Nonlinear smectic elasticity of helical state in cholesteric liquid crystals and helimagnets

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Nonlinear smectic elasticity of helical state in cholesterics liquid crystals and helimagnets

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General symmetry arguments, dating back to de Gennes dictate that at scales longer than the pitch, the low-energy elasticity of a chiral nematic liquid crystal (cholesteric) and of a Dzyaloshinskii-Morya (DM) spiral state in a helimagnet with negligible crystal symmetry fields (e.g., MnSi, FeGe) is identical to that of a smectic liquid crystal, thereby inheriting its rich phenomenology. Starting with a chiral Frank free-energy (exchange and DM interactions of a helimagnet) we present a transparent derivation of the fully nonlinear Goldstone mode elasticity, which involves an analog of the Anderson-Higgs mechanism that locks the spiral orthonormal (director/magnetic moment) frame to the cholesteric (helical) layers. This shows explicitly the reduction of three orientational modes of a cholesteric down to a single phonon Goldstone mode that emerges on scales longer than the pitch. At a harmonic level our result reduces to that derived many years ago by Lubensky and collaborators.

Dating back to the original cholesterol liquid crystal discovered by Reinitzer, chirality plays a central role in modern study of liquid crystals[1]. It is equally important for understanding helical states of noncentrosymmetric magnets (e.g., MnSi)[2–4], driven by a chiral Dzyaloshinskii-Morya (DM) interaction[5].

Among a wealth of induced phenomena[1] chirality converts uniform nematic and ferromagnetic phases into states in which the orientational (nematic or spin) field twists periodically, thereby leading to a variety of spatially modulated phases, such as the cholesteric, two- and three-dimensional "Blue" phases[1, 6, 7] and recently discovered Skyrmion line crystals in MnSi helimagnet[8]. These spontaneously break the translational and rotational symmetries, forming liquid-crystalline structures that are periodic along 1, 2 or 3 dimensions. A cholesteric and helical phases are the most ubiquitous of these, characterized by a biaxial order with

$$\hat{n}_0(\mathbf{r}) \;=\; \hat{x}_1\cos(\mathbf{q}\cdot\mathbf{r}) + \hat{x}_2\sin(\mathbf{q}\cdot\mathbf{r}), \qquad (1)$$

breaking the translational symmetry along a single, spontaneously selected axis, $\hat{x}_3 \equiv \hat{z}$, where $\hat{x}_1, \hat{x}_2, \hat{x}_3 \equiv \hat{x}_1\times\hat{x}_2$ form an orthonormal triad that is constant in the ground state.

General symmetry arguments[1] and an explicit derivation at a *harmonic* level[9] applied to this spontaneously layered helical state predict that at scales longer than the helical pitch, the low-energy (Goldstone mode) elasticity is identical to that of a smectic liquid crystal[1]. The three orientational degrees of freedom defining the cholesteric at short scales thereby reduce to a single smectic-like phonon mode, that emerges on scales longer than the pitch. The associated enhanced fluctuations in[10, 11] and disordering of these states leads to rich phenomenology[1, 13], and in the case of MnSi is believed to be associated with the striking observation of the non-Fermi liquid behavior[12].

Although by now quite familiar, the symmetry breaking of the helical state falls outside the conventional G/H paradigm[13]. Despite fully breaking the $O(3)$ rotational symmetry of the Euclidean group the state is characterized by only a *single* $U(1)$ Goldstone mode, χ, the spiral's phase related to the smectic phonon $u = -\chi/q_0$. As we will show below, the absence of the two additional orientational Goldstone modes is best understood as a mathematical equivalent of the Anderson-Higgs mechanism[1, 13, 14] that gaps them out.

In this paper we explicitly show how this single low-energy helical mode emerges and derive its nonlinear smectic energy functional, expected from the underlying rotational symmetry[1, 13]. Because the latter leads to harmonic phonon fluctuations that diverge in three dimensions and below, the inclusion of nonlinearities is essential for a sensible and self-consistent description, as anticipated long ago by Lubensky, et al.,[9] and Grinstein and Pelcovits[10, 11]. With the neglect of crystalline anisotropy all of our cholesteric results apply equally well to the description of the low-energy bosonic modes of the helical state in the DM helimagnets such as MnSi[2–4].

