Direct visualization of subcritical nuclei in molecular film growth

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The growth and structure of 4,4'-biphenyldicarboxylic-acid (BDA) on Cu(001) at temperatures between 300 K and 400 K was studied by LEEM and μ-LEED. BDA is a linear molecule consisting of two phenyl rings with a carboxylic-acid group at opposite ends. During growth on Cu(001) the adsorbed BDA molecules form first a disordered 2D gas phase. Once this phase reaches a sufficiently large density, a crystalline phase nucleates, in which the molecules form a hydrogen-bonded 2D supramolecular network. By a careful analysis of the LEEM bright-field image intensity we can measure the density of the 2D gas phase, which is up to 40% of that in the crystalline phase. From the equilibrium densities at different temperatures we can construct the 2D phase diagram and extract the cohesive energy (0.35 eV). During the distinct nucleation period we can observe a fascinating phenomenon: sub-critical nuclei form, grow up to 4000 nm² in size and decay with lifetimes of several seconds. These sizes are considerably larger than what is usually seen in epitaxial growth and we explain this observation with the relatively weak intermolecular interactions.


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