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Macroscopic response of nematic elastomers via relaxation of a class of SO(3)-invariant energies

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MACROSCOPIC RESPONSE OF NEMATIC ELASTOMERS VIA RELAXATION OF A CLASS OF SO(3)-INVARIANT ENERGIES

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Abstract. We obtain an explicit formula for the relaxation of the free energy density for nematic elastomers proposed by Bladon, Terentjev&Warner (Phys. Rev. E **47** (1993), 3838-3840). The proof is based on a characterization of the level sets of the relaxed energy. In particular, the construction uses only laminates within laminates and it identifies those deformations that correspond to simple laminates.

1. Introduction

Solid to solid phase transformations are often related to surprising mechanical properties of materials and their promising technological applications. Inspired by the success of the mathematical theory for crystalline microstructure in shape memory alloys [BJ1, BJ2, CK], we present in this paper the analysis of a model of nematic elastomers, polymeric materials that undergo an isotropic to nematic phase transformation. The local order in the material that is established in the nematic phase gives rise to a unique combination of elastic and optic features in the system with applications such as a novel design of bifocal contact lenses and light-guiding substrates for integrated optics.

The macroscopic response of materials displaying fine internal structures is governed by the so-called relaxed or effective energy $W^{\rm qc}$ defined by

$$W^{\rm qc}(F) = \inf_{\substack{\varphi\in W^{1,\infty}(\Omega;\mathbb{R}^3)\\ \varphi(x)=Fx \text{ on } \partial\Omega}} \frac{1}{|\Omega|}\int_\Omega W(D\varphi(x))\mathrm{d}x,$$

where W is the free energy of the system, because the material is free to choose locally an (asymptotically) optimal microstructure to realize a given deformation gradient F. For simplicity we assume in the following that $W \geq 0$ and that $K = \{X \in \mathbb{M}^{3\times 3} : W(X) = 0\}$ is not empty. The set of all affine deformations with approximately zero energy is then characterized by $K^{\rm qc} = \{X : W^{\rm qc}(X) = 0\}$. In this paper we derive an explicit formula both for $K^{\rm qc}$ and for the effective energy of nematic elastomers. Our results

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follow from the analysis of the family of functions

$$W(F) = \begin{cases} \dfrac{\lambda_1^p(F)}{\gamma_1^p} + \dfrac{\lambda_2^p(F)}{\gamma_2^p} + \dfrac{\lambda_3^p(F)}{\gamma_3^p} - 3 & \text{if } \det F = 1, \\ +\infty & \text{else,} \end{cases} \tag{1}$$

which includes the free energy for nematic elastomers [BTW] as a special case for $p = 2$. Here $p \in [2, \infty)$ and $\lambda_1(F) \leq \lambda_2(F) \leq \lambda_3(F)$ are the singular values of F, i.e. the eigenvalues $(F^T F)^{1/2}$. Moreover, $\gamma_1, \gamma_2, \gamma_3 \in \mathbb{R}$ are constants that satisfy $0 < \gamma_1 \leq \gamma_2 \leq \gamma_3$ and $\gamma_1\gamma_2\gamma_3 = 1$. The requirement that the energy be infinite if the determinant of the deformation gradient is different from one models (in mathematical abstraction) the incompressibility of the elastomer. The main result in this paper is the following theorem.

Theorem 1.1. *There exists a function $\psi : \mathbb{R}_+^2 \to \mathbb{R}$ which is convex and nondecreasing in its arguments such that*

$$W^{\mathrm{qc}}(F) = \begin{cases} \psi\big(\lambda_{\max}(F), \lambda_{\max}(\operatorname{cof} F)\big) & \text{if } \det F = 1, \\ +\infty & \text{else.} \end{cases}$$

We provide an explicit formula for ψ in (14).

The fundamental difference in the analysis of phase transformations in crystalline materials and in the polymers we consider lies in the fact that, due to material frame indifference and isotropy of the high temperature phase, the energy density W and the set K depend for polymers only on the singular values of the deformation gradient. In fact,

$$\begin{aligned} K &= \bigcup_{\{e_1,e_2,e_3\}\in\mathcal{E}} \mathrm{SO}(3)\big(\gamma_1 e_1 \otimes e_1 + \gamma_2 e_2 \otimes e_2 + \gamma_3 e_3 \otimes e_3\big), \\ &= \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_i(F) = \gamma_i,\, i = 1, 2, 3\big\}, \end{aligned}$$

where $\mathcal{E}$ denotes the set of all orthonormal bases of $\mathbb{R}^3$. Thus the set K is much larger than the corresponding sets in the analysis of crystalline materials which are typically finite unions of sets of the form $\mathrm{SO}(3)U$ with $U \in \mathbb{M}^{3\times 3}$ symmetric and positive definite.

The paper is organized as follows. We introduce important notation in Section 2 and characterize all affine deformations with approximately zero energy in Section 3. The idea behind the proof of Theorem 1.1 is presented for a two-dimensional model in Section 4 and the details are carried out in the three-dimensional case in Section 5. We finally present in Section 6 the relaxation result for the free energy density describing nematic elastomers.

2. Preliminaries

We begin with some notation which we use throughout the paper. The n-dimensional Euclidean space is denoted by $\mathbb{R}^n$ with scalar product $\langle u, v\rangle$ and norm $|u|^2 = \langle u, u\rangle$. We write $\mathbb{R}_+$ for the set of all nonnegative real numbers, $\overline{\mathbb{R}} = \mathbb{R} \cup \{+\infty\}$, and $\mathbb{M}^{m\times n}$ for the space of all real $m \times n$ matrices. If $m = n$, then $\operatorname{cof} F$ is defined to be the matrix of all $(n-1) \times (n-1)$ minors of F

that satisfies $F^T \operatorname{cof} F = (\det F)I$, where F^T is the transposed matrix of F and I the identity matrix in $\mathbb{M}^{n\times n}$.

The following notions of convexity are fundamental for our analysis. A function $f : \mathbb{M}^{m\times n} \to \overline{\mathbb{R}}$ is said to be polyconvex if there exists a convex function g which depends on the vector $M(F)$ of all minors of F such that $f(F) = g(M(F))$. In particular, if $m = n = 2$, then $f(F) = g(F, \det F)$, and if $m = n = 3$, then $f(F) = h(F, \operatorname{cof} F, \det F)$, where the functions $g : \mathbb{R}^5 \to \overline{\mathbb{R}}$ and $h : \mathbb{R}^{19} \to \overline{\mathbb{R}}$ are convex. The function f is called quasiconvex, if there exists an open domain Ω with $|\partial\Omega| = 0$ such that the inequality

$$\int_\Omega f(F)\mathrm{d}x \leq \int_\Omega f(F + D\varphi)\mathrm{d}x$$

holds for all $F \in \mathbb{M}^{m\times n}$ and all $\varphi \in W_0^{1,\infty}(\Omega; \mathbb{R}^m)$, whenever the right hand side exists. It follows that the foregoing inequality holds for all domains $\widetilde{\Omega}$ with $|\widetilde{\Omega}| = 0$. Finally, f is rank-one convex if $t \mapsto f(F + tR)$ is a convex function for all F, $R \in \mathbb{M}^{m\times n}$ with $\operatorname{rank}(R) = 1$. For extended valued functions, polyconvexity implies both quasiconvexity and rank-one convexity, but quasiconvexity does not imply rank-one convexity. If f is finite valued, $f : \mathbb{M}^{m\times n} \to \mathbb{R}$, then we have

$$f \text{ convex } \Rightarrow f \text{ polyconvex } \Rightarrow f \text{ quasiconvex } \Rightarrow f \text{ rank-one convex.}$$

If f is not polyconvex, then the polyconvex envelope f^{pc} of f is the largest polyconvex function less than or equal to f. The quasiconvex and the rank-one convex envelope are obtained analogously and denoted by f^{qc} and f^{rc}, respectively. Based on these notions of convexity, we define semiconvex hulls of compact sets $K \subset \mathbb{M}^{m\times n}$. The set

$$K^{\mathrm{pc}} = \big\{F : f(F) \leq \sup_{X\in K} f(X) \text{ for all } f : \mathbb{M}^{m\times n} \to \mathbb{R} \text{ polyconvex } \big\}$$

is called the polyconvex hull of K. The quasiconvex hull K^{qc} and the rank-one convex hull K^{rc} are defined analogously by replacing polyconvexity in the definition of K^{pc} by quasiconvexity and rank-one convexity, respectively. Finally, we define the lamination convex hull of K by allowing all extended valued, rank-one convex functions in the definition of K^{rc},

$$K^{\mathrm{lc}} = \big\{F : f(F) \leq \sup_{X\in K} f(X) \text{ for all } f : \mathbb{M}^{m\times n} \to \overline{\mathbb{R}} \text{ rank-one convex } \big\}.$$

Equivalently, the lamination convex hull can be defined by successively adding rank-one segments, i.e.,

$$K^{\mathrm{lc}} = \bigcup_{i=0}^{\infty} K^{(\mathrm{i})},$$

where $K^{(0)} = K$ and

$$K^{(\mathrm{i}+1)} = K^{(\mathrm{i})} \cup \big\{ F = \lambda F_1 + (1-\lambda) F_2 : F_1,\, F_2 \in K^{(\mathrm{i})}, \\ \mathrm{rank}(F_1 - F_2) = 1,\, \lambda \in (0,1) \big\}.$$

The relations between the different notions of convexity imply the inclusions

$$K^{\mathrm{lc}} \subseteq K^{\mathrm{rc}} \subseteq K^{\mathrm{qc}} \subseteq K^{\mathrm{pc}}. \tag{2}$$

We frequently use the two subsets $K^{(1)}$ and $K^{(2)}$, which we informally describe as barycenters of simple laminates and laminates within laminates supported on K, respectively. See [Pe] for the definition of laminates, [Dc] and [M] for a discussion of all the different notions of convexity and their relations, and [B1, B2] for convexity and regularity properties of frame indifferent and isotropic functions.

