operatorname{Re}(\sigma) \to 0$ ] at threshold. Then the solution of the linear stochastic problem near threshold can be cast into the form

$$\overline{\boldsymbol{u}}_{\boldsymbol{k}}(z,t) = \psi_{\boldsymbol{k}}(t)\boldsymbol{f}_{\boldsymbol{k}}(z) .$$
(6)

No bicritical or tricritical points are considered here (see Sec. IID), so we suppress the  $\mathbf{R}$  dependence apart from one control parameter, say  $R_1$ , and define the distance from threshold  $\epsilon := (R_1 - R_{1c})/R_{1c}$ .

By projecting (4) onto the respective eigenvector  $f_k^{\dagger}$  of the adjoint linear problem, one obtains with (6) a Langevin equation for the mode amplitudes

$$\partial_t \psi_k = \sigma(\mathbf{k}, \epsilon) \psi_k + \overline{\Gamma}_k(t, \epsilon), \tag{7}$$

$$\sigma(\mathbf{k}, \epsilon) = \frac{-(\mathbf{f}_k^{\dagger}, \underline{L}(i\mathbf{k}, \partial_z, \epsilon)\mathbf{f}_k)}{(\mathbf{f}_k^{\dagger}, \underline{S}(i\mathbf{k}, \partial_z, \epsilon)\mathbf{f}_k)},$$

$$\overline{\Gamma}_k(t, \epsilon) = \frac{(\mathbf{f}_k^{\dagger}, \overline{\xi}_k)}{(\mathbf{f}_k^{\dagger}, \underline{S}(i\mathbf{k}, \partial_z, \epsilon)\mathbf{f}_k)}, \tag{8}$$

with the scalar product  $(\phi, \psi) := \int_{-d/2}^{d/2} dz \phi_{\alpha}^{*}(z) \psi_{\alpha}(z)$ , where summation over dummy indices is implied. The second moments of the projected stochastic forces can be calculated using (5)

$$\langle \overline{\Gamma}_{k}^{*}(t)\overline{\Gamma}_{k}^{\prime}(t^{\prime})\rangle = \frac{1}{(2\pi)^{2}}Q_{k}\delta(\mathbf{k}-\mathbf{k}^{\prime})\delta(t-t^{\prime}),$$
$$Q_{k} = \frac{(\boldsymbol{f}_{k}^{\dagger}, \underline{O}(i\boldsymbol{k},\partial_{z},\boldsymbol{u}=\boldsymbol{0})\boldsymbol{f}_{k}^{\dagger})}{\left|(\boldsymbol{f}_{k}^{\dagger}, \underline{S}(i\boldsymbol{k},\partial_{z},\epsilon)\boldsymbol{f}_{k})\right|^{2}}.$$
(9)

Direct solution of Eq. (7) then leads to

$$\langle \psi_{\boldsymbol{k}}^{*}(t)\psi_{\boldsymbol{k}}(t')\rangle = -\frac{1}{2(2\pi)^{2}}\frac{Q_{\boldsymbol{k}}}{\operatorname{Re}}(\sigma)e^{\operatorname{Re}(\sigma)|t-t'|-i\operatorname{Im}(\sigma)(t-t')}.$$
(10)

[Note that  $\operatorname{Re}(\sigma) < 0$  below threshold.] In nondegenerate systems, modes from a vicinity  $G_k$  of only one pair of wave vectors  $\pm \mathbf{k}_c$  become critical. Therefore the solution in real space is approximately

$$\boldsymbol{u}(\boldsymbol{r},t) = \int_{\boldsymbol{G}_{\boldsymbol{k}_{c}}} d^{2}\boldsymbol{k}\psi_{\boldsymbol{k}}(t)\boldsymbol{f}_{\boldsymbol{k}}(z)e^{\boldsymbol{i}\boldsymbol{k}\cdot\boldsymbol{x}} + \text{c.c.}$$
(11)

Comparing this with the usual definition for the envelope  ${\cal A}$ 

 $\boldsymbol{u}(\boldsymbol{r},t) = A(\boldsymbol{x},t)\boldsymbol{f}(z)e^{i(k_c\cdot\boldsymbol{x}+\omega_ct)} + \mathrm{c.c.} + \mathrm{h.o.t.}$ 

 $\omega_c = \operatorname{Im}(\sigma_c), \quad (12)$ 

(h.o.t. denotes higher-order terms) one obtains with Eq. (7) in Fourier space for wave numbers near the critical one  $(|\boldsymbol{q}| \ll |\boldsymbol{k}_c|)$ 

$$\partial_t \overline{A}_q = [\sigma(\boldsymbol{k}_c + \boldsymbol{q}, \epsilon) - i\omega_c] \overline{A}_q + \overline{\Gamma}(t), \qquad (13)$$

where the approximations  $\overline{\Gamma}_{k_c+q}(t,\epsilon) \approx \overline{\Gamma}_{k_c}(t,\epsilon=0) := \overline{\Gamma}(t)$  and  $f_{k_c+q}(z,\epsilon) \approx f_{k_c}(z,\epsilon=0) := f(z)$ , valid near threshold, are invoked. In real space one obtains

$$\partial_{t} A(\boldsymbol{x}, t) = [\sigma(\boldsymbol{k}_{c} - i\boldsymbol{\nabla}_{2}, \epsilon) \\ -i\omega_{c}]A(\boldsymbol{x}, t) + \Gamma(\boldsymbol{x}, t), \qquad (14)$$
$$\langle \Gamma \rangle = \langle \Gamma \Gamma \rangle = \langle \Gamma^{*} \Gamma^{*} \rangle = 0,$$

$$\langle \Gamma^*(\boldsymbol{x},t)\Gamma(\boldsymbol{x}',t')\rangle = Q\delta(\boldsymbol{x}-\boldsymbol{x}')\delta(t-t'), \qquad (15)$$

$$Q = Q_{k_c}(\epsilon = 0), \tag{16}$$

with  $\nabla_2 := (\partial_x, \partial_y)$ . The integration in Eq. (11) was extended over all space, which is usually justified near threshold.

Fluctuations of the physical quantities are related to the fluctuations of A by virtue of the definition (12)  $(\Delta \boldsymbol{x} = \boldsymbol{x} - \boldsymbol{x}', \Delta t = t - t')$ 

$$\langle u_{\alpha}(\boldsymbol{r},t)u_{\beta}(\boldsymbol{r}',t')\rangle = 2\operatorname{Re}\{\langle A^{*}(\Delta\boldsymbol{x},\Delta t)A(0,0)\rangle f_{\alpha}^{*}(z)f_{\beta}(z')\}\cos(\boldsymbol{k}_{c}\cdot\Delta\boldsymbol{x}+\omega_{c}\Delta t) \\ + 2\operatorname{Im}\{\langle A^{*}(\Delta\boldsymbol{x},\Delta t)A(0,0)\rangle f_{\alpha}^{*}(z)f_{\beta}(z')\}\sin(\boldsymbol{k}_{c}\cdot\Delta\boldsymbol{x}+\omega_{c}\Delta t).$$

$$(17)$$

Correlations of A are obtained by solving Eqs. (14) with (15); see, e.g., [15]. For the equal-time correlations one has

$$\langle AA \rangle = \langle A^*A^* \rangle = 0,$$
  
$$\langle A^*(\Delta \boldsymbol{x}, t)A(0, t) \rangle = -\frac{1}{(2\pi)^2} \int d^2 \boldsymbol{q} \frac{e^{i\boldsymbol{q}\cdot\Delta \boldsymbol{x}}Q}{2\operatorname{Re}\{\sigma(\boldsymbol{k}_c + \boldsymbol{q}, \epsilon)\}}.$$
(18)

Near threshold, analytical expressions can be obtained by substituting for  $\sigma(\mathbf{k}, \epsilon)$  its Taylor expansion around the critical point to lowest nontrivial order  $O(\epsilon) = O[(\mathbf{k} - \mathbf{k}_c)^2]$ ,

$$\sigma(\mathbf{k}_{c} - i\nabla_{2}, \epsilon) \approx i\omega_{c} + \sigma_{\epsilon}\epsilon - i(\sigma_{\mathbf{k}_{x}}\partial_{x} + \sigma_{\mathbf{k}_{y}}\partial_{y}) \\ -\frac{1}{2}\sigma_{\mathbf{k}_{x}\mathbf{k}_{x}}\partial_{x}^{2} - \sigma_{\mathbf{k}_{x}\mathbf{k}_{y}}\partial_{x}\partial_{y} - \frac{1}{2}\sigma_{\mathbf{k}_{y}\mathbf{k}_{y}}\partial_{y}^{2},$$

$$(19)$$

with the (complex) coefficients  $\sigma_{\epsilon} = \frac{\partial \sigma}{\partial \epsilon}\Big|_{\text{crit}}, \sigma_{k_{x}} = \frac{\partial \sigma}{\partial k_{x}}\Big|_{\text{crit}}, \sigma_{k_{y}} = \frac{\partial \sigma}{\partial k_{y}}\Big|_{\text{crit}}, \sigma_{k_{x}k_{x}} = \frac{\partial^{2}\sigma}{\partial k_{x}^{2}}\Big|_{\text{crit}}, \sigma_{k_{x}k_{y}} = \frac{\partial^{2}\sigma}{\partial k_{x}\partial k_{y}}\Big|_{\text{crit}}, \text{ and } \sigma_{k_{y}k_{y}} = \frac{\partial^{2}\sigma}{\partial k_{y}^{2}}\Big|_{\text{crit}}, \text{ where crit denotes}$ ( $\mathbf{k} = \mathbf{k}_{c}, \epsilon = 0$ ). Note that the physical fluctuations (17), calculated with (18) and (16), are invariant with respect to the normalization chosen for  $\mathbf{f}$  and  $\mathbf{f}^{\dagger}$ .

