Geometry of Thin Nematic Elastomer Sheets

Hillel Aharoni and Eran Sharon
Racah Institute of Physics, The Hebrew University of Jerusalem, Jerusalem 91904, Israel

Raz Kupferman
Institute of Mathematics, The Hebrew University of Jerusalem, Jerusalem 91904, Israel

(Received 19 August 2014; published 17 December 2014)

A thin sheet of nematic elastomer attains 3D configurations depending on the nematic director field upon heating. In this Letter, we describe the intrinsic geometry of such a sheet and derive an expression for the metric induced by general nematic director fields. Furthermore, we investigate the reverse problem of constructing a director field that induces a specified 2D geometry. We provide an explicit recipe for how to construct any surface of revolution using this method. Finally, we show that by inscribing a director field gradient across the sheet's thickness, one can obtain a nontrivial hyperbolic reference curvature tensor, which together with the prescription of a reference metric allows dictation of actual configurations for a thin sheet of nematic elastomer.

DOI: 10.1103/PhysRevLett.113.257801 PACS numbers: 61.30.Vx, 02.40.Yy, 46.25.Cc, 46.70.De

The design of two-dimensional curved surfaces via the inscription of their intrinsic geometry has been a subject of extensive research in the last decade. Methods for inscribing surface geometries include the use of responsive hydrogels [1,2], the manipulation of biological tissues [3,4], and even crocheting [5]. A new approach, subject to much research in the last years, includes the incorporation of liquid crystals into elastic polymers [6,7]. Such materials exhibit many unusual properties including large deformations depending on the nematic order, unusual softness of certain deformation modes, and certain opto-mechanical peculiarities. We focus on thin sheets of nematic elastomers or glasses that were cross-linked while in the nematic phase. These sheets undergo large deformations when switching to the isotropic phase upon heating [8–10] or illumination [11–13]. Together with material properties, the nematic director field determines this deformation, thus setting the 2D intrinsic geometry of the sheet. The director field can be prescribed by pretreating the glass substrate in which the material is cast, either mechanically [14,15] or optically [16,17], or by applying an external electromagnetic field during cross-linking [10].

Theoretical work established the relation between the spatial distribution of the director field and the resultant intrinsic geometry in some specific cases. The local behavior, as well as the global behavior for a bulk of constant director field were studied [6,18]. Other works studied thin sheets with constant director field using a reduced two-dimensional model. Sheets with a gradient of either the nematic director field or the external stimuli across their thickness were found to have spontaneous curvature [19–22]. Other works study director fields resulting from a single defect in the nematic order [23,24] or from a collection of defects [25]. Such director fields, which were generated experimentally [16], lead to various 3D configurations of the sheets. However, the variety of possible director fields, and therefore 3D configurations that can be obtained, is in principle much larger. To our knowledge, no treatment has been given for the case of a general (smooth) director field. The inscription of such a field should be possible with existing experimental techniques such as the ones mentioned above, in which the thickness of the sheet is smaller than the surface extrapolation length.

In this Letter, we provide a general description for such systems. In order to describe the resulting geometry while avoiding unnecessary linearizations, we use the formalism of incompatible elasticity, previously presented in [26]. From this formalism, it is evident that the thin sheets may have no rest configuration, but will rather exhibit a competition between the stretching term (penalizing deviations of the 2D metric from its reference value) and the bending term (penalizing deviations of the surface curvature from its reference value). As the reference metric and curvature tensors may be incompatible, such competition results in a thickness-dependent equilibrium state of nonzero energy. In the case of liquid crystal elastomers, both the elastic shell stretching term and the elastic shell bending term contain contributions from both polymer elasticity and Frank energy associated with gradients in the nematic director field. Therefore, the reference metric and reference curvature result themselves from some competition between these contributions and may therefore depend upon external controls, e.g., the temperature. The (anisotropic) elastic moduli also depend upon these controls. However, in this Letter, we focus on the purely geometric problem of equilibrium configurations at the vanishing thickness limit, which is not affected by the elastic moduli.

