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Rapid laser prototyping of plasmonic components

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ABSTRACT Renewed and growing interest in the field of surface plasmon polaritons (SPPs) comes from a rapid advance of nanostructuring technologies. In this paper, we will report on the application of two-photon polymerization (2PP) technique for the fabrication of dielectric SPP-structures, which can be used for localization, guiding, and manipulation of SPPs on a subwavelength scale. This technology is based on nonlinear absorption of near-infrared femtosecond laser pulses. Resolutions down to 100 nm (and even better) are already achievable. Characterization of these structures is performed by leakage radiation microscopy. 2PP allows the fabrication of dielectric waveguides, splitters, and couplers directly on metal surfaces. The dielectric structures on metal films are demonstrated to be very efficient for the excitation of SPPs. Using these structures, one can achieve excitation and focusing of the resulting plasmon field.

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1 Introduction

For a long time, metals have been investigated for their potential in downsizing optics beyond the diffraction limit [1, 2]. Surface plasmon polaritons (SPPs) excited on nanostructured metal–dielectric interfaces turn out to be promising candidates to serve that need. SPPs are electromagnetic surface waves coupled to coherent oscillations of electrons in a metallic conduction band at the interface with a dielectric material [3]. The corresponding electromagnetic fields are strongly confined to the interface. If the interface is nanostructured, it is the spatial dimension of the nanostructure rather than the light wavelength that determines the spatial extension of the SPP field. Recently, the feasibility of nanooptics based on metal nanostructures was experimentally demonstrated and elements, such as SPP mirrors, beam splitters, and interferometers, were realized [1, 2, 4–8].

In this paper, we study possible applications of two-photon polymerization (2PP) of the prepolymer ORMOCER for the fabrication of narrow dielectric structures on thin layers of noble metals, e.g., gold or metallic structures on a dielectric substrate capable of sustaining and propagating surface plasmons polaritons. Using 2PP one can fabricate high-quality three-dimensional microobjects and photonic crystals with a resolution down to 100 nm on dielectric transparent substrates [9, 10]. The SPP structures fabricated on gold films can be used for localization, guiding, and focusing of SPPs on a sub-wavelength scale.

The dielectric structures fabricated by 2PP are not only suitable for guiding of plasmons, as described in our previous paper [11], but are also very efficient for the excitation of SPPs [12]. The plasmons propagating along the surfaces can be observed in far-field by leakage radiation microscopy (LRM) [13, 14]. The leakage radiation is emitted by plasmons, as they propagate along the substrate–metal–air system, into the substrate material having usually higher refractive index. This enables the observation of free propagating SPPs. On the other hand, strong leakage radiation is responsible for SPP losses. Using the dielectric surface structures as waveguides, splitters, etc., one can suppress the leakage radiation. Thus, the SPP losses are reduced. The SPP field in this case is confined to the dielectric guiding structure. Another aspect of the dielectric surface structures fabricated from the high refractive index ORMOCER is the efficient generation of SPPs at the refractive index discontinuity, at the air–polymer boundary. SPPs are emitted perpendicular to the structure surface which can be applied for focusing of the generated plasmon field.

2 Sample preparation and experimental analysis

The SPP dielectric optical elements are produced by two-photon polymerization (2PP) technique [9, 10]. Dielectric structures are fabricated on glass or metal substrates by 2PP of the inorganic–organic hybrid polymer ORMOCER provided by Microresist Technology. This polymer can be polymerized by using a radical photoinitiator Irgacure 369 from Ciba Specialty Chemicals Inc. For the fabrication of dielectric structures by 2PP, a femtosecond oscillator, Spectra-Physics Model Tsunami, is used. This system delivers laser pulses at a wavelength of 780 nm with pulse duration of 80 fs (FWHM), and repetition rate of 80 MHz. In present experiments, an average laser power of 40 mW is applied. The schematic setup used for the sample preparation is illustrated in Fig. 1.

