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Application of ultrashort lasers pulses in micro- and nano-technologies

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Laser induced modification of materials is used for produce micro and nanostructures. Different techniques such as laser ablation, two-photon photopolymerization (TPP), near-field laser lithography (NFLL) are developed for structuring of various shapes and geometries using laser beams with 180 fs pulse duration at 775 nm, or 400 ps pulse duration at 532 nm. The realized structures and their applications are presented.

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1. Introduction

Nonlinear multiphoton absorption in materials is easily induced by ultrashort laser pulses due to high peak power of a focused beam. Depending on the nature of target material and the laser intensity, different effects could take place. The laser ablation or melting is responsible for the structuring of metallic, dielectric or semiconductors surfaces, while multiphoton induced photopolymerization of photoresists, or modification of the refractive index by laser induced densification of glasses is the way to create 3D structures in the volume of a transparent media [1]. Such effects are employed in a laser direct-writing (LDW) technique, as an alternative to the photolithographic methods, for producing different micro and nano-structures with controlled geometry. When the laser beam is tightly focused on or inside the material, the size of the modified area can be controlled by the laser intensity, the exposure time or number of laser pulses. If the laser fluence is properly set, the nonlinear absorption takes place only in the centre of the focused spot where the laser intensity exceeds the threshold value. Then, structures with dimension much below the size of the laser spot are created, even below the diffraction limit [2-5]. When the sample is precisely translated in 2D or 3D, practically any computed design can be obtained.

Here, we present a series of laser processing techniques using ultrashort laser pulses, such as laser micromachining, two-photon photopolymerization (TPP), laser ablation by optical near-filed enhancement. For implementation of these techniques we developed a microscope for laser processing which can be coupled with different type of laser sources. Structures on gold film or copper were created for applications in microelectronics. In photopolymers 3D microstructures were realised for applications in photonics or biology. Features of tens to hundreds of nanometres in size are also produced by special effects such as optical near-field enhancement.

2. Experimental set-up

A modular microscope for LDW was build to be coupled with laser beams at different radiation wavelength depending on the experimental requirement. As shown in the figure 1, the main modules are: the laser beam delivery module; attenuation module; the focusing optics; the sample translation stages in XYZ; the visualisation system with video camera; the confocal module for light collection from sample for signal analysis.

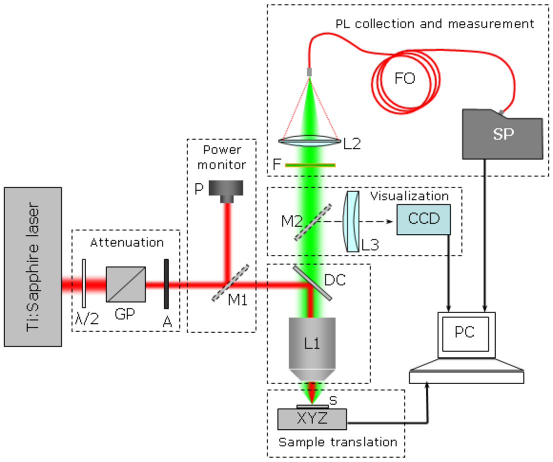


Fig. 1. Experimental set-up for laser processing and characterization. GP - Glan Polarizer; A – Neutral density filters; P – Powermeter ; DC – Dichroic mirror; L1- focusing lens; S – Sample; XYZ – translation stage; L2 – coupling lens; FO – optical fiber; SP - Spectrometer; L3 – imaging lens; CCD – video camera.

In our experiments, different laser sources were coupled with the LDW workstation. Laser beam with energy of more than tens of nano-Joules up to micro-Joules is required for laser ablation of almost all of materials. In this case, an amplified femtoseconds laser system Clark CPA-2101 we use. The laser emits femtoseconds pulses with 200 fs pulse duration, at 2 kHz repetition rate and 775 nm wavelength. The maximum laser energy is about 0.6 mJ per pulse. The second harmonic can be generated by a BBO nonlinear crystal at 387 nm with more than 40% conversion efficiency. Laser ablation was also performed with the same microscope at 1064 nm and 532 nm emitted from a Nd:YAG laser with 400 ps pulse duration at 1 Hz up to 10 Hz. High repetition rate experiments were performed with a femtoseconds laser oscillators (Femtolasers – Synergy) at 790 nm, delivering 10 fs pulse duration, and 100 nm spectral band width. The beam delivery optics is interchangeable and can be easily replaced with optics adapted for the working wavelengths.

