Nonlinear relative rotations in liquid crystalline elastomers

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Relative rotations between the coupled subsystems of a complex material can become crucial in continuum modeling. In this paper the authors focus on the macroscopic description of side-chain liquid crystalline elastomers, where relative rotations between the polymer network and the director orientation associated with the liquid crystalline component are decisive. They extend the known expression for relative rotations to the nonlinear regime. This allows the investigation of qualitatively different nonlinear effects determined by these variables, and they give an illustrative example. The formalism can easily be transferred to the macroscopic description of magnetic gels and will certainly be helpful in the characterization of other complex systems. © 2007 American Institute of Physics.
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I. INTRODUCTION

By the term "relative rotation" we refer to a situation, in which two or more coupled subsystems of a complex material are spatially rotated relative to each other, globally or locally. If such relative rotations lead to contributions to the energy density of the system under consideration and influence its macroscopic dynamics, it is frequently possible to include them as slowly relaxing variables in a generalized hydrodynamic description of the respective system. The cases in which such a procedure has successfully been applied include the macroscopic description of magnetic gels[1] and of side-chain liquid crystalline elastomers (SCLCEs). The first synthesis of SCLCEs has been reported in 1981. It was performed by chemically crosslinking polymer chains, to which low molecular weight liquid crystalline molecules, the so-called mesogenic units, had been attached as side groups via flexible spacers[2]. In the liquid crystalline phase of low molecular weight materials these mesogenic units on average orient macroscopically in a certain direction, which is characterized by the director field $\hat{n}(\mathbf{r})$ (see e.g., Ref. 3). This macroscopic orientation also occurs in liquid crystalline elastomers, however more sophisticated techniques of synthesis are necessary to achieve a macroscopic ordering of the mesogenic units over the whole sample. It was in 1991 when such liquid crystalline monodomain elastomers were generated for the first time,[4] and they were called side-chain liquid single crystal elastomers (SCLSCEs).

If the crosslinking density in the elastomers is low enough, two subsystems can be identified in SCLCEs from a macroscopic point of view[5]. On the one hand the polymer network shows the mechanical properties of a conventional elastomer, and on the other hand the entity of the oriented mesogenic units characterized by $\hat{n}(\mathbf{r})$ brings along the properties of the respective liquid crystalline phase. Both subsystems are coupled to and interact with each other, which already results from the chemical binding between the mesogenic units and the polymer backbone via the flexible spacer.

It was proposed for the first time by de Gennes to account for this coupling between the two subsystems in liquid crystalline elastomers by relative rotations between the polymer network and the director field[6]. Two of us later included relative rotations as macroscopic variables in the generalized hydrodynamic characterization of SCLSCEs[7]. The consideration of relative rotations in the macroscopic description of SCLSCEs has since turned out to be essential to understand many of the unique properties of these materials. By means of relative rotations it can be explained, for example, how mechanical deformations of SCLSCEs such as shear can lead to a reorientation of the director field[8]. On the other hand it has been investigated theoretically how the reorientation of the director field by external electric or magnetic fields can also lead to a macroscopic deformation of the elastomers via relative rotations[9–11].

All of these properties could be described as linear effects within a continuum model of SCLSCEs. For this purpose, the linear expressions for the macroscopic variables characterizing the thermodynamic state of the system were combined to symmetry-allowed quadratic terms, which then formed an expression for the generalized energy density of the system.

If we want to include nonlinear properties of the materials

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into the macroscopic characterization, the generalized energy density has to be supplemented by higher order terms. However, also the quadratic terms in the generalized energy density describe nonlinear effects, when nonlinear expressions are inserted for the macroscopic variables. Therefore, a nonlinear expression for relative rotations has to be derived.

Up to now only a linear expression for the relative rotations in liquid crystalline elastomers has been given within the framework of a continuum model. In the next section of this paper we extend this expression to the nonlinear regime using the approach that applies to the macroscopic description of SCLSCEs. With the help of the resulting formalism characteristic features of liquid crystalline elastomers can be described as nonlinear effects in a continuum model. In order to demonstrate this, in Sec. III and in the appendix we investigate as an illustrative example the nonlinear reaction of a nematic SCLSCE to an imposed static shear deformation. We discuss and summarize our results in the last two sections of this paper and also give a short perspective.

II. NONLINEAR RELATIVE ROTATIONS

In this section we derive a nonlinear expression for the variables of relative rotations between the director field and the polymer network in SCLSCEs. We first perform an extension of the linear expression up to quartic order in the variables characterizing the actual state of the system. This procedure will mainly be appropriate for practical purposes and can systematically be generalized up to any desirable order. After that we show how an exact expression for the relative rotations can be found and we give the result for a two-dimensional system.

When we compare to the ground state of a nematic SCLSCE, its actual state under the influence of a static external field is completely identified by five independent variables. Namely, these are on the one hand the three components of the displacement field $\mathbf{u}(\mathbf{r}) = \mathbf{r} - \boldsymbol{a}(\mathbf{r})$, describing local strains and rotations of the polymer network. $\mathbf{r}$ denotes the actual positions of the volume elements of the polymer network in the final state, and by the initial field $\boldsymbol{a}(\mathbf{r})$ we specify their positions in the undeformed state. Obviously we can also take the components of $\boldsymbol{a}(\mathbf{r})$ as independent variables instead. The difference between the two choices of $\mathbf{u}(\mathbf{r})$ and $\boldsymbol{a}(\mathbf{r})$ is discussed in section IV.

On the other hand there are two variables that determine the current state of the director field $\hat{\mathbf{n}}(\mathbf{r})$ and thus characterize the subsystem showing the liquid crystalline phase. $\hat{\mathbf{n}}$ is an axial unit vector, such that the states $\hat{\mathbf{n}}$ and $-\hat{\mathbf{n}}$ cannot be distinguished and only two components of $\hat{\mathbf{n}}(\mathbf{r})$ are independent. The ground state conformation of the director field will be denoted as $\hat{\mathbf{n}}_0$, whereas in general for finite deformations this will be a functional of $\mathbf{u}(\mathbf{r})$ or $\boldsymbol{a}(\mathbf{r})$,

$$\hat{\mathbf{n}}_0 = \hat{\mathbf{n}}_0(\boldsymbol{a}(\mathbf{r})). \tag{1}$$

We include this dependence because the mesogenic units are part of the polymer network and are displaced in the same way under finite mechanical deformations, in spite of forming a separate subsystem in a macroscopic description. In general,

the ground state orientation $\hat{\mathbf{n}}_0$ at a certain position $\boldsymbol{a}(\mathbf{r})$ is known as an initial condition, however, what is not known is the initial field $\boldsymbol{a}(\mathbf{r})$.

A standard procedure of finding the current state of a system consists of deriving an expression for the generalized energy density of the system and of minimizing the corresponding generalized energy. It is of major importance to perform this minimization with respect to the independent variables that correspond to the degrees of freedom of the system and that can completely characterize the current state of the system. Following this procedure it is clear that a realistic situation is described, for which conditions of compatibility as known from solving problems of elasticity theory are satisfied automatically.

