Relaxation of some transversally isotropic energies and applications to smectic A elastomers

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We determine the relaxation of some transversally-isotropic energy densities, i.e., functions $W : \mathbb{R}^{3\times3} \to [0,\infty]$ with the property $W(QFR) = W(F)$ for all $Q \in SO(3)$ and all $R \in SO(3)$ such that $Rn_0 = n_0$, where n_0 is a fixed unit vector. One physically relevant example is a model for smectic A elastomers. We discuss the implications of our result for the computation of macroscopic stress-strain curves for this material and compare with experiment.

1 Introduction

Quasiconvexity, introduced by Morrey in 1952 [27], is fundamental to understand lower semicontinuity and existence in the vectorial calculus of variations, and has proven to be a very effective tool to study microstructure formation in materials. Examples include shape-memory materials [4, 9, 5, 28], shape optimization [24, 3], composite materials [23, 7, 26], and nematic elastomers [35]. The macroscopic behavior of phase-transforming materials can in principle be determined by computing the quasiconvex envelope of the appropriate energy density; one of the few examples where this has been accomplished is that of nematic elastomers [19, 12]. A key feature that makes the analysis possible in the case of (ideally soft) nematic elastomers is the isotropy of the material, which leads to full rotational symmetry.

In this paper we address problems with cylindrical symmetry, the prototype being $W_{\mathrm{A},\infty} : \mathbb{R}^{3\times 3} \to [0,\infty]$ defined by

$$W_{\mathrm{A},\infty}(F) = \begin{cases} |F|^2 & \text{if } \det F = 1 \text{ and } |\operatorname{cof} F n_0| = d_0\,, \\ \infty & \text{else,} \end{cases}$$

where $n_0 \in \mathbb{R}^3$ is a fixed unit vector, and $d_0 > 0$ a parameter. We are interested in the macroscopic response arising from such an energy, which is characterized by the quasiconvex envelope.

Theorem 1.1. *The quasiconvex envelope of* $W_{\mathrm{A},\infty}$ *is*

$$W^{\mathrm{qc}}_{\mathrm{A},\infty}(F) = \begin{cases} |Fn_0|^2 + f(\lambda_{\max}(FP)) & \textit{if } \det F = 1 \textit{ and } |\operatorname{cof} F n_0| \le d_0\,, \\ \infty & \textit{else,} \end{cases} \tag{1.1}$$

where $P = \mathrm{Id} - n_0 \otimes n_0$*,*

$$f(x) = \begin{cases} x^2 + \dfrac{d_0^2}{x^2} & \textit{if } x > d_0^{1/2}\,, \\ 2d_0 & \textit{else.} \end{cases} \tag{1.2}$$

We recall that the quasiconvex envelope of a function $W : \mathbb{R}^{m\times n} \to [0,\infty]$ is defined as [27, 6, 21, 28, 20]

$$\begin{aligned} W^{\mathrm{qc}}(F) \;=\; \sup\big\{&\psi(F) : \psi : \mathbb{R}^{m\times n} \to [0,\infty], \psi \text{ quasiconvex}, \\ &\psi(G) \le W(G) \text{ for all } G \in \mathbb{R}^{m\times n}\big\}\,. \end{aligned} \tag{1.3}$$

A function $\psi : \mathbb{R}^{m\times n} \to [0,\infty]$ is quasiconvex if affine functions are minimizers with respect to their own boundary conditions, in the sense that

$$\psi(F) \le \int_{(0,1)^n} \psi(F + Dv)dx \qquad \forall v \in W_0^{1,\infty}((0,1)^n;\mathbb{R}^m)\,, \tag{1.4}$$

provided the integral exists. We recall that in the presence of suitable growth conditions – both from above and from below – the lower semicontinuous envelope of the integral functional $E[u] = \int W(Du)dx$ is determined by the quasiconvex envelope of W, namely, $sc^- E[u] = \int W^{\mathrm{qc}}(Du)dx$, see, e.g., [15, 28, 25]. The same has however not yet been proven, to the best of our knowledge, for the extended-valued case of interest here. Finally, the largest singular value of the matrix $F \in \mathbb{R}^{m\times n}$ is defined by

$$\lambda_{\max}(F) = \sup\left\{|Fe| : e \in \mathbb{R}^n, |e| = 1\right\}. \tag{1.5}$$

The supremum is actually a maximum; $\lambda_{\max}(F)$ is a convex function of F.

A result analogous to Theorem 1.1 can be obtained if the simpler side condition $|Fn_0| = d_0$ is used instead of $|\operatorname{cof} Fn_0| = d_0$, see Proposition 2.6 below. The resulting model describes an elastic material which is inextensible in the direction of n_0.

We then consider the case where the constraint $|\operatorname{cof} Fn_0| = d_0$ is relaxed, and replaced by an energy penalization. To be precise, let

$$W_{\mathrm{A},k}(F) = \begin{cases} |F|^2 + k\left(|\operatorname{cof} Fn_0| - d_0\right)^2 & \text{if } \det F = 1\,, \\ \infty & \text{else,} \end{cases}$$

where $k > 0$ is a parameter.

Theorem 1.2. *The quasiconvex envelope of $W_{\mathrm{A},k}$ is*

$$W^{\mathrm{qc}}_{\mathrm{A},k}(F) = \begin{cases} |Fn_0|^2 + f(\lambda^2_{\max}(FP), |\operatorname{cof} Fn_0|) & \textit{if } \det F = 1\,, \\ \infty & \textit{else,} \end{cases}$$

where $f : \{(b, d) \in \mathbb{R}^2 : 0 < d \le b\} \to \mathbb{R}$ is defined by

$$f(b, d) = \begin{cases} b + \dfrac{d^2}{b} + k(d - d_0)^2 & \textit{if } d \ge \dfrac{kd_0 b}{kb+1}\,, \\ b + \dfrac{kd_0^2}{kb+1} & \textit{if } b \ge d_0 - \dfrac{1}{k} \textit{ and } d \le \dfrac{kd_0 b}{kb+1}\,, \\ 2d_0 - \dfrac{1}{k} & \textit{if } b \le d_0 - \dfrac{1}{k}\,, \end{cases} \tag{1.6}$$

(see Figure 1) and $P = \mathrm{Id} - n_0 \otimes n_0$.

One main difficulty in obtaining this type of result is to express the energy in a way which permits to reduce the problem of quasiconvexity to convexity in a lower dimensional space. The key to this reduction is the identification of an appropriate set of variables, which reveals the hidden structural properties of the energy. Here, use of $\lambda_{\max}(FP)$ and $|\operatorname{cof} Fn_0|$ is crucial.

