

Macroscopic dynamics of the ferroelectric smectic C_F phase with C_{1h} symmetry

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We present the macroscopic dynamics of ferroelectric smectic C , smectic C_F , liquid crystals reported recently experimentally by two groups. In this fluid and tilted smectic phase, the macroscopic polarization, $\mathbf{P}$, is tilted with respect to the layer normal, thus giving rise to C_{1h} overall symmetry for this phase in the spatially homogeneous limit. A combination of linear irreversible thermodynamics and symmetry arguments is used to derive the resulting dynamic equations applicable at sufficiently low frequencies and sufficiently long wavelengths. Compared to nonpolar smectic C phases, we find two static cross-coupling terms between compression of the layering and bending of the layers, which do not lead to elastic forces but to elastic stresses. In addition, a number of static cross-coupling terms is elucidated, which can exist in smectic C_F but not in the ferroelectric A_F phase because of the polar in-plane preferred direction in smectic C_F . Due to the fact that the magnitude of the polarization is a slowly relaxing variable, the velocities of the first and second sound both reflect the monoclinic symmetry of the ground state, thus rendering all sound velocities to be biaxial. We also analyze reversible cross-coupling terms between elongational flow and electric fields as well as temperature and concentration gradients, which lend themselves to experimental detection. Such cross-coupling terms have apparently never been considered before for monoclinic symmetry since they are absent without the presence of a polar direction and thus do not exist in nonpolar smectic C phases. Among the dissipative cross-coupling terms characteristic of smectic C_F , we find that bending of the layers can couple to temperature gradients. We also address the question how the linear $\mathbf{P} \cdot \mathbf{E}$ coupling in the energy alters the macroscopic response behavior when compared to usual nonpolar smectic C phases.

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I. INTRODUCTION

Recently, two groups described the discovery of truly ferroelectric smectic C phases [1,2], which we call C_F throughout this paper, and they analyzed the physical properties of this phase [1,2]. In this phase, the macroscopic electric polarization is oriented along the average axis of the molecules and thus a liquid crystalline phase of C_{1h} symmetry results, since—when compared to the nonpolar smectic C phase [3]—the twofold axis disappears, thus reducing the symmetry from C_{2h} to C_{1h} . It is the goal of the present paper to derive macroscopic dynamic equations describing this phase and to suggest further experiments to elucidate its physical properties.

The discovery of the smectic C_F phase followed about two years after the ferroelectric analog of smectic A_F had been announced [4–6]. In smectic A_F , the macroscopic polarization is parallel to the layer normal and there is isotropic fluidity in the layer planes, giving rise to $C_{\infty v}$ symmetry in the ground state. In contrast, in the ferroelectric C_F phase, one has a tilted

polar preferred direction, since the macroscopic polarization encloses a finite angle with the layer planes.

About the same time as the first reports of the smectic A_F phase, an antiferroelectric smectic phase, Z_A , was also experimentally found and analyzed [7–11]. In this phase, the macroscopic polarization lies in the layer planes and changes its direction from one layer to the next. Thus, overall, a biaxial phase with globally D_{2h} symmetry results in the absence of external electric fields. As a consequence, the Z_A phase shares many macroscopic properties with the McMillan C_M phase [3] described [12–14] and experimentally investigated [15] quite some time ago. Naturally, the response to sufficiently high electric fields is qualitatively different from that of C_M , including a double hysteresis loop characteristic of antiferroelectric phases [8].

Therefore, the recently discovered smectic C_F phase is a natural addition to the multiferroic liquid crystalline (LC) phases described earlier. Here we denote by multiferroic LC phases, phases which show, in addition to LC order, also long-range electric and/or magnetic order. The multiferroic LC phases found so far experimentally in chronological order include ferromagnetic nematics [16–18] and cholesterics [19], ferroelectric nematics [20,21], ferroelectric smectic A_F [4–6], antiferroelectric smectic Z_A [7–11], and now the tilted ferroelectric smectic C_F [1,2]. We note that, so far, to our knowledge no truly ferromagnetic smectic phases or antiferromagnetic smectic phases have been found experimentally and that their macroscopic properties have been elucidated only very recently theoretically [22,23].

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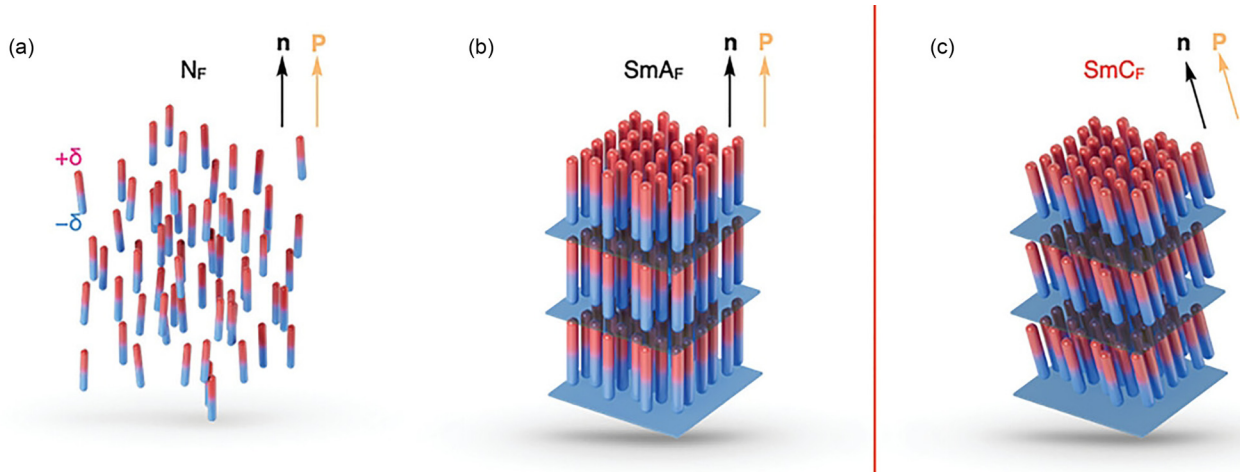


FIG. 1. Overview of ferroelectric nematic N_F (a), ferroelectric smectic A_F (b) and ferroelectric smectic C_F (c). The director is indicated by $\mathbf{n}$ and the macroscopic polarization by $\mathbf{P}$. In ferroelectric smectic C_F , the director and the macroscopic polarization are parallel and include a finite angle with the layer normal. (Adapted from Ref. [1].)

In the present paper, we characterize the ferroelectric smectic C_F phase in the framework of macroscopic dynamics, thus generalizing previous work on the macroscopic dynamics of usual nonpolar smectic C phases [3,24,25] as well as of ferroelectric nematics [26–28]. In Fig. 1, we include a plot of the three truly ferroelectric liquid crystalline phases known to date: ferroelectric nematics N_F , ferroelectric smectic A_F , and ferroelectric smectic C_F . To derive the macroscopic dynamic equations, we use the combination of irreversible thermodynamics and of symmetry arguments, including the behavior of the macroscopic variables along with that of the associated currents and quasicharges under time reversal and parity (spatial inversion) as well as under rigid rotations and Galilei transformations [24,25,29–31]. There are three classes of macroscopic variables which must be taken into account: (a) conserved quantities, (b) variables associated with a spontaneously broken continuous symmetry, and (c) macroscopic variables, which relax on a sufficiently long timescale, including in the present context the slow spatiotemporal variations of the modulus of the polarization. The approach of macroscopic dynamics has been applied to many condensed matter systems including spin waves in magnets [31,32], nematic liquid crystals [3,24,31,33], and polymeric liquids [34–37], as well as superfluids including superfluid ^4He [38,39] and the superfluid phases of ^4He [40–43], and—most recently—to ferroelectric smectic A_F [44], antiferroelectric smectic Z_A , and antiferromagnetic smectic Z_M phases [23].

The paper is organized as follows. In Sec. II, we discuss in detail the macroscopic variables used: conserved quantities, variables associated with spontaneously broken continuous symmetries and macroscopic variables, which relax on long, but finite and macroscopically relevant timescales. As additional variables associated with spontaneously broken continuous symmetries, we have the layer displacement as in all fluid smectic phases and the spatiotemporal variations of the polar in-plane director. And as the most important macroscopic variable, the modulus of the macroscopic electric polarization enters the picture. In Sec. III, we present the free-energy functional, thereby defining the thermodynamic conjugates. The macroscopic dynamic equations (Sec. IV) are

followed in Secs. V and VI by the reversible and dissipative phenomenological currents. In Sec. VII, we analyze the effects of selected cross-coupling terms which might be suitable for experimental detection in ferroelectric smectic C_F and help to identify the phase. A summary and perspective is given in Sec. VIII.

II. VARIABLES

To derive a complete set of macroscopic dynamic equations in the low-frequency, long wavelength limit, the procedure is as follows. First, one identifies the symmetries of the ground state. This is followed by the evaluation of the macroscopic variables due to conservation laws, variables associated with spontaneously broken continuous symmetries and macroscopic variables, which relax on a sufficiently long, but finite timescale to be macroscopically relevant. Next, the assumption of local thermodynamic equilibrium is used to establish the Gibbs relation containing the variations of the variables and serving to introduce the thermodynamic conjugate quantities. To derive a self-consistent set of equations is then a two-step procedure. First, one relates the thermodynamic forces and conjugate quantities to the variables or, equivalently, one expands a generalized thermodynamic potential (e.g., a generalized free energy functional) into the hydrodynamic variables and their spatial derivatives. In a second step, the currents and quasicharges arising in the dynamic equations for the macroscopic variables are related to the thermodynamic forces. Currents and quasicharges can be split into two contributions: reversible parts that do not give rise to any entropy production and dissipative parts, which lead to entropy generation thus guaranteeing that the second law of thermodynamics is fulfilled.

