Phase transitions and soft elasticity of smectic elastomers

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Smectic-C elastomers can be prepared by crosslinking, e.g., liquid crystal polymers, in the smectic-A phase followed by a cooling through the smectic-A to smectic-C phase transition. This transition from $D_{\infty h}$ to $C_{2h}$ symmetry spontaneously breaks rotational symmetry in the smectic plane as does a transition from a smectic-A to a biaxial smectic phase with $D_{2h}$ symmetry. We study these transitions and the emergent elasticity of the smectic-C and biaxial phases in three related models and show that these phases exhibit soft elasticity analogous to that of nematic elastomers.

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Smectic-C elastomers are remarkable materials that combine the orientational and positional order of liquid crystals with the elastic properties of rubber. The traditional liquid crystalline nematic, cholesteric, smectic-A (SmA), smectic-C (SmC), and smectic-C* phases all exist in elastomeric forms. In this letter we investigate the properties of SmC and biaxial phases in elastomers formed via spontaneous symmetry breaking from SmA or uniaxial phases and, within mean-field theory, the phase transitions to them. We introduce and analyze phenomenological models for these transitions involving strains only and a model involving strains and the Frank director specifying the direction of local molecular order. Our primary result is that monodomain samples of the emerging biaxial and SmC phases (see Fig. 1), both of which break the continuous rotational symmetry in the smectic layers, exhibit soft elasticity characterized by the vanishing of certain elastic moduli and the associated absence of restoring forces to strains along specific symmetry directions. As in monodomain nematic elastomers, this soft elasticity is a consequence of the Goldstone theorem that requires any phase with a spontaneously broken continuous symmetry to have modes whose energy vanishes with wavenumber.

To keep our discussion as simple as possible, we will consider only smectic elastomers crosslinked in the SmA phase so that the smectic layers are locked to the crosslinked matrix. Macroscopically, these SmA elastomers are simply uniaxial rubbers (or solids) with $D_{\infty h}$ symmetry, and we will treat them as such, though we will distinguish between the normal N to smectic layers and the direction of uniaxial anisotropy e.

We employ the usual Langrangian formalism in which mass points in the undistorted medium, which we take as the reference space, are labelled by vectors x. Mass points of the distorted medium are at positions $R(x) = x + u(x)$ in physical space, which we call the target space. Distortions of the reference medium are described by the Cauchy deformation tensor $A$ with components $A_{ij} = \partial R_i / \partial x_j \equiv \partial_i R_j$, $i, j = x, y, z$. Lagrangian elastic energies are expressed in terms of the nonlinear strain tensor $y$ with components $u_{ij} = 1/2 (\partial_i u_j + \partial_j u_i + \partial_i u_k \partial_j u_k)$, which transforms as a tensor in the reference space and which is invariant with respect rotations in the target space. The elastic free energy density of a uniaxial elastomer to harmonic order in strains can be expressed as

$$f_{\text{uni}} = \frac{1}{2} C_1 u_{zz}^2 + C_2 u_{zz} u_{ii} + \frac{1}{2} C_3 u_{ii}^2 + C_4 u_{ab}^2 + C_5 u_{az}^2,$$

(1)

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where we use the Einstein convention on repeated indices and where $z$ is along the uniaxial axis and indices at the beginning of the alphabet a, b, .. run over $x$ and $y$ only. The tensor $u_{ab} = u_{ab} - \frac{1}{3} \delta_{ab} u_{cc}$ is the two-dimensional symmetric, traceless strain tensor. The elastic constant $C_1$ describes dilation or compression along $z$. $C_4$ and $C_5$ respectively describe shears in the plane perpendicular to the anisotropy axis and in the planes containing it. Relative volume change $\delta V/V = \det A - 1$ can be approximated by $u_{ii}$ in our Landau expansion in powers of $u_{ij}$, and the incompressible limit relevant to most elastomers corresponds to $C_5 \to \infty$.

