Electrostriction tensor of the cubic blue phases: The role of amplitudes

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The phenomenological theory of electrostriction of the cubic blue phases is further developed by generalizing the model of rigid helices [H. Stark and H.-R. Trebin, Phys. Rev. A **44**, 2752 (1991)]. In the present approach not only the wave vectors of the cubic structures are distorted, but also the scalar amplitudes of the order parameter. By considering a full spectrum of the distortions exact formulas (within the Landau-deGennes approach) for the components of the electrostriction tensor are derived. Detailed results of the numerical calculations are compared with those obtained from the earlier model and with experimental data. It is shown that the electrostriction coefficients are strongly affected by the deformations of the amplitudes. Quantitatively, for low temperatures (or high chiralities) the contribution due to the deformed amplitudes exceeds that from the distortions of the wave vectors. The results are in good agreement with experimental data except for the region where the anomalous electrostriction of the blue phase I is observed. This proves that a correct description of the anomalous electrostriction is beyond the standard Landau-deGennes theory of the blue phases as suggested in our recent publication [L. Longa, M. Żelazna, H.-R. Trebin, and J. Mościcki, Phys. Rev. E **53**, 6067 (1996)]. [S1063-651X(98)02306-X]

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I. INTRODUCTION

The cubic blue phases (BPs) of chiral liquid crystals, i.e., the blue phase I [space group $O^8(\mathcal{I}4_132)$] and the blue phase II [space group $O^2(\mathcal{P}4_232)$] have been intensively studied both experimentally and theoretically [1]. In particular, a problem that has been receiving much attention is the influence of an external electric field on these BPs [2]. Depending on the field strength three different effects are observed. In a weak field the elementary cells of the cubic blue phases align with their [001] crystallographic axes parallel to the field direction [3]. With increasing field strength the unit cells are deformed continuously. This phenomenon, also known as *electrostriction*, can be observed from wavelength shifts of Bragg reflections in the visible spectral range [4]. For strong enough fields, the blue phases transform into structures with hexagonal or tetragonal symmetry [5] and, finally, for very high fields they unwind into a uniaxial nematic phase.

The most successful theoretical approach to the blue phases has been the application of the Landau-Ginzburg-deGennes (LGdG) theory of chiral liquid crystals. The theory was used by Grebel, Hornreich, and Shtrikman (GHS) [6] to explain universality of the phase diagrams of chiral liquid crystals, in particular the appearance and the structure of the blue phases. Later it was extended to include a uniform external field [7]. In this work we concentrate on the electrostriction of the cubic blue phases. Although calculations concerning this phenomenon were carried out by Dmitrienko [8] and a quantitative analysis was performed by Stark and Trebin [9] the theory is still at a preliminary stage. A major theoretical problem lies in calculating the elastic constants of the blue phases, which enter the formula for electrostriction. So far these calculations have been carried out using the so-called rigid helices model [9], in which only wave vectors of the reciprocal lattice were allowed to deform.

Our objective here is to develop a complete theory which is of the same accuracy as present calculations of the phase diagram for BPs [6]. This is achieved by extending the model of Stark and Trebin [9]. The extension takes into account both the effect of higher harmonics and the full spectrum of distortions of the alignment tensor. We successfully overcome theoretical problems in calculating elastic constants by combining symmetry considerations with algebraic techniques offered by MAPLE. The analysis seems important for we know that calculations involving phase diagrams of blue phases are extremely sensitive to approximations imposed on the $\boldsymbol{Q}$ tensor. Similar sensitivity is found for the electrostriction tensor.

The organization of the paper is as follows. Section II contains basic elements of the LGdG theory of blue phases. In Sec. III we define the electrostriction tensor $\boldsymbol{R}$ and show how it follows from our model. We also give details of the technique employed. Finally, Sec. IV contains the results and a discussion.

II. LANDAU-GINZBURG-DEGENNES THEORY OF CHOLESTERIC LIQUID CRYSTALS

The Landau-Ginzburg-deGennes theory of liquid crystals is an expansion in an order parameter which measures the degree of orientational order in the system under consideration. To identify the relevant order parameters one either refers to the orientational distribution function [10] or considers a response function of the system, e.g., a polarization $\boldsymbol{P}$ due to an applied electric field $\boldsymbol{E}$. Writing $\boldsymbol{P}$ as a power series in $\boldsymbol{E}$, we obtain

$$\boldsymbol{P}=\int d^3\boldsymbol{r}\{\boldsymbol{\chi}^{(1)}(\boldsymbol{r})+\boldsymbol{\chi}^{(2)}(\boldsymbol{r})\cdot\boldsymbol{E}+\boldsymbol{\chi}^{(3)}(\boldsymbol{r})\cdot(\boldsymbol{E}\otimes\boldsymbol{E})+\cdots\}. \tag{2.1}$$