The laser energy can be continuously and precisely attenuated with an attenuation system composed by a motorised half wave plate placed in front of a Glan polarizer providing a 300:1 extinction rate. In the case of extremely short pulse duration, a reflective polarizer is used for avoiding the temporal stretching of femtoseconds pulses due to the dispersion introduced by bulk material. A dielectric mirror reflects the laser beam to the focusing optics and transmits the other wavelength to the visualization system and to the characterisation module.

For beam focusing, different microscope objectives or lenses with a wide range of numerical aperture, adapted to a specific application, are used. The same focusing objective is used for light collection. The optical signal is collimated and sent to the upper part of the microscope trough the dichroic mirror. The light is coupled by a second focusing lens to an optical fiber in a confocal configuration. The collected optical signal is transmitted by an optical fiber to be measured or analyzed by photomultiplier or spectrometers.

The sample is translated by XYZ motorized stages and piezo drivers. A Nanocube stage (Thorlabs) has a total travel range of 4x4x4 mm^3 with hundreds of nm accuracy. The embedded piezo stage has 20 μm travel range per each axis and accuracy down to 5 nm. For longer travels, linear stages with 50 mm maximum travel are used. The maximum translation speed is 2 mm/s. The translations are computer controlled for generating any path according to a computed design. The sample focusing is done by the visualization system with CCD and a 200 mm tube lens. The resolution of the visualization system is better than 1 μm when a 100x microscope objective is mounted.

Dedicated software was realized for controlling the laser processing of samples. Common geometries such as parallel lines, grids, interdigitated structures, periodical structures of dots in rectangular or hexagonal symmetry, are included in a predefined library. More complex designs can be imported from files in bitmap format for 2D surface structuring, or in STL format in the case of 3D structuring of transparent materials. The STL is a standard format commonly used in rapid prototyping.

Using the above described microscope, we demonstrated several applications of femtoseconds lasers for micro and nanostructuring.

3. Microstructures produced by femtosecond laser ablation

Controlled surface modification and structuring can be easily induced via nonlinear absorbtion effects such as multiphoton absorption, followed by avalanche photoionization and laser ablation. When an infrared femtosecond laser beam is tightly focused at the surface of a material, due to the very high laser power density (GW-TW/cm^2), the temperature of the materials at the centre of the focused spot reaches rapidly very high temperatures, and the material is removed by ablation. Since the laser pulse duration is very short compared to the thermal diffusion time, the adjacent surface of the irradiated area remains unaffected. Therefore, femtoseconds lasers can be used for precisely processing of almost any kind of materials, such as metallic films, ceramics, polymers.

Compared to the classical lithography techniques, laser ablation is a direct writing method, no mask is required, and no corrosive chemicals are used, so it is an environmental friendly technique. It is a suitable method for producing microstructures for various electronic devices, such as interdigital capacitors, were fine electrodes has to be fabricated.

In the Fig. 2, series of interdigital capacitors with different number and width of fingers were processed by laser direct-writing technique on a copper film deposit on alumina substrate. Computer controlled algorithms are used for translating the sample in XY. The fabricated interdigital capacitors have electrodes with 1 mm length, 10 μm interdigital spacing, and digits width varying between 20 to 50 μm. The femtosecond laser beam from CPA-Clark laser system was focused by a 20X microscope objective with 0.4 numerical apertures. The diameter of the focused spot was 6 μm.

Fig. 2. Laser processed interdigital capacitors on copper film for different digit width: 20, 30, 40 and 50 μm.

The lateral resolution of the processed electrodes is generally given by the size of the focused spot and the laser fluence. Due to a finite ablation rate of the metal, the minimal width of the ablated lines is also limited by the thickness of the copper film, which in our case is 6 μm. For obtaining the complete removal of the material down to the substrate, the sample has to be processed using low scanning speed and high laser fluence. In the figure 3, profilometry measurements were recorded from a sample processed by laser at scanning speed of 0.1 mm/s, and laser fluence of 15 J/cm^2. As observed in the image, the film is completely ablated and the electrodes are not short circuited.