In our case, the five independent variables given by $\mathbf{u}(\mathbf{r})$ or $\boldsymbol{a}(\mathbf{r})$, and by $\hat{\mathbf{n}}(\mathbf{r})$ correspond to these degrees of freedom. However, an expression for the generalized energy density cannot be derived from these variables directly. This results from the fact that homogeneous translations or rotations of the whole system do not contribute to the generalized energy density. On the contrary, strain deformations of the polymer network and relative rotations between the polymer network and the director field can contribute to the generalized energy density of SCLSCEs. Therefore, in the following we will express the nonlinear variables of strain deformations and of relative rotations by the components of $\mathbf{u}(\mathbf{r})$ or $\boldsymbol{a}(\mathbf{r})$, respectively, and of $\hat{\mathbf{n}}(\mathbf{r})$.

Deformations of strain are entirely characterized by the nonlinear strain tensor $\boldsymbol{\varepsilon}$, which is defined by $dr_i^2 - da_i^2(\mathbf{r}) = 2dr_i dr_j \varepsilon_{ij}(\mathbf{r})$ in the Euler notation[12]. The Euler notation describes the appropriate approach in a hydrodynamic picture[13,14], and the complete nonlinear expressions for the components of $\boldsymbol{\varepsilon}$ then read

$$\begin{aligned}
\varepsilon_{ij} &= \frac{1}{2}\left[(\partial_i u_j) + (\partial_j u_i) - (\partial_i u_k)(\partial_j u_k)\right] \\
&= \frac{1}{2}\left[\delta_{ij} - (\partial_i a_k)(\partial_j a_k)\right]. \tag{2}
\end{aligned}$$

Here, $\boldsymbol{\varepsilon}$, $\mathbf{u}$ and $\boldsymbol{a}$ are functionals of $\mathbf{r}$, and ∂_i denotes the partial derivative $\partial/\partial r_i$ ($i = 1, 2, 3$).

In the linear regime, rotations can be described by vectors. With the two vectors $\boldsymbol{\omega}^n$ and $\boldsymbol{\omega}^{nw}$ denoting local rotations of the director and the polymer network, respectively, the expression $(\boldsymbol{\omega}^n - \boldsymbol{\omega}^{nw}) \times \hat{\mathbf{n}}$ describes relative rotations between the two subsystems. This is the way relative rotations were introduced in[6]. The use of $\hat{\mathbf{n}}$ and $\hat{\mathbf{n}}_0$ in this expression is indistinguishable in the linear regime, so we could as well write

$$\tilde{\boldsymbol{\Omega}} = (\boldsymbol{\omega}^n - \boldsymbol{\omega}^{nw}) \times \hat{\mathbf{n}}_0 \tag{3}$$

for the vector of relative rotations.

In[7] the notation was modified and relative rotations were defined by

$$\tilde{\boldsymbol{\Omega}} = \delta\mathbf{n} - \boldsymbol{\Omega}^\perp. \tag{4}$$

This expression refers to the linear regime. Here, $\delta n_i = n_i - n_{0,i}$ and $\Omega^\perp_i = n_j \Omega_{ji}$, whereas in linear order the latter expression cannot be distinguished from $n_{0,j}\Omega_{ji}$. $\boldsymbol{\Omega}$ denotes
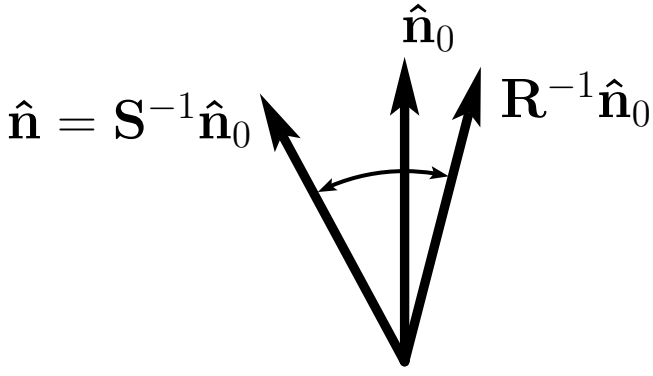
FIG. 1: The vector of relative rotations characterizes the difference between two local rotations of the director out of its ground state conformation $\hat{\mathbf{n}}_0$. Actually $\hat{\mathbf{n}}_0$ is rotated by $\mathbf{S}^{-1}$ to its local state $\hat{\mathbf{n}}$. If $\hat{\mathbf{n}}_0$ were rigidly coupled to the polymer network, it would be rotated with the polymer network by $\mathbf{R}^{-1}$.

the antisymmetric tensor of rigid rotations of the polymer network, which we obtain in the linear regime from the distortion tensor $\boldsymbol{\nabla}\mathbf{u}$ by

$$\boldsymbol{\Omega}_{ij} = (\partial_i u_j) - \varepsilon_{ij} = \frac{1}{2}\left(\partial_i u_j - \partial_j u_i\right). \quad (5)$$

The vector $\boldsymbol{\Omega}^{\perp}$ can then be interpreted as the vector of rigid rotations of the polymer network perpendicular to the director[7]. This interpretation only accounts for the linear regime as we will see in the following.

The consistency of Eqs. (3) and (4) follows from $\delta n_i = \epsilon_{ijk}\omega_j^n n_{0,k}$ and $\omega_i^{nw} = \frac{1}{2}\epsilon_{ijk}\Omega_{jk}$, where ϵ_{ijk} denotes the Levi-Cività tensor. Equations (3) and (4) imply that in the linear regime

$$n_i\tilde{\Omega}_i = 0. \quad (6)$$

In the following we want to derive a nonlinear expression for $\tilde{\boldsymbol{\Omega}}$. According to the way we introduced relative rotations at the beginning of section I, we now define relative rotations in SCLSCEs as the difference between two rotations.

On the one hand, when we consider the system in its final state, the director has locally been rotated by the rotation matrix $\mathbf{S}^{-1}$ to its final orientation. In general, for finite deformations, $\mathbf{S}^{-1}$ will be a functional of $\mathbf{r}$ and of $\mathbf{u}(\mathbf{r})$ or $\boldsymbol{a}(\mathbf{r})$, which is a consequence of Eq. (1)

$$n_{0,i}(\boldsymbol{a}(\mathbf{r})) = S_{ij}(\boldsymbol{a}(\mathbf{r}), \mathbf{r})n_j(\mathbf{r}). \quad (7)$$

Because of this dependence, $\mathbf{S}^{-1}(\boldsymbol{a}(\mathbf{r}), \mathbf{r})$ can describe how the mesogenic units have on average been rotated compared to their ground state orientation (see also section IV).

On the other hand the polymer network has locally been rotated by the rotation matrix $\mathbf{R}^{-1}(\mathbf{r})$. We derive this matrix from a comparison between the initial state and the final state of the system

$$da_i = dr_k\left(\partial_k a_i\right). \quad (8)$$

Here, $\partial_k a_i$ describes the local distortions of the polymer network. Using the polar decomposition theorem, $\partial_k a_i$ can be

rewritten as a product of a rotation matrix and a symmetric matrix as

$$\partial_k a_i = R_{ij}\,\Xi_{jk}. \quad (9)$$

Altogether we obtain $da_i = R_{ij}\Xi_{jk}dr_k$. $\boldsymbol{\Xi}$ tells us, how the polymer network in its final state locally has to be unstrained, and $\mathbf{R}$ tells us how it locally has to be rotated back to retrieve its initial state. In Refs. (13) and (14) it was shown how to obtain $\boldsymbol{\Xi}$ from Eq. (9), and $\boldsymbol{\Xi}$ was given up to quadratic order in the components of $\boldsymbol{\nabla}\mathbf{u}$. We now calculate $\boldsymbol{\Xi}$ up to quartic order and from that derive the rotation matrix $\mathbf{R}$.

