

Abstract Submitted  
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**Phase transitions in colloidal fluids: Kinetically or thermodynamically controlled?**<sup>1</sup> MIGUEL A. DURAN-OLIVENCIA, PETER YATSYSHIN, Imperial College London, JAMES F. LUTSKO, Université libre de Bruxelles, SERAFIM KALLIADASIS, Imperial College London — In recent years, a flurry of experimental observations suggests that most phase transitions occur in a multistage manner and via intermediate phases. These precursors to the final phase are commonly understood as the local minima of the free energy of the system. Inherently, the classical paradigm of nucleation has no capacity to describe neither the origin nor the role played by these precursors in the nucleation pathway. Here we present a systematic theoretical framework capable of describing the precursor phases in a self-consistent way. We demonstrate that nucleation precursors can appear even in situations involving a single free-energy barrier. This contradicts previous phenomenological approaches, which always characterise intermediate phases as the minima of a complex free-energy landscape. We show that a kinetically-induced mechanism temporarily stabilises an intermediate phase, which thus is not the result of a local minimum of the free energy but a consequence of the entropic cost of cluster formation. Moreover, the appearance of precursors does not seem to influence the overall nucleation time, which is governed by the free-energy barrier. The mechanism uncovered in this study can be used to explain recently reported experimental findings in crystallisation.

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