STRESS-INDUCED ROUGHENING INSTABILITIES ALONG SURFACES OF PIEZOELECTRIC MATERIALS

Nelly Yung Chien and Huajian Gao
Division of Applied Mechanics, Stanford University, Stanford, CA 94305-4040

ABSTRACT

The possibility of using electric field to stabilize surfaces of piezoelectric solids against stress-induced morphological roughening is explored in this paper. Two types of idealized boundary conditions are considered: 1) a traction free and electrically insulated surface and 2) a traction free and electrically conducting surface. A perturbation solution for the energy variation associated with surface roughening suggests that the electric field can be used to suppress the roughening instability to various degrees. A completely stable state is possible in the insulating case, and kinetically more stable states can be attained in the conducting case. The stabilization has importance in reducing concentration of stress and electric fields due to microscopic surface roughness which might trigger failure processes involving dislocation, cracks and dielectric breakdown.

INTRODUCTION

This paper is concerned with a class of stress-induced instabilities which causes material surfaces to roughen under diffusional mass transport. These instabilities occur, for example, in heteroepitaxial thin films where the elastic strain energy due to lattice misfit provides a thermodynamic driving force for the onset of the island-like surface morphology, also known as the Stranski-Krastonov pattern (e.g., [1]), during film growth or annealing. The conclusion that the strain energy tends to destabilize an initially flat surface and thus to promote the development of surface roughness has been reached by studying linearized kinetic equations along a slightly wavy surface [2] [3] and by showing that the strain energy is always reduced when an initially flat surface is slightly perturbed in an arbitrary manner [4]. Gao [5] [4] also studied the stress concentration along a slightly undulating surface of a stressed isotropic or anisotropic solid and found that even a slightly undulating surface can generate significant stress concentration causing deformation and fracture. It appears that a thorough investigation of the surface roughening instability and the resulting stress concentration is of importance for understanding the nucleation of misfit dislocations in heteroepitaxial thin films [6] [7] and general flaw initiation at material surfaces exposed to environmental corrosions. The significance of microscopic surface roughness is further elucidated by the recent work of Chiu and Gao [8] who adopted a cycloid surface to model periodic rough surfaces. Chiu and Gao found that the cusped cycloid surface generates a crack-like stress singularity within a thin surface layer. Under uniform tension this singularity shows identical strength (i.e. stress intensity factor) as a row of periodic parallel cracks. Even though a rough surface with cusps may not have been perceived to be as dangerous as a periodically cracked body, application of fracture mechanics predicts that the two structures should fail at the same stress level.

The essence of the surface roughening instability leading to formation of stress singularities lies in the competition between elastic energy and surface energy in a stressed material system. The elastic energy can be most efficiently released by formation of localized defects such as cusps, cracks and dislocations. However, such strain relaxation must occur at the cost of creating additional free surfaces, thus increasing the surface energy of the system. One may show that the elastic energy will dominate over surface energy at relatively long wavelengths (However, at long wavelengths, the gravitational energy may also interfere in the instability process; see [4]). A question that arises is whether other forms of energy such as those of electromagnetic origin can sometimes be utilized to interfere in the instability process so as to control the development of defects. As a first study in that direction, we explore in this paper the possibility of using electric field as a control parameter to minimize or even stabilize the stress-induced surface instabilities in piezoelectric materials.

The subject here is of physical significance. Piezoelectric materials have been widely used as electromechanical transducer, such as ultrasonic generators, filters, sensors, and actuators. Thin films technology plays an important role in many industrial processes and in the fabrication of solid state components. Recent developments involving piezoelectric thin films include applications such as force-sensing resistors [9], built-in vibration sensors [10], molecular sensing devices [11], and surface acoustic waves (SAW) generators [12]. As piezoelectric thin films may be subjected to very large stresses generated by strain sources such as thermal mismatch and lattice mismatch. Stress concentration effects due to an undulating surface may trigger processes involving nucleation of dislocations and cracks. Electric field concentrations might also occur causing dielectric breakdown. Thus, it is of interest to investigate the possibility of controlling surface instabilities with an applied electric field. To achieve our objectives, a perturbation analysis developed in Gao [4] for anisotropic elastic solids is extended to the piezoelectric case. The perturbation solution of energy variations is then used to examine the effects of mechanical and electric loading on the instability wavelength.