With δ_{jk} denoting the Kronecker delta we obtain from $R_{ij}R_{ik} = \delta_{jk}$ and from Eqs. (2) and (9):

$$\delta_{jk} - 2\varepsilon_{jk} = \Xi_{ij}\Xi_{ik}. \quad (10)$$

Inserting a power expansion of Ξ_{ij} with respect to ε into Eq. (10) we obtain

$$\begin{aligned}\Xi_{ij} &= \delta_{ij} - \varepsilon_{ij} - \frac{1}{2}\varepsilon_{ik}\varepsilon_{kj} - \frac{1}{2}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lj} - \frac{5}{8}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lm}\varepsilon_{mj} \\ &\quad + \mathcal{O}\left((\boldsymbol{\nabla}\mathbf{u})^5\right).\end{aligned} \quad (11)$$

Furthermore, by using $(\Xi^{-1})_{ij}\Xi_{jk} = \delta_{ik}$ we can show that

$$\begin{aligned}(\Xi^{-1})_{ij} &= \delta_{ij} + \varepsilon_{ij} + \frac{3}{2}\varepsilon_{ik}\varepsilon_{kj} + \frac{5}{2}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lj} \\ &\quad + \frac{35}{8}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lm}\varepsilon_{mj} + \mathcal{O}\left((\boldsymbol{\nabla}\mathbf{u})^5\right).\end{aligned} \quad (12)$$

This expression for $\boldsymbol{\Xi}^{-1}$ and Eq. (9) can then be used to obtain the components of the rotation matrix $\mathbf{R}$. As we noted above, we are interested in the way the polymer network has been rotated from the initial to the final state and so we give the components of $\mathbf{R}^{-1}$:

$$\begin{aligned}(R^{-1})_{ij} = R_{ji} &= \left(\delta_{ik} + \varepsilon_{ik} + \frac{3}{2}\varepsilon_{il}\varepsilon_{lk} + \frac{5}{2}\varepsilon_{il}\varepsilon_{lm}\varepsilon_{mk}\right. \\ &\quad \left. + \frac{35}{8}\varepsilon_{il}\varepsilon_{lm}\varepsilon_{mn}\varepsilon_{nk}\right)(\partial_k a_j) + \mathcal{O}\left((\boldsymbol{\nabla}\mathbf{u})^5\right) \\ &= \delta_{ij} + \varepsilon_{ij} + \frac{3}{2}\varepsilon_{ik}\varepsilon_{kj} + \frac{5}{2}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lj} \\ &\quad + \frac{35}{8}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lm}\varepsilon_{mj} - (\partial_i u_j) - \varepsilon_{ik}(\partial_k u_j) \\ &\quad - \frac{3}{2}\varepsilon_{ik}\varepsilon_{kl}(\partial_l u_j) - \frac{5}{2}\varepsilon_{ik}\varepsilon_{kl}\varepsilon_{lm}(\partial_m u_j) \\ &\quad + \mathcal{O}\left((\boldsymbol{\nabla}\mathbf{u})^5\right),\end{aligned} \quad (13)$$

where we have made use of $\boldsymbol{a}(\mathbf{r}) = \mathbf{r} - \mathbf{u}(\mathbf{r})$ to obtain the final expression.

For a check of consistency we calculate the matrices $\tilde{\boldsymbol{\Xi}}$ and $\tilde{\mathbf{R}}$, defined by $\partial_k a_i = \tilde{\Xi}_{ij}\tilde{R}_{jk}$. We multiply this equation by $\tilde{R}_{lk}$, and from the resulting equation we derive $\tilde{\Xi}_{li}\tilde{\Xi}_{im} = (\partial_k a_i)(\partial_n a_i)\tilde{R}_{lk}\tilde{R}_{mn}$. With the help of this expression and together with Eq. (2) we can verify that

$$\begin{aligned}\tilde{\Xi}_{ij} &= \tilde{R}_{ik}\tilde{R}_{jl}\left(\delta_{kl} - \varepsilon_{kl} - \frac{1}{2}\varepsilon_{km}\varepsilon_{ml} - \frac{1}{2}\varepsilon_{km}\varepsilon_{mn}\varepsilon_{nl}\right. \\ &\quad \left. - \frac{5}{8}\varepsilon_{km}\varepsilon_{mn}\varepsilon_{no}\varepsilon_{ol}\right) + \mathcal{O}\left((\boldsymbol{\nabla}\mathbf{u})^5\right).\end{aligned} \quad (14)$$

Introducing $\tilde{\Xi}_{ij}$ into $\partial_p a_i = \tilde{\Xi}_{ij}\tilde{R}_{jp}$, taking into account that $\tilde{R}_{jp}\tilde{R}_{jl} = \delta_{lp}$ and convincing ourselves that the inverse of the expression in parentheses in Eq. (14) is given by Eq. (12) we obtain $\tilde{\mathbf{R}} = \mathbf{R}$. There arises no problem from $\tilde{\Xi} \neq \Xi$, because in our description we will use the tensor $\boldsymbol{\varepsilon}$ as a macroscopic variable to include strain deformations, not Ξ.

We now can define the components of the vector of relative rotations by

$$\tilde{\Omega}_i = n_i - [n_k(R^{-1})_{kl}n_{0,l}](R^{-1})_{ij}n_{0,j} \qquad (15)$$

This expression for relative rotations can be interpreted in the following manner. Relative rotations give the difference between the way the director has been actually rotated starting from its ground state orientation ($n_i = (S^{-1})_{ij}n_{0,j}$) and the way it would have been rotated if it was rigidly coupled to the polymer network ($\mathbf{R}^{-1}$). We have illustrated this feature in Fig. 1. For symmetry reasons the component of $\tilde{\Omega}_i$ that is perpendicular to $(R^{-1})_{ij}n_{0,j}$ is taken in Eq. (15).

As Eq. (15) shows, the matrix $\mathbf{S}$ in general does not have to be determined explicitly for practical purposes. This is because usually for a given problem the two independent variables that define the current state of $\hat{\mathbf{n}}(\mathbf{r})$ have to be found as a solution to the problem, and because the ground state conformation $\hat{\mathbf{n}}_0(\boldsymbol{a}(\mathbf{r}))$ is known as an initial condition in general. In addition, the three components of $\boldsymbol{a}(\mathbf{r})$, or those of $\mathbf{u}(\mathbf{r})$, must be determined in a separate step. Including Eqs. (2) and (13) we have thus expressed the variables of relative rotations in the five independent variables describing the current state of the SCLSCE.

If we only consider the terms in Eqs. (13) and (15) that are linear in $\boldsymbol{\nabla}\mathbf{u}$, we obtain Eq. (4). Thus in the case of small deformations we recover the expressions for the relative rotations as noted in Refs, 6 and 7 in the linear theory. The dependences on the coordinates $\mathbf{r}$ in the final frame have been omitted in the notation of the matrices above.

At the end of this section we want to derive an exact expression for the relative rotations. For this purpose we have to find an exact expression for $\mathbf{R}^{-1}$, which can then be introduced into Eq. (15).

