Finite-time Singularities in Surface-Diffusion Instabilities are Cured by Plasticity

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A free material surface which supports surface diffusion becomes unstable when put under external non-hydrostatic stress. Since the chemical potential on a stressed surface is larger inside an indentation, small shape fluctuations develop because material preferentially diffuses out of indentations. When the bulk of the material is purely elastic one expects this instability to run into a finite-time cusp singularity. It is shown here that this singularity is cured by plastic effects in the material, turning the singular solution to a regular crack.

We address the problem of an amorphous material with a free surface on which the material can diffuse such that the surface normal velocity is proportional to $\partial^2 \mu / \partial b^2$ where $\mu$ is the local chemical potential and $b$ is the parameterization of the interface. When the material itself is purely elastic, this phenomenon leads to an instability which was termed 'thermal grooving' by Mullins [1] who discovered it. The phenomenon of grooving is equally present in crystals, where it provides an amnogmechanism for the failure of growing crystals [2,3], as it does in a range of amorphous solids that concern us here. Mullins considered the linear instability taking into account only the curvature dependence of the chemical potential. In fact, the chemical potential along the interface is strongly dependent on the elastic energy; linear stability analysis taking both effects into account [4] reveals that the surface is stable for short wavelengths but unstable for longer ones, with a usual 'fastest growing mode' whose wavelength depends on the material parameters. The late stage of development of this instability was initially studied by a handful of researchers, namely Asaro and Tiller [2] and Grinfeld [6] and followed by many [4,7,8,9]. The result is that the instability runs into a finite-time singularity, with the growing indentation forming a cusp. Clearly, this often explored [7,10,11] mathematical phenomenon cannot be physical, and its discovery leaves open the question of the physical mechanism that may cure the singularity.

The question of what might cure the finite-time singularity in the Asaro-Tiller-Grinfeld (ATG) instability remained dormant until recently Bréner and Spatschek proposed that inertial effect in the velocity of the moving boundary may tame the singularity [8]. These authors pointed out that without inertial effects the velocity of the tip $v$ appears in one dimensionless combination, i.e. $vr_0^2/D$ where $r_0$ is the radius of the tip and $D$ the diffusion coefficient (of dimension length$^2$/time, and cf. Eq. (1)). Therefore there is no mechanism to select $v$ or $r_0$, and as $r_0$ decreases without limit, $v$ increases without limit. Once inertial effects are taken into account the velocity appears also in the combination $v/v_R$ where $v_R$ is the Rayleigh wave speed. Thus a selection of both $v$ and $r_0$ can happen. While clearly correct, the present authors stress that in many cases the surface diffusion is very slow, leading to small interface velocities which do not justify the incorporation of inertial terms. We focus here on such cases where the question of taming the cusp-singularities remains open.

In this Letter we propose that the generic mechanism for the taming of the ATG instability may be plastic deformation in the stressed material, especially near the putative cusp. To test and demonstrate this proposition we will employ the recently proposed theory of elasto-plastic dynamics in amorphous systems [12]. To this theory, which is valid in the bulk of the material, we couple the surface diffusion, allowing the chemical potential to take its stress dependence from the elasto-plastic theory. For concreteness we choose to explore this interesting physics on the inner surface of a circular hole which is stressed at infinity in a radial fashion. The surface diffusion modifies the shape of the slightly perturbed circular hole, leading eventually to a highly non-linear morphology. With a sharpening interface due to the surface diffusion instability, stresses in the bulk increase rapidly, exceeding at some point in time the yield stress of the material, triggering plastic flows which are dissipated by the exertion of plastic work [12,13,14]. It is interesting to observe the coupling of both processes, namely surface diffusion and plasticity, as they become competitive and of opposite influence on the morphology, to a point where the finite time singularity is removed. In addition to shedding new light on the late stage of the ATG instability we find that the elasto-plastic theory employed here, which is sensitive to the plastic properties of matter, allows a natural understanding of this a-priori seemingly hard problem.

