203d Analysis of Flow-Induced and Impurity-Induced Step-Bunching Instabilities during the Growth of Crystals from Liquid Solutions

Bing Dai and Jeffrey J. Derby

During solution growth, crystal quality is often compromised by morphological instabilities, such as macrosteps, step bunches, and inclusions, which arise from the coupled effects of fluid dynamics, mass transport, impurities, and the growth of atomic layers (steps) across a vicinal crystal surface. The main motivation for the work presented here is the need for a coupled, multi-scale, transport-kinetic model which is sufficiently detailed to capture microscopic step growth kinetics and is in a sufficiently general form to be incorporated in a global transport model of fluid mechanics.

We present a multi-scale model of the growth of a vicinal crystal surface from a supersaturated liquid solution that couples bulk fluid dynamics with surface step growth. We consider relatively simple flows within boundary layers adjacent to the macroscopic, vicinal surface of a crystal growing from a liquid solution. There is a depletion of solute due to the crystallization at the surface leading to a concentration boundary layer, which is embedded within the momentum boundary layer. At a still smaller scale, we consider a moving group of steps on the crystal surface and construct a growth model involving a series of elementary transport and kinetic processes, starting from material transport in the bulk phase, adsorption and desorption of growth units to and from terraces between steps, followed by surface diffusion and incorporation at discrete step ledges. This phenomenological approach to describe step motion was first proposed in the classical model by Burton, Cabrera and Frank for solution growth. The governing equations are solved numerically by an efficient, moving-boundary, finite element method.

We demonstrate that sufficiently strong flows parallel to step motion destablize step trains via a mechanism consistent with prior linear stability analyses. However, due to the inherent nonlinearity of the system, the behavior of the step train under multiple-mode perturbations in flowing solution is far more complicated. Perturbation modes with longer wavelengths are most likely to lead to this flow-induced instability and step bunches. We also demonstrate that stable step bunches result when stabilizing forces are included in the computations, such as those arising from capillarity or step-strain interactions. Finally, we consider the effects of step-pinning by impurities and show that the waveforms associated with step bunching instabilities are dramatically different from those driven by flow effects.