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Nanosecond-laser plasma-mediated generation of colloidal solutions from silver films of variable thickness: Colloidal optical density versus predetermined ablated mass

Alena Alexandrovna Nastulyavichus[a,*], Sergey Ivanovich Kudryashov[a,b], Nikita Alexandrovich Smirnov[a], Andrey Andreevich Rudenko[a], Alexander Yurievich Kharin[b,c], Dmitriy Al'bertovich Zayarny[a], Andrey Alexeyevich Ionin[a]

[a] Lebedev Physical Institute, Leninskiy prospect 53, 119991 Moscow, Russia
[b] National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 115409 Moscow, Russia
[c] Lomonosov Moscow State University, 119991 Moscow, Russia

HIGHLIGHTS

- Laser ablation of silver films of different thickness in water.
- Colloidal optical density versus predetermined ablation depth.
- Linear dependence of colloidal extinction on laser intensity.

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ABSTRACT

Metal nanoparticles have attracted much attention due to their unique properties. One of the important aspects is particle size control for the use of their colloids in various applications. Colloidal solutions of silver nanoparticles were generated in water from silver films of variable thickness at different intensities of a nanosecond fiber laser marker (Bulat) on Yb3+ ions and its scanning speeds over the film surfaces in the laser-plasma ablation regime. The obtained colloidal nanoparticles were characterized by scanning electron microscopy, optical transmission spectroscopy, and dynamic light scattering. A monotonic increase in the extinction coefficient and in the size of the colloidal particles was observed as a function of laser intensity and exposure.

1. Introduction

Ligand-free laser-generated colloidal nanoparticles are basic functional form of nanomaterials in biomedicine, nanophotonics, material science and catalysis etc. [1–3]. Recently, gram-scale per hour (mg/s for the dry weight) laser production of corundum [4], pure metallic (Pt, Au, Ag, Al, Cu, Ti) [5] and selenium [6] nanoparticles in sols was demonstrated, attempting to bring lasers into industry of nanomaterials, nanoelectronics, nanophotonics and nanomedicine. Such high-temperature pulsed laser heating and vaporization (ablation) of raw solid materials at multi-Watt average powers and MHz-repetition rates enables "green" and flexible mass-production of chemically pure colloidal solutions of any possible materials, including covalent ones, in pure solvents, or composite – oxidized, carbonized, alloyed and even "core-shell" nanoparticles in chemically active solutions [7]. Formation of colloidal nanoparticles was related to laser pulse energy-dependent bubble dynamics [8,9], while bubble starting parameters – laser energy and mass inputs, which are governed by laser ablation (or ablative laser plasma via surface screening [10,11]) through different ablation mechanisms – are usually unspecified.

Comparing to femtosecond and picosecond laser irradiation regimes with predominating phase explosion mechanism [12–14], proper choice of generation regimes of colloidal nanoparticles becomes especially important during common nanosecond (ns) laser ablation, when several mechanims are well known for "dry" conditions [13,15–20]. As a function of increasing laser intensity (fluence), dry ns-laser ablation proceeds as:

* Corresponding author.
E-mail address: ganuary_moon@mail.ru (A.A. Nastulyavichus).

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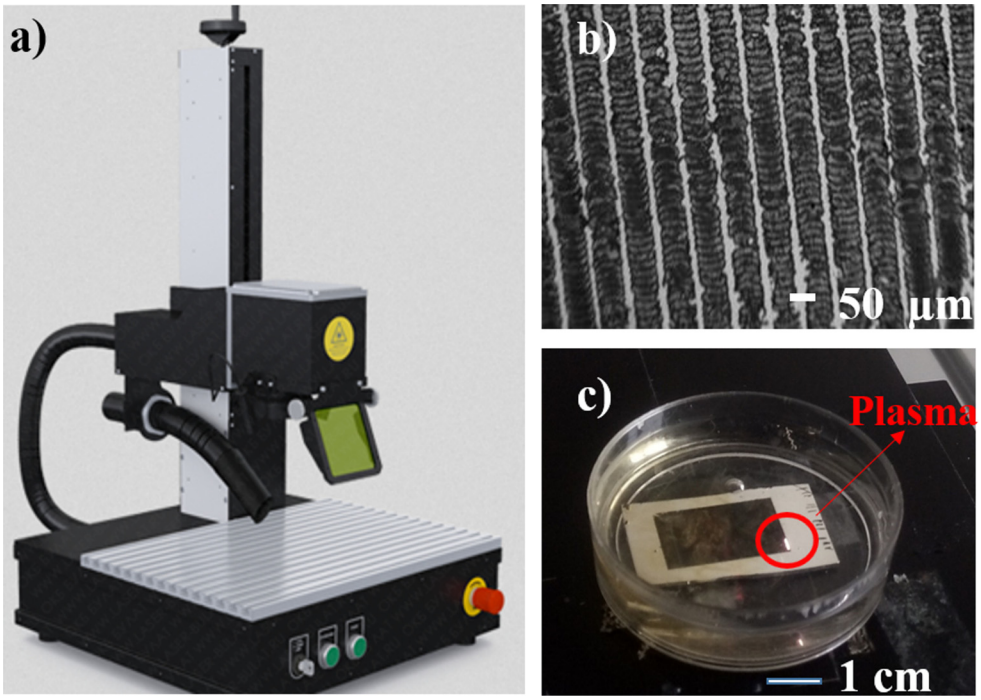

Fig. 1. (a) Experimental setup, (b) the characteristic view of the 85-nm thick silver film surface after ablation, (c) optical image of the ablated region during the laser exposure.

