

Electrochemically-driven metal nanoparticle nucleation and growth

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Electrochemical deposition is a versatile tool to grow controlled surface architectures and thin films for a variety of applications ranging from electronics to catalysis. This technology is centuries old, however, a detailed molecular level picture of the initial stages of nucleation has yet to emerge. Established electro-nucleation theories typically assume that, once a particle has reached a critical radius, subsequent growth occurs irreversibly to the particle rather than to the electrode (Vollmer-Weber growth). We report a combination of atomic-resolution transmission electron microscopy (TEM), scanning electrochemical cell microscopy, Atomic Force Microscopy, and Density Functional Theory to establish a picture of unprecedented detail in microscopic length and time resolution of initial Gold nanoparticle formation on a diamond electrode. At hand of a simplified diamond surface model, we discuss in detail the competition between thermal and electrochemically-driven adatom mobility and the coexistence of various elementary processes, which cannot be fully reconciled with existing electro-nucleation theories.