In this section of the paper, we discuss the macroscopic variables that must be taken into account for ferroelectric smectic C_F phases. Classical nonpolar smectic C phases possess monoclinic C_{2h} symmetry [3,24]. They are characterized by two truly hydrodynamic variables, namely, the layer displacement and variations of the in-plane director associated with broken rotational symmetry [24]. For the recently

discovered ferroelectric smectic C_F phases [1,2] (compare Fig. 1), the symmetry is reduced because of the presence of a macroscopic polarization, P_i . As a consequence, the symmetry is reduced from C_{2h} to C_{1h} . We still have monoclinic symmetry due to the existence of a horizontal mirror plane, while the twofold axis no longer exists due to the presence of a polar vector, P_i .

Two fluid effects as they arise in immiscible mixtures [45,46] are not the subject of this paper, since we will assume the presence of a miscible system.

The list of macroscopic variables includes the entropy density, σ , the mass density, ρ , the density of linear momentum, g_i , and the macroscopic polarization, P_i . We split P_i into its magnitude, $|P_i| = P$, and its orientation P_i/P . The latter gives rise to the normalized polar preferred direction $\hat{p}_i$ perpendicular to the layer planes,

$$\hat{p}_i = \hat{k}_i(\hat{k}_j P_j) / \sqrt{(P_l \hat{k}_l)^2}, \quad (1)$$

with the layer normal denoted by $\hat{k}_i$. We emphasize that $\hat{p}_i$ is not parallel to the macroscopic polarization, P_i , since C_F is a tilted phase in contrast to the ferroelectric smectic A_F phase. Similarly, there is a preferred normalized polar in-plane direction, $\hat{c}_i$, defined by

$$\hat{c}_i = (P_i - \hat{k}_i \hat{k}_j P_j) / \sqrt{P^2 - (P_l \hat{k}_l)^2}, \quad (2)$$

which is perpendicular to $\hat{p}_i$.

Using $\hat{p}_i$ and $\hat{c}_i$, we can construct a third direction orthogonal to both $\hat{p}_i$ and $\hat{c}_i$:

$$\hat{l}_i = \epsilon_{ijk} \hat{p}_j \hat{c}_k. \quad (3)$$

The strength of the polar order, P , is the order parameter, and δP is not a conserved quantity. For very fast relaxation times, P does not couple to the macroscopic variables and will be no longer kept on the list of variables, while for sufficiently long relaxation times its dynamic equation will be incorporated.

Since the system we study, smectic C_F , has a layered structure, we keep u , the layer displacement along the layer normal, in our list of macroscopic variables,

$$u = u_i \hat{p}_i, \quad (4)$$

similar to the nonpolar smectic C . There is, however, an important difference: In the C_F phase depicted in Fig. 1, u is a component of the general displacement vector along one of the two polar directions. Thus u is a scalar under inversion, while for the nonpolar C phase, $u = \hat{k}_i u_i$ is a pseudoscalar, since $\hat{k}_i$ is a nematic-type quantity.

In the absence of an external electric field, there is a second truly hydrodynamic variable associated with the broken rotational symmetry in the layer planes which we denote by δc :

$$\delta c = \hat{l}_i \delta \hat{c}_i. \quad (5)$$

We note that in the smectic C_F phase, δc transforms like a pseudoscalar, since $\hat{p}_i$, $\hat{c}_i$, and δc_i all transform odd under parity. We mention in passing that the number of variables associated with spontaneously broken symmetries is thus the same for nonpolar smectics C and ferroelectric smectic C_F , although the physical properties are quite different for the two different phases.

The presence of impurities, contaminants, etc. can be taken care of by adding a concentration variable ϕ . We assume the associated mass density, ρ_s , with $\rho_s = \rho\phi$ to be conserved, i.e., there are no chemical reactions involved [25].

The first law of thermodynamics relates changes of the variables to changes of the energy density ε by the Gibbs relation [25,29]:

$$d\varepsilon = T d\sigma + \mu d\rho + \Pi d\phi + h_i^u d\nabla_i u + h_i^c d\nabla_i c + h^P dP + E_i dD_i + v_i dg_i. \quad (6)$$

The electric displacement field D_i is related to the charge density ρ_e by $\nabla_i D_i = -\delta\rho_e$ as the independent variable. The conjugate field E_i is the external electric field. Our notation follows closely that of Refs. [26,47].

The Gibbs relation contains the entropy density σ , representing the thermal degree of freedom, with its thermodynamic conjugate, the temperature T . Other conjugates are the chemical potential μ , the osmotic pressure Π , the mean velocity $v_i = g_i/\rho$, the field h_i^u is conjugate to $\nabla_i u$, the field h_i^c is conjugate to $\nabla_i c$, and the molecular field h^P associated with the magnitude of the polarization P .

III. STATIC CONSIDERATIONS

After having determined the symmetry of the ground state, the macroscopic variables and the Gibbs relation in the last section, we now turn our attention to set up the generalized energy density and to determine from that expression the thermodynamic forces.

The static behavior of the macroscopic system studied here is conveniently described by the energy functional in harmonic approximation, Refs. [26,48]. Disregarding electric fields for the moment and including the kinetic energy density, we get

$$\begin{aligned} \varepsilon = & \frac{1}{2} c_P (\delta P)^2 + \frac{1}{2} K_{ij}^{(2)} (\nabla_i P) (\nabla_j P) + \frac{1}{2} K_{ij}^{(c)} (\nabla_i c) (\nabla_j c) + \frac{1}{2} B (\nabla_z u)^2 + (\nabla_z u) (C_1 \nabla_x^2 u + C_2 \nabla_y^2 u) + \frac{1}{2} A_{ijkl} (\nabla_i \nabla_j u) (\nabla_k \nabla_l u) \\ & + D_{ijk} (\nabla_i \nabla_j u) (\nabla_k c) + (\nabla_z u) (d_1 \delta P + d_2 \nabla_z P + d_3 \nabla_x P) + (\nabla_x^2 u) (d_{31} \delta P + d_{41} \nabla_z P + d_{51} \nabla_x P) + (\nabla_y^2 u) (d_{32} \delta P + d_{42} \nabla_z P \\ & + d_{52} \nabla_x P) + \frac{1}{2} c_{\rho\rho} (\delta\rho)^2 + \frac{1}{2} c_{\sigma\sigma} (\delta\sigma)^2 + \frac{1}{2} c_{\phi\phi} (\delta\phi)^2 + c_{\rho\phi} (\delta\rho) (\delta\phi) + c_{\rho\sigma} (\delta\rho) (\delta\sigma) + c_{\sigma\phi} (\delta\sigma) (\delta\phi) + (\psi_1 \delta\rho + \psi_2 \delta\sigma \\ & + \psi_3 \delta\phi + \psi_4 \delta P) \nabla_y c + (\gamma_1 \delta\rho + \gamma_2 \delta\sigma + \gamma_3 \delta\phi) \delta P + (\theta_1 \delta\rho + \theta_2 \delta\sigma + \theta_3 \delta\phi) \nabla_z P + (\theta_4 \delta\rho + \theta_5 \delta\sigma + \theta_6 \delta\phi) \nabla_x P \\ & + (\hat{\theta}_1 \delta\rho + \hat{\theta}_2 \delta\sigma + \hat{\theta}_3 \delta\phi) \nabla_z u + (\bar{\theta}_1 \delta\rho + \bar{\theta}_2 \delta\sigma + \bar{\theta}_3 \delta\phi) (\nabla_y^2 u) + (\bar{\theta}_4 \delta\rho + \bar{\theta}_5 \delta\sigma + \bar{\theta}_6 \delta\phi) (\nabla_x^2 u) + \frac{1}{2\rho} \mathbf{g}^2, \end{aligned} \quad (7)$$

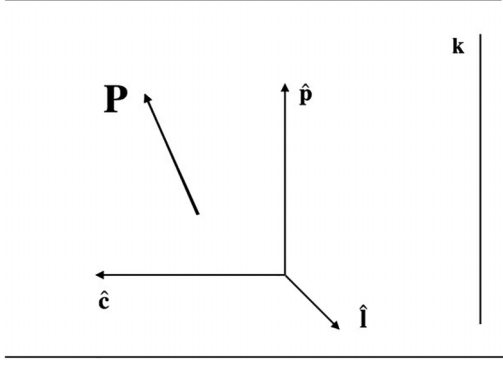


FIG. 2. Figure 2 shows one smectic layer from the side. The layer normal is along the direction $\mathbf{k}$, while $\mathbf{P}$ is the oblique polarization of the C_F phase. The tripod of unit vectors contains the polar vector $\hat{\mathbf{p}}$ [along the layer normal, Eq. (1)], the in-layer polar vector $\hat{\mathbf{c}}$ [in the $\mathbf{P}/\mathbf{k}$ plane, Eq. (2)] and $\hat{\mathbf{i}}$ is perpendicular to both, pointing out of the drawing plane. For explicit calculations, $\hat{\mathbf{p}}$ is taken as the $\hat{\mathbf{z}}$ direction, $\hat{\mathbf{c}}$ as the $\hat{\mathbf{x}}$ direction, and $\hat{\mathbf{i}}$ is taken as the $\hat{\mathbf{y}}$ direction.

where ∇_z is shorthand notation for $\hat{p}_i^0 \nabla_i$, ∇_x for $\hat{c}_i^0 \nabla_i$, and ∇_y for $\hat{i}_i^0 \nabla_i$. In Fig. 2, we show schematically the coordinate system used throughout the rest of the paper whenever referring to directions explicitly. $\hat{\mathbf{z}}$ is taken to be parallel to the layer normal $\hat{\mathbf{p}}$, $\hat{\mathbf{x}}$ parallel to the in-plane preferred direction $\hat{\mathbf{c}}$ and $\hat{\mathbf{y}}$ is in the direction orthogonal to both, $\hat{\mathbf{p}}$ as well as $\hat{\mathbf{c}}$. A δ denotes deviations from the equilibrium value, in particular, $\delta P = P - P_0$, $\delta \hat{p}_i = \hat{p}_i - \hat{p}_i^0$, $\delta \phi = \phi - \phi_0$, $\delta \rho = \rho - \rho_0$, and $\delta T = T - T_0$. As an exception, δc is given by Eq. (5), since there is no c_0 .