The model system that we consider here is an infinite 2-dimensional isotropic elasto-plastic sheet with a hole in the center whose radius is $R(\theta)$. For $r(\theta) < R(\theta)$ the system is void, whereas the elasto-plastic material occupies the region $r(\theta) \geq R(\theta)$. The boundary is traction free, meaning that on the boundary $\sigma_{ij}n_j = 0$ where $\sigma$ is the stress tensor and $n$ is the unit normal vector. The equations of acceleration and continuity are exact, reading:

$$\rho \frac{Dv}{Dt} = \nabla \cdot \sigma \quad (1)$$

$$\frac{D\rho}{Dt} = -\rho \nabla \cdot v \quad (2)$$

Here the full material derivative $D$ is defined for an arbi-
trary tensor $\mathbf{A}$ as:

$$\frac{D\mathbf{A}}{Dt} = \partial_t \mathbf{A} + v \cdot \nabla \mathbf{A} + \mathbf{A} \cdot \omega - \omega \cdot \mathbf{A},$$  \hspace{1cm} (3)

where $\omega$ is the spin tensor $\omega_{ij} \equiv \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} - \frac{\partial v_j}{\partial x_i} \right)$. Reading Eq. (11) in radial coordinates we get:

$$\rho \left( \frac{\partial v_r}{\partial t} + v_r \frac{\partial v_r}{\partial r} + \frac{v_\theta v_r}{r} - \frac{v_r^2}{r} \right) = \frac{1}{r} \frac{\partial p}{\partial \theta} - \frac{1}{r^2} \frac{\partial^2 p}{\partial r \partial \theta},$$  \hspace{1cm} (4)

$$\rho \left( \frac{\partial v_\theta}{\partial t} + v_r \frac{\partial v_\theta}{\partial r} + \frac{v_\theta v_r}{r} + \frac{v_\theta v_\theta}{r} \right) = \frac{1}{r} \frac{\partial p}{\partial r} + \frac{1}{r} \frac{\partial s}{\partial \theta} + \frac{1}{r} \frac{\partial p}{\partial \theta} + \frac{2 \tau}{r}.$$  \hspace{1cm} (5)

Here $s$ and $\tau$ are defined via the transformations

$$\sigma_{rr} = s_{rr} - p; \quad \sigma_{\theta \theta} = s_{\theta \theta} - p; \quad \sigma_{r \theta} = \tau; \quad \sigma_{r r} = -s_{\theta \theta} = -s.$$  \hspace{1cm} (6)

Below our velocities are sufficiently small to allow neglecting the nonlinear terms $-v_r^2/r$ and $v_\theta v_r/r$. On the other hand nonlinear terms containing derivatives are retained, since the derivatives are large.

The velocity at the interface, $R(\theta)$, reads:

$$\frac{\partial R}{\partial t} = \frac{v_r}{R} \partial_\theta R + v_r$$  \hspace{1cm} (7)

When the surface evolves, the stresses in the bulk evolve accordingly. A fundamental assumption of our elasto-plastic theory is that the total rate of deformation $D^{tot} \equiv \frac{1}{2}[\nabla v + (\nabla v)^\top]$ can be represented as a linear combination of its elastic and plastic components \[14\]:

$$D^{tot} = D^{el} + D^{pl}$$  \hspace{1cm} (8)

Here the elastic contribution, $D^{el}$, is assumed to be linearly dependent on the stress (linear elasticity)

$$D_{ij}^{el} = \frac{D\epsilon_{ij}}{Dt} = \epsilon_{ij} = -\frac{p \delta_{ij}}{2K} + \frac{s_{ij}}{2\mu}$$  \hspace{1cm} (9)

where $K$ and $\mu$ are the 2-dimensional bulk and shear moduli and $p$ and $s_{ij}$ are the pressure and the deviatoric stress tensor, respectively. The plastic rate of deformation, $D^{pl}$, is determined by a set of internal fields which are discussed at length in \[12\] where the elasto-plastic theory is presented in detail. For the purpose of this Letter it is enough to state that the tensorial field $m$ acts as a ‘back-stress’ due to plastic deformations, and the scalar field $\chi$ is the effective temperature that controls the amount of configurational disorder in the elasto-plastic materials. The constitutive relations that were derived for these fields read