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Due to the *local* D_∞ or D_2 symmetry of most liquid crystalline phases the permanent polarization $\boldsymbol{\chi}^{(1)}$ and the third-order nonlinear susceptibility $\boldsymbol{\chi}^{(3)}$ must vanish. Therefore, the leading term in the expansion (2.1) is the second rank tensor $\boldsymbol{\chi}^{(2)}(\boldsymbol{r})$. Its anisotropic part reads

$$Q_{ij}(\boldsymbol{r})=\chi_{ij}^{(2)}(\boldsymbol{r})-\frac{1}{3}\mathrm{Tr}[\boldsymbol{\chi}^{(2)}(\boldsymbol{r})]\delta_{ij}, \tag{2.2}$$

where

$$\mathrm{Tr}[\boldsymbol{\chi}^{(2)}(\boldsymbol{r})]=\sum_i \chi_{ii}^{(2)}(\boldsymbol{r}). \tag{2.3}$$

$\boldsymbol{Q}(\boldsymbol{r})$ in Eq. (2.2) is denoted *alignment tensor* and usually taken as a primary order parameter of liquid crystals. By construction, this symmetric and traceless quantity vanishes in the disordered phase. It becomes nonzero in any phase characterized by orientational order of liquid crystalline molecules. The LGdG free energy functional follows then from a series expansion in $\boldsymbol{Q}(\boldsymbol{r})$ and its derivatives. To the lowest nontrivial order it reads

$$\mathcal{F}_{\mathrm{LGdG}}[\boldsymbol{Q}(\boldsymbol{r})]=\mathcal{F}_{\mathrm{gradient}}[\boldsymbol{Q}(\boldsymbol{r}),\partial\boldsymbol{Q}(\boldsymbol{r})]+\mathcal{F}_{\mathrm{bulk}}[\boldsymbol{Q}(\boldsymbol{r})]. \tag{2.4}$$

In terms of dimensionless units introduced by Grebel *et al.* [6] the gradient and the bulk parts in Eq. (2.4) are

$$\mathcal{F}_{\mathrm{gradient}}[\boldsymbol{Q}(\boldsymbol{r}),\partial\boldsymbol{Q}(\boldsymbol{r})] =v^{-1}\int d^3\boldsymbol{r}\left\{\frac{1}{4}\kappa^2[\epsilon_{imn}Q_{nj,m}-Q_{ij}]^2+\rho[Q_{ij,j}]^2\right\}, \tag{2.5}$$

$$\mathcal{F}_{\mathrm{bulk}}[\boldsymbol{Q}(\boldsymbol{r})]=v^{-1}\int d^3\boldsymbol{r}\{\tau\,\mathrm{Tr}\,\boldsymbol{Q}^2-\sqrt{6}\,\mathrm{Tr}\,\boldsymbol{Q}^3+\mathrm{Tr}(\boldsymbol{Q}^2)^2\}. \tag{2.6}$$

Here κ denotes the chirality parameter, t the standard reduced temperature of the Landau theory, $\tau=\frac{1}{4}(t-\kappa^2)$ a renormalized reduced temperature, ρ the ratio of elastic constants, and v the volume of the cubic unit cell [6]. That is, the theory has only three characteristic parameters: t, κ, and ρ.

The global minimization of the free energy (2.4) at an arbitrary point in the (κ,t) plane turns out to be extremely difficult and is still unsolved. A source of this difficulty is the presence of the chiral term (proportional to ϵ_{imn}) in Eq. (2.5). It causes the bulk and the gradient free energy to favor different structures. Thus the cubic blue phases, which emerge from the LGdG theory, are an example of a *frustrated system*, i.e., a system where the condition of a local energetic minimum cannot be extended globally. Their structures arrive as a kind of compromise.

In all practical calculations based on the LGdG free energy it is convenient to parametrize the order parameter $\boldsymbol{Q}$ with respect to the symmetry of the system studied. Since we shall be interested in periodic structures of cubic symmetry we expand $\boldsymbol{Q}(\boldsymbol{r})$ into a Fourier series:

$$\boldsymbol{Q}(\boldsymbol{r})=\sum_{*\boldsymbol{k}}\frac{1}{\sqrt{N_{*\boldsymbol{k}}}}\left\{\sum_{\boldsymbol{k}\in *\boldsymbol{k}}\boldsymbol{Q}(\boldsymbol{k})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}\right\}, \tag{2.7}$$

where

$$\boldsymbol{Q}(\boldsymbol{k})=\frac{1}{v}\int d^3\boldsymbol{r}\boldsymbol{Q}(\boldsymbol{r})e^{-i\boldsymbol{k}\cdot\boldsymbol{r}}. \tag{2.8}$$