The Einstein summation convention is used throughout the paper.

Even in harmonic approximation, the energy density Eq. (7) can give rise to nonlinear effects, since material parameters generally are still functions of the state variables, like temperature, pressure, and polarization P_0 .

The stiffness of order parameter variations is given by c_P . Inhomogeneous deviations of the polarization are described by energy contributions ($\sim K_{ij}^{(2)}$), which are of the standard monoclinic form [24,49]

$$K_{ij} = K_1 \hat{c}_i \hat{c}_j + K_2 \hat{l}_i \hat{l}_j + K_3 \hat{p}_i \hat{p}_j + K_4 (\hat{p}_i \hat{c}_j + \hat{p}_j \hat{c}_i). \quad (8)$$

A surface term $\sim (\delta P)(\nabla_z P)$ has been suppressed. The same structure arises for the spatial variations of the in-plane director, $\sim K_{ij}^{(c)}$ according to Eq. (8).

Of the static contributions associated with layer displacements (gradients of u), the compression (dilation) term $\sim B$ is of the standard form familiar from nonpolar smectic A [3].

The contributions $\sim C_1$ and $\sim C_2$ are specific for polar smectic C_F , since ∇_z is a scalar, but vanish for non-polar smectic C because of parity.

It couples compression/dilation of the layers with bending of the layers. However, in linear order the terms $\sim C_1$ and $\sim C_2$ do not enter the Euler equation for the first variation of ε .

The tensor A_{ijkl} associated with pure bending of the layers assumes the form

$$A_{ijkl} = A \hat{c}_i \hat{l}_j \hat{c}_k \hat{l}_l + \tilde{A} \hat{c}_i \hat{c}_j \hat{l}_k \hat{l}_l + A_{12} \hat{c}_i \hat{c}_j \hat{c}_k \hat{c}_l + A_{21} \hat{l}_i \hat{l}_j \hat{l}_k \hat{l}_l, \quad (9)$$

where we have chosen the notation to facilitate comparison with the usual nonpolar smectic C phase. Keeping in mind that the term $\sim \tilde{A}$ can be obtained from the term $\sim A$ by partial integration up to a surface term, we thus conclude that the contribution $\sim A_{ijkl}$ has the same structure for nonpolar C [3] and the C_F phase.

For the contribution coupling spatial variations of δc to bending of the layers the associated third rank tensor D_{ijk} takes the form

$$D_{ijk} = D_1 \hat{l}_i \hat{l}_j \hat{l}_k + D_2 \hat{p}_i \hat{p}_j \hat{l}_k + D_3 \hat{c}_i \hat{c}_j \hat{l}_k + D_4 (\hat{p}_i \hat{c}_j + \hat{p}_j \hat{c}_i) \hat{l}_k + D_5 (\hat{l}_i \hat{p}_j + \hat{l}_j \hat{p}_i) \hat{p}_k + D_6 (\hat{l}_i \hat{c}_j + \hat{l}_j \hat{c}_i) \hat{p}_k + D_7 (\hat{l}_i \hat{p}_j + \hat{l}_j \hat{p}_i) \hat{c}_k + D_8 (\hat{l}_i \hat{c}_j + \hat{l}_j \hat{c}_i) \hat{c}_k. \quad (10)$$

We note that D_{ijk} must be symmetric in i and j and even under parity keeping in mind that δc is odd under parity.

There are eight independent contributions, among which four describe surface terms. Applying partial integration, the differences between the D_2 and D_5 contributions, between D_3 and D_8 , between D_4 and D_6 , and between D_4 and D_7 give rise to surface terms. These numbers are much larger than for nonpolar smectic C , which is of C_{2h} symmetry and only allows for the D_1 , D_3 , and D_8 contributions, i.e., two bulk and one surface term.

There are nine coupling terms between the polarization and layer displacement (coefficients $d_{1,2,3,31,41,51,32,42,52}$) unknown in nonpolar smectics.

In addition, the energy density of a fluid binary mixture involving only $\delta \sigma$, $\delta \rho$, and $\delta \phi$ is as in the nonpolar case. In addition, one has for the smectic C_F phase, couplings between those variables and δP , $\nabla_z P$, $\nabla_z u$, $\nabla_x^2 u$, and $\nabla_y^2 u$ which gives rise to a total of 18 new coefficients when compared to the nonpolar smectic C phase. These coupling terms are absent in a smectic C phase, since they violate the $\hat{\mathbf{n}} \rightarrow -\hat{\mathbf{n}}$ invariance. The couplings with δP have counterparts in solids that show piezoelectricity [49].

It is well-known that a phase with $\text{div } \hat{\mathbf{p}} = \text{const}$ (splay phase) does have a lower Ginzburg-Landau free energy (compared to the homogeneous state) but necessarily involves defects that increase the energy. The stability of such a splay phase depends, e.g., on boundary conditions and will not be considered here. For a hydrodynamic treatment of splay phases, see Ref. [27]. Since we are dealing with a stable homogeneous equilibrium state here, the linear surface term, $\sim \text{div } \hat{\mathbf{p}}$, can be neglected.

According to the Gibbs relation, Eq. (6), the conjugate quantities to the hydrodynamic and macroscopic variables follow from the free energy as variational or partial derivatives with respect to the appropriate variable, while all the other variables are kept constant. We provide a list of the conjugates in Appendix. Naturally, the harmonic approximation is a restriction in the sense that only sufficiently small deviations from the spatially homogeneous ground state are contained. Big changes such as, for example, a complete director re-orientation as in the Frederiks transition [3], require a fully nonlinear analysis of all the variables involved.

Nonlinear generalizations of Eq. (7) contains the replacements $P_0 \rightarrow P$ and, particularly, in the material tensors, $\hat{p}_i^0 \rightarrow \hat{p}_i$ and $\hat{c}_i^0 \rightarrow \hat{c}_i$.

Applying a constant external electric field E_i with magnitude E_0 , the orientation of polarization P_i^0 in equilibrium will be parallel to the external field due to the electric polarization coupling energy, $\varepsilon_E = -\mathbf{P} \cdot \mathbf{E}$. Thus, if one chooses the external field to be parallel to the (oblique) polarization of the C_F phase, the equilibrium structure is unchanged by the field. Only deviations from equilibrium depend on the field. We will assume that the external field is too weak to change the internal structure, in particular, the tilt angle α of the oblique polarization with respect to the layer normal, is constant. We note that E_i has been split in z and x directions, just as P_i .

Since the external field fixes the orientation of $\hat{c}_i$, deviations from the preferred direction cost an energetic penalty for not being in equilibrium $\sim (1/2) \sin^2(\alpha) P_0 E_0 (\delta \hat{c}_i)^2$. This latter expression follows from $\hat{c}_i \delta \hat{c}_i = 0$ and $\hat{c}_i^0 \delta \hat{c}_i = -\frac{1}{2} (\delta \hat{c}_i)^2$. The tilt angle dependence reflects the different orientation of the polarization and the field relative to $\hat{c}_i^0$.

In terms of the hydrodynamic degree of freedom, δc , Eq. (5), this results in a free-energy contribution

$$\varepsilon_{Ec} = \frac{1}{2} \sin^2(\alpha) P_0 E_0 (\delta c)^2. \quad (11)$$

Obviously, δc is no Goldstone mode anymore and the Gibbs relation, Eq. (6), acquires an additional contribution $h_E^c d\delta c$ with $h_E^c \equiv \partial \varepsilon_{Ec} / \partial \delta c$.

The polar orientation energy can be related to local layer rotations when applied to $\hat{p}_i$, and is written, similarly to the case of $\hat{c}_i$, as $(1/2) \cos^2(\alpha) P_0 E_0 (\delta \hat{p}_i)^2$. Thus, the electric part of the orientational energy can be written

$$\varepsilon_{Eu} = \frac{1}{2} \cos^2(\alpha) P_0 E_0 (\nabla_\perp u)^2. \quad (12)$$

As a result, the conjugate h_i^u in the Gibbs relation Eq. (6), that only allows for $\nabla_z u$ contributions in the field-free case, acquires transverse gradients describing layer rotations with respect to the field.