The helical texture $\hat{n}_0(\mathbf{r})$ minimizes the chiral Frank-Oseen free-energy density of a chiral nematic[1, 13, 15]

$$\mathcal{H} \;=\; \frac{1}{2}K_s(\nabla\cdot\hat{n})^2 + \frac{1}{2}K_b(\hat{n}\times\nabla\times\hat{n})^2, \\ +\frac{1}{2}K_t(\hat{n}\cdot\nabla\times\hat{n} + q_0)^2, \qquad (2a)$$

$$= \; \frac{1}{2}K\left[(\partial_i\hat{n}_j)^2 + 2q_0\hat{n}\cdot\nabla\times\hat{n} + q_0^2\right], \qquad (2b)$$

where in the second form we focused on the isotropic limit, $K_s = K_b = K_t \equiv K$ and dropped the total derivative saddle-splay (Gaussian curvature) contribution, that reduce the Hamiltonian to that of DM ferromagnet in the absence of crystal symmetry breaking fields[3, 5]. Within this approximation the space-spin $(\mathbf{r}-\hat{n})$ coupling only enters through the chiral (second) term, with the elastic (first) piece explicitly exhibiting independent rotational invariances of space, $\mathbf{r}$ and of the director, $\hat{n}$ (spin in the MnSi magnet context).

We now look at long-wavelength, low-energy Goldstone modes excitations about the helical ground state, $\hat{n}_0(\mathbf{r})$. A general state is described by

$$\hat{n}(\mathbf{r}) \;=\; \hat{e}_1(\mathbf{r})\cos(\mathbf{q}\cdot\mathbf{r}+\chi(\mathbf{r})) + \hat{e}_2(\mathbf{r})\sin(\mathbf{q}\cdot\mathbf{r}+\chi(\mathbf{r})), \tag{3}$$

where $\hat{e}_1(\mathbf{r}), \hat{e}_2(\mathbf{r}), \hat{e}_3(\mathbf{r}) \equiv \hat{e}_1(\mathbf{r}) \times \hat{e}_2(\mathbf{r})$ now constitute a spatially dependent orthonormal frame, describing the orientation of the local director (spin) helical plane, that is independent of the helical axis set by $\mathbf{q}\cdot\mathbf{r}+\chi(\mathbf{r}) = const$. The phase $\chi(\mathbf{r}) = -q_0 u(\mathbf{r})$ of the chiral helix also defines the phonon field $u(\mathbf{r})$ of the helical layers, which on scales longer than the pitch $a = 2\pi/q_0$ define the smectic-like displacement of these helical phase fronts. Thus, altogether on the intermediate scales there are three independent orientational degrees of freedom, $\chi(\mathbf{r})$ and $\hat{e}_3(\mathbf{r})$. The azimuthal angle $\phi(\mathbf{r})$, defining the orientation of the $\hat{e}_{1,2}$ around $\hat{e}_3$ is redundant to $\chi(\mathbf{r})$, as it can be eliminated in favor of it via a local gauge-like transformation on $\hat{e}_{1,2}$. Although naively, the low-energy coset space is isomorphic to $(S_1 \otimes S_2)/\mathbb{Z}_2 = RP_3$, as we will see below, only a single Goldstone mode, $\chi(\mathbf{r})$ will survive this helical symmetry breaking.

Substituting $\hat{n}(\mathbf{r})$ from Eq. (3) into the free-energy of the chiral nematic (helimagnet), Eq. (2b), and using

$$\begin{aligned}\partial_i \hat{n}_j \;&=\; (q_i + \partial_i\chi)[-\hat{e}_{1j}\sin(\mathbf{q}\cdot\mathbf{r}+\chi) + \hat{e}_{2j}\cos(\mathbf{q}\cdot\mathbf{r}+\chi)] \\ &\quad +\partial_i\hat{e}_{1j}\cos(\mathbf{q}\cdot\mathbf{r}+\chi) + \partial_i\hat{e}_{2j}\sin(\mathbf{q}\cdot\mathbf{r}+\chi),\end{aligned} \tag{4}$$

together with

$$\partial_i \hat{e}_{1j} \;=\; a_i \hat{e}_{2j} + c_{1i}\hat{e}_{3j}, \tag{5a}$$
$$\partial_i \hat{e}_{2j} \;=\; -a_i \hat{e}_{2j} + c_{2i}\hat{e}_{3j}, \tag{5b}$$

and gauge field "spin-connections"

$$a_i \;=\; \hat{e}_2 \cdot \partial_i \hat{e}_1, \tag{6a}$$
$$c_{1i} \;=\; -\hat{e}_1 \cdot \partial_i \hat{e}_3, \tag{6b}$$
$$c_{2i} \;=\; -\hat{e}_2 \cdot \partial_i \hat{e}_3, \tag{6c}$$