The harmonic free energy $f_{\text{uni}}$ describes any uniaxial solid or elastomer, including SmA elastomers. However, to provide a complete description of SmA and SmC elastomers, we need to add terms that describe the smectic layers and the Frank director and their interactions with each other and with the elastic medium. Before adding

![FIG. 1: Sample distortion and rotations of the Frank director n, the uniaxial anisotropy axis e, and the layer normal N in a transition from (a) SmA to (b) a sheared SmC elastomer. In this geometry, the the smectic layers do not rotate. The figure corresponds to a shear strain $\gamma \approx u_{yz} < 0$ (see text).](image)
these terms, we will first investigate phase transitions in uniaxial elastomers in which either of the elastic constants \(C_4\) or \(C_5\) go to zero. The vanishing of \(C_4\) and \(C_5\) lead, respectively, to phases with \(D_{2h}\) (orthorhombic) and \(C_{2h}\) (triclinic) symmetry. We will then consider the smectic layers and director and show that the transition to the SmC phase, which has \(C_{2h}\) symmetry, is in fact identical to the one in which \(C_5\) goes to zero.

**Strain-only theory for \(C_4 \to 0\):** If \(C_4\) or \(C_5\) become negative, as they will in response to an instability toward biaxial or SmC ordering of constituent mesogens, terms higher order in strains must be added to Eq. \((1)\) to ensure mechanical stability. If \(C_4\) becomes negative, order of the shear strain \(\delta_{ab}\) sets in and higher order terms featuring \(\delta_{ab}\) have to be added which leads to the model elastic energy

\[
f_{\text{uni}}^{(1)} = f_{\text{uni}} + A_1 u_{zz}^2 + A_2 u_{ii}^2 u_{ab} + B (u_{ab}^2)^2, \tag{2}
\]

where we have dropped qualitatively inconsequential higher order terms. It will be useful in the analysis that follows to regroup the terms in \(f_{\text{uni}}^{(1)}\) by completing the squares in \(\frac{1}{2} C_1 u_{zz}^2 + A_1 u_{zz}^2 u_{ab}\), etc., and to reexpress it as a sum of two terms

\[
f_{\text{uni}}^{(1,1)} = \frac{1}{2} C_1 v_{zz}^2 + C_2 v_{zz} v_{ii} + \frac{1}{2} C_3 v_{ii}^2 + C_5 u_{zz}^2; \tag{3}
\]

\[
f_{\text{uni}}^{(1,2)} = C_4 u_{ab}^2 + B R (u_{ab}^2)^2, \tag{4}
\]

where \(v_{zz} = u_{zz} - \alpha_1 u_{ab}^2\), \(v_{ii} = u_{ii} - \beta u_{ab}^2\), and where \(\beta\), which vanishes as \(C_3 \to 0\), is unit vector in the \(xy\) plane, is determined by minimizing \(f_{\text{uni}}^{(1,2)}\). The result is \(S = 0\) for \(C_4 > 0\), and

\[S = \pm \sqrt{-C_4 B R} \text{ for } C_4 < 0.\]

The equilibrium strain \(\vartheta = \frac{1}{2} (\Delta T - \bar{\Delta} - \delta)\) is diagonal with components \(u_{xx}^0 = \frac{1}{2} S + \frac{1}{2} (\beta - \alpha) S^2\), \(u_{yy}^0 = -\frac{1}{2} S + \frac{1}{2} (\beta - \alpha) S^2\), and \(u_{zz}^0 = \frac{1}{2} \alpha S^2\). The new state is biaxial with \(D_{2h}\) symmetry.

To determine the elastic properties of the new state, we expand \(f_{\text{uni}}^{(1)}\) in powers of \(\delta_{ab} = u - u^0\). Since the equilibrium values of \(v_{zz}, v_{ii}\) and \(u_{ab}\) are zero, the expansion of \(f_{\text{uni}}^{(1)}\) is trivial. The structure of \(f_{\text{uni}}^{(1,2)}\) is identical to that of an \(xy\) model, and it has no restoring force in the ordered phase for \(\delta u_{xy}\): \(\delta f_{\text{uni}}^{(1,2)} = B R S^2 (\delta u_{xy} - \delta u_{xy})\). Thus it is clear that \(\delta f_{\text{uni}}^{(1)}\) does not depend on \(\delta u_{xy}\) to harmonic order, i.e., the system is soft with respect to shears in the \(xy\) plane of the original reference material.