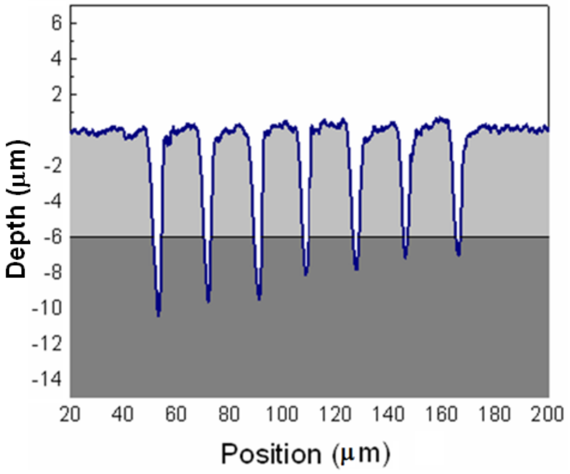


Fig. 3. Profilometry measurements of the laser ablated structures.

Such structures realized by laser ablation can be integrated in more complex circuits for microwaves, transmission line, band filters, couplers, antenna, in microstrip or Coplanar Waveguide configuration [6-8].

4. Submicrometer structuring by femtosecond laser ablation

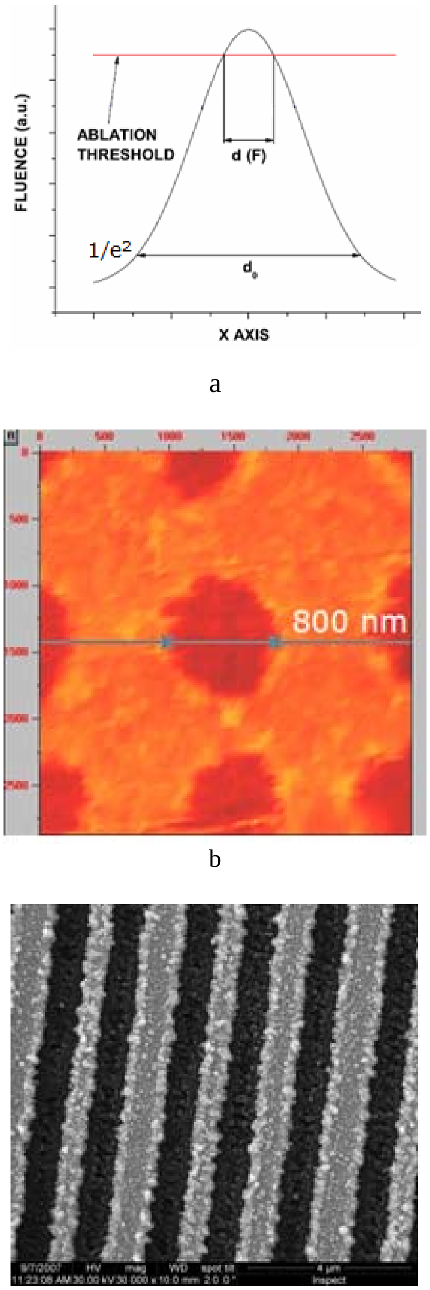
For thinner films, with thickness below the size of the focused laser spot, the dimension of the processed structures can be at submicron level. The fines of the fabricated structures can be controlled by the focusing optics and the laser energy deposited into the material. The following relations describe the dependence of the size of the ablated spot with the laser fluence:

$$d(F) = \frac{d_0}{\sqrt{2}}\sqrt{\ln(F\ /F_{th})} \qquad (1)$$

where d_0 is the focused beam diameter, given by the laser wavelength - λ, beam quality factor - M^2, and the focusing numerical aperture - NA:

$$d_0 = \frac{2M^2\lambda}{\pi NA} \approx \frac{\lambda}{NA} \qquad (2)$$

Fig. 4 (a) shown the principle of laser processing with resolution below the size of the focused laser spot d_0. When the laser intensity is kept just above the ablation threshold, the material modification takes place only in the center of the focused beam, where the laser fluence exceeds the intensity of modification threshold. Therefore, when the laser beam is tightly focused in a very small spot and the intensity is well controlled and kept at threshold, laser processing is possible even below the optical diffraction limit.



a

b

c

Fig. 4. (a) The principle of laser ablation below the diffraction limits. (b) and (c) submicron structures obtained on 100 nm thick gold films.

