Laser-fabricated dielectric optical components for surface plasmon polaritons

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Fabrication of dielectric optical components for surface plasmon polaritons (SPPs) by two-photon polymerization (2PP) is studied. This direct-write femtosecond laser technology provides a low-cost and flexible method for the fabrication and investigation of plasmonic structures and optical components. Using the 2PP technique, we fabricated narrow dielectric ridges with dimensions as small as 150 nm on metal surfaces. SPP excitation with the laser-fabricated structures and guiding along them are demonstrated. © 2006 Optical Society of America

OCIS codes: 220.4000, 240.6680, 140.3390.

The science of surface plasmon polaritons (SPPs), electromagnetic excitations propagating along and bound to an interface between a metal and a dielectric, has attracted much attention during the past few years.[1–5] Renewed and growing interest in this field comes from a rapid advance of nanostructuring technologies. Using nanostructured surfaces, one can study basic properties and develop possible applications of SPPs. The desired nanostructures are usually fabricated by electron- or ion-beam lithography.

An alternative approach to the rapid and low-cost fabrication of nanostructured surfaces is the application of two-photon polymerization[6–9] (2PP) or maskless nonlinear lithography.[10] Both of these technologies are based on nonlinear absorption of near-infrared femtosecond laser pulses by a photosensitive material. Using 2PP, one can fabricate high-quality three-dimensional micro-objects and photonic crystals with a resolution down to 100 nm upon dielectric transparent substrates.[11,12]

In this Letter we study possible applications of 2PP for the fabrication of narrow dielectric structures upon thin metal surfaces. We demonstrate that these dielectric structures can be used for excitation, guiding, and manipulation of SPPs on a subwavelength scale. Dielectric structures are fabricated upon gold surfaces by 2PP of the commercial inorganic–organic hybrid polymer ORMOCER provided by Microresist Technology. ORMOCER is a liquid and can be polymerized by use of a radical photoinitiator, for example, Irgacure 369. The refractive indices of the uncured and cured materials are slightly different,[13] facilitating online observation of the polymerization process.[11]

For investigations, a commercial femtosecond oscillator, Spectra-Physics Model Tsunami, is used. This system delivers laser pulses at $\lambda = 780$ nm with a pulse duration of $\tau = 80$ fs (FWHM) and repetition rate $\nu = 80$ MHz. In our experiments, average powers up to $P = 40$ mW are applied. Femtosecond laser pulses are focused by a $100 \times$ Nikon microscope objective with a numerical aperture of 1.3. The space be-

tween the microscope objective and the sample is filled with a refractive-index-matching oil ($n_{oil} = 1.515$). For the fabrication of dielectric structures, the ORMOCER droplet is sandwiched between two $18 \text{ mm} \times 18 \text{ mm}$ glass slides of 150 μm thickness. The 100 μm distance between these slides is set by a thin plastic frame. The lower slide of the sample carries a gold layer on its upper side, and the top slide is used to prevent contact with the immersion oil. The gold layers of (50 ± 5) nm thickness are prepared by electron sputtering onto glass substrates. For fabrication of dielectric structures, the laser beam is focused through the upper glass plate and the ORMOCER layer onto the gold surface; see Fig. 1. During structuring, the sample is kept fixed and the beam is scanned through the microscope objective with a galvo-scanner system from Scanlab. In this way, an area of $40 \text{ μm} \times 40 \text{ μm}$ can be structured. After polymerization, 4-methyl-2-pentanone is used to remove the nonirradiated material.

Structuring of ORMOCER onto transparent dielectric substrates by 2PP has recently been studied.[9,11] In contrast, the fabrication of polymerized structures by 2PP directly onto metal surfaces has not been explored so far. Laser-induced polymerization on the reflecting metal surface is more problematic owing to interference effects and poor polymer adhesion to metals. In our investigations we observed that 2PP of ORMOCER and fabrication of dielectric structures
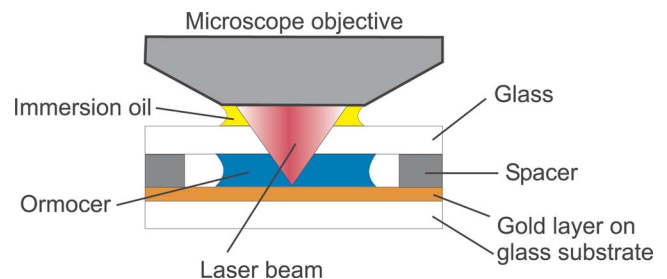