Since Ξ is symmetric it follows from Eq. (10) that $\boldsymbol{\varepsilon}$ and Ξ are diagonalized by the same matrix $\mathcal{R}$,

$$\boldsymbol{\varepsilon} = \mathcal{R}\,\boldsymbol{\varepsilon}^D\mathcal{R}^T, \qquad \Xi = \mathcal{R}\,\Xi^D\mathcal{R}^T, \qquad (16)$$

where in our notation diagonalized matrices are marked by $\cdot^D$ and transposed matrices by $\cdot^T$. Since we know $\boldsymbol{\varepsilon}$ from Eq. (2) we can calculate exact expressions for its eigenvalues e_i ($i = 1, 2, 3$) and its eigenvectors. From the eigenvalues e_i the eigenvalues of Ξ follow with the help of Eq. (10) as $X_i = \sqrt{1 - 2e_i}$ ($i = 1, 2, 3$). The eigenvectors of $\boldsymbol{\varepsilon}$ lead us to an exact expression for $\mathcal{R}$.

Introducing all these ingredients into Eq. (9) we can calculate the exact expressions for the components of the rotation matrix $\mathbf{R}$

$$R_{ij} = (\partial_k a_i)\mathcal{R}_{kl}((\Xi^D)^{-1})_{lm}(\mathcal{R}^T)_{mj}. \qquad (17)$$

Here, the components of the matrix $(\Xi^D)^{-1}$ are simply given by $((\Xi^D)^{-1})_{ij} = X_i^{-1}\delta_{ij}$ (no summation over i in this for-

mula; $i, j \in \{1, 2, 3\}$). $\mathbf{R}^{-1}$ then of course follows as $\mathbf{R}^{-1} = \mathbf{R}^T$.

Concerning the existence of the expressions above no problems arise, and all expressions remain real. First, $\boldsymbol{\varepsilon}$ and Ξ are symmetric and thus can be diagonalized in real space. Next, we consider the relation $da_i = dr_k(\partial_k a_i)$ in the local principal frame of $\boldsymbol{\nabla}\boldsymbol{a}$. It reads $da_i = \lambda_i^{-1}dr_i$ ($i = 1, 2, 3$; no summation over i in this formula), λ_i^{-1} being the eigenvalues of $\boldsymbol{\nabla}\boldsymbol{a}$. Rewriting the latter equation by $dr_i = \lambda_i da_i$ implies that λ_i can be interpreted as the stretch of the system parallel to the ith principal axis ($i = 1, 2, 3$). For physical reasons $0 < \lambda_i < \infty$ (for a discussion of this point in the Lagrangian description see e.g. Ref. 15. Furthermore, in the principal frame of $\boldsymbol{\nabla}\boldsymbol{a}$ no rotations occur and thus $(\boldsymbol{\nabla}\boldsymbol{a})^D = \Xi^D$. Due to this fact $X_i = \lambda_i^{-1}$, which includes $0 < X_i < \infty$ and $e_i < \frac{1}{2}$ ($i = 1, 2, 3$).

Applying this procedure in the two-dimensional case we can derive a result which still can be written in a manageable form:

$$\mathbf{R}^{-1} = \frac{1}{2}\begin{pmatrix} A_- & \pm A_+ \\ \mp A_+ & A_- \end{pmatrix}\begin{pmatrix} X_1^{-1} & 0 \\ 0 & X_2^{-1} \end{pmatrix}$$
$$\times \begin{pmatrix} A_- & \mp A_+ \\ \pm A_+ & A_- \end{pmatrix}\begin{pmatrix} \partial_x a_x & \partial_x a_y \\ \partial_y a_x & \partial_y a_y \end{pmatrix}. \quad (18)$$

Here we used as abbreviations $A_\pm = \sqrt{1 \pm \alpha}$, with $\alpha = (\varepsilon_{xx} - \varepsilon_{yy})/e$ and $e = \sqrt{(\varepsilon_{xx} - \varepsilon_{yy})^2 + 4\varepsilon_{xy}^2}$. As defined above, we have $X_i = \sqrt{1 - 2e_i}$ ($i = 1, 2$), where the eigenvalues of $\boldsymbol{\varepsilon}$ read $e_{1;2} = \frac{1}{2}(\varepsilon_{xx} + \varepsilon_{yy} \mp e)$. From the multiple signs in Eq. (18) the upper sign has to be chosen in the case of $\varepsilon_{xy} > 0$ and the lower one for $\varepsilon_{xy} < 0$ at the local position $\mathbf{r}$. This ensures the right handedness of our coordinate system in the principal frame.

Introducing the rotation matrix into Eq. (15) then leads us to an exact expression for the variables of relative rotations in the two-dimensional case.

III. STATIC SHEAR DEFORMATION OF A NEMATIC SCLSCE

As already discussed in the introduction relative rotations play a role of major importance in a continuum model of SCLSCEs (see e.g. also Ref. 16).Up to now various unique effects of SCLSCEs have been characterized in a linear macroscopic description that directly result from relative rotations and cannot be explained without these.[9–11,16–18]

In this section we want to demonstrate that also qualitatively different nonlinear effects have to be attributed solely to relative rotations. For this purpose we investigate the case of a nematic SCLSCE under a static shear deformation. The geometry we have in mind is depicted in Fig. 2. In the ground state the nematic elastomer is oriented such that the mesogenic units are aligned on average parallel to the $\hat{\mathbf{x}}$ axis. Thus the director in its ground state conformation reads

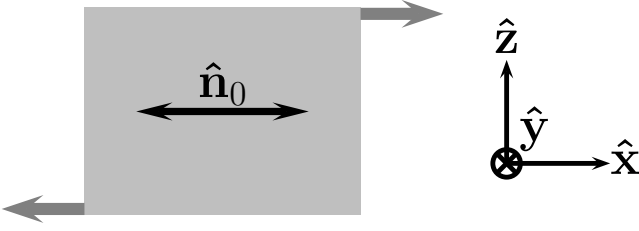$$\hat{\mathbf{n}}_0 = (1, 0, 0). \qquad (19)$$

FIG. 2: Geometry of the static shear deformation investigated in the text. The bulk volume element of the nematic SCLSCE is oriented such that $\hat{\mathbf{n}}_0 \parallel \hat{\mathbf{x}}$. The shear is applied within the x-z plane as indicated by the arrows.

This conformation is spatially homogeneous and we do not have to explicitly account for an $\boldsymbol{a}(\boldsymbol{r})$ dependence of $\hat{\mathbf{n}}_0$ according to Eq. (1), which simplifies the problem. In order to parametrize the current state of the director field we set

$$\hat{\mathbf{n}} = \left([1 - \sin^2 n_y - \sin^2 n_z]^{1/2}, \; \sin n_y, \; \sin n_z \right). \quad (20)$$

Here, n_y and n_z describe the angles between the director orientation and the planes of $y = 0$ and $z = 0$, respectively.

On the other hand we have to characterize the deformations of the polymer network by three independent variables. We will use the three components of the more intuitive field $\mathbf{u}(\mathbf{r})$, rather than those of $\boldsymbol{a}(\mathbf{r})$.

Furthermore, for illustration, we will only study the bulk effect of an external shear imposed on the elastomer. In other words we will neglect influences of the boundaries and only look for spatially homogeneous solutions of the director reorientation and the distortion of the polymer network. For this reason terms containing components of $\boldsymbol{\nabla}\hat{\mathbf{n}}$ are not explicitly listed in the following.

We now want to derive an expression for the generalized energy density F of the system. By minimizing the generalized energy $\mathcal{F} = \int F \, d^3r$ we then can find the current state of the elastomer under an imposed shear deformation.