The first sum in the expansion (2.7) runs over different stars of $\boldsymbol{k}$ vectors from the reciprocal cubic lattice and the second sum over all $N_{*\boldsymbol{k}}$ members of the star $*\boldsymbol{k}$. The coefficients $\boldsymbol{Q}(\boldsymbol{k})$ are traceless second-rank tensors and can further be represented in the basis of spherical tensors with momentum $L=2$. Explicitly

$$\boldsymbol{Q}(\boldsymbol{k})=\sum_{m=-2}^{2}Q_m(\boldsymbol{k})\boldsymbol{M}_m(\boldsymbol{k}), \tag{2.9}$$

where m labels different *helicity modes*. The basis matrices $\boldsymbol{M}_m(\boldsymbol{k})$ are defined separately for each wave vector $\boldsymbol{k}$. With the help of a right-handed local system of orthonormal unit vectors $\{\hat{\boldsymbol{\xi}},\hat{\boldsymbol{\eta}},\hat{\boldsymbol{k}}=\boldsymbol{k}/|\boldsymbol{k}|\}$, where $\hat{\boldsymbol{k}}$ defines the local $\hat{\boldsymbol{z}}$ axis, the basis matrices read

$$\boldsymbol{M}_0(\boldsymbol{k})=\frac{1}{\sqrt{6}}\{3\hat{\boldsymbol{k}}\otimes\hat{\boldsymbol{k}}-\mathbf{1}\},$$

$$\boldsymbol{M}_{\pm1}(\boldsymbol{k})=\pm\frac{1}{2}\{(\hat{\boldsymbol{\xi}}\pm i\hat{\boldsymbol{\eta}})\otimes\hat{\boldsymbol{k}}+\hat{\boldsymbol{k}}\otimes(\hat{\boldsymbol{\xi}}\pm i\hat{\boldsymbol{\eta}})\}, \tag{2.10}$$

$$\boldsymbol{M}_{\pm2}(\boldsymbol{k})=\frac{1}{2}\{(\hat{\boldsymbol{\xi}}\pm i\hat{\boldsymbol{\eta}})\otimes(\hat{\boldsymbol{\xi}}\pm i\hat{\boldsymbol{\eta}})\}.$$

In the high chirality limit, which was extensively studied by GHS [6], the quadratic part of the free energy (2.4) can be minimized separately. In particular, it is found that for $\kappa>0$ only the $m=2$ helicity mode of $\boldsymbol{Q}$ is important. This rules out the elastic constant ρ from the free energy and simplifies the order parameter expansion. Now it reads

$$\boldsymbol{Q}(\boldsymbol{r})=\sum_{*\boldsymbol{k}}\frac{1}{\sqrt{N_{*\boldsymbol{k}}}}\left\{\sum_{\boldsymbol{k}\in *\boldsymbol{k}}\boldsymbol{Q}_2(\boldsymbol{k})\boldsymbol{M}_2(\boldsymbol{k})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}\right\}. \tag{2.11}$$

A detailed analysis of the cholesteric phase diagrams further shows that for the blue phase II only two symmetry-allowed amplitudes need to be considered in Eq. (2.11), while for the blue phase I the three lowest-lying states are relevant [6]. A purpose of this paper is to carry out calculations of the electrostriction tensor with the same accuracy as that for the phase diagrams, and without any further approximations.

III. ELECTROSTRICTION OF THE CUBIC BLUE PHASES

The phenomenon of electrostriction can be viewed as a competition between electric and elastic forces. The emerging structure results from a continuous deformation of the zero-field state. This distortion can be described by the strain tensor

