A Finsler Geometry Modeling of the Liquid Crystal Elastomer

Hiroshi Koibuchi
Department of Mechanical and Systems Engineering, National Institute of Technology, Ibaraki College, Nakane 866, Hitachinaka, Ibaraki 312-8508, Japan
E-mail: koibuchi@mech.ibaraki-ct.ac.jp

Andrey Shobukhov
Faculty of Computational Mathematics and Cybernetics, Lomonosov Moscow State University, 119991, Moscow, Leninskiye Gory, MSU, 2-nd Educational Building, Russia

Liquid crystal elastomer (LCE) is a rubbery material composed of polymer chains and liquid crystals (LC). LCE is well known to undergo a shape transformation from the isotropic to the anisotropic phase. This shape transformation is caused by the nematic transition of the LC included in the LCE. However, the mechanism of this transformation is unknown because the interaction of LC with the bulk polymers is too complex. In this presentation, we extend the two-dimensional Finsler geometry model for membranes to a three-dimensional model for LCE. The Finsler geometry model for LCE is a coarse grained one: the Gaussian bond potential \( S_1 \) is obtained by extending the one for membranes, which is originally obtained by a simple extension of the Gaussian bond potential for the linear chain polymer model. The continuous Hamiltonian, which contains \( S_1 \) and the curvature energy \( S_2 \), is discretized using a three-dimensional rigid sphere composed of tetrahedrons. We study the shape transformation as a phase transition between the isotropic and anisotropic phases and report the results of the transition order, obtained by the Monte Carlo simulations.

Keywords: Liquid crystal elastomer, Finsler geometry, Anisotropy, Monte Carlo

1. Introduction
The linear chain model for polymer is defined by the Gaussian bond potential, which is a function of the sum of chain (or bond) length squares [1]. This model is extended to a model for membranes, where the extended Gaussian bond potential plays the role of surface tension energy [2]. Therefore it is natural to suppose that the Gaussian bond potential for membranes can also be extended to the one for the three-dimensional liquid crystal elastomer (LCE) [3].

The LCE is composed of polymer chains and liquid crystals (LC) and is well known because of its interesting properties such as the anisotropic shape transformation and the soft-elasticity [3]. At high temperature, the LC is in a random phase, and as a consequence the LCE has no preferred direction, and hence it is in the symmetric phase. On the contrary, as the temperature is lowered, the LCE starts to elongate in a spontaneously chosen direction. This phenomenon, which is the so-called anisotropic shape transformation, has its origin in the nematic transition of the constituent LC. However, the mechanism of the shape change is unknown. This is because...
the interaction of the LC and the bulk polymer is too complex. Therefore, a coarse grained model is helpful for better understanding of the LCE properties. In this paper, we extend the 2D Finsler geometry model for membranes [4] to a 3D model for LCE. The discrete Hamiltonian is defined on the spherical body, which is discretized by tetrahedrons. The curvature energy is assumed on the surface of the sphere with the bending rigidity $\kappa$. By using the Monte Carlo (MC) technique, we study the anisotropic shape transformation and its dependence of the surface elasticity, and report the order of the transition of the anisotropic shape transformation.

2. Model

![Figure 1](image_url)

**Figure 1.** (a) A sphere of size $N = 2431$ and (b) its sectional slice. (c) A tetrahedron in which the discrete Gaussian bond potential $S_1$ is defined, and (d) the variable $\sigma_i$ and the unit tangential vector $t_{ij}$ along the edge vector $\ell_{ij}$.