$$D_{ij}^{pl} = e^{-\chi} C(\check{s}) \left( \frac{s_{ij}}{\check{s}} - m_{ij} \right)$$  \hspace{1cm} (10)

$$\frac{Dm_{ij}}{Dt} = 2e^{-\chi} D_{ij}^{pl} - \Gamma(s_{ij}, m_{ij}) m_{ij}$$  \hspace{1cm} (11)

In these equations all the stresses were normalized by the yield stress of the material $s_y$, using $\check{s} = \sqrt{s_{ij} s_{ij}/2s_y^2}$.

To this theory we need to couple now the surface diffusion, expressed in terms of the normal velocity, $v_n(\theta)$ on the boundary. Without the effects of elasto-plasticity in the bulk the normal velocity satisfies

$$v_n = -\frac{D_s \Omega^2 \delta}{k_BT} \frac{\partial^2 \mu}{\partial \theta^2}$$  \hspace{1cm} (15)

where $D_s$, the surface diffusion constant, $\Omega$, particle volume and $\delta$, the number of particles per unit area. In solving the coupled problem the total normal velocity should be computed as a sum of this contribution and the one coming from Eqs. (11) and (13).

The chemical potential on the boundary, $\mu(\theta)$, is associated on the one hand with the destabilizing curvature and on the other hand with the stabilizing surface energy \[4\]:

$$\mu = \mu_0 - \gamma \kappa + \mathcal{E}.$$  \hspace{1cm} (16)

Here $\mu_0$ is the chemical potential of the unperturbed surface, the curvature $\kappa(\theta) = (R^2 + 2R^2 - R''^2)/(R^2 + R''^2)^{3/2}$ and the strain energy density $\mathcal{E} = \frac{1}{2} \sigma_{ij} \epsilon_{ij} \[15\]. $\gamma$ is the surface energy.

In terms of these effects on the chemical potential one derives the equation for the normal velocity due to surface diffusion alone:

$$v_n = -\frac{D_s \Omega^2 \delta}{k_BT} \frac{\partial^2 \mu}{\partial \theta^2} \left[ \left( -\gamma \kappa(\theta) \right) + \left( \frac{1 - \nu^2}{2E} \sigma_{ij}^{\kappa}(\theta) \right) \right],$$  \hspace{1cm} (17)

where $\nu$ is the Poisson ratio and $E$ Young’s Modulus. After non-dimensionalization and projecting from $v_n$ to
\[ \tilde{v}_r = -\partial_\theta \frac{1}{\sqrt{R^2 + R'^2}} \partial_\theta \left[ -\tilde{\kappa}(\theta) + \frac{(1 - \nu^2)}{2} \tilde{\sigma}^{ii}(\theta) \right] \] (18)

As noted, we need to couple Eqs. \([4], [5]\) and \([18]\) to be solved together, such that the surface normal velocity is made from the sum of contributions coming from the bulk dynamics and the surface dynamics respectively. This should be done while keeping the traction free boundary conditions and the initial condition of a pure elastic solution of a slightly perturbed circle. In practice we used the fact that the elastic response is the fastest process in this problem. Accordingly we solved at each iteration first the elastic part of the model (with \(D^{pl} = 0\)) to find the stress fields which is in agreement with the given interface, without taking into account any plastic deformation. Second, elasto-plastic relaxation was allowed to take place, until the system reached elasto-plastic equilibrium. Here ‘equilibrium’ means that \(D^{pl}\) is smaller than \(10^{-4}\). Lastly, a step of surface diffusion was allowed to take place using an adaptive time step such as to bound the maximal movement of the boundary by \(10^{-4}\). The last step changes the morphology of the boundary again, necessitating a re-calculation of the elastic fields around the new boundary, etc. Since the effect on the velocity of the interface in the last two steps is additive, these steps (being infinitesimal) could be also done simultaneously with impunity. We chose to separate the last two steps since the plastic and surface diffusion processes are non-dimensionalized independently and have different normalization values of characteristic times and stresses.