If $W : \mathbb{M}^{m\times n} \to \overline{\mathbb{R}}$ is a given function and $K \subset \mathbb{M}^{3\times 3}$, then we say that $\widetilde{W}(F)$ is obtained by averaging W with respect to a simple laminate supported on K if $F \in K^{(1)}$ and if there exist $\lambda \in (0,1)$ and $F_1,\, F_2 \in K$ with $\mathrm{rank}(F_1 - F_2) = 1$ such that

$$F = \lambda F_1 + (1-\lambda) F_2 \quad \text{and} \quad \widetilde{W}(F) = \lambda W(F_1) + (1-\lambda) W(F_2). \tag{3}$$

Similarly, we say that $\widetilde{W}(F)$ is obtained by averaging W with respect to a laminate within a laminate supported on K if $F \in K^{(2)}$ and if the following assertion is true: There exist $F_1, \ldots, F_4 \in K$ and $\lambda_1, \ldots, \lambda_4 \in [\,0,1\,]$ such that $\lambda_1 + \lambda_2 \neq 0$, $\lambda_3 + \lambda_4 \neq 0$, $\lambda_1 + \ldots + \lambda_4 = 1$, $\mathrm{rank}(F_1 - F_2) = 1$ and $\mathrm{rank}(F_3 - F_4) \le 1$. Moreover, if we define

$$G_1 = \frac{\lambda_1}{\lambda_1+\lambda_2} F_1 + \frac{\lambda_2}{\lambda_1+\lambda_2} F_2, \quad G_2 = \frac{\lambda_3}{\lambda_3+\lambda_4} F_3 + \frac{\lambda_4}{\lambda_3+\lambda_4} F_4,$$

then $\mathrm{rank}(G_1 - G_2) = 1$, and

$$F = \sum_{i=1}^{4} \lambda_i F_i \quad \text{and} \quad \widetilde{W}(F) = \sum_{i=1}^{4} \lambda_i W(F_i). \tag{4}$$

Our relaxation result in Section 5 is based on the construction of optimal laminates for which the resulting value for $\widetilde{W}$ is minimal.

3. Affine deformations with approximately zero energy

We begin with the characterization of the set K^{qc}. In the next proposition we use the convention that $\lambda_0 = \lambda_3$, $\lambda_4 = \lambda_1$ and $\xi_0 = \xi_3$, $\xi_4 = \xi_1$ and we write $\Lambda(F) = \big\{\lambda_1(F), \lambda_2(F), \lambda_3(F)\big\}$ for the set of the singular values of F.

Theorem 3.1. *Assume that* $0 < \xi_1 \le \xi_2 \le \xi_3$ *with* $\xi_1\xi_2\xi_3 = 1$ *and that*

$$K = \big\{ F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_i(F) = \xi_i,\, i = 1,2,3 \big\}.$$

Then the sets M_i defined by

$$M_i = \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\ \xi_i \in \Lambda(F),\\ \Lambda(F) \setminus \{\xi_i\} \subset [\,\min\{\xi_{i-1}, \xi_{i+1}\}, \max\{\xi_{i-1}, \xi_{i+1}\}\,]\big\}$$

are contained in $K^{(1)}$ for $i = 1, 2, 3$. Moreover, we have

$$K^{(1)} = M_1 \quad \textit{if} \quad \xi_1 = \xi_2 \qquad \textit{and} \qquad K^{(1)} = M_3 \quad \textit{if} \quad \xi_2 = \xi_3.$$

Remark 3.2. *In general the inclusion $M_1 \cup M_2 \cup M_3 \subset K^{(1)}$ is strict.*

Proof. To prove the first part of the theorem, we assume that $i = 1$, and we write $M = M_1$. The argument is analogous for $i = 2$ and $i = 3$. The assertion of the proposition is now equivalent to $M \subseteq K^{(1)}$ where

$$M = \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\ \lambda_1(F) = \xi_1, \lambda_2(F), \lambda_3(F) \in [\,\xi_2, \xi_3\,]\big\}.$$

Let $F \in M$. Since $QFR \in M$, for all Q, $R \in \mathrm{SO}(3)$ and $F \in M$, we may suppose that F is diagonal, $F = \mathrm{diag}(\xi_1, \mu_2, \mu_3)$, with $\mu_2, \mu_3 \in [\,\xi_2, \xi_3\,]$. Note that $\mu_2\mu_3 = \xi_2\xi_3$ since $\det F = 1$. There is nothing to prove if $\mu_2 = \xi_2$ or $\mu_3 = \xi_3$, since the condition $\det F = 1$ implies in this case that $F \in K$. Following [WT, DSD] we show that there exists for $\mu_2, \mu_3 \in (\xi_2, \xi_3)$ a $\delta > 0$ (which depends on μ_2 and μ_3) such that

$$F^{\pm} = \mathrm{diag}(\xi_1, \widehat{F}^{\pm}) \in K \quad \text{where} \quad \widehat{F}^{\pm} = \begin{pmatrix} \mu_2 & \pm\delta \\ 0 & \mu_3 \end{pmatrix}.$$

Then

$$F^+ - F^- = 2\delta e_2 \otimes e_3 \quad \text{and} \quad F = \frac{1}{2}F^+ + \frac{1}{2}F^- \in K^{(1)}.$$

We define

$$\widehat{C}^{\pm} = (\widehat{F}^{\pm})^T \widehat{F}^{\pm} = \begin{pmatrix} \mu_2^2 & \pm\delta\mu_2 \\ \pm\delta\mu_2 & \mu_3^2 + \delta^2 \end{pmatrix}.$$

The eigenvalues $t^{\pm}$ of $\widehat{C}^{\pm}$ are the solutions of

$$\det(\widehat{C}^{\pm} - tI) = t^2 - (\mu_2^2 + \mu_3^3 + \delta^2)t + \mu_2^2\mu_3^2 = 0,$$

and the requirement that t^+ defined by

$$t^{\pm} = \frac{\mu_2^2 + \mu_3^2 + \delta^2}{2} \pm \sqrt{\Big(\frac{\mu_2^2 + \mu_3^2 + \delta^2}{2}\Big)^2 - \mu_2^2\mu_3^2}$$

be equal to ξ_3^2 leads to

$$\delta = \frac{1}{\xi_3}\sqrt{\xi_3^2 - \mu_2^2}\,\sqrt{\xi_3^2 - \mu_3^2} > 0. \tag{5}$$

Since $t^+t^- = \mu_2^2\mu_3^2 = \xi_2^2\xi_3^2$, this choice of δ also yields $t^- = \xi_2^2$ and we conclude that for the value of δ given in (5) the matrices $F^{\pm}$ are contained in K and this proves the first assertion of the theorem.

It remains to prove the characterization of $K^{(1)}$ if two of the parameters in the description of K coincide. Without loss of generality we assume that $\xi_1 = \xi_2 < \xi_3$, and we have to prove that $K^{(1)} \subseteq M_1$. Suppose thus that

$F \in K^{(1)} \setminus K$ and choose $F_1, F_2 \in K$ such that there exists a $\lambda \in (0,1)$ and $a, n \in \mathbb{R}^3$, $a, n \neq 0$ with

$$F = \lambda F_1 + (1-\lambda)F_2, \qquad F_1 - F_2 = a \otimes n.$$

Since $QFR \in K$, for all $Q, R \in \mathrm{SO}(3)$ and $F \in K$, we may choose Q and $R \in \mathrm{SO}(3)$ such that $\widetilde{F}_1 = QF_1R \in K$ is diagonal, $\widetilde{F}_1 = \mathrm{diag}(\xi_3, \xi_1, \xi_1)$. We define analogously $\widetilde{F}_2 = QF_2R \in K$, $\widetilde{F} = QFR \in K^{(1)}$, $\widetilde{a} = Qa$ and $\widetilde{n} = R^T n$. Then

$$\widetilde{F} = \lambda \widetilde{F}_1 + (1-\lambda)\widetilde{F}_2, \qquad \widetilde{F}_1 - \widetilde{F}_2 = \widetilde{a} \otimes \widetilde{n}.$$