The strain \(\delta u\) describes distortions relative to the new biaxial reference state measured in the coordinates of the original uniaxial state. It is customary, however, to express the elastic energy in terms of a strain \(\vartheta' = (\Delta T)^{-1} \delta \vartheta (\Delta T)^{-1}\) measured in the coordinates \(x_i' = x_i + u_i^0 = \Lambda_0 x_i\) of the new state. \(\Lambda_0\) is not uniquely determined by \(\vartheta\); rotations in the target space change \(\Lambda_0^0\) but do not change \(\vartheta^0\). In the present case, it is natural to choose \(\Lambda_0^0\) to be diagonal, i.e., not to rotate the strain field after the transition. The elastic energy of the biaxial state to harmonic order in the new strains is then

\[
f_{\text{uni}}^{(2)} = \frac{1}{2} C_{zzzz} (u_{zz}^2) + C_{xxzz} (u_{xx}^2) + C_{yzz} (u_{yy}^2) + C_{zzzx} u_{xx}' u_{xx} + C_{xxyy} u_{zz}' u_{yy} + \frac{1}{2} C_{zzxx} (u_{xx}')^2 + \frac{1}{2} C_{yyyy} (u_{yy}')^2 + C_{zzxy} u_{xx}' u_{yy}' \tag{5}
\]

where the elastic constants depend on the original elastic constants featured in Eq. \((2)\) and the order parameter \(S\), and for \(S \to 0\) this energy reduces to the uniaxial energy \((\text{1})\). If we take the incompressible limit, \(C_3 \to \infty\), the specifics of the new elastic constants are affected but the form of the elastic energy \((\text{4})\) remains the same.

Because there was no \(\delta u_{xy}\) term in the expansion of \(f_{\text{uni}}^{(1)}\), there is no term proportional to \(u_{zz}^2\), as there would be in conventional orthorhombic systems. Thus, there is no restoring force to \(xy\)-stresses, i.e., to opposing forces along \(\pm x\) applied to opposite surfaces perpendicular to \(y\) or opposing forces along \(\pm y\) applied to opposite surfaces perpendicular to \(x\). In addition, as is the case for stresses perpendicular to the anisotropy axis in a nematic elastomer \((\text{14} \text{ 15})\), it requires no stress to stretch the sample along \(y\) direction, up to a critical strain value. The same soft mode and many more were predicted by Warner and Kutter \((\text{3})\) for biaxial nematics forming spontaneously from an isotropic elastomer.

**Strain-only theory for \(C_5 \to 0\):** When \(C_5\) is driven negative the uniaxial state becomes unstable to shear in the planes containing the anisotropy axis, and the uniaxial energy \((\text{1})\) must be augmented with higher order terms involving \(u_{zz}\) to stabilize the system, namely

\[
f_{\text{uni}}^{(3)} = f_{\text{uni}} + D_1 u_{zz} u_{zz}^2 + D_2 u_{ii} u_{zz}^2 + D_3 u_{ab} u_{zz} u_{zz}^2 + E (u_{zz}^2)^2, \tag{6}
\]

where we omitted all unimportant symmetry-compatible higher order terms. To study the ordered phase of this free energy when \(C_5 < 0\), we proceed in much the same way as we did for the biaxial state of \(f_{\text{uni}}^{(1)}\). We complete squares to write \(f_{\text{uni}}^{(2)}\) as the sum of two terms:

\[
f_{\text{uni}}^{(2,1)} = \frac{1}{2} C_1 u_{zz}^2 + C_2 w_{ii} u_{zz} + \frac{1}{2} C_3 w_{ii}^2 + C_4 w_{ab}^2, \tag{7}
\]

\[
f_{\text{uni}}^{(2,2)} = C_5 u_{zz}^2 + E R (u_{zz}^2)^2, \tag{8}
\]

