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34 - Dynamics verses thermodynamics in two-step nucleation

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Nucleation is the process by which a bulk metastable phase undergoes a phase transition to a stable phase via the formation of a local fluctuation (the critical nucleus) of sufficient size to be able to grow spontaneously to macroscopic scale. In the case of "two-step nucleation" (TSN), the first step in the phase transformation process consists of the appearance in the bulk metastable phase of a local fluctuation that resembles an intermediate phase distinct from the stable phase. In the second step of TSN, this intermediate fluctuation undergoes a transition in which the stable phase emerges from within the intermediate phase. Evidence for TSN has been observed experimentally in a wide range of molecular and colloidal systems, including important cases relevant to understanding protein crystallization and biomineralization. In a recent work, the free energy surface (FES) describing TSN as it occurs in a simple 2D model of a metamagnet was evaluated [1]. This FES shows that at a well-defined size for the growing nucleus, the stable phase becomes more stable than the intermediate phase, providing a thermodynamic prediction for the nucleus size at which the second step of TSN begins. Here we identify conditions at which the spontaneous transition within the nucleus occurs at much larger size than predicted by thermodynamics, demonstrating that the system dynamics can have a dramatic impact on how TSN is observed in practice.

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