Fig. 1. (Color online) Sample preparation and experimental setup for the fabrication of polymerized ORMOCER structures upon gold-coated glass substrates.
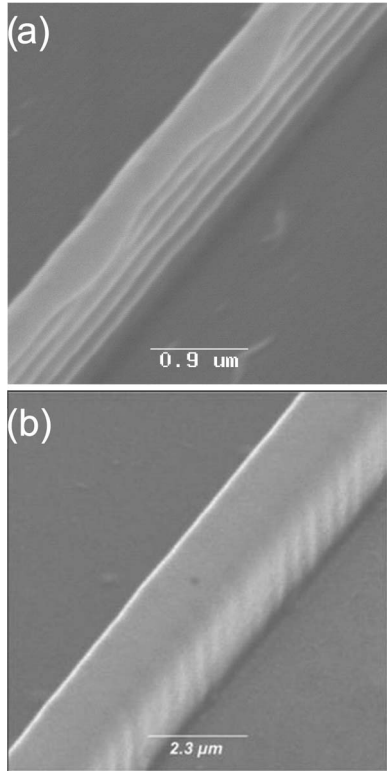
Fig. 2. SEM images of the waveguide structures fabricated on (a) gold and (b) glass substrates.

upon a gold layer is possible for a smaller range of laser powers than 2PP on a glass substrate. Simultaneously, the required threshold power for 2PP decreases. The first effect could be related to adhesion problems and laser heating of the gold layer. The reduction of the 2PP threshold can be explained by the interference of the incoming and reflected laser beams in the vicinity of the gold surface. In the interference maxima the polymerization threshold is reduced owing to higher laser intensity.

In Fig. 2, scanning electron microscope (SEM) images of waveguide structures fabricated upon gold and glass surfaces are shown. Longitudinal modulations of the waveguide sidewalls are evident in the structure fabricated on the gold surface, whereas they are not observed in the transparent glass substrate. This modulation appears to be due to the interference effects. The measured period of the interference pattern is (251 ± 4) nm. This value is in good agreement with interference period $d = \lambda/2n_{uc}$ ~ 254 nm determined by the half of the laser wavelength in the uncured ORMOCER. The slight transversal modulation of the sidewalls observed on the glass substrate can be attributed to small mechanical vibrations in the system.

To compare resolution limits of the 2PP technique on transparent (glass) and reflecting (gold) substrates, we fabricated single polymerized voxels in ORMOCER on several surfaces, using the same experimental setup. The diameters of the voxels fabricated on the glass and gold surfaces were measured as a function of the applied laser power. The experimental results are fitted by a simple expression[9]:

$$d(N_0,t) = r_0[\ln(\sigma_2 N_0^2 n \tau / C)]^{1/2}, \qquad (1)$$

where $\sigma_2 = 3 \times 10^{-55}$ cm^4 s^{-1} is the effective two-photon polymerization cross section measured previously[9] and $\tau = 80$ fs is the laser-pulse duration. The number of applied laser pulses, $n = \nu t$, is determined by the repetition rate $\nu = 80$ MHz of the laser system and the illumination time of a single voxel t, which is set to $t = 200$ ms in these experiments. Dimensionless parameter $C = \ln(\rho_0/\rho_0 - \rho_{th})$ depends on primary initiator density ρ_0 and the threshold density of radicals required for polymerization, ρ_{th}. For the ORMOCER material used, we have $\rho_0 = 1.8$ wt. % and $\rho_{th} = 0.25$ wt. %. The photon flux is defined by $N_0 = 2PT/(\pi r_0^2 \tau \nu \hbar \omega)$, where r_0 is the beam radius, P is the average laser power, ω is the laser frequency, and T is a factor that takes into account transmittance of light through the scanner and the microscope objective; $T = 0.20$ in the present case.

The experimental measurements of the voxel diameter and the results of calculations with Eq. (1) are shown in Fig. 3. In calculations of the voxel diameter on the glass surface, beam radius $r_0 = 520$ nm is used as a single fit parameter. To reproduce the experimental data for the voxel diameter on the gold surface, one must take into account the influence of the reflected beam. In this case one can reproduce the experimental data by replacing the photon flux in Eq. (1), N_0, by $N_0 \eta$, with $\eta = 1.6$. This value is below the maximum possible flux enhancement in the interference maxima. The reason for this is that, in our experiments, tightly focused laser pulses with a focus position slightly above the surface are used. Therefore the reflected beam is divergent and has lower intensity. The interference enhancement factor of 1.6 corresponds to the expected value.

