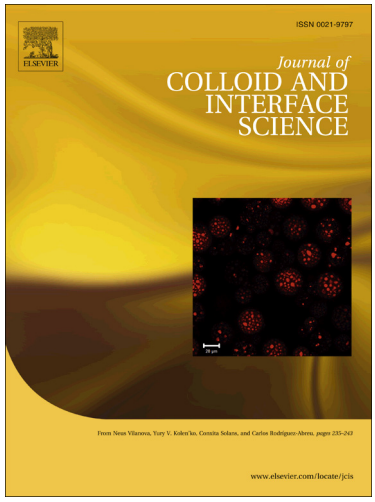

Atomistic modeling of nanoparticle generation in short pulse laser ablation of thin metal films in water

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Abstract

Laser ablation in liquids is actively used for generation of clean colloidal nanoparticles with unique shapes and functionalities. The fundamental mechanisms of the laser ablation in liquids and the key processes that control the nanoparticle structure, composition, and size distribution, however, are not yet fully understood. In this paper, we report the results of first atomistic simulations of laser ablation of metal targets in liquid environment. A model combining a coarse-grained representation of the liquid environment (parameterized for water), a fully atomistic description of laser interactions with metal targets, and acoustic impedance matching boundary conditions is developed and applied for simulation of laser ablation of a thin silver film deposited on a silica substrate. The simulations, performed at two laser fluences in the regime of phase explosion, predict a rapid deceleration of the ejected ablation plume and the formation of a dense superheated molten layer at the water-plume interface. The water in contact with the hot metal layer is brought to the supercritical state and transforms into an expanding low density metal-water mixing region that serves as a precursor for the formation of a cavitation bubble. Two distinct mechanisms of the nanoparticle formation are predicted in the simulations: (1) the nucleation and growth of small (mostly $\leq$ 10 nm) nanoparticles in the metal-water mixing region and (2) the formation of larger (tens of nm) nanoparticles through the breakup of the superheated molten metal layer triggered by the emergence of complex morphological features attributed to the Rayleigh-Taylor instability of the interface between at the superheated metal layer and the supercritical water. The first mechanism is facilitated by the rapid cooling of the growing nanoparticles in the supercritical water environment, resulting in solidification of the nanoparticles located in the upper part of the mixing region on the timescale of nanoseconds. The computational prediction of the two mechanisms of nanoparticle formation yielding nanoparticles with different characteristic sizes is consistent with experimental observations of two distinct nanoparticle populations appearing at different stages of the ablation process.