where \(w_{zz} = u_{zz} - \sigma u_{zz}^2\), \(w_{ii} = u_{ii} - \tau u_{zz}^2\), and \(u_{ab} = u_{ab} - \omega (u_{zz} u_{zz} - \frac{1}{2} \delta_{ab} u_{zz}^2)\) where \(\sigma, \tau \to 0\) as \(C_5 \to \infty\), \(\omega, \) and \(E\) are functions of the parameters in \(f_{\text{uni}}^{(2)}\). The equilibrium value of \(u_{zz}\) is determined by minimizing \(f_{\text{uni}}^{(2)}\), which has \(xy\) symmetry. For \(C_5 > 0\), \(u_{zz} = 0\); for \(C_5 < 0\) and order along \(x\), \(u_{zz}^0 \equiv S = \pm \sqrt{-C_5 / (2 E R)}\), and \(u_{yz} = 0\). The other components of \(u_{zz}^0\) are \(u_{zz}^0 = \sigma S^2,\)
\[ u_{xy}^0 = \frac{1}{2}(\tau - \omega)S^2, \] and \[ u_{yy}^0 = \frac{1}{2}(\tau + \omega)S^2. \] Unlike the biaxial case, \( u^0 \) is not diagonal; it has nonvanishing \( zz \) and \( xx \) components that lead to \( D_{2h} \) rather than \( D_{3h} \) symmetry. Expanding \( f_{uu}^{(2)} \) in powers of \( \delta u = \mathbf{u} - \mathbf{u}^0 \), we find that \( \delta f_{uu}^{(2)} = 4E_R S^2 (\delta u_{zz})^2 \) is independent of \( \delta u_{yz} \), and we might naively expect the system to exhibit softness with respect to \( u_{yz} \). This, however, is not the case because \( f_{uu}^{(2,1)} \) depends on \( \delta u_{yz} \) via the \( C_4 \) term: \[ 2C_4 (\delta u_{xy} - \omega S \delta u_{yz})^2. \] Thus, the softness of the ordered phase with \( D_{2h} \) symmetry is more subtle than that of the biaxial phase with \( D_{3h} \) symmetry.

To determine the energy of strains relative to the new ground state, we need to choose how we define our new coordinate system relative to it. It is easiest to visualize the new state as emerging from a simple shear as shown in Fig. 1 in which \( \Lambda^0_{zz} = \partial R^0_{xi}/\partial x \equiv \gamma \) is nonzero but \( \Lambda^0_{xx} = \partial R^0_{xi}/\partial x = 0 \). In this case, the only nonzero components of \( \Lambda^0 \) are the diagonal components and \( \Lambda^0_{zz} \) which can be expressed in terms of \( u^0: \) \[ \Lambda^0_{zz} = \sqrt{1 + 2u_{yy}^0}, \] \[ \Lambda^0_{yy} = \sqrt{1 + 2u_{xy}^0}, \] \[ \Lambda^0_{xx} = \gamma = 2u_{xy}^0/\Lambda^0_{zz}, \] and \( \Lambda^0_{zz} = \sqrt{1 + 2u_{yy}^0 - \gamma^2}. \] The elastic energy can now be written in terms of \( u' = (\Lambda^0)^{-1} \delta u (\Lambda^0)^{-1} \) as

\[ f_{uu}^{(2)} = \frac{1}{2} \bigg[ C_x \cos \theta u_{xy}^2 + \sin \theta u_{yy}^2 + \frac{1}{2} C_{zzzz} (u_{zz}^2)^2 \bigg] + C_{zxxz} (u_{zz}^2) + C_{zzxx} u_{xz}^2 + C_{zzyy} u_{yz}^2 + C_{zzzy} u_{yz}^2 + \frac{1}{2} C_{zzxx} (u_{zz}^2 + u_{xx}^2) + C_{zzxx} u_{xz}^2 + C_{zzyy} u_{yz}^2 + \frac{1}{2} C_{zzxx} (u_{zz}^2 + u_{xx}^2) + C_{zzyy} u_{yz}^2 + C_{zzzy} u_{yz}^2 + (9) \]

