

Hydrodynamical modes and light scattering in the liquid-crystalline cubic blue phases. I. Elastic theory

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Light-scattering experiments reveal a strange behavior of thermal excitations in the liquid-crystalline cubic blue phases. To interpret the experiments we identify the hydrodynamical modes of the orientational pattern with pure *displacements* of the order parameter field. There are further modes that deform the orientational pattern, e.g., local *rotations* of the order parameter. An investigation of the elastic free energy shows that our identification of the hydrodynamical modes is valid as long as the wavelength is much smaller than the lattice constant. In this long-wavelength limit the additional deformation modes merely renormalize the elastic constants of the displacement modes. We study the influence of two characteristic deformations, rotational and $m = 2$ modes, and discuss the temperature and chirality behavior of the elastic constants. Also Keyes's [Phys. Rev. Lett. **65**, 436 (1990)] idea of considering the phase transition to blue phase III as a melting of the cubic blue phases is critically reviewed.

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I. INTRODUCTION

Blue phases are attracting considerable attention because they have much more in common with the crystalline state than other liquid crystals. The centers of mass of the molecules do not show positional order, but the molecular axes align to form a complex long-range orientational pattern. In the cubic blue phases (BPs) I and II this pattern is periodic along all three spatial directions with cubic space group symmetries O^8 and O^2 , respectively. The lattice constant is of the order of several hundred nanometers and the unit cell contains 10^7 molecules. The deviation of the distribution of the molecular axes from isotropy leads to local anisotropic physical properties, for example, in the dielectric response. We therefore choose as an order parameter a tensor field $\boldsymbol{\mu}(\mathbf{r})$ proportional to the anisotropic part $\delta\boldsymbol{\varepsilon}(\mathbf{r})$ of the dielectric tensor $\boldsymbol{\varepsilon}(\mathbf{r})$:

$$\boldsymbol{\mu}(\mathbf{r}) \propto \delta\boldsymbol{\varepsilon}(\mathbf{r}) = \boldsymbol{\varepsilon}(\mathbf{r}) - \frac{1}{3} \mathbf{1} \operatorname{tr}\boldsymbol{\varepsilon}(\mathbf{r}) . \quad (1)$$

It governs light scattering. The dielectric tensor is symmetric and so is characterized by its eigenvectors and eigenvalues. For a visualization of the tensor field in the cubic unit cell see Barbet-Massin and Pieranski [1].

The orientational pattern can be deformed in different ways. For example, the tensor $\boldsymbol{\mu}(\mathbf{r})$ may be rotated

locally, and with it its eigenvectors are rotated. Alternatively the directions of the main axes may be kept fixed and the eigenvalues are changed, altering the degree of the molecular alignment. Also a deformation of the whole unit cell as in normal crystals is possible. In this and the following paper [2] we shall deal with the elastic and dynamic properties of special deformations in the cubic blue phases, the hydrodynamical modes. We will see that, in a certain limit, these are described just by pure displacements of the order parameter $\boldsymbol{\mu}(\mathbf{r})$. Therefore they are called *displacement modes*, equivalent to acoustic phonons in crystals.

Modes are denoted hydrodynamical [3] if for a plane wave ansatz

$$\exp(-z\mathbf{t} + i\mathbf{q} \cdot \mathbf{r}) \quad (2)$$

the relaxation frequency $\operatorname{Re}z(\mathbf{q})$ goes to zero and the lifetime $[\operatorname{Re}z(\mathbf{q})]^{-1}$ becomes infinite for $\mathbf{q} \rightarrow \mathbf{0}$. Usually one finds $\operatorname{Re}z(\mathbf{q}) \sim \mathbf{q}^2$. The lifetime of hydrodynamical modes is much larger than that of microscopic excitations.

Light scattering from thermally excited hydrodynamical modes is an efficient tool for studying their dynamics. We introduce two methods to identify them [4,5] and briefly review what one can learn from such experiments. In an isotropic liquid a frequency analysis of the scattered light reveals the dispersion relations for three hydrodynamical modes belonging to two sound waves traveling in opposite directions and a heat diffusion mode. One can extract material parameters such as compressibility, longitudinal viscosity, and thermal conductivity [3,5]. The three modes result from three conserved quantities: the mass, the longitudinal component of momentum, and the

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energy. For example, to smooth an inhomogeneous mass distribution corresponding to a mode of wavelength λ , mass must be transported over a distance of $\lambda/2$, requiring infinite time as $q \sim \lambda^{-1}$ tends to zero. Not only conserved quantities but also broken continuous symmetries lead to hydrodynamical modes, which are then denoted as *Goldstone* modes [5]. In the nematic phase, for example, the full rotational symmetry of physical space is broken by the director. A homogeneous rotation of the director field described by a rotation vector $\varphi(\mathbf{q} = \mathbf{0})$ does not cost any elastic free energy δf , and for spatially modulated rotations [$\varphi(\mathbf{q}) \neq \mathbf{0}$] the elastic free energy $\delta f[\varphi(\mathbf{q})]$ becomes zero in the long-wavelength limit, as is the case for the Frank-Oseen free energy. Therefore there exist large, thermally activated director fluctuations which scatter light strongly. Their relaxation frequency is proportional to the elastic restoring force $\partial \delta f[\varphi(\mathbf{q})]/\partial \varphi(\mathbf{q})$ of the director field and also vanishes with q . The director modes are calculated from the Leslie-Ericksen equations [6,7]. Using different geometries, one can measure the Frank-Oseen elastic constants and the viscosities appearing in the dynamical equations by light scattering experiments.

In periodic liquid-crystalline systems the identification of the hydrodynamical modes is more complicated because, in addition, the translational symmetry of physical space is broken. For the cholesteric phase hydrodynamical modes and their equations have been studied by Lubensky [4]. Further analysis was done in Refs. [8,9] and the first light-scattering experiments were performed by Domberger [10].

In 1984 Marcus [11] observed Bragg reflections from the orientational pattern of the cubic blue phases and noticed strong fluctuations of the scattered light intensity. These evidently had their origin in thermal modes which are easily excitable. Candidates are the above mentioned displacement modes with a hydrodynamical character due to the broken translational symmetry (Sec. II). This light diffraction is equivalent to diffuse x-ray scattering from acoustic phonons in normal crystals, as first suggested by Dmitrienko [12]. In further experiments, carried out in foreshattering, Marcus [13] and later Domberger [10] identified two purely diffusive modes. Surprisingly both authors found finite relaxation frequencies for $q \rightarrow \mathbf{0}$ on the order of 1000 s^{-1} , an unusual behavior for hydrodynamical modes as pointed out by Marcus [13]. This puzzle will be studied in the following paper [2].

We will follow a strategy of three steps: (1) identification of the displacement modes with hydrodynamical modes and investigation of their elastic free energy, (2) formulation and study of the hydrodynamical equations, and (3) calculation of light-scattering intensities from the displacement modes. In this article we deal with the first step. The second and third steps will be treated in the following paper [2].

II. DISPLACEMENT MODES

In cubic blue phases the translational and rotational symmetry of physical space is broken to certain space

group symmetries. For the hydrodynamical modes one expects a combination of translations and rotations of the local order parameter $\boldsymbol{\mu}(\mathbf{r})$. As will become evident later, only the translational symmetry must be considered. We introduce a deformed order parameter field $\boldsymbol{\mu}_d(\mathbf{r})$ by shifting the undeformed order parameter field $\boldsymbol{\mu}(\mathbf{r})$:

$$\boldsymbol{\mu}(\mathbf{r}) \longrightarrow \boldsymbol{\mu}_d(\mathbf{r}) = \boldsymbol{\mu}[\mathbf{r} - \mathbf{u}(\mathbf{r})] ; \quad (3)$$

$\mathbf{u}(\mathbf{r})$ is a field of displacement vectors. Restriction to small displacements gives

$$\boldsymbol{\mu}_d(\mathbf{r}) = \boldsymbol{\mu}(\mathbf{r}) + \delta \boldsymbol{\mu}_u(\mathbf{r}) \approx \boldsymbol{\mu}(\mathbf{r}) - [\mathbf{u}(\mathbf{r}) \cdot \nabla] \boldsymbol{\mu}(\mathbf{r}) \quad (4)$$

and the use of the Fourier expansions

$$\boldsymbol{\mu}(\mathbf{r}) = \sum_{\mathbf{k}} \boldsymbol{\mu}(\mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{r}) , \quad (5)$$

$$\mathbf{u}(\mathbf{r}) = \sum_{\mathbf{q}} \mathbf{u}(\mathbf{q}) \exp(i\mathbf{q} \cdot \mathbf{r}) \quad (6)$$

(\mathbf{k} is a reciprocal lattice vector and \mathbf{q} a wave vector satisfying periodic boundary conditions) leads to

$$\delta \boldsymbol{\mu}_u(\mathbf{r}) = - \sum_{\mathbf{q}, \mathbf{k}} [i\mathbf{u}(\mathbf{q}) \cdot \mathbf{k}] \boldsymbol{\mu}(\mathbf{k}) \exp[i(\mathbf{k} + \mathbf{q}) \cdot \mathbf{r}] . \quad (7)$$

The deviation $\delta \boldsymbol{\mu}_u(\mathbf{r})$ from the undeformed order parameter field appears as a sum of Bloch functions with wave vectors \mathbf{q} . These are the displacement modes. Our approximation is valid only for $|\mathbf{u}(\mathbf{q}) \cdot \mathbf{k}| \ll 1$, which is satisfied for thermal excitations.

