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LETTER TO THE EDITOR

On the morphology of the stress-driven corrugations of the phase boundary between the solid and its melt

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Abstract. The predictions of possible 2D patterns of corrugations (the 'islands') of the unstable flat phase boundary separating the pre-stressed solid substance and the melt are carried out on the basis of a quasi-static evolution. The dispersion relations for the increment of small disturbances are announced for the model of an isotropic solid of arbitrary thickness and in-plane pre-stresses. It seems that the predictions can be checked in various experiments mentioned by Nozieres in 1990 and, in particular, in experiments with ^{4}He similar to those of Torii and Balibar; also, they can be compared with numerous experimental data related to solid epitaxial films.

Recently, Torii and Balibar [2] presented several elegant and reliable results pertaining to the first quantitative experiment on the equilibrium shape of ^{4}He crystals under non-hydrostatic stress. In particular, '... above a certain threshold in strain, large grooves appear on the crystal surface, in agreement with the instability first predicted...' in [3] (as was first announced in [3, 4] and demonstrated theoretically in [5–7], in the absence of surface tension a flat boundary of non-hydrostatically stressed solid of any symmetry is always unstable with respect to 'mass rearrangement'). The other, much more simple and transparent consideration and explanation of the instability was proposed in [8, 9]. The physical mechanisms of the rearrangement can be as different as, for instance, a) melting–freezing or vaporization–sublimation processes at liquid–solid or vapour–solid phase boundaries, b) surface diffusion of particles along free or interfacial boundaries, c) adsorption–desorption of the atoms in epitaxial crystal growth, etc. This universal instability delivers new insights and provides new opportunities in different branches of materials science, a part of which is discussed in [10]. In particular, in addition to successful prediction of the corrugations of crystalline solid ^{4}He, it has allowed an explanation of the phenomenon of dislocation-free Stranski–Krastanov pattern of growth of epitaxial films of GaAs on Si substrates which are not tractable in the framework of the classical theory of that pattern of epitaxial growth (see [11–13]). Regardless of tremendous differences in the physical properties of the substances, the 'corrugation' of ^{4}He and the 'islanding' of growing epitaxial film have a common nature and can be treated in the framework of one and the same theory. Moreover, the experimental study of pre-stressed ^{4}He films (in particular, the control of the kinetics of corrugation growth and migration and,

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also, the opportunity of a direct observation of islanding) is much easier than with typical epitaxial films investigated and used in contemporary nanomechanics. Thus, further experimental and theoretical studies of the ^{4}He films might well be instructive for the simulation and understanding of epitaxial crystal growth and the fracture of thin solid films, in addition to low-temperature physics.

Though conceptually the experiments of [2] have confirmed the predictions of [3, 9] there are certain distinctions between the experimental set up and dynamic regimes investigated in [2], on the one hand, and the two-phase system studied theoretically in [3–7, 10], on the other hand. First, we relied on the investigation of positiveness of the second energy variation, which appears to be a specific integro-differential quadratic form of the 'naturally' defined fields of allowable variations. Actually, this purely static approach to the problem of stability is deeply bound up with the dynamic approach resting on the study of the evolution of small disturbances. Indeed, singular values of the second energy variation admit the transparent physical interpretation: they coincide with the eigenfrequencies of the two-phase heterogeneous system in question with the 'instantaneous' kinetics of the phase transformation at the boundary. It seems, however, that neither static approximation nor the inertia of the substance (which significantly affects the spectrum of oscillation) are the satisfactory theoretical schemes relevant to the experiments of Torii and Balibar [2] and others [1]. The processes studied in [2] are instead quasi-static with a sharp dependence of the kinetics of the irreversible processes on temperature. Secondly, in the theoretical studies mentioned above we have used 2D elasticity theory, and the 3D approach is becoming an urgent necessity in view of the wide opportunities of experiments with solid ^{4}He films (in addition, typical misfit stresses in the problems of epitaxy are two-dimensional rather than uniaxial). Therefore, in this short note we abandon both of the above-mentioned assumptions of our earlier studies and investigate a quasi-static evolution of the pre-stressed isotropic elastic film attached to a rigid substrate and bordering its melt. However, in order to elucidate the role of non-hydrostatic stresses we ignore here the influence of thermal fields and choose the simplest models of the surface tension and mass flux: the mass flux across the phase boundary is assumed to be proportional to the difference in chemical potential between the two phases (see [8] for the required justification). In this note we announce the dispersion relation of the rate of amplification/decay for different surface corrugation, Fourier components, which allows one to study morphological patterns of the unstable corrugations (islands) in pre-stressed solid films and those possible symmetry changes of the phase (or the free) surface morphology which accompany growth of the film thickness.