Following [6] and others, our model assumes a liquid crystalline solid to expand (contract), upon some

environmental stimulus, by a factor of $\alpha^{1/2}$ along the director field and by a factor of $\alpha^{-\nu_t/2}$ along the perpendicular direction (where ν_t is the *thermal Poisson ratio*, see [25]). This description applies both for elastomeric materials (sparsely cross-linked) or glassy materials (densely cross-linked); however, the two classes differ in their typical values of α and ν_t. This deformation is the most general local deformation in 2D, our assumptions are that α and ν_t are constant throughout the sheet and that the principal expansion direction is aligned with the dictated director field. The process is illustrated in Fig. 1. A nematic sheet originally prepared on a Euclidean plane will therefore have, after stimulation, a reference metric of the form

$$\bar{a}(u,v) = R[\theta(u,v)]\begin{pmatrix}\alpha & 0\\ 0 & \alpha^{-\nu_t}\end{pmatrix}R[\theta(u,v)]^T, \quad (1)$$

where $R[\theta]$ is the 2D rotation matrix and $\theta(u,v)$ is the nematic director field. Equation (1) assumes that (u,v) is a Cartesian coordinate system; hence, the metric prior to the deformation was represented by the identity matrix. The director field θ is assumed to be smooth, but might contain isolated defects, in which the field is smooth everywhere except for a small region around the defect. We shall also assume constant ambient conditions so that α is constant throughout the sheet. Prior to stimulation, $\alpha = 1$ and the metric is Euclidean. We can rewrite Eq. (1) in terms of the scalar field $\phi(u,v) = \tan\theta(u,v)$:

$$\bar{a}(u,v) = \frac{1}{\phi^2+1}\begin{pmatrix}\alpha+\alpha^{-\nu_t}\phi^2 & (\alpha-\alpha^{-\nu_t})\phi\\ (\alpha-\alpha^{-\nu_t})\phi & \alpha\phi^2+\alpha^{-\nu_t}\end{pmatrix}. \quad (2)$$

Equation (2) expresses the metric of the 2D nematic elastomer sheet in terms of its director field. The reference Gaussian curvature of the sheet (determined by $\bar{a}$ through *Gauss' Theorema Egregium*) is

$$\bar{K}_{\bar{a}}(u,v) = (\alpha-\alpha^{-\nu_t})\left[\frac{(\phi^2-1)\phi_{uv}-\phi(\phi_{uu}-\phi_{vv})}{(\phi^2+1)^2} + \frac{(3\phi^2-1)(\phi_u^2-\phi_v^2)-2\phi(\phi^2-3)\phi_u\phi_v}{(\phi^2+1)^3}\right]. \quad (3)$$

We now exemplify our formalism by examining two classes of director fields. The first will be the director field of a point disclination, that was previously studied in [25]. The director field for a point disclination of charge m is of the form $\theta(u,v) = m\arctan(v/u)$. Using Eq. (3), we obtain the Gaussian curvature field resulting from this point defect:

$$\bar{K}_{\bar{a}}^{\mathrm{def}}(r,\varphi) = (\alpha^{\nu_t}-\alpha^{-1})m(m-1)\frac{\cos[2(m-1)\varphi]}{r^2}, \quad (4)$$

where $r = \sqrt{u^2+v^2}$ and $\varphi = \arctan(v/u)$ are the polar coordinates. Equation (4) is exactly as was obtained in [25], showing indeed that nontrivial Gaussian curvature is induced not only at the point defect, but also everywhere around it for $m \neq 1$.

Our second example involves a continuously varying director field, i.e., without any defects. Assume a director field that only depends on one of the coordinates, say u. Equation (3) for the Gaussian curvature simplifies into

$$\bar{K}_{\bar{a}}^{\mathrm{sym}}(u) = (\alpha-\alpha^{-\nu_t})\left[\frac{3\phi^2-1}{(\phi^2+1)^3}\phi_u^2 - \frac{\phi}{(\phi^2+1)^2}\phi_{uu}\right] = \frac{\alpha-\alpha^{-\nu_t}}{2}\left(\frac{1}{\phi^2+1}\right)_{uu}. \quad (5)$$

Equation (5) is analytically solvable for ϕ for any given $\bar{K}_{\bar{a}}(u)$. This implies that any such symmetric geometry can be "reverse engineered" to find a director field that will induce that geometry. Indeed, reverse engineering in such a symmetric case is feasible, and we discuss it below.