With the same procedure we could have introduced the acoustic phonons for the periodic mass density of normal crystals, but there is an essential difference. In crystals, atoms are shifted, i.e., unchangeable quantities which we can identify in principle before and after a deformation. On the other hand, if the order parameter $\boldsymbol{\mu}(\mathbf{r})$ changes its orientation and shape during the displacement, as explained in the Introduction, we cannot identify it before and after and the use of displacement modes makes no sense. Only if the displacement modes are eigenmodes of the elastic free energy of the blue phases do they not couple to other deformations and therefore do not change.

Extending Eq. (7) we introduce a *general* deviation

$$\delta \boldsymbol{\mu}(\mathbf{r}) = \delta \boldsymbol{\mu}_u(\mathbf{r}) + \delta \tilde{\boldsymbol{\mu}}(\mathbf{r}) \quad (8)$$

from the undeformed order parameter field $\boldsymbol{\mu}(\mathbf{r})$ and assume an expansion into Bloch functions:

$$\delta \boldsymbol{\mu}(\mathbf{r}) = \sum_{\mathbf{q}, \mathbf{k}} \delta \boldsymbol{\mu}(\mathbf{k} + \mathbf{q}) \exp[i(\mathbf{k} + \mathbf{q}) \cdot \mathbf{r}] . \quad (9)$$

In addition to the displacement modes the Fourier coefficient

$$\delta \boldsymbol{\mu}(\mathbf{k} + \mathbf{q}) = \delta \tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) - [i\mathbf{u}(\mathbf{q}) \cdot \mathbf{k}] \boldsymbol{\mu}(\mathbf{k}) \quad (10)$$

now includes further deformation modes expressed by the amplitude $\delta \tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})$. In Sec. II B we will study the elastic free energy of such a general deformation. We will show that to a good approximation the displacement modes

are elastic eigenmodes if we restrict ourselves to long-wavelength deformations where the wavelengths must be much larger than the lattice constant b of the cubic blue phases:

$$q \ll \frac{2\pi}{b} . \quad (11)$$

Nevertheless, coupling to other deformations leads to corrections in the elastic tensor of the displacement modes. We will study these corrections for special deformations in the rest of this section. First we briefly summarize the theory of blue phases.

A. Theory of cubic blue phases

For the thermodynamical description of the blue phases, a free energy according to Landau-de Gennes theory is used, in scaled units [14]:

$$\begin{aligned} f[\boldsymbol{\mu}(\mathbf{r})] = & \frac{1}{2} \int d^3r \left(\frac{t}{2} \text{tr} \boldsymbol{\mu}^2 + \frac{\kappa^2}{2q_c^2} (\nabla \otimes \boldsymbol{\mu}) \cdot (\nabla \otimes \boldsymbol{\mu}) \right. \\ & \left. + \frac{c_2}{c_1} \frac{\kappa^2}{2q_c^2} \nabla \boldsymbol{\mu} \cdot \nabla \boldsymbol{\mu} - \frac{\kappa^2}{q_c} (\nabla \times \boldsymbol{\mu}) \cdot \boldsymbol{\mu} \right) \quad (12) \\ & - \sqrt{6} \int d^3r \text{tr} \boldsymbol{\mu}^3 + \int d^3r (\text{tr} \boldsymbol{\mu}^2)^2 . \end{aligned}$$

Characteristic parameters are the chirality κ , which is proportional to the wave number q_c of the cholesteric helix, the reduced temperature t , and the ratio c_2/c_1 of elastic constants. Throughout we will use a coordinate-free representation of tensor analysis with the following notations:

$$\begin{aligned} \text{tr} \boldsymbol{\mu}^2 &= \mu_{ij} \mu_{ji}, & [\nabla \otimes \boldsymbol{\mu}]_{ijk} &= \mu_{jk,i}, \\ [\nabla \boldsymbol{\mu}]_j &= \mu_{ij,i}, & [\nabla \times \boldsymbol{\mu}]_{in} &= \varepsilon_{ijl} \mu_{ln,j}, \end{aligned} \quad (13)$$

where ε_{ijl} are the components of the Levi-Civita tensor $\boldsymbol{\varepsilon}_{LC}$. The dot between two tensors always stands for a contraction over all indices from left to right and the comma means partial derivative.

The minimization of the free energy starts with the Fourier expansion (5) of the order parameter field $\boldsymbol{\mu}(\mathbf{r})$. The Fourier coefficient $\boldsymbol{\mu}(\mathbf{k})$, also a symmetric and traceless tensor of second rank, is written in a spherical tensor basis [15]:

$$\boldsymbol{\mu}(\mathbf{k}) = \sum_{m=-2}^2 \mu_m(\mathbf{k}) \mathbf{M}_m(\mathbf{k}) . \quad (14)$$

Thus $\boldsymbol{\mu}(\mathbf{r})$ is expanded into tensor modes of helicity m . For later use we give the definition of $\mathbf{M}_2(\mathbf{k})$:

$$\mathbf{M}_2(\mathbf{k}) = \mathbf{m}(\mathbf{k}) \otimes \mathbf{m}(\mathbf{k}) , \quad \mathbf{m}(\mathbf{k}) = \frac{1}{\sqrt{2}} (\boldsymbol{\xi} + i\boldsymbol{\eta}) , \quad (15)$$

where $\{\boldsymbol{\xi}, \boldsymbol{\eta}, \mathbf{k}/k\}$ is a right-handed system of orthonormal vectors.

When the order parameter field possesses a space group symmetry \mathcal{G} , it is useful to subdivide the sum over the \mathbf{k} vectors in Eq. (5) in the following way:

$$\begin{aligned} \boldsymbol{\mu}(\mathbf{r}) = & \sum_{\mathbf{k}_R} \sum_{S \in \mathcal{P}(\mathbf{k}_R)} \sum_{m=-2}^2 \mu_m(\mathbf{S}\mathbf{k}_R) \mathbf{M}_m(\mathbf{S}\mathbf{k}_R) \\ & \times \exp(i\mathbf{S}\mathbf{k}_R \cdot \mathbf{r}) . \end{aligned} \quad (16)$$

\mathbf{k}_R is the *representative* of a star of \mathbf{k} vectors which is constructed by application of all elements \mathbf{S} of the point group \mathcal{P} , for example, the cubic point group O , to \mathbf{k}_R . In special stars the number $N(\mathbf{k}_R)$ of \mathbf{k} vectors is smaller than the order of the point group and only a subset $\mathcal{P}(\mathbf{k}_R)$ of \mathcal{P} is needed for the construction. Because of the invariance of the order parameter field:

$$\{[\mathbf{S}|\mathbf{t}]\boldsymbol{\mu}\}(\mathbf{r}) := \mathbf{S}\boldsymbol{\mu}(\{\mathbf{S}|\mathbf{t}\}^{-1}\mathbf{r}) = \boldsymbol{\mu}(\mathbf{r}) \quad (17)$$

($\mathbf{S}\boldsymbol{\mu}$ is the symbol for the rotated $\boldsymbol{\mu}$), there is only one independent complex amplitude $\mu_m(\mathbf{k}_R)$ for each star and each helicity m . If \mathbf{k} and $-\mathbf{k}$ belong to the same star, the phase factor of the complex amplitude is fixed up to a sign because the order parameter is real [16].

An exact minimization of the free energy (12) is currently not possible. In our quantitative discussion we use the results of the high chirality limit, i.e., for $\kappa \rightarrow \infty$. A separate consideration of the elastic part in the free energy reveals that modes of helicity $m = 2$ or -2 are favored [15]. The amplitudes $\mu_2(\mathbf{k}_R)$ together with the lattice constant b , which is comparable to $2\pi/q_c$, follow from the minimization of the entire free energy for a restricted number of stars. With this method Grebel, Hornreich, and Shtrikman (GHS) [14,15,17] could reproduce the phase diagram of the cubic blue phases, the control parameters being the temperature t and the chirality κ . For the space groups O^2 of BP II and O^8 of BP I they had to take into account the two stars (100) and (110) and the four stars (110), (200), (112), and (220), respectively.