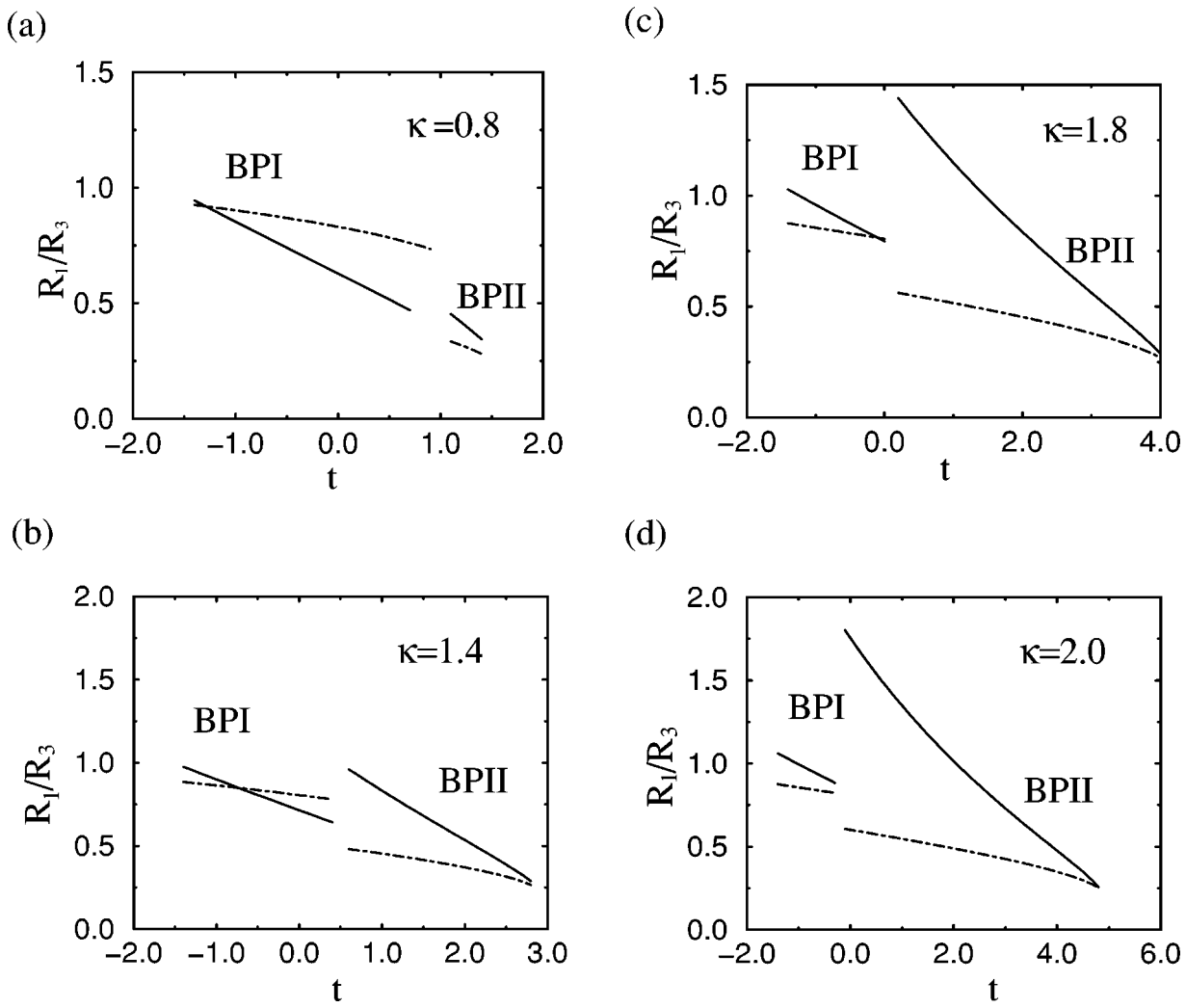

FIG. 1. The ratio R_1/R_3 of the independent components of the electrostriction tensor for four different chiralities: (a) $\kappa=0.8$, (b) $\kappa=1.4$, (c) $\kappa=1.8$, and (d) $\kappa=2.0$. The dashed and the solid lines correspond to the model without and with the deformation of scalar amplitudes, respectively.

$$\varepsilon_{ij}=\frac{1}{2}(\partial_i v_j+\partial_j v_i), \tag{3.1}$$

where the displacement vector $\boldsymbol{v}$ represents the shift of the lattice points due to the deformation. Since the unit cells of the blue phases are defined by a *complex orientational pattern* and *not* by a positional order of the molecules, this shift cannot be associated with a molecular motion.

The excess free energy due to the distortion reads

$$\mathcal{F}_{\text{distortion}}=\underbrace{\frac{1}{2}\boldsymbol{\lambda}\cdot(\boldsymbol{\varepsilon}\otimes\boldsymbol{\varepsilon})}_{\mathcal{F}_{\text{elastic}}}\underbrace{-\frac{\delta'}{8\pi}\boldsymbol{\epsilon}\cdot(\boldsymbol{E}\otimes\boldsymbol{E})}_{\mathcal{F}_{\text{electric}}}, \tag{3.2}$$

where $\boldsymbol{\lambda}$ is the tensor of elastic constants, δ' relates the external and internal electric field, and $\boldsymbol{\epsilon}$ is the dielectric tensor. The latter is expanded into a power series of the strain tensor $\boldsymbol{\varepsilon}$ and the electric field $\boldsymbol{E}$,

$$\boldsymbol{\epsilon}=\boldsymbol{\epsilon}^{(0)}+\boldsymbol{b}\boldsymbol{\varepsilon}+\boldsymbol{\epsilon}^{(4)}(\boldsymbol{E}\otimes\boldsymbol{E}). \tag{3.3}$$

The first term on the right-hand side denotes the isotropic part. The second one describes the elasto-optic effect, where $\boldsymbol{b}$ is the elasto-optic tensor. Finally, the last term accounts for a nonlinear response in $\boldsymbol{E}$.

