

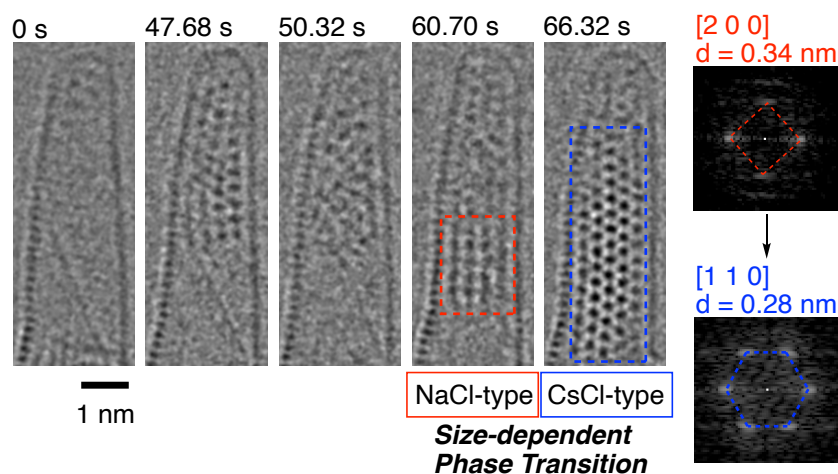
Size-Dependency in Crystal Polymorph Selection of Cesium Chloride

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The mechanism of crystal polymorph selection and its control have attracted much interest because solid-state properties are strongly related to crystal structure. However, the correlation between the size of the clusters and the crystal structures in the crystallization process has not been well studied due to the lack of experimental methods.

Using single-molecule atomic-resolution time-resolved electron microscopy (SMART-EM), which has been developed in our laboratory, the crystallization process of cesium chloride (CsCl) was investigated in detail by continuous video recordings. CsCl takes NaCl-type structure and CsCl-type structure depending on the conditions of temperature and pressure,¹ and we conducted SMART-EM experiments to clarify the size-dependent behavior in crystallization. Crystallization took place in the nano-space of carbon nanotube² shown in the attached figure. In the nucleation, dynamic structural fluctuations of the nucleating cluster of CsCl were observed (0 – 50.32 s). At 60.70 s, NaCl-type structure of CsCl appeared immediately after nucleation and it was transformed into CsCl-type structure in the subsequent growth phase (66.32 s). Two-dimensional FFT analysis gave us quantitative insights in this structural transformation.



1) M. Watanabe *et al.*, *Acta Cryst.* **1977**, A 33, 294. 2) T. Nakamuro *et al.*, *J. Am. Chem. Soc.* **2021**, 143, 1763.