The visibility of the interference pattern can be reduced when a structure is written several times with gradually increasing laser power. With this procedure, also the possibility of structural defects decreases. In this case the field enhancement effects at pointlike inhomogeneities are reduced because a thin layer of polymerized material is created on top of
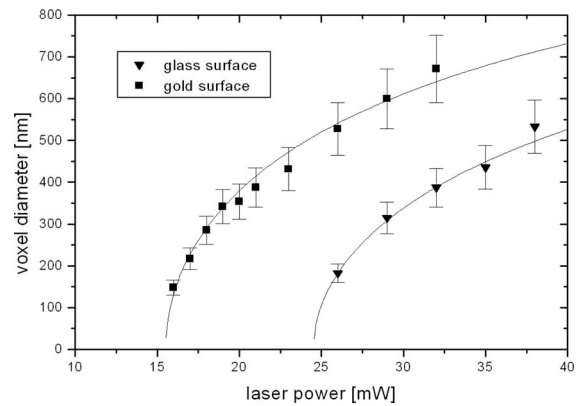

Fig. 3. Dependencies of the voxel's diameter on average laser power for the structures fabricated on the gold and glass surfaces by 2PP at a fixed illumination time of 200 ms. The curves provide theoretical fits of the experimental data.
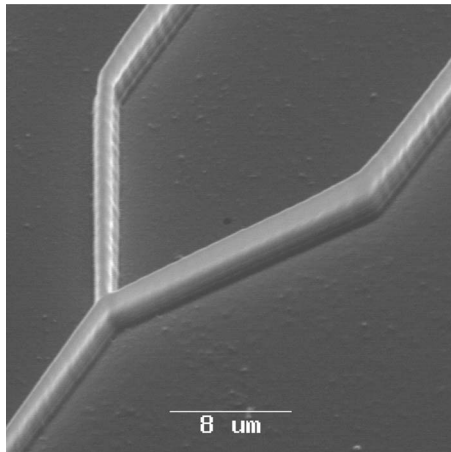
Fig. 4.   SEM image of a Y splitter waveguide fabricated with gradually increasing laser power from 6 to 14 mW.
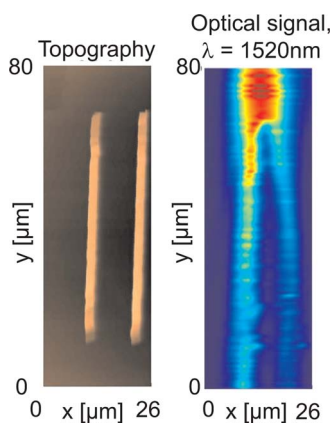

Fig. 5.   (Color online) Topographic image and SPP near-field intensity distribution at wavelength of 1520 nm recorded simultaneously by scanning near-field optical microscopy.

these inhomogeneities during the first, lower-power scan. To fabricate SPP structures such as waveguides, bends, and beam splitters with a smooth surface quality, we set the laser power initially to 6 mW and gradually increased it to 14 mW. For every laser power the structure is scanned twice. The resultant structures still show a weak interference pattern close to the metal surface but have much better surface quality. A SEM image of a Y splitter fabricated by this procedure is shown in Fig. 4.

Guiding properties of the dielectric SPP structures are investigated with straight 60 μm long ridges fabricated by 2PP. The SPPs are launched at the gold–air interface by a focused laser beam at a wavelength of 1520 nm in the Kretschmann-excitation configuration. The topography of the ORMOCER ridges and

the SPP near-field distribution imaged with a scanning near-field optical microscope are shown at the left and the right, respectively, Fig. 5. It can be seen that a relatively wide SPP beam is partially coupled into, propagating along, and coupled out of the polymer ridge. At the same time, the rest of the (diverging) SPP beam continues to propagate along the air–gold interface and is reflected by the neighboring ridge. Similar scanning near-field optical microscopes images were also obtained at other telecom wavelengths in the range 1500–1600 nm.

In conclusion, by use of a two-photon polymerization (2PP) technique, what are to our knowledge the first two-dimensional dielectric structures on metal surfaces for excitation and guiding of SPPs have been fabricated and studied. These results are promising for future applications and demonstrate high flexibility of the 2PP technique together with a high functionality of the fabricated dielectric structures.

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