B. Elastic free energy

The elastic free energy δf of a deformed order parameter field $\boldsymbol{\mu}_d(\mathbf{r}) = \boldsymbol{\mu}(\mathbf{r}) + \delta\boldsymbol{\mu}(\mathbf{r})$ is

$$\delta f = f[\boldsymbol{\mu}_d(\mathbf{r})] - f[\boldsymbol{\mu}(\mathbf{r})] . \quad (18)$$

With the free energy of Eq. (12) we obtain up to second order in $\delta\boldsymbol{\mu}(\mathbf{r})$

$$\begin{aligned} \delta f \approx & \frac{1}{2} \int d^3r \left(\frac{t}{2} \text{tr}[\delta\boldsymbol{\mu}^2] + \frac{\kappa^2}{2q_c^2} (\nabla \otimes \delta\boldsymbol{\mu}) \cdot (\nabla \otimes \delta\boldsymbol{\mu}) \right. \\ & \left. + \frac{c_2}{c_1} \frac{\kappa^2}{2q_c^2} \nabla \delta\boldsymbol{\mu} \cdot \nabla \delta\boldsymbol{\mu} - \frac{\kappa^2}{q_c} (\nabla \times \delta\boldsymbol{\mu}) \cdot \delta\boldsymbol{\mu} \right) \quad (19) \\ & - 3\sqrt{6} \int d^3r \text{tr}[\boldsymbol{\mu} \delta\boldsymbol{\mu}^2] \\ & + \int d^3r \left\{ 4(\text{tr}[\boldsymbol{\mu} \delta\boldsymbol{\mu}])^2 + 2\text{tr}[\delta\boldsymbol{\mu}^2] \text{tr}[\boldsymbol{\mu}^2] \right\} . \end{aligned}$$

The integration is carried out over the volume V of the system with periodic boundary conditions. The term linear in $\delta\boldsymbol{\mu}(\mathbf{r})$ must vanish because $\boldsymbol{\mu}(\mathbf{r})$ minimizes the free energy (12). With the Fourier expansion (5) there follows, as the condition for an extremum,

$$\begin{aligned} & \frac{1}{2} \left[t\boldsymbol{\mu}(\mathbf{k}) + \frac{\kappa^2}{q_c^2} k^2 \boldsymbol{\mu}(\mathbf{k}) + \frac{c_2}{c_1} \frac{\kappa^2}{2q_c^2} \left(\mathbf{k} \otimes \boldsymbol{\mu}(\mathbf{k}) \mathbf{k} + \boldsymbol{\mu}(\mathbf{k}) \mathbf{k} \otimes \mathbf{k} - \frac{2}{3} \text{tr}[(\mathbf{k} \otimes \mathbf{k}) \boldsymbol{\mu}(\mathbf{k})] \mathbf{1} \right) - i \frac{\kappa^2}{q_c} \left\{ \mathbf{k} \times \boldsymbol{\mu}(\mathbf{k}) + [\mathbf{k} \times \boldsymbol{\mu}(\mathbf{k})]^T \right\} \right] \\ & - 3\sqrt{6} \sum_{\mathbf{k}'} \left[\boldsymbol{\mu}(\mathbf{k} - \mathbf{k}') \boldsymbol{\mu}(\mathbf{k}') - \frac{1}{3} \text{tr}[\boldsymbol{\mu}(\mathbf{k} - \mathbf{k}') \boldsymbol{\mu}(\mathbf{k}')] \mathbf{1} \right] + 4 \sum_{\mathbf{k}', \mathbf{k}''} \text{tr}[\boldsymbol{\mu}(\mathbf{k} - \mathbf{k}' - \mathbf{k}'') \boldsymbol{\mu}(\mathbf{k}')] \boldsymbol{\mu}(\mathbf{k}'') = \mathbf{0} . \end{aligned} \quad (20)$$

This takes into account that $\boldsymbol{\mu}(\mathbf{k})$ is a traceless tensor.

We now introduce the general deformation of Eq. (9) for $\delta\boldsymbol{\mu}(\mathbf{r})$. The evaluation of δf needs tensor manipulations, permutations of \mathbf{k} vectors, and the application of condition (20). After lengthy calculations we obtain

$$\delta f \approx \delta f_{\mathbf{u}, \mathbf{u}} + \delta f_{\delta\tilde{\boldsymbol{\mu}}, \mathbf{u}} + \delta f_{\delta\tilde{\boldsymbol{\mu}}, \delta\tilde{\boldsymbol{\mu}}} \quad (21)$$

with the three terms

$$\delta f_{\mathbf{u}, \mathbf{u}} = \frac{V}{2} \sum_{\mathbf{k}, \mathbf{q}} \left[\frac{\kappa^2}{2q_c^2} \left\{ \text{tr}[\boldsymbol{\mu}(\mathbf{k}) \boldsymbol{\mu}(-\mathbf{k})] \mathbf{1} + \frac{c_2}{c_1} \boldsymbol{\mu}(\mathbf{k}) \boldsymbol{\mu}(-\mathbf{k}) \right\} \otimes \mathbf{k} \otimes \mathbf{k} \right] \cdot [\mathbf{q} \otimes \mathbf{q} \otimes \mathbf{u}(\mathbf{q}) \otimes \mathbf{u}^*(\mathbf{q})] , \quad (22)$$

$$\begin{aligned} \delta f_{\delta\tilde{\boldsymbol{\mu}}, \mathbf{u}} & \approx i \frac{V}{2} \sum_{\mathbf{k}, \mathbf{q}} \left[\frac{\kappa^2}{q_c^2} \left\{ 2\text{tr}[\boldsymbol{\mu}(-\mathbf{k}) \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})] \mathbf{1} + \frac{c_2}{c_1} [\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) \boldsymbol{\mu}(-\mathbf{k}) + \boldsymbol{\mu}(-\mathbf{k}) \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})] \right\} \mathbf{k} \otimes \mathbf{k} \right. \\ & \left. + i \frac{2\kappa^2}{q_c} \boldsymbol{\varepsilon}_{LV} [\cdot, \boldsymbol{\mu}(-\mathbf{k}) \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})] \otimes \mathbf{k} \right] \cdot [\mathbf{q} \otimes \mathbf{u}^*(\mathbf{q})] , \end{aligned} \quad (23)$$

$$\begin{aligned} \delta f_{\delta\tilde{\boldsymbol{\mu}}, \delta\tilde{\boldsymbol{\mu}}} & = \frac{V}{2} \sum_{\mathbf{k}, \mathbf{q}} \left\{ \frac{t}{2} \text{tr}[\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) \delta\tilde{\boldsymbol{\mu}}^*(\mathbf{k} + \mathbf{q})] + \frac{\kappa^2}{2q_c^2} [(\mathbf{k} + \mathbf{q}) \otimes \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})] \cdot [(\mathbf{k} + \mathbf{q}) \otimes \delta\tilde{\boldsymbol{\mu}}^*(\mathbf{k} + \mathbf{q})] \right. \\ & \left. + \frac{c_2}{c_1} \frac{\kappa^2}{2q_c^2} [\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) (\mathbf{k} + \mathbf{q})] \cdot [\delta\tilde{\boldsymbol{\mu}}^*(\mathbf{k} + \mathbf{q}) (\mathbf{k} + \mathbf{q})] - i \frac{\kappa^2}{q_c} [(\mathbf{k} + \mathbf{q}) \times \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})] \cdot \delta\tilde{\boldsymbol{\mu}}^*(\mathbf{k} + \mathbf{q}) \right\} \\ & - 3\sqrt{6} V \sum_{\Delta\mathbf{k}=\mathbf{0}, \mathbf{q}} \text{tr}[\boldsymbol{\mu}(\mathbf{k}'') \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) \delta\tilde{\boldsymbol{\mu}}^*(-\mathbf{k}' + \mathbf{q})] \\ & + V \sum_{\square\mathbf{k}=\mathbf{0}, \mathbf{q}} \left\{ 4\text{tr}[\boldsymbol{\mu}(\mathbf{k}'') \delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})] \text{tr}[\boldsymbol{\mu}(\mathbf{k}''') \delta\tilde{\boldsymbol{\mu}}^*(-\mathbf{k}' + \mathbf{q})] + 2\text{tr}[\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) \delta\tilde{\boldsymbol{\mu}}^*(-\mathbf{k}' + \mathbf{q})] \text{tr}[\boldsymbol{\mu}(\mathbf{k}'') \boldsymbol{\mu}(\mathbf{k}''')] \right\} . \end{aligned} \quad (24)$$

