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## Single-shot laser-assisted nanofabrication of plasmonic nanorings

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### Abstract

Simple high-performing two-step technique for fabrication different functional plasmonic nanostructures including nanorods, separated and crossed nanorings, as well as more complex hybrid structures on both glass and silicon substrates was proposed. In this technique the noble metal film covering bulk glass or silicon substrates is irradiated by single tightly focused nanosecond laser pulse followed by slow polishing of the fabricated nanostructures by accelerated argon ion ( $\text{Ar}^+$ ) beam. Nanosecond laser pulse locally modifies its initial thickness of metal film through the initiation of ultrafast melting and subsequent hydrodynamic processes, while the following  $\text{Ar}^+$  polishing reveals only the features of its topography - plasmonic structures on the glass/Si substrate. We demonstrate that both the type and lateral size of the resulting functional plasmonic nanostructure are determined by the pulse energy, metal film thickness as well as the optical spot size, while subsequent  $\text{Ar}^+$  polishing allows varying the height of the resulting nanostructures. The proposed simple two-step high-throughput technique represents the next step towards direct laser-induced fabrication of complex functional plasmonic nanostructures and is well-suited for both large-scale fabrication of ordered arrays comprising hundreds of nanoelements and single nanostructure at a given point on the sample surface.

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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

Functional plasmonic nanostructures (FPN) arranged into ordered arrays and fabricated on a transparent dielectric substrate are the objects of increasingly growing scientific interest in the last decade. Such structures exhibit unique optical and spectral properties allowing manipulation and amplification of the incoming optical radiation at nanoscale through excitation of coherent electron plasma oscillations referred to as localized plasmon resonances (LPRs). Many promising applications of LPRs in such rapidly developing fields of science as biosensing, photovoltaics, nanophotonics, near-field optical microscopy and nanomedicine were theoretically predicted and experimentally demonstrated in the past two decades (Schuller J.A. et al. (2010)). Modern scientific problems arising in these scientific areas impose stringent requirements on the FPN fabrication techniques. Such fabrication techniques must combine the ability to wide-range control over the geometric dimensions of each individual nanoelement in the ordered array with high performance and repeatability, relatively low cost of the fabricated FPN arrays as well as the possibility to fabricate a single nanostructure at a given point on the sample surface, for example at the end of an optical fiber or the scanning probe tip. Advanced bottom-up approaches utilizing direct impact of the focused accelerated ion beam or combination of electron milling and subsequent post-processing steps can effectively solve the problem of single FPN fabrication or small array of such nanoelements with very high precision down to angstrom level. However, these approaches become extremely expensive and time-consuming, when the number of nanoelements in the FPN array reaches  $10^4$ - $10^6$ .

Meanwhile, to date the applicability of the direct laser nanostructuring technique was limited to fabrication of relatively simple nanostructures (spherical nanoparticles, 2D surface gratings or nanojets), remaining the fabricated structures attached to the unprocessed metal film. Additionally, rather expensive femtosecond lasers are often used for this purpose. In this paper, we demonstrate for the first time that the use direct single-shot exposure of the metal film by tightly focused nanosecond (ns) laser pulse followed by slow polishing of the fabricated nanostructures by accelerated argon ion ( $\text{Ar}^+$ ) beam allows one to fabricate arrays of isolated plasmonic nanorods, separated and crossed nanorings, as well as more complex hybrid structures on both glass and silicon substrates. In this approach, the ns pulse irradiated metal film locally modifies its initial thickness through the initiation of ultrafast melting and subsequent hydrodynamic processes, while the following  $\text{Ar}^+$  polishing reveals only the features of its topography - plasmonic structures on the glass/Si substrate. We demonstrate that both the type and lateral size of the resulting FPNs are determined by the pulse energy, metal film thickness as well as the optical spot size, while subsequent  $\text{Ar}^+$  polishing allows to vary the height of the resulting nanostructures. The proposed simple two-stage high-throughput technique represents the next step towards direct laser-induced fabrication of complex nanostructures and is well-suited for the fabrication of both large-scale ordered arrays comprising hundreds nanoelements and single nanoantenna at a given point on the surface of the sample.

## 2. Methods

In our experiments, laser nanostructuring of the noble metal (Au, Ag, Cu) films of variable thickness on glass/Si substrate was carried out using second-harmonic (532 nm) linearly-polarized pulses of the Nd:YAG laser (Quantel Ultra) (Kuchmizhak A.A. et al. (2014)). The output radiation from the laser via the fiber coupler containing the micrometer positioner and the lens ( $\text{NA} = 0.25$ ) was inputted into a segment of single-mode optical fiber (Thorlabs SM405) providing the radiation filtering into the spot with a nearly Gaussian intensity distribution profile, and then was focused onto the sample surface by a high-NA lens ( $\times 40$ ,  $\text{NA} = 0.6$ ) under complete filling of their input aperture providing the irradiation of the samples by laser pulses with almost perfect lateral energy distribution profile at the focal spot with the size  $R_{\text{opt}} = 1.22\lambda \cdot (2\text{NA})^{-1} \sim 0.34 \mu\text{m}$ .

The sample was arranged on a PC-driven motorized micropositioning platform (Newport XM series) with a minimal translation step of 50 nm along three axes and moves from pulse to pulse. The pulse energy  $E$  was varied by means of a tunable filter and measured by a sensitive pyroelectric photodetector. Noble metal film with the thicknesses ranging from 15 to 120 nm deposited onto the optically smooth bulk glass or Si substrates by e-beam evaporation procedure (Ferrotec EV M-6) at a pressure of  $5 \cdot 10^{-6}$  bar and an average speed  $\sim 8 \text{ \AA}/\text{c}$  were used as a samples for laser nanostructuring experiments. To increase the adhesion of the deposited material to the glass substrate, the latter was pre-cleaned by the build-in ion source (KRI EH200). The film thickness was measured at

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