The energy $W_{\mathrm{A},k}$ is a model for smectic A elastomers, a particular phase of liquid crystal elastomers, materials which display a number of interesting mechanical and optical properties due to the coupling between the ordering transition as in ordinary liquid crystals and the rubber elasticity of the underlying network [35]. Smectic A elastomers are characterized by rod-like molecules (mesogens) assembled in a layered structure, the rods being normal to the layers. The mesogens are attached to polymer chains, which are cross-linked to obtain a rubber-like solid; the coupling to liquid crystal ordering leads to rubbery response in the tangential directions, and solid-like along the layer normal. Monodomain smectic A elastomers were recently synthesized

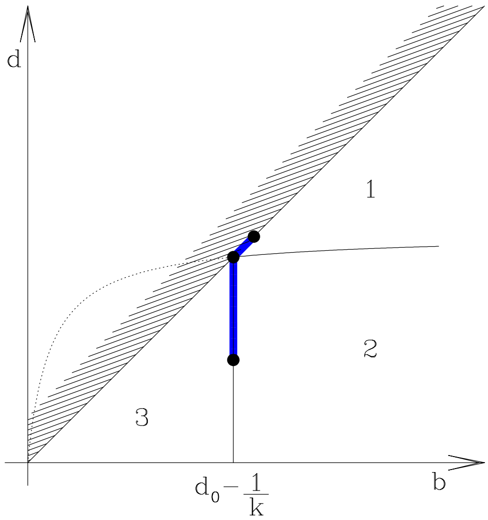

FIGURE 1: Phase diagram. The regions 1,2,3 represent the three domains in (1.6). Only the region $b \geq d$ is accessible. The thick blue line joining the three dots sketches the path followed in the experiments, see discussion in Section 3.

[31, 33] (see also discussion and references in [35]), mechanical experiments were performed in [30], measuring both the stress-strain diagram and the evolution of the director (via X-ray).

The energy $W_{\mathrm{A},k}$ is very closely related to the one proposed by Adams and Warner [2], based on statistical mechanics. After a change of coordinates in the reference configuration and a rearrangement of the terms (see discussion in the Appendix), this takes the form

$$W_{\mathrm{AW}}(F) = \begin{cases} |F|^2 + kd_0^2 \left(\dfrac{d_0}{|\operatorname{cof} F n_0|} - 1 \right)^2 & \text{if } \det F = 1\,, \\ \infty & \text{else.} \end{cases} \tag{1.7}$$

The two expressions agree to leading order in $|\operatorname{cof} F n_0| - d_0$, which is the relevant regime since $k \gg 1$.

Knowledge of the quasiconvex envelope of the energy density permits computation of the macroscopic mechanical behavior of the material. The results for the experimental geometry of [30] are given in Figure 2; comparison with the experiments from [8] is shown in Figure 3. The figure reports the result of our exact relaxation of $W_{\mathrm{A},k}$, the one obtained by Adams and Warner via partial relaxation of W_{AW}, and the experimental measurements. The two theoretical results are indistinguishable on the scale of the figure, and both agree very well with experiment. While the full relaxation of W_{AW} is unknown, one

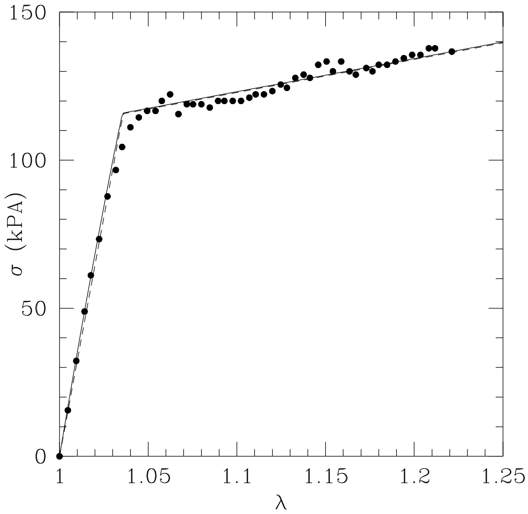

FIGURE 2: Comparison of the computed stress-strain response with experimental measurements from [30] (dots). Our curve (full) is on this scale indistinguishable from the one that was obtained in [2] (dashed). We used $d_0 = 0.82$, $k = 61$, and set the vertical scale by multiplying the stress by the scaling factor 94.5 kPa.

should expect peculiar effects originating from the lack of convexity and coercivity of the penalization term at very large values of $|\operatorname{cof} F n_0|$, see our discussion in Section 3. Phenomenological models of smectic A elastomers from Lagrangian elasticity theory are also capable of similar predictions [34].

The stress-strain response in Figures 2 and 3 is computed assuming a macroscopically affine deformation. Knowledge of $W^{\mathrm{qc}}_{\mathrm{A},k}$ is physically relevant, in that it permits accurate modeling of realistic experimental conditions, where clamps may rule out macroscopically affine deformations. An example where this has been accomplished is the related case of nematic elastomers, leading to a detailed understanding of the spatial modulation of the loading-induced microstructure [11, 12, 13, 19]. It would be interesting to pursue a similar program for the case of smectic A elastomers, but this is beyond the scope of this work.

2 Relaxation results

All proofs are based on showing that the expression we derive is both an upper bound and a lower bound for the quasiconvex envelope. Precisely, we first show that the expressions considered are polyconvex, and lie below the energy. We recall that a function $\varphi : \mathbb{R}^{3\times 3} \to \mathbb{R} \cup \{\infty\}$ is polyconvex if there is

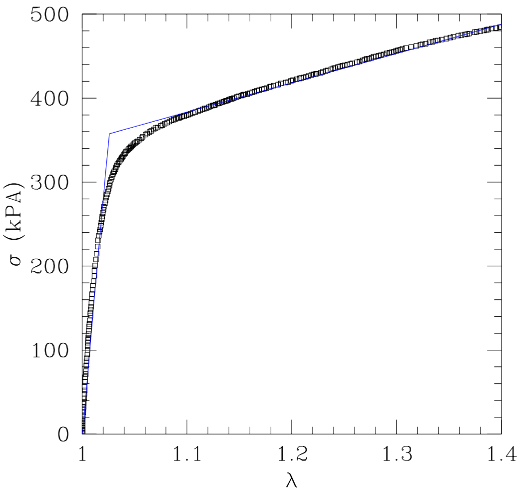

FIGURE 3: Comparison of the computed stress-strain response (full curve) with experimental measurements from [8] (squares). We used $d_0 = 0.9$, $k = 60$, and set the vertical scale by multiplying the stress by the scaling factor 318 kPa. The smoothness in the experimentally measured transition between the "hard" regime at small strain and the "soft" one at large strains can be due, e.g., to the fact that the sample does not have the time to fully relax at each strain, or to spatial inhomogeneities.

a convex function $g : \mathbb{R}^{19} \to \mathbb{R} \cup \{\infty\}$ such that $\varphi(F) = g(F, \operatorname{cof} F, \det F)$ for all F. Further, if φ is polyconvex and $\varphi \le W$, then it follows that $\varphi \le W^{\mathrm{qc}}$. This is the key tool used in proving the lower bound.

