Interfacial Energy and Fine Defect Structures
for Incoherent Films

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This note summarizes recent results [1] in which modern techniques of the calculus of variations are used to obtain qualitative features of film-substrate interfaces for a broad class of interfacial energies. In particular, we show that the existence of a critical thickness for incoherence and the formation of interfacial dislocations depend strongly on the convexity and smoothness of the interfacial energy function.

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I. INTRODUCTION

The structure of an interface separating crystalline solids is strongly affected by the competition between the relaxation of the bulk crystals to their respective equilibrium configurations, and the tendency of the interface to maintain the exact matching (coherency) of the atoms of the two solids across the interface (cf. Leo & Hu [2,3] and the review paper [4]). When the stresses due to the deviation from equilibrium of the bulk material reach a certain threshold, interfacial dislocations appear that relax the bulk stresses and an extreme situation may be reached in which all regularity of the atomic bonding at the interface is lost.

We restrict attention to a two-dimensional framework with corresponding cartesian coordinates \((x, y)\) and basis \((i, j)\). We take the layer to be infinite in the \(x\)-direction, denote by \(h\) its height in the \(y\)-direction, and assume that \(y = 0\) represents the interface between the layer and the substrate, which we assume to be rigid. Letting \(u(x, y)\) denote the displacement of the layer, \(\epsilon = \frac{1}{2}(\nabla u + \nabla u^\top)\) the infinitesimal strain, and

\[
e_0 = e_0 i \otimes i,
\]

with \(e_0 > 0\), the mismatch strain, we consider the layer as elastic with energy density

\[
w(\epsilon - e_0) = \frac{E}{2(1 + \nu)} \left\{ \frac{\nu}{1 - 2\nu} [\text{tr}(\epsilon - e_0)]^2 + \text{tr}((\epsilon - e_0)^2) \right\},
\]

with \(E\) and \(\nu\) Young’s modulus and Poisson ratio, such that \(E > 0\) and \(-1 < \nu < \frac{1}{2}\).

We consider displacement fields \(u(x, y)\) that are periodic in \(x\), \(u(1, y) = u(0, y) + (\text{const.})i\) for \(y \in [0, h]\); therefore, modulo a rescaling, we may restrict attention to a cell of unit length \(\Omega = [0, 1] \times [0, h]\). We assume that the layer cannot separate from the substrate, so that \(u(x, 0) - j = 0\) for \(x \in [0, 1]\). Thus, letting \(u(x) = u(x, 0) - i\) denote the tangential displacement of the layer at the interface, we define the incoherence strain \(\gamma(x)\) by

\[
\gamma(x) = \frac{du(x)}{dx},
\]

and refer to the interface as coherent if

\[
\gamma(x) = 0 \quad x \in [0, 1],
\]

and incoherent otherwise.

We assume that the interfacial energy is an even, continuous function \(f(\gamma)\) of the incoherence strain, and that \(f(0) = 0\), while \(f(\gamma) > 0\) for \(\gamma \neq 0\). The total energy of the system is then given by

\[
J(u) = \int_{\Omega} w(\epsilon - e_0) \, dx \, dy + \int_0^1 f(\gamma) \, dx. \quad (1.1)
\]

II. CONVEXITY OF THE INTERFACIAL ENERGY DENSITY AND EXISTENCE OF SMOOTH EQUILIBRIUM STATES

Equilibrium configurations of the layer correspond to states which minimize the total energy: classically, such configurations correspond to displacement fields \(\Pi(x, y)\) such that

\[
J(\Pi) = \min_{u \in W} J(u)
\]

over a suitable space \(W\) of displacement fields on \(\Omega\).

1Assuming that \(f\) satisfies the growth condition \(f(\gamma) \leq C(1 + |\gamma|^q)\), with \(C > 0\) and \(q \geq 1\), we minimize \(J\) on the space \(W\) of all functions \(u\) in the Sobolev space \(W^{1,2}(\Omega, \mathbb{R}^2)\), whose restriction to \(\partial \Omega\) belong to \(W^{1,q}(\partial \Omega, \mathbb{R}^2)\), which satisfy the periodicity condition and are tangential to the interface.
The chief problem here is that when \( f \) is not convex such minimizers \( u \) may not exist. Even so, one can obtain valuable physical insight by studying minimizing sequences: that is, sequences \( \{ u_n \} \) that tend to the infimum of the functional \( J(u) \) in the sense that
\[
J(u_n) \to \inf_{u \in W} J(u),
\]
as \( n \to \infty \). In fact, as we shall see, such minimizing sequences help to characterize the microstructures resulting from various choices of the interfacial energy \( f(\gamma) \).