Conducting and Insulated Surfaces of a Stressed Piezoelectric Medium

Details concerning the theory of piezoelectricity can be found, for example, in [13] and [14]. The constitutive equations of a piezoelectric medium is usually expressed as

$$\begin{aligned} T_{ij} &= C_{ijkl}S_{kl} - e_{kij}E_k \\ D_i &= e_{ikl}S_{kl} + \epsilon_{ij}E_j \ , \end{aligned} \tag{1}$$

where C_{ijkl} are the stiffness constants measured at a constant electric field, e_{kij} the piezoelectric stress constants, ϵ_{ij} the dielectric constants measured at constant strains, S_{ij} the mechanical strains, T_{ij} the mechanical stresses, D_i the electrical displacements, and E_i the electric field derivable from an electric potential ϕ by $E_i = -\phi_{,i}$. For linear, quasi-static piezoelectricity in absence of body forces and free charges, these quantities satisfy the equilibrium equations $T_{ij,i} = 0$ (mechanical) and $D_{i,i} = 0$ (electrical). Two types of idealized boundary conditions along the free surface are of importance: 1) a traction free and electrically insulated surface and 2) a traction free and electrically conducting surface. Fig. 1 shows these two boundary value problems. The

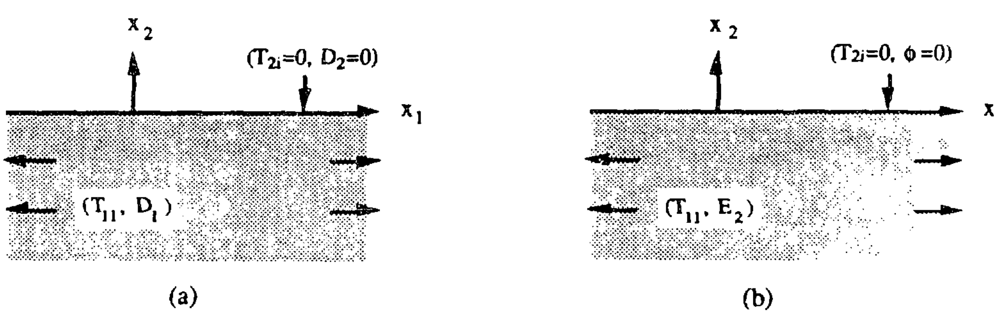

Figure 1: Surfaces of a Piezoelectric Medium with (a) Insulating Boundary Conditions and (b) Conducting Boundary Conditions.

electrically insulating boundary condition corresponds to an adjoining medium with zero dielectric constant and having no free charges residing on the piezoelectric surface. Even though it is not physical to have a medium with zero dielectric constant, it has been argued in [15] [16] [17] that this condition is approximately attained if the piezoelectric medium has much higher dielectric constant and stronger piezoelectric coupling than the adjoining medium such as air. The electrically conducting boundary condition corresponds to an adjoining medium having much higher electric conductivity. Following Lothe and Barnett [15], we attach a superscript Φ to quantities relating to the insulating case and a superscript F to quantities relating to the conducting case.

In view of thin film applications, we consider a piezoelectric medium subjected to fixed misfit strains S_{11} and S_{33}. To illustrate some typical results, 3% biaxial misfit strain is assumed to exist in the absence of an externally applied electric field. For an insulated surface, we examine the effects of an additional applied electric field E_1 in the longitudinal direction. For a conducting surface, we consider an externally applied electric field E_2 in the transverse direction. Before surface instabilities occur, both the stress field and the electric field quantities are constants, so that the equilibrium equations are automatically satisfied; the unperturbed solutions can be readily deduced from the constitutive equations using appropriate boundary conditions.

