Two-photon 3D lithography: A Versatile Fabrication Method for Complex 3D Shapes and Optical Interconnects within the Scope of Innovative Industrial Applications

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Laser material processing using intense laser pulses, which are provided from ultrafast laser systems, enable sophisticated structuring methods such as the two-photon 3D lithography. This method is based on the simultaneous absorption of two photons in a photosensitive material that is transparent for the laser wavelength. The non linear laser-matter interaction induces material changes that are tightly confined around the laser focus and build up a structure in the volume of the material. These intrinsic 3D capabilities allow easy fabrication of a physical structure from a CAD model. In combination with suitable materials, it is possible to realize complex 3D structures and photonic micro- and nano-systems. We introduce our experimental setup comprising laser source, three axes sample stage and 3D registration of the sample. We present two distinct applications of the versatile two-photon 3D lithography on organic photosensitive materials: first, the fabrication of complex arbitrarily shaped 3D micro-structures and second, the fabrication of direct laser-written, embedded multimode waveguides that are aligned relative to preconfigured printed wiring boards (PWBs). In the first case, the high resolution aspect of the method is addressed. We could fabricate small structures with feature sizes less than 1µm in a material with a very efficient photo initiator at a low concentration of only 0.025 wt%. In the case of waveguide fabrication, the lithographic method is combined with astigmatic beam focusing for a larger interaction volume as required for writing multimode waveguides. Due to the sample registration prior to the waveguide fabrication, the alignment of the waveguides becomes an intrinsic part of the fabrication process itself. A single organic-inorganic hybrid material is used for both, the waveguide core and its cladding, because the material exhibits a sufficiently large increase of the refractive index upon laser irradiation. The function of such waveguides is demonstrated by monitoring transmitted light at the waveguide exit and the optical loss of the fabricated waveguides is found to be approx. 0.64 dB/cm from cut back measurements. The total optical loss of a 12 cm optical interconnect on the PWB (including coupling losses at both waveguide caps and propagation loss) is in the range 11-20 dB. The ultimate goal of this approach is the large scale fabrication of leading-edge PWBs with an integrated optical communication layer.

Keywords: two-photon 3D lithography, embedded waveguides, photosensitive organic material, printed wiring board

1. Introduction

Lasers are versatile instruments for material processing, amongst which femtosecond lasers show superior properties in terms of structure quality compared to nanosecond lasers. The ultrafast energy deposition enables the fabrication of structures with clear and smooth shapes and boundaries of virtually any material. Additionally, femtosecond lasers are used for processing transparent materials where the laser-matter interaction is based on non-linear processes, which requires a high peak power of the incident laser light. Hence, femtosecond lasers are not only used for an improved surface processing based on laser ablation, but also for high-resolution three-dimensional patterning of transparent media, such as the two-photon 3D lithography relying on the simultaneous absorption of two photons. In contrast to linear absorption, the two-photon based nonlinear laser-matter interaction exhibits specific features that can be exploited for advanced and functional microfabrication [1]: the energy transfer from the laser to the material is strongly confined to the vicinity of the laser focus, where enough photons per pulse and interaction volume are present and the simultaneous absorption of two or more photons by the material becomes probable. Hence, the alteration of the material due to absorbed laser light is also confined to a small volume around the laser focus. This small exposed volume element is usually called a voxel. Its size depends on the light intensity profile in the area, where a two-photon absorption is triggered and is influenced by various parameters such as numerical aperture of the focusing optics, laser wavelength, pulse duration, exposure time, repetition rate of the laser, material diffusion etc. [2]. Since the influence of the laser on the material is confined to the voxel, an arbitrary 3D shape can be easily build up in a voxel-by-voxel manner inside a photosensitive material by scanning the laser focus through the volume of the material. The patterning inside the material requires no special experimental conditions such as vacuum or inert gas atmosphere during the sample processing.