where the angle \( \theta \) and the elastic constants \( C \) and \( C_{zzzz} \) and so on depend on the original elastic constants in Eq. 10 and \( S \) so that one retrieves the uniaxial energy 11 for \( S \rightarrow 0 \). To lowest order in \( S \), \( \tan \theta = \omega S \). The elastic energy 12 has only 12 (including \( \theta \)) rather than the 13 elastic constants of conventional triclinic solids 13. There are only two rather than three independent elastic constants in the subspace spanned by \( u_{xx}^0 \) and \( u_{yy}^0 \). With the introduction of \( \vec{v} = (u_{xx}^0, u_{yy}^0) \), elastic energy in this subspace can be expressed as \( \frac{1}{2} \vec{v} \cdot \tilde{m} \cdot \vec{v} \) where \( \tilde{m} \) is a 2 \( \times \) 2 matrix. In general \( \tilde{m} \) has two independent eigenvalues \( m_1 \) and \( m_2 \) with respective associated orthogonal eigenvectors \( \vec{e}_1 \) and \( \vec{e}_2 \) with respect to which \( \tilde{m} \) is diagonal: \( \vec{v} \cdot \tilde{m} \cdot \vec{v} = m_1 (\vec{e}_1 \cdot \vec{v})^2 + m_2 (\vec{e}_2 \cdot \vec{v})^2 \). The first terms in Eq. 10 is of the form \( \frac{1}{2} m_1 (\vec{e}_1 \cdot \vec{v})^2 \) with \( m_1 = C_x \) and \( \vec{e}_1 = (\cos \theta, \sin \theta) \), and we conclude that \( m_2 = 0 \). Thus distortions along \( \vec{e}_2 = (-\sin \theta, \cos \theta) \), i.e., distortions for which \( \vec{v} \parallel \vec{e}_2 \) cost no energy (see Fig. 4). Stated differently, there are no restoring forces to stress \( -\sin \theta \sigma_{xy} + \cos \theta \sigma_{yy} = c_{xy} \sigma_{yy} \), i.e., to stress in the \( xx \) plane directed along \( \vec{e}_2 \) or stress in the plane perpendicular to \( \vec{e}_2 \) directed along \( y \).

Theory with director and smectic layers: The following theory generalizes the achiral limit of a continuum theory for SmC* elastomers by Terentjev and Warner 10 in a formalism that ensures invariance with respect to arbitrary rather than infinitesimal rotations of both the director and mass points.

In traditional uncrosslinked liquid crystals, there is no reference space, and all physical fields like the smectic layer-displacement field \( U \), the layer normal \( \mathbf{N} \), and the Frank director \( \mathbf{n} \) are defined at real or target space points \( \mathbf{R} \) and they transform as scalars, vectors, and tensors under rotations in the target space. In the Lagrangian theory of elasticity, fields are defined at reference space points \( x \), and they transform into themselves under the symmetry operations of that space. To develop a theory of liquid-crystalline elastomers, it is necessary to combine target-space liquid crystalline fields and reference space elastic variables to produce scalars that are invariant under arbitrary rotations in the target space and under symmetry operations of the reference space. This requires that we be able to represent vectors and tensors in either space [3]. The matrix polar decomposition theorem [11] applied to the Cauchy tensor \( \Lambda \) provides a route to this representation. Like any non-singular matrix, \( \Lambda \) can be decomposed into the product of an orthogonal matrix \( Q \) times a symmetric matrix: \( \Lambda = Q M^{1/2} \), where \( M^{1/2} \) is the symmetric square root of the symmetric matrix \( M = (\Lambda^T \Lambda) = (I + 2n) \), which depends only on the symmetric strain \( n \) and \( = \Lambda M^{1/2} \). The orthogonal matrix \( Q \) converts (or rotates) any reference space vector \( \tilde{a} \) to a target space vector \( a \) via \( a = Q \cdot \tilde{a} \) and a target-space vector to a reference space vector via \( \tilde{a} = Q^{-1} \cdot a \). Equipped with \( Q \), we can rotate the target space director \( \mathbf{n} \) to a reference space vector \( \mathbf{n} \equiv (c, n_z) \), where \( n_z = \sqrt{1 - c_z^2} \), and rotate the reference space anisotropy vector \( \mathbf{e} = (0, 0, 1) \) to a target-space vector \( \mathbf{e} = Q \cdot \mathbf{e} \). From these, we can form invariant couplings like \( (n \cdot \mathbf{e})^2 = (n \cdot e)^2 = 1 - c_z^2, c_a u_{ab} c_b, \) and \( \tilde{c}_a u_{ab} n_z \) as building blocks the elastic energy.