#### C. Generalization to periodically driven systems

We start again from an equation of the form (4), but  $\underline{S}$  and  $\underline{L}$  are now allowed to be periodic in time through the control parameters. Suppressing the index k and the dependence on z and  $\mathbf{R}$ , it reads

$$[\underline{\boldsymbol{S}}(t)\partial_t + \underline{\boldsymbol{L}}(t)]\,\boldsymbol{u}(t) = \boldsymbol{\xi}(t),\tag{20}$$

where  $\underline{S}(t + 2\pi/\omega_0) = \underline{S}(t)$ ,  $\underline{L}(t + 2\pi/\omega_0) = \underline{L}(t)$ , and  $\langle \xi^*_{\alpha}(t)\xi_{\beta}(t') \rangle = O_{\alpha\beta}\delta(t-t')$ . (Here we assume the noise strength to be independent of t.) From Floquet theory one knows that the mode solutions of the deterministic part of (20) can be written as  $u(t) = e^{\sigma t}w(t)$ , where w is  $2\pi/\omega_0$  periodic. Expanding  $\underline{S}$ ,  $\underline{L}$ , and w in the form  $x = x^{(n)}e^{in\omega_0 t}$  with  $x = \underline{S}$ ,  $\underline{L}$ , or w (summation over doubly occurring indices is implied), and projecting (20) onto  $e^{il\omega_0 t}$  yields

$$\left[ (\sigma + im\omega_0) \underline{S}^{(l-m)} + \underline{L}^{(l-m)} \right] \boldsymbol{w}^{(m)} = \boldsymbol{\xi}^{(l)}$$
(21)

with  $\boldsymbol{\xi}^{(l)} = \frac{\omega_0}{2\pi} \int_0^{2\pi/\omega_0} dt \boldsymbol{\xi}(t) e^{-(\sigma+il\omega_0)t}$ . To determine the strength  $\langle \boldsymbol{\xi}_{\alpha}^{\star(l)} \boldsymbol{\xi}_{\beta}^{(l')} \rangle$  of the fluctuating forces, we write  $\sigma$  as  $s + i\omega$  ( $s, \omega$  real) and assume  $|s| \ll \omega_0$ , which becomes exact at the critical point. In addition, our restriction to the linear part imposes the condition that  $\boldsymbol{u}$  must always remain small. (See Ref. [33].)  $\boldsymbol{\xi}^{(l)}$  can then be identified with the Fourier component at frequency  $\omega + l\omega_0$  of the white noise  $\boldsymbol{\xi}(t)$ , which yields  $\langle \boldsymbol{\xi}_{\alpha}^{\star(l)} \boldsymbol{\xi}_{\beta}^{(l')} \rangle = O_{\alpha\beta} \delta_{ll'}$ . To see the explicit structure of (21), we write the dynamical variables  $\boldsymbol{w}^{(m)}$  as a vector  $\tilde{\boldsymbol{w}} := (\dots, \boldsymbol{w}^{(-1)}, \boldsymbol{w}^{(0)}, \boldsymbol{w}^{(1)}, \dots)$  and the stochastic variables  $\boldsymbol{\xi}^{(m)}$  as  $\tilde{\boldsymbol{\xi}} := (\dots, \boldsymbol{\xi}^{(-1)}, \boldsymbol{\xi}^{(0)}, \boldsymbol{\xi}^{(1)}, \dots)$ , leading to

$$(\underline{T}\sigma + \underline{A})\,\tilde{\boldsymbol{w}} = \tilde{\boldsymbol{\xi}} \tag{22}$$

with

$$T_{\alpha\beta}^{(n,m)} = S_{\alpha\beta}^{(n-m)},\tag{23}$$

$$A_{\alpha\beta}^{(n,m)} = im\omega_0 S_{\alpha\beta}^{(n-m)} + L_{\alpha\beta}^{(n-m)}, \qquad (24)$$

$$\langle \tilde{\xi}^{*(n)}_{\alpha} \tilde{\xi}^{(m)}_{\beta} \rangle = O_{\alpha\beta} \delta_{nm}.$$
<sup>(25)</sup>

With the ansatz  $\tilde{\boldsymbol{u}}(t) := e^{\sigma t} \tilde{\boldsymbol{w}}$  this is equivalent to

$$[\underline{\boldsymbol{T}}\partial_t + \underline{\boldsymbol{A}}]\,\tilde{\boldsymbol{u}}(t) = \tilde{\boldsymbol{\xi}}(t), \tag{26}$$

$$\langle \tilde{\xi}^{*(n)}_{\alpha}(t) \tilde{\xi}^{(m)}_{\beta}(t') \rangle = O_{\alpha\beta} \delta_{nm} \delta(t-t').$$
<sup>(27)</sup>

This autonomous Langevin equation has the form of Eq.

(4). Together with the representation

$$u_{\alpha}(t) = \tilde{u}_{\alpha}^{(n)}(t)e^{in\omega_0 t},\tag{28}$$

the problem is reduced to the autonomous case of Sec. IIB.

#### D. Systems with degenerate critical modes

In many pattern-forming systems more than one linear solution become simultaneously unstable at threshold. Some examples are (i) traveling or standing waves in systems with reflection symmetry (i.e, no drift or throughflow)

$$\boldsymbol{u}_{c}^{(\pm)}(\boldsymbol{r},t) = \boldsymbol{f}(z)e^{i(\boldsymbol{k}_{cz}\boldsymbol{x}\pm\boldsymbol{\omega}_{c}t)} + \text{c.c.} , \qquad (29)$$

(ii) stationary oblique rolls or rectangles in systems with axial anisotropy

$$u_{c}^{(\pm)}(\mathbf{r},t) = f(z)e^{i(k_{cx}x\pm k_{cy}y)} + \text{c.c.}$$
, (30)

(iii) two-dimensional isotropic systems with a transition to stationary periodic patterns, and (iv) various types of codimension-two bifurcations. The above degeneracies follow either directly from the spontaneously broken symmetries [examples (i)-(iii)] or result from "accidental" coincidence of the real parts of the two most unstable eigenvalues for special points in parameter space [example (iv)]. The number of degenerated modes is either finite, breaking a discrete symmetry [examples (i) and (ii)] or infinite, breaking a continuous symmetry [example (iii)]. One can often make a continuous transition from the degenerate to the nondegenerate case, e.g.,  $\omega_c = \operatorname{Im}(\sigma_c) \rightarrow 0$  in example (i),  $k_{cy} \rightarrow 0$  in example (ii), by varying a second control parameter. Such points also have special properties and will be considered below. The above examples can be combined, resulting, e.g., in oblique traveling waves [34] or the codimension-two bifurcation in binary mixtures where a stationary solution and a pair of waves become simultaneously critical [15]. In the rest of this subsection we treat the most important cases.

## 1. Discrete symmetry breaking of critical modes: Oblique rolls and traveling waves

We denote the amplitude of the *m*th critical mode by  $A^{(m)}$  so that the physical quantities are related to the amplitudes by a generalization of (12)

$$u = \sum_{m} A^{(m)}(x, t) f^{(m)}(z) e^{i(k_c^{(m)} \cdot x + \omega_c^{(m)} t)} + \text{c.c.} + \text{h.o.t.}, \qquad (31)$$

$$\partial_t A^{(m)} = \left[ \sigma(\boldsymbol{k}_c^{(m)} - i\boldsymbol{\nabla}_2, \epsilon) - i\omega_c^{(m)} \right] A^{(m)} + \Gamma^{(m)}. \quad (32)$$

The deterministic parts of the envelope equations are closely related to each other and we may assume that the eigenfunctions are identical for all critical modes, which yields identical fluctuating forces and fluctuation correlations for all  $A^{(m)}$ . We here assume that the different modes are sufficiently well separated such as, e.g., left- and right-traveling waves with sufficiently high frequency or oblique rolls with sufficient angle of obliqueness, so that the Fourier-transformed wave packets associated with these modes do not overlap in  $(\omega, \mathbf{k})$  space

(this restriction will be relaxed below). The fluctuating forces are then uncoupled and determined by (16) with the respective eigenfunctions  $f^{(m)}(z)$  and critical wave vectors [35]. Equation (17) then becomes

$$\langle \boldsymbol{u}_{\alpha}(\boldsymbol{r},t)\boldsymbol{u}_{\beta}(\boldsymbol{r}',t')\rangle = \langle A^{*}(\boldsymbol{x},t)A(\boldsymbol{x}',t')\rangle f_{\alpha}(z)f_{\beta}(z') \times \begin{cases} 1 & (\text{unstructured transition})\\ 2\cos k_{cx}\Delta x & (\text{normal rolls})\\ 4\cos k_{cx}\Delta x\cos k_{cy}\Delta y \text{ (oblique rolls)}\\ 4\cos k_{cx}\Delta x\cos \omega_{c}\Delta t & (\text{traveling waves}). \end{cases}$$
(33)

Note that the sine terms of (17) vanish due to symmetry and that in contrast to each separate mode the correlations exhibit the full symmetry of the system.

## 2. Continuous symmetry breaking of critical modes: Isotropic systems

In this case it is preferable to extract from the critical solutions only the z dependence and to define (for a stationary bifurcation) a real amplitude  $\psi$  as

$$\boldsymbol{u} = \boldsymbol{\psi}(\boldsymbol{x}, t) \boldsymbol{f}(z). \tag{34}$$

This ansatz leads with the same approximations as in Sec. IIB to

$$\partial_t \psi(\boldsymbol{x}, t) = \sigma(\boldsymbol{\nabla}_2^2, \epsilon) \psi + \Gamma(\boldsymbol{x}, t),$$
(35)

$$\sigma(\boldsymbol{k}^2,\epsilon) = \sigma_{\epsilon}\epsilon + \frac{1}{2}\sigma_{\boldsymbol{k}\boldsymbol{k}}(|\boldsymbol{k}| - k_c)^2 + O(|\boldsymbol{k}| - k_c)^3, \quad (36)$$

with the fluctuating force given by (16), calculated at  $|\mathbf{k}| = k_c$ .

Since the eigenfunctions  $\boldsymbol{f}$  and  $\boldsymbol{f}^{\dagger}$  are taken at the critical wave number, Eqs. (34)– (36) are valid only near threshold. Therefore it is consistent to replace the actual growth rate by a generic expression which has the same lowest-order Fourier expansion as (36). The choice  $\sigma = \sigma_{\epsilon} [\epsilon - \xi_0^4 (\boldsymbol{\nabla}_2^2 + k_c^2)^2]$  with  $\sigma_{\epsilon} \xi_0^4 = \sigma_{kk}/(8k_c^2)$  leads to a stochastic Swift-Hohenberg model for isotropic systems with an instability at  $\boldsymbol{k}_c \neq \boldsymbol{0}$ . For Rayleigh-Bénard convection in simple fluids the strength of the fluctuations was first calculated by Swift and Hohenberg by a different scheme [13]; for arbitrary isotropic systems the strength is given by (16).