THE STABILITY ANALYSIS AND A CRITICAL WAVELENGTH

We now investigate whether the surfaces in Fig. 1 are energetically stable, i.e. whether infinitesimal deviations from flatness will be magnified by some kinetic processes such as mass diffusion along the surface. The stability analysis requires the solution to a first order perturbation problem depicted in Fig. 2 where an infinitesimal cosine wave perturbation with amplitude A and wavelength λ is assumed along an otherwise perfectly flat surface. This problem can be solved following a perturbation method used by Gao [4]. For

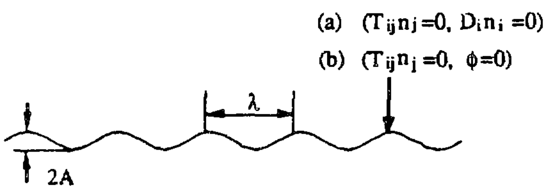

Figure 2: A Cosine Wave Surface with (a) Insulating Boundary Conditions (b) Conducting Boundary Conditions; n_i is the Surface Normal.

conciseness, the details of the mathematical derivations are neglected here. Essentially, the undulating cosine surface problem is converted into a reference flat surface subjected to a distribution of effective surface tractions. The perturbation problem is then solved using a Stroh-like formalism [18] [15] [19] [20] for piezoelectric elasticity problems. The most important result for the stability analysis is the energy change as an initially flat surface (Fig. 1) evolves into the cosine wave surface (Fig. 2). The total energy change (internal energy plus surface energy) per wavelength is found to be

$$\Delta E_{tot} = \frac{\pi^2 \gamma A^2}{\lambda} - \frac{\mathbf{t}_0^T \mathcal{R}e[\mathbf{Y}]\mathbf{t}_0}{2}\pi A^2 + O\left[(A/\lambda)^4\right]. \tag{2}$$

The first term on the right hand side represents the change in the surface energy where γ denotes the surface energy density constant and the second term is the change in internal energy. Superscript T implies vector transpose, $\mathbf{t}_0$ is a 4×1 loading vector, and $\mathbf{Y}$ is a 4×4 constant matrix usually referred to as the surface admittance tensor for a piezoelectric medium [15] [19].

The energy expression in Eq. (2) is formally identical to that derived by Gao [4] for the anisotropic elastic case. The reader may be referred to [4] for some helpful insights and discussions. In the piezoelectric case, both the loading vector $\mathbf{t}_0$ and the admittance tensor $\mathbf{Y}$ take different meanings. In the anisotropic elastic case, $\mathbf{t}_0$ is a 3×1 vector and $\mathbf{Y}$ is a 3×3 matrix. The presence of piezoelectric coupling is exhibited as an added fourth dimension in these quantities.

The loading vector $\mathbf{t}_0$ depends on the electrical boundary condition. In the insulating case $\mathbf{t}_0$ takes the form

$$\mathbf{t}_0^{\Phi} = (T_{11}, 0, T_{13}, D_1)^T \quad (3)$$

where T_{11} and T_{13} denote the mechanical stresses and D_1 is the electric displacement in the x_1 direction induced by an applied electric field. In the conducting case, the loading vector becomes

$$\mathbf{t}_0^{F} = (T_{11}, 0, T_{13}, -E_2)^T \ . \quad (4)$$

where E_2 is the electric field in the x_2 direction. Note that a conducting surface necessarily implies that E_1 and E_3 vanish.

The admittance matrix $\mathbf{Y}$ also has a strong dependence on the electrical boundary condition. Lothe and Barnett [15] [19] showed that the calculation of $\mathbf{Y}$ can be reduced to an eight-dimensional eigenvalue problem involving the material stiffness C_{ijkl}, piezoelectric coefficients e_{kij} and dielectric constants ϵ_{ij}. In the insulating case, $\mathbf{Y}$ is more explicitly written as $\mathbf{Y}^{\Phi}$, while in the conducting case $\mathbf{Y}^{F}$. The properties of $\mathbf{Y}^{\Phi}$ and $\mathbf{Y}^{F}$ have been investigated extensively by Lothe and Barnett [15] [19]. In particular, it was shown that both $\mathbf{Y}^{\Phi}$ and $\mathbf{Y}^{F}$ are Hermitian matrices with symmetric real parts and anti-symmetric imaginary parts, and have real eigenvalues; $\mathbf{Y}^{\Phi}$ has three positive and one negative eigenvalues, while all four eigenvalues of $\mathbf{Y}^{F}$ are positive.