The upper bound is instead obtained by explicit constructions. The construction is composed of two main steps. Firstly, a geometric argument in matrix space makes it possible to decompose deformation gradients into laminates, the key lamination steps are given in Lemma 2.1 and Lemma 2.2 below. We say that a matrix F is the average of a laminate supported on F_1 and F_2 if

$$\operatorname{rank}(F_1 - F_2) \le 1 \quad \text{and} \quad F = \lambda F_1 + (1-\lambda) F_2 \ \text{ for some } \lambda \in [0,1]\,. \tag{2.1}$$

It is easy to see that, given such a pair, one can construct a sequence of functions u_j such that $Du_j \in \{F_1, F_2\}$ and $u_j \overset{*}{\rightharpoonup} Fx$ in $W^{1,\infty}$. A simple truncation gives then a function with affine boundary data, which can be used in (1.4) provided that W is locally bounded. In the case of interest here, however, a delicate point is that the energy is infinite if the determinant constraint is violated. In the case of Theorem 1.1 the same holds also for the condition on the cofactor. We deal with this issue by constructing a boundary layer ensuring that the constraints are satisfied pointwise. The construction scheme we use for this purpose uses a deep result on convex integration due to Müller and Šverák [29] (see Theorem 2.3 below); for alternative approaches to similar problems, see [16, 22]. A direct explicit construction can be found in [10].

The key point for our results is the identification of an appropriate set of variables that reflect the structural properties of the free energy density. In the situation at hand, and denoting by $P = \mathrm{Id} - n_0 \otimes n_0$ the projection onto the plane normal to n_0, and by $\lambda_{\max}(F)$ the largest singular value of F, defined as in (1.5), we use

$$\det F\,, \qquad |Fn_0|\,, \qquad |\operatorname{cof} Fn_0|\,, \qquad \lambda_{\max}(FP)\,.$$

These are transversally isotropic polyconvex functions. Moreover, as we shall show below, it is possible to change each of the last three variables by lamination, without modifying the others. Thus, these functions provide basic building blocks for constructing both lower bounds and upper bounds for the transversally isotropic energies considered in this paper. For a list of many transversally-isotropic polyconvex polynomials, see [32].

We begin with the geometric construction of the laminates. In all proofs we assume, after a change of coordinates, that $n_0 = e_3$; with this choice, $\operatorname{cof} Fn_0 = Fe_1 \wedge Fe_2$. Further, we identify $\mathbb{R}^{3\times 2}$ with $\mathbb{R}^{3\times 3}P$ (i.e., the set of 3×3 matrices whose third column is zero), and $\mathbb{R}^{2\times 2}$ with $P\mathbb{R}^{3\times 2}$.

Lemma 2.1. *Let $F \in \mathbb{R}^{3\times 3}$, with $\det F = 1$. Then for any $b > \lambda^2_{\max}(FP)$ there is a pair F_1, F_2 as in (2.1) such that*

$$\det F_j = 1\,, \quad |F_j n_0| = |Fn_0|\,, \quad |\operatorname{cof} F_j n_0| = |\operatorname{cof} Fn_0|\,, \quad j = 1, 2\,,$$

and

$$\lambda^2_{\max}(F_j P) = b\,, \quad j = 1, 2\,.$$

Proof. We assume without loss of generality that $n_0 = e_3$. Since $\det F = 1$ we have $Fe_1 \neq 0$. Consider the rank-one line

$$t \mapsto F_t = F + tFe_1 \otimes e_2 = F(\mathrm{Id} + te_1 \otimes e_2).$$

Clearly $\det F_t = \det F$, $F_t e_3 = Fe_3$, and $\operatorname{cof} F_t e_3 = (F_t e_1) \wedge (F_t e_2) = Fe_1 \wedge Fe_2 = \operatorname{cof} Fe_3$ for all $t \in \mathbb{R}$. At the same time the function $t \mapsto \lambda^2_{\max}(F_t P)$ is continuous, and diverges for $t \to \pm\infty$. Since by assumption $\lambda^2_{\max}(FP) < b$, there are $t_1 > 0 > t_2$ such that the matrices $F_j = F_{t_j}$ satisfy $\lambda^2_{\max}(F_j P) = b$. This concludes the proof. $\square$

Lemma 2.2. *Let $F \in \mathbb{R}^{3\times 3}$, with $\det F = 1$. Then for any $d > |\operatorname{cof} F n_0|$ there is a pair F_1, F_2 as in (2.1) such that*

$$\det F_j = 1\,, \quad |F_j n_0| = |F n_0|\,, \quad |\operatorname{cof} F_j n_0| = d\,, \quad j = 1, 2\,,$$

and

$$\lambda^2_{\max}(F_j P) = \max\left(\lambda^2_{\max}(FP), \frac{d^2}{\lambda^2_{\max}(FP)}\right)\,, \quad j = 1, 2\,.$$

Proof. We assume without loss of generality that $n_0 = e_3$ and that Fe_1 and Fe_2 are perpendicular. The latter can be enforced by replacing F by $F' = FQ$, where $Q \in SO(3)$ obeys $Qe_3 = e_3$ and $FQe_1 \cdot FQe_2 = 0$. Let $v \in \operatorname{span}\{Fe_1, Fe_3\}$ be nonzero and perpendicular to Fe_1, and consider the rank-one line

$$t \mapsto F_t = F + tv \otimes e_2\,.$$

Clearly, $\det F_t = \det F$ and $F_t e_3 = Fe_3$. Since $F_t e_2 = Fe_2 + tv$ is perpendicular to $F_t e_1 = Fe_1$ for all t, we obtain

$$|\operatorname{cof} F_t e_3| = |F_t e_1 \wedge F_t e_2| = |Fe_1|\,|F_t e_2|$$

and

$$\lambda_{\max}(F_t P) = \max\left\{|Fe_1|, |F_t e_2|\right\}\,.$$

Since $d > |\operatorname{cof} F n_0| = |Fe_1|\,|Fe_2|$ and $|F_t e_2|$ is a continuous function that diverges as $t \to \pm\infty$, we may find $t_1 > 0 > t_2$ such that

$$|F_t e_2| = \frac{d}{|Fe_1|}, \quad t = t_j\,.$$

The matrices $F_j = F_{t_j}$ have all the asserted properties. $\square$

Before proving the main results of the present paper we recall the convex integration result we are going to use. Müller and Šverák [29] have shown that solutions to the partial differential inclusion

$$\begin{aligned} Du &\in K \qquad &&\text{a.e. in } \Omega\,, \\ u(x) &= Fx &&\text{on } \partial\Omega\,, \end{aligned}$$

can be obtained for all boundary data F contained in an in-approximation of the set K. An in-approximation of the set $K \subset \mathbb{R}^{3\times 3}$ is a sequence of uniformly bounded, relatively open sets $U_i \subset \{F \in \mathbb{R}^{3\times 3} : \det F = 1\}$, $i \geq i_0$, such that

$$U_i \subset (U_{i+1})^{\mathrm{rc}}, \qquad \text{and} \qquad U_i \to K. \tag{2.2}$$