The intersection of the plane spanned by the unit vectors $e_2 = (0,1,0)$ and $e_3 = (0,0,1)$ intersects the plane $\{w \in \mathbb{R}^3 : \langle w, \widetilde{n}\rangle = 0\}$ with normal $\widetilde{n}$ at least in a one-dimensional line through the origin parallel to some unit vector $v \in \mathbb{S}^2$. This implies

$$0 = \langle \widetilde{n}, v\rangle \widetilde{a} = (\widetilde{a} \otimes \widetilde{n})v = (\widetilde{F}_1 - \widetilde{F}_2)v = \xi_1 v - \widetilde{F}_2 v,$$

and therefore v is an eigenvector of $\widetilde{F}_1$ and $\widetilde{F}_2$ with corresponding eigenvalue ξ_1. Consequently, $\widetilde{F}v = \xi_1 v$ and

$$\lambda_1(\widetilde{F}) = \lambda_{\min}(\widetilde{F}) = \min_{e \in \mathbb{S}^2} |\widetilde{F}e| \leq |\widetilde{F}v| = \xi_1.$$

To prove that ξ_1 is the smallest singular value of $\widetilde{F}$, let $(\cdot)^+$ denote the convex, nondecreasing function $t \mapsto (t)^+ = \max\{t, 0\}$. Then the functions

$$\begin{aligned} g_1(F) &= \big(\sup_{e \in \mathbb{S}^2} |Fe| - \gamma_3\big)^+, \\ g_2(F) &= \big(\sup_{e \in \mathbb{S}^2} |\operatorname{cof} Fe| - \frac{1}{\gamma_1}\big)^+ \end{aligned}$$

are polyconvex, and since $F \in K^{(1)} \subseteq K^{\mathrm{pc}}$ we deduce $\lambda_i(\widetilde{F}) \in [\,\xi_1, \xi_3\,]$. Therefore

$$\lambda_1(\widetilde{F}) = \xi_1 \quad \text{ and } \quad \xi_1 = \min\{\xi_2, \xi_3\} \leq \lambda_2(\widetilde{F}) \leq \lambda_3(\widetilde{F}) \leq \max\{\xi_2, \xi_3\}.$$

We obtain $\widetilde{F} \in M_1$. The matrices $\widetilde{F}$ and F have the same singular values, and hence $F \in M_1$. This concludes the proof of the theorem. □

The foregoing theorem implies immediately a formula for the set of all approximately zero-energy deformations for the density (1) (see [DcT] for results for sets defined by singular values in arbitrary dimensions).

Corollary 3.3. *Assume that* $0 < \gamma_1 \leq \gamma_2 \leq \gamma_3$ *with* $\gamma_1\gamma_2\gamma_3 = 1$ *and that*

$$K = \{F \in \mathbb{M}^{3\times 3} : \det F = 1,\ \lambda_i(F) = \gamma_i,\ i = 1,2,3\}.$$

Then

$$K^{(2)} = K^{\mathrm{lc}} = K^{\mathrm{rc}} = K^{\mathrm{qc}} = K^{\mathrm{pc}}, \tag{6}$$

and these sets are given by

$$\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\ \lambda_i(F) \in [\,\gamma_1, \gamma_3\,],\ i = 1,2,3\}. \tag{7}$$

Remark 3.4. *A formula for the convex hull of K can be found in* [Do]*. Not surprisingly,* $\operatorname{conv} K \cap \{F \in \mathbb{M}^{3\times 3} : \det F = 1\} \neq K^{\mathrm{pc}}$*. This is due to the fact that the lower bound on the singular values in the description of K^{pc} follows from the polyconvex condition $|\operatorname{cof} Fe| \leq \gamma_1^{-1}$ for all $e \in \mathbb{S}^2$.*

Proof. Let $\mathcal{A}$ be the set given in (7). Since $\det F = 1$, we have

$$\lambda_{\min}(F) = \lambda_1(F) = \frac{1}{\lambda_{\max}(\operatorname{cof} F)} = \frac{1}{\lambda_3(\operatorname{cof} F)}, \tag{8}$$

and therefore we may rewrite the definition of $\mathcal{A}$ as

$$\mathcal{A} = \big\{F \in \mathbb{M}^{3\times 3} : g_1(F) \leq 0,\, g_2(F) \leq 0,\, g_3(F) \leq 0\big\},$$

where g_1 and g_2 were defined in the proof of Theorem 3.1 and

$$g_3(F) = (\det F - 1)^2.$$

The functions g_i are polyconvex, hence $K^{\mathrm{pc}} \subseteq \mathcal{A}$. It only remains to prove that $\mathcal{A} \subseteq K^{(2)}$. We then obtain by the chain of inclusions (2) that $K^{\mathrm{pc}} \subseteq K^{(2)} \subseteq K^{\mathrm{pc}}$ and the equation (6) is thus an immediate consequence. Since $QFR \in \mathcal{A}$, for all Q, $R \in \mathrm{SO}(3)$ and $F \in \mathcal{A}$ we may assume that $F \in \mathcal{A}$ is a diagonal matrix, $F = \operatorname{diag}(\mu_1, \mu_2, \mu_3)$ with $\gamma_1 \leq \mu_1 \leq \mu_2 \leq \mu_3 \leq \gamma_3$. If $\gamma_2 \leq \mu_3 \leq \gamma_3$, then

$$\frac{\gamma_2}{\mu_3} \leq 1 \leq \frac{\gamma_3}{\mu_3} \quad \text{and} \quad \gamma_2 \leq \frac{\gamma_2\gamma_3}{\mu_3} \leq \gamma_3.$$

By Theorem 3.1,

$$M_1 = \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_1(F) = \gamma_1,\\ \lambda_2(F) = \min\{\frac{\gamma_2\gamma_3}{\mu_3}, \mu_3\},\, \lambda_3(F) = \max\{\frac{\gamma_2\gamma_3}{\mu_3}, \mu_3\}\big\}$$

is contained in $K^{(1)}$. Now $\gamma_1 \leq \mu_1 \leq \mu_2 \leq \frac{\gamma_2\gamma_3}{\mu_3}$ since

$$\mu_2 \leq \frac{\gamma_2\gamma_3}{\mu_3} \quad \Leftrightarrow \quad \mu_2\mu_3 \leq \gamma_2\gamma_3 \quad \Leftrightarrow \quad \gamma_1 \leq \mu_1.$$

If $\gamma_2\gamma_3 \leq \mu_3^2$, then $\lambda_3(F) = \mu_3$ for $F \in M_1$ while for $\gamma_2\gamma_3 > \mu_3^2$ one has $\lambda_2(F) = \mu_3$ for $F \in M_1$. Another application of Theorem 3.1 with $i = 3$ or $i = 2$, respectively, implies that

$$\big\{F \in \mathbb{M}^{3\times 3} : \lambda_i(F) = \mu_i,\big\} \subseteq M_1^{(1)} \subseteq K^{(2)}.$$

Suppose now that $\gamma_1 \leq \mu_1 \leq \mu_2 \leq \mu_3 \leq \gamma_2$. Then $\gamma_1 \leq \frac{\gamma_1\gamma_2}{\mu_1} \leq \gamma_2$, and we conclude from Theorem 3.1 that

$$M_3 = \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_3(F) = \gamma_3,\\ \lambda_1(F) = \min\{\mu_1, \frac{\gamma_1\gamma_2}{\mu_1}\},\, \lambda_2(F) = \max\{\mu_1, \frac{\gamma_1\gamma_2}{\mu_1}\}\big\}$$

is contained in $K^{(1)}$. In this situation, $\frac{\gamma_1\gamma_2}{\mu_1} \leq \mu_2 \leq \mu_3 \leq \gamma_3$, since

$$\frac{\gamma_1\gamma_2}{\mu_1} \leq \mu_2 \quad \Leftrightarrow \quad \gamma_1\gamma_2 \leq \mu_1\mu_2 \quad \Leftrightarrow \quad \mu_3 \leq \gamma_3,$$

and we can apply Theorem 3.1 once more (with $i = 1$ or $i = 2$) to deduce

$$\big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_i(F) = \mu_i,\, i = 1,2,3\big\} \subseteq M_3^{(1)} \subseteq K^{(2)}.$$

This concludes the proof of the corollary. □

Corollary 3.5. *Assume that* $0 < \xi_1 \le \xi_2 \le \xi_3$ *with* $\xi_1\xi_2\xi_3 = 1$, *and let*

$$\widetilde{K} = \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_1(F) = \xi_1,\, \lambda_2(F) = \xi_2,\, \lambda_3(F) = \xi_3\big\}.$$

Assume that $g : \mathbb{M}^{3\times 3} \to \overline{\mathbb{R}}$ *depends only on the singular values of its argument and that* g *is rank-one convex. Then*

$$g(F) \le g\big(\operatorname{diag}(\xi_1,\xi_2,\xi_3)\big) \text{ for all } F \in \widetilde{K}^{\mathrm{pc}}.$$

