Reproduction of hysteresis in crystal growth

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Step dynamics is one of the fundamental physical processes of crystal growth. There are numerous steps with atomistic-scale heights on the crystal surface. A growth component such as an atom or molecule that lands on the surface usually migrates laterally until it reaches the steps –or until it desorbs into the mother phase. A growth component that reaches a step becomes incorporated into the crystal, leading to advancement of the step. Every passages of the steps increase the height of the surface by a monolayer (layer-by-layer growth). Therefore, the understanding of what determines the generation of steps and the rate of the advancement is essential to elucidate the crystal growth mechanism.
The step dynamics is significantly affected by the existence of impurities. The growth hysteresis is one of the well-known phenomena induced by impurities [e.g., 1]. Let us consider the crystal growth from aqueous solution. When the solution includes impurities, the crystal growth rate when the supersaturation is increased is different from that when it is decreased. It has been considered that the hysteresis results from the interaction between two effects: (i) retardation of the step advancement by impurities adsorbed on the crystal surface, and (ii) prevention of the impurity adsorption by frequent step passages. In previous theories of the growth hysteresis, the physical quantities such as the density of adsorbed impurities and the step velocity were averaged both in space and time (mean-field theory [e.g., 2-4]). However, these quantities differ from the position to position on the crystal surface and vary with time in actual situations, so it was not obvious whether the mean-field theory is applicable to the actual systems or not.
In this meeting, we report the results of the numerical simulations of growth hysteresis, which was recently accepted for publication in Crystal Growth and Design [5]. Recently, we have developed a new numerical scheme to simulate the step dynamics based on a phase-field (PF) method [6,7]. We introduced the adsorption/desorption processes of impurities to this method based on the Monte Carlo (MC) method in order to simulate the random impurity adsorption and its time variation. We investigated the change in the step velocity during down-and-up cycles of supersaturation, and found that the appearance of the growth hysteresis in all cycles. The average trace of the step velocity agreed with that predicted by the mean-field theory [3]. This is the first result that reproduces the growth hysteresis using the numerical simulations.

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