Here $U_i \to K$ means that $F_i \to F$, $F_i \in U_i$ implies $F \in K$; U^{rc} denotes the rank-one convex hull of U; relatively open means that there are open sets $V_i \subset \mathbb{R}^{3\times 3}$ such that $U_i = V_i \cap \{F : \det F = 1\}$. The properties of U^{rc} of relevance here are that $U \subset U^{\mathrm{rc}}$, and that U^{rc} is closed under lamination, in the sense that if F is the average of a laminate supported on F_1, F_2 (as in (2.1)) with F_1, $F_2 \in U^{\mathrm{rc}}$, then necessarily $F \in U^{\mathrm{rc}}$. For a more detailed presentation, including the precise definition of the rank-one convex hull of a set, we refer to [15, 28, 20] (see [19, 14] for other applications in related contexts).

Theorem 2.3 ([29], Theorem 1.3)**.** *Let $\{U_i\}$ be an in–approximation of the compact set $K \subset \mathbb{R}^{m\times n}$. Then, for any i, any $F \in U_i$ and any open domain $\Omega \subset \mathbb{R}^n$ there exists a Lipschitz solution $u \in W^{1,\infty}(\Omega; \mathbb{R}^m)$ of the partial differential inclusion*

$$\begin{aligned} Du &\in K && \text{a.e. in } \Omega, \\ u(x) &= Fx && \text{on } \partial\Omega. \end{aligned}$$

In order to employ this result, we shall use the following construction of an in-approximation.

Proposition 2.4. *Given a, b, $d > 0$, with $b \geq d$ and $ad^2 > 1$, let*

$$K_{a,b,d} = \left\{F \in \mathbb{R}^{3\times 3} : \det F = 1,\ |Fn_0|^2 = a,\ \lambda^2_{\max}(FP) = b,\ |\operatorname{cof} Fn_0| = d\right\}. \tag{2.3}$$

Then the sets

$$\begin{aligned} U_i \;=\; \big\{&F \in \mathbb{R}^{3\times 3} : \det F = 1,\ a - 2^{-i} < |Fn_0|^2 < a, \\ &b - 2^{-i} < \lambda^2_{\max}(FP) < b,\ d - 2^{-i} < |\operatorname{cof} Fn_0| < d\big\} \end{aligned}$$

(for $i \in \mathbb{Z}$) constitute an in-approximation of $K_{a,b,d}$.

Notice that the conditions $b \geq d$ and $ad^2 > 1$ are equivalent to the fact that the mentioned sets are nonempty.

Proof of Proposition 2.4. The sets U_i are relatively open subsets of the set of matrices with determinant one, are uniformly bounded since $|F|^2 \leq |Fn_0|^2 + 2\lambda^2_{\max}(FP)$, and converge to K since all considered quantities are continuous.

It remains to show that $U_i \subset (U_{i+1})^{\mathrm{rc}}$. To see this, let $F \in U_i$. We first choose d' such that

$$\max\left(|\operatorname{cof} F n_0|, d - 2^{-(i+1)}\right) < d' < d\,,$$

and then b' such that

$$\max\left(\lambda^2_{\max}(FP), b - 2^{-(i+1)}, d'\right) < b' < b\,.$$

This is always possible since $\lambda^2_{\max}(FP) < b$, and $d' < d \le b$.

By Lemma 2.1 there is a laminate with average F supported on matrices F_1, F_2 with unit determinant and such that

$$|F_j n_0| = |F n_0|\,, \quad |\operatorname{cof} F_j n_0| = |\operatorname{cof} F n_0|\,, \quad \lambda^2_{\max}(F_j P) = b'\,, \quad j = 1, 2\,.$$

Consider one of them, say F_j. By Lemma 2.2 there is a laminate with average F_j, supported on F_{j1} and F_{j2}, such that

$$\det F_{jk} = 1\,, \quad |F_{jk} n_0| = |F_j n_0|\,, \quad |\operatorname{cof} F_{jk} n_0| = d'\,,$$

and

$$\lambda^2_{\max}(F_{jk}P) = \max\left(\lambda^2_{\max}(F_j P), \frac{(d')^2}{\lambda^2_{\max}(F_j P)}\right)\,.$$

Since $\lambda^2_{\max}(F_j P) = b' > d'$, the latter reduces to

$$\lambda^2_{\max}(F_{jk}P) = b'\,.$$

Finally, choose a' such that

$$\max\left(|F n_0|^2, a - 2^{-(i+1)}\right) < a' < a\,,$$

and for a given pair (j, k) consider the rank-one direction

$$t \mapsto F_t = F_{jk} + t F_{jk} e_1 \otimes e_3\,.$$

Clearly

$$\det F_t = 1\,,\ \ F_t P = F_{jk} P\,,\ \ \forall t \in \mathbb{R}\,.$$

The function $t \mapsto |F_t n_0|^2$ is continuous, and diverges for $t \to \pm\infty$. Therefore we can find two solutions $t_1 > 0 > t_2$ of the equation

$$|F_t n_0|^2 = a'\,,$$

and we can write F_{jk} as the average of a laminate supported on the matrices F_{jkl}, $l = 1, 2$, such that

$$\det F_{jkl} = 1\,,\ \ |F_{jkl} n_0|^2 = a'\,,\ \ |\operatorname{cof} F_{jkl} n_0|^2 = d'\,,\ \ \lambda^2_{\max}(F_{jkl}P) = b'\,.$$

These conditions imply $F_{jkl} \in U_{i+1} \subset U^{\mathrm{rc}}_{i+1}$ for all $j, k, l \in \{1, 2\}$. Since U^{rc}_{i+1} is closed under lamination, this implies $F_{jk} \in U^{\mathrm{rc}}_{i+1}$ for all $j, k \in \{1, 2\}$; for the same reason $F_j \in U^{\mathrm{rc}}_{i+1}$ and finally $F \in U^{\mathrm{rc}}_{i+1}$. This concludes the proof. $\square$

We now come to the proof of our main results.

