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ScienceDirect

Pacific Science Review 16 (2014) 183–188

www.elsevier.com/locate/pscr

Novel hydrodynamic instability of the molten Au/Pd alloy film irradiated by tightly focused femtosecond laser pulses

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Received 9 December 2014; accepted 6 February 2015
Available online 17 March 2015

Abstract

Features of nanoscale surface hydrodynamic instabilities induced on the thin Au/Pd alloy films by single femtosecond pulses shaped into the tightly focused quasi-Bessel beam by means of a fiber microaxicon were studied. The thickness of the metal film as well as the pulse energy were found to be the key parameters determined the types of the hydrodynamic instability of the molten film, which results in the formation of different frozen surface relief microstructures: nanojets, nanocrowns and hybrid structures. Single nanojets forms on the surface of the 80-nm-thick film at the pulse energies ranging from 6 to 7 nJ, while for thicker films formation of hybrid structure, a nanojet surrounded by the nanocrown, is observed. Single nanocrown can be fabricated at film thicknesses ranging from 120 to 240 nm at near-threshold pulse energies. The number of nanospikes of the nanocrown was found to be linearly dependent on the pulse energy and the inverse film thickness.

Keywords: Laser-assisted nanostructuring; Femtosecond pulses; Fiber microaxicon; Molten film; Hydrodynamic instability

Introduction

Laser-driven instability of a molten metal film surface under the action of tightly focused single nano-, pico- and femtosecond laser pulses leads to the formation of a variety of unique sub-micron surface structures [1–4]. Among such laser-induced microstructures nanojet (also referred to as nano-waterdrops), a single standing sharp spike of the molten material with a submicron particle atop, and nanocrown representing a set of sharp nanosized spikes with the sub-100 nm nanoparticles atop [4] periodically placed along the interface of the molten and unmodified metal film attract the great researcher's attention. These structures show a local electromagnetic fields enhancement [5], resonant optical absorption [6], enhanced photoemission [6], etc. Unique properties along with the versatility, high efficiency and low cost of laser "up-down" fabrication techniques make these structures very promising candidates for practical application as optical nanoantennas for manipulation of the electromagnetic fields at

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Peer review under responsibility of Far Eastern Federal University, Kangnam University, Dalian University of Technology, Kokushikan University.

http://dx.doi.org/10.1016/j.pscr.2015.02.005

nanoscale, sensitive elements in the plasmonic refractive index sensors, the functional elements of micro- and nanoelectronics vacuum devices for enhanced electron yield, substrates for SERS signal amplification, etc. It is noteworthy that the geometric shape of the frozen laser-driven instabilities (nanojets and nanocrowns) resembles the impact of the falling water drop on the water or other liquid surface detected by using the high-speed camera [7], which presumably indicates the similarity of the formation mechanisms occurring both in the aqueous layer and the molten metal film heated by the focused ultrashort laser pulse. While the particular instability type in the aqueous layer is determined by the physical properties and the thickness of this layer, ambient conditions, impact velocity and drop size, the nanojets and nanocrowns formation under the action of focused laser pulses is determined by the properties and the thickness of the metal film, pulse energy, as well as the focal spot size [1–8]. Nevertheless, by varying these experimental parameters so far only two types of laser-driven surface instabilities of the melt metal film were experimentally obtained, while a number of experiments in fluid dynamics demonstrate the formation of more complex types of instabilities (Pelegrine Sheet, Crown Splash, Microdroplet Splash, as well as their intermediate cases) pointing out the possibility to obtain novel laser-induced structures with new unique properties.

In this paper, we experimentally demonstrate for the first time that the irradiation of the Au/Pd alloy film by single tightly focused femtosecond pulses under appropriate experimental conditions leads to the a novel laser-induced instability of the molten metal film, which results in the formation of a hybrid microstructure resembling both the nanojet and the nanocrown. In this paper we will show that the thickness of the metal film along with the pulse energy are the key parameter determined the type of obtained laser-induced nanostructures. Underlying mechanisms responsible for the formation of these laser-induced nanostructures are also discussed in this paper.

Experimental details

In our experiments to create a laser-induced instabilities of the surface relief of the metal film, the second harmonic (λ = 400 nm) of a commercial femtosecond laser system (Tsunami Femtosecond Oscillator and Spitfire Amplifier, Spectra Physics) generated 80-fs pulses with a maximal frequency up to 1 kHz is used. Thin Au/Pd films (wt. 80/20%) of variable thickness (80, 120, 160 and 250 nm) were deposited on the smooth cleaved fiber endface by magnetron sputtering with the average deposition rate ~ 0.17 nm/s. Laser pulses were focused on the Au/Pd film surface by a fiber microaxicon (FMA) made on the flat endface of the 5-mm long section of the commercial optical fiber (optical core diameter ~ 1.5 μm) by using a modified chemical etching [9].

FMA's geometric parameters (full cone angle $\theta \approx 110°$ and the cone base diameter D = 2 μm, (Fig. 1(b)) were optimized to provide tightly focused laser spot with lateral FWHM diameter ~ λ and the focus depth ~3.5 λ at λ = 400 nm, thus ensuring the ability to move the FMA during the laser modification process at a distance from the metal film surface sufficient for unobstructed growth of the nanojets and nanocrowns (in our experiments, maximal nanojet's height does not exceed 1 μm). The laser spot at the FMA output represents a central symmetrical quasi-Bessel beam (Fig. 1(c)), surrounded by an additional low-intensity maximum, which is not involved in the laser modification process. The sample was placed on a linear motorized nanopositioners system (Newport XM series), which provides a 50-nm movement precision along three axes. The FMA is placed perpendicularly to the metal film surface, while the probe-to-sample distance (in this case ~ 3 λ) was maintained at a constant level using a tuning fork feedback system. All laser-induced structures were fabricated by single-pulse irradiation under ambient conditions and post-characterized using scanning electron microscope (SEM, Hitachi S3400N).

Results and discussions

The result of the single femtosecond pulse impact on the 80-nm-thick Au/Pd film is demonstrated in Fig. 3 for different pulse energies E. The visible modification of the metal film by a femtosecond pulse occurs at the pulse energy ~3 nJ representing a molten area with recrystallization grains of metal nanocrystallites.

The lateral size of the molten area (~600–700 nm) is about half times larger than the initial optical spot (~410 nm) on the film surface, which presumably indicates the sufficient lateral heat transfer during the heated laser pulse. Note that significant exfoliation of the metal film on the glass substrate at these pulse energies is not observed. However, at further pulse energy increase up to 6 nJ the microbump structure is formed, with the molten material being tended to concentrate mainly at the forming frozen standing