## 3. Continuous transition between symmetries by varying a second control parameter: Transition from normal to oblique rolls in systems with axial anisotropy

According to Eq. (33) intensity and correlation of the physical fluctuations depend on symmetry and degeneracy of the critical solutions. Now the question arises in which way the fluctuations behave in systems where the symmetry of the critical solutions can be changed continuously by varying a second control parameter. This scenario occurs for instances in EHC at the Lifshitz point [36, 29], the frequency  $\omega_0$  of the driving voltage usually being the second control parameter. Equations (14) and (12) remain valid in the vicinity of the Lifshitz point  $(\omega_z, \epsilon = 0)$  if one relates the amplitude to normal rolls and includes in the Taylor expansion (19) lowest-order terms in  $\Delta \omega_0 := \omega_0 - \omega_z$  assuming  $O(\epsilon) = O(q)^2 =$  $O(p^4) = O(p^2 \Delta \omega_0)$  with  $\mathbf{k} = (k_c, 0) + (q, p)$ 

$$\sigma = \sigma_{\epsilon}\epsilon + \frac{1}{2}\sigma_{qq}q^{2} + \frac{1}{2}\sigma_{pp\omega}p^{2}\Delta\omega_{0} + \frac{1}{2}\sigma_{qpp}qp^{2} + \frac{1}{4!}\sigma_{pppp}p^{4},$$
(37)

where all coefficients are real [36]. The equal-time fluctuations of the amplitude (18), calculated with this growth rate, show an interesting scaling behavior; see Sec. IV B.

## E. Fluctuations at and above threshold

The essential new effects which enter into the description of fluctuations above threshold are phase fluctuations and deterministic nonlinear selection. The first effect is connected with the spontaneously broken translation symmetry above threshold in systems with large aspect ratio. The nonlinear selection determines the stable pattern configurations and thus the linearization of the stochastic envelope equations around this configuration. The nonlinear envelope equations are obtained from (14), (35), or (32) by simply adding the nonlinear deterministic terms. This is justified for the following reasons.

(i) As will be shown explicitly for EHC in Sec. III the nonlinear (multiplicative) part of the correlation matrix  $\underline{O}(\nabla, u)$  is small within the range of applicability of the envelope equation. The stochastic term therefore remains the same.

(ii) Nonlinear envelope equation are usually obtained by systematic expansion [37, 32]. This method can be extended to include the stochastic terms as has been done by Graham [12] in the case of RBC. It can be shown that the linear deterministic and stochastic terms of both the projection procedure described in this paper and the systematic expansion method are identical.

Note that the stochastic Newell-Whitehead equation [37] for isotropic systems, derived by Graham, can only be applied locally and only above threshold, because it is not rotationally invariant. In the frame of our formulation, its fluctuating force follows from the stochastic Swift-Hohenberg equation by applying Eqs. (12)-(16) where u stands for the variable  $\psi$  of the Swift-Hohenberg equation and A for the amplitude of the Newell-Whitehead equation.

#### STOCHASTIC ENVELOPE EQUATIONS FOR NONEQUILIBRIUM ...

## III. FLUCTUATIONS IN ELECTROHYDRODYNAMIC CONVECTION

#### **A. Basic equations**

The starting point are dynamical equations for the independent macroscopic variables of EHC in nematic liquid crystals, the potential  $\phi$  of the electrical field distortion (see below), the director N with  $N^2 = 1$ , and the fluid velocity v. One has to combine hydrodynamic equations for the velocity field with a quasistatic balance equation for the angular momentum of the director and quasistatic Maxwell equations (for the deterministic part see, e.g., [29])

$$d_t \rho^{(\mathrm{el})} + \boldsymbol{\nabla} \cdot \boldsymbol{j}^{(\mathrm{cond})} = \boldsymbol{\xi}^{(\rho)}, \qquad (38)$$

$$\epsilon_{ijk}n_j\left(\frac{\delta F}{\delta n_k} + S'_k\right) = \xi_i^{(n)},\tag{39}$$

$$\epsilon_{ijk}\partial_j \left[ \rho^{(m)} d_t v_k + \partial_l \left( \frac{\partial F}{\partial n_{m,l}} n_{m,k} - t_{lk} \right) - f_k^{(\text{el})} \right] = \xi_i^{(v)},$$
(40)

$$\boldsymbol{\nabla} \cdot \boldsymbol{v} = 0. \tag{41}$$

Here  $d_t = \partial_t + \boldsymbol{v} \cdot \boldsymbol{\nabla}$  denotes the material derivative,  $\rho^{(\text{el})}$  the electric charge density,  $\boldsymbol{j}^{(\text{cond})}$  the current due to conductivity,  $F = F^{(el)} + F^{(\text{mech})}$  the director free energy density, and  $\frac{\delta F}{\delta n_i} = \frac{\partial F}{\partial n_i} - \partial_j \frac{\partial F}{\partial n_{i,j}}$  the electric and elastics forces on the director  $(n_{i,j} = \partial_j n_i)$ . S' denotes the viscous coupling with fluid motion,  $\rho^{(m)}$  the mass density,  $\boldsymbol{t}$  the dissipative part of the stress tensor, and  $\boldsymbol{f}^{(\text{el})}$  the electric volume force acting on the fluid.  $\epsilon_{ijk}$  is the totally antisymmetric unit tensor. The specific material properties are contained in constitutive relations for  $\rho^{(\text{el})}, F, \boldsymbol{f}^{(\text{el})}, \boldsymbol{j}^{(\text{cond})}, S'$  and  $\boldsymbol{t}$  (for simplicity we neglect flexoelectric polarizations),

$$\rho^{(el)} = \epsilon_0 \nabla \cdot \underline{\epsilon} E, \quad \epsilon_{ij} = \epsilon_\perp \delta_{ij} + \epsilon_a n_i n_j, \qquad (42)$$

$$F^{(\text{mech})} = \frac{1}{2} \{ K_{11} (\boldsymbol{\nabla} \cdot \boldsymbol{n})^2 + K_{22} [\boldsymbol{N} \cdot (\boldsymbol{\nabla} \times \boldsymbol{n})]^2 + K_{33} [\boldsymbol{N} \times (\boldsymbol{\nabla} \times \boldsymbol{n})]^2 \},$$
(43)

$$F^{(\mathrm{el})} = -\frac{1}{2}\epsilon_0\epsilon_a (\boldsymbol{N}\cdot\boldsymbol{E})^2, \qquad (44)$$

$$\boldsymbol{f}^{(\mathrm{el})} = \boldsymbol{\rho}^{(\mathrm{el})} \boldsymbol{E} + \mathrm{h.o.t.}, \qquad (45)$$

$$\boldsymbol{j}^{(\mathrm{cond})} = \underline{\boldsymbol{\sigma}} \boldsymbol{E}, \ \ \sigma_{ij} = \sigma_{\perp} \delta_{ij} + \sigma_a n_i n_j,$$
 (46)

$$\mathbf{S}' = \gamma_1 \mathbf{N} + \gamma_2 \underline{\mathbf{d}} \mathbf{N}, \tag{47}$$

$$t_{ij} = \alpha_1 n_i n_j n_k n_l d_{kl} + \alpha_2 n_i N_j + \alpha_3 n_j N_i + \alpha_4 d_{ij} + \alpha_5 n_i n_k d_{kj} + \alpha_6 n_j n_k d_{ki}.$$
(48)

The material parameters  $\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp}$  and  $\sigma_a = \sigma_{\parallel} - \sigma_{\perp}$ are the dielectric and conductive anisotropies of a uniaxial medium;  $K_{11}, K_{22}$ , and  $K_{33}$  pertain to splay, twist and bend deformations of the director, respectively;  $\alpha_1, ..., \alpha_6$  with  $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$  are viscosity coefficients,  $\alpha_4/2$  corresponding to the usual isotropic viscosity, and  $\gamma_1 = \alpha_3 - \alpha_2$  and  $\gamma_2 = \alpha_3 + \alpha_2$  are rotational viscosities.  $N = d_t N + \frac{1}{2} N \times (\nabla \times v)$  is the angular velocity of the director relative to the moving fluid and  $d_{ij} = \frac{1}{2}(\partial_i v_j + \partial_j v_i)$  the (symmetric) strain-rate tensor. The higher-order terms of  $f^{(el)}$  are discussed, e.g., in [39]. The constitutive relations (42)-(45) relate to purely conservative effects while (46)-(48) contain, in addition to some conservative terms, all dissipative processes. The latter are relevant for determining the fluctuating forces of the EHC equations.

The quasistatic Maxwell equations  $\nabla \times E = 0$  and the condition  $N^2 = 1$  are automatically satisfied by the representation

$$N = (1 - \sqrt{n_y^2 + n_z^2}, n_y, n_z), \tag{49}$$

$$\boldsymbol{E}(t) = \frac{\sqrt{2}V_0}{d} \cos \omega_{0,abs} t - \boldsymbol{\nabla}\phi, \qquad (50)$$

with the applied ac voltage  $\sqrt{2}V_0 \cos \omega_{0,abs}t$ . Only two components of the director equation (39) and of the curl of the momentum equation (40) are independent. Selecting the y and the z component of either of these equations yields, together with charge conservation and the incompressibility condition, six equations for the six fields  $\boldsymbol{u} := (\phi, n_y, n_z, \boldsymbol{v})$ . Apart from the external control parameters  $V_0$  and  $\omega_{0,abs}$ , the equations depend only on material parameters. Taking into consideration the planar boundary conditions  $\boldsymbol{N}|_{boundary} = (1,0,0)$ , the unstructured state is given by  $\boldsymbol{u} = 0$  and the deterministic part of the EHC equations takes the symbolic form (1) with  $2\pi/\omega_{0,abs}$ -periodic operators  $\underline{S}, \underline{L}$ , and N.

To make the relative magnitudes of the fluctuating forces and the relevant internal time scales explicit we write the resulting equations in terms of dimensionless quantities given in the table below:

Length x = dx'Time  $t = T_d t', T_d = \frac{\gamma_1 d^2}{K_{11} \pi^2}$ Electric potential  $\phi = \sqrt{\frac{K_{11}}{\epsilon_0 \epsilon_\perp}} \phi'$ Orientational elastic constants  $K_{ii} = K_{11} K'_{ii}, i = 2, 3$ Viscosities  $\alpha_i = \gamma_1 \alpha'_i, \gamma_j = \gamma_1 \gamma'_j, i = 1, ...6, j = 1, 2$ Electrical constants  $\sigma_{ij} = \sigma_\perp \sigma'_{ij}, \epsilon_{ij} = \epsilon_\perp \epsilon'_{ij}$ (51) After dropping the primes, the dimensionless EHC equations in standard form read

.(a)

$$P_{1}d_{t}\rho^{(\mathrm{el})} + \boldsymbol{\nabla} \cdot \boldsymbol{j}^{(\mathrm{cond})} = \xi^{(\rho)},$$

$$\left[\epsilon_{ijk}n_{j}\left(\frac{\delta F}{\delta n_{k}} + S_{k}^{\prime}\right)\right]_{i=y,z} = \xi_{y,z}^{(n)},$$

$$\left\{\epsilon_{ijk}\partial_{j}\left[P_{2}d_{t}v_{k} + \partial_{l}\left(\frac{\partial F}{\partial n_{m,l}}n_{m,k} - t_{lk}\right)\right.$$

$$\left.-f_{k}^{(\mathrm{el})}\right]\right\}_{i=y,z} = \xi_{y,z}^{(v)},$$

$$\left.\boldsymbol{\nabla} \cdot \boldsymbol{v} = 0,$$
(52)

where  $E_i := \sqrt{2R_0} \cos \omega_0 t \delta_{i3} - \partial_i \phi$  contains the two dimensionless control parameters

$$R_0 = \frac{\epsilon_0 \epsilon_\perp}{K_{11}} V_0^2, \quad \omega_0 = \pi^2 T_d \omega_{0,\text{abs}}, \tag{53}$$

and  $P_1$  and  $P_2$  are two ratios of time scales

$$P_1 = T_q / (\pi^2 T_d), \quad P_2 = T_{visc} / T_d,$$
 (54)

with the director relaxation time  $T_d$  defined in (51), the charge relaxation time  $T_q = (\epsilon_0 \epsilon_\perp)/\sigma_\perp$ , and the viscous diffusion time  $T_{\rm visc} = (\rho^{(m)} d^2)/(\gamma_1 \pi^2)$ . The new expressions for the dimensionless quantities  $\underline{\epsilon}, \underline{\sigma}, F, S'$ , and  $\underline{t}$ are again given by Eqs. (42)-(48) with the material parameters replaced by the dimensionless ones and  $\epsilon_0 \rightarrow 1$ . Note that all these expressions are of order unity.