Let us consider a stretched thin solid film of thickness H attached to a rigid substrate having a planar matching surface S. The stretches can be produced a) by the externally provided displacements of the side-walls in two-phase systems containing solid ^{4}He and its melt (as in the experiments of [2]) or b) by the misfit in the lattice parameters of the epitaxial film and the substrate. In what follows we use the Eulerian description of the solid and the summation convention regarding repeated indices. We use the notation z^i (the indices $i, j, k, l, \ldots$ take on the values 1, 2, 3) of the Cartesian spatial coordinates with the in-plane axes z^a (the indices $a, b, c, \ldots$ take on the values 1, 2). The symbols ρ, ψ and P^{ji} are used for the mass density, specific (per unit mass) free bulk energy and the (Cauchy) stress tensor of the solid film. We assume that at the upper boundary Σ, the solid can exchange mass with its melt or vapour, which have unaltered pressure p_a and chemical potential μ_a. The

surface energy of the phase boundary Σ is assumed proportional (with the coefficient of surface tension σ) to the surface area in the deformed (actual) configuration.

In what follows we investigate the quasi-static evolution of the above system; thus in each of the consequent configurations the standard equilibrium equation and boundary conditions have to be satisfied:

$$\nabla_j P^{ji} = 0 \tag{1a}$$

$$P^{ji} N_j = -pN^i$$

$$p_a - p = -2\sigma/R \tag{1b}$$

where $-p$ is the normal component of the stress at Σ (the tangential component vanishes at the boundary with liquid or vapour); N_i is the unit normal vector of Σ pointed into the film, R is the mean curvature of the phase boundary. We make use of the notation v_i and C for velocities of the film particles and the phase boundary, respectively, and we assume that the mass flux across the boundary

$$J \equiv \rho(C - v^i N_i)$$

is proportional to the jump in the chemical potentials of the phases:

$$J = -\kappa(\mu_a - \psi - p/\rho) \tag{2}$$

where κ is a positive coefficient.

Let us consider the equilibrium configuration in which the film is uniformly stressed and has a flat phase boundary. We shall investigate the evolution of small disturbances of this configuration and use the 'degree' mark for the equilibrium fields. It is convenient to introduce the tensor

$$D^{ij} \equiv P^{ij\circ} + p_a \delta^{ij}$$

which vanishes identically if the film is hydrostatically stressed (and, thus, it is the measure of equilibrium shear stresses in the film). One can easily check that only the in-plane components of

$$D^{ij} - D^{ab} \equiv T^{ab}$$

can deviate from zero. In the case of epitaxial films it is natural to call T^{ab} the misfit-stress tensor.

The equations governing the evolution of small disturbances can be derived as the result of linearization of the system (1), (2). A routine computation leads to the following master equations for small disturbances in the vicinity of uniform equilibrium configuration: a) within the film

$$C^{ijkl} \nabla_j \nabla_l v_k = 0 \tag{3}$$

b) at the boundary Σ

$$\left(C^{ijkl} \nabla_l v_k - \sigma \delta^{ij} \nabla_a \nabla^a C\right) N_j^\circ - D^{ji} z_j^{a\circ} \nabla_l C = 0 \tag{4}$$

$$\delta C/\delta t = (\partial v^j/\partial t)_\Sigma N_j^\circ + \left(\kappa/\rho_0^2\right)\left(D^{ij} \nabla_i v_j - \sigma \nabla_a \nabla^a C\right) \tag{5}$$

where C^{ijkl} is the tensor of elastic modulae; the symbols $z^{a\circ}_j$ and $\delta/\delta t$ are used for the 'shift-tensor' of the embedded surface (the operator of tangential projection on the surface) and for the time-derivative along the normal vector to the surface (both operators are understood in the sense of [14,7]); the subscript Σ denotes the limit at the phase boundary.

We are looking for the solution of equations (3)–(5) of the form

$$v^i(z^j,t) = F^i(z)e^{\eta t + ik_a z^a}$$
$$C(z^a,t) = Fe^{\eta t + ik_a z^a} \tag{6}$$

where $z \equiv z^3$ is the vertical coordinate, k_a is the in-plane wave-vector and η is the increment/decrement of the amplitude of the disturbances.