The macroscopic variables that can contribute to the energy density F are on the one hand $\boldsymbol{\nabla}\hat{\mathbf{n}}$ and all the variables already present in the description of a simple liquid, which are the mass density ρ, the density of momentum of mass $\mathbf{g}$ and the density of entropy σ. On the other hand we have the components of the strain tensor ε_{ij} given by Eq. (2) and the components of the relative rotations $\tilde{\Omega}_i$ given by Eqs. (13), (15), (19) and (20). In order to obtain an expression for F we combine these variables taking into account symmetry requirements such as invariance under parity and under the transformation $\hat{\mathbf{n}} \rightarrow -\hat{\mathbf{n}}$. We include the uniaxial symmetry of the system, which is determined by $\hat{\mathbf{n}}$. Nevertheless, isotropic behavior is assumed for all the terms that are solely connected to the elastic behavior of the polymer network. In this way we guarantee that the behavior of the nematic SCLSCE we will predict is directly connected to the influence of the relative rotations and cannot be found by simply including the anisotropic elastic behavior of the polymer network. (In the Appendix we demonstrate that our results are not changed qualitatively if in addition an anisotropic elastic behavior of the polymer network is taken into account.)

It is then straightforward to write down a nonlinear convex expression for the generalized energy density of the system up to quartic order in the variables n_y, n_z, and in the components of $\boldsymbol{\nabla}\mathbf{u}$. However, if in the following for demonstrative purposes we are only interested in the small-amplitude first order corrections to the linear theory, quartic terms are negligible and it is a legitimate procedure to only consider terms up to cubic order. Therefore we do not write down explicitly a convex expression for the energy density, but quartic terms rendering the energy density convex can easily be included. Furthermore, in the illustrative example we will investigate below, the amplitude of the solution will be imposed onto the system externally. This additionally guarantees the stability of our solution.

We obtain

$$
\begin{aligned}
F &= F_0 + c_1 \varepsilon_{ij}\varepsilon_{ij} + \frac{1}{2}c_2 \varepsilon_{ii}\varepsilon_{jj} + \frac{1}{2}D_1 \tilde{\Omega}_i\tilde{\Omega}_i + D_2 \tilde{\Omega}_i\varepsilon_{ij}n_j + D_{2,n} n_i\tilde{\Omega}_i n_j\varepsilon_{jk}n_k + D_{2,tr} n_i\tilde{\Omega}_i \varepsilon_{jj} + \zeta_1 \varepsilon_{ii}\varepsilon_{jj}\varepsilon_{kk} \\
&\quad + \zeta_2 \varepsilon_{ii}\varepsilon_{jk}\varepsilon_{jk} + \zeta_3 \varepsilon_{ij}\varepsilon_{jk}\varepsilon_{ki} + \phi_1 n_i\varepsilon_{ij}\tilde{\Omega}_j \varepsilon_{kk} + \phi_2 n_i\varepsilon_{ij}\varepsilon_{jk}\tilde{\Omega}_k + \phi_3 n_i\varepsilon_{ij}\tilde{\Omega}_j n_k\varepsilon_{kl}n_l + \psi_1 \tilde{\Omega}_i\tilde{\Omega}_i \varepsilon_{jj} \\
&\quad + \psi_2 \tilde{\Omega}_i\varepsilon_{ij}\tilde{\Omega}_j + \psi_3 \tilde{\Omega}_i\tilde{\Omega}_i n_j\varepsilon_{jk}n_k + \text{hot},
\end{aligned} \quad (21)
$$

where hot stands for higher order terms not explicitly listed here.

In the first line F_0 incorporates all the terms that contain variables other than ε and $\tilde{\Omega}$. Their influence will not be studied in this section.

What follows are the quadratic terms with the coefficients c_1, c_2, D_1 and D_2, which are already well known from the linear theory[6,7]. There are two new quadratic terms with the coefficients $D_{2,n}$ and $D_{2,tr}$ in addition to the linear theory because in the nonlinear regime Eq. (6) does not apply anymore.

Next we listed the three cubic terms arising from the deformation of an isotropic elastic body[19] with coefficients ζ_i ($i = 1, 2, 3$), and afterward the cubic terms containing relative rotations are listed with coefficients ϕ_i and ψ_i ($i = 1, 2, 3$).
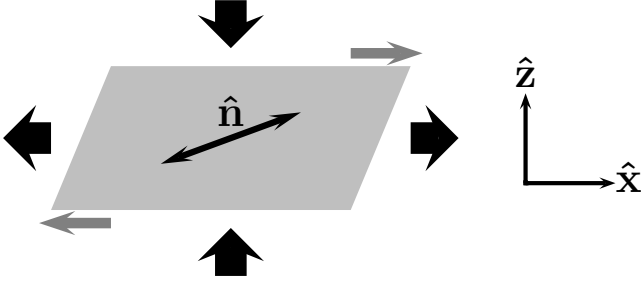
FIG. 3: Consequences of a static mechanical shear deformation of a bulk volume element of a nematic SCLSCE. The director is rotated within the plane of the applied shear. Additionally dilative and compressive strains occur, where the black arrows indicate one possible case of the resulting deformations. Relative rotations are the mediator between these effects.

The terms $\sim \psi_1$ and $\sim \psi_3$ can be thought of as modifications of the contribution $\sim D_1$, while the terms $\sim \phi_1$ and $\sim \phi_3$ can be viewed as modifications of the contribution $\sim D_2$. The components of ε and $\tilde{\Omega}$ have to be introduced into the cubic terms only to linear order, and as a consequence Eq. (6) applies. For this reason we did not list the cubic terms containing $n_i \tilde{\Omega}_i$.

Before turning to an example we want to recall once again that nonlinearities arising from Eq. (21) have two different sources: On the one hand these are the explicitly nonlinear cubic terms in Eq. (21). On the other hand, the quadratic terms contain nonlinear contributions because the nonlinear expression for ε and the new nonlinear expression for $\tilde{\Omega}$ derived in the preceding section must be inserted in these terms. That is the reason why the material parameters c_1, D_1 and D_2 will significantly contribute to the nonlinear results listed later on.

We now want to analyze the consequences of a shear deformation of the bulk of the nematic SCLSCE as indicated in Fig. 2. Denoting the shear amplitude by A_0 and looking only for homogeneous solutions due to the reasons elucidated above we make the Ansätze

$$u_x(\mathbf{r}) = A_0 z + A_1 x, \tag{22}$$
$$u_y(\mathbf{r}) = B_1 y, \tag{23}$$
$$u_z(\mathbf{r}) = C_1 z, \tag{24}$$
$$n_y(\mathbf{r}) = n_y, \tag{25}$$
$$n_z(\mathbf{r}) = n_z. \tag{26}$$

If we assume the system to be incompressible, which is a good approximation for the elastomers under investigation, we obtain

$$B_1 = \frac{A_1 C_1 - A_1 - C_1}{1 + A_1 C_1 - A_1 - C_1}. \tag{27}$$

Furthermore the terms with the coefficients ζ_1, ζ_2, ϕ_1 and ψ_1 are of higher order due to incompressibility and thus vanish in our consideration of the problem.