#### **B.** Determination of the stochastic forces

The stochastic properties of the fluctuating forces of Eqs. (52) are given by the fluctuation-dissipation theorem [21]. We formulate it here for extended linear systems with the relevant dynamical variables  $u_{\alpha}^{(\text{diss})}(\mathbf{r},t)$ , where the equations of motion are written in the generalized Onsager form (GOF) [40]

$$\dot{u}_{\alpha}^{(\text{diss})}\left(\boldsymbol{r},t\right) = \tilde{M}_{\alpha\beta}(\boldsymbol{\nabla})\frac{\delta S}{\delta u_{\beta}^{(\text{diss})}} + \eta_{\alpha}(\boldsymbol{r},t), \qquad (55)$$

with the coarse grained entropy functional  $S[u^{(diss)}]$ . The FDT then states that for local thermal equilibrium the fluctuating forces are given by

$$\langle \eta_{\alpha}(\boldsymbol{r},t) \rangle = 0,$$
(56)

$$\langle \eta_{\alpha}(\boldsymbol{r},t)\eta_{\beta}(\boldsymbol{r}',t')\rangle = (\underline{\tilde{M}} + \underline{\tilde{M}}^{\dagger})_{\alpha\beta}\delta(\boldsymbol{r}-\boldsymbol{r}')\delta(t-t'),$$

with  $[\underline{\tilde{M}}^{\mathsf{T}}(\nabla)]_{\alpha\beta} = \tilde{M}_{\beta\alpha}(-\nabla)$ . The variables  $\boldsymbol{u}^{(\text{diss})}$ need not be identical to the variables  $\boldsymbol{u}$  of the original system as long as they express all dissipative effects.

To get a linear GOF in terms of suitable variables for EHC or any other extended system we follow the method of Landau and Lifshitz [22], hereafter referred to as the Landau approach. A brief comparison with the "global approach" [20] employing a nonlinear formulation of the FDT [41] is given in Appendix A.

In the Landau approach one compares the total entropy production, expressed in terms of independent thermodynamical forces  $F_{\alpha}$  and fluxes  $J_{\alpha}$ ,

$$\dot{S} = \int d^3 \boldsymbol{r} F_{\alpha}(\boldsymbol{r}) J_{\alpha}(\boldsymbol{r}), \qquad (57)$$

with the entropy production obtained from the GOF (55),

$$\dot{S} = \int d^3 \boldsymbol{r} \frac{\delta S}{\delta u_{\alpha}^{(\text{diss})}} \ \dot{u}_{\alpha}^{(\text{diss})} = \int d^3 \boldsymbol{r} \frac{\delta S}{\delta u_{\alpha}^{(\text{diss})}} \underline{\tilde{M}}_{\alpha\beta} \frac{\delta S}{\delta u_{\beta}^{(\text{diss})}}.$$
(58)

One may identify  $u_{\alpha}^{(\text{diss})}$  with  $J_{\alpha}$  and  $\frac{\delta S}{\delta u_{\alpha}^{(\text{diss})}}$  with  $F_{\alpha}$ . One then sees that the deterministic linear relations between the fluxes and the forces,

$$J_{\alpha} = M_{\alpha\beta}F_{\beta},\tag{59}$$

represent a GOF of the form (55) where the coefficient matrix M is identical to the Onsager matrix M, and the FDT requires that these relations be supplemented by stochastic terms

$$J_{\alpha} = M_{\alpha\beta}F_{\beta} + \tilde{J}_{\alpha}, \tag{60}$$

$$\langle \tilde{J}_{\alpha}(\boldsymbol{r},t)\tilde{J}_{\beta}(\boldsymbol{r}',t')\rangle$$

$$= [\underline{\boldsymbol{M}}(\boldsymbol{u}) + \underline{\boldsymbol{M}}^{T}(\boldsymbol{u})]_{\alpha\beta}\delta(\boldsymbol{r}-\boldsymbol{r}')\delta(t-t'), \quad (61)$$

where  $\underline{M}$  contains no derivatives.

In EHC, the dimensionless entropy production can be written as [42, 44]

$$\frac{k_B T}{K_{11} d} \dot{S} = \int d^3 \boldsymbol{r} \left\{ t_{ij}^{(s)} d_{ij} - t_{ij}^{(as)} \Omega_{ij} + \frac{1}{P_1} \boldsymbol{j}^{(\text{cond})} \cdot \boldsymbol{E} -\frac{1}{T} \boldsymbol{j}^{(\text{therm})} \cdot \boldsymbol{\nabla} T \right\},$$
(62)

with the (anti)symmetric part of the stress tensor  $t_{ij}^{(as)} =$  $(t_{ij} - t_{ji})/2$ ,  $t_{ij}^{(s)} = (t_{ij} + t_{ji})/2$ , and the antisymmetric tensor  $\underline{\Omega}$  defined by  $N_i = \Omega_{ij}n_j$ . The dimensionless quantities of Sec. III A were used and the entropy is measured in units of  $k_B$ . Neglecting the temperature gradient as usual [28] one can identify three independent fluxes

$$\underline{J}^{(1)} = \underline{t}^{(s)}, \quad \underline{J}^{(2)} = \underline{t}^{(as)}, \quad J^{(3)} = \underline{j}^{(cond)}, \quad (63)$$

and the corresponding forces

$$\underline{F}^{(1)} = \frac{K_{11}d}{k_B T} \underline{d},$$

$$\underline{F}^{(2)} = -\frac{K_{11}d}{k_B T} \underline{\Omega},$$

$$F^{(3)} = \frac{K_{11}d}{P_1 k_B T} \underline{E}.$$
(64)

The deterministic linear relations between the fluxes and forces,  $J_{ij}^{(m)} = M_{ij,kl}^{(mn)} F_{kl}^{(n)} + M_{ij,k}^{(m3)} F_{k}^{(3)}$  and  $J_{i}^{(3)} = M_{i,kl}^{(3n)} F_{kl}^{(n)} + M_{i,k}^{(33)} F_{k}^{(3)}$ , where i, j, k, l = 1, ..., 3 and m, n = 1, 2, are obtained by comparing the dissipative constitutive relations (46)–(48) with the definition (60) of the Onsager matrix and (63) and (64). The explicit expressions (B1) are given in Appendix B. Note the Onsager relations  $M_{ij,kl}^{(nm)} = M_{kl,ij}^{(mn)}$ , which lead to  $\alpha_2 + \alpha_3 = \alpha_6 + \alpha_5$ . With the FDT (61) the fluctuations of the fluxes are then known.

In order to get the stochastic forces of the basic equations (52), all dissipative terms of them have to be expressed in terms of these fluxes. For the charge conservation and the momentum balance equations the expressions are evident. For the director equations one uses conservation of angular momentum, which is separately fulfilled for the conservative and dissipative parts [42]. With the relation for the dissipative part,  $n_i S'_j - n_j S'_i = 2t^{(as)}_{ij}$ , the stochastic forces are

$$\xi^{(\rho)} = -\partial_i \tilde{J}_i^{(3)},$$
  

$$\xi_i^{(n)} = -2\epsilon_{ijk} \tilde{J}_{jk}^{(2)},$$
  

$$\xi_i^{(v)} = \epsilon_{ijk} \partial_j \partial_l (\tilde{\underline{J}}^{(1)} + \tilde{\underline{J}}^{(2)})_{lk}.$$
(65)

The vector of the stochastic forces  $\boldsymbol{\xi} := (\boldsymbol{\xi}^{(p)}, \boldsymbol{\xi}^{(n)}_{\boldsymbol{y}}, \boldsymbol{\xi}^{(n)}_{\boldsymbol{x}}, \boldsymbol{\xi}^{(n)}_$ 

$$\underline{O}(\nabla \boldsymbol{u}] = \underline{D}(\nabla)[\underline{M}(\boldsymbol{u}) + \underline{M}^{T}(\boldsymbol{u})]\underline{D}^{T}(-\nabla) \qquad (66)$$

and for EHC to the following nonvanishing elements of the correlation matrix:

$$O_{11} = -2\partial_i M_{ij}^{(33)} \partial_j,$$

$$O_{22} = 8M_{31,31}^{(22)},$$

$$O_{33} = 8M_{12,12}^{(22)},$$

$$O_{23} = -8M_{31,12}^{(22)},$$

$$O_{2\alpha} = -4D_{ij}^{(\alpha)} (\underline{M}^{(21)} + \underline{M}^{(22)})_{31,ij}, \quad \alpha = 4, 5$$

$$O_{\alpha\beta} = 2D_{ij}^{(\alpha)} (\underline{M}^{(11)} + \underline{M}^{(12)} + \underline{M}^{(21)} + \underline{M}^{(22)})_{ij,kl} D_{kl}^{(\beta)}, \quad \alpha, \beta = 4, 5$$

$$O_{3\alpha} = O_{2\alpha}(31 \rightarrow 12), \quad \alpha = 4, 5,$$

$$O_{\beta\alpha} = O^{\dagger}_{\alpha\beta} , \quad \alpha, \beta = 1, ..., 6, \tag{67}$$

with the matrix differential operators

 $\underline{D}^{(4)} := (\partial_x, \partial_y, \partial_z)(-\partial_y, \partial_x, 0)^T$ and

$$\underline{\boldsymbol{D}}^{(5)} := (\partial_{\boldsymbol{x}}, \partial_{\boldsymbol{y}}, \partial_{\boldsymbol{z}})(\partial_{\boldsymbol{z}}, 0, -\partial_{\boldsymbol{x}})^T.$$

The fluctuating forces introduce via the Onsager coefficients  $\underline{M}$  a further dimensionless quantity

$$Q_0 := \frac{k_B T}{K_{11} d}.$$
 (68)

The length  $k_B T/K_{11} \approx 10^{-9}$  m for N-(p-methoxybenzylidene)-p'-butylaniline (MBBA)] can be interpreted as the range of the molecular correlations. Thus  $Q_0$  is the ratio of this length and the cell thickness d.

