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# Fabrication of porous metal nanoparticles and microbumps by means of nanosecond laser pulses focused through a fibre microaxicon

Alexander A. Kuchmizhak [a,*], Oleg B. Vitrik [b], Yuri N. Kulchin [b]

[a] Optoelectronics Lab., Institute of Automation and Control Processes, FEB RAS, Russia
[b] Far Eastern Federal University, FEFU, Russia

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## Abstract

We present a novel optical element: a fibre microaxicon (FMA) for laser radiation focussing into a diffraction-limited spot with a Bessel-like profile as well as for precision laser nanostructuring of metal film surfaces. Using the developed FMA for single-pulse irradiation of Au/Pd metal films on a quartz substrate, we demonstrated the formation of submicron hollow microbumps, each with a small spike atop it as well as hollow spherical nanoparticles. The experimental conditions for the controllable and reproducible formation of ordered arrays of such microstructures were defined. The internal structure of the fabricated nanoparticles and nanobumps was experimentally studied using both argon-ion polishing and scanning electron microscopy. These methods revealed a porous inner structure of laser-induced nanoparticles and nanobumps, which presumably indicates that a subsurface boiling of the molten metal film is a key mechanism determining the formation process of such structures.



*Keywords:* Laser-assisted nanostructuring; Nanosecond laser pulses; Fibre microaxicon

## Introduction

Nanostructuring using short and ultra-short laser pulses provides opportunities for the fabrication of different micro- and nano-scale structures (nanojets [1,2], nanowhiskers [3], bumps [4], spherical nanoparticles [5,6], through nanoholes [7], nanocrowns [8], etc.) on sample surfaces. Single or periodically arranged laser-induced nanostructures can exhibit the desired properties of local field enhancement and strong plasmonic response [9] and high electron emission [10], which make such nanostructures promising candidates for use in different sensors as well as in nanophotonic and plasmonic devices [11]. Research interest in this area has continued unabated for several decades, largely due to the versatility, non-contact nature, high efficiency and relatively low cost of the laser nanostructuring methods compared with electron- and ion-beam milling methods as well as the

* Corresponding author.
*E-mail addresses:* alex.iacp.dvo@mail.ru (A.A. Kuchmizhak), oleg_vitrik@mail.ru (O.B. Vitrik), kulchin@iacp.dvo.ru (Y.N. Kulchin).
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possibility to fabricate unique types of nanostructures via laser–matter interaction.

The fabrication of nanostructures onto the sample surface is mainly performed by using nano-, pico-, and femto-second laser pulses focused using classical high-NA focussing optics [1,2,5,6]. The minimal lateral size of the focal spot in this case is limited by a fundamental diffraction limit, which, under ideal conditions, is equal to $\lambda/2$ ($\lambda$ – laser wavelength), i.e., ~200 nm for visible light. Fabrication of sub-100-nm structures using focussing optics is still possible and can be performed despite the diffraction limit effect by using a non-linear threshold response of the modified sample. However, in this case, the requirements for homogeneity and intensity distribution in the focal spot are very high, which often necessitates the use of additional diffraction optical elements. In practice, the achievement of such extreme light localisation with the high-NA optics is a difficult task and requires expensive high-quality focussing lenses. However, several papers [7,12–14] report that the high spatial focussing of the laser radiation into a high-quality focal spot as well as precise positioning of the focal spot on the sample surface can be achieved by using a single optical element – a bare fibre taper (BFT). This element is similar to a standard aperture-type probe of a scanning near-field optical microscopy (SNOM) but differs due to the absence of a metal coating and a nanosized aperture. Such BFTs, despite the reduced focussing capabilities compared with conventional SNOM probes, exhibit a significantly higher throughput and damage threshold and are widely used in precision laser nanostructuring [7], laser-induced breakdown spectroscopy [12], local spectroscopy of quantum objects [13], reflection-mode SNOM [14], etc. To achieve the maximal lateral localisation of the laser radiation by using the BFT, its tip must be shaped into a truncated cone with an upper cone base diameter ~$\lambda/2$ [15]. However, such geometric shape optimisation of the probe tip requires not only an expensive and the time-consuming ion-beam milling, which significantly complicates the fabrication process of probes, but also causes the focussing element to be tied to a specified wavelength of the input laser radiation.

In our previous work [16], we demonstrated that near-$\lambda/2$ lateral localisation of laser radiation can be achieved by using a fibre microaxicon (FMA) fabricated on the flat endface of a single-mode optical fibre (OF) that is axially symmetric to its core. We also modified a chemical etching method to fabricate the FMAs [16] using different commercial OFs. In this paper, we will use for the first time the FMA as a compact, universal and highly efficient optical element for the spatial filtering and focussing of nanosecond laser pulses into a diffraction-limited spot with a Bessel-like spatial distribution as well as demonstrate the single-pulse laser-assisted fabrication of various nanostructures on the surfaces of metal films. We also report the fabrication of submicron bumps through holes and porous spherical nanoparticles as well as discuss the possible underlying formation mechanisms of such micron- and nano-scale structures.

## Experimental details

Linearly polarized third-harmonic ($\lambda$ = 355 nm) pulses of a Nd:YAG-laser with the pulse width $\tau_p$ ~7 ns, maximum energy $E$ ~10 mJ and energy stability ~10% were used for surface nanostructuring (Fig. 1(a)). Spatial filtering of the output laser beam and its focussing into the diffraction-limited spot were performed by means of the FMA (Fig. 1(b))

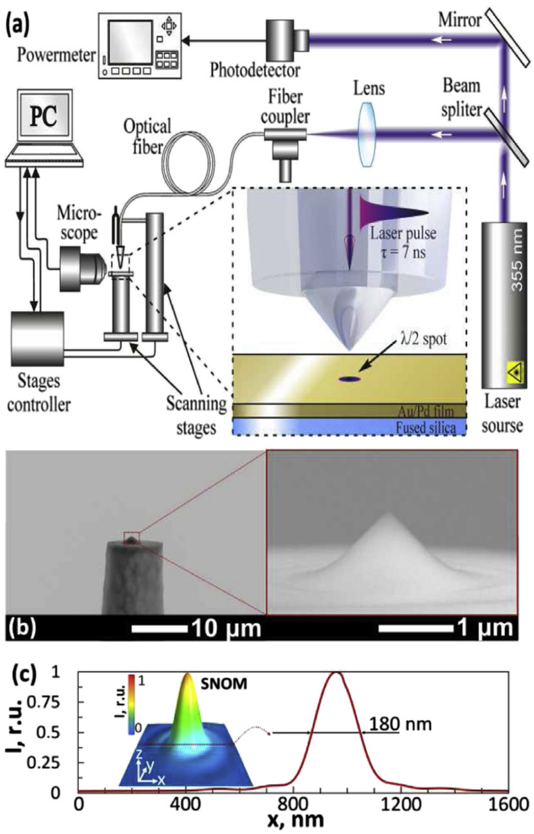


Fig. 1. (a) Experimental setup; (b) SEM image of the fibre microaxicon with a full cone angle $\theta \approx 90°$ and the cone base diameter $D = 2$ μm used for the laser nanostructuring experiments; (c) typical laser intensity distribution measured at the FMA focal plane using the aperture-type collection-mode SNOM.