Since we study a spatially homogeneous solution, $\mathcal{F}$ is minimized simultaneously with F. Thus we can find the actual state of the system by solving the set of equations $\partial F / \partial A_1 = 0$, $\partial F / \partial C_1 = 0$, $\partial F / \partial n_y = 0$ and $\partial F / \partial n_z = 0$. For this purpose we expand the coefficients in the Ansätze (22)-(26) in a small parameter ϵ up to quadratic order,

$$A_0 = A_0^{(1)} \epsilon, \tag{28}$$
$$A_1 = A_1^{(1)} \epsilon + A_1^{(2)} \epsilon^2, \tag{29}$$
$$C_1 = C_1^{(1)} \epsilon + C_1^{(2)} \epsilon^2, \tag{30}$$
$$n_y = n_y^{(1)} \epsilon + n_y^{(2)} \epsilon^2, \tag{31}$$
$$n_z = n_z^{(1)} \epsilon + n_z^{(2)} \epsilon^2, \tag{32}$$

and introduce them into the set of equations. Here $A_0^{(1)} \epsilon$ has been used as an input. Up to quadratic order in ϵ we obtain the following results, which are also depicted in Fig. 3

The director $\hat{\mathbf{n}}$ acquires a z component with an angle

$$n_z = -\frac{D_1 + D_2}{2D_1} A_0. \tag{33}$$

As we can see this is an effect linear in the shear amplitude A_0, and indeed this effect has already been predicted by the linear theory[8]. Up to the order investigated there is no correction to this result for n_z: $n_z^{(2)} = 0$. Furthermore, we find that the director remains oriented within the x-z plane,

$$n_y = 0. \tag{34}$$

which is not surprising for a spatially homogeneous solution due to symmetry reasons. It turns out that $A_1^{(1)}$ and $C_1^{(1)}$ vanish identically. Thus, in addition to the reorientation of the director, we observe a compression and/or dilation of the SCLSCE parallel to the $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, and $\hat{\mathbf{z}}$ directions, described by

$$A_1 = \frac{A_0^2}{24 c_1 D_1^2} \left(4 c_1 D_1^2 + D_1 D_2^2 + D_2^3 - 2 D_{2,n} D_2^2 - 3 \zeta_3 D_1^2 + \phi_2 D_1 D_2 + 2 \phi_3 D_1 D_2 + \psi_2 D_2^2 - 2 \psi_3 D_2^2 \right), \tag{35}$$

$$B_1 = \frac{A_0^2}{48 c_1 D_1^2} \left(-16 c_1 D_1^2 + 2 D_1 D_2^2 + 2 D_2^3 + 2 D_{2,n} D_2^2 + 12 \zeta_3 D_1^2 - 4 \phi_2 D_1 D_2 - 2 \phi_3 D_1 D_2 + 2 \psi_2 D_2^2 + 2 \psi_3 D_2^2 \right), \tag{36}$$

$$C_1 = \frac{A_0^2}{48 c_1 D_1^2} \left(8 c_1 D_1^2 - 4 D_1 D_2^2 - 4 D_2^3 + 2 D_{2,n} D_2^2 - 6 \zeta_3 D_1^2 + 2 \phi_2 D_1 D_2 - 2 \phi_3 D_1 D_2 - 4 \psi_2 D_2^2 + 2 \psi_3 D_2^2 \right). \tag{37}$$

The amplitudes given by these strain coefficients are proportional to the square of the shear amplitude A_0. So they de-

scribe nonlinear effects, which cannot be predicted by a linear theory.

We can interpret these results in the following way. Due to the external mechanical shear deformation the director is reoriented, which is a linear effect arising from the coupling between the polymer network and the director, mediated by the relative rotations. However, this reorientation of the director itself acts back onto the polymer network and leads to compressive and/or dilative strains. From that point of view the nonlinear character of the compressive and/or dilative strain deformations becomes clear, and again this action of the director reorientation back onto the polymer network is mediated by the relative rotations. The described effects cannot be attributed to the anisotropy of the elastic behavior of the polymer network. We excluded anisotropies from all terms of F that are solely related to the elastic behavior of the polymer network. All the terms in Eqs. (33)-(37) directly depend on the coefficients of Eq. (21) that are connected to relative rotations. If in those terms of Eq. (21) containing relative rotations we furthermore take into account the isotropy of the elastic behavior of the polymer network, we have to set $D_{2,n}$, ϕ_3, and ψ_3 equal to zero. However, this does not affect our results qualitatively. On the other hand, as already mentioned, in the Appendix we investigated the effect of an anisotropic elastic behavior of the polymer network. Up to the inspected order we do not find a correction to the reorientation of the director field as given by Eqs. (33) and (34). The corrections to the expressions in Eqs. (35)-(37) do not change the results given above qualitatively. We also demonstrate in the Appendix that concentrating only on the anisotropic elastic behavior of the polymer network and neglecting relative rotations one does not recover the compressive and/or dilative deformations described above.

The interesting physics mainly occurs in the x-z plane of the system. In this plane the rotation of the director takes place, and the relative rotations between the director orientation and the polymer network also occur within this plane. The compression or dilation in the $\hat{\mathbf{y}}$ direction only results from the incompressibility condition (27) and influences the coefficients A_1 and C_1. We qualitatively obtain the same result concerning the physics in the x-z plane, if we treat the system as two dimensional. In this case and up to the order investigated, including incompressibility of the system, we obtain

$$n_z = -\frac{D_1 + D_2}{2D_1} A_0, \tag{38}$$

$$A_1 = A_0^2 \frac{D_2}{32 c_1 D_1^2} \left(2D_2^2 + 2D_2(D_1 - D_{2,n}) \right.$$
$$\left. + 2D_1 \phi_3 + 2D_2(\psi_2 - \psi_3) \right), \tag{39}$$

$$C_1 = -A_1. \tag{40}$$

Therefore the director reorients in the same way as described before. In the two-dimensional case compression or dilation in $\hat{\mathbf{x}}$ direction coincides with dilation or compression in $\hat{\mathbf{z}}$ direction, respectively. This is the situation indicated by the arrows in Fig. 3.

IV. DISCUSSION

When we were investigating the static shear deformation of the nematic SCLSCE in the last section, we used the components of the displacement field $\mathbf{u}(\mathbf{r})$ instead of those of $\boldsymbol{a}(\mathbf{r})$ in order to characterize the current state of the polymer network. This was done, because $\mathbf{u}(\mathbf{r})$ gives the more intuitive and illustrative variables, and this procedure is also convenient when considering static deformations. However, in anisotropic systems, the use of $\mathbf{u}(\mathbf{r})$ should be handled with care when dynamic deformations are studied because it can lead to erroneous results. This feature arises from the fact that the components of $\mathbf{u}(\mathbf{r})$ connect the space in which the initial/ground state of the system is described and the space in which its final state is characterized[13,14]. We can simply avoid this problem by replacing $\mathbf{u}(\mathbf{r})$ by $\mathbf{r} - \boldsymbol{a}(\mathbf{r})$ in the expressions of Sec. II. In doing this we correctly distinguish between initial space and final space.