In order to obtain the equilibrium value of the strain tensor $\boldsymbol{\varepsilon}$ we minimize $\mathcal{F}_{\text{distortion}}$ for a given electric field $\boldsymbol{E}$,

$$\frac{\partial\mathcal{F}_{\text{distortion}}}{\partial\boldsymbol{\varepsilon}}=\boldsymbol{\lambda}\boldsymbol{\varepsilon}-\frac{\delta'}{8\pi}\boldsymbol{b}(\boldsymbol{E}\otimes\boldsymbol{E})=\boldsymbol{0}. \tag{3.4}$$

Solving Eq. (3.4) for $\boldsymbol{\varepsilon}$ yields the electrostriction tensor $\boldsymbol{R}$,

$$\boldsymbol{\varepsilon}=\boldsymbol{R}(\boldsymbol{E}\otimes\boldsymbol{E}),\quad \boldsymbol{R}=\frac{\delta'}{8\pi}\boldsymbol{\lambda}^{-1}\boldsymbol{b}. \tag{3.5}$$

Clearly, to find $\boldsymbol{R}$ we need to calculate $\boldsymbol{\lambda}$ and $\boldsymbol{b}$ from the LGdG model of the blue phases. The starting point of such calculations is the elastic free energy, i.e., the free energy difference between the distorted and undistorted states. In the LGdG approach it reads

$$\mathcal{F}_{\text{elastic}}=\frac{1}{2}\boldsymbol{\lambda}(\boldsymbol{\varepsilon}\otimes\boldsymbol{\varepsilon})=\mathcal{F}[\tilde{\boldsymbol{Q}},\partial\tilde{\boldsymbol{Q}}]-\mathcal{F}[\boldsymbol{Q},\partial\boldsymbol{Q}], \tag{3.6}$$

where $\tilde{\boldsymbol{Q}}$ denotes the order parameter of the deformed structure. To obtain its precise form we need a model for the deformation $\tilde{\boldsymbol{Q}}$.

For that purpose we note that when the real lattice is distorted proportionally to the strain tensor $\boldsymbol{\varepsilon}$ the reciprocal lattice behaves inversely [8]. Hence, the distorted order parameter must depend on the deformed wave vectors $\tilde{\boldsymbol{k}}=(1-\boldsymbol{\varepsilon})\boldsymbol{k}$. Additionally, it acquires a homogeneous term, proportional to the strain tensor, which is forbidden in systems with cubic symmetry. Thus in a symbolic way this deformation can be written as

$$\begin{aligned}\boldsymbol{Q}(\boldsymbol{r})\xrightarrow{\text{deformation}}\tilde{\boldsymbol{Q}}(\boldsymbol{r})&=\boldsymbol{b}\boldsymbol{\varepsilon}+\sum_{\tilde{\boldsymbol{k}}}Q_2(\tilde{\boldsymbol{k}})\boldsymbol{M}_2(\tilde{\boldsymbol{k}})e^{i\tilde{\boldsymbol{k}}\cdot\boldsymbol{r}}\\&=\sum_{*\tilde{\boldsymbol{k}}}\frac{1}{\sqrt{N_{*\tilde{\boldsymbol{k}}}}}\\&\quad\times\left\{\sum_{\tilde{\boldsymbol{k}}\in{}^*\tilde{\boldsymbol{k}}\cup\boldsymbol{0}}\boldsymbol{Q}_2(\tilde{\boldsymbol{k}})\boldsymbol{M}_2(\tilde{\boldsymbol{k}})e^{i\tilde{\boldsymbol{k}}\cdot r}\right\},\end{aligned} \tag{3.7}$$

where $\boldsymbol{b}$ is the elasto-optic tensor, which we introduced in Eq. (3.3). Additionally, ${}^*\tilde{\boldsymbol{k}}$ stands for the set of $\tilde{\boldsymbol{k}}$ vectors obtained from the deformation of ${}^*\boldsymbol{k}$, $\boldsymbol{Q}_2(\boldsymbol{0})\boldsymbol{M}_2(\boldsymbol{0})\equiv\sqrt{N_{*\tilde{\boldsymbol{k}}}}\boldsymbol{b}\boldsymbol{\varepsilon}$, and $N_{*\tilde{\boldsymbol{k}}}=N_{*\boldsymbol{k}}$

In the earlier work on electrostriction by Stark and Trebin [9], also referred to as the *model of rigid helices*, the scalar amplitudes $Q_2(\tilde{\boldsymbol{k}})$ of $\tilde{\boldsymbol{Q}}(\boldsymbol{r})$ were kept fixed: $Q_2(\tilde{\boldsymbol{k}})\equiv Q_2(\boldsymbol{k})$. It is the main purpose of the present work to extend the model of rigid helices by additionally including the deformation of the amplitudes. Such analysis seems important as the response of the blue phases to an external field is very sensitive to the underlying structure [2].