Proof of Theorem 1.1. Let

$$\varphi(F) = |Fn_0|^2 + f(\lambda_{\max}(FP)) + \begin{cases} 0 & \text{if } \det F = 1 \text{ and } |\operatorname{cof} Fn_0| \le d_0\,, \\ \infty & \text{else} \end{cases}$$

be the function on the right-hand side of (1.1). The function f defined in (1.2) is nondecreasing and convex; $\lambda_{\max}(FP)$ is a convex function of F, therefore $f(\lambda_{\max}(FP))$ is convex in F. The constraints $\det F = 1$ and $|\operatorname{cof} Fn_0| \le d_0$ are convex in $\det F$ and $\operatorname{cof} F$, therefore φ is polyconvex. Further, if $|Fe_1 \wedge Fe_2| = d_0$ then $|FP|^2 \ge \lambda^2_{\max}(FP) + d_0^2/\lambda^2_{\max}(FP)$, therefore $W_{\mathrm{A},\infty} \ge \varphi$, and $W^{\mathrm{qc}}_{\mathrm{A},\infty} \ge \varphi$.

It remains to construct a laminate to prove the upper bound. It suffices to do so for a generic matrix F such that $\det F = 1$ and $|Fe_1 \wedge Fe_2| < d_0$.

First, we write F as the average of a second-order laminate; then we show that F belongs to an in-approximation of the set $K_{a,b,d}$ as defined in (2.3). This will then permit to obtain a good test function via application of Theorem 2.3.

The laminate is constructed by a direct combination of Lemma 2.1 and Lemma 2.2. Indeed, if $\lambda^2_{\max}(FP) < d_0$ we can, by Lemma 2.1, represent F as average of a laminate supported on matrices with $\lambda^2_{\max}(FP) = d_0$, without changing $|\operatorname{cof} Fn_0|$, $\det F$, and $|Fn_0|$. In a second step, if $|\operatorname{cof} Fn_0| < d_0$ we use Lemma 2.2 to represent each of the remaining matrices as the average of a laminate supported on matrices with $|\operatorname{cof} Fn_0| = d_0$. Evaluating the energy on the final laminate gives the formula in the theorem, and proves that φ is the rank-one convex and polyconvex envelope of $W_{\mathrm{A},\infty}$.

To prove that φ is also the quasiconvex envelope of W^∞, one has to show that the mentioned laminates are limits of functions satisfying the nonlinear constraints $\det Du$ and $|(\operatorname{cof} Du)n_0| = d_0$ pointwise. This can be done using Proposition 2.4. Precisely, for any F such that $\varphi(F) < \infty$ and $|\operatorname{cof} Fn_0| < d_0$, and any $\varepsilon > 0$, let $a = |Fn_0|^2 + \varepsilon$, $d = d_0$, and $b = \max(\lambda^2_{\max}(FP) + \varepsilon, d_0)$. By Proposition 2.4 the sets U_i are an in-approximation of $K_{a,b,d}$; and it is clear that there is i_0 such that $F \in U_i$ for $i \le i_0$. Therefore by Theorem 2.3 there is a function $u \in W^{1,\infty}((0,1)^3; \mathbb{R}^3)$ such that

$$Du \in K \text{ in } (0,1)^3\,, \qquad u(x) = Fx \text{ for } x \in \partial(0,1)^3\,.$$

Setting $v(x) = u(x) - Fx$ in (1.4) gives that for all ψ quasiconvex, $\psi \le W_{\mathrm{A},\infty}$,

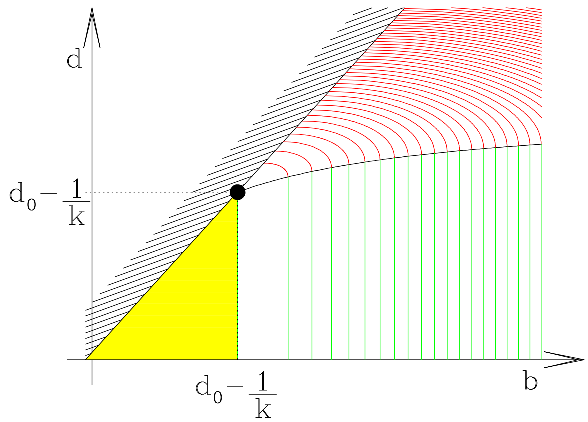

FIGURE 4: Contour plot of the function $f(b,d)$ defined in (1.6). Only the region $d \leq b$ is accessible.

one has

$$\begin{aligned}\psi(F) &\leq \int_{(0,1)^3} \psi(Du)dx \\ &\leq \int_{(0,1)^3} W_{\mathrm{A},\infty}(Du)dx = a + b + \frac{d_0^2}{b} \\ &\leq |Fn_0|^2 + \varepsilon + f(\lambda_{\max}^2(FP) + \varepsilon)\,,\end{aligned}$$

where f was defined in (1.2). Since ε was arbitrary, we conclude that

$$\psi(F) \leq |Fn_0|^2 + f(\lambda_{\max}^2(FP))$$

for all ψ entering (1.3), and therefore for $W_{\mathrm{A},\infty}^{\mathrm{qc}}$. This concludes the proof. $\square$

Proof of Theorem 1.2. We start with the lower bound. The unrelaxed energy can be written as

$$W_{\mathrm{A},k}(F) = \begin{cases} |Fn_0|^2 + g(\lambda_{\max}^2(FP), |\operatorname{cof} F n_0|) & \text{if } \det F = 1\,, \\ \infty & \text{else,} \end{cases}$$

where

$$g(b,d) = b + \frac{d^2}{b} + k(d - d_0)^2\,.$$

The function g is convex, as a function from $(0,\infty)^2$ to $\mathbb{R}$. To see this, we compute

$$\frac{\partial^2 g}{\partial b^2} = 2\frac{d^2}{b^3}\,, \quad \frac{\partial^2 g}{\partial b \partial d} = -2\frac{d}{b^2}\,, \quad \frac{\partial^2 g}{\partial d^2} = 2\frac{1}{b} + 2k\,.$$

The function g is increasing in d if

$$0 \leq \frac{\partial g}{\partial d} = 2\frac{d}{b} + 2k(d - d_0)\,, \qquad \text{i.e.} \qquad d \geq \frac{kd_0}{k + 1/b}\,.$$

Further, g achieves its minimum at the point

$$b = d = d_0 - \frac{1}{k}\,.$$

We conclude that the function f defined in (1.6) is the largest nondecreasing function below g, and it is convex.