The question of whether a perfectly flat piezoelectric surface is stable amounts to whether the total energy change ΔE_{tot} in going from an initially flat to a slightly undulating surface is positive. In other words, $\Delta E_{tot} > 0$ implies that the flat surface is stable in that any perturbation would tend to increase the energy in the system. On the other hand, if ΔE_{tot} is negative for at least one perturbation wavelength, then a wavy surface is preferred energetically because the energy can be further lowered by roughening. Following Eq. (2), a critical wavelength exists so that the stability condition can be stated as $\lambda < \lambda_{cr}$ where

$$\lambda_{cr} = \frac{2\gamma\pi}{\mathbf{t}_0^T Re[\mathbf{Y}]\mathbf{t}_0} \ . \quad (5)$$

Discussions

Having established the stability condition for a stressed piezoelectric surface, i.e. $\lambda < \lambda_{cr}$, we investigate whether it is possible to suppress surface instabilities by using the electric field as a control parameter. Two types of stabilization can occur leading to a completely stabilized surface or a kinetically more stable surface. The first pertains to applying an external electric field to the system such that $\Delta E_{tot} > 0$ where any perturbation would increase the total energy of the system. This corresponds mathematically to λ_{cr} being negative. For the second type of stabilization, a kinetically more stable surface can be achieved by applying an electric field to the system with a net effect of increasing λ_{cr}. Here, the stressed surface is not truly stable but matter has to diffuse a longer distance in the roughening process, and hence requires a longer period of time. In both cases, what we hope to achieve reduces mathematically to minimizing the product of $\mathbf{t}_0^T Re[\mathbf{Y}]\mathbf{t}_0$. Since the properties of $\mathbf{Y}^{\Phi}$ for the insulating problem is different from $\mathbf{Y}^{F}$ for the conducting problem, we will discuss these two cases separately.

To stabilize an insulated piezoelectric surface, we try to minimize the following:

$$\mathbf{t}_0^T Re[\mathbf{Y}]\mathbf{t}_0 = \mathrm{y}_{11}^{\Phi}(T_{11})^2 + 2\mathrm{y}_{13}^{\Phi}T_{11}T_{13} + 2\mathrm{y}_{14}^{\Phi}T_{11}D_1 + \mathrm{y}_{33}^{\Phi}(T_{13})^2 + 2\mathrm{y}_{34}^{\Phi}T_{13}D_1 + \mathrm{y}_{44}^{\Phi}(D_1)^2 \ , \quad (6)$$

where y_{ij} representing the real part of Y_{ij}.

There exist three mechanisms for stabilization in an insulating problem. The first arises from the negative nature of y_{44}^{Φ}. It has been shown by Lothe and Barnett [15] that, for stable materials, the upper 3×3 block of the $\mathbf{Y}^{\Phi}$ is positive definite, while Y_{44}^{Φ} is negative definite. If we increase the external electric loading E_1, an increase in electric displacement D_1 will occur in all piezoelectric materials. With a negative y_{44}^{Φ} in Eq. (6), and using Eq. (5), we see that any increase in D_1 will enlarge λ_{cr} creating a kinetically more stable state. Furthermore, if D_1 is increased beyond a critical value, λ_{cr} will become negative corresponding to $\Delta E_{tot} > 0$, and a totally stabilized surface can be realized. This y_{44}^{Φ} stabilizing effect exists for all piezoelectric insulated surface. As a demonstration, we consider a zinc oxide surface under a 3% biaxial misfit strain. The orientation in this example is such that the six-fold rotational axis of symmetry of the crystal coincides with the x_2 axis in our problem. For plotting convenience, we normalized the critical wavelength with respect to its value in the absence of an electric field. This normalized critical wavelength $\hat{\lambda}_{cr}$ is plotted as a function of E_1 in Fig. 3. From this example, we see that an electric field of the order of gigavolts is needed to completely stabilize

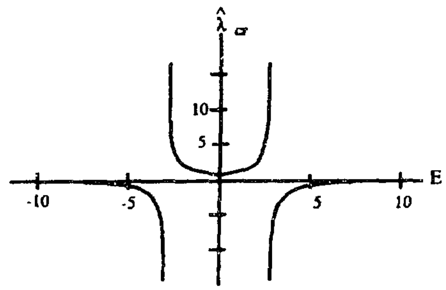

Figure 3: Normalized $\hat{\lambda}_{cr}$ vs. E_1 in Gigavolts for Zinc Oxide in an Insulating Problem

an insulated zinc oxide surface with a 3% misfit strain.