Plugging (6) into (3)–(5) and the boundary condition at the matching surface and making the standard routine computation one can find the required formula of η. In particular, in the case of an isotropic film (with shear modulus μ and Poisson ratio ν) and small pre-deformations we arrive at the following dispersion relation:

$$\eta\rho_0^2/|k|\kappa\mu = \left((1-\nu)\left(\tau^{ab}e_a e_b\right)^2[(3-4\nu)\sinh(2h)+2h]\right.$$
$$\left. + 2\Delta\left[\left(\tau^{cd}e_c q_d\right)^2\tanh(h) - (\sigma/\mu)|k|\right]\right)$$
$$\times\left(2\Delta + (\sigma/\mu)|k|(1-\nu)[(3-4\nu)\sinh(2h)-2h]\right)^{-1} \tag{7}$$

where $\Delta \equiv 4(1-\nu)^2 + h^2 + (3-4\nu)\sinh^2 h$. We use the notation $|k|$ for the modulus of the wave-vector k^a and the notation e_a, q_a for unit in-plane vectors parallel and orthogonal to k^a, respectively; $\tau^{ab} \equiv T^{ab}/\mu$ and $h \equiv H|k|$ are the dimensionless misfit stresses and thickness, respectively; ν is the Poisson ratio.

In the asymptotic case of infinitely thick film the master formula (7) gives

$$\eta\rho_0^2/|k|\kappa\mu = \left((1-\nu)\left(\tau^{ab}e_a e_b\right)^2 + \left(\tau^{cd}e_c q_d\right)^2 - (\sigma/\mu)|k|\right)$$
$$\times\left(1+(1-\nu)(\sigma/\mu)|k|\right)^{-1}. \tag{8}$$

The maxima of $\eta(k^a)$ correspond to the Fourier components of corrugations having the fastest rate of growth which can be investigated with the help of formulas (7) and (8). Formula (7) leads, in particular, to the following value of the critical film thickness

$$H_{\text{crit}} = \mu\sigma/T^2_{\max} \tag{9}$$

where $T_{\max}$ is the greater of two principal in-plane misfit stresses. Formula (9) generalizes a similar formula for the critical thickness given in [7, 10, 13, 15] established originally in the framework of the 2D approach. The investigation of formulas (7) and (8) shows that at $H < H_{\text{crit}}$ the film is stable with respect to arbitrary surface

corrugations while at $H > H_{crit}$ it becomes unstable. At this initial stage the most unstable corrugations are in the shape of 'trenches' parallel to the direction of minimal in-plane principal stress T_{min}. As the film thickens the most unstable mode changes. The evolution scenario depends significantly on the misfit stresses: in particular, on the dimensionless parameter

$$s \equiv (T_{max} + T_{min})/(T_{max} - T_{min}).$$

In the case of uniaxial misfit stresses ($|s| = 1$) the most unstable corrugations take the shape of an array of parallel trenches collinear with the direction of a smaller in-plane stress T_{min}, the distance between the trenches diminishes as the thickness increases. In the case of the misfit stresses of a pure shear (when $T_{max} = -T_{min}$ and $s = 0$) the most unstable mode has the shape of an array of squares the size of which diminishes with increase in thickness.

At the final stage of growth when the film is infinitely thick the pattern of fastest growth corresponds to the array of trenches if the following inequality is valid

$$|s| > (1 - \nu)/\nu. \tag{10}$$

If inequality (10) is violated, then the pattern of fastest growth corresponds to the array of rectangular islands; the ratio Ra of legs of the elementary cell is the following

$$Ra = \sqrt{\frac{1 - \nu - s\nu}{1 - \nu + s\nu}}. \tag{11}$$

Thus, in the latter case at a certain (second!) critical thickness, the growth pattern changes from trench-like to island-like.

The above-mentioned results can easily be generalized for some other models of surface tension and mass-transport mechanisms. In particular, for models with distinct surface tension σ and surface energy σ^* (see [8, 16]) we arrive at the dispersion relations (7) and (8) with the extra term $-|k|\sigma^*$ on the RHS. On the other hand, multiplying these relations by $|k|^2$ we arrive at the dispersion relations for the case of mass transport produced by surface diffusion. All these complications as well as other sorts of misfit stresses will be described in detail elsewhere.

It seems highly desirable to verify the above predictions experimentally with solid ^{4}He and to compare them with various experimental data pertaining to epitaxial solid films.

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