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1. Formation of grain boundaries

As a liquid solidifies, crystals grow from separate nucleation sites, each with a random orientation of crystal planes. Eventually these crystals grow and meet at a grain boundary. These can be observed as shallow grooves on metal surfaces. The density of grain boundaries affects the appearance of metal surfaces, as well as the strength of the solid. For example, grain boundaries are used to create a spangled appearance on the surface of some metal sheet products. Through an electron microscope, grain boundaries beginning at the nanoscale, can be observed to grow in thickness and depth. They may be several millimetre in length between triple grain junctions, in comparison to a thickness of a few micron. Therefore grain boundary evolution may be described as a two-dimensional cross section profile, $y(x,t)$ in Cartesian coordinates. From Herring (1951), the surface energy per particle is

$$\Phi(\phi) = \Omega[\gamma_s''(\phi) + \gamma_s(\phi)]\kappa, \tag{1}$$

where Ω is the mean particle volume, γ_s is surface tension, κ is curvature and $\phi = \arctan(y_x)$ is the orientation angle of the surface. Mullins' 1957 theory of surface diffusion applies the Nernst–Einstein relation to express material area flux due to surface diffusion,

$$J = -\nu\Omega\frac{D_s(\phi)}{kT}\frac{\partial\Phi}{\partial s},$$

where ν is areal density of particles, D_s is surface mobility, kT is absolute temperature multiplied by Boltzmann's constant, and s is arclength. For an ideal isotropic material, D_s and Φ are independent of surface orientation. In that case, the equation of continuity for conservation of volume, results in the surface diffusion equation

$$y_t = -B\,\partial_x\left\{\left(1+y_x^2\right)^{-1/2}\partial_x\frac{y_{xx}}{\left(1+y_x^2\right)^{3/2}}\right\}\ ;B>0\ \text{(constant)} \tag{2}$$

$$\begin{aligned}\text{or}\quad \theta_t &= -B\,\partial_x^2\left\{f(\theta)\,\partial_x\left[\theta_x f(\theta)\sqrt{[f'(\theta)]^2+[\theta f'(\theta)+f(\theta)]^2}\right]\right\} \\ &= -B\,\partial_x^2\left\{\left(1+\theta^2\right)^{-1/2}\partial_x\left[\left(1+\theta^2\right)^{-3/2}\theta_x\right]\right\},\end{aligned} \tag{3}$$

$$\text{where}\quad \theta = y_x\ ,\ f(\theta) = \frac{1}{\sqrt{1+\theta^2}} = \cos\phi.$$

This equation is invariant under Euclidean isometries. By way of contrast, the small-slope approximation $f=1$ gives the linear fourth-order diffusion equation

$$y_t = -By_{xxxx}$$

that is not invariant under rotations in the (x,y) plane.

2. Integrable nonlinear models.

A rotation transforms the linear equation to a nonlinear equation of the form (3) with $f(\theta) = \alpha/(\beta+\theta)$. This is a useful integrable nonlinear model, since exact solutions can be constructed when $f(\theta)$ approximates the isotropic version at both small slopes and large slopes. These solutions are exact solutions for surface diffusion on a non-isotropic material, which in general leads to a fourth-order nonlinear diffusion equation of the form

$$\theta_t = -\partial_x^2\{D(\theta)\partial_x[E(\theta)\theta_x]\}. \tag{4}$$

The surface of the recently solidified material is assumed to be horizontal and flat, with initial condition

$$\theta(x,0) = 0. \tag{5}$$

Just as on a material surface, within the disordered region at a grain boundary, atoms are at a higher energy state than in the crystal lattice. This gives rise to a grain boundary tension γ_b, just as for surface tension γ_s, has the units of energy per unit area or force per unit length. The groove angle is such that the grain boundary tension balances surface tension on both arms of the groove. Hence for a symmetric groove centred at $x = 0$,

$$\gamma_b = 2\gamma_s \sin(\phi).$$

This leads to the boundary condition

$$\theta(0,t) = m = \left[\left(\frac{2\gamma_s}{\gamma_b}\right)^2 - 1\right]^{-1/2}. \tag{6}$$

For a symmetric groove, there must be zero mass flux in either direction out of the groove, implying

$$\partial_x[E(\theta)\theta_x] = 0, \;\; x = 0. \tag{7}$$

Finally, far from the grain boundary, the initial condition is not disturbed, so

$$\theta(x,t) \to 0, \;\; x \to 0, \tag{8}$$

$$\text{and } \theta_x(x,t) \to 0, \;\; x \to 0. \tag{9}$$

The balance between grain boundary tension and surface tension is depicted in Figure 1.