A sphere in $\mathbb{R}^3$ is randomly discretized by Voronoi tessellation (Fig. 1(a),(b)). In this sphere, the primitive object is the tetrahedron, which is composed of vertices, bonds, and triangles. The discrete model is defined by the Hamiltonian $S(\mathbf{r}, \sigma)$ which is given by

$$S(\mathbf{r}, \sigma) = \lambda S_0(\sigma) + S_1(\mathbf{r}, \sigma) + \kappa S_2(\mathbf{r}, \sigma) + U_{3D} + U_{2D},$$

$$S_0(\sigma) = \sum_{ij} (1 - \sigma_i \cdot \sigma_j),$$

$$S_1(\mathbf{r}, \sigma) = \frac{1}{4} \sum_{\text{tet}} \left( \gamma_{12} \ell_{12}^2 + \gamma_{13} \ell_{13}^2 + \gamma_{14} \ell_{14}^2 + \gamma_{23} \ell_{23}^2 + \gamma_{24} \ell_{24}^2 + \gamma_{34} \ell_{34}^2 \right),$$

$$S_2(\mathbf{r}) = \sum_{ij} (1 - \mathbf{n}_i \cdot \mathbf{n}_j),$$

$$U_{3D} = \sum_{\text{tet}} U_{3D}(\text{tet}), \quad U_{3D}(\text{tet}) = \begin{cases} 0 & (\text{Vol(\text{tet})} > 0) \\ \infty & (\text{otherwise}) \end{cases},$$

$$U_{2D} = \sum_{\Delta_i, \Delta_j} U_{2D}(i, j), \quad U_{2D}(i, j) = \begin{cases} 0 & (i, j \text{ intersect}) \\ \infty & (\text{otherwise}) \end{cases},$$

where $\mathbf{r} \in \mathbb{R}^3$ is the vertex position and $\sigma_i \in S^2$ (unit sphere) represents the LC defined at the vertex $i$. In $S_1$, $\ell_{ij}^2$ denotes the bond length squares between the vertices $i$ and $j$ (Fig.1(c)), and $\gamma_{ij}$ are defined by

$$\gamma_{12} = \frac{v_{12}}{v_{13}v_{14}} + \frac{v_{21}}{v_{23}v_{24}}, \quad \gamma_{13} = \frac{v_{13}}{v_{12}v_{14}} + \frac{v_{31}}{v_{32}v_{34}}.$$
\[ \gamma_{14} = \frac{v_{14}}{v_{12}v_{13}} + \frac{v_{41}}{v_{43}v_{42}}, \quad \gamma_{23} = \frac{v_{23}}{v_{21}v_{24}} + \frac{v_{32}}{v_{31}v_{34}}, \]
\[ \gamma_{24} = \frac{v_{24}}{v_{23}v_{21}} + \frac{v_{42}}{v_{41}v_{43}}, \quad \gamma_{34} = \frac{v_{34}}{v_{31}v_{32}} + \frac{v_{43}}{v_{41}v_{42}}. \]

The variable \( v_{ij} \) in Eq. (2) is the tangential component of \( \sigma_i \) along the bond \( ij \) (Fig.1(b))
\[ v_{ij} = |\mathbf{t}_{ij} \cdot \mathbf{\sigma}_i|, \quad \mathbf{t}_{ij} = \mathbf{\ell}_{ij}/l_{ij}. \]

Note that in general \( v_{ij} \neq v_{ji} \). The \( \sum_{tet} \) in \( S_1 \) denotes the sum over tetrahedrons. The symbol \( \mathbf{n}_i \) in \( S_2 \) denotes the unit normal vector of the triangle \( i \) on the surface, and \( \kappa \) is the bending rigidity. The potential \( U_{3D} \) protects the tetrahedron volume \( \text{Vol}(tet) \) from being negative. The potential \( U_{2D} \) also protects the surface (of a spherical body) from intersecting. We should note that the interaction of \( U_{3D} \) is local and the computation is not so time consuming, while that of \( U_{2D} \) is non-local and hence the computational time increases with increasing lattice size.

The partition function \( Z \) is given by
\[ Z(\lambda, \kappa) = \sum_{\sigma} \int \prod_{i=1}^N d\mathbf{r}_i \exp[-S(\mathbf{r}, \sigma)], \]

where \( \int \prod_{i=1}^N d\mathbf{r}_i \) are the multiple three-dimensional integrations, that are performed by fixing the center of mass to the origin of \( \mathbb{R}^3 \). \( \sum_{\sigma} \) denotes the sum over the spin variables \( \sigma \).

