

## Accepted Manuscript

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PII: S1359-0294(17)30133-4  
DOI: doi:[10.1016/j.cocis.2018.04.002](https://doi.org/10.1016/j.cocis.2018.04.002)  
Reference: COCIS 1193

To appear in:

Received date: 12 November 2017  
Revised date: 12 April 2018  
Accepted date: 18 April 2018

Please cite this article as: Jinhui Tao, Michael H. Nielsen, James J. De Yoreo , Nucleation and phase transformation pathways in electrolyte solutions investigated by in situ microscopy techniques. The address for the corresponding author was captured as affiliation for all authors. Please check if appropriate. Cocis(2018), doi:[10.1016/j.cocis.2018.04.002](https://doi.org/10.1016/j.cocis.2018.04.002)

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## Nucleation and phase transformation pathways in electrolyte solutions investigated by *in situ* microscopy techniques

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Keywords: Nucleation, Phase transformation, *in situ* TEM, *in situ* AFM

### Abstract

Identification of crystal nucleation and growth pathways is of fundamental importance for synthesis of functional materials, which requires control over size, orientation, **polymorph**, and hierarchical structure, often in the presence of additives used to tune the energy landscape defining these pathways. Herein we summarize the recent progress in application of *in situ* TEM and AFM techniques to monitor or even tune the pathway of crystal nucleation and growth.

### Introduction

The study of nucleation is of paramount importance because it represents the seminal event in the development of a new phase. Classical descriptions of crystal nucleation in solution envision a stable nucleus arising from unstable density fluctuations. These create clusters that grow by monomer addition with free energy increasing until the critical nucleus size is exceeded [1]. At this point, continued growth is energetically favorable and proceeds unhindered through the continued attachment of individual growth units [2]. However, the phase transformation pathway may in fact be complex, because the free energy landscape traversed by each growth unit typically possesses many local minima representing different structural states, each of which is separated from the others by barriers of varying height [3,4,5]. These barriers can be finely tuned in the presence of additives, to bias the nucleation pathway towards a specific outcome. For example, during biologically mediated formation of mineralized tissues, nucleation and growth are tightly regulated by surrounding organic matrices [6]. These often elegant biomineral structures are produced by living organisms at ambient conditions, which typically consist of an

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