In addition to the electric polarization coupling energy, which leads to effects linear in the field, there are dielectric anisotropy effects that are quadratic in the field. The dielectric energy, $\sim \chi_{ij} E_i E_j$, with χ_{ij} of the form of Eq. (8), contains four contributions for monoclinic symmetry and its anisotropic part can be written as

$$\varepsilon_{da} = \frac{1}{2} E_0^2 [\chi_{a1} (\nabla_x^2 u)^2 + \chi_{a3} (\nabla_z^2 u)^2 + 2\chi_{a4} (\nabla_z \nabla_x u)^2] \quad (13)$$

with the anisotropic dielectric coefficients $\chi_{a1} = \chi_1 - \chi$, $\chi_{a3} = \chi_3 - \chi$, and $\chi = \chi_2$ is taken as the isotropic coefficient.

It is the linear field dependence on P_0 that can lead to a linear E dependence of certain material parameters. This is in contrast to ordinary nematics and smectics, where the material parameters can only be a function of E^2 . While for small external fields the electric fields the coupling energy to the polarization will dominate, for high enough fields the dielectric energy will become important.

The electric field contributions, Eqs. (11)–(13), are incorporated into the list of conjugate, h_i^u , Φ_{ij}^u , and h_E^c , in the Appendix.

The external field can also change the absolute value of the polarization, but this is a rather unimportant effect and we use P_0 for the cases with or without the external field.

An applied constant magnetic field (taken along the layer normal for simplicity) leads to an energy contribution $\frac{1}{2} H^2 [\chi_{a1}^m (\nabla_x^2 u)^2 + \chi_{a2}^m (\nabla_y^2 u)^2]$ involving the diamagnetic anisotropies χ_a^m . It is quadratic in the field strength, since H_i is odd under time reversal.

IV. DYNAMICS

Following the procedure outlined in the first paragraph of Sec. II, we now turn to the derivation of the dynamic equations for the macroscopic variables and to the determination of the reversible (nondissipative) and irreversible parts of the currents and quasicurrents.

In the following, we will disregard electrodynamic effects of the electric field, assuming $\nabla \times \mathbf{E} = 0$. The dynamic equations have the form

$$\dot{\sigma} + \nabla_i (\sigma v_i + j_i^{\sigma R} + j_i^{\sigma D}) = \frac{2R}{T}, \quad (14)$$

$$\dot{\rho} + \nabla_i (\rho v_i) = 0, \quad (15)$$

$$\dot{g}_i + \nabla_j (g_i v_j + \psi \delta_{ij} - \hat{p}_i h_j^u + \sigma_{ij}^{\text{th}} + \sigma_{ij}^R + \sigma_{ij}^D) = 0, \quad (16)$$

$$\dot{\phi} + v_j \nabla_j \phi + \nabla_i (j_i^{\phi R} + j_i^{\phi D}) = 0, \quad (17)$$

$$\dot{u} + v_j \nabla_j u - v_z + X^{uR} + X^{uD} = 0, \quad (18)$$

$$\dot{c} + v_j \nabla_j c + X^{cR} + X^{cD} = 0, \quad (19)$$

$$\dot{P} + v_i \nabla_i P + X^{PR} + X^{PD} = 0, \quad (20)$$

$$\dot{\rho}_e + \nabla_j (\rho_e v_j) + \nabla_i (j_i^{eR} + j_i^{eD}) = 0. \quad (21)$$

The conserved quantities and the entropy density contain phenomenological currents ($\sim j_i$), while the quasicurrents ($\sim X$) are associated with spontaneously broken continuous symmetry variables or macroscopic variables. The superscripts D and R on the currents denote, respectively, the dissipative and reversible parts.

The energy conservation law

$$\dot{\varepsilon} + \nabla_i (\varepsilon + \psi) v_i + \nabla_i (j_i^{\varepsilon R} + j_i^{\varepsilon D}) = 0 \quad (22)$$

is redundant, since it follows from Eqs. (14)–(21) due to the Gibbs relation (6).

We use the pressure ψ including the isotropic part of the Maxwell stress

$$\psi = \frac{\partial (\int \varepsilon dV)}{\partial V} = -\varepsilon + \mu \rho + T \sigma + \mathbf{v} \cdot \mathbf{g} + D_i E_i \quad (23)$$

and the off-diagonal terms of the Maxwell and the Ericksen-type stresses [50]:

$$\begin{aligned} 2\sigma_{ij}^{\text{th}} = & -(E_i D_j + D_i E_j) + \Phi_j^P \nabla_i P + \Phi_i^P \nabla_j P \\ & + \Phi_{ki}^u \nabla_k \nabla_j u + \Phi_{kj}^u \nabla_k \nabla_i u \\ & + \Phi_{ki}^c \nabla_k \nabla_j c + \Phi_{kj}^c \nabla_k \nabla_i c. \end{aligned} \quad (24)$$

The Maxwell stress is of the standard form [30,51] with $D_i = E_i + P_i$ and has been symmetrized with the help of the

requirement that the energy density should be invariant under rigid rotations [25]. In detail, one first obtains directly from the condition of zero entropy production in Eq. (28)

$$\sigma_{ij}^{\text{th}} = -D_j E_i + \Phi_j^P \nabla_i P + \Phi_{kj}^u \nabla_k \nabla_i u + \Phi_{kj}^c \nabla_k \nabla_i c \quad (25)$$

and uses the requirement of rotational invariance of the Gibbs relation [24]. Compare also Ref. [24] for a detailed exposition.

With a redefinition of the pressure, $\psi = \tilde{\psi} + (1/2)E^2$, another useful form of the momentum conservation Eq. (16) can be found,

$$\begin{aligned} \dot{g}_i + \nabla_j (g_i v_j + \tilde{\psi} \delta_{ij} - \hat{p}_i h_j^u + \Phi_j^P \nabla_i P + \sigma_{ij}^R + \sigma_{ij}^D) \\ = \rho_e E_i + P_j \nabla_j E_i, \end{aligned} \quad (26)$$

with the Coulomb and Kelvin external forces as source terms. Using the Gibbs relation Eq. (6), the pressure gradient can be written as

$$\nabla_i \tilde{\psi} = \rho \nabla_i \mu + \sigma \nabla_i T - \Pi \nabla_i \phi + P_j \nabla_j E_i. \quad (27)$$

The source term of Eq. (14) contains R , the dissipation function, which represents the energy dissipation of the system. Due to the second law of thermodynamics, R must satisfy $R \geq 0$: For reversible processes, this dissipation function is equal to zero, while for irreversible processes it must be positive,

$$\begin{aligned} 2R = -j_i^{\sigma*} \nabla_i T - j_i^{\phi*} \nabla_i \Pi - \sigma_{ij}^* \nabla_j v_i - j_i^{e*} E_i \\ - X^{u*} \nabla_i h_i^u - X^{c*} (\nabla_i h_i^c - h_E^c) + X^{P*} h^P \geq 0, \end{aligned} \quad (28)$$

where positive entropy production ($R > 0$) applies for $*$ = D and vanishing entropy production ($R = 0$) for $*$ = R. The superscripts D refer to the dissipative part of the currents and quasicurrents in Eqs. (14)–(21) while the superscripts R refer to the reversible parts in Eqs. (14)–(21).

The phenomenological currents and quasicurrents are the sum of the reversible and the dissipative part, as can be seen in Eqs. (14)–(21). The various transport contributions in Eqs. (14)–(21) (as well as ψ and σ_{ij}^{th}) are reversible and add up to zero in the entropy production. We include E_i as a thermodynamic force, describing electric fluctuations that are assumed to be small enough not to change the static structure.

These phenomenological currents and quasicurrents are treated in the following subsections within linear irreversible thermodynamics (guaranteeing general Onsager relations), i.e., as linear relations between currents and thermodynamic forces. The resulting expressions are nevertheless nonlinear, since all material parameters can be functions of the scalar state variables (e.g., σ , ρ , P , ϕ).

The form of Eq. (17) reflects the assumption that the impurity density is conserved, while Eq. (21) describes the polar order parameter modulus as a slowly relaxing quantity (similar to, e.g., the nematic order parameter modulus [52] or the superfluid degree of order [38,53]).

V. REVERSIBLE CURRENTS

To obtain the reversible currents, one expands all currents and quasicurrents systematically into the thermodynamic forces and conjugates, taking into account the behavior under time reversal, spatial inversion, rigid rotations and, most

importantly, zero entropy production. For a more detailed exposition of the method, we refer to Ref. [25].