Rather than looking at the metric emerging from some given director field, in the following, we construct a director field given a desired metric (as was done for hydrogel systems in [27]). This problem is important when attempting to implement nematic elastomers for methods of design. We start by inducing a metric which is symmetric along one of the coordinates, i.e., embeddable as a surface of revolution. Suppose we are given a metric of the form:

$$\bar{a}(\xi,\eta) = \Omega^2(\xi)\begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix}. \quad (6)$$

This form of the metric is known as isothermal. Every two-dimensional metric can be locally brought to this form [generally, with $\Omega(\xi,\eta)$] by a proper choice of coordinates [28]. For a surface of revolution given by the radius R as a function of the height z, defining $\xi \equiv \int(\sqrt{1+R'^2}/R)dz$ and setting η to be the azimuthal angle, we obtain a metric in the form Eq. (6).

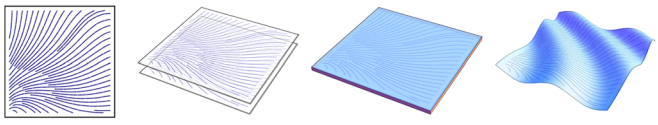

FIG. 1 (color online). Inscription of geometry of a nematic elastomer sheet (left-to-right). The director field can be imposed by substrate pretreatment or by applying an electromagnetic field. After cross-linking, we are left with an elastic plate, within which is a nematic liquid crystal with a prescribed director field. Upon heating, the material switches to the disordered state and the resulting sheet locally expands or contracts by a factor $\sqrt{\alpha}$ in the direction parallel to the director field and by a factor $\sqrt{\alpha^{-\nu_t}}$ in the perpendicular direction. This (in general) results in a non-Euclidean two-dimensional metric, which causes out-of-plane deformations if the sheet is thin enough.

We construct a new set of coordinates (u, v) defined by

$$u(\xi,\eta) \equiv \alpha^{-(1-\nu_t)/2} \int \Omega^2(\xi) d\xi,$$
$$v(\xi,\eta) \equiv \eta - \alpha^{-(1-\nu_t)/2} \int \sqrt{(\alpha - \Omega^2(\xi))(\Omega^2(\xi) - \alpha^{-\nu_t})}\, d\xi. \tag{7}$$

It is easy to verify that in these new coordinates the metric takes the form Eq. (2) with

$$\phi^2(u,v) = \frac{\Omega^2(\xi(u)) - \alpha^{-\nu_t}}{\alpha - \Omega^2(\xi(u))}, \tag{8}$$

where $\xi(u)$ is the inverse function of $u(\xi)$ given by Eq. (7). Equation (8) gives us an explicit recipe for the manufacture of any metric of the type Eq. (6), i.e., any surface of revolution. A few examples are given in Fig. 2. It also gives us a clue as for the limitations of this construction. In order for this scheme to work, we need the conformal factor Ω^2 to be within the range $[\alpha^{-\nu_t}, \alpha]$, as ϕ^2 needs be positive. This can be guaranteed in a small enough neighborhood of every point, but may pose a global problem. The larger the expansion factor α is, the easier it is to satisfy this constraint.

As an example, we construct a (pseudo)spherical surface with constant Gaussian curvature K_0. This suggests $\Omega(\xi) = A/\cosh(A\sqrt{K_0}\xi)$, where A is a constant and $\sqrt{K_0}$ is either real or imaginary (depending on the sign of K_0). From Eq. (8), we obtain the director field:

$$\theta(u) = \arctan \sqrt{\frac{\alpha^{1-\nu_t} K_0 u^2 - A^2(A^2 - \alpha^{-\nu_t})}{A^2(A^2 - \alpha) - \alpha^{1-\nu_t} K_0 u^2}}, \tag{9}$$

which can now be imprinted onto a flat surface in order to get a thin sheet that becomes (pseudo)spherical upon heating (Fig. 2). The constant A may be chosen to best suit our needs, and specifically, to maximize the size of the domain in which $\Omega^2 \in [\alpha^{-\nu_t}, \alpha]$.