Here we use the symbols $\Delta\mathbf{k} = \mathbf{k} + \mathbf{k}' + \mathbf{k}''$ and $\square\mathbf{k} = \mathbf{k} + \mathbf{k}' + \mathbf{k}'' + \mathbf{k}'''$. The elastic free energy does not contain any coupling between deformation modes of different wave vectors \mathbf{q} because of the long-wavelength limit (11) and the restriction to second order in $\delta\boldsymbol{\mu}$. In the following arguments the order of \mathbf{q} plays an important role. \mathbf{q} always appears relative to q_c and according to the condition (11), q/q_c is a very small number. $\delta f_{\mathbf{u}, \mathbf{u}}$, the elastic free energy of pure displacement modes, contains \mathbf{q} only in second order as expected from the broken translational symmetry. $\delta f_{\delta\tilde{\boldsymbol{\mu}}, \delta\tilde{\boldsymbol{\mu}}}$, the elastic free energy of further deformations, and $\delta f_{\delta\tilde{\boldsymbol{\mu}}, \mathbf{u}}$, which describes the coupling to displacement modes, depend on \mathbf{q} in zeroth and first order, respectively. If we introduce a large column vector $\Delta\tilde{\boldsymbol{\mu}}(\mathbf{q})$, comprising all the components of $\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})$ of all \mathbf{k} vectors, δf can be rewritten in a Hermitian form:

$$\delta f = \frac{V}{2} \sum_{\mathbf{q}} \begin{pmatrix} \mathbf{u}(\mathbf{q}) \\ \Delta\tilde{\boldsymbol{\mu}}(\mathbf{q}) \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{\lambda}_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q}) & \mathbf{W}(\mathbf{q}) + \mathcal{O}(\mathbf{q} \otimes \mathbf{q}) \\ \mathbf{W}^\dagger(\mathbf{q}) + \mathcal{O}(\mathbf{q} \otimes \mathbf{q}) & \boldsymbol{\Theta} + \mathcal{O}(\mathbf{q}) \end{pmatrix} \begin{pmatrix} \mathbf{u}^*(\mathbf{q}) \\ \Delta\tilde{\boldsymbol{\mu}}^*(\mathbf{q}) \end{pmatrix} . \quad (25)$$

The Hermitian block matrix contains the elastic tensors $\boldsymbol{\lambda}_{\mathbf{u}}$ and $\boldsymbol{\Theta}$, the coupling tensor \mathbf{W} , and further tensors \mathcal{O} of higher order in \mathbf{q} , as indicated by the arguments \mathbf{q} and $\mathbf{q} \otimes \mathbf{q}$. \mathbf{W}^\dagger is the symbol for the Hermitian conjugate tensor of \mathbf{W} . We now calculate the elastic eigenmodes, but only to the lowest order in \mathbf{q} . With the unitary transformation

$$U = \begin{pmatrix} \mathbf{1} & -\mathbf{W}(\mathbf{q})\boldsymbol{\Theta}^{-1} \\ [\mathbf{W}(\mathbf{q})\boldsymbol{\Theta}^{-1}]^\dagger & \mathbf{1} \end{pmatrix} , \quad (26)$$

the coupling tensor $\mathbf{W}(\mathbf{q})$ is eliminated. The variables

$\mathbf{u}(\mathbf{q})$ and $\Delta\tilde{\boldsymbol{\mu}}(\mathbf{q})$ do not mix, whereas the elastic tensor $\boldsymbol{\lambda}_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q})$ is renormalized:

$$\boldsymbol{\lambda}(\mathbf{q} \otimes \mathbf{q}) = \boldsymbol{\lambda}_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q}) - \mathbf{W}(\mathbf{q})\boldsymbol{\Theta}^{-1}\mathbf{W}^\dagger(\mathbf{q}) . \quad (27)$$

The elastic free energy is then

$$\begin{aligned} \delta f & \approx \frac{V}{2} \sum_{\mathbf{q}} \begin{pmatrix} \mathbf{u}(\mathbf{q}) \\ \Delta\tilde{\boldsymbol{\mu}}(\mathbf{q}) \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{\lambda}(\mathbf{q} \otimes \mathbf{q}) & \mathcal{O}(\mathbf{q} \otimes \mathbf{q}) \\ \mathcal{O}(\mathbf{q} \otimes \mathbf{q}) & \boldsymbol{\Theta} \end{pmatrix} \\ & \times \begin{pmatrix} \mathbf{u}^*(\mathbf{q}) \\ \Delta\tilde{\boldsymbol{\mu}}^*(\mathbf{q}) \end{pmatrix} . \end{aligned} \quad (28)$$

A calculation of the eigenvalues of the new block matrix, using the definition for the determinant, shows that a further diagonalization can be performed for $\lambda(\mathbf{q} \otimes \mathbf{q})$ and Θ separately. So we can state the following important result: The hydrodynamical modes of the cubic blue phases are identical to the displacement modes for small wave vectors \mathbf{q} . Thus we can restrict ourselves to the displacement field $\mathbf{u}(\mathbf{r})$, as the hydrodynamical variable, to describe their dynamics.

Finally, we present the full form of the elastic tensor $\lambda(\mathbf{q} \otimes \mathbf{q})$. Restriction to $m = 2$ modes for the undeformed order parameter in the elastic free energy $\delta f_{\mathbf{u},\mathbf{u}}$ of Eq. (22) leads to

$$\delta f_{\mathbf{u},\mathbf{u}} = \frac{V}{2} \sum_{\mathbf{q}} \lambda_{\mathbf{u}} \cdot [\mathbf{q} \otimes \mathbf{q} \otimes \mathbf{u}(\mathbf{q}) \otimes \mathbf{u}^*(\mathbf{q})] \quad (29)$$

with

$$\lambda_{\mathbf{u}} = \sum_{\mathbf{k}_R} |\mu_2(\mathbf{k}_R)|^2 \sum_{\mathcal{P}(\mathbf{k}_R)} \frac{\kappa^2}{2q_c^2} \left[\mathbf{1} \otimes \mathbf{S}\mathbf{k}_R \otimes \mathbf{S}\mathbf{k}_R + \frac{c_2}{c_1} \mathbf{S}\mathbf{m}_R \otimes \mathbf{S}\mathbf{m}_R^* \otimes \mathbf{S}\mathbf{k}_R \otimes \mathbf{S}\mathbf{k}_R \right]. \quad (30)$$

$\lambda_{\mathbf{u}}$ is a tensor of rank four, invariant under cubic point operations. Its general form is

$$\lambda_{\mathbf{u}} = \lambda_{\mathbf{u}} \mathbf{1} \otimes \mathbf{1} + (\lambda_{\mathbf{u}} + \lambda'_{\mathbf{u}}) \mathbf{1}_S^{(4)} + \lambda_{K,\mathbf{u}} \sum_{i=1}^3 \mathbf{P}_i \otimes \mathbf{P}_i, \quad (31)$$

$\mathbf{1}_S^{(4)}$ denotes the symmetrized identity tensor of rank four with the components

$$[\mathbf{1}_S^{(4)}]_{ijkl} = \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \quad (32)$$

and $\mathbf{P}_i = \mathbf{e}_i \otimes \mathbf{e}_i$, a projector on one of the fourfold axes whose directions are given by the unit vectors \mathbf{e}_i . $\lambda_{\mathbf{u}}$ and $\lambda'_{\mathbf{u}}$ are Lamé's constants for an isotropic elastic solid. $\lambda_{K,\mathbf{u}}$ is the third elastic constant, allowed by cubic point symmetry and a measure of the elastic anisotropy. $\lambda_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q})$ follows from $\lambda_{\mathbf{u}}$ by contraction over the first two indices:

$$\begin{aligned} \lambda_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q}) &:= \lambda_{\mathbf{u}}(\mathbf{q}, \mathbf{q}, \cdot, \cdot) \\ &= \lambda_{\mathbf{u}} q^2 \mathbf{1} + (\lambda_{\mathbf{u}} + \lambda'_{\mathbf{u}}) \mathbf{q} \otimes \mathbf{q} \\ &\quad + \lambda_{K,\mathbf{u}} \sum_{i=1}^3 (\mathbf{P}_i \mathbf{q})^2 \mathbf{P}_i \end{aligned} \quad (33)$$

with the elastic constants

$$\lambda_{\mathbf{u}} = \frac{\kappa^2}{6} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R^2}{q_c^2}, \quad (34)$$

$$\lambda'_{\mathbf{u}} = -\frac{\kappa^2}{6} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R^2}{q_c^2} \left[1 + \frac{c_2}{c_1} f(\mathbf{k}_R) \right], \quad (35)$$

$$\lambda_{K,\mathbf{u}} = \frac{\kappa^2}{3} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R^2}{q_c^2} \frac{c_2}{c_1} f(\mathbf{k}_R). \quad (36)$$

Here the abbreviation

$$f(\mathbf{k}_R) = \frac{h^2 k^2 + h^2 l^2 + k^2 l^2}{(h^2 + k^2 + l^2)^2} \quad (37)$$

is used, where (hkl) are the components of \mathbf{k}_R .

In the following two subsections we study the corrections to the elastic tensor $\lambda_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q})$ for two characteristic deformation modes in the blue phases.