Note that by means of the N dependence of the Onsager coefficients the fluctuating forces have nonlinear multiplicative parts if interpreted in terms of N rather than in terms of the fluxes. We show now that for our purposes the multiplicative parts of the fluctuating forces can be dropped. To this end we expand  $\underline{O}$ with respect to N around  $N_0$ ,  $O_{ij}(N) = O_{ij}(N_0) + \frac{\delta O_{ij}}{\delta n_k}(n_k - n_{k0}) + \mathcal{O}(N - N_0)^2$ . Since the variational derivative of the scaled correlation matrix (67) is of the same order of magnitude as  $O_{ij}$  itself, the approximation  $O_{ij}(N) \approx O_{ij}(N_0)$  holds as long as  $|N - N_0| \ll 1$ , a condition which has to be fulfilled anyway when using the envelope equations. Therefore we truncate all multiplicative parts of  $\underline{O}$  in the following calculations setting  $\underline{O}(\nabla, \mathbf{u}) \approx \underline{O}(\nabla, \mathbf{u} = 0)$  as in Sec. II B.

#### C. Langevin equations for Fourier modes and the stochastic envelope equation in the normal-roll regime

The stochastic envelope equation for the nonautonomous system (52) will be calculated according to Sec. IIC. With the external field (50), the linear deterministic parts  $\underline{S}(t)\partial_t$  and  $\underline{L}(t)$  of (52) contain terms  $\propto 1, \cos \omega_0 t, \cos^2 \omega_0 t$  and therefore setting  $\cos \omega_0 t =$  $1/2(e^{i\omega_0 t} + e^{-i\omega_0 t})$ , the Fourier coefficients of the expansion  $\underline{S}^{(n)}, \underline{L}^{(n)}$  [Eq. (21) vanishes for  $|n| \geq 2$ . Besides, only odd time Fourier expansion coefficients of  $\phi$  couple with even coefficients of the other fields (conductive mode) and vice versa (dielectric mode). In the following we restrict ourselves to the conductive regime  $\omega_0 < \omega_{\rm cutoff}$  with the cutoff frequency  $\omega_{\rm cutoff} P_1$  =  $\omega_{\rm cutoff,abs}T_q \approx 2$  [29] (the suffix abs denotes unscaled quantities). We also use the lowest-order time Fourier expansion, applicable for  $\omega_0 \gg 1$ , i.e.,  $\omega_{0,abs} \gg 1/T_d$ ; see the discussion in [29]. The effective autonomous Langevin equations (26) and (27) then become a  $7 \times 7$ system for the fields

$$\tilde{\boldsymbol{u}} = (\phi^{(1)}, \phi^{(-1)}, n_{\boldsymbol{y}}^{(0)}, n_{\boldsymbol{z}}^{(0)}, \boldsymbol{v}^{(0)}), \tag{69}$$

where the linear operators  $\underline{A}, \underline{T}$  and the fluctuating forces  $\tilde{\boldsymbol{\xi}} = (\xi_1^{(1)}, \xi_1^{(-1)}, ..., \xi_6^{(0)})$  are determined by Eqs. (23)–(25), using Eqs. (52), (50), and (67). Note that according to (25) the cross correlations  $\langle \tilde{\xi}_1 \tilde{\xi}_\alpha \rangle = \langle \xi_1^{(1)} \xi_\alpha^{(0)} \rangle, \ \alpha \neq 1$ , vanish due to different time symmetries, even if the original correlation matrix  $O_{\alpha\beta}$  had nonvanishing components

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 $O_{1\alpha}$ . Now we specialize the above expressions to Fourier modes of normal rolls by substituting  $\partial_x o iq, \; \partial_y o$ 0. For  $q \neq 0$ ,  $n_y^{(0)}$  and  $v_y^{(0)}$  vanish and we are left with a  $5 \times 5$  system for the remaining variables. After elimination of  $v_x$  by means of the continuity equation  $v_x = -\partial_z v_z/(iq)$  [43] we finally obtain a 4 × 4 system for  $\tilde{\boldsymbol{u}}_q := (\phi_q, \overline{\phi}_q, n_{zq}, v_{zq})$ , the Fourier components of

$$\begin{split} \tilde{\underline{S}}_{q} &= \begin{pmatrix} P_{1}E & 0 & iqP_{1}\sqrt{2R_{0}}\epsilon_{a} & 0 \\ 0 & P_{1}E & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & q^{2} & P_{2}S_{vv} \end{pmatrix}, \\ \tilde{\underline{L}}_{q} &= \begin{pmatrix} S & iP_{1}E\omega_{0} & iq\sqrt{2R_{0}}\sigma_{a} & 0 \\ iP_{1}E\omega_{0} & S & -P_{1}q\sqrt{2R_{0}}\epsilon_{a}\omega_{0} & 0 \\ \frac{iq}{\sqrt{2}}\sqrt{R_{0}}\epsilon_{a} & 0 & K-\epsilon_{a}R_{0} & -iq \\ \frac{iq}{\sqrt{2}}\sqrt{R_{0}}E & 0 & -\epsilon_{a}q^{2}R_{0} & \frac{O_{vv}}{iq} \\ \tilde{\underline{O}}_{q} &= 2Q_{0} \begin{pmatrix} 2P_{1}S & 0 & 0 & 0 \\ 0 & 2P_{1}S & 0 & 0 \\ 0 & 0 & 1 & q^{2} \\ 0 & 0 & q^{2} & O_{vv} \end{pmatrix}, \end{split}$$

where

$$E = \epsilon_{\parallel} q^{2} - \partial_{z}^{2},$$

$$S = \sigma_{\parallel} q^{2} - \partial_{z}^{2},$$

$$K = K_{33}q^{2} - \partial_{z}^{2},$$

$$S_{vv} = (q^{2} - \partial_{z}^{2})/(iq),$$

$$O_{vv} = \frac{1}{2}(2 + \alpha_{4} + \alpha_{6})q^{4} - (1 + \alpha_{1} + \alpha_{4} + \alpha_{6})q^{2}\partial_{z}^{2}$$

$$+ \frac{1}{2}(\alpha_{4} + \alpha_{6})\partial_{z}^{4}.$$
(74)

To simplify the expressions we have set the quantity  $\alpha_3$ , which is for ordinary nematics very small, equal to zero. The lowest branch of the dispersion relation  $\sigma(q, R_0) := \sigma(\boldsymbol{k} = (q, 0), R_0), ext{ the respective modal eigen-}$ functions  $\tilde{f}_{q}(z), \tilde{f}_{q}^{\dagger}(z)$ , and the critical point  $(k_{c}, R_{c})$  of the deterministic eigenvalue problem

$$[\underline{\tilde{\boldsymbol{S}}}_{\boldsymbol{q}}\sigma(\boldsymbol{q},\boldsymbol{R}_{0}) + \underline{\tilde{\boldsymbol{L}}}_{\boldsymbol{q}}]\tilde{\boldsymbol{f}}_{\boldsymbol{q}}(z) = 0$$
(75)

are obtained using a Galerkin method described elsewhere [29]. Now all quantities for calculating the fluctuating forces  $Q_k$  (9) of the Fourier modes and the fluctuating force (16) of the amplitude equation are determined for normal rolls in the conductive regime. With Eqs. (6) and (10) the Fourier components of the measurable fluctuations of the z component of the director in the middle of the slab are given by

$$\langle |\overline{\theta}_q|^2 \rangle(\epsilon, P_1, P_2) = -\frac{\tilde{f}_{q,n_\star}^2(0)}{2(2\pi)^2} \frac{Q_q(\epsilon, P_1, P_2)}{\sigma(q, \epsilon, P_1, P_2)}, \qquad (76)$$

where  $\theta_q(t)$  is the Fourier transform of  $\theta(\boldsymbol{x},t)$  :=  $n_{z}(\boldsymbol{x}, z = 0, t)$  at the wave number  $\boldsymbol{k} = (q, 0)$  and  $\epsilon = (R_0 - R_c)/R_c$ . The amplitude equation (14) together with (19) and the relation to the physical quantities (12)

$$\begin{split} \phi^{(1)} + \phi^{(-1)}, \phi^{(1)} - \phi^{(-1)}, n_z^{(0)}, \text{ and } v_z^{(0)} \text{ at } \boldsymbol{k} &= (q, 0), \\ (\underline{\tilde{\boldsymbol{S}}}_q \partial_t + \underline{\tilde{\boldsymbol{L}}}_q) \tilde{\boldsymbol{u}}_q(z, t) &= \tilde{\boldsymbol{\xi}}_q(z, t), \quad \langle \tilde{\boldsymbol{\xi}}_q^*(z, t) \tilde{\boldsymbol{\xi}}_q(z', t') \rangle \\ &= \underline{\tilde{\boldsymbol{O}}}_q \delta(z - z') \delta(t - t') \end{split}$$
(70)

( **a** )

with the matrix-differential operators

$$\begin{pmatrix} -iq\\ -iq\\ \frac{i}{2} \end{pmatrix}$$
, (72)

(73)

reads in the usual notation (multiplication by  $T_0$ ) including the lowest-order nonlinearity

$$\boldsymbol{u} = (\phi, \overline{\phi}, n_z, v_z) = A(x, y, t) \tilde{f}_{q_c}(z) e^{iq_c x} + \text{c.c.},$$
(77)

$$T_0 \partial_t A(x, y, t) = (\epsilon + \xi_x^2 \partial_x^2 + \xi_y^2 \partial_y^2) A$$
$$-g |A|^2 A + F_A(x, y, t), \tag{78}$$

with the linear coefficients  $T_0 = 1/\sigma_{\epsilon}$ ,  $\xi_x^2 = -\sigma_{k_x k_x}/(2\sigma_{\epsilon})$ ,  $\xi_y^2 = -\sigma_{k_y k_y}/(2\sigma_{\epsilon})$ , and  $\sigma$  given by Eq. (75). With (9) and (16) the fluctuating force is given by

$$= T_0^2 Q_{q_c}(\epsilon = 0) \delta(x - x') \delta(y - y') \delta(t - t'), \quad (79)$$

$$Q_{q} = \frac{1}{|N_{q}|^{2}} \int_{-1/2}^{1/2} dz \tilde{\boldsymbol{f}}_{q}^{\dagger *}(z) \underline{\tilde{\boldsymbol{O}}}_{q}(\partial_{z}) \tilde{\boldsymbol{f}}_{q}^{\dagger}(z), \qquad (80)$$

$$N_{q} = \int_{-1/2}^{1/2} dz \tilde{\boldsymbol{f}}_{q}^{\dagger *}(z) \underline{\tilde{\boldsymbol{S}}}_{q}(\partial_{z}) \tilde{\boldsymbol{f}}_{q}(z).$$
(81)