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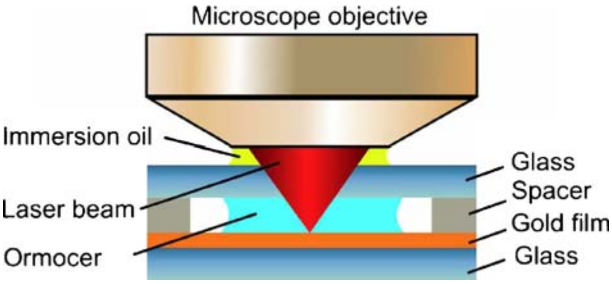

FIGURE 1 Schematic setup for the fabrication of SPP surface structures

Femtosecond laser pulses are focused by an immersion-oil objective (Nikon with a 100× magnification and a numerical aperture of 1.3). A liquid polymer droplet is sandwiched between the substrate and a cover glass with a thickness of 150 μm. Their separation is fixed by a plastic frame with the size of $18 \times 18\ \text{mm}^2$ and thickness of 100 μm. For the fabrication of surface structures, the laser beam is focused through the cover glass and the ORMOCER layer on the substrate surface. During the structuring, the laser beam is scanned along the sample surface by a galvo-scanner system from Scanlab. After completion of the 2PP and development of the surface polymer structures, the samples are washed in isobutyl-methylketone (4-methyl-2-pentanone) to remove liquid non-irradiated polymer. At the final fabrication stage, 50 nm-thin gold films can be deposited on the dried samples with surface dielectric structures by electron sputtering. A scanning electron microscope (SEM) is used for the visual inspection of the fabricated samples. Examples of dielectric couplers fabricated by 2PP are shown in Fig. 2. A possibility for guiding of SPPs in straight line ORMOCER waveguides has been demonstrated before by scanning near-field optical microscopy [11].

3 Leakage radiation microscopy of SPP focusing structures

Leakage radiation microscopy (LRM) is used for investigations of dielectric structures fabricated by 2PP technique and their SPP excitation properties [10, 13]. SPP focusing is studied on a curved (circular) line of protrusions (dielectric polymerized nanoparticles) covered with gold (Fig. 3, right).

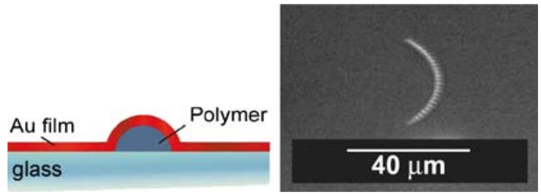

FIGURE 3 Schematic cross-section of the sample (*left*) and corresponding SEM image of the "*dot*" dielectric ring segment (*right*)

SPPs are excited at a wavelength of 800 nm using a laser beam focused on the sample surface by a microscope objective. The incident radiation scattered by nanoparticles is partially coupled into SPPs propagating along the metal surface. In our experimental setup, the laser spot size and its position on the structure could be easily adjusted by changing position of the objective. The LRM imaging technique is based on coupling of SPPs into leaky light modes propagating through the substrate. The direct transmitted light of the laser beam is blocked by Fourier-filtering [10, 15]. SPP focusing by the curved chains of nanoparticles (Fig. 3, left) can be readily observed by the LRM technique for moderate sizes of the incident laser beam. SPPs are excited by focusing laser beam onto the centre of the nanoparticle chain. In this case, the diameter of the exciting laser spot is approximately 10 μm. Interference of the divergent SPPs excited by different nanoparticles results in the SPP focusing, with a focal point located at the centre of curvature of the nanoparticle chain. Note that the SPP focal waist is very well localized and has relatively small value of less than 800 nm, as it has been measured from the images shown in Fig. 4. In addition to the SPP focusing, one can see that there is a system of SPP rays on the other side from the surface structure. The origin of this effect is again in the interference between the SPPs produced by different chain particles, as will be demonstrated below with numerical simulations, see Fig. 4, right. These features can be described

