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Investigating protein controls on crystal growth: how competing timescales and electrostatic interactions lead to bi-stable and catalytic growth

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Structural relationships, chemical interactions, and mechanistic impacts of proteins at the surfaces of growing crystals are poorly understood, despite of their central role in directing formation of mineralized tissues. Here we describe results of in situ AFM investigations into the interactions of aspartic acid-rich peptides and proteins with single crystals of calcium oxalates and carbonates. Using specially designed cantilevers, we have obtained true single molecule resolution and directly imaged protein interactions with atomic steps. We show how the slow adsorption dynamics, strong electrostatic interactions and tendency towards clustering peculiar to macromolecules lead alternately to acceleration and inhibition, as well as switching of growth between two distinct states. We provide a mechanistic model for the observed behavior in terms of altered activation energies and competing timescales for macromolecule adsorption and solute attachment.