In analogy to the calculations performed in [9] the expressions for the deformed basis modes $\boldsymbol{M}_2(\tilde{\boldsymbol{k}})$ are derived by rotating $\boldsymbol{M}_2(\boldsymbol{k})$ about an axis perpendicular to $\boldsymbol{k}$ and $\tilde{\boldsymbol{k}}$. They read [9]

$$\boldsymbol{M}_2(\tilde{\boldsymbol{k}})=\boldsymbol{M}_2(\boldsymbol{k})+[\boldsymbol{M}_1(\boldsymbol{k})\otimes\boldsymbol{M}_1(\boldsymbol{k})]\boldsymbol{\varepsilon}+\cdots. \tag{3.8}$$

Consequently, the free energy $\mathcal{F}[\tilde{\boldsymbol{Q}},\partial\tilde{\boldsymbol{Q}}]$ of the deformed state which enters the definition of the elastic free energy can be written as

$$\begin{aligned}\mathcal{F}[\tilde{\boldsymbol{Q}},\partial\tilde{\boldsymbol{Q}}]=&\sum_{*\tilde{\boldsymbol{k}}}\frac{1}{N_{*\tilde{\boldsymbol{k}}}}\sum_{\tilde{\boldsymbol{k}}\in{}^*\tilde{\boldsymbol{k}}\cup\boldsymbol{0}}\left[\frac{t}{4}+\frac{1}{4}\kappa^2\tilde{\boldsymbol{k}}^2-\frac{1}{2}\kappa^2|\tilde{\boldsymbol{k}}|\right]\boldsymbol{Q}_2(\tilde{\boldsymbol{k}})\boldsymbol{Q}_2(-\tilde{\boldsymbol{k}})\\&-\sqrt{6}\sum_{*\tilde{\boldsymbol{k}}_1,*\tilde{\boldsymbol{k}}_2,*\tilde{\boldsymbol{k}}_3}\frac{1}{\sqrt{N_{*\tilde{\boldsymbol{k}}_1}N_{*\tilde{\boldsymbol{k}}_2}N_{*\tilde{\boldsymbol{k}}_3}}}\sum_{\tilde{\boldsymbol{k}}_\alpha\in{}^*\tilde{\boldsymbol{k}}_1\cup\boldsymbol{0}}\sum_{\tilde{\boldsymbol{k}}_\beta\in{}^*\tilde{\boldsymbol{k}}_2\cup\boldsymbol{0}}\sum_{\tilde{\boldsymbol{k}}_\gamma\in{}^*\tilde{\boldsymbol{k}}_3\cup\boldsymbol{0}}\tilde{Q}_\alpha\tilde{Q}_\beta\tilde{Q}_\gamma\mathrm{Tr}[\tilde{\boldsymbol{M}}_\alpha\tilde{\boldsymbol{M}}_\beta\tilde{\boldsymbol{M}}_\gamma]\delta(\tilde{\boldsymbol{k}}_\alpha+\tilde{\boldsymbol{k}}_\beta+\tilde{\boldsymbol{k}}_\gamma)\\&+\sum_{*\tilde{\boldsymbol{k}}_1,*\tilde{\boldsymbol{k}}_2,*\tilde{\boldsymbol{k}}_3,*\tilde{\boldsymbol{k}}_4}\frac{1}{\sqrt{N_{*\tilde{\boldsymbol{k}}_1}N_{*\tilde{\boldsymbol{k}}_2}N_{*\tilde{\boldsymbol{k}}_3}N_{*\tilde{\boldsymbol{k}}_4}}}\sum_{\tilde{\boldsymbol{k}}_\alpha\in{}^*\tilde{\boldsymbol{k}}_1\cup\boldsymbol{0}}\sum_{\tilde{\boldsymbol{k}}_\beta\in{}^*\tilde{\boldsymbol{k}}_2\cup\boldsymbol{0}}\sum_{\tilde{\boldsymbol{k}}_\gamma\in{}^*\tilde{\boldsymbol{k}}_3\cup\boldsymbol{0}}\sum_{\tilde{\boldsymbol{k}}_\mu\in{}^*\tilde{\boldsymbol{k}}_4\cup\boldsymbol{0}}\\&\times\tilde{Q}_\alpha\tilde{Q}_\beta\tilde{Q}_\gamma\tilde{Q}_\mu\mathrm{Tr}[\tilde{\boldsymbol{M}}_\alpha\tilde{\boldsymbol{M}}_\beta][\tilde{\boldsymbol{M}}_\gamma\tilde{\boldsymbol{M}}_\mu]\delta(\tilde{\boldsymbol{k}}_\alpha+\tilde{\boldsymbol{k}}_\beta+\tilde{\boldsymbol{k}}_\gamma+\tilde{\boldsymbol{k}}_\mu),\end{aligned} \tag{3.9}$$

where $\tilde{Q}_\alpha\equiv Q_2(\tilde{\boldsymbol{k}}_\alpha)$, $\tilde{\boldsymbol{M}}_\alpha\equiv\boldsymbol{M}_2(\tilde{\boldsymbol{k}}_\alpha)$, and where δ is the Kronecker delta function. In order to extract the tensor of elastic constants from the formula (3.9) we still need to introduce the deformation of the amplitudes. Up to linear order in $\boldsymbol{\varepsilon}$ it can be written as

$$Q_2(\tilde{\boldsymbol{k}})=Q_2(\boldsymbol{k})+\boldsymbol{\psi}(\boldsymbol{k})\cdot\boldsymbol{\varepsilon}+\cdots, \tag{3.10}$$

