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Cavitation bubble dynamics and nanoparticle size distributions in laser ablation in liquids

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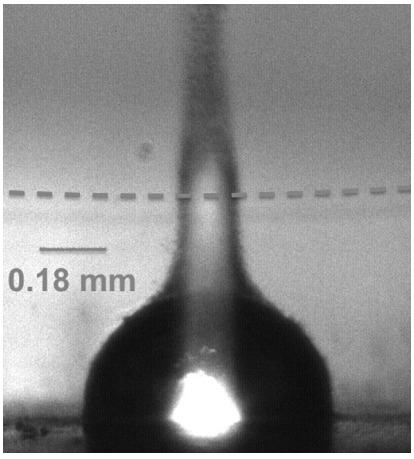
HIGHLIGHTS

- The height of the liquid column is found the strongly affect the cavitation bubble dynamics in laser ablation in liquids.
- The cavitation bubble collapse time determines the size distribution of nanoparticles produced.
- The results are found to be in good agreement with simulations of laser ablation in liquids.

GRAPHICAL ABSTRACT



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ABSTRACT

The influence of cavitation bubble dynamics is investigated during nanoparticle synthesis by *ps* laser ablation of *Au* targets in distilled and deionized water. The height of the liquid column and the ambient pressure is found to strongly influence both the maximum bubble radius and the collapse time, and thus the nanoparticle size distribution. The bubble radius is fit to the Rayleigh–Plesset equation and the results are compared to earlier studies with *ns* lasers. These results are interpreted in the light of recent computational work and the strong body of experimental evidence that shows the fundamental role the central cavitation bubble plays in determining the size distribution of nanoparticles in laser ablation in liquids.

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1. Introduction

Laser ablation in liquids (LAL) is a technique used to produce colloidal nanoparticle dispersions by focusing an intense laser beam onto a target surface immersed in liquid. Despite a wealth of experimental results (*c. f.* [1]), there is significantly less in the way of theoretical/computational framework to understand the separate influence of liquid properties, ambient conditions, materials properties, and laser parameters in LAL [2,3]. It is a very challenging problem to computationally model as there is all the attendant complexity of laser ablation with the added challenges of modeling a highly confined plasma and plume hydrodynamics in a dense medium. When one adds the cavitation bubble dynamics that occur immediately over the ablated laser spot, it is understandable why computational studies lag somewhat behind experimental results. Itina, Povarnitsyn, et al., have provided a comprehensive overview of the first nanosecond following the arrival of a 100 fs pulse [4,5], but it is clear that molecular dynamical simulations must be linked with continuum models to capture the rich variety of length and time scales present during this process. Such knowledge is key to the control of size and shape necessary for application-related work in this area [6].

Understanding the role of the cavitation bubble and the target–bubble interaction during collapse appears to be key to manipulating the size and shapes of nanostructures formed during the LAL process [2,7]. Ibrahimkutty, Wagener, et al., have promulgated the idea that one must control the cavitation bubble dynamics if one wishes to specify the size distribution of nanoparticle produced by LAL [8,9]. Through the use of small X-ray and light scattering, they have shown that nascent nanoparticles form within the central cavitation bubble and that the dispersity index of the particles is greatly influenced by the rate of collapse of the bubble. In particular, they find that particles are re-deposited on the target surface during ablation with a *ns* laser and that they can be irreversibly fused to produce larger aggregates in the high temperature and pressure of the collapsed bubble. Since their measurements are occurring *within* the bubble, they can obtain an accurate assessment of the nanoparticle size distribution that is not influenced by laser-nanoparticle interactions that complicate the understanding of ex-situ analyses.

In the present work, we have either utilized a small chamber as shown in Fig. 1 in which we have varied the ambient pressure within the chamber from 20 kPa to 180 kPa, or we have placed the target on an elevated platform to specify the height of the liquid column over the target. The volume of liquid was kept constant and a stir-bar was utilized to minimize screening from highly concentrated nanoparticles right above the target surface.