For the reversible dynamic behavior of our macroscopic system, we obtain the following expressions for the reversible currents containing phenomenological parameters:

$$j_i^{\sigma R} = \varphi_{ijk}^{\sigma} A_{jk}, \quad (29)$$

$$j_i^{eR} = \varphi_{ijk}^e A_{jk}, \quad (30)$$

$$\begin{aligned} \sigma_{ij}^R = \lambda_{ij}^P h^P + \xi_{ij} (h_E^c - \nabla_l h_l^c) \\ - \varphi_{kji}^{\sigma} \nabla_k T - \varphi_{kji}^{\phi} \nabla_k \Pi - \varphi_{kji}^e E_k, \end{aligned} \quad (31)$$

$$X^{PR} = \lambda_{ij}^P A_{ij}, \quad (32)$$

$$X^{uR} = 0, \quad (33)$$

$$X^{cR} = \xi_{ij} \nabla_i v_j, \quad (34)$$

$$j_i^{\phi R} = \varphi_{ijk}^{\phi} A_{jk}, \quad (35)$$

with the symmetrized velocity gradient A_{ij} , where $2A_{ij} = \nabla_i v_j + \nabla_j v_i$. There is a coupling of the modulus of the polarization and the density of linear momentum provided by the symmetric tensor λ_{ij}^P , which is of the standard monoclinic form, as given in Eq. (8) reflecting the biaxial nature of C_F . The experimental implications of this coupling are discussed in Sec. VII B. The coupling terms $\sim \xi_{ij}$ describe a cross coupling between velocity gradients and the molecular field associated with the in-plane director, δc . It contains an antisymmetric contribution from the coupling to rotations and a symmetric part describing the flow alignment terms. Explicitly, we have

$$\begin{aligned} \xi_{ij} = \frac{1}{2} (\hat{l}_i \hat{c}_j - \hat{l}_j \hat{c}_i) + \xi^{(1)} \frac{1}{2} (\hat{l}_i \hat{c}_j + \hat{l}_j \hat{c}_i) \\ + \xi^{(2)} \frac{1}{2} (\hat{l}_i \hat{p}_j + \hat{l}_j \hat{p}_i). \end{aligned} \quad (36)$$

It is easily checked that the flow alignment terms and the term associated with rigid rotation are isomorphic in nonpolar smectic C and ferroelectric smectic C , C_F . Therefore, flow alignment should also be observable in freely suspended films of smectic C_F , just in analogy to predictions made [54] and experimental observations [55] for usual nonpolar smectic C phases.

A third class of reversible couplings are provided by the third-rank material tensors φ_{ijk}^{α} for $\alpha \in \{\sigma, e, \phi\}$ with the structure

$$\begin{aligned} \varphi_{ijk}^{\alpha} = \varphi_1^{\alpha} \hat{c}_i \hat{c}_j \hat{c}_k + \varphi_2^{\alpha} \hat{p}_i \hat{p}_j \hat{p}_k + \varphi_3^{\alpha} \hat{p}_i \hat{c}_j \hat{c}_k \\ + \varphi_4^{\alpha} \hat{c}_i \hat{p}_j \hat{p}_k + \varphi_5^{\alpha} (\hat{p}_i \hat{p}_j \hat{c}_k + \hat{p}_i \hat{p}_k \hat{c}_j) \\ + \varphi_6^{\alpha} (\hat{c}_i \hat{c}_j \hat{p}_k + \hat{c}_i \hat{c}_k \hat{p}_j) \\ + \varphi_7^{\alpha} \hat{c}_i \hat{l}_j \hat{l}_k + \varphi_8^{\alpha} (\hat{c}_j \hat{l}_i \hat{l}_k + \hat{c}_k \hat{l}_i \hat{l}_j) \\ + \varphi_9^{\alpha} \hat{p}_i \hat{l}_j \hat{l}_k + \varphi_{10}^{\alpha} (\hat{p}_j \hat{l}_i \hat{l}_k + \hat{p}_k \hat{l}_i \hat{l}_j). \end{aligned} \quad (37)$$

They connect currents of scalar conserved quantities with A_{ij} and, reciprocally, their conjugate forces with the stress tensor σ_{ij} . Such couplings require the presence of at least one parity breaking vector. We refer to Ref. [28], where the possibility of this type of reversible cross coupling has been elucidated

for polar nematics. In the present case of C_F , we have two, $\hat{c}_i$ and $\hat{p}_i$. All parts of the tensors φ_{ijk}^α therefore contain an odd number of $\hat{p}_i$ or $\hat{c}_i$ in order to restore parity symmetry. These reversible dynamic cross-coupling terms are absent in nonpolar systems such as nonpolar smectic A and nonpolar smectic C . We note that the structure of the tensors $\sim \varphi_{ijk}^\alpha$ has not been elucidated before for systems with monoclinic symmetry, to the best of our knowledge. It appears worthwhile to mention that because of the broken parity symmetry associated with these contributions, their symmetry under parity is the same as that of the third-rank piezoelectric tensor. Of course, the behavior under time reversal is the opposite, since we are discussing contributions to reversible currents. For C_{1h} symmetry, we therefore find ten contributions in parallel with the ten piezoelectric coefficients for this symmetry. In Sec. VII B, we discuss consequences of those contributions that can be detected experimentally.

VI. DISSIPATIVE CURRENTS

To describe dissipative processes, it is convenient to expand the dissipation function, R , the source term in the dynamic equation for the entropy density, Eq. (14), into a positive definite expression quadratic in the thermodynamic forces. Then taking variational derivatives (or partial derivatives when applicable) of R with respect to forces, one obtains linear relations (listed below) between the currents and the quasi-currents on the one hand and thermodynamic forces on the other. The entropy production is a scalar under all transformations compatible with symmetry including time reversal, spatial parity, and rigid rotations. For a detailed exposition of the method we refer to Ref. [25]. The dissipative dynamic behavior of our macroscopic system is characterized by the dissipation function R

$$\begin{aligned} 2R = & \kappa_{ij}(\nabla_i T)(\nabla_j T) + D_{ij}(\nabla_i \Pi)(\nabla_j \Pi) \\ & + 2D_{ij}^{T\Pi}(\nabla_i T)(\nabla_j \Pi) + \sigma_{ij}^E E_i E_j \\ & + 2D_{ij}^{TE} E_j \nabla_i T + 2D_{ij}^{\Pi E} E_j \nabla_i \Pi \\ & + \frac{1}{\gamma_u} (\nabla_i h_i^u)(\nabla_j h_j^u) + v_{ijkl}(\nabla_j v_i)(\nabla_l v_k) \\ & + 2D_i^{PE} E_i h^P + 2D_i^{\Pi\Pi}(\nabla_i \Pi)(\nabla_j h_j^u) \\ & + 2D_i^{PT} h^P \nabla_i T + 2D_i^{P\Pi} h^P \nabla_i \Pi + \Gamma h^P \nabla_i h_i^u \\ & + 2D_i^{uT}(\nabla_i T)(\nabla_j h_j^u) + 2D_i^{u\Pi} E_i \nabla_j h_j^u \\ & + \frac{1}{\gamma_c} (\nabla_i h_i^c - h_E^c)(\nabla_j h_j^c - h_E^c) + b_{||} h^P h^P. \end{aligned} \quad (38)$$

The symmetric second rank tensors κ_{ij} , $D_{ij}^{T\Pi}$, D_{ij} , σ_{ij}^E , D_{ij}^{TE} , and $D_{ij}^{\Pi E}$ are of the standard form for tensors of monoclinic symmetry [25,49], Eq. (8).

We note that also v_{ijkl} is of the standard form for a fourth-rank tensor of monoclinic symmetry [24,25,49] and has 13 coefficients. The contribution $\sim b_{||}$ in the entropy production describes the relaxation of the polarization modulus P . This term has an analog in ordinary nematics (with the order parameter modulus included). Specific for polar systems including polar nematics, polar nematics with a solvent, polar

smectic A_F as well as the polar smectic C_F phase, studied here are the dissipative crosscouplings between polarization on the one hand and gradients of temperature, gradients of osmotic pressure and electric fields on the other governed by the first rank material tensors D_i^{PT} , $D_i^{P\Pi}$, and D_i^{PE} , which take for monoclinic symmetry the form [49]

$$D_i^{PT} = D_1^{PT} \hat{c}_i + D_2^{PT} \hat{p}_i. \quad (39)$$

These contributions can only exist in polar systems of sufficiently low symmetry, since they are odd in $\hat{c}_i$ and $\hat{p}_i$. The same reasoning applies to the first rank material tensors D_i^{uT} , $D_i^{u\Pi}$, and D_i^{uE} . Finally, we emphasize that Eq. (38) contains all the terms of a nonpolar usual smectic C phase as a special case [24].

To obtain the dissipative parts of the currents and quasicharacteristics, we take the partial derivatives of R with respect to the appropriate thermodynamic force:

$$\begin{aligned} j_i^{\sigma D} = - \frac{\partial R}{\partial (\nabla_i T)} \Big|_{\dots} = & -\kappa_{ij} \nabla_j T - D_{ij}^{T\Pi} \nabla_j \Pi \\ & - D_{ij}^{TE} E_j - D_i^{TP} h^P - D_i^{uT} (\nabla_j h_j^u), \end{aligned} \quad (40)$$

$$\begin{aligned} j_i^{eD} = - \frac{\partial R}{\partial E_i} \Big|_{\dots} = & -\sigma_{ij}^E E_j - D_{ij}^{\Pi E} \nabla_j \Pi \\ & - D_{ij}^{TE} \nabla_j T - D_i^{PE} h^P - D_i^{uE} (\nabla_j h_j^u), \end{aligned} \quad (41)$$

$$\begin{aligned} j_i^{\phi D} = - \frac{\partial R}{\partial (\nabla_j \Pi)} \Big|_{\dots} = & -D_{ij} \nabla_j \Pi - D_{ij}^{T\Pi} \nabla_j T \\ & - D_{ij}^{\Pi E} E_j - D_i^{P\Pi} h^P - D_i^{u\Pi} (\nabla_j h_j^u), \end{aligned} \quad (42)$$

$$\sigma_{ij}^D = - \frac{\partial R}{\partial (\nabla_j v_i)} \Big|_{\dots} = -v_{ijkl} A_{kl}, \quad (43)$$

$$\begin{aligned} X^{PD} = \frac{\partial R}{\partial h^P} \Big|_{\dots} = & b_{||} h^P + D_i^{P\Pi} \nabla_i \Pi + \Gamma \nabla_i h_i^u \\ & + D_i^{PE} E_i + D_i^{PT} \nabla_i T, \end{aligned} \quad (44)$$

$$\begin{aligned} X^{uD} = - \frac{\partial R}{\partial (\nabla_i h_i^u)} \Big|_{\dots} = & -\frac{1}{\gamma_u} \nabla_i h_i^u - D_i^{u\Pi} \nabla_i \Pi - \Gamma h^P \\ & - D_i^{uE} E_i - D_i^{uT} \nabla_i T, \end{aligned} \quad (45)$$

$$X^{cD} = \frac{\partial R}{\partial (h_E^c - \nabla_i h_i^c)} \Big|_{\dots} = \frac{1}{\gamma_c} (h_E^c - \nabla_i h_i^c). \quad (46)$$