where the matrices $\boldsymbol{\psi}(\boldsymbol{k})$ represent the derivatives of $Q_2(\tilde{\boldsymbol{k}})$ with respect to the components of the strain tensor $\boldsymbol{\varepsilon}$. In general, there are as many different $\boldsymbol{\psi}(\boldsymbol{k})$ matrices as $\boldsymbol{k}$ vectors modeling the ground state. Fortunately, not all of them are independent. This is a consequence of the symmetry properties of the order parameter. To clarify this statement let us write the deformed order parameter field as

$$\tilde{\boldsymbol{Q}}(\boldsymbol{r})=\boldsymbol{Q}_0(\boldsymbol{r})+\boldsymbol{b}\boldsymbol{\varepsilon}+\boldsymbol{Q}_1(\boldsymbol{r})\boldsymbol{\varepsilon}+\cdots. \tag{3.11}$$

Clearly, the coefficients $\boldsymbol{Q}_0(\boldsymbol{r})$ and $\boldsymbol{Q}_1(\boldsymbol{r})$ should possess the symmetry of the cubic ground state which requires that

$$[\{\boldsymbol{S}|\boldsymbol{t}\}\boldsymbol{Q}_n](\boldsymbol{r})=\boldsymbol{Q}_n(\boldsymbol{r}),\quad n=0,1 \tag{3.12}$$

where $\{\boldsymbol{S}|\boldsymbol{t}\}$ is an element of the space group O^8 or O^2. It consists of a combination of a rotation $\boldsymbol{S}$ from the cubic point group and a translation $\boldsymbol{t}$ [11]. The first condition provides a relation for the ground state amplitudes $Q_2(\boldsymbol{k})$,

$$Q_2(\boldsymbol{k})=Q_2(\boldsymbol{S}k), \tag{3.13}$$

whereas the second yields restrictions on the matrices $\boldsymbol{\psi}(\mathbf{k})$

$$\boldsymbol{S}\boldsymbol{\psi}(\boldsymbol{k})=\boldsymbol{\psi}(\boldsymbol{S}\boldsymbol{k}), \tag{3.14}$$

where we choose a symbolic notation $\boldsymbol{S}\boldsymbol{k}$ and $\boldsymbol{S}\boldsymbol{\psi}$ for the rotated $\boldsymbol{k}$ and $\boldsymbol{\psi}$. Equation (3.14) means that for each star of $\boldsymbol{k}$ vectors we need to know only *one* of the $\boldsymbol{\psi}(\boldsymbol{k})$ matrices. All the others can be systematically generated by acting with the point group elements $\boldsymbol{S}$ on this representative. For a general star, the matrix $\boldsymbol{\psi}(\boldsymbol{k})$ possesses six independent components. In the case of a degenerate star ($\boldsymbol{S}\boldsymbol{k}=\boldsymbol{k}, \boldsymbol{S}\neq\mathbf{1}$) Eq. (3.14) gives

$$\boldsymbol{S}\boldsymbol{\psi}(\boldsymbol{k})=\boldsymbol{\psi}(\boldsymbol{k}), \tag{3.15}$$

which means that $\boldsymbol{\psi}$ has the little group of $\boldsymbol{k}$ as its symmetry group. For $\boldsymbol{k}$ and $-\boldsymbol{k}$ belonging to the same star we have a further restriction on $\boldsymbol{\psi}$'s that comes from the reality condition of the order parameter, namely,

$$\boldsymbol{\psi}^{*}(\boldsymbol{k})=\boldsymbol{\psi}(-\boldsymbol{k}). \tag{3.16}$$

Taking into account the relations (3.15) and (3.16) we can calculate the general form of the $\boldsymbol{\psi}(\boldsymbol{k})$ matrices for the relevant, leading stars of $\boldsymbol{k}$ vectors that describe BPI and BPII. The result is

$$\boldsymbol{\psi}([n00])=\begin{pmatrix} a1_n & 0 & 0 \\ 0 & b1_n & 0 \\ 0 & 0 & b1_n \end{pmatrix},$$

$$\boldsymbol{\psi}([nn0])=\begin{pmatrix} a2_n & b2_n & 0 \\ b2_n & a2_n & 0 \\ 0 & 0 & c2_n \end{pmatrix}, \tag{3.17}$$

$$\boldsymbol{\psi}([n2n2n])=\begin{pmatrix} a3_n & b3_n & c3_n \\ b3_n & a3_n & c3_n \\ c3_n & c3_n & d3_n \end{pmatrix}.$$