While the pressure range used in this study is admittedly narrow, the maximum cavitation bubble radius and the collapse time are observed to change by a factor of more than two. This change in bubble dynamics strongly influences the nanoparticle size distribution obtained from LAL and the results can be understood in terms of the models that follow from the Rayleigh–Plesset equation [10,11].

2. Results and discussion

2.1. Experimental details

The procedure for obtaining particles has been extensively described previously [13]. Briefly, a 1064 nm laser with a 25 ps pulse duration and 250 Hz repetition rate (Ekspla PL-2241) was focused onto an *Au* target immersed in distilled and deionized (DDI) water ($R > 18\ M\Omega$). 4.7 mJ laser pulses with a beam waist of 100 μm (determined via knife-edge method) were utilized, leading to a fluence of 20 J/cm^2. The platform was rastered to avoid excessive structuring of the target. Ablation occurred for 2 min and the solutions were immediately scanned to obtain UV–Vis spectra (Cary 20). Individual drops of solution were placed on transmission electron microscope (TEM) (Zeiss 902) grids where they evaporated to leave behind nanoparticles. To obtain histograms, 400 particles were counted for each sample with ImageJ software. Finally, images of the cavitation bubble were obtained in the same fashion as earlier work [13,14].

2.2. Experimental results

2.2.1. Influence of liquid height

. If the liquid height above the target is increased as shown in Fig. 2, we see that the nanoparticle diameter increases up until a value of 8 mm and remains roughly constant after. As in our previous studies [13], we plot the geometric mean and the error bars reflect the same range as a normal distribution plus or minus one standard deviation would. While Ibrahimkutty et al. [9] argue for the use of the mass-weighted average, we believe that the geometric mean most accurately captures the average particle size and we do not observe enough large nanoparticles to assert the existence

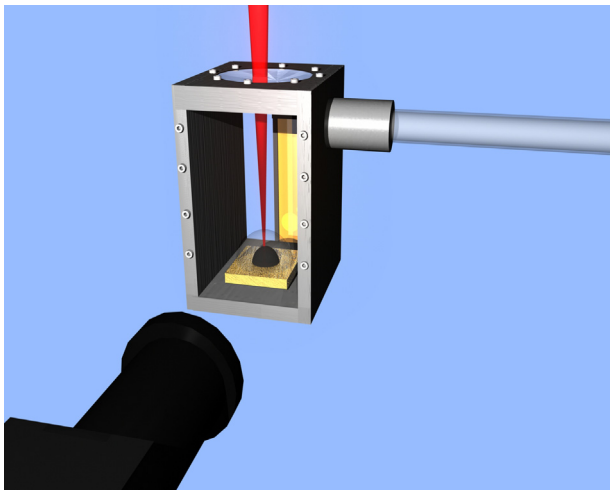

Fig. 1. Schematic of chamber used in these experiments. The camera lens is visible in the left of the frame.

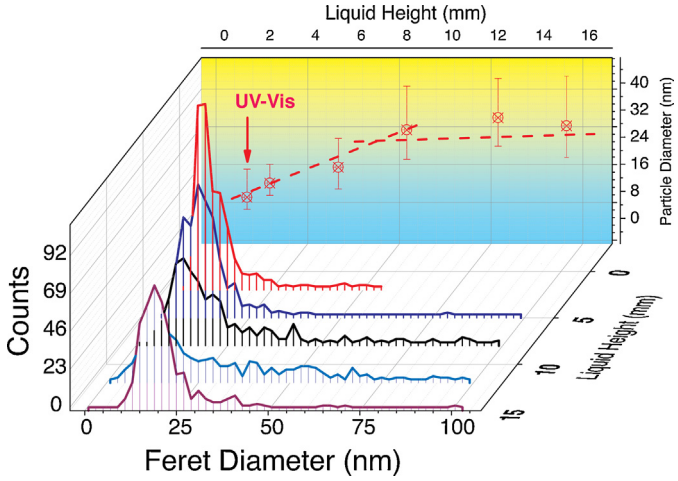

Fig. 2. Nanoparticle diameter and histograms as a function of the height of the liquid column above the target surface. All data points are obtained from TEM measurements except for the first point in the back panel which was obtained by UV–Vis fitting [12]. The lines in the back panel serve as visual guides.

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