Central to this method of analysis is the convex envelope \( f^{**}(\gamma) \) of \( f(\gamma) \), which is the largest convex energy \( f^{**}(\gamma) \) with \( f^{**}(\gamma) \leq f(\gamma) \) for all \( \gamma \); precisely, \( f^{**} \) is the supremum of all convex functions \( g \) such that \( g \leq f \). Then \( f \) is convex if and only if \( f = f^{**} \) (Fig. 1).

![FIG. 1. The interfacial energy density (thick line) and its convex envelope (thin line).](image)

The convexity of \( f^{**} \) allows us to obtain a unique solution \( u^{**} \) of the "regularized problem"
\[
J(u^{**}) = \inf_{u \in W} \left\{ \int_{\Omega} w(\epsilon - \epsilon_0) \, dx \, dy + \int_0^1 f^{**}(\gamma) \, dx \right\}.
\]
In fact, this solution has the explicit form
\[
u^{**}(x, y) := \nu^{**} x i + \frac{\nu(\epsilon_0 - \nu^{**})}{(1 - \nu)} y j,
\]
and corresponds to the constant value \( \nu^{**} \) of the incoherence strain \( \nu \) defined in Section IV below.

Consider the case in which \( f \) is not convex at \( \nu^{**} \). Here (cf. [4]), any minimizing sequence must satisfy \[ u_n \to u^{**} \text{ in } \Omega, \text{ and } \int_0^1 \gamma_n \, dx \to \gamma^{**}, \]
as \( n \to \infty \).

Thus minimizing sequences always converge in bulk to the homogeneous deformation (2.1). As we shall see, minimizing sequences may not have a classical limit at the interface, but the "generalized limit" [4] however it be visualized, corresponds to a well defined average incoherence strain \( \gamma^{**} \).

Of course, \( u^{**} \) is a candidate minimizer of the total energy, but since
\[
J(u^{**}) = \frac{Eh}{2(1 - \nu^2)} (\nu^{**} - \epsilon_0)^2 + f(\gamma^{**}),
\]
then
\[
(a) \ f(\gamma^{**}) = f^{**}(\gamma^{**}) \iff J(u^{**}) = \inf_{u \in W} J(u),
\]
\[
(b) \ f(\gamma^{**}) > f^{**}(\gamma^{**}) \iff J(u^{**}) > \inf_{u \in W} J(u).
\]
Hence only when \( f \) is convex at \( \gamma^{**} \), does a minimum exist for the energy functional, at least in the classical sense.

III. MINIMIZING SEQUENCES AND INTERFACIAL MICROSTRUCTURES

Fix a thickness of the layer \( h > 0 \), let \( \gamma^{**} \) be the average incoherence strain corresponding to the infimum of \( J \), and assume that \( \gamma^{**} \neq 0 \) and \( f(\gamma^{**}) > f^{**}(\gamma^{**}) \), so that no smooth equilibrium state exists. In this case, physically meaningful results may be still be obtained by inspection of the minimizing sequences \( \{ u_n \} \).

By (2.2) we may indeed restrict attention to the corresponding sequences of incoherence strains \( \gamma_n \) at the interface, which must, in turn, be minimizing sequences of the interfacial energy (cf. [4]):
\[
\lim_{n \to \infty} \int_0^1 f(\gamma_n(x)) \, dx = f^{**}(\gamma^{**}),
\]
with \( f^{**}(\gamma^{**}) < f(\gamma^{**}) \). We shall consider two cases.

• Oscillating sequences (Fig. 2). Consider the situation in Figure 2(a), in which there is a \( \lambda, 0 < \lambda < 1 \), such that
\[
\gamma^{**} = \lambda \gamma_a + (1 - \lambda) \gamma_b, \quad f^{**}(\gamma^{**}) = \lambda f(\gamma_a) + (1 - \lambda) f(\gamma_b).
\]

![FIG. 2. (a) The interfacial energy density and its convex envelope; (b) a typical element of a minimizing sequence \( \gamma_n \).](image)

Then (3.1) may be satisfied by a minimizing sequence \( \gamma_n \) oscillating between \( \gamma_a \) and \( \gamma_b \) on two subsets mixing finely as \( n \to \infty \). More precisely, we may take
\[
\gamma_n(x) = \begin{cases} 
\gamma_a & x \in \left[ \frac{k - \lambda}{h} m, \frac{k - \lambda - 1}{h} m \right), \\
\gamma_b & x \in \left[ \frac{k + \lambda}{h} m, \frac{k + \lambda - 1}{h} m \right), 
\end{cases}
\]

2 Convergence of \( u_n \) is here in the sense of \( W^{1,2}(\Omega, \mathbb{R}^2) \).
3 The generalized or weak limit of a sequence can be thought of as the limit of its "local integral averages".
with \( k = 0, \ldots, n - 1 \) (cf. Figure 2(b)). Thus, since for any \( n \)
\[
\int_0^1 f(\gamma_n) \, dx = \lambda f(\gamma_a) + (1 - \lambda) f(\gamma_b) = f^{**}(\gamma^{**}),
\]
then \( (3.3) \) is satisfied. Indeed, the \( \gamma_n \) are minimizers of the interfacial energy functional for fixed \( \gamma^{**} \), but their weak limit \( \gamma(x) \equiv \gamma^{**} \) is not.