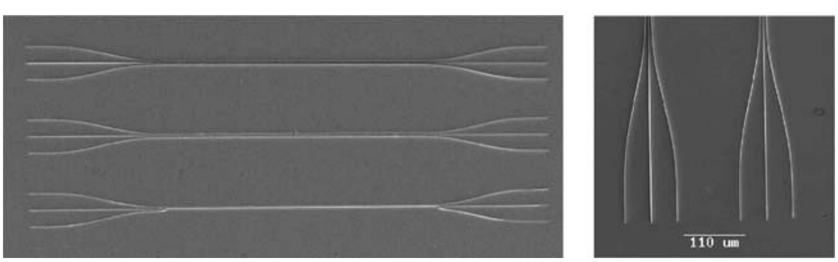

FIGURE 2 Example of bend three-fold coupling structures fabricated from ORMOCER

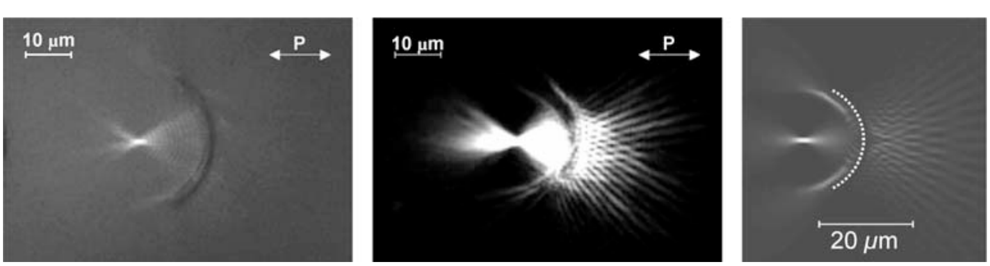

FIGURE 4 LRM images of focused SPPs exited on the "*dot*" dielectric ring segment. In order to make low-intensity details visible grey levels in the middle image are saturated. The *right image* shows results of numerical modelling

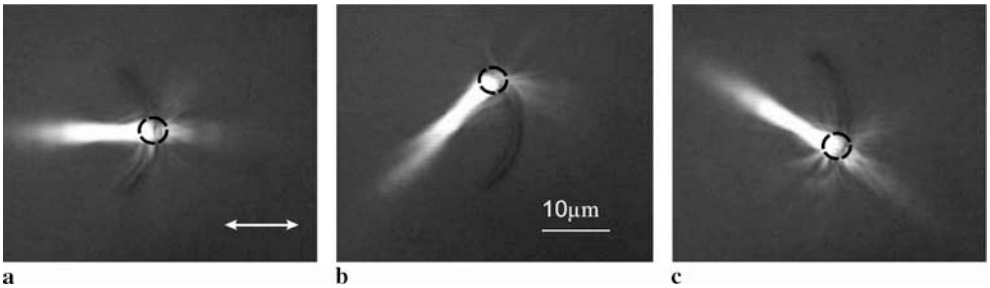

FIGURE 5 Experimental LRM images of the SPPs excited by the surface structure shown in Fig. 2b. The diameter of the incident light spot for the SPP excitation (*dashed circles*) is equal to 3 µm. (**a**) The exciting laser spot is at the centre of the structure, (**b**) and (**c**) the laser spot is shifted away from the centre. The *arrow* indicates the incident light polarization

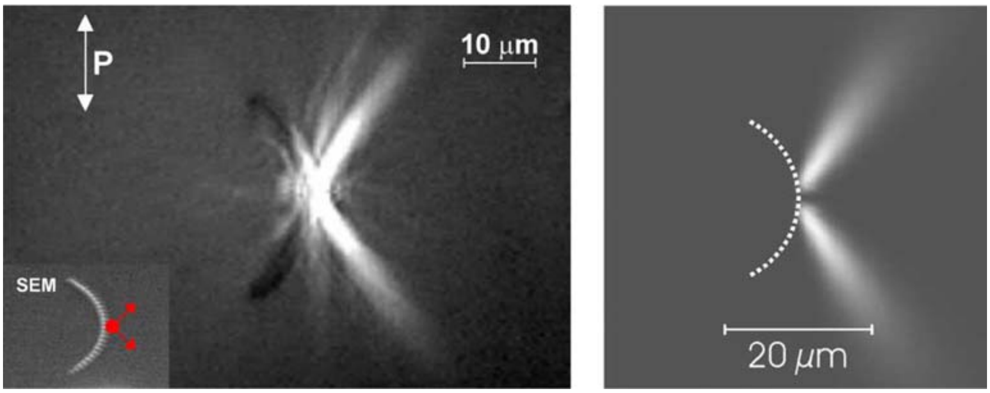

FIGURE 6 LRM images of SPP excitation on the "*dot*" dielectric ring segment by polarized laser beam (*left*). The *spots* and *arrows* in the SEM image in the *insets* indicate the laser focus position and SPP propagation directions, respectively. The *right image* shows results of numerical simulations