Let now

$$\varphi(F) = \begin{cases} |Fn_0|^2 + f(\lambda^2_{\max}(FP), |\operatorname{cof} Fn_0|) & \text{if } \det F = 1\,, \\ \infty & \text{else,} \end{cases}$$

be the function given in the statement. Since $\lambda^2_{\max}(FP)$ and $|\operatorname{cof} Fn_0|$ are polyconvex, it follows that φ is polyconvex. Since $f \leq g$, we have $\varphi \leq W_{\mathrm{A},k}$, and therefore $\varphi \leq W^{\mathrm{qc}}_{\mathrm{A},k}$.

To prove equality, it suffices to perform a construction, analogously to what was done for Theorem 1.1. Since in this case the only constraint is on the determinant, by [10] we know that the function $W^{\mathrm{qc}}_{\mathrm{A},k}$ is convex on rank-one directions. If we express it as a function of the usual variables $|Fn_0|^2$, $b = \lambda^2_{\max}(FP)$ and $d = |\operatorname{cof} Fn_0|$, Lemma 2.1 shows that it is nondecreasing in b, and Lemma 2.2 shows that it is nondecreasing in d. The largest nondecreasing function which is below g is exactly f, as defined in the statement. This concludes the proof.

For completeness we give also an explicit proof along the lines of Theorem 1.1. Let F be a matrix with $\det F = 1$. If $|\operatorname{cof} Fn_0| \geq d_0 k\lambda^2_{\max}(FP)/(k\lambda^2_{\max}(FP)+1)$, then $\varphi(F) = W_{\mathrm{A},k}(F) \geq W^{\mathrm{qc}}_{\mathrm{A},k}(F)$, and there is nothing to prove. Otherwise, let $\varepsilon > 0$,

$$a = |Fn_0|^2 + \varepsilon\,, \;\; b = \max\left(d_0 - \frac{1}{k}, \lambda^2_{\max}(FP)\right)\,, \;\; d = d_0\frac{kb}{kb+1}\,.$$

Clearly $b \geq d$, and $ad^2 > 1$. Therefore we can apply Proposition 2.4. Again, for some $i \in \mathbb{Z}$ we have $F \in U_i$, and by Theorem 2.3 we obtain a solution $u \in W^{1,\infty}((0,1)^3;\mathbb{R}^3)$ of the partial differential inclusion

$$Du \in K \text{ in } (0,1)^3\,, \qquad u(x) = Fx \text{ for } x \in \partial(0,1)^3\,.$$

Setting $v(x) = u(x) - Fx$ in (1.4) gives that for all ψ quasiconvex, $\psi \le W_{\mathrm{A},\infty}$, one has

$$\begin{aligned}\psi(F) &\le \int_{(0,1)^3} \psi(Du)dx \\ &\le \int_{(0,1)^3} W_{\mathrm{A},k}(Du)dx = a + \varepsilon + g(b,d) \\ &\le |Fn_0|^2 + \varepsilon + f(\lambda^2_{\max}(FP), |\operatorname{cof} Fn_0|)\,.\end{aligned}$$

Since ε was arbitrary, the proof is concluded as in the case of Theorem 1.1. □

We now characterize the region where second laminates need to be used.

Lemma 2.5. *If $\lambda^2_{\max}(FP) < d_0 - \frac{1}{k}$ then there is no simple laminate which achieves the optimal energy, i.e., there is no pair F_1, $F_2 \in \mathbb{R}^{3\times 3}$ and $\lambda \in [0,1]$ such that* $\operatorname{rank}(F_1 - F_2) = 1$*, $F = \lambda F_1 + (1-\lambda)F_2$, and $W^{\mathrm{qc}}_{\mathrm{A},k}(F) = \lambda W_{\mathrm{A},k}(F_1) + (1-\lambda)W_{\mathrm{A},k}(F_2)$.*

Proof. We use the notation of the proof of Theorem 1.2. Since $W^{\mathrm{qc}}_{\mathrm{A},k}(F) = |Fn_0|^2 + \min g$, and the first term is convex, the laminate must be supported on the set where g achieves its minimum, i.e., on the set where

$$b = d = d_0 - \frac{1}{k}\,.$$

If there were such a single laminate, there would be matrices Q, $R \in O(2,3)$ such that

$$FP \in \left(d_0 - \frac{1}{k}\right)^{1/2} [Q, R]\,, \qquad \operatorname{rank}(Q - R) = 1\,.$$

Here $[Q, R]$ denotes the convex hull of the set $\{Q, R\}$, i.e., the segment with endpoints Q and R, and $O(2,3) = \{F \in \mathbb{R}^{3\times 2} : F^T F = \mathrm{Id}\}$. But the last condition implies that

$$\lambda_{\max}(G) = 1\,, \qquad \text{for all } G \in [Q, R]\,.$$

This implies $\lambda^2_{\max}(FP) = d_0 - 1/k$, contradicting the assumption. □

Finally, we consider the case where the constraint on $|\operatorname{cof} Fn_0|$ is replaced by the simpler one on $|Fn_0|$.

Proposition 2.6. *The quasiconvex envelope of*

$$W_{\mathrm{B},\infty}(F) = \begin{cases} |F|^2 & \text{if } \det F = 1 \text{ and } |Fn_0| = d_0\,, \\ \infty & \text{else,} \end{cases}$$

is given by

$$W^{\mathrm{qc}}_{\mathrm{B},\infty}(F) = \begin{cases} |FP|^2 + d_0^2 & \text{if } \det F = 1 \text{ and } |Fn_0| \leq d_0\,, \\ \infty & \text{else.} \end{cases}$$

Proof of Proposition 2.6. Consider the matrices $F_s = F + sFe_1 \otimes e_3$, and choose $s_\pm$ as the two solutions to

$$|F_s e_3| = |Fe_3 + sFe_1| = d_0\,.$$

A laminate between F_{s_+} and F_{s_-} shows that the given expression is an upper bound on the relaxation. The rest of the proof follows the same steps as the proof of Theorem 1.1. ☐

Proposition 2.7. *The quasiconvex envelope of*

$$W_{\mathrm{B},k}(F) = \begin{cases} |F|^2 + k(|Fn_0| - d_0)^2 & \text{if } \det F = 1\,, \\ \infty & \text{else,} \end{cases}$$

is given by

$$W^{\mathrm{qc}}_{\mathrm{B},k}(F) = \begin{cases} |FP|^2 + f(|Fn_0|) & \text{if } \det F = 1\,, \\ \infty & \text{else,} \end{cases}$$

where

$$f(d) = \begin{cases} d^2 + k(d - d_0)^2 & \text{if } d \geq \dfrac{k}{k+1} d_0\,, \\ d_0^2 \dfrac{k}{k+1} & \text{else.} \end{cases}$$