Proof. Theorem 3.1 implies that $F \in \widetilde{K}^{\mathrm{qc}}$ if and only if $F \in \widetilde{K}^{(2)}$. We now have from the definition of $\widetilde{K}^{\mathrm{lc}}$ that

$$g(F) \le \sup_{X\in\widetilde{K}} g(X) = g\big(\operatorname{diag}(\xi_1,\xi_2,\xi_3)\big),$$

and this proves the assertion of the corollary. □

4. The construction for a two-dimensional model problem

Before we present the relaxation result in three dimensions, we describe the ideas behind the construction for the corresponding two-dimensional energy density given by

$$f(F) = \begin{cases} \dfrac{\lambda_1^p(F)}{\gamma_1^p} + \dfrac{\lambda_2^p(F)}{\gamma_2^p} - 2 & \text{if } \det F = 1, \\ +\infty & \text{else,} \end{cases} \tag{9}$$

where $0 < \gamma_1 < \gamma_2$, $\gamma_1\gamma_2 = 1$, and $p \in [2,\infty)$. Since $\lambda_1 = \frac{1}{\lambda_2}$ and $\gamma_1 = \frac{1}{\gamma_2}$, we may rewrite the energy as

$$f(F) = \begin{cases} \dfrac{\gamma_2^p}{\lambda_2^p(F)} + \dfrac{\lambda_2^p(F)}{\gamma_2^p} - 2 & \text{if } \det F = 1, \\ +\infty & \text{else.} \end{cases}$$

The key idea in the three-dimensional case is analogous: we first eliminate λ_2 from the formulae by $\lambda_2 = \frac{1}{\lambda_1\lambda_3}$, and then use the identity (8) between $\lambda_{\min}(F)$ and $\lambda_{\max}(\operatorname{cof} F)$.

The density (9) is only finite on the curve $\lambda_1\lambda_2 = 1$ in the (λ_1, λ_2) plane and converges to infinity as λ_2 tends to infinity. It is easy to see that the function

$$g(\lambda) = \big(\frac{\gamma_2}{\lambda}\big)^p + \big(\frac{\lambda}{\gamma_2}\big)^p - 2$$

has a minimum at

$$\lambda^* = \gamma_2 \quad \text{with} \quad \big(\frac{\lambda}{\gamma_2}\big)^p\Big|_{\lambda=\lambda^*} = \big(\frac{\gamma_2}{\lambda}\big)^p\Big|_{\lambda=\lambda^*} = 1 \tag{10}$$

and $g(\lambda^*) = 0$. Therefore

$$K = \{F \in \mathbb{M}^{2\times 2} : f(F) = 0\} = \{F \in \mathbb{M}^{2\times 2} : \det F = 1,\ \lambda_2 = \gamma_2\}.$$

The construction of the relaxed energy now proceeds in three steps:

1) Finding an upper bound $\widetilde{f}$ for $f^{\rm rc}$.
2) Establishing that $\widetilde{f}$ is polyconvex; this implies $\widetilde{f} = f^{\rm rc} = f^{\rm pc}$.
3) Showing that $f^{\rm qc} \le \widetilde{f} = f^{\rm pc}$; since polyconvexity implies quasiconvexity for extended valued functions, we conclude that $f^{\rm rc} = f^{\rm qc} = f^{\rm pc}$.

We briefly discuss the first two steps in the proof, since the calculations are more transparent in the two-dimensional case and illustrate the strategy we use in the demonstration of the three-dimensional result. The third step is identical in the two-dimensional and the three-dimensional situation.

Step 1: Finding an upper bound $\widetilde{f}$.

The arguments in the proof of Theorem 3.1 show that

$$K^{(1)} = K^{\rm rc} = K^{\rm qc} = K^{\rm pc} = \{F \in \mathbb{M}^{2\times 2} : \det F = 1,\ \lambda_2(F) \le \lambda^*\}.$$

Moreover, for all $F \in K^{\rm qc} \setminus K$ there exist pairs (s, F_1) and $(1-s, F_2)$ with $F_1,\ F_2 \in K$ and $\operatorname{rank}(F_1 - F_2) = 1$ such that

$$F = sF_1 + (1-s)F_2, \quad \text{and} \quad f(F_1) = f(F_2) = 0.$$

This implies

$$f^{\rm rc}(F) \le sf(F_1) + (1-s)f(F_2) = 0,$$

and suggests to define

$$\widetilde{f}(F) = \begin{cases} 0 & \text{if } \lambda_2 \le \lambda^*,\ \det F = 1, \\ f(F) & \text{if } \lambda_2 \ge \lambda^*,\ \det F = 1, \\ +\infty & \text{if } \det F \ne 1. \end{cases} \tag{11}$$

In the three dimensional situation we use an analogous construction also for values of f different from zero, and obtain an upper bound $\widetilde{f}$ by characterizing (parts of) its level sets.

Step 2: Establishing that $\widetilde{f}$ is polyconvex.

Motivated by (11) we define

$$\psi(s) = \begin{cases} 0 & \text{if } s \le \lambda^*, \\ \left(\frac{\gamma_2}{s}\right)^p + \left(\frac{s}{\gamma_2}\right)^p - 2 & \text{if } s \ge \lambda^*. \end{cases}$$

It is easy to see that ψ is convex and nondecreasing since

$$\psi'(s) = -\frac{p\gamma_2^p}{s^{p+1}} + \frac{ps^{p-1}}{\gamma_2^p} \ge 0 \quad \Leftrightarrow \quad s \ge \gamma_2 = \left(\frac{\gamma_2}{\gamma_1}\right)^{1/2} = \lambda^*.$$

This implies that the function $\Psi_1(F) : \mathbb{M}^{2\times 2} \to \mathbb{R}$ given by

$$\Psi_1(F) = \psi(\sup_{e\in\mathbb{S}^1} |Fe|) = \psi(\lambda_{\max}(F))$$

is convex on $\mathbb{M}^{2\times 2}$ and therefore polyconvex (see the proof of Proposition 5.3 below). We now define the function $\Psi_2 : \mathbb{M}^{2\times 2} \to \overline{\mathbb{R}}$ by

$$\Psi_2(F) = I_1(\det F) \quad \text{where} \quad I_1(t) = \begin{cases} 0 & \text{if } t = 1, \\ \infty & \text{else.} \end{cases}$$

Then Ψ_2 is polyconvex and

$$\Psi_1(F) + \Psi_2(F) = \begin{cases} 0 & \text{if } \lambda_2 \leq \lambda^*, \ \det F = 1, \\ \big(\frac{\gamma_2}{\lambda_2}\big)^p + \big(\frac{\lambda_2}{\gamma_2}\big)^p - 2 & \text{if } \lambda_2 \geq \lambda^*, \ \det F = 1, \\ +\infty & \text{else.} \end{cases}$$

It follows from $\lambda_1\lambda_2 = 1$ for $\det F = 1$ that $\widetilde{f} = \Psi_1 + \Psi_2$ is a polyconvex function. The arguments in the proof of Theorem 5.6 in Section 5 apply also in the two-dimensional setting and we rediscover that $\widetilde{f}$ is in fact the relaxation for the two-dimensional model, see [DSD].

5. Construction of the relaxed energy

After these preparations, we describe in this section the analysis of the three-dimensional case. The construction becomes particularly transparent if one considers W on the set $\{\det F = 1\}$ as a function of two variables in the $\big(\lambda_{\max}(F), \lambda_{\max}(\operatorname{cof} F)\big)$-plane, see Figure 1. In order to simplify notation, we write $s = \lambda_{\max}(F)$ and $t = \lambda_{\max}(\operatorname{cof} F)$. With this abbreviation, the region in the (s,t)-plane, on which W is finite, is bounded by the two curves $t = \sqrt{s}$ and $t = s^2$. This is due to the fact that $\det F = 1$ implies

$$\lambda_{\max}(\operatorname{cof} F) = \frac{1}{\lambda_{\min}(F)}, \ \lambda_{\max}(F) \leq \frac{1}{\lambda^2_{\min}(F)} \ \text{ and } \ \lambda_{\min}(F) \geq \frac{1}{\lambda^2_{\max}(F)}\,.$$

In the three-dimensional situation, the single value λ^* in Section 4 is replaced by two curves in the phase plane along which two terms in the energy are equal, see (10). They are determined from

$$\big(\frac{\lambda_1}{\gamma_1}\big)^p = \big(\frac{\lambda_2}{\gamma_2}\big)^p = \big(\frac{1}{\gamma_2\lambda_1\lambda_3}\big)^p \quad \text{and} \quad \big(\frac{\lambda_2}{\gamma_2}\big)^p = \big(\frac{1}{\gamma_2\lambda_1\lambda_3}\big)^p = \big(\frac{\lambda_3}{\gamma_3}\big)^p,$$

and these two conditions are equivalent to

$$\lambda_{\max}(\operatorname{cof} F) = \big(\frac{\gamma_2}{\gamma_1}\big)^{1/2}\sqrt{\lambda_{\max}(F)} \quad \text{or} \quad t = \big(\frac{\gamma_2}{\gamma_1}\big)^{1/2}\sqrt{s}$$

and

$$\lambda_{\max}(\operatorname{cof} F) = \frac{\gamma_2}{\gamma_3}\,\lambda^2_{\max}(F) \quad \text{or} \quad t = \frac{\gamma_2}{\gamma_3}\,s^2,$$

respectively. In the sequel we write

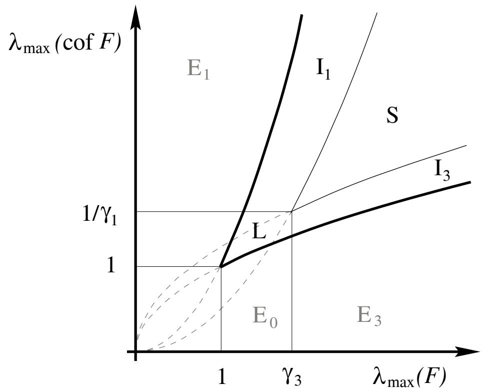

FIGURE 1. The phase diagram for the relaxed energy.