The continuous Gaussian bond potential \( S_1 \) is given by
\[ S_1 = \int \sqrt{g} d^3x g^{ab} \partial_a \mathbf{r} \cdot \partial_b \mathbf{r}, \]
where \( \mathbf{r} \) is a mapping from a three-dimensional space \( M \) to \( \mathbb{R}^3 \), \( g^{ab} \) is the inverse of the Finsler metric on \( M \), and \( g \) is its determinant. The discrete form of \( g_{ab} \) is given by
\[ g_{ab} = \begin{pmatrix} 1/v_{12}^2 & 0 & 0 \\ 0 & 1/v_{13}^2 & 0 \\ 0 & 0 & 1/v_{14}^2 \end{pmatrix}, \]

where \( v_{ij} \) are defined in Eq. (3).

The partial derivatives of \( \mathbf{r} \) in \( S_1 \) are replaced by \( \partial_1 \mathbf{r} \rightarrow \mathbf{r}_2 - \mathbf{r}_1 \), \( \partial_2 \mathbf{r} \rightarrow \mathbf{r}_3 - \mathbf{r}_1 \), and \( \partial_3 \mathbf{r} \rightarrow \mathbf{r}_4 - \mathbf{r}_1 \) on the tetrahedron in Fig. 1(c), where the coordinate origin is at the vertex 1. Since there are four possible coordinate origins in a tetrahedron, the partial derivatives \( \partial_a \mathbf{r} \) have several different discrete forms depending on the coordinate origin. Thus, summing over all possible forms of \( \partial_a \mathbf{r} \), we have \( S_1 \) in Eq. (1).

3. Simulation Results

The model has a continuous transition of shape transformation between the spherical phase and the oblong phase shown in Figs. 2(a), (b), where the oblong direction in Fig. 2(b) is chosen spontaneously. The bending energy \( S_2/N_B^2 \) on the surface rapidly changes in the region of \( \lambda \approx 0.23 \) in Fig.2(c), where \( N_B^2 \) is the total number of bonds on the surface. The specific heat for \( S_2 \) is defined by
\[ C_{S_2} = (1/N_S)(\langle S_2^2 \rangle - \langle S_2 \rangle)^2, \]

where the factor \( \kappa^2 \) is eliminated, and \( N_S \) denotes the total number of vertices on the surface. The peak value \( C_{S_2}^{\text{max}} \), growing with increasing \( N \), indicates the existence of a phase transition.
Figure 2. Snapshots in (a) the isotropic phase at $\lambda = 0.224$ and (b) anisotropic phase at $\lambda = 0.54$, where $\kappa = 1$ and $N = 4601$. (c) The two-dimensional bending energy $S_2/N_B'$ on the surface, and (d) the specific heat $C_{S_2}$.

The scaling coefficient for $C_{S_2}^{\text{max}} \sim (N_0)^{\alpha}$ is $\alpha = 0.44 \pm 0.06$. This implies that the transition is of second order. This continuous transition resembles the one of the two-dimensional surface model [5]. Also the order of this transition is consistent with that of two-dimensional self-avoiding surface model in [6], although the model in [6] is fluid. The difference between the LCE surface and the membrane surface is that the former surface has neither the crumpled phase nor the branched polymer phase.

4. Summary

We have presented preliminary results of a Finsler geometry model for 3D liquid crystal elastomer (LCE). The anisotropic shape transformation, caused by the nematic transition of the liquid crystal, is understood as a continuous transition in the model. We expect that the dependence of the LCE length $L$ on $\lambda$ (or inverse temperature) and the soft elasticity [3] such as the strain vs. stress of a cylindrical LCE, the height of which is fixed, are obtained from the Finsler geometry model. Three dimensional energy for the rigidity of LCE can also be included in the Hamiltonian. The detailed information of the model and those MC data will be reported elsewhere.

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