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Laser-assisted nanostructuring of metal films by means of a fibre dielectric microprobe

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Abstract

A simple apertureless dielectric microprobe in the form of a section of the tapered optical fibre was proposed for surface laser nanomodification. This probe enables surface $\lambda/2$-localisation of laser beam, as demonstrated both numerically and experimentally. The controllable formation of single through nanoholes with the minimum size down to 35 nm (~$\lambda/15$) in the 50-nm Au/Pd film was shown using this probe and a 532-nm pump nanosecond laser. We also report for the first time on the formation of micro- and nanobumps, jet-like microstructures and microholes on optically thick gold films using single nanosecond laser pulses focused through the fibre dielectric apertureless probe. Both the shape and the sizes of the obtained microstructures were demonstrated to be determined by the pulse energy and film thickness.

Keywords: Laser-assisted nanostructuring; Nanosecond laser pulses; Fibre apertureless microprobe

Introduction

Nanoscale interaction of short and ultrashort laser pulses with solid surfaces is of consistently growing scientific and technological interest. Such interaction typically results in the fabrication of single and periodic surface nanofeatures [1,2], bringing new optical properties to the nanomodified surfaces [3]. Sub-100-nm surface nanofeatures are efficiently fabricated by spatial localisation of a driving laser electric field transmitted through an exit hole of a scanning near-field optical microscope (SNOM) probe [4,5]. Such SNOM probes are composed of a single-mode optical fibre (OF) with a conically tapered and metal-coated tip, ending up with a nanoscale exit aperture. A SNOM probe can be moved along the modified surface with a high positioning accuracy of tens nanometre. However, the low optical transmittance through the exit nanoaperture of the SNOM probe (~10^{-6} at the aperture diameter D = 50 nm) [6] decreases the transmitted intensity to well below the modification

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threshold for most of materials. This problem cannot be solved by merely increasing the input energy because this results in strong heating and degradation of the tip due to the high dissipative losses in its metallic coating [7]. Consequently, the output aperture diameter of the SNOM probe, which is used for the surface modification with high energy pulses, typically cannot be made smaller than ~λ [4], where λ is the laser wavelength.

Another laser-based method of formation of surface nanostructures with high spatial resolution consists of using the effect of electromagnetic field amplification near the nanoscale tip [6,8]. An atomic-force microscope probe irradiated with the external laser source can be used, e.g., a nanosized tip [9–11]. A record lateral resolution of ~10 nm was obtained [9] using this method with femtosecond laser pulses. However, the vertical resolution is also limited by this value, which restricts the application of this method.

As a result, high-NA focussing optics are still frequently used to localise laser fields on a diffraction scale ~$\lambda/2$. Nevertheless, sub-100-nm features can be fabricated in such a case via non-linear near-threshold effects (e.g., laser modification or ablation), multi-beam interference and excitation of surface plasmon-polaritons [1–3]. However, these methods require additional diffractive optical elements to obtain a homogeneous focal spot with the required energy distribution. In addition, the addressing of light to a specific point on the sample surface with nanometre precision is significantly complicated because of the rather large dimensions of the far-field focussing element.

One potential solution for sub-100-nm surface nanostructuring could be the combination of the high SNOM positioning accuracy and the high transmittance of far-field optics. In particular, a standard SNOM device can be equipped with a piece of a single-mode fibre with a conical output tip (an apertureless dielectric microprobe, ADM) [12–15]. The homogeneous Gaussian-like spatial distribution of the fibre fundamental mode does not require correction and is well suited for further focussing of the transmitted laser energy, due to total internal reflection at the "dielectric/air" interface, into a $\lambda/2$-spot [12]. Such a probe differs from its apertured SNOM analogue only by the absence of the metallic coating on its tip and exhibits significantly higher optical damage threshold and transmittance than the aperture SNOM probe. As a result, ADMs have been used in laser ablation, photochemical etching, laser-induced breakdown spectroscopy [13–15], etc. However, the fabrication process of the ADM tip is usually very complex [12].

In this paper, we present a simple fibre dielectric probe in the form of a section of the tapered optical fibre with a tip in the form of a truncated cone, as well as demonstrate the applicability of the developed ADM for precise laser-assisted nanostructuring of metal films surfaces and fabrication of various nanostructures with high spatial resolution.

Experimental

Linearly polarized second-harmonic ($\lambda = 532$ nm) pulses of a Nd:YAG laser (Solar LS LQ215) with the FWHM-pulsewidth ~7 ns and maximum energy $E < 10$ mJ in the TEM_{00}-mode were used for surface nanostructuring (Fig. 1(a)). Each p-polarized laser pulse was focused onto the sample surface (Fig. 1(a)) by means of an apertureless dielectric microprobe (ADM), a tapered 20-mm long section of a single-mode optical fibre (Thorlabs SM400) with a constant taper angle ~12° (Fig. 1(b)) and a flat endface with a diameter ~250 nm [16,17]. In accordance to our finite-difference time-domain (FDTD) simulation such an ADM provides spatial filtering and focussing of a laser beam (Fig. 1(c)), resulting in a diffraction-limited output spot ($R_{1/e} \approx 0.3$ μm at normal incidence) with a nearly Gaussian spatial profile and relatively deep focal depth (~$\lambda/2$) [16]. The laser pulses were effectively coupled to the ADM using a fibre coupler (Thorlabs MBT612D/M).

During laser nanostructuring, the ADM tip was located 50 nm above the sample surface and inclined at an angle of 60° (inset in Fig. 1) to the sample surface normal, yielding an elliptical Gaussian spot with $R_{x,1/e} \approx 0.65$ μm and $R_{y,1/e} \approx 0.4$ μm. The probe-to-sample distance was controlled using the tuning fork feedback. Visual control of the ADM motion and observation of the Au/Pd film damage was performed by means of a high-resolution optical microscope Hirox KH7700 (optical magnification ~700–7000×, working distance ~3.4 mm). The laser pulse energy entering the fibre was varied by means of a polarizing attenuator. To measure the pulse energy E at the ADM tip, the output laser radiation was collected by means of an objective (Olympus, NA = 0.65) and focused onto a photodetector (J-10SI-HE Energy Sensor, Coherent EPM2000). The laser-structured films were characterised using an atomic force microscope (AFM, NanoDST Pacific Nanotechnology) in the close-contact mode and using a scanning electron microscope (Hitachi S3400). All