C. Coupling to rotational modes

Local rotations of the — in general biaxial — order parameter $\boldsymbol{\mu}(\mathbf{r})$ are a generalization of director modes in the nematic phase. They are favored by the free energy (12) in the low chirality limit $\kappa \rightarrow 0$ because only the elastic terms contribute to rotations of $\boldsymbol{\mu}(\mathbf{r})$. We introduce the deformed order parameter field $\boldsymbol{\mu}_d(\mathbf{r})$ as a displacement of $\boldsymbol{\mu}(\mathbf{r})$ followed by a rotation:

$$\boldsymbol{\mu}(\mathbf{r}) \longrightarrow \boldsymbol{\mu}_d(\mathbf{r}) = \mathbf{R}[\mathbf{S}(\mathbf{r})] \boldsymbol{\mu}(\mathbf{r} - \mathbf{u}(\mathbf{r})) \mathbf{R}^{-1}[\mathbf{S}(\mathbf{r})]. \quad (38)$$

$\mathbf{R}[\mathbf{S}(\mathbf{r})]$ is the rotation operator

$$\mathbf{R}[\mathbf{S}(\mathbf{r})] \approx \mathbf{1} + \boldsymbol{\varphi}(\mathbf{r}) \times, \quad [\boldsymbol{\varphi}(\mathbf{r}) \times]_{ij} = \varepsilon_{ikj} \varphi_k \quad (39)$$

in which we have restricted ourselves to small rotations [18]. $\boldsymbol{\varphi}(\mathbf{r})$ denotes a vector whose direction and modulus are given by the local rotation axis and the angle of rotation, respectively. Up to first order it follows from Eq. (38) that

$$\boldsymbol{\mu}_d(\mathbf{r}) \approx \boldsymbol{\mu}(\mathbf{r}) - [\mathbf{u}(\mathbf{r}) \cdot \nabla] \boldsymbol{\mu}(\mathbf{r}) + [\boldsymbol{\varphi}(\mathbf{r}) \times, \boldsymbol{\mu}(\mathbf{r})]. \quad (40)$$

The symbol $[\cdot, \cdot]$ stands for a commutator. Using the Fourier expansion

$$\boldsymbol{\varphi}(\mathbf{r}) = \sum_{\mathbf{q}} \boldsymbol{\varphi}(\mathbf{q}) \exp(i\mathbf{q} \cdot \mathbf{r}), \quad (41)$$

we finally obtain the Fourier coefficients of the rotational modes

$$\delta \tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) = [\boldsymbol{\varphi}(\mathbf{q}) \times, \boldsymbol{\mu}(\mathbf{k})]. \quad (42)$$

They are used to calculate $\delta f_{\boldsymbol{\varphi},\mathbf{u}}$ and $\delta f_{\boldsymbol{\varphi},\boldsymbol{\varphi}}$ from Eqs. (23) and (24), the index $\delta \tilde{\boldsymbol{\mu}}$ being replaced by $\boldsymbol{\varphi}$. Restriction to $m = 2$ modes in the undeformed order parameter field yields the result

$$\delta f_{\boldsymbol{\varphi},\boldsymbol{\varphi}} = V c_{\boldsymbol{\varphi}} \sum_{\mathbf{q}} |\boldsymbol{\varphi}(\mathbf{q})|^2, \quad (43)$$

$$\delta f_{\boldsymbol{\varphi},\mathbf{u}} = -i V c_{\boldsymbol{\varphi}} \sum_{\mathbf{q}} \boldsymbol{\varepsilon}_{LV} \cdot [\mathbf{q} \otimes \boldsymbol{\varphi}(\mathbf{q}) \otimes \mathbf{u}^*(\mathbf{q})], \quad (44)$$

with the constant

$$c_{\boldsymbol{\varphi}} = \frac{\kappa^2}{6} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R}{q_c} \left[1 + \frac{c_2}{2c_1} \frac{k_R}{q_c} \right]. \quad (45)$$

$\delta f_{\boldsymbol{\varphi},\boldsymbol{\varphi}}$ and $\delta f_{\boldsymbol{\varphi},\mathbf{u}}$ contain the only tensors invariant under cubic point operations: the unit tensor $\mathbf{1}$ of second rank and the Levi-Civita tensor $\boldsymbol{\varepsilon}_{LV}$. The difficult task was to

determine the constant c_φ . In our compact formulation (25) of δf we have to replace $\Delta\tilde{\boldsymbol{\mu}}(\mathbf{q})$ by $\boldsymbol{\varphi}(\mathbf{q})$ and we can identify the coupling tensor $\mathbf{W}_{\varphi,\mathbf{u}}$ and $\boldsymbol{\Theta}_{\varphi,\varphi}$ with

$$\mathbf{W}_{\varphi,\mathbf{u}} = \mathbf{W}_{\varphi,\mathbf{u}}^\dagger = -ic_\varphi \boldsymbol{\varepsilon}_{LV}(\mathbf{q}, \cdot, \cdot), \quad (46)$$

$$\boldsymbol{\Theta}_{\varphi,\varphi} = 2c_\varphi \mathbf{1}. \quad (47)$$

The correction $\lambda_\varphi(\mathbf{q} \otimes \mathbf{q})$ to $\lambda_u(\mathbf{q} \otimes \mathbf{q})$ follows from Eq. (27):

$$\begin{aligned} \lambda_\varphi(\mathbf{q} \otimes \mathbf{q}) &= -\mathbf{W}_{\varphi,\mathbf{u}}(\mathbf{q}) \boldsymbol{\Theta}_{\varphi,\varphi}^{-1} \mathbf{W}_{\varphi,\mathbf{u}}^\dagger(\mathbf{q}) \\ &= \lambda_\varphi q^2 \mathbf{1} + (\lambda_\varphi + \lambda'_\varphi) \mathbf{q} \otimes \mathbf{q} \end{aligned} \quad (48)$$

with the elastic constants

$$\lambda_\varphi = -\frac{1}{2}c_\varphi, \quad \lambda'_\varphi = c_\varphi, \quad \lambda_{K,\varphi} = 0. \quad (49)$$

$\lambda_\varphi(\mathbf{q} \otimes \mathbf{q})$ contains only isotropic terms.

$\boldsymbol{\Theta}_{\varphi,\varphi}$ does not vanish for $\mathbf{q} \rightarrow \mathbf{0}$. Therefore the rotational modes are not hydrodynamical modes, in contrast to the director modes of the nematic phase. The reason is the lattice structure of the cubic blue phases, which leads to inequivalent directions, the crystallographic axes. Changing the orientation of $\boldsymbol{\mu}(\mathbf{r})$ relative to these directions requires energy. We can compare the situation to ferromagnetic cubic crystals. There are the directions of easy magnetization, for example, the four-fold axes. Changing the orientation of the magnetization needs the *magnetocrystalline energy* [19].

D. Coupling to $m = 2$ modes

Modes of helicity $m = 2$ are characteristic for the structure of the blue phases as explained in Sec. II A. We therefore introduce further deformation modes by restricting the Fourier coefficient $\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q})$ to the helicity $m = 2$:

$$\delta\tilde{\boldsymbol{\mu}}(\mathbf{k} + \mathbf{q}) = \delta\tilde{\boldsymbol{\mu}}_2(\mathbf{k} + \mathbf{q}) \mathbf{M}_2(\mathbf{k} + \mathbf{q}). \quad (50)$$

With the approximation

$$\mathbf{M}_2(\mathbf{k} + \mathbf{q}) \approx \mathbf{M}_2(\mathbf{k}), \quad (51)$$

due to our limitation $q \ll k$, it follows that

$$\delta f_{2,2} \approx \frac{V}{2} \sum_{\mathbf{q}} \sum_{\mathbf{k}, \mathbf{k}'} [\boldsymbol{\Theta}_{2,2}]_{\mathbf{k}, \mathbf{k}'} \delta\tilde{\boldsymbol{\mu}}_2(\mathbf{k} + \mathbf{q}) \delta\tilde{\boldsymbol{\mu}}_2^*(\mathbf{k}' + \mathbf{q}), \quad (52)$$

$$\begin{aligned} \delta f_{2,u} \approx \frac{V}{2} \sum_{\mathbf{q}} \sum_{j=1}^3 \sum_{\mathbf{k}} \left\{ [\mathbf{W}_{2,u}(\mathbf{q})]_{j,\mathbf{k}} u_j(\mathbf{q}) \delta\tilde{\boldsymbol{\mu}}_2^*(\mathbf{k} + \mathbf{q}) \right. \\ \left. + [\mathbf{W}_{2,u}^\dagger(\mathbf{q})]_{\mathbf{k},j} \delta\tilde{\boldsymbol{\mu}}_2(\mathbf{k} + \mathbf{q}) u_j^*(\mathbf{q}) \right\} \end{aligned} \quad (53)$$

with the components of the tensors $\boldsymbol{\Theta}_{2,2}$ and $\mathbf{W}_{2,u}(\mathbf{q})$ given by