Thus, in the limit as \( n \to \infty \), the above sequence describes an interfacial microstructure whose corresponding incoherence strain takes the values \( \gamma_a \) and \( \gamma_b \) on infinitesimal patches of length fractions \( \lambda \) and \( 1 - \lambda \) respectively.

- **Concentrating sequences (Fig. 3).** Condition \( (3.3) \) may not be satisfied in the important case in which \( f \) is strictly concave (see also Fig. 6). We have

\[
f^{**}(\gamma) = m|\gamma|, \quad \text{with} \quad m = \lim_{\gamma \to \infty} \frac{f(\gamma)}{\gamma}.
\]

![Fig. 3](image)

(a) The interfacial energy density and its convex envelope; (b) a typical element of a minimizing sequence \( \gamma_n \).

Fix \( h > 0 \) as above and assume that \( \gamma^{**} > 0 \). Then \( (3.3) \) can only be satisfied by sequences \( \gamma_n \) which become unbounded on smaller and smaller sets, for instance

\[
\gamma_n(x) = \begin{cases} 
\gamma^{**}n & x \in [\frac{k}{n}, \frac{k + 1}{n}), \\
0 & x \in [\frac{k}{n} + \frac{1}{n^2}, \frac{k + 1}{n}],
\end{cases} \tag{3.4}
\]

for \( k = 0, \ldots, n - 1 \). In this case, \( (3.3) \) is satisfied since

\[
\int_0^1 f(\gamma_n(x)) \, dx = \frac{f(\gamma^{**}n)}{n} \to m\gamma^{**} = f^{**}(\gamma^{**}).
\]

Note that, since the \( \gamma_n \) tend to grow without bound on small sets, the corresponding displacements \( u_n \) at the interface tend to develop microscopic jumps; these may be identified with interfacial dislocations.

IV. CRITICAL THICKNESS AND SMOOTHNESS OF THE CONVEXIFIED INTERFACIAL ENERGY DENSITY

We now study the behavior of the minimizers when \( h \) varies. We show that, when \( f^{**} \) is smooth at zero, the interface is incoherent (i.e., \( \gamma^{**} \neq 0 \)) for any \( h > 0 \), while if \( f^{**} \) is non-smooth at zero, there exists a critical thickness \( h_c \) such that for \( h < h_c \) the interface is coherent (so that \( \gamma^{**} = 0 \)).

Consider first a smooth \( f^{**} \). Let \( j(\gamma) \) denote the right side of \( (2.3) \) when \( \gamma^{**} \) is replaced by \( \gamma \). Then \( j \) is smooth and convex and \( \gamma^{**} \) is the unique solution of \( j'(\gamma) = 0 \). Since a direct calculation shows that \( j'(0) \) can only vanish when \( h = 0 \) then, for \( h > 0 \), \( \gamma^{**} \neq 0 \) and the interface is incoherent.

Assume now that \( f^{**} \), and thus \( j \), is non-smooth at zero. Then the condition \( j'(0) = 0 \) must be replaced by \( 0 \in [j'_-(0), j'_+(0)] \), where \( j'_\pm \) are the left and right derivatives of \( j \). This condition is in general satisfied by an interval of values for the thickness (cf. \( [0] \)), and thus the equilibrium interface is coherent for all \( h \leq h_c \), with \( h_c \) a suitable critical thickness.

In general, the convexified interfacial energy \( f^{**} \) may be non-smooth at other values \( \gamma \) of the incoherence strain; if this is the case, we assume that \( f^{**}(\gamma) = f(\gamma) \), so that a regular solution exists to the minimization problem. Proceeding as above, we see that the condition which assures that \( \gamma^{**} = \gamma \) is that \( 0 \in [j'_-(\gamma), j'_+(\gamma)] \), and this condition again determines an interval for the thickness for which the incoherence strain remains fixed at the value \( \gamma(x) \equiv \gamma^{**} = \gamma \), the film remains ‘glued’ to the substrate.

V. DISCUSSION

We now turn to the analysis of specific forms of the interfacial energy density \( f \) and discuss the existence of smooth equilibrium configurations for the film, the formation of microstructures at the interface, and the critical thickness for incoherence. Remark that the height of the film \( h \) is now allowed to vary, so that the average incoherence strain \( \gamma^{**} \) also varies accordingly.