We now relieve the symmetry along one of the coordinates and examine the more general case. As demonstrated in the symmetric case, the main task in designing a director field resulting in a desired metric is to find a coordinate system in which the metric tensor is of the form Eq. (1). We can find this coordinate transformation, without solving for the director field $\phi(u, v)$, by satisfying the two equations

$$\det(J^T \bar{a} J) = \alpha^{1+\nu_t}, \tag{10a}$$
$$\mathrm{tr}(J^T \bar{a} J) = \alpha + \alpha^{-\nu_t}, \tag{10b}$$

where J is the Jacobian of the coordinate transformation. As before, we shall assume that the metric is given in its isothermal form:

$$\bar{a}(\xi,\eta) = \Omega^2(\xi,\eta) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \tag{11}$$

(otherwise, a coordinate transformation that will bring it to that form can be found by solving a Beltrami equation [28]). Equations (10) now take the form

$$u_\xi v_\eta - u_\eta v_\xi = \alpha^{(\nu_t - 1)/2} \Omega^2, \tag{12a}$$
$$u_\xi^2 + u_\eta^2 + v_\xi^2 + v_\eta^2 = (\alpha^{\nu_t} + \alpha^{-1}) \Omega^2. \tag{12b}$$

We can rewrite Eqs. (12) in terms of the complex function $w(z)$, where $w = u + iv$ and $z = \xi + i\eta$:

$$\left|\frac{\partial w}{\partial z}\right| = P(z), \tag{13a}$$
$$\left|\frac{\partial w}{\partial \bar{z}}\right| = t \left|\frac{\partial w}{\partial z}\right|, \tag{13b}$$

where $t \equiv |(\alpha^{\nu_t/2} - \alpha^{-1/2})/(\alpha^{\nu_t/2} + \alpha^{-1/2})|$ and $P : \mathbb{C} \to \mathbb{R}^+$ is defined by $P(\xi + i\eta) = \frac{1}{2}|\alpha^{\nu_t/2} + \alpha^{-(1/2)}|\Omega(\xi,\eta)$. Note that $0 \le t < 1$, with $t = 0$ only in the trivial case, $\alpha = 0$ or $\nu_t = -1$.

We see from Eqs. (13) that $w(z)$ is some (extremal) quasiconformal mapping; however, it is difficult to find in the general case. One possible tactic for dealing with Eqs. (13) is to consider the Beltrami equation

$$\frac{\partial w}{\partial \bar{z}} = t e^{i\varphi(z)} \frac{\partial w}{\partial z}, \tag{14}$$

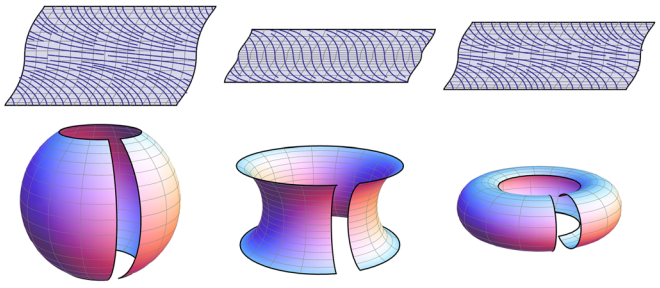

FIG. 2 (color online). Director field (top) and resulting surface of revolution (bottom) for $\alpha = 2$ and $\nu_t = 1$, for spherical ($\Omega(\xi) = \sqrt{2}/\cosh\xi$), pseudospherical ($\Omega(\xi) = \sqrt{1/2}/\cos\xi$), and toroidal ($\Omega(\xi) = \sqrt{2}/1 + (1 + \sqrt{5}/2)\sin^2\xi$) surfaces. The $\sqrt{2}$ factors were introduced in order to maximize the size of the domain, given that $\Omega(\xi)$ must, by Eq. (8), lie between $\sqrt{1/2}$ and $\sqrt{2}$. The curved boundaries of the planar domains at the top panel become longitudes of the resulting surfaces upon heating.

where $\varphi(z)$ is some real-valued function. The absolute value of this equation is Eq. (13b). Like all smooth Beltrami equations, Eq. (14) has a unique solution (modulo composition with analytic functions). This defines an operator B, which takes a real-valued phase function $\varphi(z)$ and returns $P(z)$, which is the absolute value of the partial derivative of the solution of the corresponding Beltrami Eq. (14). The operator B needs then to be inverted, in order to find the phase $\varphi(z)$ which gives the desired $P(z)$, so that both Eq. (13a) and Eq. (13b) are satisfied.