With Eq. (33) the correlations of the director fluctuations  $n_z$  in the middle of the slab are

$$egin{aligned} & heta(oldsymbol{x},t) heta(oldsymbol{x}',t') 
angle \end{aligned}$$

 $\langle F_A^*(x,y,t)F_A(x',y',t')\rangle$ 

$$=2\tilde{f}_{q,n_z}^2(0)\langle A^*(\boldsymbol{x},t)A(\boldsymbol{x}',t')\rangle\cos q_c\Delta x.$$
 (82)

Note that, for equal times and near threshold, the spatial Fourier component  $k_c$  of the correlations (82) and the expression (76) both converge to  $\langle |\theta_{q_c}|^2 \rangle =$  $\tilde{f}_{q,n_z}^2(0)Q_{q_c}T_0/(8\pi^2\epsilon)$ . Equation (76) together with the growth rate from (70)-(75) explicitly give, for normal rolls and all values of q and  $\epsilon < 0$ , the fluctuations of the mode becoming critical at threshold in terms of the linear deterministic quantities. Equations (77)–(81) give the fluctuating strength of the stochastic amplitude equation in the conductive normal-roll regime. Together with the connection to the measurable director fluctuations, Eqs. (76) and (82), these equations are the main result of this section.

## IV. RESULTS AND COMPARISON WITH EXPERIMENT

#### A. Intensity of director fluctuations in the normal-roll regime

We calculated the director fluctuations for the parameter set I of MBBA (see [29]) in lowest-order Galerkin approximation

$$\tilde{\boldsymbol{f}}_{q} = \left[ c_{\phi} g_{\phi}(z), c_{\overline{\phi}} g_{\overline{\phi}}(z), g_{\boldsymbol{n}_{z}}(z), c_{v} g_{\boldsymbol{v}_{z}}(z) \right], \\ \tilde{\boldsymbol{f}}_{q}^{\dagger} = \left[ c_{\phi}^{\dagger} g_{\phi}(z), c_{\overline{\phi}}^{\dagger} g_{\overline{\phi}}(z), g_{\boldsymbol{n}_{z}}(z), c_{v}^{\dagger} g_{\boldsymbol{v}_{z}}(z) \right].$$
(83)

We assumed realistic rigid planar boundary conditions and the same trial functions for  $\phi, \overline{\phi}, n_z$  and  $v_z$  as in Ref. [29],  $g_{\phi}(z) = g_{\overline{\phi}}(z) = g_{n_z}(z) = \sqrt{2} \cos \pi z$  and  $g_{v_z} = C_1(z)$ , where  $C_n(z)$  denotes the *n*th normalized Chandrasekhar function, see, e.g., [29]. Thus the  $4 \times 4$ matrices  $\underline{\tilde{S}}_q$ ,  $\underline{\tilde{L}}_q$ , and  $\underline{\tilde{O}}_q$  of Eq. (70) become algebraic with the components  $A_{\alpha\beta}^{(\text{Gal})} = \int dz g_\alpha(z) A_{\alpha\beta}(\partial_z) g_\beta(z)$ , where  $\underline{A} = \underline{\tilde{S}}_q$ ,  $\underline{\tilde{L}}_q$ , or  $\underline{\tilde{O}}_q$  and  $\alpha, \beta = \phi, \overline{\phi}, n_z$ , or  $v_z$ , and no sum convention is applied.

The eigenvalue equation (75) and its Hermitian conjugate become an algebraic problem for the eigenvectors  $\mathbf{c} = (c_{\phi}, c_{\overline{\phi}}, 1, c_v)$  and  $\mathbf{c}^{\dagger}$ . The normalization with respect to the  $n_z$  component ensures that with  $\tilde{f}_{q,n_z}^2(0) = 2$  the amplitude is directly connected to the director fluctuations (82) and (76). Figure 1 shows the growth rate  $\sigma$ 

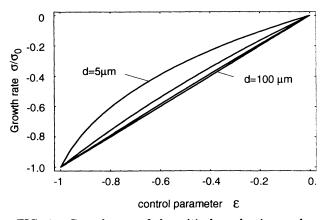


FIG. 1. Growth rate of the critical conductive mode of EHC as function of  $\epsilon$  at applied frequencies  $\omega_{0,abs}$  with  $T_d \ll 2\pi/\omega_{0,abs} < T_q$ . The parameter is the cell thickness d = 5, 10, 20, and 100  $\mu$ m. The growth rate is normalized relative to  $\sigma_0 = \sigma(\epsilon = -1)$  and the parameter set I of MBBA [29] is used. The critical value of the control parameter in dimensionless units is  $R_c = 307.3$ .

at the critical wave number for the conductive mode as a function of  $\epsilon = (R_0 - R_c)/R_c$  with  $R_0 = V_0^2 \epsilon_0/K_{11}$ for dimensionless external frequencies  $2\pi \ll \omega_0 \ll 1/P_1$ corresponding to  $\omega_{0,abs}T_d \gg 1$  and  $\omega_{0,abs}T_q \ll 1$ . The parameter is the cell thickness. Since usually  $P_1$  is small and  $P_2$  is negligible, an expansion of the growth rate at the critical wave vector in terms of  $P_1, P_2$  and  $\epsilon$  is useful

 $\sigma(q_c,\epsilon,P_1,P_2)$ 

$$= -\sigma_0 \epsilon \left[ 1 - 92.4(1+\epsilon)P_1 + 8540(1+\epsilon)P_1^2 \right] -0.86(1+\epsilon)P_2 + O(P_1^3, P_2^2, \epsilon^3),$$
(84)

where  $R_c = 307.3$ ,  $k_c = (1.52\pi, 0)$ ,  $P_1 = 1.50(\mu m/d)^2$ ,  $P_2 = 5.6 \times 10^{-7}$ , and the zero-field growth rate  $\sigma_0 = -72.7$ . Analytical expressions for  $R_c$  and  $\sigma_0$ are given in Appendix C, Eqs. (C1) and (C3). The expansion shows that in the limit  $P_1 \rightarrow 0$ , i.e.,  $d \rightarrow \infty$ , and  $P_2 = 0$  the quantity  $\sigma/\epsilon = -\sigma_0$  does not depend on  $\epsilon$  and that only the correction terms with  $P_1$  on the right-hand side of Eq. (84) can become important. For  $\epsilon = -1$  (V<sub>0</sub> = 0) all corrections must vanish. For typical thicknesses used in experiments, e.g.,  $d = 23 \ \mu m$  in Ref. [8] or  $d = 13 \ \mu m$  in [6], the correction is only a few percent and could not be measured. Note that  $R_c$  and  $k_c$ do not depend on  $P_1$  or  $P_2$  for  $\omega_0 P_1 \ll 1$ . Figure 2 shows the dependence of the noise strength  $Q_{q_c}$ , Eq. (80), on  $\epsilon$  for the critical mode. For  $\epsilon = 0$  one obtains the noise strength  $T_0^2 Q$  of the corresponding stochastic envelope equation (78) with

$$Q = Q_{q_c}(\epsilon = 0) = 2Q_0 B\left(\frac{1 + 147P_1}{(1 + 92.4P_1 + 3.6P_2)^2}\right),$$
(85)

where  $Q_0 = k_B T/(K_{11}d)$  is the small parameter,  $B \approx 1.84$  includes the deterministic effects of the velocity field and the expression in large parentheses contains the correction due to the fluctuating forces of the electric field and of the velocities. For analytical expressions and the

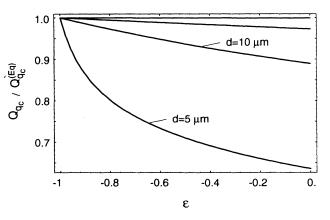


FIG. 2. The fluctuation strength  $Q_{qc}$  of the noise term in the Langevin equation for the critical Fourier mode as function of  $\epsilon$ , for the same cell thicknesses as in Fig. 1. The fluctuation strength is plotted relative to the constant value for the equipartition limit  $P_1 = P_2 \rightarrow 0$ . For  $\epsilon = 0$ , the stochastic force of the amplitude equation is obtained.

definition of B, see Appendix C. Although Q decreases slightly with increasing time-scale ratios  $P_1$  and  $P_2$ , the noise strength  $T_0^2 Q$  of the amplitude equation and the modal director fluctuations  $\langle |\bar{\theta}_q|^2 \rangle = -1/(2\pi)^2 Q_q/\sigma$  increase, because  $1/T_0^2$  and  $\sigma$  decrease more rapidly than  $Q_q$ . The fluctuations can be compared with the estimate of Refs. [6-8], based on two assumptions: (i) the stochastic forces are insensitive to the applied electric fields, so their value at  $R_0 \to 0$ , where the equipartition theorem holds, can be used; and (ii) the growth rate of the critical mode is proportional to  $\epsilon$  for  $-1 \leq \epsilon < 0$ . This yields in scaled units for a slab of volume V (for a factor of 2 see [47])

$$\left\langle |\overline{\theta}_{q_c}|^2 \right\rangle_{\text{est}} = \frac{2Q_0 d^3}{V(K_{33}q_c^2 + \pi^2)} \left(-\frac{1}{\epsilon}\right). \tag{86}$$

In Appendix D it is shown that in the limit  $\epsilon \to -1, P_2 \to 0$  at arbitrary  $P_1$  (or  $P_1 = P_2 \to 0$  at arbitrary  $\epsilon$ ) our result (76) coincides with the estimate and with the equipartition theorem, an important check of consistency. In Fig. 3 the estimate is compared with our theory (76) for nonzero driving forces by plotting the ratio

$$\frac{\langle |\overline{\theta}_{q_c}|^2 \rangle}{\langle |\overline{\theta}_{q_c}|^2 \rangle_{\text{est}}} = 1 + (0.179P_1 + 2.81P_1^2)R_0 + \text{h.o.t.}$$
(87)

for  $P_2 = 0$ . For realistic  $P_1$  corresponding, e.g., to d = 13  $\mu m$  in [6] or  $d = 23 \ \mu m$  in [8] our model yields at  $\epsilon = 0$  fluctuations which are enhanced by a factor 1.1 or 1.03, respectively. The fact that the corrections are so small must be considered as fortuitous since Q and  $\sigma$  differ separately considerably more from the simple estimates.