Laser ablation with submicron resolution was performed on gold thin film 100 nm deposit of glass substrate. In the figures 4 (b) and (c) different films processed by focused femtosecond laser beam are shown. For laser processing, the focusing optics was a 100X microscope objective with NA = 0.5. The laser fluence is kept just above the ablation threshold, as shown in the figure 4(a), in order to obtain small features on sample surface. Periodical structures such as holes, parallel lines, or grids were created by predefined structures library from the software. The parallel lines were created by translating the sample with the scanning speed of 0.2 mm/s. The width of the laser structured electrodes is below 1 µm. The period of the structures was 2 µm. At this level of structuring, the periodicity of the structures is limited by positioning accuracy of the translation stages, as observed in the figure 4(c). With piezo translation stages more accurate structuring can be realized, however the travel range is limited to hundreds of micrometers even in the case of the most outstanding piezo drivers available.

5. Nanostructuring by nearfield enhancement of femtosecond laser beam

While the resolution of direct processing method is limited by focusing optics and the accuracy of the mechanics, other laser techniques demonstrated to be able for producing structures with nanosize dimensions. Recent experiments have shown that light enhancement can produce a hot spot, resulting in the formation of a small pit on a silicon substrate using femtosecond or nanosecond pulsed lasers [9,10]. Near-field enhancement in the vicinity of nanometer-size particles is capable of producing nanostructures on large areas in a parallel processing regime, without complicated focusing and scanning systems. In this configuration, the enhanced field is confined in an area defined by the particle size to produce nanosize modification on a substrate. The optical field enhancement mechanism can be due to the lens effect or Mie's scattering depending on the particle size when transparent dielectric particles are used [11,12]. Using this method nanoholes array are fabricated by laser irradiation mediated by hexagonally arrayed polystyrene particles [13].

Fig. 5(a) presents the numerical simulation of the propagation of the light in vicinity of microsphere with 3 µm in diameter. In this simulation, the Maxwell equations are numerically resolved by finite difference time domain algorithm (FTDT). The Rsoft package from Rsoft Design was used for this calculation.

Experimentally, for obtaining a monolayer of microfocusing objects, a droplet of colloidal particles is placed on the substrate using a pipette. Following the water evaporation, layers of colloidal particles are self-assembled on the surface. Further details of the self-assembly process can be found elsewhere [14]. The substrates covered with a well organized monolayer of colloidal particles were irradiated with a single laser pulse.