VII. POSSIBLE EXPERIMENTS

A. On the two static coupling terms between compression and deformations of the layering

As noted briefly in Sec. III, there are two rather interesting static cross-coupling term, when compared to usual smectic C , namely, the contributions

$$(\nabla_z u)(C_1 \nabla_x^2 u + C_2 \nabla_y^2 u) \quad (47)$$

in Eq. (7), which couple layer compressions to a bending of the layers in a biaxial fashion, where we make use of the coordinate system indicated in Fig. 2.

From Eqs. (A3) and (A4), it is easy to see that these contributions do not generate bulk effects in the dissipative currents, Eqs. (40)–(46), since their first variational derivative, $\nabla_i h_i^u = \nabla_i (h_i^u - \nabla_j \Phi_{ij}^u)$ vanishes identically. On the other hand, the contributions $\sim C_1$ and $\sim C_2$ are not pure surface terms, in contrast, e.g., to $\nabla_i P_i$ in the energy. We are aware of only one other cross-coupling term in the hydrostatic regime for which such a situation arises, namely, for ferroelectric smectic A_F phases [44]. Indeed, the terms given in Eq. (47) can be interpreted as a biaxial generalization of the contribution $C(\nabla_z u)(\nabla_z^2 u)$ given in Eq. (3) of Ref. [44].

The only instance where h_i^u appears (and not its divergence) is the stress tensor σ_{ij} in Eq. (16), where $\hat{p}_i h_j^u$ represents the elastic tensions. It consists of two parts,

$$\sigma_{ij}^C = \hat{p}_i \hat{p}_j (C_1 \nabla_x^2 u + C_2 \nabla_y^2 u) - \hat{p}_i (\hat{c}_j C_1 \nabla_x \nabla_z u + \hat{l}_j C_2 \nabla_y \nabla_z u), \quad (48)$$

describing a relative compression—shear tension weighted with $\nabla_x^2 u$ and $\nabla_y^2 u$ (for the part of compressional tensions) and $\nabla_x \nabla_z u$ and $\nabla_y \nabla_z u$ (for the contribution from shear tensions). It seems nontrivial, however, to experimentally detect these various effects.

Thus, so far, one has found the effects described for two fluid and ferroelectric smectic phases, namely, the uniaxial A_F phase depicted in Fig. 1 and the biaxial C_F phase. Essential for the existence for this type of contribution is a polar direction and a layering. We emphasize that for ferromagnetic smectics there is no analog because of the different behaviors under parity and time reversal of the magnetization M_i .

B. Reversible dynamic coupling of flow with the magnitude of the polarization P

Here we investigate the influence of biaxiality on sound propagation in a tilted ferroelectric fluid smectic phase, smectic C_F . In simple fluids, ordinary sound is known as the only propagating low- q excitation, $\exp i(q_i r_i - \omega t)$, with $\omega^2 \sim q^2$. In a smectic A liquid crystal, ferromagnetic or paramagnetic, there is an additional propagating mode, sometimes called second sound. It is due to the spontaneously broken translational symmetry along the normal of the layers. This one-dimensional compression (dilation) mode, with susceptibility B , Eq. (7), is anisotropic and leads to an anisotropic part in the first sound spectrum. In the tilted ferroelectric smectic phase, C_F the magnitude of the macroscopic polarization δP provides additionally a low- q coupling with gradients of the velocity field via λ_{ij}^P in Eqs. (31) and (32). As a result, sound-like excitations become more complex. We do not consider an external electric field and neglect the concentration degree of freedom.

Linearizing the dynamic equations given in Sec. IV, we find to lowest order in the gradients and discarding dissipation

$$\dot{\rho} + \rho_0 \nabla_i v_i = 0, \quad (49)$$

$$\dot{\sigma} + \sigma_0 \nabla_i v_i = 0, \quad (50)$$

$$\dot{g}_i + \nabla_j (\delta_{ij} \psi - \hat{p}_i h_j^u + \lambda_{ij}^P h^P) = 0, \quad (51)$$

$$\dot{u} - v_z = 0, \quad (52)$$

$$\dot{P} + \lambda_{ij}^P A_{ij} = 0, \quad (53)$$

where we have in linearized form $h_j^u = \hat{p}_j (B \nabla_z u + d_1 \delta P)$, $h^P = c_P \delta P + d_1 \nabla_z u$, and $\nabla_i \psi = \rho_0 \nabla_i \mu + \sigma_0 \nabla_i T$, with $\nabla_i \mu$ and $\nabla_i T$ related to the variables $\delta \sigma$, $\delta \rho$, and δP by Eqs. (A6) and (A7).

Thus, we have seven equations for the seven variables $\delta \rho$, $\delta \sigma$, g_i , δP , and δu . As a consequence, we obtain as a solvability condition after a Fourier transform in time and space with wave vector q_i and frequency ω a real cubic polynomial for the squares of the eigenfrequencies ω_n^2 where $n = 1, 2$, and 3. Because the resulting cubic polynomial has a quite complicated appearance, we refrain from giving it explicitly, while the sum of the three ω_n^2 is quite simple and reflects the monoclinic symmetry of the smectic C_F phase. Suppressing the superscript “P” at the tensor λ_{ij}^P in the displayed formulas throughout this subsection for simplicity, we find

$$\begin{aligned} \omega_1^2 + \omega_2^2 + \omega_3^2 &= c_{10}^2 q^2 + \frac{1}{\rho_0} B q_z^2 + \frac{2d_1}{\rho_0} (\lambda_3 q_z^2 + \lambda_4 q_x q_z) \\ &+ \frac{c_P}{\rho_0} (\lambda_2^2 q_y^2 + [\lambda_3 q_z + \lambda_4 q_x]^2 + [\lambda_1 q_x + \lambda_4 q_z]^2) \\ &+ \bar{\gamma} (\lambda_1 q_x^2 + \lambda_2 q_y^2 + \lambda_3 q_z^2 + 2\lambda_4 q_x q_z), \end{aligned} \quad (54)$$

with $\bar{\gamma} = \gamma_1 + (\sigma_0/\rho_0)\gamma_2$. For the first sound velocity of simple fluids, we have in the present notation $\rho_0 c_{10}^2 = c_{\rho\rho}^2 \rho_0^2 + 2c_{\rho\sigma}^2 \rho_0 \sigma_0 + c_{\sigma\sigma}^2 \sigma_0^2$. Equation (54) contains the results well known for smectic A [24] as a special case, if all reactive terms associated with the magnitude of the polarization, λ_n^P ($n \in \{1, 2, 3, 4\}$), are discarded.

It is important to emphasize that one can extract from Eq. (54) a corresponding result for the ferroelectric smectic A_F phase shown in Fig. 1, since the static contributions $\sim \gamma_1$, γ_2 and d_1 also exist already in the uniaxial A_F phase. Specializing Eq. (54) to smectic A_F , we have

$$\begin{aligned} \omega_1^2 + \omega_2^2 + \omega_3^2 &= c_{10}^2 q^2 + \frac{1}{\rho_0} B q_z^2 + \frac{2d_1}{\rho_0} \lambda_3 q_z^2 \\ &+ \frac{c_P}{\rho_0} (2\lambda_1^2 q_\perp^2 + \lambda_3^2 q_z^2) + \bar{\gamma} (2\lambda_1 q_\perp^2 + \lambda_3 q_z^2). \end{aligned} \quad (55)$$

From inspection of Eq. (55), we see immediately two main differences to the case of smectic C_F : the interference terms $\sim \lambda_4^P$ have disappeared and the results reflect the uniaxiality of smectic A_F . We observe that there is no change regarding the contributions $\sim c_{10}^2$ and $\sim B$ for the unperturbed velocities of first and second sound, respectively. Finally, we note that one must make the identifications $\lambda_1 \equiv \lambda_1^{A_F}$, $\lambda_3 \equiv \lambda_3^{A_F}$ and where $q_\perp$ is the wave vector perpendicular to the layer normal $\hat{p}_i$ when comparing with Ref. [23].

Although Eq. (54) does not show the results for the individual sound velocities in smectic C_F , a number of quite general conclusions can be drawn. Most importantly, we conclude that the monoclinic structure of λ_{ij}^P is reflected in all three pairs of sound modes via the interference terms $\sim \lambda_4^P q_x q_z$. These are absent in uniaxial systems like classical smectic A phases or the uniaxial ferroelectric smectic A_F phases. Thus, probing sound waves in a monodomain sample of smectic C_F will give

rise to a rather intricate angular dependence on the direction of a general wave vector q_i .