Measurements of director fluctuations in [6-8] were performed by means of the shadowgraph method [45, 46]; For normal-roll fluctuations in Fourier space [47] the ratio of the measured intensity and the value of the estimate was 1.4 in Ref. [6]  $(d = 13 \ \mu m)$  and 1.18 in Ref. [8]  $(d = 23 \ \mu m)$ .

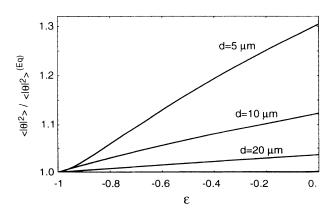


FIG. 3. Fluctuations of the Fourier-transformed director component  $\overline{\theta} = n_z(z = 0)$  at the critical wave number as function of  $\epsilon$ . Shown is the ratio of our result and the equipartition-theorem estimate. The parameter is the slab thickness d = 5, 10, and 20  $\mu$ m.

#### B. Scaling behavior of the director fluctuations near the Lifshitz point

The evaluation of fluctuations at equal times with (76), (18), (16), and (37), near the Lifshitz point  $\omega_0 = \omega_z$  reveals interesting exponents. Some results for correlations of the field  $\theta_{q_c}(y,t) := \frac{1}{2\pi} \int dx e^{-iq_c x} \theta(x,y,t)$ , i.e., the director component  $n_z$ , Fourier transformed only in the x direction, are (see Fig. 4.)the following.

(i) The fluctuation intensity shows a crossover from the usual one-dimensional law

$$|| heta_{q_c}(y,t)|^2
angle=Q/(2\pi)|2\sigma_\epsilon\sigma_{m{pp\omega}}|^{-1/2}|\Delta\omega_0\epsilon|^{-1/2}$$

to

(

$$|| heta_{q_c}(y,t)|^2
angle = Q/(4\pi)|\sigma_\epsilon^3\sigma_{pppp}/6|^{-1/4}|\epsilon|^{-3/4}$$

The crossover occurs at  $|\epsilon| \approx (\Delta\omega_0)^2/\alpha$ , where  $\alpha = 2\sigma_{pppp}\sigma_{\epsilon}/(3\sigma_{pp\omega}^2)$ , and  $\Delta\omega_0 = \omega_0 - \omega_z$  denotes the distance from the Lifshitz frequency  $\omega_z$ .

(ii) A similar crossover from  $|\epsilon|^{-1/2}$  to  $|\epsilon|^{-1/4}$  occurs for the correlation length in the same region of parameter space.

In the normal-roll limit  $\Delta\omega_0 \gg |\alpha\epsilon|^{1/2}$  the expressions for normal rolls are obtained by identifying  $\sigma_{pp\omega}\Delta\omega_0$ with  $\sigma_{pp}$ . In the oblique-roll limit  $\Delta\omega_0 \ll -|\alpha\epsilon|^{1/2}$ , an expansion of (37) around the new minima  $p_{\min}$  =

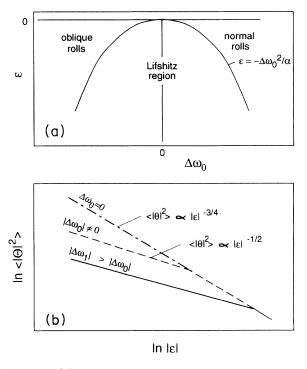


FIG. 4. (a) Regions in control-parameter space where the intensity and correlation function exhibit normal-roll or oblique-roll behavior, or the different scaling near the Lifshitz point (see text). (b) Fluctuation intensity of the director mode, Fourier transformed only in x direction at  $k_{xc}$ , as function of  $\epsilon$  for two values of the distance  $|\Delta \omega_0|$  from the Lifshitz point (straight and dashed line) and at the Lifshitz point (dash-dotted line).

 $\pm(-\Delta\omega_0\sigma_{pp\omega}/\sigma_{pppp})^{1/2}$  yields the results for oblique rolls with  $p_c=p_{\rm min}.$ 

Preliminarily measurements appear to show the predicted behavior, at least qualitatively [30].

## **V. DISCUSSION**

In this paper an efficient scheme to calculate thermal fluctuations of physical fields in extended patternforming nonequilibrium systems is presented. It contains two main steps: the derivation of the stochastic forces of the Langevin equation associated with the macroscopic equations of motion and the calculation of the stochastic terms of the amplitude equations describing the nearthreshold fluctuations. The scheme is then applied to calculate the director fluctuations of nematic liquid crystals in a thin slab near the threshold of EHC.

In Sec. II general expressions for the fluctuation strength of the stochastic amplitude equations are given for general classes of systems including RBC, TCF, and the various electrically driven transitions in liquid crys-Important types of bifurcations such as those tals. in isotropic systems, codimension-two bifurcations to oblique rolls in anisotropic systems, and traveling waves were discussed. The case of time-periodic control parameters of the system is included (sufficiently rapid and nonresonant). Below theshold, fluctuations of all critical modes have to be considered and the resulting correlations exhibit the full symmetry of the system. This is confirmed by experiments for RBC [1, 3] and for EHC in the oblique-roll regime [30]. We showed that above threshold the fluctuating forces of the stochastic amplitude equations are the same as below within the range of applicability of these equations. Then phase fluctuations become important.

In Sec. III the stochastic forces of the macroscopic Langevin equations for electroconvection using the Landau approach are calculated. Thus we wrote all constitutive relations containing dissipative effects in terms of a generalized Onsager form and added fluctuating forces determined by the fluctuation-dissipation theorem. The matrix of the fluctuating forces takes the form  $\underline{O}(\nabla, u) = \underline{D}(\underline{M} + \underline{M}^T)\underline{D}^{\dagger}$ , where  $\underline{M}$  is the Onsager matrix and  $\underline{D}$  the matrix-differential operator, which acts on the Onsager fluxes in the macroscopic equations. This form should be generic also for other systems as long as one can write the entropy production in terms of suitable Onsager forces and fluxes and the fluxes can be identified in the macroscopic equations.

The crucial assumption in this approach is that, although the systems considered are in general far from global equilibrium, the fluctuation-dissipation theorem, derived near equilibrium, can be applied in terms of local variables (local equilibrium). This in turn is essentially equivalent to linear responses (Onsager fluxes) to local gradients (Onsager forces) and is therefore fulfilled if the Onsager matrix  $\underline{M}$  does not depend on the forces. In anisotropic systems, however,  $\underline{M}$  depends on the macroscopic variables  $\boldsymbol{u}$  itself, in our case on the local director orientation. This leads to multiplicative contributions in the noise terms of the macroscopic equations, reminiscent of the nonlinear generalization of the fluctuationdissipation theorem as discussed in Appendix A. The multiplicative terms can be neglected at least for the case of EHC within the range of applicability of the amplitude equation, so that the subtleties inherent in nonlinear Langevin equations with multiplicative stochastic forces [38] do not arise. Since we made no specific assumptions, this should be valid also generally.

In contrast to systems already investigated such as ordinary fluids at rest [22], Rayleigh-Bénard convection in simple fluids [12, 14], and binary mixtures [15], calculations for EHC are much more complex due to the anisotropy of the fluid leading to complicated constitutive relations with many material parameters. Moreover the identification of the local Onsager fluxes in the macroscopic equations is not obvious for the director equations. In the scaled units used here [see Eq. (51)], the resulting matrix  $\underline{O}$  of the fluctuation strengths is proportional to the small quantity  $Q_0 = (kT)/(K_{11}d)$ , the ratio of a molecular interaction length and the slab thickness d.

In Sec. III C the general stochastic equations of electroconvection derived in the Secs. III A and III B are specialized to normal rolls in a periodically driven system with planar boundary conditions. It is shown that in the conduction regime cross correlations between the stochastic forces of the charge conservation equation and those of the other equations cancel out due to different time symmetries. This would hold even if there were nonvanishing cross correlations in the original stochastic forces.

In Sec. IV the resulting equal-time director fluctuations are calculated and compared with experimental results and a simple estimate based on the equipartition theorem. In the limit of zero external field our method yields the same fluctuations as the equipartition theorem. an important check of consistency. For nonzero external fields there are corrections proportional to the system parameters  $P_1 = T_q/(\pi^2 T_d)$  and  $P_2 = T_{\text{visc}}/T_d$  denoting charge and velocity relaxation time, respectively, in units of the director relaxation time. The corrections in  $P_2$  can be neglected while the  $P_1$  terms explain at least in part the difference between the measured fluctuations and the equipartition-theorem estimate and they give the right trend, because they increase with  $P_1 \propto 1/d^2$ . Since the estimate is based on director fluctuations alone, corrections can be attributed to charge and velocity fluctuations. For  $P_1 \rightarrow 0$  the fluctuations are equivalent to an adiabatic elimination of the electric potential  $\phi$  via  $\boldsymbol{\nabla}\cdot\boldsymbol{j}^{(\mathrm{cond})}=0$  from the beginning on, so all terms containing  $P_1$  or  $P_2$  can be attributed to the effects of charge and velocity fluctuations, respectively. This confirms the assumption that in a composite system the relative influence of its constituents to fluctuations is proportional to the respective time scales. The relative influence of charge fluctuations  $\propto P_1 \approx 0.285 \; (\mu {
m m}/d)^2$  cannot be neglected for thin slabs and becomes of the order of the director fluctuations for  $d \approx 5 \ \mu \text{m}$ . For thinner slabs, however, our calculations would have to be generalized because the assumptions  $\omega_{0,abs}T_d \gg 1$  and  $\omega_{0,abs}T_q < 1$ can no longer be fulfilled simultaneously. In nematic materials the relative influence of the velocity fluctuations is

of the order  $10^{-6}$  and therefore can always be neglected, in accordance with [28]. In fact, one could have eliminated the velocities adiabatically right from the beginning. Note, however, that the velocity field influences the physical fluctuations Eq. (76) by virtue of its backflow effect on the growth rate  $\sigma$ . To obtain simple expressions, all the calculations in Sec. IV have been done in the limit  $\omega_0 \ll \omega_{\rm cutoff}$ , where  $R_c$  is independent of the ac frequency  $\omega_0$ . The extension to larger external frequencies in the conduction regime, starting from Eq. (70), is straightforward.

We have presented the methods in some generality before applying them to EHC, so the work may serve as a starting point for other complex fluids. To test the method it was applied to RBC in simple fluids and in binary mixtures, yielding the same results as Graham [12], Hohenberg and Swift [14], and Schöpf and Zimmermann [15]. Application to Taylor-Couette flow is also straightforward.