For example, for any harmonic $\varphi(z)$, we define

$$f = \int e^{-(i/2)[\varphi(z)+i\tilde{\varphi}(z)]}dz, \tag{15}$$

where $\tilde{\varphi}$ is the harmonic conjugate of φ. f is obviously analytic. We observe that $w(z,\bar{z}) = f(z) + t\overline{f(z)}$ is a solution to Eq. (14). Therefore, for any harmonic function $\varphi(z)$:

$$B\varphi(z) = |w_z| = |f'(z)| = e^{(1/2)\tilde{\varphi}(z)}. \tag{16}$$

This relation can be easily inverted, which means we have a solution for any $P(z)$ [hence, $\Omega(z)$] whose logarithm is an harmonic function. However, this is somewhat unhelpful since such Ω's represent Euclidean metrics. Solutions for other phase functions $\varphi(z)$ can be found, analytically or numerically. These will give us a collection of metric functions [represented by their conformal factor $\Omega(z)$] that can be brought via coordinate transformation to the form Eq. (1), and can therefore be induced using liquid crystal elastomers. However, since we were unable to establish an existence theorem for solutions of Eq. (13), the mathematical question of whether such a coordinate system locally exists for *any* smooth metric remains open.

Finally, we briefly discuss the direct inscription of curvature on the sheet by prescribing different director fields on its top and bottom layers, as was done with uniform fields in [14,21,22]. Assuming director fields $\theta_\pm(u,v)$ at the top and bottom layers, we will obtain expressions $\bar{a}_\pm(u,v)$ of the form Eq. (1) for the lateral components of the metric on the top and bottom layers. To linear order in z (the perpendicular coordinate), the full 3D metric is of the form

$$\bar{g}(u,v) = \begin{pmatrix} \frac{1}{2}(\bar{a}_+ + \bar{a}_-) + \frac{z}{h}(\bar{a}_+ - \bar{a}_-) & 0 \\ 0 & 0 \end{pmatrix}, \quad 1 \tag{17}$$

hence, obtaining 2D metric and curvature tensors

$$\bar{a} = \frac{1}{2}(\bar{a}_- + \bar{a}_+), \tag{18a}$$

$$\bar{b} = \frac{1}{2h}(\bar{a}_- - \bar{a}_+). \tag{18b}$$

In terms of the average director field $\langle\theta\rangle = \frac{1}{2}(\theta_- + \theta_+)$ and the director field difference $\Delta\theta = (\theta_+ - \theta_-)$, Eq. (18) reads

$$\begin{aligned}\bar{a} &= R[\langle\theta\rangle]\left[\begin{pmatrix}\alpha & 0\\ 0 & \alpha^{-\nu_t}\end{pmatrix}\cos^2\left(\frac{\Delta\theta}{2}\right)\right.\\ &\quad\left.+\begin{pmatrix}\alpha^{-\nu_t} & 0\\ 0 & \alpha\end{pmatrix}\sin^2\left(\frac{\Delta\theta}{2}\right)\right]R[\langle\theta\rangle]^T\\ &= R[\langle\theta\rangle]\begin{pmatrix}\alpha & 0\\ 0 & \alpha^{-\nu_t}\end{pmatrix}R[\langle\theta\rangle]^T + O(\Delta\theta^2),\end{aligned} \tag{19a}$$

$$\begin{aligned}\bar{b} &= \frac{\alpha-\alpha^{-\nu_t}}{2h}R[\langle\theta\rangle]\begin{pmatrix}0 & \sin(\Delta\theta)\\ \sin(\Delta\theta) & 0\end{pmatrix}R[\langle\theta\rangle]^T\\ &= \frac{\alpha-\alpha^{-\nu_t}}{2h}R[\langle\theta\rangle]\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix}R[\langle\theta\rangle]^T\Delta\theta + O(\Delta\theta^3).\end{aligned} \tag{19b}$$

Thus, inscribing director fields $\theta_\pm(u,v)$ on the top and bottom surfaces results in a sheet with spontaneous metric $\bar{a}$ and spontaneous curvature $\bar{b}$ given by Eq. (19). These results hold even if we relax the assumption of director field linearity in the z direction [made in Eq. (17)]; however, the effective thickness h may slightly differ from the actual thickness. Dictating an average director field $[\theta]$ with difference $\Delta\theta$ allows us to design not only the metric but also the curvature tensor, hence, allowing us to differentiate between different isometries of the same metric, even for very thin sheets (Fig. 3).