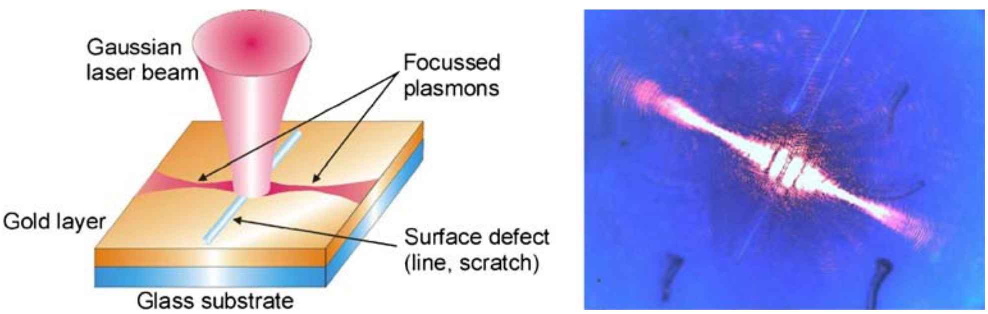

FIGURE 7 Schematic and realization of plasmon focusing on a straight line structure (*left*), and the realization visualized by the detection of leakage radiation

by a simple model in which it is assumed that each particle is the centre of a cylindrical wave and that the emission has a $\cos^2$-characteristic with respect to the polarization direction. Here particle sizes are neglected, only the interparticle angular distance of 4.5°, as derived from the SEM image in Fig. 3, is taken into account. Details of the numerical modelling will be presented elsewhere [16].

If the incident light spot is decreased down to 3 µm, the SPP focusing effect becomes less pronounced (Fig. 5), since only a few scattering centres are excited by the laser beam. The position where the laser beam is focussed on the structure is indicated by the dashed circles. The SPP focal waist broadens resulting in a nearly parallel and relatively narrow SPP beam. It is interesting to note that one can change the propagation direction of the SPP beam (always directed perpendicular to a local tangent of the particle chain) simply by changing the incident light spot position along the chain, see again Fig. 5. This effect is useful for the SPP manipulation in complex micro-optical elements utilizing chains of scatterers. One should keep in mind that the efficiency of SPP excitation decreases for directions deviating from the polarization of the incident light beam.

An experimental LRM image of the SPPs generated by the nanoparticle chain structure, when the incident light polariza-

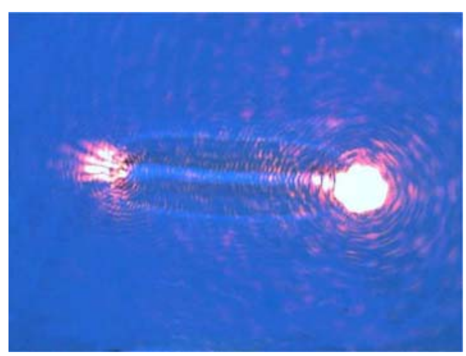

FIGURE 8 Plasmon propagation in a dielectric waveguide fabricated by 2PP. SPP is excited on the right hand side

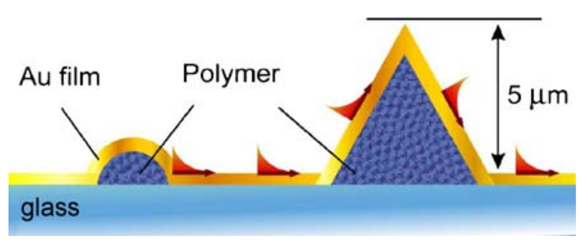

FIGURE 9 Schematic of a 3D surface structure for investigations of SPP propagation