The above mentioned spatially limited modification of the material upon laser exposure alters either chemical and/or physical properties of the target material, for example the solubility, or the refractive index. This effect is utilized for the fabrication of 3D micro-devices and building blocks for M(N)EMS/M(N)OEMS, and other photonic micro- and nano-systems. Femtosecond lasers have been used in the field of photonics for waveguide fabrication in glass [3, 4], complex three-dimensional structures employing two-photon micro-stereo-lithography in organicinorganic hybrid materials [5], photonic crystals [5, 6, 7], and many more. A very detailed and comprehensive survey over the development is found in [8].

Basically, the two-photon 3D lithography is capable to fabricate complex 3D photonic structures in one laser writing process. It is possible to think of photonic crystal structures combined with waveguides that are manufactured in one process step. Such parts can be considered as building blocks for the integration of optical communication paths into printed wiring boards (PWB), which is motivated by the increasing demand for faster data rates and puts optical information paths into the focus of research. In contrast to the conventional core-cladding approach for the formation of an embedded waveguide, the presented application of the two-photon 3D lithography requires only one (suitable) material for core and cladding of the buried waveguide structure. Beside requirements regarding the optical performance of the waveguide (low optical loss), adequate polymer materials must be sufficiently photo-sensitive at the laser wavelength (sufficiently large two-photonabsorption cross section) and must be compatible to the industrial process flow (high temperature and high pressure during lamination, chemical treatments etc.).

Fabrication concepts of optical core layers in PWBs usually use two different polymer materials with different refractive indices for the core and the cladding layer, respectively, where the layers are usually built up sequentially. Embedded waveguides are photo-lithographically patterned and buried in the cladding material [9]. This approach requires sophisticated coupling methods between waveguides and electro-optical components on the board. Poor coupling increases optical losses drastically and leads to higher bit errors and reduced communication bandwidth.

In this paper, the adaptation of the two-photon 3D lithography for the fabrication of 3D shapes and integrated waveguide systems is presented as a novel approach. Provided that a suitable material is available, the need for separate cladding and core materials becomes redundant, and the coupling between waveguides and electro-optical components on the board becomes an inherent part of the patterning process, which ultimately leads to a significant cost reduction regarding processing and integration.

2. Materials and Methods

The material matrix and its photo-sensitive component are essential for the practical application of the two-photon 3D lithography. Special developed and high efficient photo initiators are superior to commonly used UV photo starters which show a very low two-photon absorption cross section. In contrast to UV photo starters, efficient two-photonphoto-initiators based on π -conjugated compounds allow a more practical and widespread application of the two photon based lithography because such two-photon excitable materials require less laser power and can be used e.g. for 3D data storage [10].

For the 3D shapes presented in this work, a 1:1 blend of Genomer 1330 (trimethylolpropane triacrylate) and Sartomer 415 (ethoxylated trimethylolpropane triacrylate) is used. The matrix material is mixed with a donor substituted 1,5-diphenylpenta-1,4-diyn-3-one (DPD) derivative (only 0.025 wt% photo initiator for the most efficient D- π -A- π -D system are required for two photon induced polymerization). Details about the specially developed photo initiators are published elsewhere [11]. The material and all photo starters are provided by the technical university of Vienna. The photo-polymerization changes the material solubility: after writing the 3D structures, the unexposed material is washed away with methanol and the developed structure is examined by means of SEM.

For waveguide fabrication, the sol-gel synthesis based material ORMOCER[®] is used, which has already proved to be suitable for a two-photon based micro- and nanofabrication [12].

The synthesis of ORMOCER[®] comprises the establishment of a Si-O-Si network via hydrolysis/ (poly) condensation reactions of alkoxysilanes, which yields organically modified nanoscaled inorganic-oxidic units, resulting in a pre-polymer sol. The sizes of these units usually range from about 1 to 10 nm, dependent on the material system synthesized. An organic cross-linking is performed either photochemically and/or thermally, which leads to (patterned) ORMOCER[®] layers. (Oligo-) methacryl, acryl, styryl, or epoxy groups are often used in order to account for the organic cross-linking, and the resulting material thus behaves as a negative resist [12]. The material properties can be chemically tuned by the synthesis conditions, which permit tailoring the materials as required by the application. An overview about applications of inorganicorganic hybrid materials is given in Refs. [14, 15]. The material used within this work is described in Ref. [17]. The resin is highly transparent at 850 nm (0.02 dB/cm) and 1310 nm (0.028 dB/cm). The material can withstand the PWB processing at temperatures as high as 200 °C under a pressure of about 20 bar [16].