we find

$$\begin{aligned}\mathcal{H} \;&=\; \frac{K}{2}\Big[(a_i\hat{e}_{2j} + c_i\hat{e}_{3j})\cos(\mathbf{q}\cdot\mathbf{r}+\chi) + (-a_i\hat{e}_{1j} + c_{2i}\hat{e}_{3j})\sin(\mathbf{q}\cdot\mathbf{r}+\chi) \\ &\quad +(q_i+\partial_i\chi)\big[-\hat{e}_{1j}\sin(\mathbf{q}\cdot\mathbf{r}+\chi) + \hat{e}_{2j}\cos(\mathbf{q}\cdot\mathbf{r}+\chi)\big] \\ &\quad +q_0(\hat{e}_{2i}\hat{e}_{3j} - \hat{e}_{3i}\hat{e}_{2j})\cos(\mathbf{q}\cdot\mathbf{r}+\chi) + q_0(\hat{e}_{3i}\hat{e}_{1j} - \hat{e}_{1i}\hat{e}_{3j})\sin(\mathbf{q}\cdot\mathbf{r}+\chi)\Big]^2,\end{aligned} \tag{7a}$$

$$\;=\; \frac{K}{2}(\nabla\chi + \mathbf{a} + \mathbf{q} - q_0\hat{e}_3)^2 + \frac{K}{4}(\mathbf{c}_1 + q_0\hat{e}_2)^2 + \frac{K}{4}(\mathbf{c}_2 - q_0\hat{e}_1)^2 \tag{7b}$$

In obtaining Eq.(7b), we dropped the constant and oscillatory parts, that average away upon spatial integration.

We first note that the requirement of well-defined helical phase fronts, i.e., the absence of dislocations and disclinations in the layer structure, can be enforced by the compatibility condition $\nabla \times \mathbf{a} = 0$, consistent with Mermin-Ho relation[16]. This allows us to take $\mathbf{a} = \nabla\phi$ and thereby shift ("gauge") away $\phi(\mathbf{r})$ in favor of $\chi(\mathbf{r})$, according to $\chi(\mathbf{r}) + \phi(\mathbf{r}) \to \chi(\mathbf{r})$.

Without loss of generality, we next take $\mathbf{q} = q_0\hat{z}$ with $\hat{z}$ defining the orientation of the helical axis in the laboratory coordinate system $\hat{x}, \hat{y}, \hat{z}$. The long-wavelength free-energy density, reexpressed in terms of the smectic-like phonon field $u(\mathbf{r})$ and the local nematic helical frame orientation $\hat{e}_3$ then reduces to

$$\begin{aligned}\mathcal{H} \;&=\; \frac{Kq_0^2}{2}(\nabla u + \hat{e}_3 - \hat{z})^2 + \frac{K}{4}(\mathbf{c}_1^2 + \mathbf{c}_2^2) \\ &\quad + \frac{Kq_0}{2}(\mathbf{c}_1\cdot\hat{e}_2 - \mathbf{c}_2\cdot\hat{e}_1).\end{aligned} \tag{8}$$

Clearly the first term, above, accounts for the energetic cost of the deviation of the local nematic frame $\hat{e}_3$ from the local orientation of the helical layers. A minimization of this term (or equivalently at long wavelengths, in a statistical mechanical treatment integrating out the independent $\hat{e}_3(\mathbf{r})$ degree of freedom), at low-energies locks the orientations of the cholesteric layers and the nematic frame. In a perturbative treatment this leads to the expected relation

$$\nabla_\perp u \approx -\hat{e}_{3\perp}, \tag{9}$$

that is an example of a Higg's-like mechanism (akin to thermotropic smectic liquid crystals[1, 13, 17]), that at long scales effectively gaps out the orientational Goldstone modes.