Proof. The proof can be done with the arguments used to prove Proposition 2.6, choosing values $s_\pm$ so that the function

$$|F_s e_3|^2 + k(|F_s e_3| - d_0)^2$$

is minimized. Precisely, consider the function $h : [|Fe_3|, \infty) \to \mathbb{R}$ defined by

$$t \mapsto h(t) = t^2 + k(t - d_0)^2\,.$$

Let t_0 be the point where h achieves its minimum. Let $s_\pm$ be the two solutions (possibly both zero) of the quadratic equation

$$|F_s e_3|^2 = t_0^2\,,$$

which is soluble since $t_0 \geq |Fe_3|$. If $s_\pm$ are not both zero, they have different sign, and the upper bound is obtained considering a laminate supported on the two matrices F_{s_-} and F_{s_+}. ☐

3 Physical implications

We now consider the energy $W_{\mathrm{A},k}$, which is the one relevant for smectic A elastomers, and make predictions for the macroscopic material behavior. The parameters in experiments are $d_0 \sim 1$, and $k \gg 1$ (see Figure 2 and Figure 3), therefore we focus on the asymptotic behavior for large k.

The global minimum of $W_{\mathrm{A},k}(F)$ is achieved by

$$F_0 = \begin{pmatrix} \lambda_0 & 0 & 0 \\ 0 & \lambda_0 & 0 \\ 0 & 0 & 1/\lambda_0^2 \end{pmatrix}$$

where λ_0 is the minimizer of

$$2\lambda^2 + \lambda^{-4} + k(\lambda^2 - d_0)^2 \,.$$

An explicit computation shows that

$$\lambda_0^2 = d_0 + \frac{1}{k}\left(\frac{1}{d_0^3} - 1\right) + O(k^{-2})\,.$$

The minimum of $W_{\mathrm{A},k}(F)$ is not located at $F = \mathrm{Id}$ since we chose an ideal reference configuration, which is not the initial configuration in experiments, see (A.1) below. It does also not correspond to the minimum of the function g used in the proof of Theorem 1.2, since we are here minimizing the full energy, which includes the term $|Fn_0|^2$, subject to the constraint on the determinant.

We now consider a stretching experiment along $n_0 = e_3$. This means, we start from the global minimum F_0, impose a uniaxial stretch λ in the 33 direction, and assume that the macroscopic deformation gradient remains diagonal,

$$F(\lambda) = \begin{pmatrix} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda/\lambda_0^2 \end{pmatrix}.$$

This is the geometry used in [30, 8]. The corresponding energy is

$$E(\lambda) = \min_{\lambda_1,\lambda_2} W^{\mathrm{qc}}_{\mathrm{A},k}(F(\lambda))\,.$$

The minimization can be carried out explicitly, and leads to

$$E(\lambda) = \begin{cases} \dfrac{\lambda^2}{\lambda_0^4} + 2\dfrac{\lambda_0^2}{\lambda} + k\left(\dfrac{\lambda_0^2}{\lambda} - d_0\right)^2 & \text{if } \lambda \le \lambda_{\mathrm{crit}}\,, \\ \dfrac{\lambda^2}{\lambda_0^4} + 2d_0 - \dfrac{1}{k} & \text{else.} \end{cases}$$

Here $\lambda_{\text{crit}} = \lambda_0^2/(d_0 - 1/k) = 1 + 1/(kd_0^4) + O(k^{-2})$.

It is also interesting to analyze the values of $\lambda_{1,2}$ obtained by minimization. If $d = \lambda_0^2/\lambda \geq d_0 - 1/k$ then the minimum is attained by $\lambda_1 = \lambda_2 = \lambda_0/\lambda^{1/2}$. If instead $d = \lambda_0^2/\lambda < d_0 - 1/k$ then the energy-minimizing state is not uniquely determined. There are many minima which correspond to second laminates, as, e.g., $\lambda_1 = \lambda_2 = \lambda_0/\lambda^{1/2}$. However there is only one which corresponds to a single laminate, namely, the state $\lambda_1^2 = d_0 - 1/k$, $\lambda_2 = \lambda_0^2/\lambda\lambda_1$ (see Lemma 2.5). Within the model considered here all these states have the same energy, but they will be distinguished by higher-order terms. In particular, since interfaces are expected to be penalized, one can expect that the state with first laminates will be preferred to the ones with second laminates.

Computing the derivative of $E(\lambda)$ with respect to λ we obtain the stress,

$$\sigma(\lambda) = \begin{cases} 2\dfrac{\lambda}{\lambda_0^4} - 2\dfrac{\lambda_0^2}{\lambda^2} - 2k\left(\dfrac{\lambda_0^2}{\lambda} - d_0\right)\dfrac{\lambda_0^2}{\lambda^2} & \text{if } \lambda \leq \lambda_{\text{crit}}\,, \\ 2\dfrac{\lambda}{\lambda_0^4} & \text{else.} \end{cases}$$

One can check that the stress is continuous at λ_{crit}. This prediction is compared with experimental results from [30] in Figure 2, and with experimental results from [8] in Figure 3.

Physically, the transition between a "hard" regime at small deformation and a "soft" regime at large deformation can be understood as due to the onset of microstructure formation, much as in the related case of nematic elastomers [35]. While there are many microstructures leading to the optimal energy, the simplest one is a laminate, and we expect it to be favored over the others if interfacial energies are included in the model. Indeed, a break-up of the Bragg reflection corresponding to the layer spacing has been observed in X-ray experiments after the threshold deformation λ_{crit}, confirming the appearance of microstructure [30]. Even more, experiment has shown a decrease in the X-ray intensity [30]. As discussed in [2], this decrease is consistent with the appearance of a more complex microstructure, with many layer normals distributed symmetrically around the direction of the applied force: For any given λ, the layer normals would be uniformly distributed on a ring, and only two such points would meet the Bragg condition. We recall, however, that full cylindrical symmetry is appropriate in the present experimental geometry only if the microstructure length–scale is much smaller than the film thickness. Hence, this specific X–ray observation gives some indirect information on the microstructure size. Another experimental confirmation of the emergence of complex microstructure, with layer normals distributed with isotropic symmetry, is the observed isotropic Poisson ratio of 1/2. Both

Poisson's ratio and the distribution of layer normals are expected to change in the case of very thin films (i.e., thinner than the expected microstructure size).