The system (4)–(9) is the initial-boundary problem for evolution of a grain boundary by surface diffusion. For a near-isotropic material, this problem was solved exactly in [4] by approximating $f(\theta)$ by a reciprocal linear spline $f(\theta) = \alpha_i/(\beta_i + \theta)$ between node points $\theta_{i-1} \leq \theta \leq \theta_i$ for $i = 1, \cdots, N$. In practice, increasing the number of spline segments made little difference to the solution when N was increased from 4 to 16. The spline method can be used for this particular problem because it has a similarity solution of the form $\theta = g(xt^{-1/4})$. It then follows that the location of the node value $\theta = \theta_i$ is $g^{-1}(\theta_i)t^{1/4}$.

It is desirable to be able to solve some form of the anisotropic surface diffusion equation for other useful boundary conditions that are not compatible with the similarity reduction. Then the location of the node values can no longer be determined

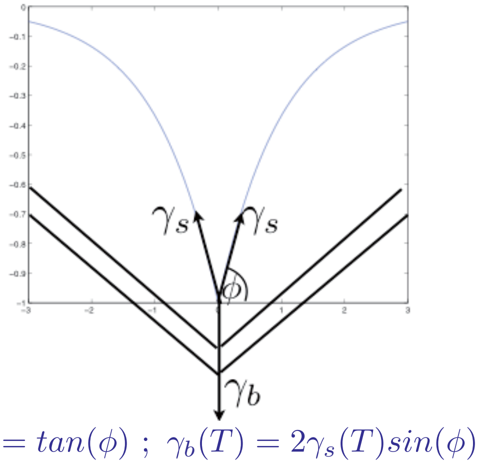

FIGURE 1. Balance between grain boundary tension and surface tension.

by elementary methods, so the spline representation of $D(\theta)$ and $E(\theta)$ can no longer be used. Neverheless the integrable model $D(\theta) = \beta/(\beta+\theta)$, $E(\theta) = E_0\beta^3/(\beta+\theta)^3$, still gives a reasonable representation of a near-isotropic material over a wider range of inclination angles ϕ, than does the linear model. Since it can be easily transformed to the linear model, it can be solved exactly for a variety of initial and boundary conditions. The next problem to be solved, has a time dependent slope $m(t)$ prescribed at $x = 0$. The dihedral angle of the groove may be controlled by heating or cooling. As the solid surface cools, surface tension will increase, so that the equilibrium slope will decrease. In the simplest generalisation, the expression (1 for surface energy has an extra factor $\xi(T)$ that depends on temperature T. After defining a new time-like coordinate

$$\tau = \frac{1}{\nu k}\int_0^t \frac{\xi(T(t_1))}{T(t_1)} dt_1,$$

the surface diffusion equation is still autonomous, reducing again to (4) but with τ replacing t. After rotation by angle $\cot^{-1}\beta$, the integrable model transforms to the linear fourth-order diffusion equation $\bar{y}_\tau = -\bar{y}_{\bar{x}\bar{x}\bar{x}\bar{x}}$. When m is time dependent, the solution $\bar{y}(\bar{x},\tau)$ can no longer be a function of the scaling invariant $X = \bar{x}\tau^{-1/2}$. However $\bar{y}$ may be conveniently expanded as a series of separated solutions $\tau^{i/4}$ multiplying a function of X that is expressed in terms of generalised hypergeometric functions.

$$\bar{y} = g_0(X) + \sum_{j=1}^{\infty} \tau^{j/4}\Big[\ K_{1j}\ {}_1F_3\left(\frac{-j}{4};[\frac{1}{4},\frac{1}{2},\frac{3}{4}],\frac{X^4}{256}\right) + K_{2j}\ X\ {}_1F_3\left(\frac{1}{4}-\frac{j}{4};[\frac{1}{2},\frac{3}{4},\frac{5}{4}],\frac{X^4}{256}\right)$$
$$+K_{3j}\ X^2\ {}_1F_3\left(\frac{1}{2}-\frac{j}{4};[\frac{3}{4},\frac{5}{4},\frac{3}{2}],\frac{X^4}{256}\right) + K_{4j}\ X^3\ {}_1F_3\left(\frac{3}{4}-\frac{j}{4};[\frac{5}{4},\frac{3}{2},\frac{7}{4}],\frac{X^4}{256}\right)\Big].$$