$$\begin{aligned} [\boldsymbol{\Theta}_{2,2}]_{\mathbf{k}, \mathbf{k}'} &= \left(\frac{t}{2} + \frac{\kappa^2}{2q_c^2} k^2 - \frac{\kappa^2}{q_c} k \right) \delta_{\mathbf{k}, \mathbf{k}'} - 6\sqrt{6} \text{tr}[\mathbf{M}_2(\mathbf{k}) \mathbf{M}_2(-\mathbf{k}') \mathbf{M}_2(\mathbf{k}' - \mathbf{k})] \mu_2(\mathbf{k}' - \mathbf{k}) \\ &+ \sum_{\mathbf{k}''} \left\{ 8\text{tr}[\mathbf{M}_2(\mathbf{k}'') \mathbf{M}_2(\mathbf{k})] \text{tr}[\mathbf{M}_2(\mathbf{k}' - \mathbf{k} - \mathbf{k}'') \mathbf{M}_2(-\mathbf{k}')] \right. \\ &\left. + 4\text{tr}[\mathbf{M}_2(\mathbf{k}) \mathbf{M}_2(-\mathbf{k}')] \text{tr}[\mathbf{M}_2(\mathbf{k}'') \mathbf{M}_2(\mathbf{k}' - \mathbf{k} - \mathbf{k}'')] \right\} \mu_2(\mathbf{k}'') \mu_2(\mathbf{k}' - \mathbf{k} - \mathbf{k}''), \end{aligned} \quad (54)$$

$$[\mathbf{W}_{2,u}(\mathbf{q})]_{j,\mathbf{k}} = -i \frac{\kappa^2}{q_c^2} \mu_2(\mathbf{k}) \left(1 - \frac{q_c}{k} \right) \mathbf{k} \cdot \mathbf{q} k_j. \quad (55)$$

The indices of the components $[\boldsymbol{\Theta}_{2,2}]_{\mathbf{k}, \mathbf{k}'}$, \mathbf{k} and \mathbf{k}' , cover all the \mathbf{k} vectors which we take into account for the deformation modes. Like for the displacement and rotational modes, we consider all \mathbf{k} vectors which are used to construct the undeformed order parameter field. If the amplitudes $\mu_2(\mathbf{k}' - \mathbf{k})$ and $\mu_2(\mathbf{k}' - \mathbf{k} - \mathbf{k}'')$ belong to higher stars, they are chosen to be zero in agreement with the results of GHS [17]. The index j of $[\mathbf{W}_{2,u}(\mathbf{q})]_{j,\mathbf{k}}$ ranges from 1 to 3, according to the three components of the displacement vector \mathbf{u} , while the index \mathbf{k} is handled as explained above. Again we introduce a column vector

$$\Delta\tilde{\boldsymbol{\mu}}_2(\mathbf{q}) = \begin{pmatrix} \delta\tilde{\boldsymbol{\mu}}_2(\mathbf{k}^{(1)} + \mathbf{q}) \\ \vdots \\ \delta\tilde{\boldsymbol{\mu}}_2(\mathbf{k}^{(n)} + \mathbf{q}) \end{pmatrix} \quad (56)$$

and write the elastic free energy in the compact form

$$\begin{aligned} \delta f \approx \frac{V}{2} \sum_{\mathbf{q}} \begin{pmatrix} \mathbf{u}(\mathbf{q}) \\ \Delta\tilde{\boldsymbol{\mu}}_2(\mathbf{q}) \end{pmatrix} \cdot \begin{pmatrix} \lambda_u(\mathbf{q} \otimes \mathbf{q}) & \mathbf{W}_{2,u}(\mathbf{q}) \\ \mathbf{W}_{2,u}^\dagger(\mathbf{q}) & \boldsymbol{\Theta}_{2,2} \end{pmatrix} \\ \times \begin{pmatrix} \mathbf{u}^*(\mathbf{q}) \\ \Delta\tilde{\boldsymbol{\mu}}_2^*(\mathbf{q}) \end{pmatrix}. \end{aligned} \quad (57)$$

The correction

$$\lambda_2(\mathbf{q} \otimes \mathbf{q}) = -\mathbf{W}_{2,u}(\mathbf{q}) \boldsymbol{\Theta}_{2,2}^{-1} \mathbf{W}_{2,u}^\dagger(\mathbf{q}) \quad (58)$$

of the elastic tensor involves too complex quantities for an analytic evaluation to be possible. Nonetheless $\lambda_2(\mathbf{q} \otimes \mathbf{q})$ has to reflect the cubic point symmetry of the blue phases and therefore has the form

$$\begin{aligned} \lambda_2(\mathbf{q} \otimes \mathbf{q}) &= \lambda_2 q^2 \mathbf{1} + (\lambda_2 + \lambda'_2) \mathbf{q} \otimes \mathbf{q} \\ &+ \lambda_{K,2} \sum_{i=1}^3 (\mathbf{P}_i \mathbf{q})^2 \mathbf{P}_i. \end{aligned} \quad (59)$$

The elastic constants λ_2, λ'_2 , and $\lambda_{K,2}$ are determined numerically for each set of parameters κ and t and the corresponding amplitudes $\mu_2(\mathbf{k})$.

E. Coupling to rotational and $m = 2$ modes

In a last step we admit both the rotational and $m = 2$ modes as deformations in addition to the displacement modes and introduce the Fourier coefficient

$$\delta\tilde{\mu}(\mathbf{k} + \mathbf{q}) = [\varphi(\mathbf{q}) \times, \mu(\mathbf{k})] + \delta\tilde{\mu}_2(\mathbf{k} + \mathbf{q}) M_2(\mathbf{k} + \mathbf{q}) . \quad (60)$$

The different contributions to the elastic free energy (21)

$$\delta f = \frac{V}{2} \sum_{\mathbf{q}} \begin{pmatrix} \mathbf{u}(\mathbf{q}) \\ \varphi(\mathbf{q}) \\ \Delta\tilde{\mu}_2(\mathbf{q}) \end{pmatrix} \cdot \begin{pmatrix} \lambda_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q}) & W_{\varphi, \mathbf{u}}(\mathbf{q}) & W_{2, \mathbf{u}}(\mathbf{q}) \\ W_{\varphi, \varphi}^\dagger(\mathbf{q}) & O(\mathbf{q}) & O(\mathbf{q}) \\ W_{2, \mathbf{u}}^\dagger(\mathbf{q}) & O(\mathbf{q}) & \Theta_{2,2} \end{pmatrix} \begin{pmatrix} \mathbf{u}^*(\mathbf{q}) \\ \varphi^*(\mathbf{q}) \\ \Delta\tilde{\mu}_2^*(\mathbf{q}) \end{pmatrix} . \quad (64)$$

The corrected elastic tensor for the displacement modes follows from

$$\lambda(\mathbf{q} \otimes \mathbf{q}) = \lambda_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q}) - (W_{\varphi, \mathbf{u}}(\mathbf{q}) W_{2, \mathbf{u}}(\mathbf{q})) \times \begin{pmatrix} \Theta_{\varphi, \varphi} & O(\mathbf{q}) \\ O(\mathbf{q}) & \Theta_{2,2} \end{pmatrix}^{-1} \begin{pmatrix} W_{\varphi, \mathbf{u}}^\dagger(\mathbf{q}) \\ W_{2, \mathbf{u}}^\dagger(\mathbf{q}) \end{pmatrix} . \quad (65)$$

For the inverse block matrix we need only the zeroth order in \mathbf{q} :

$$\begin{pmatrix} \Theta_{\varphi, \varphi} & O(\mathbf{q}) \\ O(\mathbf{q}) & \Theta_{2,2} \end{pmatrix}^{-1} = \begin{pmatrix} \Theta_{\varphi, \varphi}^{-1} & O(\mathbf{q}) \\ O(\mathbf{q}) & \Theta_{2,2}^{-1} \end{pmatrix} . \quad (66)$$

The corrections turn out to be additive:

$$\lambda(\mathbf{q} \otimes \mathbf{q}) = \lambda_{\mathbf{u}}(\mathbf{q} \otimes \mathbf{q}) + \lambda_{\varphi}(\mathbf{q} \otimes \mathbf{q}) + \lambda_2(\mathbf{q} \otimes \mathbf{q}) . \quad (67)$$

F. Symmetry aspects

In this last subsection the symmetry properties of the elastic free energy of the displacement modes are investigated. This will help us to find eigenmodes with a special polarization, say, transverse and longitudinal modes. The elastic free energy reads

$$\delta f_{\mathbf{u}, \mathbf{u}} = \frac{V}{2} \sum_{\mathbf{q}} \lambda(\mathbf{q} \otimes \mathbf{q}) \cdot [\mathbf{u}(\mathbf{q}) \otimes \mathbf{u}^*(\mathbf{q})] . \quad (68)$$

The elastic tensor $\lambda(\mathbf{q} \otimes \mathbf{q})$ follows from a tensor λ of rank four with cubic point symmetry

$$\lambda = 2\lambda \mathbf{1}_S^{(4)} + \lambda' \mathbf{1} \otimes \mathbf{1} + \lambda_K \sum_{i=1}^3 \mathbf{P}_i \otimes \mathbf{P}_i \quad (69)$$

are

$$\delta f_{\delta\tilde{\mu}, \mathbf{u}} \approx \delta f_{\varphi, \mathbf{u}} + \delta f_{2, \mathbf{u}} , \quad (61)$$

$$\delta f_{\delta\tilde{\mu}, \delta\tilde{\mu}} \approx \delta f_{\varphi, \varphi} + \delta f_{2,2} + \delta f_{\varphi, 2} . \quad (62)$$