#### ACKNOWLEDGMENTS

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## APPENDIX A: COMPARISON OF THE LANDAU APPROACH WITH THE GLOBAL APPROACH

In this approach adopted by San Miguel and Sagues to study magnetic field induced transient patterns in planarly aligned nematic liquid crystals [20], the original variables of the system itself are taken as the variables of the GOF, Eq. (55). This requires that one can find an appropriate entropy functional or alternatively, in the isothermal case, a free-energy functional  $\mathcal{F} = -TS$ , which allows the representation of the deterministic part of the original equations as

$$\partial_t \boldsymbol{u} = \underline{\boldsymbol{S}}^{-1}[-\underline{\boldsymbol{L}}\boldsymbol{u} + \boldsymbol{N}(\boldsymbol{u})] = \underline{\tilde{\boldsymbol{M}}}(\boldsymbol{u})\frac{\delta \boldsymbol{S}}{\delta \boldsymbol{u}}.$$
 (A1)

The GOF of the global approach is necessarily nonlinear, so that the FDT has to be formulated for nonlinear systems. This has been done in Ref. [41]. The result is that the FDT remains unchanged, if an additional condition

$$\int d^3 r \frac{\delta}{\delta u_{\alpha}} \left( \underline{\underline{M}}^{(as)}(\underline{u})_{\alpha\beta} \frac{\delta}{\delta u_{\beta}} \right) = 0$$
 (A2)

is fulfilled. This corresponds to a generalization of the Liouville theorem for the conservative part of the dynamics.

The Landau approach has the following advantages.

(i) It has a broader range of applicability: While the Landau approach requires the system only to be near local equilibrium, there are many systems, including EHC, for which a transformation to a GOF in terms of the original variables is not yet found or does not exist.

(ii) The additional condition (A2) restricts the range of applicability of the global approach further. Note that for a linear GOF, this condition is always fulfilled.

(iii) Since the constitutive relations are autonomous in any case, the Landau approach can be extended to nonautonomous systems such as EHC.

## APPENDIX B: THE ONSAGER MATRIX FOR THE EHC SYSTEM OF SEC. III C (SCALED UNITS)

$$\begin{split} M_{ij,kl}^{(11)} &= Q_0 \Bigg[ \alpha_1 n_i n_j n_k n_l + \frac{\alpha_4}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \\ &+ \frac{\alpha_5 + \alpha_6}{4} (\delta_{ik} n_j n_l + \delta_{il} n_j n_k \\ &+ \delta_{jk} n_i n_l + \delta_{jl} n_i n_k) \Bigg], \end{split}$$

$$M_{ij,kl}^{(12)} = Q_0 \Bigg[ -rac{lpha_2 + lpha_3}{4} (\delta_{ik} n_j n_l - \delta_{il} n_j n_k + \delta_{jk} n_i n_l - \delta_{jl} n_i n_k) \Bigg],$$

$$\begin{split} M^{(22)}_{ij,kl} &= Q_0 \Bigg[ \left. \frac{\alpha_3 - \alpha_2}{4} (\delta_{ik} n_j n_l - \delta_{il} n_j n_k - \delta_{jk} n_i n_l \right. \\ &+ \delta_{jl} n_i n_k ) \Bigg], \end{split}$$

$$M(33)_{ij} = Q_0 P_1 \left[ \sigma_\perp \delta_{ij} + \sigma_a n_i n_j \right]. \tag{B1}$$

## APPENDIX C: ANALYTICAL EXPRESSIONS FOR THE FLUCTUATING FORCE OF THE STOCHASTIC AMPLITUDE EQUATION AND FOR THE DIRECTOR FLUCTUATIONS

In lowest-order Galerkin approximation and for  $1 \ll \omega_0 \ll 1/P_1$  the threshold  $R_c$  and the time constant  $T_0$  of the amplitude equation (78) are given in the normal-roll regime by

$$R_c = -\frac{SK\sigma_0}{SK\epsilon_a - \sigma_0\sigma_a q^2 E_q},\tag{C1}$$

$$T_0 = -\frac{1}{\sigma_0} (1 + S_1 P_1 + S_2 P_2), \tag{C2}$$

$$\sigma_0 = \frac{-K}{1 - q^4 I^2 / O_{vv}},\tag{C3}$$

$$E_q = \frac{q^2 E I^2}{O_{vv}} - \epsilon_a,\tag{C4}$$

$$S_1 = -(\epsilon_a R_c + \sigma_0) \left(\frac{E}{S} - \frac{\epsilon_a}{\sigma_a}\right), \tag{C5}$$

$$S_2 = -\frac{q\sigma_0 S_{vv}}{KO_{vv}} \left[ K + \epsilon_a R_c \left( q^2 \frac{\sigma_a}{S} - 1 \right) \right].$$
(C6)

The fluctuating force  $\langle f_A^* f_A \rangle = T_0^2 Q \delta(x - x') \delta(y - y') \delta(t - t')$ , Eq. (79), is given by

$$Q = 2Q_0 B\left(\frac{1+O_1 P_1}{(1+S_1 P_1+S_2 P_2)^2}\right),$$
 (C7)

$$O_1 = -\frac{KS}{\sigma_0 R_c} \left(\frac{\sigma_0 + \epsilon_a R_c}{q \sigma_a}\right)^2,\tag{C8}$$

$$B = -\frac{\sigma_0}{K}.$$
 (C9)

The quantities  $E, S, K, S_{vv}$ , and  $O_{vv}$  are given by the Galerkin integrals of Eq. (74)

$$E = \epsilon_{\parallel} q^{2} + \pi^{2},$$

$$S = \sigma_{\parallel} q^{2} + \pi^{2},$$

$$K = K_{33}q^{2} + \pi^{2},$$

$$S_{vv} = [q^{2} - (\partial_{z}^{2})_{vv}]/(iq),$$

$$O_{vv} = \frac{1}{2}(2 + \alpha_{4} + \alpha_{6})q^{4} - (1 + \alpha_{1} + \alpha_{4} + \alpha_{6})q^{2}(\partial_{z}^{2})_{vv}$$

$$+ \frac{1}{2}(\alpha_{4} + \alpha_{6})(\partial_{z}^{4})_{vv},$$
(C10)

with

$$I = \int_{-1/2}^{1/2} dz \sqrt{2} \cos \pi z \ \mathcal{C}_1(z) \approx 0.986, \qquad (C11)$$

$$(\partial_z^2)_{vv} = \int_{-1/2}^{1/2} dz \mathcal{C}_1(z) \partial_z^2 \mathcal{C}_1(z) \approx -12.3,$$
 (C12)

$$(\partial_z^4)_{vv} = \int_{-1/2}^{1/2} dz \mathcal{C}_1(z) \partial_z^4 \mathcal{C}_1(z) \approx 501.$$
(C13)

All quantities are taken at  $q = q_c$ .

Near threshold and for  $P_2 = 0$ , the resulting director fluctuations, Eq. (76), are enhanced with respect to the simple model by a factor

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$$\frac{\langle |\bar{\theta}_q|^2 \rangle}{\langle |\bar{\theta}_q|^2 \rangle_0} = \frac{1 + O_1 P_1}{1 + S_1 P_1} \approx \frac{1 + 147 P_1}{1 + 92.4 P_1},\tag{C14}$$

where  $\langle |\overline{\theta}_q|^2 \rangle_{\text{est}} = \langle |\overline{\theta}_q|^2 \rangle_{\text{eq}} (-1/\epsilon)$  and  $\langle |\overline{\theta}_q|^2 \rangle_{\text{eq}}$  is the equilibrium result (D3).

## APPENDIX D: COMPARISON OF THE DIRECTOR FLUCTUATIONS FOR ZERO EXTERNAL FIELD WITH THE EQUIPARTITION THEOREM

The starting point is Eq. (76), which gives the fluctuations of a Fourier component of  $n_z(z=0)$  with normalroll wave number  $\mathbf{k} = (q, 0)$ . Inserting (9) and (8) yields

$$\langle |\bar{\theta}_{q}|^{2} \rangle = \frac{\tilde{f}_{q,n_{z}}^{2}}{2(2\pi)^{2}} \frac{(\tilde{f}_{q}^{\dagger}, \underline{\tilde{O}}_{q} \tilde{f}_{q}^{\dagger})}{(\tilde{f}_{q}^{\dagger}, \underline{\tilde{S}}_{q} \tilde{f}_{q})(\tilde{f}_{q}^{\dagger}, \underline{\tilde{L}}_{q} \tilde{f}_{q})}.$$
 (D1)

With the lowest-order Galerkin ansatz one gets

$$\langle |\bar{\theta}_{q}|^{2} \rangle = \frac{1}{(2\pi)^{2}} \frac{\boldsymbol{f}^{\dagger} \cdot \boldsymbol{\underline{O}}^{(\text{Gal})} \cdot \boldsymbol{f}^{\dagger}}{(\boldsymbol{c}^{\dagger *} \cdot \boldsymbol{\underline{S}}^{(\text{Gal})} \cdot \boldsymbol{c})(\boldsymbol{c}^{\dagger *} \cdot \boldsymbol{\underline{L}}^{(\text{Gal})} \cdot \boldsymbol{c})}, \qquad (\text{D2})$$

where the eigenvectors  $\mathbf{c} = (c_{\phi}, c_{\overline{\phi}}, 1, c_v)$  and  $\mathbf{f}^{\dagger} = (c_{\phi}^{\dagger}, c_{\overline{\phi}}^{\dagger}, 1, c_v^{\dagger})$  and the Galerkin matrices depend on q and  $\epsilon$ . For  $\epsilon \to -1$  the charge components and the other components of  $\underline{\tilde{L}}_q$  are uncoupled, so the charge component of eigenvectors with a nonzero director component vanishes. For the director-dominated mode one gets for  $P_2 = 0$  the zero-field growth rate  $\sigma(\epsilon = -1) = \sigma_0$  and the eigenvectors  $\mathbf{c} = (0, 0, 1, c_v)$  and  $\mathbf{f}^{\dagger} = (0, 0, 1, -q^2 I/O_{vv})$ . Inserting all this into (D2) yields for arbitrary  $P_1$ 

$$\begin{aligned} \langle |\bar{\theta}_{q}|^{2} \rangle (\epsilon = -1) &= \frac{2Q_{0}}{(2\pi)^{2}K} = \frac{2Q_{0}}{(2\pi)^{2}(K_{33}q^{2} + K_{11}\pi^{2})} \\ &= \langle |\bar{\theta}_{q}|^{2} \rangle_{\mathrm{Eq}}. \end{aligned} \tag{D3}$$

This result is derived for a laterally infinite system. The transformation to finite lateral dimensions  $L_x$  and  $L_y$  of the slab yields a factor  $(2\pi)^2 d^2/(L_x L_y) = (2\pi)^2 d^3/V$ , which gives the equipartition result (86) of the estimate for  $\epsilon = -1$ .

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