A Appendix

We briefly discuss here the relation between $W_{\mathrm{A},k}$ and the energy density W_{AW} derived by Adams and Warner. In [2] they obtain the expression

$$\widetilde{W}_{\mathrm{AW}}(\Lambda) = \begin{cases} \frac{1}{2}\mu \left[\mathrm{Tr}(\Lambda U_0^2 \Lambda^T U_n^{-2}) - 3\right] \\ \qquad\qquad + \frac{1}{2} B \left(\frac{1}{|\Lambda^{-T} n_0|} - 1 \right)^2 & \text{if } \det \Lambda = 1\,, \\ +\infty & \text{else,} \end{cases}$$

where $\Lambda \in \mathbb{R}^{3\times 3}$ is the deformation gradient with respect to the cross-linking configuration, n_0 is the unit normal to the smectic layers at cross-linking, which for the present case (smectic A) coincides with the smectic director at cross-linking, and the smectic director n is a unit vector parallel to $\Lambda^{-T} n_0$. Further,

$$U_n = r^{-1/6} \left[\mathrm{Id} + (r^{1/2} - 1) n \otimes n\right]$$

is a uniaxial stretch along n, and $U_0 = U_{n_0}$, $r > 1$ and $B \gg \mu$ are material parameters. The expressions in [2] are written in terms of $\ell_n = U_n^2$.

As in the case of nematic [17, 18, 19, 13] and smectic C [1] elastomers, it is convenient to change reference configuration, in order to better exploit the symmetry of the problem. In particular, we replace the variable Λ with F, defined by

$$F = \Lambda U_0 = r^{-1/6} \Lambda \left[\mathrm{Id} + (r^{1/2} - 1) n_0 \otimes n_0\right]\,, \tag{A.1}$$

and compute

$$\Lambda^{-T} n_0 = (F U_0^{-1})^{-T} n_0 = F^{-T} U_0 n_0 = r^{1/3} F^{-T} n_0\,,$$

$$n = \frac{\Lambda^{-T} n_0}{|\Lambda^{-T} n_0|} = \frac{F^{-T} n_0}{|F^{-T} n_0|}\,,$$

and

$$|F^T n| = \frac{|F^T F^{-T} n_0|}{|F^{-T} n_0|} = \frac{1}{|F^{-T} n_0|}\,.$$

Since

$$\begin{aligned} \mathrm{Tr}(\Lambda U_0^2 \Lambda^T U_n^{-2}) &= \mathrm{Tr}(F F^T U_n^{-2}) = r^{1/3}\, \mathrm{Tr}\left(F F^T \left[\mathrm{Id} + (r^{-1} - 1) n \otimes n\right]\right) \\ &= r^{1/3} |F|^2 + r^{1/3} (r^{-1} - 1) |F^T n|^2\,, \end{aligned}$$

we obtain

$$
\begin{aligned}
W'_{\rm AW}(F) &= \widetilde{W}_{\rm AW}(FU_0^{-1}) \\
&= \begin{cases} \frac{1}{2}\mu\left[r^{1/3}|F|^2 + r^{1/3}\left(\frac{1}{r}-1\right)\frac{1}{|F^{-T}n|^2} - 3\right] & \\ \qquad\qquad + \frac{1}{2}B\left(\frac{1}{r^{1/3}|F^{-T}n_0|}-1\right)^2 & \text{if } \det F = 1\,, \\ \infty & \text{else}\,. \end{cases}
\end{aligned}
$$

Rearranging terms we obtain

$$
W'_{\rm AW}(F) = \frac{\mu r^{1/3}}{2}\begin{cases} |F|^2 + kd_0^2\left(\frac{d_0}{|\operatorname{cof} Fn_0|}-1\right)^2 + c & \text{if } \det F = 1\,, \\ \infty & \text{else,} \end{cases}
$$

where

$$
d_0 = \frac{1}{r^{1/3}}\left[1 + \frac{\mu r}{B}\left(\frac{1}{r}-1\right)\right]\,, \qquad k = \frac{1}{d_0^3}\frac{B}{\mu r^{2/3}}\,,
$$

and c is a suitable constant (depending on r and B/μ). After eliminating the irrelevant additive and multiplicative constants c and $\mu r^{1/3}/2$, this coincides with (1.7).

The two energies $W_{\rm AW}$ and $W_{{\rm A},k}$ are equivalent to leading order in $|\operatorname{cof} Fn_0| - d_0$, which is expected to be the relevant regime since in experiments $k \gg 1$. However, the energy $W_{\rm AW}$ has an additional instability at large values of $|\operatorname{cof} Fn_0|$. To see this, consider for example the family of deformation gradients

$$
F_t = \begin{pmatrix} 2 & 0 & 0 \\ 0 & 0.5 & 0 \\ 0 & t & 1 \end{pmatrix}\,.
$$

The energy is (for simplicity we work for $d_0 = 1$ and $n_0 = e_3$)

$$
W_{\rm AW}(F_t) = 5 + \frac{1}{4} + t^2 + k\left(\frac{1}{\sqrt{1+4t^2}}-1\right)^2\,. \tag{A.2}
$$

This function is concave at intermediate values of t (see Figure 5). Consider for definiteness the deformation gradient $F_{0.5}$ for $k = 100$. Then $F_{0.5}$ is unstable against formation of laminates supported on the matrices $F_{t_{1,2}}$, where $t_1 \sim 0.28$, $t_2 \sim 9.09$. Physically, this corresponds to the fact that a uniform moderate shear of the layers is unstable against a laminate in which about 3% of the layers are very strongly sheared, and most layers are only very weakly sheared. The corresponding energy diagram along the considered rank-one line is illustrated in Figure 5.

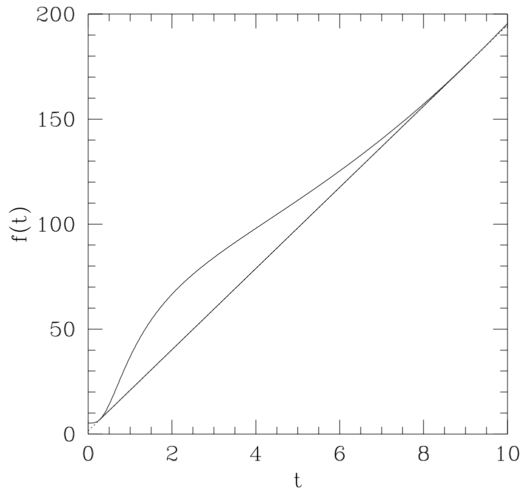

FIGURE 5: Plot of the function $f(t) = W_{\mathrm{AW}}(F_t)$ defined in (A.2) and of its convex hull, for $k = 100$.

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