We then obtain expressions for the macroscopic variables $\boldsymbol{\varepsilon}$ and $\tilde{\boldsymbol{\Omega}}$ which are even under parity in the initial as well as in the final space. For $\boldsymbol{\varepsilon}$ this can directly be confirmed from Eq. (2). Referring for example to Eq. (11) we can convince ourselves that the same also applies to $\boldsymbol{\Xi}$. Next, from Eq. (9) it follows that the inverse rotation matrix of the polymer network $\mathbf{R}^{-1}$ is odd under parity in the initial as well as in the final space, which is also implied by Eqs. (13) and (17). $\mathbf{R}^{-1}$ connects the two spaces, and more exactly we have to write it in the form $\mathbf{R}^{-1}(\boldsymbol{a}(\mathbf{r}), \mathbf{r})$. The other rotation matrix connecting initial and final spaces is $\mathbf{S}(\boldsymbol{a}(\mathbf{r}), \mathbf{r})$, which describes the local rotations of the director according to Eq. (7). Both matrices must explicitly depend on $\boldsymbol{a}(\mathbf{r})$ as well as on $\mathbf{r}$ because a local state in the final space is compared to the corresponding one in the initial space: For each volume element of the elastomer at position $\mathbf{r}$ in the final state the matrices must contain the information of how the polymer network and the director have been rotated compared to their initial state, in which the volume element was located at $\boldsymbol{a}(\mathbf{r})$. The states $\hat{\mathbf{n}}$ and $-\hat{\mathbf{n}}$ as well as the states $\hat{\mathbf{n}}_0$ and $-\hat{\mathbf{n}}_0$ are indistinguishable. Inserting Eq. (7) into Eq. (15) we see that the relative rotations $\tilde{\boldsymbol{\Omega}}$ are even under parity in the initial as well as in the final space. The same applies for the expression of the energy density [Eq. (21)].

In the last few years there have been several different attempts of finding nonlinear model descriptions of the specific properties of SCLSCEs. On the one hand, by a generalization of classical Gaussian rubber elasticity to the case of anisotropic polymer networks, some nonlinearities were included and the concept of "soft elastic distortions" was suggested (see e.g. Ref. 20). On the other hand, nonlinear macroscopic descriptions were also proposed (see e.g. Ref. (21), which again included the concept of soft elastic distortions[22]. A number of interesting experiments have been performed, and several of them have been interpreted in the context of these models. Examples are the reorientation process of the director field of a nematic SCLSCE when the material is stretched perpendicularly to the ground state director orientation[23], a thereby emerging stripe instability[24], and also reorientation processes of the director field in swollen nematic

SCLSCEs when exposed to an external electric field[25].

It has been demonstrated in Ref. 26 that the molecular model formally contains the D_1 and D_2 terms from Eq. (21) proposed in Ref. 6, however, the material parameters D_1 and D_2 are not independent in this model. Also the terms $\sim \psi_2$ and $\sim \psi_3$ are shown to be included but again with dependent coefficients[20]. However, the authors of Ref. 20 always use the linear expression of relative rotations as proposed in Ref. 6. Consequently, these authors conclude that the D_1 and D_2 terms cannot describe nonlinear effects, and cubic terms such as the ones $\sim \psi_2$ and $\sim \psi_3$ in Eq. (21) must be incorporated in a macroscopic characterization. On the contrary, by our illustrative example we demonstrated that the D_1 and D_2 terms can model such effects already, when an appropriate nonlinear expression is inserted for the relative rotations[27].

Our model qualitatively differs from the one proposed in Ref. 21 as we explicitly include nonlinear relative rotations between the polymer network and the director field as variables when we obtain the generalized energy density of the system. We derive for the first time a nonlinear expression for these relative rotations within the framework of a macroscopic theory, and we formulate it as a function of $\mathbf{u}(\mathbf{r})$ (or $\boldsymbol{a}(\mathbf{r})$) and $\hat{\mathbf{n}}(\mathbf{r})$, which completely describe the state of the material.

Furthermore, we do not incorporate the assumption of "soft elasticity" in our considerations. It has already been demonstrated in Ref. 28 by a constitutive model that the experimental results of Refs. 23 and 24 can also be obtained without the concept of soft elasticity. In our case, we have shown qualitatively by our illustrative example that the underlying effects are also covered by our model description without the assumption of soft elastic responses. In particular, as already mentioned, this means that the variables of relative rotations are explicitly included when setting up the expression for the energy density, and that the values of the material parameters are independent of each other. This renders the characterization of the materials more general.

V. CONCLUSIONS

In this paper we have shown how the linear macroscopic description of SCLSCEs can be extended to the nonlinear regime. For this purpose we have derived the nonlinear expression for the variables of relative rotations. To achieve this, the major problem consisted of finding the matrix of local rotations of the polymer network $\mathbf{R}$. We demonstrated how this matrix can be approximated by a series expansion and how it can be determined using a local transformation to the principal system of the strain tensor $\boldsymbol{\varepsilon}$.

Using the example of a static shear deformation of a nematic SCLSCE we demonstrated that relative rotations be-

tween the director orientation and the polymer network are essential in the macroscopic description of many of the unique properties of SCLSCEs. Including into this description nonlinear contributions of the relative rotations, we could account for qualitatively different nonlinear properties of SCLSCEs, which improves our understanding of these materials. In our example of shearing a bulk volume element of a nematic SCLSCE in a plane that contains the director, a reorientation of the director within this plane occurs as a linear effect. By including nonlinearities, we found, in addition, compressive and dilative strains of the bulk volume element as nonlinear effects arising from the influence of the relative rotations. Finally, we have shown that our results are consistent with general symmetry requirements, and we have related them to the work already performed in the field.

As a perspective we emphasize that the nonlinear expressions for the relative rotations in Sec. II will also be helpful in the macroscopic description of other complex systems. For example, these expressions can directly be included in the macroscopic characterization of magnetic gels. In this case one only has to replace the components of the director $\hat{\mathbf{n}}$ by those of the unit vector of the local magnetization $\mathbf{m}$[1].

Clearly, a nonlinear macroscopic description which includes the nonlinear contributions of relative rotations will reveal more of the unique features of SCLSCEs.

APPENDIX: STATIC SHEAR DEFORMATION OF AN ELASTICALLY ANISOTROPIC SCLSCE

In Sec. III we studied the reaction of a nematic SCLSCE to an imposed static shear deformation. There, we concentrated on the role the relative rotations play during this kind of deformation. Because of that we assumed the elastic behavior of the polymer network to be isotropic. We now demonstrate that an anisotropic elastic behavior of the polymer network does not qualitatively change the results derived in Sec. III.

If we want to include an anisotropic elastic behavior of the polymer network into our description, we have to supplement our expression for the energy density (21) by some additional terms (compare e.g. Ref. 29)

$$c_3 \, \varepsilon_{ii} \, n_j \varepsilon_{jk} n_k + c_4 \, n_i \varepsilon_{ij} \varepsilon_{jk} n_k + c_5 \, n_i \varepsilon_{ij} n_j \, n_k \varepsilon_{kl} n_l + \zeta_4 \, \varepsilon_{ij} \varepsilon_{ij} \, n_k \varepsilon_{kl} n_l + \zeta_5 \, \varepsilon_{ii} \, \varepsilon_{jj} \, n_k \varepsilon_{kl} n_l + \zeta_6 \, \varepsilon_{ii} \, n_j \varepsilon_{jk} \varepsilon_{kl} n_l$$
$$+ \zeta_7 \, \varepsilon_{ii} \, n_j \varepsilon_{jk} n_k \, n_l \varepsilon_{lm} n_m + \zeta_8 \, n_i \varepsilon_{ij} \varepsilon_{jk} \varepsilon_{kl} n_l + \zeta_9 \, n_i \varepsilon_{ij} \varepsilon_{jk} n_k \, n_l \varepsilon_{lm} n_m + \zeta_{10} \, n_i \varepsilon_{ij} n_j \, n_k \varepsilon_{kl} n_l \, n_m \varepsilon_{mn} n_n. \tag{A1}$$

Following the procedure described in Sec. III, we obtain the results listed hereafter.