All terms are known except for $\delta f_{\varphi, 2}$, which describes the coupling between rotational and $m = 2$ modes. When the undeformed order parameter is restricted to $m = 2$ modes it contains \mathbf{q} only in first order:

$$\delta f_{\varphi, 2} = O(\mathbf{q}) . \quad (63)$$

The elastic free energy is then

by contraction:

$$\begin{aligned} \lambda(\mathbf{q} \otimes \mathbf{q}) &:= \lambda(\mathbf{q}, \cdot, \mathbf{q}, \cdot) \\ &= \lambda q^2 \mathbf{1} + (\lambda + \lambda') \mathbf{q} \otimes \mathbf{q} + \lambda_K \sum_{i=1}^3 (\mathbf{P}_i \mathbf{q})^2 \mathbf{P}_i . \end{aligned} \quad (70)$$

Note that we have chosen slightly different definitions compared to Eqs. (31) and (33) to get the same form of λ as in the linear elastic theory of solids. λ is invariant under operations of the cubic point group O_h , which also contains reflections, in contrast to O , the point group of the blue phases. In a symbolic notation this reads

$$\mathbf{S} \lambda = \lambda \mathbf{S} , \quad \mathbf{S} \in O_h . \quad (71)$$

The eigenvectors of $\lambda(\mathbf{q} \otimes \mathbf{q})$ are calculated as usual:

$$\lambda(\mathbf{q} \otimes \mathbf{q}) \mathbf{u}(\mathbf{q}) = \lambda_{\text{eff}} q^2 \mathbf{u}(\mathbf{q}) , \quad (72)$$

where λ_{eff} is the effective elastic constant of the eigenmode $\{\mathbf{q}, \mathbf{u}(\mathbf{q})\}$. We investigate the rotated eigenvalue problem

$$\mathbf{S}[\lambda(\mathbf{q} \otimes \mathbf{q}) \mathbf{u}(\mathbf{q})] = \lambda_{\text{eff}} q^2 \mathbf{S} \mathbf{u}(\mathbf{q}) . \quad (73)$$

In components one can show, using Eq. (71), that

$$\lambda(\mathbf{S} \mathbf{q} \otimes \mathbf{S} \mathbf{q}) \mathbf{S} \mathbf{u}(\mathbf{q}) = \lambda_{\text{eff}} q^2 \mathbf{S} \mathbf{u}(\mathbf{q}) . \quad (74)$$

The last equation can also be proven by considering λ , \mathbf{q} , and \mathbf{u} as geometrical objects and by applying the rotation step by step:

$$\begin{aligned} \mathbf{S}[\lambda(\mathbf{q} \otimes \mathbf{q}) \mathbf{u}(\mathbf{q})] &= \mathbf{S}[\lambda(\mathbf{q} \otimes \mathbf{q})] \mathbf{S} \mathbf{u}(\mathbf{q}) \\ &= [\mathbf{S} \lambda](\mathbf{S} \mathbf{q} \otimes \mathbf{S} \mathbf{q}) \mathbf{S} \mathbf{u}(\mathbf{q}) . \end{aligned} \quad (75)$$

In addition to the eigenmode $\{\mathbf{q}, \mathbf{u}(\mathbf{q})\}$ we have found a second one $\{\mathbf{S} \mathbf{q}, \mathbf{S} \mathbf{u}(\mathbf{q})\}$ with the same effective elastic constant.

TABLE I. Wave vectors \mathbf{q} with a nontrivial little group $\mathcal{K}(\mathbf{q})$ and their possible longitudinal and transverse eigenmodes. The eigenvector $\mathbf{u}(\mathbf{q})$, the effective elastic constant, and the degree d of degeneracy are listed. The polarization p of the eigenmodes is abbreviated by l and t for longitudinal and transverse, respectively.

$\mathcal{K}(\mathbf{q})$	\mathbf{q}	p	$\mathbf{u}(\mathbf{q})$	λ_{eff}	d
C_{4v}	\mathbf{e}_1	l	\mathbf{e}_1	$2\lambda + \lambda' + \lambda_K$	1
		t	$\cos \Phi \mathbf{e}_2 + \sin \Phi \mathbf{e}_3$	λ	2
C_{3v}	$(\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3)/\sqrt{3}$	l	$(\mathbf{e}_1 + \mathbf{e}_2 + \mathbf{e}_3)/\sqrt{3}$	$2\lambda + \lambda' + \lambda_K/3$	1
		t	\mathbf{t}^a	$\lambda + \lambda_K/3$	2
C_{2v}	$(\mathbf{e}_1 + \mathbf{e}_2)/\sqrt{2}$	l	$(\mathbf{e}_1 + \mathbf{e}_2)/\sqrt{2}$	$2\lambda + \lambda' + \lambda_K/2$	1
		t	$(\mathbf{e}_1 - \mathbf{e}_2)/\sqrt{2}$	$\lambda + \lambda_K/2$	1
		t	\mathbf{e}_3	λ	1
C_s	$\cos \Phi \mathbf{e}_1 + \sin \Phi \mathbf{e}_2$	t	\mathbf{e}_3	λ	1
C_s	$\cos \Phi (\mathbf{e}_1 - \mathbf{e}_2)/\sqrt{2} + \sin \Phi \mathbf{e}_3$	t	$(\mathbf{e}_1 + \mathbf{e}_2)/\sqrt{2}$	$\lambda + \cos^2 \Phi \lambda_K/2$	1

$${}^a \mathbf{t} = \cos \Phi (\mathbf{e}_1 - \mathbf{e}_2)/\sqrt{2} + \sin \Phi (\mathbf{e}_1 + \mathbf{e}_2 - 2\mathbf{e}_3)/\sqrt{6}.$$

We now restrict ourselves to special wave vectors \mathbf{q} invariant under a subgroup of O_h , the *little group* $\mathcal{K}(\mathbf{q})$ of \mathbf{q} . With $\mathbf{S}\mathbf{q} = \mathbf{q}$ we get, from Eq. (74),

$$\lambda(\mathbf{q} \otimes \mathbf{q}) \mathbf{S}\mathbf{u}(\mathbf{q}) = \lambda_{\text{eff}} q^2 \mathbf{S}\mathbf{u}(\mathbf{q}). \quad (76)$$

If $\mathbf{S}\mathbf{u}(\mathbf{q}) \neq \pm \mathbf{u}(\mathbf{q})$, there exists a second eigenmode $\{\mathbf{q}, \mathbf{S}\mathbf{u}(\mathbf{q})\}$ to the degenerate eigenvalue $\lambda_{\text{eff}} q^2$. Reducing this degeneracy leads to transverse and longitudinal eigenmodes. Consider, for example, \mathbf{q} parallel a fourfold axis. Applying all possible rotations to $\mathbf{u}(\mathbf{q})$ yields, in general, three linear independent eigenvectors. One can reduce the degeneracy if one chooses one eigenvector parallel to \mathbf{q} and a plane of eigenvectors perpendicular to \mathbf{q} . Then the effective elastic constants can easily be calculated using Eq. (70). In Table I we list all wave vectors \mathbf{q} with a nontrivial little group $\mathcal{K}(\mathbf{q})$ and their possible longitudinal and transverse eigenmodes. It shows the eigenvector $\mathbf{u}(\mathbf{q})$, the effective elastic constant, and the degree d of degeneracy. The polarization p of the eigenmodes is abbreviated by l and t for longitudinal and transverse, respectively. We will refer to this table when we discuss the elastic properties of the displacement modes in the next section.

III. DISCUSSION

The elastic constants for the displacement modes including all corrections are

$$\begin{aligned} \lambda &= \lambda_u + \lambda_\varphi + \lambda_2 \\ &= \frac{\kappa^2}{6} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R^2}{q_c^2} \left(1 - \frac{1}{2} \left[\frac{q_c}{k_R} + \frac{c_2}{2c_1} \right] \right) \\ &\quad + \lambda_2, \end{aligned} \quad (77)$$

$$\begin{aligned} \lambda' &= \lambda'_u + \lambda'_\varphi + \lambda'_2 \\ &= -\frac{\kappa^2}{6} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R^2}{q_c^2} \left(1 + \frac{c_2}{c_1} f(\mathbf{k}_R) \right. \\ &\quad \left. - \left[\frac{q_c}{k_R} + \frac{c_2}{2c_1} \right] \right) + \lambda'_2, \end{aligned} \quad (78)$$

$$\begin{aligned} \lambda_K &= \lambda_{K,u} + \lambda_{K,2} \\ &= \frac{\kappa^2}{3} \sum_{\mathbf{k}_R} N(\mathbf{k}_R) |\mu_2(\mathbf{k}_R)|^2 \frac{k_R^2}{q_c^2} \frac{c_2}{c_1} f(\mathbf{k}_R) + \lambda_{K,2}. \end{aligned} \quad (79)$$

The corrections due to the rotational modes are written in square brackets. All effective elastic constants of the longitudinal modes contain the combination $2\lambda + \lambda'$ (Table I). Together with Eqs. (77) and (78) we notice that λ_{eff} does not involve any correction from rotational modes.