$$\gamma^* = \frac{\gamma_2}{\gamma_3} < 1 \quad \text{and} \quad \Gamma^* = \frac{\gamma_2}{\gamma_1} > 1. \tag{12}$$

We now define the following sets of matrices:

$$\begin{aligned}
\mathrm{L} &= \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_{\max}(F) \le \gamma_3,\, \lambda_{\max}(\operatorname{cof} F) \le \frac{1}{\gamma_1}\big\},\\
\mathrm{I}_1 &= \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_{\max}(\operatorname{cof} F) \ge \frac{1}{\gamma_1},\\
&\qquad \gamma^* \lambda^2_{\max}(F) \le \lambda_{\max}(\operatorname{cof} F) \le \lambda^2_{\max}(F)\big\},\\
\mathrm{I}_3 &= \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_{\max}(F) \ge \gamma_3,\\
&\qquad \sqrt{\lambda_{\max}(F)} \le \lambda_{\max}(\operatorname{cof} F) \le \sqrt{\Gamma^* \lambda_{\max}(F)}\big\},\\
\mathrm{S} &= \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1,\\
&\qquad \sqrt{\Gamma^* \lambda_{\max}(F)} \le \lambda_{\max}(\operatorname{cof} F) \le \gamma^* \lambda^2_{\max}(F)\big\}.
\end{aligned}$$

These sets of deformation gradients correspond to deformations for which the material has a liquid-like (L), an intermediate, solid-liquid-like (I), and a solid-like (S) behaviour, respectively, see our discussion in Section 6. With some abuse of notation we denote by L, S, I_1, and I_3 also the corresponding subsets in the (s,t)-plane which are formally given by replacing $\lambda_{\max}(F)$ by s and $\lambda_{\max}(\operatorname{cof} F)$ by t in the foregoing definition. See Figure 1 for a sketch of these curves and domains in the phase plane.

We now deduce in Propositions 5.1 and 5.3 the results corresponding to Steps 1 and 2 in Section 4.

Proposition 5.1. *Suppose that W is given by (1) with $0 < \gamma_1 \le \gamma_2 \le \gamma_3$ and $\gamma_1\gamma_2\gamma_3 = 1$. Let*

$$\widetilde{W}(F) = \begin{cases} 0 & \text{if } F \in \mathrm{L}, \\ \Big(\dfrac{\lambda_1(F)}{\gamma_1}\Big)^p + 2\Big(\dfrac{\gamma_1}{\lambda_1(F)}\Big)^{p/2} - 3 & \text{if } F \in \mathrm{I}_1, \\ W(F) & \text{if } F \in \mathrm{S}, \\ \Big(\dfrac{\lambda_3(F)}{\gamma_3}\Big)^p + 2\Big(\dfrac{\gamma_3}{\lambda_3(F)}\Big)^{p/2} - 3 & \text{if } F \in \mathrm{I}_3, \\ +\infty & \text{else.} \end{cases} \tag{13}$$

Then $W^{\mathrm{rc}} \le \widetilde{W}$. Moreover, for all matrices $F \in \mathrm{L}$ the function $\widetilde{W}$ is obtained by averaging with respect to laminates within laminates, and for all matrices $F \in \mathrm{I}_1 \cup \mathrm{I}_3$ by averaging with respect to simple laminates. In both cases, we find pairs $(\lambda_i, F_i)_{i=1,\dots,N(F)}$ with $N(F) \le 4$ satisfying (3) or (4), respectively, such that $\widetilde{W}(F) = W(F_i)$, i.e., the matrix F and the matrices F_i are contained in the level set $\{X : \widetilde{W}(X) = W(F_1)\}$.

Proof. The proof is based on several applications of Theorem 3.1, which we describe for the regions L, I_1 and I_3 separately. There is nothing to show for the region of solid behaviour since the relaxed energy coincides there with the original energy. Moreover, the assertion is an immediate consequence of Proposition 3.5 for $F \in \mathrm{L} = K^{\mathrm{qc}}$, since W^{rc} is rank-one convex and $W \equiv 0$ on K.

Suppose now that $F \in \mathrm{I}_3$. We may assume without loss of generality that F is diagonal, $F = \mathrm{diag}(\mu_1, \mu_2, \mu_3)$, with $0 \le \mu_1 \le \mu_2 \le \mu_3$ and $\mu_3 \ge \gamma_3$. Consider the line parallel to the t-axis through the point $(\mu_3, 1/\mu_1)$, and let $(\overline{s}, \overline{t})$ be the intersection point of this line with the curve $\big(s, \sqrt{\Gamma^* s}\big)$, i.e., $\overline{s} = \mu_3$ and $\overline{t} = \sqrt{\Gamma^*\mu_3}$. This point in the phase plane corresponds to the set $\widetilde{K}$ of all matrices G with the corresponding singular values $\overline{s}$ and $1/\overline{t}$, namely

$$\widetilde{K} = \big\{G \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_1(G) = \xi_1, \lambda_2(G) = \xi_2,\, \lambda_3(G) = \mu_3\big\},$$

where

$$\xi_1 = \frac{1}{\sqrt{\Gamma^*\mu_3}} \quad \text{and} \quad \xi_2 = \frac{\sqrt{\Gamma^*}}{\sqrt{\mu_3}}.$$

The idea behind the subsequent estimates is to show, based on Theorem 3.1, that the matrix F is contained in the quasiconvex hull of $\widetilde{K}$. We first observe that

$$\frac{1}{\sqrt{\Gamma^*\mu_3}} = \xi_1 \le \xi_2 = \frac{\sqrt{\Gamma^*}}{\sqrt{\mu_3}} = \Big(\frac{\gamma_2}{\gamma_1}\Big)^{1/2}\frac{1}{\sqrt{\mu_3}} \le \mu_3,$$

since the last inequality is equivalent to $\gamma_2^2\gamma_3 \le \mu_3^3$. Moreover, $F \in \mathrm{I}_3$ implies that

$$\frac{1}{\mu_1} \le \sqrt{\Gamma^*\mu_3} \quad \Leftrightarrow \quad \frac{1}{\sqrt{\Gamma^*\mu_3}} \le \mu_1 \quad \Leftrightarrow \quad \mu_2 = \frac{1}{\mu_1\mu_3} \le \frac{\sqrt{\Gamma^*}}{\sqrt{\mu_3}}.$$

Consequently $\xi_1 \le \mu_1 \le \mu_2 \le \xi_2$, and we conclude from Theorem 3.1 and Corollary 3.5 that $F \in \widetilde{K}^{(1)}$ and

$$W^{\mathrm{rc}}(F) \le W\big(\mathrm{diag}(\xi_1, \xi_2, \mu_3)\big) = 2\big(\frac{\gamma_3}{\mu_3}\big)^{p/2} + \big(\frac{\mu_3}{\gamma_3}\big)^p - 3,$$

as asserted.

It only remains to consider the case $F \in \mathrm{I}_1$, where we may assume that $F = \mathrm{diag}(\mu_1, \mu_2, \mu_3)$ with $\mu_1 \le \gamma_1$. In this case we consider the line parallel to the s axis through $(\mu_3, 1/\mu_1)$ and consider the intersection point of this line with the curve $\big(s, \gamma^* s^2\big)$ at $\big(1/\sqrt{\gamma^*\mu_1}, 1/\mu_1\big)$. Here the goal is to show that F is contained in the quasiconvex hull of the set

$$\widetilde{K} = \big\{G \in \mathbb{M}^{3\times 3} : \det F = 1,\, \lambda_1(G) = \mu_1, \lambda_2(G) = \xi_2,\, \lambda_3(G) = \xi_3\big\},$$

where

$$\xi_2 = \frac{\sqrt{\gamma^*}}{\sqrt{\mu_1}} \quad \text{and} \quad \xi_3 = \frac{1}{\sqrt{\gamma^*\mu_1}}.$$

We now have that

$$\mu_1 \le \big(\frac{\gamma_2}{\gamma_3}\big)^{1/2}\frac{1}{\sqrt{\mu_1}} = \frac{\sqrt{\gamma^*}}{\sqrt{\mu_1}} = \xi_2 \le \xi_3 = \frac{1}{\sqrt{\gamma^*\mu_1}},$$

since the first inequality is equivalent to $\mu_1^3 \le \gamma_1\gamma_2^2$. In addition,

$$\gamma^*\mu_3^2 \le \frac{1}{\mu_1} \quad \Leftrightarrow \quad \frac{\sqrt{\gamma^*}}{\sqrt{\mu_1}} \le \frac{1}{\mu_1\mu_3} = \mu_2 \quad \Leftrightarrow \quad \mu_3 \le \frac{1}{\sqrt{\gamma^*\mu_1}}.$$