To treat smectic layers, we need to discuss in more detail the smectic displacement field \( U \) and the layer normal \( \mathbf{N} \). The smectic mass-density-wave amplitude for a system with layer spacing \( d \) has a phase \( \phi(\mathbf{R}) = q_0 |R_z - U(\mathbf{R})| \) where \( q_0 = 2\pi/\lambda \). Since there is a one-to-one mapping from the reference space points \( x \) to the target-space points \( \mathbf{R}(x) \), we can express \( \phi \) as a function of \( x \) as \( \phi(x) = q_0[z + u_z(x) - U(\mathbf{R}(x))] \). We are only considering systems crosslinked in the smectic phase. In these systems, the smectic mass-density wave cannot translate freely relative to the reference material, and there is a term in the free-energy density \( \frac{1}{2} A(u_z - U)^2 \) that locks the smectic field \( U \) to the displacement field \( u_z \). In what follows, we will take this lock-in as given and set \( U = u_z \). The layer normal in the target space is \( \mathbf{N}_i = \mathbf{v}_i/|\mathbf{v}_i| \), where \( \mathbf{v}_i \equiv \partial \phi/\partial R_i = \partial \phi/\partial n_{ji}^{-1} \). Thus, when \( U \) is locked to \( u_z \), \( N_i = [(M^{-1})_{zz}]^{-1/2} \), and to harmonic order in \( \tilde{c}_a \)
and \( u_{ij}, \tilde{\mathbf{N}} \cdot \tilde{\mathbf{n}} = \mathbf{N} \cdot \mathbf{n} = 1 - c_{ij}^0 - u_{za}\tilde{c}_a + \cdots. \)

We can now develop a full phenomenological free energy for the SmA–to–SmC transition in an elastomer. In the equilibrium SmA phase, the director is parallel to both the layer normal \( \mathbf{N} \) and the anisotropy axis \( \mathbf{e} \), which are parallel to each other, and there are energy costs proportional to \((\tilde{\mathbf{N}} \cdot \tilde{\mathbf{n}})^2\) and \((\tilde{\mathbf{e}} \cdot \tilde{\mathbf{n}})^2\) associated with deviations from this equilibrium. These combine to yield a term in the free-energy density proportional to \( c_{ij}^0 \) and higher order terms involving the strain and strain-director coupling. With the addition of higher order terms in \((\tilde{\mathbf{N}} \cdot \tilde{\mathbf{n}})^2\) and \((\tilde{\mathbf{e}} \cdot \tilde{\mathbf{n}})^2\) to stabilize the SmC phase, the free energy up to inconsequential higher order terms becomes

\[
f = f_{\text{uni}} + f_{\text{tri}} + f_{\text{coup}},
\]

where \( f_{\text{uni}} \) is the uniaxial energy \[11\] and

\[
f_{\text{tri}} = \frac{1}{2} r c_{ij}^2 + \frac{1}{4} g (c_{ij}^0)^2,
\]

\[
f_{\text{coup}} = \lambda_1 c_{ij}^0 u_{zz} + \lambda_2 c_{ij} c_{ab} u_{ab} + \lambda_3 \tilde{c}_a \tilde{u}_{ab} \tilde{c}_b + \lambda_4 \tilde{c}_a u_{az} \tilde{n}_z.
\]

When \( \lambda_1 = \lambda_2 = \lambda_3 = 0 \), this model is equivalent to that studied in Ref. \[10\] when polarization is ignored.

We can now analyze the transition to the SmC phase in exactly the same way as we did in the strain only model. We complete the squares involving the strains \( c_{ij} \) and \( \tilde{c}_a \) only on \( \tilde{\mathbf{c}} \) and \( \tilde{\mathbf{n}} \). This expression shows clearly that the softness identical to that of Eq. \(10\) in terms of \( \tilde{\mathbf{c}} \) and \( \tilde{\mathbf{n}} \) is massive, it can be integrated out. Converting the energy \(10\) in terms of \( \tilde{\mathbf{c}} \) and \( \tilde{\mathbf{n}} \), the result is identical to

\[
A_0 N f
\]

both the layer normal \( \mathbf{N} \) and the smectic layers remain parallel to the \( x \)-axis as one would intuitively expect in the chosen geometry.

**Concluding remarks:** We have presented models for transitions from uniaxial SmA elastomers to biaxial and SmC elastomers, and we have calculated the nature of the soft elasticity, required by symmetry, of monodomain samples of these phases. We hope that our work encourages experiments to probe this soft elasticity.

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