Concerning the reorientation of the director field, Eqs. (33) and (34) are recovered identically. We find, however, that the amplitudes of the compression and/or dilation of the SCLSCE as given by Eqs. (35)-(37) are slightly modified in the anisotropic case

$$A_1 = \frac{A_0^2}{8D_1^2(3c_1 + 2c_4 + 2c_5)}\Big(4c_1D_1^2 + D_1D_2^2 + D_2^3 - 2D_{2,n}D_2^2 - 3\zeta_3D_1^2 + \phi_2D_1D_2 + 2\phi_3D_1D_2 + \psi_2D_2^2 - 2\psi_3D_2^2$$
$$+ 4c_3D_1^2 + 6c_4D_1^2 + 2c_4D_1D_2 + 8c_5D_1^2 + 8c_5D_1D_2 - 4\zeta_4D_1^2 - 3\zeta_8D_1^2 - 2\zeta_9D_1^2\Big), \tag{A2}$$

$$B_1 = \frac{A_0^2}{16c_1D_1^2}\Big(-4c_1D_1^2 + D_1D_2^2 + D_2^3 + 3\zeta_3D_1^2 - \phi_2D_1D_2 + \psi_2D_2^2 - 2c_4D_1^2 - 2c_4D_1D_2 + \zeta_8D_1^2$$
$$+ \frac{c_1}{3c_1 + 2c_4 + 2c_5}\Big(-4c_1D_1^2 - D_1D_2^2 - D_2^3 + 2D_{2,n}D_2^2 + 3\zeta_3D_1^2 - \phi_2D_1D_2 - 2\phi_3D_1D_2 - \psi_2D_2^2 + 2\psi_3D_2^2$$
$$- 4c_3D_1^2 - 6c_4D_1^2 - 2c_4D_1D_2 - 8c_5D_1^2 - 8c_5D_1D_2 + 4\zeta_4D_1^2 + 3\zeta_8D_1^2 + 2\zeta_9D_1^2\Big)\Big), \tag{A3}$$

$$C_1 = \frac{A_0^2}{16c_1D_1^2}\Big(4c_1D_1^2 - D_1D_2^2 - D_2^3 - 3\zeta_3D_1^2 + \phi_2D_1D_2 - \psi_2D_2^2 + 2c_4D_1^2 + 2c_4D_1D_2 - \zeta_8D_1^2$$
$$+ \frac{c_1}{3c_1 + 2c_4 + 2c_5}\Big(-4c_1D_1^2 - D_1D_2^2 - D_2^3 + 2D_{2,n}D_2^2 + 3\zeta_3D_1^2 - \phi_2D_1D_2 - 2\phi_3D_1D_2 - \psi_2D_2^2 + 2\psi_3D_2^2$$
$$- 4c_3D_1^2 - 6c_4D_1^2 - 2c_4D_1D_2 - 8c_5D_1^2 - 8c_5D_1D_2 + 4\zeta_4D_1^2 + 3\zeta_8D_1^2 + 2\zeta_9D_1^2\Big)\Big). \tag{A4}$$

However, all terms appearing in these expressions directly depend on those coefficients of Eq. (21) that are directly related to relative rotations. This means that without including the variables of relative rotations compressive and dilative deformations are not found in this description at all. The material parameters ζ_5, ζ_6, and ζ_7 do not enter the expressions listed above because the respective terms in the energy density are of higher order due to incompressibility. From Eqs. (A2)-(A4) the special case of an isotropic elastic behavior of the polymer network is simply recovered by letting the coefficients appearing in expression (A1) tend to zero.

Overall we find that the results derived in this appendix for an elastically anisotropic nematic SCLSCE qualitatively coincide with those obtained in the isotropic case in Sec. III.

[1] S. Bohlius, H. R. Brand, and H. Pleiner, Phys. Rev. E **70**, 061411 (2004).

[2] H. Finkelmann, H.-J. Kock, and G. Rehage, Makromol. Chem., Rapid Commun. **2**, 317 (1981).

[3] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1993).

[4] J. Küpfer and H. Finkelmann, Makromol. Chem., Rapid Commun. **12**, 717 (1991).

[5] J. Küpfer, E. Nishikawa, and H. Finkelmann, Polym. Adv. Techn. **5**, 110 (1994).

[6] P. G. de Gennes, in *Liquid Crystals of One- and Two-Dimensional Order*, edited by W. Helfrich and G. Heppke (Springer, Berlin, 1980), p. 231 ff.

[7] H. R. Brand and H. Pleiner, Physica A **208**, 359 (1994).

[8] P. Martinoty, P. Stein, H. Finkelmann, H. Pleiner, and H. R. Brand, Eur. Phys. J. E **14**, 311 (2004).

[9] O. Müller and H. R. Brand, Eur. Phys. J. E **17**, 53 (2005).

[10] A. M. Menzel and H. R. Brand, Phys. Rev. E **75**, 011707 (2007).

[11] A. M. Menzel and H. R. Brand (to be published).

[12] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, 1995).

[13] H. Temmen, H. Pleiner, M. Liu, and H. R. Brand, Phys. Rev. Lett. **84**, 3228 (2000).

[14] H. Pleiner, M. Liu, and H. R. Brand, Rheol. Acta **39**, 560 (2000).

[15] R. W. Ogden, *Non-Linear Elastic Deformations* (Dover, Mineola, 1997).

[16] H. R. Brand, H. Pleiner, and P. Martinoty, Soft Matter **2**, 182 (2006).

[17] J. Weilepp and H. R. Brand, Europhys. Lett. **34**, 495 (1996).

[18] A. M. Menzel and H. R. Brand, J. Chem. Phys. **125**, 194704 (2006).

[19] W. P. Mason, *Physical Acoustics and the Properties of Solids* (Van Nostrand Reinhold, New York, 1958).

[20] M. Warner and E. M. Terentjev, *Liquid Crystal Elastomers* (Clarendon, Oxford, 2003).

[21] T. C. Lubensky, R. Mukhopadhyay, L. Radzihovsky, and X. Xing, Phys. Rev. E **66**, 011702 (2002).

[22] L. Golubović and T. C. Lubensky, Phys. Rev. Lett. **63**, 1082 (1989).

[23] G. R. Mitchell, F. J. Davis, and W. Guo, Phys. Rev. Lett. **71**, 2947 (1993).

[24] I. Kundler and H. Finkelmann, Macromol. Rapid Commun. **16**, 679 (1995).

[25] K. Urayama, S. Honda, and T. Takigawa, Macromolecules **39**, 1943 (2006).

[26] P. D. Olmsted, J. Phys. II **4**, 2215 (1994).

[27] More precisely the authors of Ref. 20 refer to a situation corresponding to the experimental situation of e.g. Refs. 23 and 24, in which compression or dilation couples to the reorientation of the director field. In the linear description the D_2 term in Eq. (21) or in Ref. 6 does not contain this coupling, as linear relative rotations are always perpendicular to $\hat{\mathbf{n}}(\mathbf{r})$ [see Eq. (6)].

[28] E. Fried and S. Sellers, J. Appl. Phys. **100**, 043521 (2006).

[29] O. Müller, Diploma thesis, University of Bayreuth, 2001; H. R. Brand and O. Müller, Macromol. Theory Simul. **11**, 154 (2002); due to the transverse isotropy of the system only nine of

the ten terms with the coefficients ζ_i ($i = 1, ..., 10$) are independent.