Theorem 3.1 and Corollary 3.5 thus imply that $F \in \widetilde{K}^{(1)}$ with

$$W^{\mathrm{rc}}(F) \le W\big(\mathrm{diag}(\mu_1, \xi_2, \xi_3)\big) = \big(\frac{\mu_1}{\gamma_1}\big)^p + 2\big(\frac{\gamma_1}{\mu_1}\big)^{p/2} - 3.$$

The proof of the proposition is now complete. $\square$

We can state the foregoing proposition equivalently by saying that

$$W^{\mathrm{rc}}(F) \le \begin{cases} \psi\big(\lambda_{\max}(F), \lambda_{\max}(\mathrm{cof}\, F)\big) & \text{if } \det F = 1, \\ +\infty & \text{else,} \end{cases}$$

where $\psi : \mathbb{R}^2_+ \to \mathbb{R}$ is given by

$$\psi(s,t) = \begin{cases} 0 & \text{if } (s,t) \in \mathrm{E}_0, \\ \big(\frac{1}{\gamma_1 t}\big)^p + 2(\gamma_1 t)^{p/2} - 3 & \text{if } (s,t) \in \mathrm{I}_1 \cup \mathrm{E}_1, \\ \big(\frac{1}{\gamma_1 t}\big)^p + \big(\frac{t}{\gamma_2 s}\big)^p + \big(\frac{s}{\gamma_3}\big)^p - 3 & \text{if } (s,t) \in \mathrm{S}, \\ \big(\frac{s}{\gamma_3}\big)^p + 2\big(\frac{\gamma_3}{s}\big)^{p/2} - 3 & \text{if } (s,t) \in \mathrm{I}_3 \cup \mathrm{E}_3. \end{cases} \tag{14}$$

Here we denote by E_0, E_1, and E_3 the following domains in the phase plane (see Figure 1):

$$\begin{aligned} \mathrm{E}_0 &= [\,0,\gamma_3\,] \times [\,0, \frac{1}{\gamma_1}\,] \supset L, \\ \mathrm{E}_1 &= \big\{(s,t) : t \geq \frac{1}{\gamma_1},\, t \geq s^2\big\}, \\ \mathrm{E}_3 &= \big\{(s,t) : s \geq \gamma_3,\, t \leq \sqrt{s}\big\}. \end{aligned}$$

The next proposition is the key ingredient in the proof of the polyconvexity of $\widetilde{W}$, since ψ is a finite extension of $\widetilde{W}$ which coincides with $\widetilde{W}$ for all matrices in $\big\{F \in \mathbb{M}^{3\times 3} : \det F = 1\big\}$.

Proposition 5.2. *The function ψ defined in (14) is convex and nondecreasing in its arguments.*

Proof. In order to simplify the notation we define the three functions

$$g_1 : \mathrm{E}_1 \cup \mathrm{I}_1 \to \mathbb{R}, \quad g_2 : \mathrm{S} \to \mathbb{R}, \quad g_3 : \mathrm{E}_3 \cup \mathrm{I}_3 \to \mathbb{R}$$

by

$$\begin{aligned} g_1(s,t) &= \big(\frac{1}{\gamma_1 t}\big)^p + 2(\gamma_1 t)^{p/2} - 3, \\ g_2(s,t) &= \big(\frac{1}{\gamma_1 t}\big)^p + \big(\frac{t}{\gamma_2 s}\big)^p + \big(\frac{s}{\gamma_3}\big)^p - 3, \\ g_3(s,t) &= \big(\frac{s}{\gamma_3}\big)^p + 2\big(\frac{\gamma_3}{s}\big)^{p/2} - 3. \end{aligned}$$

We first prove that ψ is continuous. To see this, it suffices to consider ψ on $\partial(\mathrm{E}_3 \cup \mathrm{I}_3)$ and $\partial(\mathrm{E}_1 \cup \mathrm{I}_1)$. If $s = \gamma_3$, then $g_3(\gamma_3, t) = 0$, and along the curve $t = \sqrt{\Gamma^* s}$ we have

$$g_2(s,t) = \big(\frac{1}{\gamma_1\gamma_2 s}\big)^{p/2} + \big(\frac{1}{\gamma_1\gamma_2 s}\big)^{p/2} + \big(\frac{s}{\gamma_3}\big)^p - 3 = g_3(s,t).$$

Similarly, $g_1(s, \frac{1}{\gamma_1}) = 0$ and along the curve $s = \sqrt{t/\gamma^*}$ we obtain

$$g_2(s,t) = \big(\frac{1}{\gamma_1 t}\big)^p + \big(\frac{t}{\gamma_2\gamma_3}\big)^{p/2} + \big(\frac{t}{\gamma_2\gamma_3}\big)^{p/2} - 3 = g_1(s,t).$$

In order to prove that ψ is nondecreasing, we calculate

$$\begin{aligned} Dg_1(s,t) &= \big(0, -\frac{p}{\gamma_1^p t^{p+1}} + p\gamma_1^{p/2} t^{p/2-1}\big), \\ Dg_2(s,t) &= \big(-\frac{pt^p}{\gamma_2^p s^{p+1}} + \frac{ps^{p-1}}{\gamma_3^p}, -\frac{p}{\gamma_1^p t^{p+1}} + \frac{pt^{p-1}}{\gamma_2^p s^p}\big), \\ Dg_3(s,t) &= \big(-\frac{p\gamma_3^{p/2}}{s^{p/2+1}} + \frac{ps^{p-1}}{\gamma_3^p}, 0\big). \end{aligned}$$

We obtain from these formulae that

$$\begin{aligned} \partial_t g_1(s,t) \geq 0 \quad &\Leftrightarrow \quad t^{3p/2} \geq \Big(\frac{1}{\gamma_1}\Big)^{3p/2}, \\ \partial_s g_2(s,t) \geq 0 \quad &\Leftrightarrow \quad s^{2p} \geq \Big(\frac{t}{\gamma^*}\Big)^p, \\ \partial_t g_2(s,t) \geq 0 \quad &\Leftrightarrow \quad t^{2p} \geq (\Gamma^* s)^p, \\ \partial_s g_3(s,t) \geq 0 \quad &\Leftrightarrow \quad s^{3p/2} \geq \gamma_3^{3p/2}. \end{aligned}$$

All these inequalities are satisfied in the domains of the functions g_i, and we conclude that ψ is nondecreasing in its arguments. We now show that ψ is continuously differentiable. Since

$$\partial_s g_3(\gamma_3, t) = 0, \quad \partial_t g_1(s, \frac{1}{\gamma_1}) = 0,$$

we only need to check this along the curves $t = \sqrt{\Gamma^* s}$ and $t = \gamma^* s^2$, respectively. A short calculation shows that

$$Dg_2\big(s, \big(\frac{\gamma_2}{\gamma_1}\big)^{1/2}\sqrt{s}\big) = Dg_3(s,t), \quad Dg_2\big(\big(\frac{\gamma_3}{\gamma_2}\big)^{1/2}\sqrt{t}, t\big) = Dg_1(s,t),$$

and this establishes the differentiability of the function ψ.

It remains to prove the convexity of ψ. It is clear that g_1 and g_3 are convex since the functions $s \mapsto s^q$ and $s \mapsto s^{-q}$ are convex on $\mathbb{R}_+$ for $q \geq 1$. We obtain for g_2 that

$$D^2 g_2(s,t) = \begin{pmatrix} \frac{p(p+1)t^p}{\gamma_2^p s^{p+2}} + \frac{p(p-1)s^{p-2}}{\gamma_3^p} & -\frac{p^2 t^{p-1}}{\gamma_2^p s^{p+1}} \\ -\frac{p^2 t^{p-1}}{\gamma_2^p s^{p+1}} & \frac{p(p+1)}{\gamma_1^p t^{p+2}} + \frac{p(p-1)t^{p-2}}{\gamma_2^p s^p} \end{pmatrix}$$

and thus the determinant of the matrix of the second derivatives is given by

$$\frac{p^2(p+1)^2\gamma_3^p}{s^{p+2}t^2} - \frac{p^2 t^{2p-2}}{\gamma_2^{2p} s^{2p+2}} + \frac{p^2(p^2-1)\gamma_2^p s^{p-2}}{t^{p+2}} + \frac{p^2(p-1)^2\gamma_1^p t^{p-2}}{s^2}.$$

By assumption, $\frac{1}{s^2} \leq \frac{\gamma^*}{t}$ and thus for $p \geq 2$ and $(p-1)^2 \geq 1$,

$$\frac{p^2(p-1)^2\gamma_1^p t^{p-2}}{s^2} - \frac{p^2 t^{2p-2}}{\gamma_2^{2p} s^{2p+2}} \geq \frac{p^2\gamma_1^p t^{p-2}}{s^2} - \frac{p^2 t^{2p-2}}{\gamma_3^p\gamma_2^p t^p s^2} = 0.$$

Since also $(D^2 g)_{11} > 0$, we conclude that g_2 is convex on its domain and this finishes the proof of the proposition. $\square$

Proposition 5.3. *Assume that* $\psi : \mathbb{R}^2_+ \to \mathbb{R}$ *is given by (14). Then the function* $\Psi_1 : \mathbb{M}^{3\times 3} \to \mathbb{R}$, *given by*

$$\Psi_1(F) = \psi\big(\lambda_{\max}(F), \lambda_{\max}(\operatorname{cof} F)\big)$$

is polyconvex.