When $m(\tau)$ is an analytic function in $\tau^{1/4}$, the boundary $x = 0$ can be specified as $\bar{x}$ being a power series in $\tau^{1/4}$,

$$\bar{x} = \beta\tau^{1/4}\sum_{i=0}^{\infty} b_i\tau^{i/4}.$$

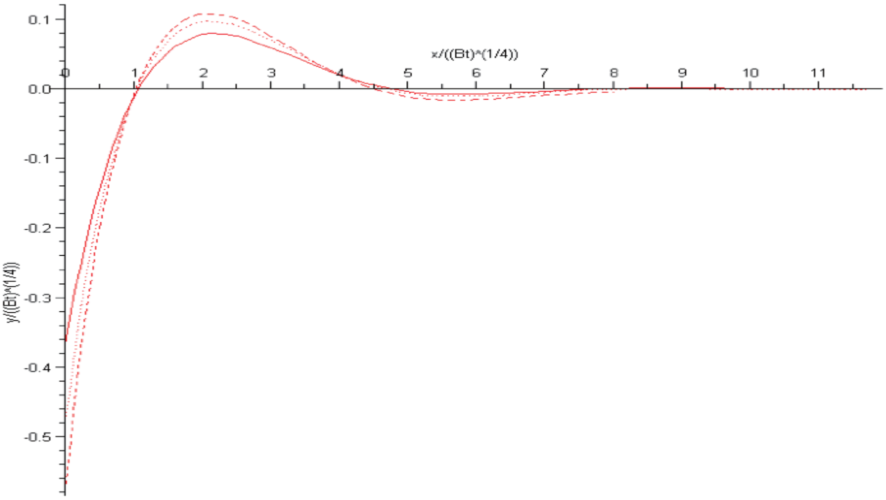

FIGURE 2. Groove with slope $m = 0.5 + 0.5\sqrt{\tau}$ at the root. Output times are $\tau = 0.0002,\ 0.1$ and 1.0

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The coefficients b_i and K_{ij} are then uniquely determined by the boundary conditions. This solution method was completed fully for the steepening groove $m = 0.5 + 0.5\tau^{1/4}$ by Broadbridge and Goard [5]. This is shown here in Figure 2.

3. Method of lines with central finite differences

Although it solves a very challenging problem, the exact solution method is complicated even after a minor change to the boundary conditions. We need also to have a back-up numerical method that can easily handle a variety of boundary conditions. One standard approach that is relatively easy to implement is the method of lines that constructs a a semi-discrete model by central finite differencing of spatial derivatives. The resulting system of ordinary differential equations may be run through a numerical solver for stiff systems. For a uniform grid with spacing h, at $x = jh$, the approximate second and fourth derivatives are

$$\frac{\partial^2\theta}{\partial x^2} = \frac{\theta_{j-1} - 2\theta_j + \theta_{j+1}}{h^2} + \mathcal{O}(h^2) \quad \text{and}$$
$$\frac{\partial^4\theta}{\partial x^4} = \frac{\theta_{j-2} - 4\theta_{j-1} + 6\theta_j - 4\theta_{j+1} + \theta_{j+2}}{h^4} + \mathcal{O}(h^2).$$

The boundary at infinity is replaced by a boundary at some moderately large value $x = Nh$, where the same boundary conditions are assumed as before. At each of the boundaries $x = 0$ and $x = Nh$, the second-order boundary conditions for θ are implemented by introducing one fictitious grid point x_{-1} or x_{N+1}. However the initial and boundary conditions on an isotropic material for the grain boundary with constant dihedral angle, together imply a quadratic equation for $\theta_{-1}(0)$:

$$-3m\theta_{-1}^2 + 4(1+m^2)\theta_{-1} - 8m(1+m^2) = 0.$$

This does not have a real solution unless $m < 1/\sqrt{5}$. This is despite the finite real solution of the original continuum model having been constructed for an arbitrary groove slope, even for an infinite slope of a cuspoid groove. This problem with finite differencing [6] arises only for higher-order nonlinear PDEs with initial and boundary conditions with a corner singularity at $(x, t) = (0, 0)$. The boundary and initial conditions for the integral y of θ do not have such a singularity. The flux condition at $x = 0$ is now third-order in $y(x, t)$, for which we incorporate two fictitious grid points at $x = -h, -2h$. The boundary and initial conditions now imply a unique solution for $y_{-1}(0)$ and $y_{-2}(0)$. The real solution can be constructed for all later times, except that the zero-flux boundary condition at a finite value of x eventually leads to an unacceptable errors.

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