Atomistic Dynamics of "Floating Island" Transiently Formed in Two-Dimensional Nucleation

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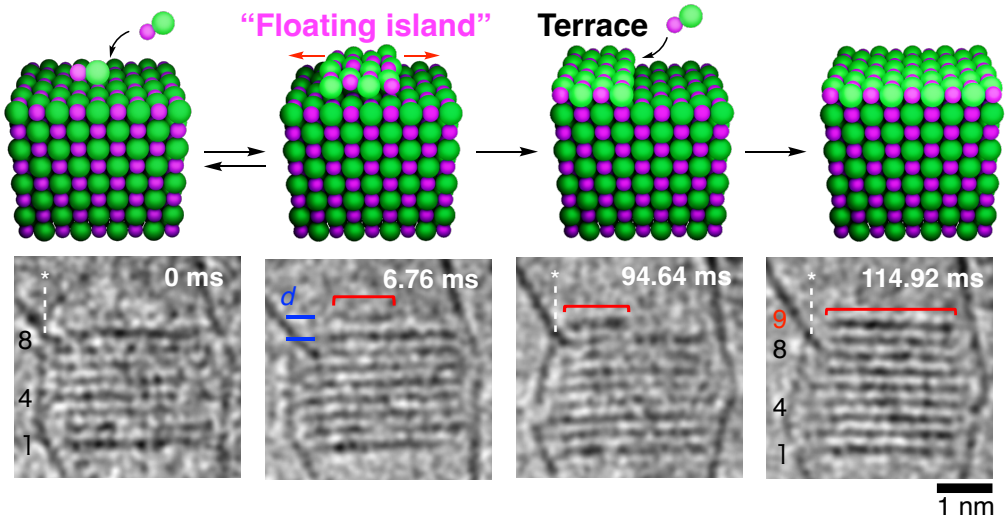
Two-dimensional (2-D) nucleation is the first step of epitaxial growth, in which crystal components adsorbed on the crystal surface assemble to form 2-D nuclei [1]. Since this process determines the direction and rate of crystal growth, detailed understandings of its mechanism are essential for realizing precise control of the properties of resulting crystals. However, the atomistic mechanism of crystal growth has not been clarified due to the lack of observation techniques with high spatial and temporal resolutions. In this context, we have demonstrated direct observation of crystal nucleation of sodium chloride (NaCl) inside carbon nanotube (CNT) by single-molecule atomic-resolution time-resolved electron microscopy (SMART-EM) imaging [2]. Here, we combined this technique with a state-of-the-art high-speed camera (K3-IS) to achieve direct observation of atomistic dynamics in the 2-D nucleation of NaCl [3]. Under SMART-EM observation, the formation of a "floating island (FI)", dynamically diffusing 2-D cluster on a crystal surface with interlayer distance *d* significantly larger than that in a crystal, was recorded (6.76 ms). The existence of such a dynamic intermediate of 2-D nucleation was not assumed in conventional theory. After several tens of milliseconds (94.64 ms), the FI stopped surface migration and landed on the nanocrystal surface to form a stable 2-D nucleus. Then the nucleus rapidly spread on the surface to form a new crystalline layer (114.92 ms). Theoretical calculation suggests that the dynamic nature of the FI originates from shortened Na-Cl bond lengths in the FI. The vessel wall's catalytic activity for forming FIs was also quantitatively evaluated. Nano-space temporally formed between the crystal surface and graphitic wall of CNT effectively hosts NaCl ion pairs and mechanical vibration of the vessel accelerated the incorporation of the FI into the crystal surface by changing the local environment.



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