- \( f \) convex and smooth (Fig. 4(a)). Since in this case \( f(\gamma) = f^{**}(\gamma) \), a regular solution exists and is given by \( u^{**} \) in \( (2.1) \). In fact, the restriction of \( u^{**} \) to the interface, namely \( u(x) = \gamma^{**}x \), is a smooth minimizer of the interfacial energy functional.

![Fig. 4](image)

(a) Interfacial energy density: (a) smooth and convex; (b) non-smooth but convex.

Thus the film is uniformly incoherently strained with respect to the substrate, but no fine structure appears. Moreover, since \( f^{**}(\gamma) \) is smooth, the interface relaxes to incoherence for any thickness of the layer.
• \( f \) convex but non-smooth at \( \gamma = 0 \) (Fig. 4(b)). Again \( f(\gamma) = f^{**}(\gamma) \), so that the homogeneous deformation \( u^{**} \) in (2.3) is a minimizer of \( J \), and no fine structure develops at the interface.

Now, since \( f \) is non-differentiable at \( \gamma = 0 \), there exists a critical thickness for the transition to incoherency such that for \( h \leq h_{c} \) the interface is coherent, while for \( h > h_{c} \) the interface is uniformly strained with respect to the substrate.

This form of interfacial energy might be appropriate to describe “glassy” interfaces between crystals with large mismatch.

• \( f \) nonconvex and nonsmooth (Fig. 5). We assume here that \( f \) has pointed minima at values \( \gamma_{i} \in \{ \gamma_{0} = 0 , \pm \gamma_{1} , \pm \gamma_{2} , ... \} \) of the incoherency strain, so that the convexified interfacial energy \( f^{**} \) is piecewise linear in the intervals \((0, \gamma_{1}),(\gamma_{1}, \gamma_{2}),...\) but non smooth at \( \gamma = \gamma_{i} \).

(i) Existence: since \( f(\gamma) = f^{**}(\gamma) \) only for \( \gamma = \gamma_{i} \), the homogeneous deformation \( u^{**} \) in (2.3) is a minimizer only when \( \gamma^{**} = \gamma_{i} \). When \( \gamma^{**} = 0 \) the interface is coherent, while for \( \gamma^{**} = \pm \gamma_{1}, \pm \gamma_{2}, ... \) the film is uniformly strained with respect to the substrate.

(ii) Critical thickness: since \( f^{**} \) is non-smooth at \( \gamma_{i} \), the requirement that \( \gamma^{**} = \gamma_{j} \) has the form \( 0 \in [j^{-}_{\gamma_{i}}(\gamma_{j}) , j^{+}_{\gamma_{i}}(\gamma_{j})] \). Thus, there exists a whole interval for \( h \) for which this condition is satisfied. More precisely, there exist critical intervals \([0 , h_{0}] , ... , [h_{i} , h_{i+1}] \) such that if \( h \in [h_{i} , h_{i+1}] \), then \( \gamma^{**} = \gamma_{i} \). This means that the interface remains “glued” to the substrate at this fixed incoherency strain for all values of the thickness in the critical interval.

(iii) Microstructures: for all other values of \( h \) we have \( \gamma^{**} \neq \gamma_{j} \), and (2.3) is not a minimizer of \( J \). But since condition (2.3) holds, the minimizing sequences have an oscillating character, and are finer and finer mixtures of patches on which \( \gamma \) takes the values \( \gamma_{a} = 0 \) and \( \gamma_{b} = \gamma_{1} \) (if \( \gamma^{**} \in (0, \gamma_{1}) \) ), or \( \gamma_{a} = \gamma_{1} \) and \( \gamma_{b} = \gamma_{2} \) (if \( \gamma^{**} \in (\gamma_{1}, \gamma_{2}) \) ), and so on.

This behavior is reminiscent of “coincidence boundaries”, which are special incoherent interfaces between crystals with a large difference in lattice parameters.

• \( f \) concave and non-smooth at \( \gamma = 0 \) (Fig. 6). We have seen that in this case

\[
\gamma^{**} = 0 \text{, a coherent interface. For values of } h \text{ at which } \gamma^{**} \neq 0 \text{ coherency is lost and fine microstructures appear at the interface.}
\]

More precisely, observe first that \( f^{**} \) is non-smooth at zero, so that there exists a critical thickness \( h_{c} \) for incoherency. When \( h > h_{c} \), since \( \gamma^{**} \neq 0 \), the displacement field \( u^{**} \) in (2.3) is no longer a solution; the minimizing sequences are concentrating, and energy is minimized by allowing the incoherency strain to become infinitely large on infinitesimal intervals: accordingly, the interfacial displacement \( u_{\gamma} \) tends to develop microscopic jumps, which we may interpret as interfacial dislocations.

These energies might describe small-misfit epitaxial layers.

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