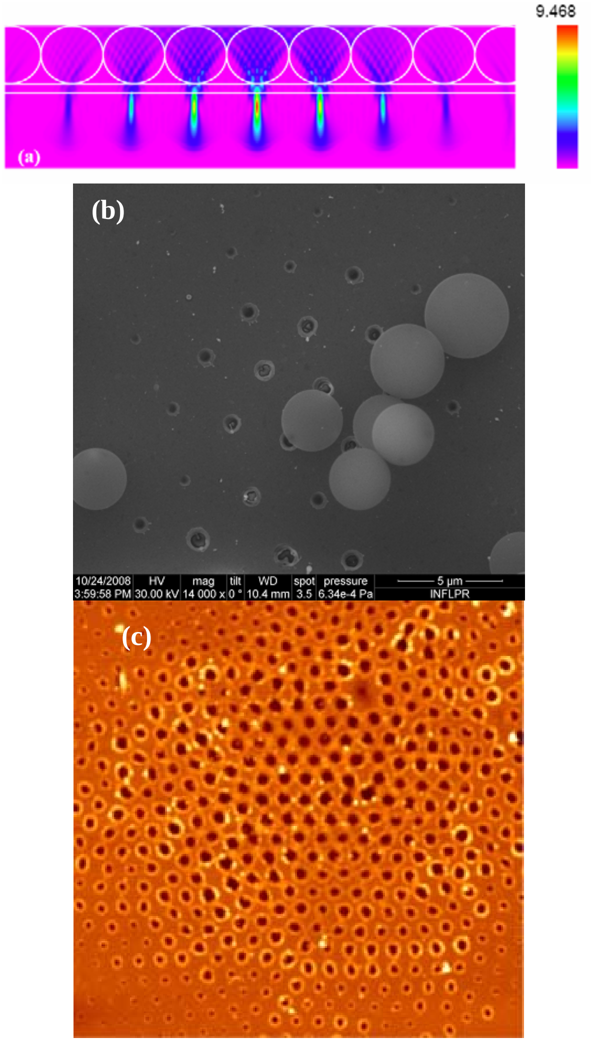


Fig. 5. Near field laser lithography. a) FDTD simulation of optical field enhancement underneath a microsphere b) near-field laser patterning of a 100 gold film, and c) of 50 nm Cu film.

Fig. 5(b) presents the SEM image of a glass substrate after laser irradiation in near-field regime. The diameter of the irradiated spheres was 3 µm, and the processing laser had pulse duration of 400 ps and 532 nm wavelength. The optical enhanced field at the interface of spheres with the substrate leads to laser ablation in the very small volume of the glass substrate. Simultaneously, the spheres are removed from de surface. A pattern of nanoholes with diameter of about 300 nm remain of the surface.

In the Fig. 5(c) spheres with diameter of 700 nm were deposit on 50 nm Cu thin film. The spheres are irradiated by laser with 200 fs pulse duration at 775 nm wavelength. The created holes have diameter down to 100 nm, smaller compared to the case of processing with longer laser pulses at 400 ps. The very short pulse duration, 180 fs, shorter than the thermal diffusing time in Cu layer, allows the evaporation of the metal in the area of the intensified optical field without affecting the surface of the adjacent zone. As seen in the figure 5(c) the diameters of the holes depend on the laser irradiance across the Gaussian profile of the beam.

6. Two-photon photopolymerization in photoresists using femtosecond laser pulses

The above mentioned techniques allow the fabrication of 2D micro and nanostructures on the surface of materials. In some applications 3D structures in transparent materials are needed, and techniques for fabrication of such microstructures have to be developed.

The nonlinear effect of two-photon photopolymerization (TPP) in photoresists was intensively used in the last years for developing the micro-stereo-lithography technique [15-19]. 3D microstructures can be realized by NIR femtosecond lasers processing in materials which are normally transparent to the NIR radiation. When a femtosecond laser beam is focused in the volume of a transparent photorezist, due to the high peak intensity in the beam waist, a high probability of two-photon [20] or multiphoton absorption occurs [21].

A large series of photoresists such as SU-8, Ormoces, KMPR, PMMA, etc. has the maximum of the absorption band in the UV-blue spectral range. In such photopolymers the two-photon absorption of NIR femtosecond laser pulses induces photochemical reactions and then photopolymerisation, just like in the case of a single UV photon absorption. In contrast with the single photon processing, the two-photon absorption occurs in a very tiny volume of material, at the center of the focused spot. If the laser fluence is keeped low enough, small features can be created with resolution down to tens of nm's [22,23]. After laser irradiation the polymerized sample is rinsed in a specific solvent for removing the non irradiated material. Then, complex microstructures are produced. However, in applications such as micro/nanophotonics or microfluidics, some designs are difficult to be realized because of the limited aspect-ratio of the structures, shrinkage and deformation of the polymerized structure, or collapsing of the structure on the substrate after removal of sample from the solvent.

In Fig. 6 a periodic network of parallel lines produced by TPP in SU-8 photoresist is presented. The period of the structures is 10 μm. The focusing optics was a microscope objective with 0.4 NA. The diameter of the focused spot was about 6 μm, the scanning speed was 0.2 mm/s and the laser energy was 1.5 mJ per pulse. From AFM measurement we observe the transversal profile of polymerized structure. Such structures could be used for producing diffractive optics elements, integrated optics or focusing optics for near-field laser processing.