From Eq. (19b), it is evident that only curvature tensors which are everywhere saddlelike can be induced. The principal curvature directions are at $\pm 45°$ from $\langle\theta\rangle$ and

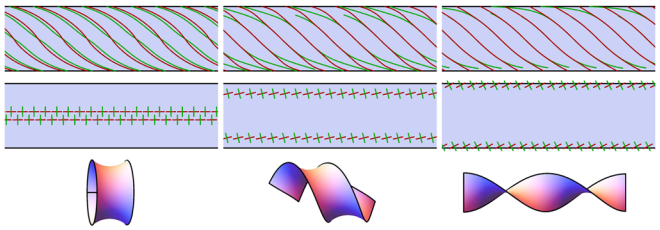

FIG. 3 (color online). Top—director field on the top (red) and bottom (green) surfaces, in all of which $\langle\theta\rangle$ is the same (hence, inscribing the same metric); however, $\Delta\theta$ differs (hence, inscribing different curvature tensors). Center—the resulting curvature field dictated (red and green mark the two principal curvatures). By applying different $\Delta\theta$ fields, we can "activate" the curvature tensor wherever its directions match the surface we wish to design. Bottom—the resulting surfaces. All surfaces are exact isometries of the inscribed metric (and of each other); however, their curvature tensors best fit the inscribed ones, and are therefore energetically preferable among isometries due to their lower bending energy.

the curvature magnitude is proportional to $\Delta\theta$, and therefore, these can be chosen independently (since we can independently control θ_+ and θ_-). The Gaussian curvature induced by the curvature tensor, which is implied by Eq. (19b), is everywhere nonpositive and reads:

$$\bar{K}_{\bar{b}}(u,v) = -\frac{\sin^2(\Delta\theta)}{h^2\left(\left(\frac{\alpha+\alpha^{-\nu_t}}{\alpha-\alpha^{-\nu_t}}\right)^2-\cos^2(\Delta\theta)\right)} = -\left|\frac{(\alpha^{(1+\nu_t)/2}-\alpha^{-(1+\nu_t)/2})\Delta\theta}{2h}\right|^2 + O(\Delta\theta^4) \quad (20)$$

(note the interesting contrast to sheets constructed by isotropic expansion, in which only non-negative Gaussian curvature can be dictated to the reference curvature tensor). It is also important to observe that sheets produced using that method are (generally) elastically incompatible and will be residually stressed for any $\alpha \neq 1$.

In conclusion, the prescription of local director fields in thin liquid crystalline solid sheets is one of the most promising techniques for programming inducible 3D configurations of thin sheets. Until now, there has been no complete theoretical work that links between the director field and the resultant sheet geometry. In this Letter, we developed a general framework for describing the intrinsic geometry of a thin nematic elastomer or glass sheet. We provided explicit relations between the nematic director field at preparation and the reference metric and curvature fields of the resultant elastic sheet. We investigated the inverse problem, trying to construct the director field needed to obtain a desired 2D metric. We obtained a Beltrami-like set of equations whose solution gives us the coordinate system in which such a director field exists. We demonstrated how to construct the 2D geometry of any surface of revolution using this method. Finally, we showed that by inscribing a director field gradient across the sheet's thickness, we can obtain a nontrivial reference curvature tensor. We proved that this reference curvature must be of negative Gaussian curvature everywhere and suggested a way to add it to the reference metric field in order to differentiate between isometries of the same 2D geometry. Such combinations of reference metric and curvature fields allow us to accurately dictate equilibrium configurations for the nematic elastomer thin sheets.

It is our hope that results described in this Letter will be used by experimental groups in the design of responsive elastic surfaces. More theoretical work is needed as well. Specifically, the mathematical question of which 2D metrics are (locally or globally) accessible via this method remains open. The mathematical formulation of this question in the form of Eq. (13) seems like a good starting point for any further investigation of this problem. Other extensions to this work may include homeotropic boundary conditions in addition to the planar ones, and the use of cholesteric or smectic liquid crystal elastomers.

We thank J. V. Selinger for useful discussions. H. A. and E. S. were supported by the Israel-US Binational Foundation (Grant No. 2008432) and by the European Research Council SoftGrowth project. R. K. was supported by the Israel-US Binational Foundation (Grant No. 2010129) and by the Israel Science Foundation (Grant No. 661/13).

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