To bring this out more clearly, it is convenient to introduce spherical polar coordinates for q_i making use of the directions of $\hat{c}_i$, $\hat{l}_i$ and $\hat{p}_i$ via $q_z = q \cos(\Theta)$, $q_x = q \sin(\Theta) \cos(\varphi)$ and $q_y = q \sin(\Theta) \sin(\varphi)$. In the most general case with q_i oblique to all preferred directions, these substitutions lead, on the right hand side of Eq. (54), to the anisotropic contributions

$$\begin{aligned} & \left(\frac{B}{\rho_0} + \frac{c_P}{\rho_0} [\lambda_3^2 + \lambda_4^2] + \left[\bar{\gamma} + \frac{2d_1}{\rho_0} \right] \lambda_3 \right) q^2 \cos^2(\Theta) \\ & + \left(\frac{c_P}{\rho_0} \lambda_2^2 + \bar{\gamma} \lambda_2 \right) q^2 \sin^2(\Theta) \sin^2(\varphi) \\ & + \left(\frac{c_P}{\rho_0} [\lambda_1^2 + \lambda_4^2] + \bar{\gamma} \lambda_1 \right) q^2 \sin^2(\Theta) \cos^2(\varphi) \\ & + \left(\frac{c_P}{\rho_0} [\lambda_1 \lambda_4 + \lambda_3 \lambda_4] + \left[\bar{\gamma} + \frac{d_1}{\rho_0} \right] \lambda_4 \right) q^2 \sin(2\Theta) \cos(\varphi). \end{aligned}$$

From the last line of Eq. (54), we see immediately the influence of the monoclinic symmetry for the smectic C_F phase studied here. We also note that for a vanishing value of λ_4^P the result applies to an orthorhombic biaxial phase, and, if additionally $\lambda_1 = \lambda_2$, to an uniaxial phase.

It thus emerges that the presence of a relaxing variable such as the magnitude of the macroscopic polarization for ferroelectric smectic C_F , influences all soundlike excitations to lowest order in the wave vector. It is expected that the influence of the relaxation of δP is strongest, and therefore most easily measurable, near a second order phase transition from smectic C_F to a phase of higher symmetry such as smectic A_F . This is based on the fact that in the vicinity of such a phase transition the lifetime of order parameter fluctuations is growing.

C. Reversible shear and elongational flows

In this subsection, we discuss some of the implications of the reversible cross-coupling terms described by the tensors φ_{ijk}^α , Eq. (37), between velocity gradients and gradients of temperature ($\alpha = \sigma$) and concentration ($\alpha = \phi$), or electric fields ($\alpha = e$). Since the consequences of these cross-couplings has never been elucidated before for a biaxial system like smectic C_F , we will be quite detailed in our description. Using the notation introduced in Fig. 2, we take $\hat{p}_i$ parallel to the z direction with the perpendicular layering in the $x - y$ planes resembling a free-standing film geometry. In addition, we take $\hat{c}_i$ to be parallel to the x direction.

Using this notation, we obtain explicitly for the structure given in Eq. (37) for the case of the three components of the entropy current density, j_i^σ [Eq. (29)]:

$$\begin{aligned} j_x^\sigma &= \varphi_1^\sigma A_{xx} + \varphi_4^\sigma A_{zz} + 2\varphi_6^\sigma A_{xz} + \varphi_7^\sigma A_{yy}, \\ j_y^\sigma &= 2\varphi_8^\sigma A_{xy} + 2\varphi_{10}^\sigma A_{zy}, \\ j_z^\sigma &= \varphi_2^\sigma A_{zz} + \varphi_3^\sigma A_{xx} + 2\varphi_5^\sigma A_{xz} + \varphi_9^\sigma A_{yy}. \end{aligned} \quad (56)$$

As it is easily checked the uniaxial case, which arises for polar and ferroelectric nematics as well as for ferroelectric smectic A_F , is contained in the Eq. (56) given above as a special case.

From Eq. (56), a number of possible results for applied shear or extensional flows emerges. Applying a shear flow in the $y - z$ plane, meaning in the plane containing the layer normal and the in-plane direction perpendicular to the in-plane preferred direction, $\hat{c}_i$, one obtains a heat current only in the y direction,

$$j_y^{\sigma,R} = 2\varphi_{10}^\sigma A_{yz}, \quad (57)$$

a result found previously for the uniaxial smectic A_F phase [44]. However, to obtain a heat current only in the y direction, one can also a shear flow in the layer planes via A_{xy} :

$$j_y^{\sigma,R} = 2\varphi_8^\sigma A_{xy}. \quad (58)$$

This result emerges because we have in smectic C_F a polar biaxial phase, two polar preferred directions. The third case for applying simple shear is via A_{xz} , that is, in the $x - z$ plane, resulting in two components of the heat current, namely, in x as well as in the z direction, another result specific for a system with two polar preferred directions,

$$\begin{aligned} j_x^\sigma &= 2\varphi_6^\sigma A_{xz}, \\ j_z^\sigma &= 2\varphi_5^\sigma A_{xz}, \end{aligned} \quad (59)$$

with vanishing j_y^σ .

Other examples are uniaxial elongational or compressional flow in one of the two in-plane directions. Starting with A_{yy} , we obtain

$$\begin{aligned} j_x^\sigma &= \varphi_7^\sigma A_{yy}, \\ j_z^\sigma &= \varphi_9^\sigma A_{yy}, \end{aligned} \quad (60)$$

with vanishing j_y^σ . We note that the result for j_z^σ has also been obtained before for the uniaxial case. Applying a uniaxial elongational or compressional flow in x - direction, A_{xx} , that is parallel to $\hat{c}_i$, results in

$$\begin{aligned} j_x^\sigma &= \varphi_1^\sigma A_{xx}, \\ j_z^\sigma &= \varphi_2^\sigma A_{xx}, \end{aligned} \quad (61)$$

with vanishing j_y^σ . We note that the result for j_z^σ is also known from the uniaxial A_F phase, while the result for j_x^σ is again specific for a system with two polar directions.

In principle, also another uniaxial elongational flow (along the layer normal $\hat{p}_i$) results in a heat current with two components, namely, $j_x^{\sigma,R} = \varphi_4^\sigma A_{zz}$ and $j_z^{\sigma,R} = \varphi_2^\sigma A_{zz}$. However, the flow $\sim A_{zz}$ is incompatible with a constant layer spacing.

Analogously, we obtain concentration and electric currents of the same form (replacing in the φ tensor superscript σ by ϕ and e , respectively).

Reciprocally, one can apply a temperature gradient, a concentration gradient or an electrical field to a sample and then obtain nonvanishing elements of the stress tensor.

For a temperature gradient parallel to x , $\nabla_x T = G_x$, we find

$$\begin{aligned} \sigma_{xx} &= \varphi_1^\sigma G_x; & \sigma_{yy} &= \varphi_7^\sigma G_x; \\ \sigma_{zz} &= \varphi_4^\sigma G_x; & \sigma_{xz} &= \sigma_{zx} = \varphi_6^\sigma G_x; \\ \sigma_{xy} &= \sigma_{yx} = \sigma_{zy} = \sigma_{yz} \equiv 0. \end{aligned} \quad (62)$$

For a temperature gradient parallel to y , $\nabla_y T = G_y$, we have

$$\begin{aligned}\sigma_{xy} = \sigma_{yx} &= \varphi_8^\sigma G_y, & \sigma_{yz} = \sigma_{zy} &= \varphi_{10}^\sigma G_y, \\ \sigma_{xx} = \sigma_{yy} = \sigma_{zz} &= \sigma_{xz} = \sigma_{zx} \equiv 0.\end{aligned}\quad (63)$$

Finally, for a temperature gradient parallel to z , $\nabla_z T = G_z$, we find

$$\begin{aligned}\sigma_{xx} &= \varphi_3^\sigma G_z; & \sigma_{yy} &= \varphi_9^\sigma G_z; \\ \sigma_{zz} &= \varphi_2^\sigma G_z; & \sigma_{xz} = \sigma_{zx} &= \varphi_5^\sigma G_z; \\ \sigma_{xy} = \sigma_{yx} &= \sigma_{zy} = \sigma_{yz} \equiv 0.\end{aligned}\quad (64)$$

It is easily checked that all ten coefficients are appearing precisely once in Eqs. (62)–(64). Such stresses can lead to flows via the dynamic equation for the density of linear momentum only for spatially inhomogeneous external temperature or concentration gradients or inhomogeneous electric fields.

VIII. SUMMARY AND PERSPECTIVE

In the present paper, we have derived the complete set of macroscopic dynamic equations for ferroelectric smectic C_F liquid crystals with the spontaneous average polarization tilted with respect to the layer normal of the layering.