The second mechanism for stabilization can be termed the bulk stress reduction effect where an increase in E_1 may lead to a decrease in the bulk stress T_{11}. This effect occurs in materials where piezoelectric coupling exists between the T_{11} stress and the electric field E_1. Lithium Niobate and Lithium Tantalate, belonging to crystal class *Trigonal* 3*m*, are examples of such materials. In addition to the Y_{44}^{Φ} stabilization effect, an insulated surface with this specific coupling behavior can also be stabilized by a reduction of the bulk stress. It must be noted that this stabilizing effect only occurs within a range, if the electric field is increased beyond this range, it might destabilizes the system by overshooting. As this behavior also applies to a conducting problem, additional details and an example on this effect will be presented shortly.

The third mechanism for stabilization occurs in materials, such as quartz of the *Trigonal* 32 class, where piezoelectric coupling exists between the shear stress T_{13} and the electric field E_1. This stabilization effect stems from the cross terms such as $2y_{13}^{\Phi}T_{11}T_{13}$ and $2y_{34}^{\Phi}T_{13}D_1$ in Eq. (6). As it is possible to have a negative shear stress T_{13} induced by E_1 and a negative y_{34}^{Φ}, the net effect might decrease the product $\mathbf{t}_0^T Re[\mathbf{Y}]\mathbf{t}_0$ and allowing the surface to attain a kinetically more stable state. We call this the shear stress coupling effect. Since the Y_{44}^{Φ} effect is always present in the isolating case, it is not possible to isolate an example where the shear stress coupling effect alone accounts for stabilization. Thus, an example on this mechanism is not included in this paper.

In general, all three stabilizing mechanisms can be present simultaneously. For the materials we have considered, the Y_{44}^{Φ} stabilization effect is a much more dominant effect than the bulk stress reduction and the shear stress coupling effect. As the Y_{44}^{Φ} effect can completely stabilize an insulated piezoelectric surface, the effects of bulk stress reduction and shear stress coupling are relatively unimportant in the insulating case. However, this is not true in the conducting case.

For conducting surfaces, the product that we try to minimize for stabilization is

$$\mathbf{t}_0^T Re[\mathbf{Y}]\mathbf{t}_0 = y_{11}^F(T_{11})^2 + 2y_{13}^F T_{11}T_{13} - 2y_{14}^F T_{11}E_2 + y_{33}^F(T_{13})^2 - 2y_{34}^F T_{13}E_2 + y_{44}^F(E_2)^2 \; . \tag{7}$$

In this case, the 4×4 $\mathbf{Y}^F$ matrix has been shown by Lothe and Barnett [15] to be positive definite. Thus, unlike the insulating case where the surface can be stabilized by the Y_{44}^{Φ} effect, stabilization of a traction free and conducting piezoelectric surface can only be achieved by bulk stress reduction or by shear stress coupling. These two mechanisms do not completely stabilize a conducting surface, only leading to a kinetically more stable state.

To demonstrate the effects of bulk stress reduction in a conducting problem, we consider the example of zinc oxide under a 3% misfit strain. The orientation of the crystal here is identical to that in the insulating example. The result for the normalized critical wavelength $\hat{\lambda}_{cr}$ is plotted in Fig. 4. The bulk stress reduction effect is the only stabilization mechanism present in this example, and we found that an applied electric field of one gigavolts in the x_2 direction causes a 25% increase in λ_{cr}. In several other cases that we have studied.

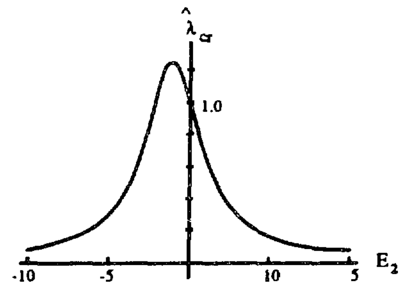

Figure 4: Normalized λ_c vs. E_2 in Gigavolts for Zinc Oxide in a Conducting Problem

the percentage increase in λ_{cr} is less than 10%. This seems to suggest that the bulk stress reduction effect is not particularly effective in controlling instabilities in conducting piezoelectric surfaces. However, in materials exhibiting quadratic effects such as electrostriction and magnetostriction, an increase in electric or magnetic field might lead to a significant reduction in the bulk stress. These effects should be investigated as they might provide better alternatives for stabilization of stressed surfaces.

