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## Plasmon-mediated enhancement of Rhodamine 6G spontaneous emission on laser-spalled nanotextures

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### Abstract

Biosensing characteristics of the laser-spalled nanotextures produced under single-pulse irradiation of a 500-nm thick Ag film surface were assessed by measuring spontaneous emission enhancement of overlaying Rhodamine 6G (Rh6G) molecules utilizing polarization-resolved confocal microspectroscopy technique. Our preliminary study shows for the first time that a single spalled micro-sized crater covered with sub-100 nm sharp tips at a certain excitation conditions provides up to 40-fold plasmon-mediated enhancement of the spontaneous emission from the 10-nm thick Rh6G over-layer indicating high potential of these easy-to-do structures for routine biosensing tasks.

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*Keywords:* nanosecond pulses; metal films; Ar ion polishing; functional plasmonic nanostructures; nanorings; nanorods; hybrid nanostructures

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### 1. Introduction

Metal nanostructures made of noble metals have extensively studied during past three decades owing to their unique ability to localize and enhance the electromagnetic energy at nanoscale volumes (Maier S.A. (2007)), providing an increasing need for cheap and high-performing fabrication techniques, in its turn. In this lieu, laser material processing of bulk noble-metal targets and their thin films with tightly focused femtosecond (fs) laser

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pulses was proven to be an unrivalled tool for “green”, controllable, high-throughput and easy-to-do fabrication of different individual nano-elements (Zywietz U. et al. (2014), Kuchmizhak A.A. et al. (2016)), highly-ordered or disordered nanotextured and nanopatterned surfaces (Makarov S.V. et al. (2016)), colloidal nanoparticle solutions (Kabashin A.V. et al. (2003)), which can be readily applied for plasmonics, biosensing, photovoltaics, chemical catalysis, nanophotonics, etc.

The majority of studies were considered the case of single-pulse ablation of “thermally thin” (usually, thinner than 150 nm) noble- and semi-noble metal films covering low transparent heat-insulating substrate under tight focusing irradiation (Koch J., et al., (2005)). Such experimental conditions provides strong lateral conductance of absorbed heat in the excited metal film as well as its uniform distribution along the film thickness during the two-temperature stage, resulting in its homogeneous melting and subsequent formation of well-known surface nanostructures, namely nanobumps, nanovoids, nanojets, micro- and nanosized holes, etc. The applications of these nanostructures for plasmonics (Reininghaus M. et al. (2013)), biosensing (Kuznetsov A.I. et al. (2011), Chen L. et al. (2014)), field-emission devices were shown in numerous of papers. To the contrary, both experimental studies (Savolainen J.M. et al. (2011), Zayarny D.A. et al. (2015)) and theoretical predictions (Ashitkov S.I. et al. (2012)) of the bulk targets ablation under sub-threshold excitation conditions reveal the pronounced material swelling (Savolainen J.M. et al. (2011)) accompanied by formation of surface nanoholes and subsurface nanovoids indicating distinctly different ablation character associated with inertial “stress confinement” near the surface layer (Wu C. et al. (2015)) or even subsurface boiling process (Ionin A.A. et al. (2016), Kuchmizhak A.A. et al., (2014)). The cumulative action of these processes leads to spallation of the molten layer at increased fluence accompanied by appearance of ablative nanofeatures – nano-protrusions, nanotips, nanopores and nanobridges – densely packed into disordered nanopatterned areas.

Meanwhile, potential applications of such nanopatterned area for biosensing were highlighted in recent publication (Ionin A.A. et al. (2016)), to the best of our knowledge, no direct attempts were made to measure plasmonic response of such nanotextured surfaces as well as to justify their applicability for routine biosensing tasks. In this preliminary study, to the best of our knowledge, we make first attempt to assess biosensing characteristics of single micro-sized nanotextures produced under irradiation of the 500-nm thick silver films on a thermally isolated glass substrate with single femtosecond (fs) laser pulses. To do this, we have examined the features of the spontaneous emission of Rhodamine 6G (Rh6G) molecules covering produced nanotextures using polarization-resolved confocal microspectroscopy technique. Our study shows that at a certain nanotexture geometry and excitation conditions one can achieve up to 40-fold plasmon-mediated enhancement of the Rh6G spontaneous emission indicating great biosensing potential of the laser-spalled surfaces.

## 2. Methods

### 2.1. Film deposition

We have deposited 500-nm thick silver films onto optically smooth bulk glass substrate at the pressure of  $5 \cdot 10^{-6}$  bar and the average rate  $\sim 1$  nm/s by an e-beam evaporation procedure (Ferrotec EV M-6) while rotating the sample holder for uniform deposition and applying no additional adhesive sublayers. The substrate was pre-cleaned with a build-in  $\text{Ar}^+$  source (KRI EH200) for 5 min to increase adhesion of the metal films. The resulting film thickness was measured by a calibrated quartz crystal microbalance (Sycon STC-2002) and verified by atomic force microscopy.

### 2.2. Laser processing

Nanopatterning of the metal films was performed with 200-fs 400-nm pulses generated by Ti: Sapphire laser system, comprised by an oscillator and an amplifier (Tsunami and Spitfire, Spectra Physics). The laser pulses were focused onto the sample surface by lens (NA=0.3, Nikon) yielding in a Gaussian-like focal spots with the characteristic radius  $R \sim 1.22\lambda \cdot (2NA)^{-1} \sim 1.08 \mu\text{m}$ . The samples were arranged on a PC-driven three-dimensional motorized micropositioning platform (Newport XM series) providing a minimal translation step of 50 nm along each axis. The pulse energy (E) was varied by means of a variable energy filter and measured by a sensitive pyroelectric photodetector. The topographical features of the nanopatterned surfaces were studied by means of a scanning electron microscope (SEM, Hitachi S3400).

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