Inserting Eqs. (3.8), (3.10), and (3.17) into Eq. (3.7) yields the deformed tensor field $\tilde{\boldsymbol{Q}}(\boldsymbol{r})$, which is accurate up to terms linear in $\boldsymbol{\varepsilon}$. Since the expansion (3.7) is performed around the minimum value of the free energy this linear approximation is sufficient to obtain exact formulas for the tensor $\boldsymbol{\lambda}$. The calculations are involved due to the presence of cubic and quartic terms in the expansion (3.9) and are carried out using MAPLE package for symbolic calculations. The procedure is the following: in the first step all the basis tensors $\boldsymbol{M}_m(\boldsymbol{k})$ are expressed in a common coordinate system. Next, the closed loops formed by three or four wave vectors $\tilde{\boldsymbol{k}}_\alpha$ are identified by noting the relation between deformed and undeformed vectors that sum up to zero: $\tilde{\boldsymbol{k}}_1+\tilde{\boldsymbol{k}}_2+\tilde{\boldsymbol{k}}_3+\cdots=0=\boldsymbol{k}_1+\boldsymbol{k}_2+\boldsymbol{k}_3+\cdots$. For each such loop the corresponding contribution to Eq. (3.9) is calculated by appropriate multiplication and tracing of the associated matrices. The huge number of contributions is partly reduced by observing permutation symmetry of the trace operation.

The fourth-rank tensor of elastic constants $\boldsymbol{\lambda}$ so determined has a cubic symmetry and therefore it has only three independent elements. We denote them $\lambda_1=\lambda_{1111}$, $\lambda_2=\lambda_{1122}$, and $\lambda_3=\lambda_{2323}$. Since the formulas are about four pages long we will not present them here. On request we could provide them in the form of an ASCII file. They depend on the elements of the $\boldsymbol{\psi}(\boldsymbol{k})$ matrices and on the components of the elasto-optic tensor $\boldsymbol{b}$. The latter is also a fourth-rank tensor of cubic symmetry and its three independent components usually are denoted as $b_1=b_{1111}$, $b_2=b_{1122}$, and $b_3=b_{2323}$ [12]. Assuming that the deformation leaves the mass density unchanged an additional relation holds, namely, $b_1=-2b_2$ [9].

The equilibrium value of the $\boldsymbol{\psi}(\boldsymbol{k})$ matrices and of the elasto-optic tensor $\boldsymbol{b}$ are then determined by minimizing the elastic free energy for arbitrary distortions $\boldsymbol{\varepsilon}$,

$$\frac{\partial\boldsymbol{\lambda}\cdot(\boldsymbol{\varepsilon}\otimes\boldsymbol{\varepsilon})}{\partial\boldsymbol{b}}=0, \quad \frac{\partial\boldsymbol{\lambda}\cdot(\boldsymbol{\varepsilon}\otimes\boldsymbol{\varepsilon})}{\partial\boldsymbol{\psi}(\boldsymbol{k})}=0. \tag{3.18}$$

Finally, the equilibrium tensor $\boldsymbol{\lambda}$ with its three independent elements is evaluated by substituting the solutions for $\boldsymbol{\psi}(\boldsymbol{k})$ and $\boldsymbol{b}$. Together with $\boldsymbol{b}$, it allows for the determination of the electrostriction tensor $\boldsymbol{R}$.

IV. RESULTS AND SHORT SUMMARY

The independent components of $\boldsymbol{R}$ read

$$R_1=\frac{\delta'}{8\pi}\frac{b_1}{\lambda_1-\lambda_2}=-2R_2, \quad R_3=\frac{\delta'}{8\pi}\frac{b_3}{\lambda_3}. \tag{4.1}$$

All of them are functions of the cubic ground state amplitudes $Q_2(\boldsymbol{k})$ where the latter are obtained by minimizing the LGdG free energy function for fixed values of t and κ. The temperature dependence of the ratio R_1/R_3 for different values of chirality κ is shown in Fig. 1.

Now we compare the results of present calculations with the model of rigid helices [9]. The predictions arriving from these two models differ quantitatively up to 50%. For the blue phase II the model proposed in this paper fits better to experimental data, which are summarized in Table I.

The experimental values for R_1/R_3 are in general higher than the predictions of the model of rigid helices and agree with the calculations presented in this paper. The situation is more complex for the blue phase I. Our model can either lower or enlarge the ratio R_1/R_3 depending on the chirality. But it cannot yield the negative values predicted by the

TABLE I. Experimental values of the R_1/R_3 for different chiral compounds [4].

Blue phase II			
Compound	1	2	3
R_1/R_3	0.42	0.83	1.03
Blue phase I			
Compound	1	2	3
R_1/R_3	−0.07	−0.38	−0.27

experiments, i.e., cannot describe the anomalous electrostriction of the blue phase I.

Summarizing, we have developed a complete theory of the electrostriction of the cubic blue phases within the standard Landau-Gizburg-deGennes model. The rigid helices approximation proposed earlier by Stark and Trebin [9] was generalized by taking into account the full spectrum of deformations of the tensor order parameter. Exact expressions (within LGdG theory) for the independent components of the electrostriction tensor $\boldsymbol{R}$ were obtained. Interestingly, the results for the blue phase II are in good agreement with the experimental data. However, the anomalous electrostriction of the blue phase I cannot be explained by the generalized model. Therefore we conclude that a proper description of this phenomenon is beyond the standard LGdG theory. One possible explanation can be obtained within the bond-order model [13].

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