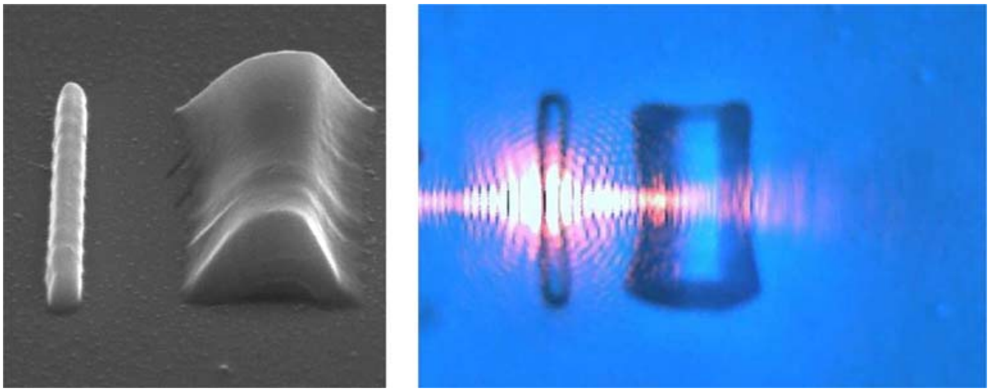

FIGURE 10 SEM image of the 3D test structure fabricated by 2PP (*left*) and LRM image of the SPPs excited on the dielectric line and propagating over the 3D ramp structure (*right*)

tion is turned by 90° compared to Figs. 4 and 5, is shown in Fig. 6. By illumination of the outer side of the dot ring segment with a vertically polarized laser beam, two non-collinear SPP beams are generated, which are not observed in case of the continuous dielectric ring segment. The right image in Fig. 6 illustrates results of numerical simulations.

Another possibility to achieve plasmon focusing can be realized by using curved laser beam wavefronts for the excitation of SPPs on straight surface defects [10]. Placing the laser beam focus below the surface results in a phase delay between the SPPs excited in the centre of the laser beam compared to that excited at the outer regions, see Fig. 7.

By changing position of the laser beam waist with respect to the metal surface, one can adjust the plasmon focus distance relative to the surface defect structure.

4 Guiding of SPPs by dielectric structures

Observation of SPP guiding in dielectric structures can be performed in the far field using LRM [11]. The structure under investigation is a straight ORMOCER waveguide with a height of about 1 μm. In this case SPPs inside the waveguide structure are non-radiative due to the same values of the refractive indices of the substrate and polymer material. SPPs can be excited by scattering on one (right hand) side of the waveguide and propagation of the SPPs can be observed behind the other end, as shown in Fig. 8.

In this experiment, the directly transmitted laser light used for the excitation of plasmons has not been blocked. Due to the high intensity of this directly transmitted light, the rapidly diverging free propagating SPPs are not visible. The SPPs transmitted through the waveguide still have relatively high intensity due to the confinement in the guiding structure and can clearly be observed by LRM at the left side.

5 SPP propagation on 3D surface structures

2PP provides possibilities for the realization and investigation of plasmon propagation on 3D surface structures. The schematic of first experiment is shown in Fig. 9, where plasmons are excited on the left defect structure and propagate over a ramp structure.

In this case leakage radiation from plasmons on the ramp structure is less visible compared to the leakage radiation from SPPs on the plane surface.

The LRM image together with the corresponding SEM image of the test structure is demonstrated in Fig. 10. This result clearly indicates that SPPs can propagate on smoothly curved surfaces with curvatures larger than the SPP wavelength.

6 Conclusion

Laser fabrication of dielectric 2D and 3D surface plasmon polariton (SPP) structures on metal surfaces by two-photon induced polymerization of a high refractive index inorganic–organic hybrid polymer has been studied. Optical properties of the fabricated dielectric SPP structures have been investigated by leakage radiation microscopy of the propagating SPPs. Effective excitation and focusing of the SPPs have been demonstrated with dielectric structures. The demonstrated results on excitation and manipulation of SPP fields and the simplicity of laser fabrication technique provide interesting prospects for the realization of future plasmonic devices.

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