For photonic purposes, the fact that the photopolymerization locally increases the refractive index of the ORMOCER[®] material upon laser irradiation in the focal volume is exploited. Thus, regarding ORMOCER[®] waveguide structures, the exposed area forms the core of an embedded waveguide without the need for a further development of the unexposed material which acts as a cladding. This simple method enables also the in situ coupling of laser-written waveguides to optoelectronic components as an intrinsic part of the patterning process.

2.1 Experimental Setup

An amplified ultrafast Ti:Sapphire laser system from Spectra Physics (Mai Tai[®] - Spitfire[®] combination) is used for the direct laser processing. The laser system operates in the NIR and provides pulses at 800 nm wavelength with a pulse duration of approx. 130 fs, and operates at a repetition rate of 1 kHz (Spitfire - amplified pulses). Maximum pulse energy of approx. 1mJ can be achieved. An extracavity electro-optical switch is used for synchronizing the laser to the sample motion and for attenuating the laser power. For improving the fundamental spatial Gaussian beam distribution, the amplified laser pulses are subsequently focused through a spatial filter before being guided to the sample and additional diagnostic tools, such as a Femtos single shot autocorrelator and a Spiricon laser beam analyzer (Fig. 1). A He-Ne laser is collinearly guided to the sample and used for depth gauging of the sample: the light from the He-Ne laser, which is back-reflected from the sample interfaces is detected by a photodiode in a confocal setup. This setup is used for the measurement of the sample's surface position at the laser beam axis, and by means of scanning the sample laterally; the sample surface can be mapped. A CCD camera which is placed close to the microscope objective is used for lateral registering laser and photodiodes that are attached to the sample PWB, and are supposed to be connected by a laser-written waveguide. The handling of large-area samples with the photosensitive material exposed to ambient atmosphere requires yellow light and clean-room environment for the entire system.



Fig. 1: Scheme of optical setup. Main components of the system are the Ti:Sapphire laser system, the electro-optical pulse picker / attenuator (Pockels cell), spatial filter, confocal z gauging setup with photo diode PD, 3-axes high precision stage, and machine vision by means of a CCD camera next to the three axes stage for sample registration. The collinearly He-Ne laser for sample depth profiling is also shown. The optical paths between sample gauging and laser writing are switched by means of flipping mirrors as indicated by the arrows.

The laser is astigmatically focused by a 70mm planconvex lens, which is combined with a 1:3 cylindrical telescope and a 20x microscope objective. These additional optics reduce the laser beam diameter in one dimension, and allow symmetric shaping of the waveguide's crosssection, which was successfully shown for multi-photon absorption based waveguide fabrication in an Er:Yb doped glass [13].

3. Fabrication of **3D** structures

3.1 Material testing for 3D structures

Material testing is performed prior to the fabrication of 3D structures or waveguides. Simple 2.5D mechanical stable designs were generated and arranged with CAD software. All structures were identical and written with the same optical setup, whereas only one parameter is changed, (*e.g.* the laser power) for different structures in the arrangement. After development, the optimized laser writing conditions and system parameters were determined from SEM images (Fig. 2). Finally, more complex 3D structures were fabricated with optimized system parameters.



Fig. 2: Material testing of Genomer/Sartomer blend (1:1) with 0.025 wt% N-DPD photo initiator. a) The sample feedrate was set to 0.5 mm/min and the laser power was varied between 20 μ W and 10 μ W as indicated in the figure. The structures at the corners are fabricated at 25 μ W and serve as markers. The laser is focused by a 100x (NA=0.95) microscope objective. b) Item fabricated at approx. 10 μ W. Best results were obtained for the given setup with the laser power set to less than 14 μ W.

It is observed from the material tests that both, the system parameters of the equipment as well as the photo initiator are crucial for the quality of the structure. A non optimized two-photon initiator for a given material system may result in significantly worse structures than a very efficient photo starter and the parameter screening may not lead to a set of optimized process parameters as compared to a more suitable material / photo initiator combination (Fig. 3).