The temperature behavior of the effective elastic constants $\lambda + \lambda_K/2$ and $2\lambda + \lambda' + \lambda_K/3$ of a transverse and a longitudinal mode, respectively, is shown in Figs. 1 and

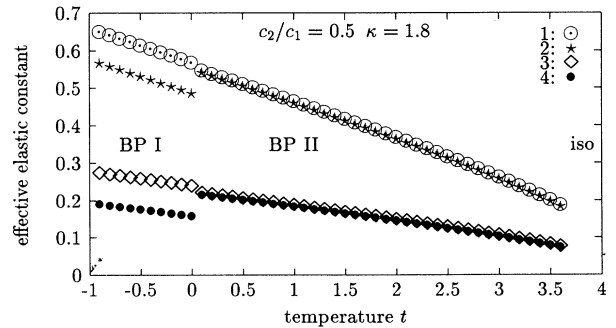


FIG. 1. Temperature behavior of the effective elastic constant $\lambda + \lambda_K/2$ with different corrections: 1, pure displacement mode; 2, with the correction from $m = 2$ modes; 3, with the correction from rotational modes; 4, with both corrections.

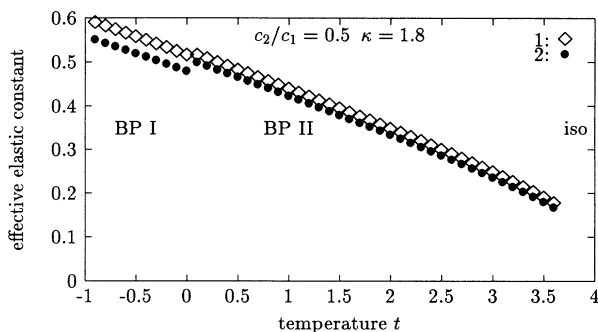


FIG. 2. Temperature behavior of the effective elastic constant $2\lambda + \lambda' + \lambda_K/3$ with different corrections: 1, pure displacement mode; 2, with the correction from $m = 2$ modes.

2. We choose $c_2/c_1 = 0.5$ [20,21] and $\kappa = 1.8$. The chirality is too high compared to the experiments [17], but for this value the phase sequence BP I – BP II – isotropic liquid is realized in the phase diagram of GHS [17]. The first curve shows the elastic constant for a pure displacement mode. The second and third (only in Fig. 1) take into account the corrections from $m = 2$ and rotational modes and the fourth (only in Fig. 1) includes both corrections. All curves have negative curvature. We notice that λ_{eff} is decreased by the additional deformations because these make the orientational pattern softer. The rotational modes lower the elastic constant by a factor 2.5. The corrections due to the $m = 2$ modes are much smaller. They are more significant in BP I and therefore λ_{eff} displays a jump to higher values at the phase transition BP I – BP II. The jump is not so large when we only look at λ because λ_K has different signs in BP I and II (Fig. 4). Since the rotational modes have no influence on the elastic constants of the longitudinal mode λ_{eff} is a factor 2 – 3 larger than for transverse modes. A chirality $\kappa \approx 0.8$ fits the experiments better [17]. Here we find values of 0.01 for the scaled elastic constants. Using the estimate $\beta^4/36\gamma^3 \approx 10^5$ ergs/cm³ for the scaling factor of the free-energy density [22] gives a value of 1000 ergs/cm³ for the elastic constant which is a factor 10^9 smaller than in normal crystals.

Figure 3 shows the chirality behavior of three effective elastic constants. Equations (77)–(79) suggest first that they are proportional to κ^2 , but the amplitudes $\mu_2(\mathbf{k}_R)$ depend also on κ , and this fact makes the situation more complicated. For λ we find by a fit to κ^e an exponent 3.44 instead of 2. The other elastic constants have larger exponents. We emphasize this point because Keyes [23] argues that the phase transition to the blue fog, which only appears at high chiralities, can be understood as a melting of the cubic blue phases at high chiralities. Therefore he calculates the mean square displacement

$$\frac{\sqrt{\langle \mathbf{u}^2 \rangle}}{b} \sim \sqrt{\frac{\kappa^3}{\lambda_{\text{eff}}}} \quad (80)$$

of the orientational pattern following the Landau-Peierls estimate. Assuming $\lambda_{\text{eff}} \sim \kappa^2$, the mean square displace-

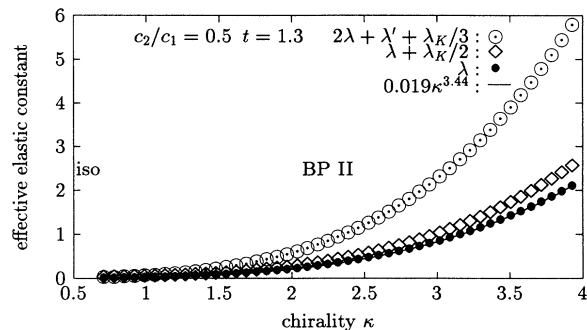


FIG. 3. Chirality behavior of effective elastic constants with both corrections. The solid line is a fit to κ^e .

ment increases with κ and the cubic blue phase melts according to the Lindemann melting criterion. Our result seems to contradict this argument because $\lambda_{\text{eff}} \sim \kappa^{3.44}$ leads to the opposite behavior. However, we have to remember that for increasing chirality the phase transition from the cubic blue phases to the isotropic liquid (the blue fog appears somewhere between) takes place at increasing temperatures, i.e., λ_{eff} decreases. If we take this point into account, there are hints that the mean square displacement shows the behavior as suggested. Recently calculations were presented which identify the blue fog as a liquid of purely cubic bond orientational order (cubic phase) which remains when the cubic blue phases melt [24].

Figure 4 shows the temperature behavior of λ' and λ_K relative to λ . The effective elastic constant λ must be positive to ensure the stability of the elastic free energy. The temperature dependence in the BP I is stronger than in the BP II. λ_K/λ , which is a measure of the anisotropy of the elastic properties of the cubic blue phases, has a negative sign in the BP I and changes sign at the phase transition. In the BP II the elastic anisotropy is larger, but nevertheless we can consider it as small.

Mechanical experiments were performed in polycrystalline materials to measure an average shear coefficient [25–27]. The values lie in the same order of magnitude proposed by the theory. The experimental curves of

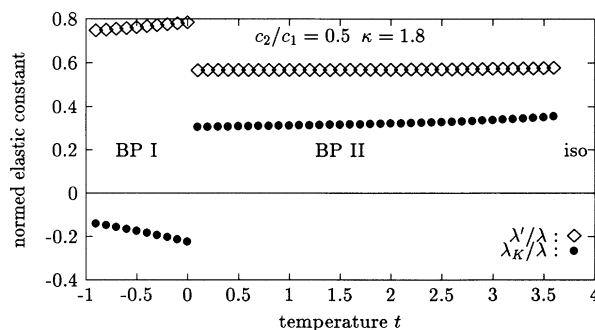


FIG. 4. Temperature behavior of the elastic constants λ' and λ_K relative to λ with both corrections.

Kleiman *et al.* [27] for the temperature behavior show a negative curvature as predicted by the theory, but in contrast to the results of Cladis *et al.* [26] and Clark *et al.* [25]. They also find a jump of the shear coefficient to higher values at the phase transition BP I – BP II.

A measurement of all three elastic constants requires a monocrystal of the cubic blue phases. The investigations by mechanical experiments are too rough because they destroy it. Therefore more subtle methods such as light scattering experiments must be used. Rakes and Keyes [28] measured the Debye-Waller factor [29] of different Bragg reflections. From these measurements one can only extract an estimate for the elastic constants because the Debye-Waller factor depends on the whole wave vector range of the excitation spectrum. Thus we have to rely on the scattering experiments of Marcus and Domberger. They can answer the interesting question about the amount of the elastic anisotropy in the cubic blue phases. Further, we have made an important step

towards understanding the dynamic behavior of the blue phases, the signs of which are hidden in the fluctuating scattered light intensity. With the displacement modes we have identified the origin of the fluctuations and we also know that the displacement field $\mathbf{u}(\mathbf{r})$ has to appear in the hydrodynamical equations, at least for small wave vectors \mathbf{q} . In the following paper [2] we will therefore use dynamical equations known from colloidal crystals [25] to analyze the dynamics of the displacement modes and of the light-scattering experiments. The same equations were also used by the groups which performed the above mentioned mechanical experiments [25–27].

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