The shear stress coupling cross terms in Eq. (7) for a conducting surface are identical to that for an insulated surface in Eq. (6). Therefore, as discussed in the insulating case, shear stress coupling effect is also a feasible mechanism, mathematically, for controlling instabilities in a conducting surface. However, we are unable to find a physical example to illustrate this effect. For the materials we have considered, a non-zero shear stress T_{13} either has no effect on stabilization or it leads to instabilities.

Both stabilizing mechanisms for a conducting surface depends on piezoelectric coupling between T_{11}, T_{13} and E_2, thus stabilization is impossible for materials in an orientation where such coupling effect is absent. In fact, an applied E_2 in these cases will only destabilize the system.

Based on the above discussions, it seems possible, at least mathematically, to control the surface stability of a piezoelectric solid by varying an applied electric field in both the insulating and the conducting case. However, we must also address the difficulty in applying these electrical loading physically. With a conducting piezoelectric surface, it might be possible to apply E_2 by imposing an electric potential on the surface. Since x_2 is the thickness direction, typically small on a thin film, a small potential difference might generate a large electric field. A possible setup of this is to grow an insulated thin film on a capacitor plate. Applying E_1 to an insulated surface is an obviously more difficult task, it might be done by setting the surface transverse to a set of parallel capacitor plates.

Also, as an applied electric field of the order of gigavolts is needed to stabilize surface instabilities in thin film applications, we must also investigate other possible adverse effects on the piezoelectric medium caused by an electric field of such magnitude.

Conclusions

In this paper, we have extended the morphological stability analysis for anisotropic materials by Gao [4] to address surface roughening in piezoelectric medium. In contrast to the analysis for an anisotropic medium, where Gao concluded that the surface is always unstable under sufficiently large bulk stress, we found that the electric field can be used as a control parameter to stabilize a stressed piezoelectric surface. Two sets of boundary conditions were considered: A traction free and insulated surface and a traction free and conducting surface.

The stability condition is obtained from analyzing the total energy change associated with roughening of a flat surface. To completely stabilize a stressed flat surface, we try to apply an electric field such that $\Delta E_{tot} > 0$ in Eq. (2). In this case, any perturbation of the flat surface would result in an overall increase of energy for the system. The stability condition can also be expressed in terms of a critical wavelength $\lambda < \lambda_{cr}$. If we can increase this critical wavelength λ_{cr} by applying an electric field, a kinetically more stable state can be achieved.

Three types of stabilizing mechanisms are possible. The first is referred to as the Y_{44}^{Φ} effect and it is applicable only to an insulated surface. This effect arises from the non-positive definiteness of the admittance matrix for an insulating problem. With a large enough electric field in the x_1 direction, this Y_{44}^{Φ} effect can completely stabilize a flat insulated surface. For a conducting surface, a stabilizing mechanism is the bulk stress reduction effect. This effect stems from the piezoelectric coupling of the T_{11} stress and the electric field component E_1. By applying an optimal E_1, we can attain a kinetically most stable state for a conducting surface via bulk stress reduction. The third stabilizing mechanism is the shear stress coupling effect and it arises from a coupling between the shear stress T_{13} and electric components E_1 or E_2. Mathematically this effect can be present in both the insulating and conducting cases. However, the influence of this effect in the insulating case is minimal compared to the Y_{44}^{Φ} stabilization. Furthermore, in the conducting case, a physical example cannot be found where shear stress coupling stabilizes the surface.

As stress and electric field concentrations associated with an undulating surface might lead to mechanical failures, the stabilization of stressed piezoelectric surfaces should be an important concern. In the insulating case, we have shown that a possible mechanism exists, at least mathematically, for complete stabilization of a stressed surface. For a conducting surface, even though a completely stable surface is not attainable by the stabilizing mechanisms presented here; we have shown that it is possible, for some materials, to achieve a kinetically more stable state.

Due to the length restriction imposed on this paper, many of the necessary mathematical theories and details relating to the derivation of our results are not presented here. They will be reported in another paper in the near future.

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