Proof. By definition, Ψ_1 is polyconvex if there exists a convex function

$$g : \mathbb{M}^{3\times 3} \times \mathbb{M}^{3\times 3} \times \mathbb{R} \to \mathbb{R}$$

such that $\Psi_1(F) = g(F, \operatorname{cof} F, \det F)$. We define

$$g(X, Y, \delta) = \psi\big(\sup_{e\in\mathbb{S}^2} |Xe|, \sup_{e\in\mathbb{S}^2} |Ye|\big).$$

It follows that for all matrices X_1, X_2, Y_1, $Y_2 \in \mathbb{M}^{3\times 3}$, scalars δ_1, $\delta_2 \in \mathbb{R}$ and $\lambda \in [0,1]$

$$\begin{aligned}
&g\big(\lambda(X_1, Y_1, \delta_1) + (1-\lambda)(X_2, Y_2, \delta_2)\big)\\
&= \psi\big(\sup_{e\in\mathbb{S}^2} |(\lambda X_1 + (1-\lambda)Y_1)e|, \sup_{e\in\mathbb{S}^2} |(\lambda X_2 + (1-\lambda)Y_2)e|\big)\\
&\le \psi\big(\lambda \sup_{e\in\mathbb{S}^2} |X_1 e| + (1-\lambda) \sup_{e\in\mathbb{S}^2} |Y_1 e|, \lambda \sup_{e\in\mathbb{S}^2} |X_2 e| + (1-\lambda) \sup_{e\in\mathbb{S}^2} |Y_2 e|\big)\\
&= \psi\big(\lambda\big(\sup_{e\in\mathbb{S}^2} |X_1 e|, \sup_{e\in\mathbb{S}^2} |X_2 e|\big) + (1-\lambda)\big(\sup_{e\in\mathbb{S}^2} |Y_1 e|, \sup_{e\in\mathbb{S}^2} |Y_2 e|\big)\big)\\
&\le \lambda\psi\big(\sup_{e\in\mathbb{S}^2} |X_1 e|, \sup_{e\in\mathbb{S}^2} |Y_1 e|\big) + (1-\lambda)\psi\big(\sup_{e\in\mathbb{S}^2} |X_2 e|, \sup_{e\in\mathbb{S}^2} |Y_2 e|\big)\\
&= \lambda g\big(X_1, Y_1, \delta_1\big) + (1-\lambda) g\big(X_2, Y_2, \delta_2\big).
\end{aligned}$$

Here we used the triangle inequality for the norm and the fact that ψ is nondecreasing in the first inequality, and the convexity of ψ for the second inequality. This establishes the polyconvexity of Ψ_1 and concludes the proof. $\square$

Theorem 5.4. *The rank-one convex and the polyconvex envelope of* W *coincide and are given by*

$$W^{\mathrm{rc}}(F) = W^{\mathrm{pc}}(F) = \begin{cases} \psi\big(\lambda_{\max}(F), \lambda_{\max}(\operatorname{cof} F)\big) & \text{if } \det F = 1,\\ +\infty & \text{else.}\end{cases}$$

Proof. We define

$$\Psi_2(F) = I_1(\det F) \quad \text{where} \quad I_1(t) = \begin{cases} 0 & \text{if } t = 1,\\ \infty & \text{else.}\end{cases}$$

Then $\widehat{W}(F) = \Psi_1(F) + \Psi_2(F)$ is a polyconvex function which is finite only on the set $\{F \in \mathbb{M}^{3\times 3} : \det F = 1\}$. This implies that

$$\lambda_1(F) = \lambda_{\min}(F) = \frac{1}{\lambda_{\max}(\operatorname{cof} F)} \quad \text{and} \quad \lambda_2(F) = \frac{1}{\lambda_{\min}(F)\lambda_{\max}(F)}$$

whenever the energy is finite. In view of the definition of ψ,

$$\widehat{W}(F) = \begin{cases} 0 & \text{if } F \in L, \\ \big(\frac{\lambda_{\min}(F)}{\gamma_1}\big)^p + 2\big(\frac{\gamma_1}{\lambda_{\min} F}\big)^{p/2} - 3 & \text{if } F \in \mathrm{I}_1, \\ W(F) & \text{if } F \in \mathrm{S}, \\ \big(\frac{\lambda_{\max}(F)}{\gamma_3}\big)^p + 2\big(\frac{\gamma_3}{\lambda_{\max}(F)}\big)^{p/2} - 3 & \text{if } F \in \mathrm{I}_3, \\ +\infty & \text{else,} \end{cases}$$

and a comparison with (13) shows that $\widehat{W} = \widetilde{W} \le W^{\mathrm{rc}}$. Therefore

$$W^{\mathrm{rc}} \le \widetilde{W} = \widehat{W} \le W^{\mathrm{pc}} \le W^{\mathrm{rc}},$$

and hence equality holds throughout this chain of inequalities. This proves the assertion of the theorem. □

The final step is to prove that the quasiconvex envelope of W is equal to the polyconvex and the rank-one convex hull. This does not follow automatically since quasiconvexity does not imply rank-one convexity for extended valued functions. To close this gap, we use an explicit construction based on the following result by Müller&Šverák.

Lemma 5.5 ([MS99], Lemma 4.1). *Let Σ be given by*

$$\Sigma = \{F \in \mathbb{M}^{m\times n} : M(F) = t\},$$

where M is a minor of F and $t \neq 0$. Let V be an open set in Σ, let $F \in V^{rc}$, and let $\varepsilon > 0$. Then there exists a piecewise linear map $u : \Omega \subset \mathbb{R}^n \to \mathbb{R}^m$ such that $Du \in V^{rc}$ a.e. in Ω and

$$\big|\{x : Du(x) \notin V\}\big| < \varepsilon|\Omega|, \qquad u(x) = Fx \text{ on } \partial\Omega.$$

Theorem 5.6. *The quasiconvex envelope of W,*

$$W^{\mathrm{qc}}(F) = \inf_{\substack{\varphi \in W^{1,\infty}(\Omega;\mathbb{R}^3) \\ \varphi(x) = Fx \text{ on } \partial\Omega}} \frac{1}{|\Omega|}\int_\Omega W(D\varphi(x))\mathrm{d}x,$$

is equal to the rank-one convex and the polyconvex envelope and given by

$$W^{\mathrm{qc}}(F) = \begin{cases} \psi\big(\lambda_{\max}(F), \lambda_{\max}(\operatorname{cof} F)\big) & \text{if } \det F = 1, \\ +\infty & \text{else,} \end{cases}$$

where ψ is given by (14).

Proof. We have to construct for all $F \in \mathbb{M}^{3\times 3}$ with $\det F = 1$ and for all $\delta > 0$ a function $\varphi_{F,\delta} \in W^{1,\infty}(\Omega;\mathbb{R}^3)$ such that $\varphi_{F,\delta} = Fx$ on $\partial\Omega$ and

$$\int_\Omega W(D\varphi_{F,\delta})\mathrm{d}x \leq |\Omega| W^{\mathrm{pc}}(F) + \mathcal{O}(\delta),$$

where $\mathcal{O}(\delta) \to 0$ as $\delta \to 0$. This implies $W^{\mathrm{qc}}(F) \leq W^{\mathrm{pc}}(F)$, and since W^{pc} is quasiconvex, we conclude $W^{\mathrm{qc}} = W^{\mathrm{pc}}$.