In order to realize an infinite sheet it is convenient to transform the \((r, \theta)\) coordinate system through a conformal transformation to a new, finite domain \((\zeta, \theta)\) with \(\zeta \in [0, 1]\):

\[ \zeta(\theta) = R(\theta, t)/r \ . \] (19)

In the finite space all the derivatives are computed using finite differences and are redefined for the transformed space using the chain rule: \(\partial_x = x_k \partial_{x_k}\). Explicitly

\[ \frac{\partial}{\partial r} \rightarrow \frac{-\zeta^2}{R} \frac{\partial}{\partial \zeta} , \quad \frac{\partial}{\partial \theta} \rightarrow \frac{R'}{R} \frac{\partial}{\partial \zeta} + \frac{\partial}{\partial \theta} , \quad \frac{\partial}{\partial t} \rightarrow \frac{R'}{R} \frac{\partial}{\partial \zeta} + \frac{\partial}{\partial t} \] (20)

At \(\zeta = 0\) all the derivatives vanish and on the boundary \(\zeta = 1\) the time derivatives are estimated by linear extrapolation in the \(\zeta\) direction. For the sake of numerical stability small viscosity terms were added to the acceleration components. The coupled equations were solved using \(K = 100\) and \(\mu = 50\). Since the speed of sound is orders of magnitude larger than the velocity of the interface, we could safely neglect the effects of Eq. \([2]\), giving up on seeing the sound waves. The initial condition on the perturbed circle were

\[ R(t = 0, \theta) = 1 + 0.01 \cos 2\theta \ , \] (21)

the stress at infinity was chosen to be \(\sigma_\infty = 0.9\sigma_y\). The Poisson ratio \(\nu\) in Eq. \([18]\) is \(1/3\). The surface diffusion equations contain fourth order derivatives, calling for spectral techniques for sufficiently stable evaluation. All the other derivatives were computed by finite differences.

**Results and Discussion:** The typical morphology of the unstable interface is shown in Fig. \(\text{II}\) The elastic solution for the finite-time singularity is well established and was reproduced in our numerics. The curvature \(\kappa\) in the growing cusp first grows exponentially and rapidly switches to faster regime that agrees with the growth law

\[ \kappa(t) \propto (t^* - t)^{-1/2} \ . \] (22)

To make this growth obvious we plotted the curvature of the elastic solution in Fig. \(\text{IV}\) as a function of \((t^* - t)^{-1/2}\) with \(t^* = 5.37 \times 10^{-4}\). Once plasticity is allowed to intervene, it prevents the finite-time singularity by blunting the tip and by dissipating the stress.
FIG. 3: The tip velocity (panel A), its first and second time derivatives (panels B and C respectively). We see that the singularity is cured and the velocity decelerates due to the plastic effects.

On the boundary, the smoothening of the interface in the vicinity of the cusp via blunting is the “cure” of the singularity. The ever-increasing curvature occurring in the elastic solution is prevented in the plastic solution by the plastic flow induced by stress concentration. The avoidance of the singularity is shown by the deceleration in the tip velocity, see Fig. 3 panels B and C.

It is important to stress that although the plasticity in the bulk succeeds to cure the finite-time cusp-singularity, the role of surface diffusion is far from being negligible. Without it, the stressed circle would remain stable to small shape fluctuations, as was demonstrated recently in [16]. The surface diffusion makes the circle unstable, and the instability results in the growth of a groove. Without plasticity in the bulk the solution loses its meaning at \( t = t^* \), whereas now, with plasticity playing its useful role, the solutions continue to exist at times \( t > t^* \), in a form of a lengthening groove, or crack, whose tip is protected from cusping by the plastic effects. At some point the crack will increase its velocity due to the Griffith mechanism, and then the problem becomes inertial again.

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