We followed the procedure used to derive systematically dynamic equations for generalized hydrodynamics [24,25,31,38]. First, we identified the symmetries of the ground state, followed by the evaluation of the macroscopic variables due to conservation laws, variables associated with spontaneously broken continuous symmetries and macroscopic variables. Then we used the assumption of local thermodynamic equilibrium to establish the Gibbs relation containing the variations of the variables and serving to introduce the thermodynamic conjugate quantities. To derive a self-consistent set of equations was then a two-step procedure. First we related the thermodynamic forces to the variables. In a second step the currents and quasicurrents arising in the dynamic equations for the macroscopic variables were related to the thermodynamic forces. Currents and quasicurrents were split into two contributions: reversible quantities that do not give rise to any entropy production and dissipative parts, which lead to entropy generation thus guaranteeing that the second law of thermodynamics is fulfilled.

Compared to usual smectic C phases, we found a number of static and reversible as well as irreversible dynamic cross-coupling terms. As additional macroscopic variables when compared to simple fluids we have the layer displacement u , familiar from smectic liquid crystals, the variations of the in-plane director and the polarization density, $\mathbf{P}$.

Specifically, compared to nonpolar smectic C phases, we found two static cross-coupling terms between compression of the layering and bending of the layers, which does not lead

to elastic forces, but to elastic stresses. In addition, a number of static cross-coupling terms has been elucidated, which can exist in smectic C_F , but not in the ferroelectric A_F phase, because of the polar in-plane preferred direction in smectic C_F .

We found that both the velocity of first and second sound become biaxial reflecting the monoclinic symmetry due to the influence of the modulus of the polarization. We also analyzed the consequences of reversible cross-coupling terms between elongational flow and electric fields as well as temperature and concentration gradients, which lend themselves to experimental detection. Such cross-coupling terms are absent without the presence of a polar direction and thus do not exist in nonpolar smectic C phases. Finally, we also examined the question how the linear $\mathbf{P} \cdot \mathbf{E}$ coupling in the energy alters the macroscopic response behavior when compared to usual nonpolar smectic C phases.

There are several directions in which the results presented here can be generalized. One possibility is to chiralize a smectic C_F phase, thus reducing the symmetry from C_{1h} to C_1 , the lowest symmetry possible, by removing the horizontal mirror plane. This way, one can obtain a phase without symmetry by tilting once, a situation which is different from C_G , which is tilted twice [3,56].

Another direction to go into would be to crosslink a smectic C_F phase, thus producing a ferroelectric liquid crystalline elastomer or gel, thus generalizing previous work on nematic elastomers and gels [57,58]. Such a smectic C_F elastomer or gel would have two polar directions due to the tilt in contrast to a smectic A_F elastomer or gel. Using the concept of Kato's group [59] for generating liquid crystalline physical gels, one could induce a reversible transition from a ferroelectric gel or a soft solid to a fluid ferroelectric smectic C_F phase by changing, for example, temperature or pressure.

It will also be most interesting to generate the antiferroelectric analog of the smectic C_F phase, for which one would have a changing direction of the macroscopic electric polarization (for both, the in-plane component as well as for the component parallel to the layer normal) from one layer to the next. This would then be a complement to the antiferroelectric smectic Z_A phase, which was shown to be orthogonal and biaxial [7–11] and whose macroscopic properties have been studied very recently [23].

Note added in proof. It turns out that gradients of δc can couple to gradients of u , in particular giving rise to the static contribution $\sim (\nabla_z u)(\nabla_y c)$, leading to an interesting connection between rotation and compression in a tilted smectic phase.

DATA AVAILABILITY

No data were created or analyzed in this study.

APPENDIX: EXPLICIT EXPRESSIONS FOR THE THERMODYNAMIC FORCES

In the following, we list the explicit expressions for the thermodynamic forces, including the electric energies Eqs. (11)–(13), into the energy functional Eq. (7), explicitly using the coordinate system shown in Fig. 2 and introduced after Eq. (7):

$$h^P = \frac{\partial \varepsilon}{\partial \delta P} \Big|_{\dots} = c_P \delta P + \gamma_1 \delta \rho + \gamma_2 \delta \sigma + \gamma_3 \delta \phi + \psi_4 \nabla_y c + d_1 \nabla_z u + d_{31} \nabla_x^2 u + d_{32} \nabla_y^2 u, \quad (\text{A1})$$

$$\begin{aligned} \Phi_i^P = \frac{\partial \varepsilon}{\partial (\nabla_i P)} \Big|_{\dots} &= K_{ij}^{(2)} \nabla_j P + \hat{p}_i (d_2 \nabla_z u + d_{41} \nabla_x^2 u + d_{42} \nabla_y^2 u) + \hat{c}_i (d_3 \nabla_z u + d_{51} \nabla_x^2 u + d_{52} \nabla_y^2 u) \\ &+ \hat{p}_i (\theta_1 \delta \rho + \theta_2 \delta \sigma + \theta_3 \delta \phi) + \hat{c}_i (\theta_4 \delta \rho + \theta_5 \delta \sigma + \theta_6 \delta \phi), \end{aligned} \quad (\text{A2})$$

with $h^P = h'^P - \nabla_i \Phi_i^P$,

$$\begin{aligned} h_i^u = \frac{\partial \varepsilon}{\partial \nabla_i u} \Big|_{\dots} &= \hat{p}_i (B \nabla_z u + C_1 \nabla_x^2 u + C_2 \nabla_y^2 u + \hat{\theta}_1 \delta \rho + \hat{\theta}_2 \delta \sigma + \hat{\theta}_3 \delta \phi + d_1 \delta P + d_2 \nabla_z P + d_3 \nabla_x P) \\ &+ \cos^2(\alpha) E_0 P_0 (\delta_{ij} - \hat{p}_i \hat{p}_j) \nabla_j u, \end{aligned} \quad (\text{A3})$$

$$\begin{aligned} \Phi_{ij}^u = \frac{\partial \varepsilon}{\partial \nabla_i \nabla_j u} \Big|_{\dots} &= D_{ijk} \nabla_k c + A_{ijkl} \nabla_k \nabla_l u + \hat{c}_i \hat{c}_j C_1 \nabla_z u + \hat{c}_i \hat{c}_j (d_{31} \delta P + d_{41} \nabla_z P + d_{51} \nabla_x P) \\ &+ \hat{c}_i \hat{c}_j (\bar{\theta}_4 \delta \rho + \bar{\theta}_5 \delta \sigma + \bar{\theta}_6 \delta \phi) + \hat{l}_i \hat{l}_j (C_2 \nabla_z u + d_{32} \delta P) + \hat{l}_i \hat{l}_j (d_{42} \nabla_z P + d_{52} \nabla_x P) \\ &+ \hat{l}_i \hat{l}_j (\bar{\theta}_1 \delta \rho + \bar{\theta}_2 \delta \sigma + \bar{\theta}_3 \delta \phi) + E_0^2 (\chi_{a1} \hat{c}_i \hat{c}_j \nabla_x^2 u + \chi_{a2} \hat{p}_i \hat{p}_j \nabla_z^2 u + 2\chi_4 (\hat{c}_i \hat{p}_j + \hat{p}_i \hat{c}_j) \nabla_x \nabla_z u), \end{aligned} \quad (\text{A4})$$

with $h_i^u = h_i'^u - \nabla_j \Phi_{ij}^u$:

$$h_E^c = \frac{\partial \varepsilon_{Ec}}{\partial \delta c} \Big|_{\dots} = \sin^2(\alpha) E_0 P_0 \delta c, \quad (\text{A5})$$

$$h_i^c = \frac{\partial \varepsilon}{\partial \nabla_i c} \Big|_{\dots} = K_{ij}^{(c)} \nabla_j c + D_{jki} \nabla_j \nabla_k u + \hat{l}_i (\psi_1 \delta \rho + \psi_2 \delta \sigma + \psi_3 \delta \phi + \psi_4 \delta P) \quad (\text{A6})$$

$$\delta \mu = \frac{\partial \varepsilon}{\partial \delta \rho} \Big|_{\dots} = \gamma_1 \delta P + \theta_1 \nabla_z P + \theta_4 \nabla_x P + \bar{\theta}_1 \nabla_y^2 u + \bar{\theta}_4 \nabla_x^2 u + \hat{\theta}_1 \nabla_z u + \psi_1 \nabla_y c + c_{\rho\rho} \delta \rho + c_{\rho\phi} \delta \phi + c_{\rho\sigma} \delta \sigma, \quad (\text{A7})$$

$$\delta T = \frac{\partial \varepsilon}{\partial \delta \sigma} \Big|_{\dots} = \gamma_2 \delta P + \theta_2 \nabla_z P + \theta_5 \nabla_x P + \bar{\theta}_2 \nabla_y^2 u + \bar{\theta}_5 \nabla_x^2 u + \hat{\theta}_2 \nabla_z u + \psi_2 \nabla_y c + c_{\sigma\sigma} \delta \sigma + c_{\rho\sigma} \delta \rho + c_{\sigma\phi} \delta \phi, \quad (\text{A8})$$

$$\delta \Pi = \frac{\partial \varepsilon}{\partial \delta \phi} \Big|_{\dots} = \gamma_3 \delta P + \theta_3 \nabla_z P + \theta_6 \nabla_x P + \bar{\theta}_3 \nabla_y^2 u + \bar{\theta}_6 \nabla_x^2 u + \hat{\theta}_3 \nabla_z u + \psi_3 \nabla_y c + c_{\phi\phi} \delta \phi + c_{\phi\rho} \delta \rho + c_{\phi\sigma} \delta \sigma, \quad (\text{A9})$$

$$v_i = \frac{\partial \varepsilon}{\partial g_i} \Big|_{\dots} = g_i / \rho. \quad (\text{A10})$$

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