The exact minimization over the unit vector $\hat{e}_3(\mathbf{r})$ can also be carried out using a Lagrange multiplier λ to impose the constraint $\hat{e}_3 \cdot \hat{e}_3 = 1$. Minimization over $\hat{e}_3(\mathbf{r})$ gives

$$\nabla u + \hat{e}_3 - \hat{z} = -\lambda\hat{e}_3, \tag{10}$$

with the solution

$$\lambda + 1 = \sqrt{(\nabla u - \hat{z})^2} = \sqrt{1 - 2u_{zz}}, \quad (11a)$$
$$\hat{e}_3 = (\hat{z} - \nabla u)/\sqrt{1 - 2u_{zz}}, \quad (11b)$$

where

$$u_{zz} = \partial_z u - \frac{1}{2}(\nabla u)^2 \quad (12)$$

is the standard fully nonlinear smectic strain tensor, that encodes the full rotational invariance of the helical state[10, 13]. Using Eqs.(11) to eliminate λ and $\hat{e}_3(\mathbf{r})$ in favor u_{zz} (valid at long scales), we find

$$\mathcal{H} = \frac{Kq_0^2}{2}(\sqrt{1 - 2u_{zz}} - 1)^2 + \frac{K}{4}\left[(\hat{e}_1 \cdot \partial_i \hat{e}_3)(\hat{e}_1 \cdot \partial_i \hat{e}_3) + (\hat{e}_2 \cdot \partial_i \hat{e}_3)(\hat{e}_2 \cdot \partial_i \hat{e}_3)\right] + \frac{Kq_0}{2}\hat{e}_{1i}\hat{e}_{2j}(\partial_i \partial_j u - \partial_j \partial_i u), \quad (13a)$$
$$\approx \frac{B}{2}u_{zz}^2 + \frac{\overline{K}}{2}(\partial_i \partial_j^{\perp} u)^2, \quad (13b)$$
$$\approx \frac{B}{2}\left[\partial_z u - \frac{1}{2}(\nabla u)^2\right]^2 + \frac{\overline{K}}{2}(\nabla_{\perp}^2 u)^2, \quad (13c)$$

where to obtain our main result, Eq. (13c) we used the condition of well-defined helical layers with no dislocations in $u(\mathbf{r})$, i.e., a single-valued phase field $\chi(\mathbf{r})$, neglected the boundary terms, expanded to lowest order in the nonlinear strain tensor u_{zz}, and defined the compressional and bending elastic moduli

$$B = Kq_0^2, \quad \overline{K} = K/2. \quad (14)$$

Thus, as advertised, we have demonstrated that on scales longer than the helical pitch $2\pi/q_0$, the low-energy deformations (Goldstone modes) of the helical state are characterized by a fully rotationally invariant, nonlinear smectic elastic theory[1, 10, 13]. The latter can be derived by spontaneously ordering the density $\rho(\mathbf{r})$ of the isotropic fluid into a one-dimensional periodically modulated state (smectic), characterized by $\rho(\mathbf{r}) = \rho_0 + \rho_q \cos[(\mathbf{q} \cdot \mathbf{r} - q_0 u)]$[10]. Alternatively, the above nonlinear compressional form (first term in (13a)) emerges directly from the de Gennes' gauge theory of the smectic[1], after condensing $\rho_q = |\rho_q|e^{i\Phi(\mathbf{r})}$ to give

$$\mathcal{H}_{sm} = \frac{1}{2}B(\nabla\Phi - \hat{n})^2 + \frac{1}{2}K(\nabla \cdot \hat{n})^2, \quad (15)$$

and then minimizing over the unit director $\hat{n}(\mathbf{r})$ as in (10)-(12).

We note that as required, $\mathcal{H}$ in Eq.(13a) is a function of the fully nonlinear strain u_{zz} as it must to preserve full rotational invariance. Furthermore, it is a nonlinear function of this strain, that reduces to the familiar "harmonic nonlinear" form[10] in Eq.(13c) only for small u_{zz}. It is worth observing that through the introduction of the phase field $\Phi = -z + u$, this nonlinear in u_{zz} term in (13a) can be written as $(|\nabla\Phi| - 1)^2$. This form has been used in recent analyses of various nonlinear properties of smectics.[18]

Our result in Eq. (13c) then in turn implies that the helical state inherits all the novel nonlinear elastic effects previously discovered in the context of conventional, thermotropics and lyotropic smectic liquid crystals and other spontaneously layered states that emerge from an isotropic state. These include thermal fluctuations[10, 11, 19] and heterogeneous[20] anomalous elasticity effects, the undulation instability[21], and many others.

One important distinction from a conventional smectic, however, is the underlying chirality of the helical layered state. Although as in the chiral smectic the effective Anderson-Higgs mechanism expels the expression of chirality (e.g., twist) inside the helical state (as the magnetic flux density [twist of the vector potential] is expelled from the Meissner state)[1, 13, 17], inclusion of the chiral terms (encoded in the departure from the $\nabla \times \mathbf{a} = 0$ condition used to get to (8)) is essential to understanding the topological defects and melting of the helical state[22].

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