Crystal growth hysteresis in birth-and-spread mechanism

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Crystal growth hysteresis is a remarkable phenomenon induced by impurities: the crystal growth velocity when the driving force (supersaturation or supercooling) is increased is different from that when it is decreased. It has been considered that the growth hysteresis is caused by slow adsorption of impurities onto the crystal surface. On the surface, there are steps in atomic scale that advance independently due to continuous incorporations of growth units, therefore the surface is grown layer by layer (layer-by-layer growth mechanism). The step advancement is inhibited by impurities adsorbed on the surface. However, the amount of the adsorbed impurities is also affected by the step passages: frequent step passages reduce the amount of adsorbed impurities. The competition between two timescales —impurity adsorption timescale and time interval of surface-updating (terrace exposure time)— causes complex nonlinear phenomena such as crystal growth hysteresis.

The mechanism of the crystal growth hysteresis was elucidated by a mean field theory. Punin & Artamonova [1] and Miura & Tsukamoto [2] formulated the following two effects —inhibition of step advancement by adsorbed impurities (pinning effect) and removal of adsorbed impurities by step passages (impurity sweeping)— to clarify the existence of multiple steady solutions that satisfies both of two formulas at a certain range of supersaturation. In the case of constant step interval, numerical simulations based on a phase-field method demonstrated that the growth hysteresis emerges as predicted by the mean field theory even when the physical quantities are fluctuated around these averages [3]. The phase-field simulations also confirmed the emergence of hysteresis in the case that the surface is grown by a screw dislocation [4]. However, the growth hysteresis has not been investigated in the case that the surface is grown by two-dimensional nucleation (birth-and-spread mechanism), which is another mechanism to generate new steps on the surface. The birth-and-spread growth dominates the spiral growth at high supersaturation, so it is important to elucidate the impurity-induced effect for that. In this study, we theoretically investigate the growth hysteresis in the birth-and-spread growth mechanism.

At first, we formulated the interdependence between the step velocity and the amount of adsorbed impurities based on the mean field theory, in the same way in the case of the spiral growth. As the result, we found that multiple steady solutions exist at a certain range of supersaturation. This suggests that the growth hysteresis can emerge even in the birth-and-spread growth in principle. At second, we carried out the phase-field calculation with the equivalent condition of the mean field theory. We confirmed the emergence of growth hysteresis in down-and-up cycles of supersaturation. In addition, we found that the range in supersaturation at which the growth hysteresis is observed depends on the rate of supersaturation variation: slow change in supersaturation makes the hysteresis behavior unclear.

References:
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