We give the proof for the situation that W^{pc} is obtained from W by averaging with respect to laminates within laminates. If follows from Proposition 5.1 that there exist pairs $(\lambda_i, F_i)_{i=1,\ldots,4}$ such that

$$F = \sum_{i=1}^{4} \lambda_i F_i, \quad \text{and} \quad W^{\mathrm{pc}}(F) = W(F_i),\ i = 1,\ldots,4.$$

Moreover, $F \in \widetilde{K}^{(2)}$ where $\widetilde{K} = \{F_1, F_2, F_3, F_4\}$. We choose

$$\Sigma = \big\{F \in \mathbb{M}^{3\times 3} : \det F = 1\big\},$$

and define for $\delta > 0$

$$V_\delta = \big\{F \in \Sigma : \operatorname{dist}(F, \widetilde{K}) < \delta\big\},\ \omega_\delta = \sup\big\{W(X) : X \in V_\delta\big\} - W^{\mathrm{pc}}(F).$$

Since W is continuous on Σ we have $\omega_\delta \to 0$ as $\delta \to 0$. Lemma 5.5 guarantees the existence of a piecewise linear map $\varphi_{F,\delta} : \Omega \to \mathbb{R}^3$ with $D\varphi_{F,\delta}(x) \in V_\delta^{\mathrm{rc}}$ a.e. and

$$\varphi_{F,\delta}(x) = Fx \text{ on } \partial\Omega, \quad \text{and} \quad \big|\big\{x \in \Omega : D\varphi_{F,\delta}(x) \notin V_\delta\big\}\big| \leq \delta|\Omega|.$$

Therefore, if M is an upper bound for W on V_1,

$$\begin{aligned}\int_\Omega W(D\varphi_{F,\delta})\mathrm{d}x &\leq \big|\big\{D\varphi_{F,\delta}(x) \in V_\delta\big\}\big|\big(W^{\mathrm{pc}}(F) + \omega_\delta\big) + \delta M|\Omega| \\ &\leq |\Omega| W^{\mathrm{pc}}(F) + |\Omega|\big(\omega_\delta + \delta M\big).\end{aligned}$$

The assertion of the theorem follows as $\delta \to 0$. □

6. Perspectives for nematic elastomers

Nematic elastomers are rubbery solids which combine the entropic elasticity of a network of cross-linked polymeric chains with the peculiar optical properties of nematic liquid crystals, see Figure 2 for a sketch. These features have already attracted considerable attention in the chemical and physical literature and more recently also in the mathematical literature from the point of view of continuum mechanics, see [ACF, DS].

The motivation behind the successful efforts to synthesize nematic elastomers [FKR, KpF] was the attempt to reproduce the mesophases typical of a liquid crystal within an amorphous, polymeric solid. The resulting physical system has the translational order of a solid phase coupled to the orientational order of a nematic phase. Soft deformation modes whose occurrence had been predicted by theory [GL], were observed experimentally

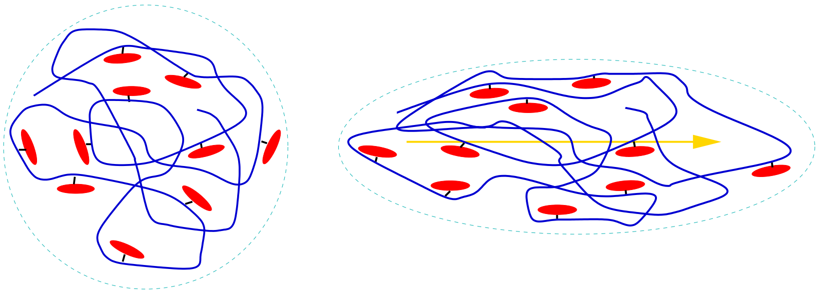

FIGURE 2. *Sketch of the isotropic-nematic phase transformation in nematic elastomers. They consist of cross-linked backbone chains to which nematic elements (rigid, rod-like molecules) are attached. The nematic mesogens have a random orientation in the high temperature (isotropic) phase due to thermal fluctuations. A local alignment of the mesogens in the low temperature (nematic) phase causes a stretch of the network in direction of n (indicated by the arrow in the right figure) and a contraction in the directions perpendicular to it.*

[KnF], in association with the appearance of domain patterns with a characteristically layered texture. The formation of these microstructures was explained by energy minimization in the framework of continuum models in [WB], [WT]. One of the proposed models [BTW] is based on minimization of the free energy density,

$$W_{BTW}(F,n) = \begin{cases} \frac{\mu}{2}\Big(r^{1/3}\big[\,|F|^2 - \frac{r-1}{r}|F^T n|^2\,\big] - 3\Big) & \text{if } \det F = 1, \\ +\infty & \text{otherwise.} \end{cases}$$

Here μ and r are positive, temperature dependent material constants, the rubber energy scale and the backbone anisotropy parameter (i.e. the statistical average of the ratio of the dimensions of a chain in directions parallel and perpendicular to n), and n is the director describing the local directional order in the nematic phase. In the isotropic phase, $r = 1$, and the system is governed by the neo-hookean energy of the rubber. For temperatures below the transformation temperature, $r > 1$, and a minimization of the energy in n leads to

$$W(F) = \min_{n\in\mathbb{S}^2} W_{BTW}(F,n) = \begin{cases} \frac{\mu}{2}\Big(r^{1/3}\big[\,\lambda_1^2 + \lambda_2^2 + \frac{1}{r}\lambda_3^2\big] - 3\Big), & \det F = 1, \\ +\infty & \text{else.} \end{cases}$$

Here we have chosen the stress-free state of the isotropic parent phase as reference configuration rather than one of the stress-free states of the product uniaxial phase as it is done in [BTW, WT].

The mathematical interest in this energy lies in its nonconvexity caused by the factor $1/r < 1$ in front of the largest singular value. Its expression is up to the factor $\frac{\mu}{2}$ identical to (1) with

$$\gamma_1 = \gamma_2 = \frac{1}{r^{1/6}} < r^{1/3} = \gamma_3 \quad \text{and} \quad \gamma^* = \frac{1}{\sqrt{r}},\, \Gamma^* = 1,$$

where the parameters γ^* and Γ^* have been defined in (12).

Our results in Sections 3 and 5, Theorems 3.1 and 5.6, imply immediately the following result (see the phase diagram in Figure 3):

Theorem 6.1. *The relaxed energy W^{qc} of the system is given by*

$$W^{\mathrm{qc}}(F) = \begin{cases} 0 & \text{if } F \in \mathrm{L} \\ \frac{\mu}{2}\Big(\frac{2}{r^{1/6}\lambda_{\min}(F)} + r^{1/3}\lambda^2_{\min}(F) - 3\Big) & \text{if } F \in \mathrm{I}_1, \\ W(F) & \text{if } F \in \mathrm{S}, \\ +\infty & \text{else}, \end{cases}$$

where

$$\begin{aligned} \mathrm{L} &= \big\{F \in \mathbb{M}^{3\times3} : \det F = 1,\, \lambda_{\max}(F) \le r^{1/3}\big\}, \\ \mathrm{I}_1 &= \big\{F \in \mathbb{M}^{3\times3} : \det F = 1,\, \lambda_{\min}(F)\lambda^2_{\max}(F) \le r^{1/2}\big\}, \\ \mathrm{S} &= \big\{F \in \mathbb{M}^{3\times3} : \det F = 1,\, \lambda_{\min}(F)\lambda^2_{\max}(F) \ge r^{1/2}\big\}. \end{aligned}$$

The set K^{qc} of all affine, asymptotically zero-energy deformations is equal to $K^{(2)}$. Moreover, deformations in $K^{(1)}$ and $K^{(2)}$ can be realized by simple laminates and laminates within laminates, respectively, and can be represented by

$$\begin{aligned} K^{(1)} &= \Big\{F \in \mathbb{M}^{3\times3} : \det F = 1,\, \lambda_{\min}(F) = \frac{1}{r^{1/6}}\Big\}, \\ K^{(2)} &= \Big\{F \in \mathbb{M}^{3\times3} : \det F = 1,\, \frac{1}{r^{1/6}} \le \lambda_{\min}(F) \le \lambda_{\max}(F) \le r^{1/3}\Big\}. \end{aligned}$$

Finally, the relaxed energy in I_1 can be achieved by averaging with respect to simple laminates.

From the expression of W^{qc} it is clear that in the region (L) of the phase diagram drawn in Figure 3 the response of the system is completely soft, and only constrained by incompressibility. Thus, in the region (L), the material behaves essentially like a liquid. On the other hand, in the region (S) of the phase diagram, the expression for the energy is very similar to the one describing a neo-hookean rubber, hence the material behaves like an elastic solid. In the intermediate region (I_1), the energy depends only on the smallest singular value of F (the dependence on the other two singular values is indirect, through the incompressibility constraint). Thus the material response is intermediate between a liquid-like and a solid-like one.

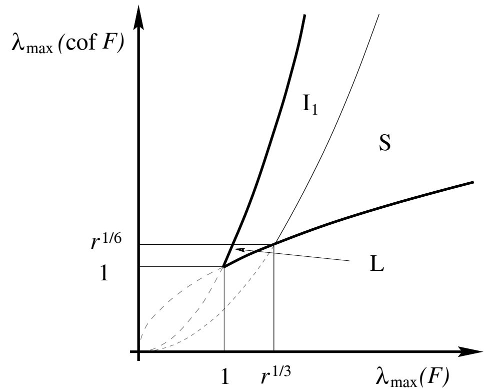

FIGURE 3. *The phase diagram for nematic elastomers. The phase boundary between the intermediate phase and the solid phase is given by* $\lambda_{\min}(F)\lambda^2_{\max}(F) = r^{1/2}$.

The significance of Theorem 6.1 from the point of view of Continuum Physics is that it represents the first explicit relaxation result for an SO(3)-invariant energy related to solid-solid phase transitions. In particular, it allows one to explore analytically and numerically realistic loading conditions and boundary value problems that involve microstructures whose characteristic features are not spatially homogeneous. Numerical experiments with a thin sheet of a nematic elastomer in a geometry for which experimental results are available in the literature are presented in [CDD].

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