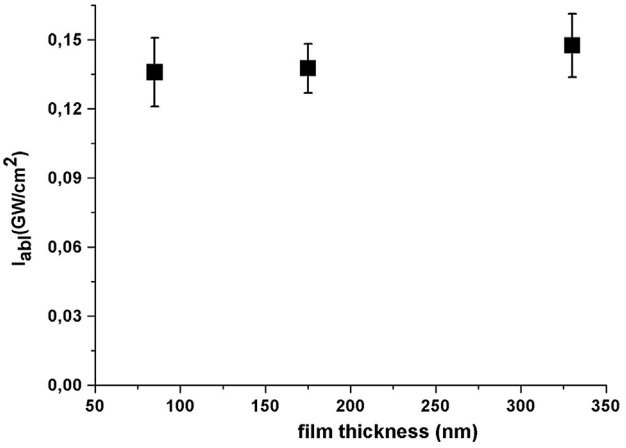

Fig. 2. Dependence of the threshold ablation intensity on the silver film thickness.

(1). surface vaporization of molten materials along (somewhat lower in vapor pressure) their "melt-vapor" binodes, ending up by short condensation of corresponding low-density atomic or small-cluster vapors, in the characteristic ns-laser intensity range ~0.01–0.1 GW/cm^2 (f1uence range ~0.1–10 J/cm^2 for laser pulsewidths of 10–100 ns) [13,21];

(2). surface phase explosion (homogeneous boiling) in the proximity of their "melt-vapor" spinodes, expelling high-density vapor-droplet mixture with its bimodal size distribution, in the characteristic ns-laser intensity range ~0.1–1 GW/cm^2 (f1uence range ~1–100 J/cm^2 for laser pulsewidths of 10–100 ns) [13,15–20,22–23]. This event almost coincides with dense-plume facilitated optical breakdown above the ablated surface, the related onset of sub-critical plasma and ambient shock-wave emission [8,19];

(3). the following sub-critical density plasma regulates in the characteristic ns-laser intensity range ~1–100 GW/cm^2 (fluence range ~10–10^4 J/cm^2 for laser pulsewidths of 10–100 ns) a number of basic laser-matter interaction parameters [24], which are also important during wet ablation – laser energy coupling (fraction) to plasma

$$\eta = 1.7 \times 10^{-6} \frac{\Psi^{9/8} I^{1/2} \tau^{3/4}}{A^{1/4} \mu \lambda^{1/2}} \quad [\%], \tag{1}$$

plasma pressure and surface pressurization

$$P_a = 0.6 \frac{\Psi^{9/16} I^{3/4}}{A^{1/8} \lambda^{1/4} \tau^{1/8}} \quad [\text{Pa}], \tag{2}$$

and ablation rate mediated by plasma through screening

$$\dot{M} = 2.66 \times 10^{-6} \frac{\Psi^{9/8} I^{1/2}}{A^{1/4} \lambda^{1/2} \tau^{1/4}} \quad [\text{g/cm}^2\text{s}], \tag{3}$$

on the surface described by the well-established scaling relationships as functions of material (atomic mass A, average ion charge Z, $\Psi = 0.5A[Z^2(Z+1)]^{-1/3}$) and laser (intensity I [W/cm^2], wavelength λ [cm] and pulsewidth τ [s]) parameters and result in dissociation/ionization of ablation products in the hot plasma core above the surface prior their recombination/condensation [22,23];

(4). finally, deep material melting and superheating by transient bremsstrahlung and recombination plasma emission and its mechanical unloading during plasma adiabatic expansion [25–27] results in intense, microsecond-delayed expulsion of micro-droplets [19,20,28], which is generally not accounted for in Eq. (3).

Meanwhile, possible appearances of these well-known ns-laser ablation mechanisms in mass removal [29], in driving near-surface vapor bubbles [2,8–11,29–31] and, finally, in generation of colloidal nanoparticles under wet ablation conditions [1–3,29] were not firmly observed and justified yet.

In the present work, we have studied the generation of colloidal particles as a function of nanosecond laser intensity and scanning speed, for the first time using thin (in this case, silver) films of variable thickness. This allowed the first time to trace the output of colloidal nanoparticles in correlation with their size and quantity for the predetermined mass of ablation.