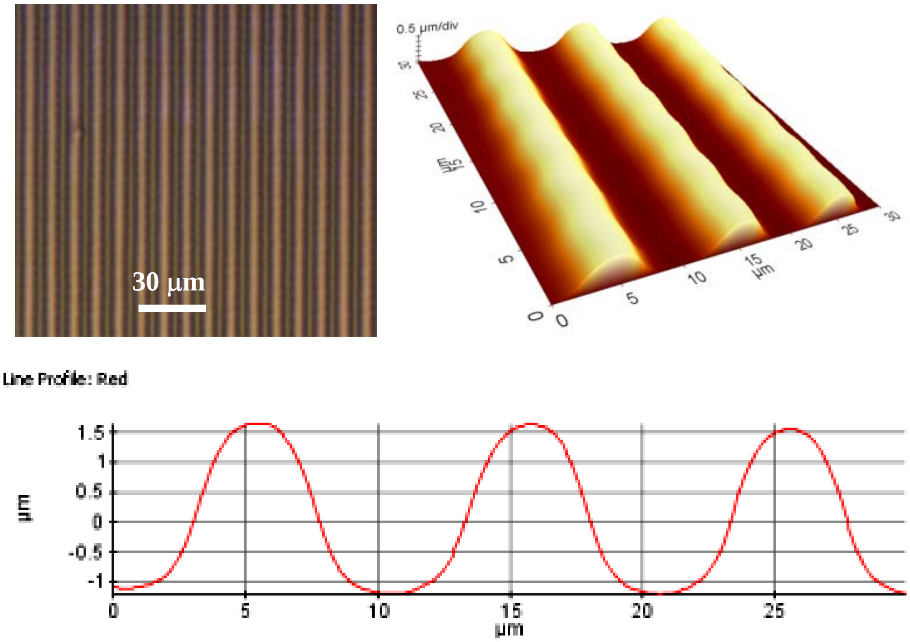


Fig. 6. Periodic structures produced by TPP in SU-8.

More other polymerized structures are presented in figures 7. An optical ring resonator with diameter of 100 μm was realized in Ormocer photoresist. The width of the waveguides is about 5 μm.

By TPP a scaffold structure with XY periodicity was produced. The Ormocer is a biocompatible photopolymer, and then such structures are suitable for fabrication of supports for biology applications and for tissue engineering.

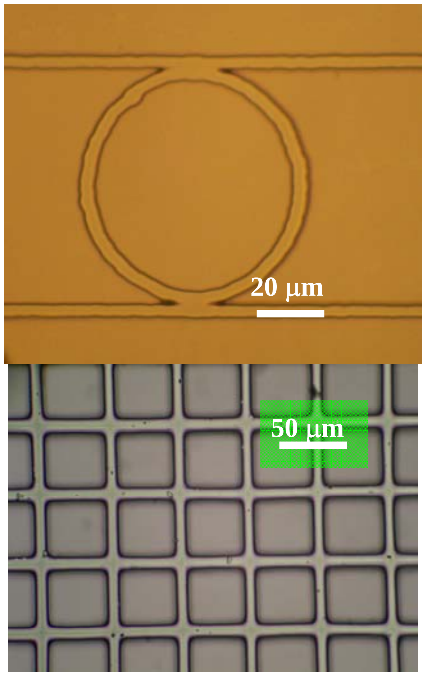


Fig. 7. Polymerized structures fabricated in Ormocer photoresist: a) Optical ring resonator, b) Periodical structure of XY lines.

The demonstrated TPP technique can be used for fabrication of photonic devices such as photonic crystals, optical couplers, diffractive elements, or 3D structures for microfluidics, scaffold for tissue engineering or other MEMS.

7. Conclusions

Many of devices for microelectronics are commonly produced by classical lithography. In some applications, such as high frequency microwaves or millimeter wave devices, the geometrical dimensions of the circuit layout reach the limits of classical photolithography. So, the laser ablation could be a valuable technique to process such devices. We presented a complex but low cost microscope for laser processing and characterization. We demonstrated various experimental techniques using short laser pulses at pico and femtoseconds time duration. Direct-writing techniques such as laser ablation, near-field laser ablation, two-photon photopolymerization, were used in order to produce microstructures on different material surfaces and in transparent materials. Nanostructuring was demonstrated by laser ablation in near-field regime using colloidal microspheres as focusing micro-optics. Features down to 100 nm were produced.

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