Fig. 3: Test structure of Genomer/Sartomer blend (1:1) with a) 1.5 wt% Irgacure 369 and b) with 0.025 wt% N-DPD photo initiator. c) and d) show details of structure b). The sample feedrate was set to 1mm/min and the laser power is set to best values for each material / photo starter combination (40 μ W for a) and approx. 15 μ W for b)). The laser is focused by a 40x (NA=0.65) microscope objective

3.2 Two-photon 3D lithography of 3D structures

The fabrication of 3D structures comprises a simple sample preparation: a glass substrate is cleaned (rinsed with isopropanol and the use of opti-clean polymer peel tabs) and coated with a 5 nm Au film, which helps to identify the surface position of the substrate and the alignment of the structure relative to the substrate surface. Subsequently, a drop of the polymer material is drop-cast on the substrate for the formation of a thick film. In case of smaller structure sizes (max. height of approx. 25µm), the material is spin-coated on the substrate. After sample preparation, the CAD data is converted to CNC code for the three axes high precision stages. For this purpose, the CAD model is sliced and the vertex coordinates of the contour line stack are extracted (Fig. 4). Finally, the outer shell of the model is build up layer by layer starting at the substrate surface in order to have good adhesion on the substrate, which prevents wiping off the structure from the substrate during the development process.



Fig. 4: Stack of contour lines (yellow) generated from a CAD model. The vertices of the contour lines represent the coordinates for the linear interpolating sample stage motion.

After development, the unexposed material is removed and a free standing structure is obtained. Since only the shell of the model is solidified upon laser irradiation, the core is still liquid and a post development UV flood exposure is applied to the structures for approx. 1-2 minutes. This totally cures the structures, which are subsequently Au-coated and examined by SEM (Fig. 5).



Fig. 5: Samples of 3D structures fabricated by means of twophoton 3D lithography. The material was the Genomer-Sartomer blend with 0.025 wt% N-DPD photo initiator.

4. Fabrication of embedded ORMOCER[®] waveguides

4.1 Material testing for waveguides

In terms of ORMOCER[®] waveguide writing, parameters (laser power and sample feed rate) and optic settings (alignment of the cylindrical telescope for a nearly circular waveguide cross-section etc.) are screened prior to waveguide writing. The ORMOCER® is mixed with 1.8 wt% Irgacure 369, which is a common UV initiator. In addition to optical microscopy, the waveguides were characterized by means of refracted near field (RNF) measurements, which yield the refractive index distribution across the waveguide. It was found that the increase of the refractive index at the waveguide centre is typically < 0.005, which is sufficient for embedded waveguides with a bending radius > 20 mm. Although the increase of the UV photo initiator yields a larger increase of the refractive index under same exposure conditions [18], the index change upon femtosecond laser irradiation did not increase with 3 wt% photo initiator in first measurements.

Investigations of the material with more efficient twophoton excitable photo initiators and concentrations regarding the maximization of the refractive index increase at the waveguides are currently running.







For this purpose, ORMOCER[®] films were prepared on pieces of a scored board and examined after the laser writing of the waveguides. The film preparation comprised drop casting, softbake on a hotplate at 90°C for approx. 2 minutes and UV flood pre-cure under Argon atmosphere for approx. 10s. This treatment of the sample prior to laser writing stabilizes the film and leads to a very flat and smooth surface of the film, which is essential for laser writing through the sample surface. Subsequently, crosssections of these pieces are investigated by means of optical microscopy, where the depth and the cross-section of the individual waveguides are characterized by their shape and diameter. Due to the refractive index contrast, the laser written structures can be identified by means of transmission optical microscopy. At appropriate laser parameters, nearly circular and transparent waveguides are found in the optical film. The same material tests were performed for verification of the waveguide bundle geometry (Fig. 6a-c).

4.2 Concept of a PWB with optical layer and twophoton 3D lithography of embedded waveguides

Many different and strict requirements must be fulfilled by a suitable material for the fabrication of embedded waveguides: it must exhibit a sufficient two-photon sensitivity to the NIR laser light for photo-patterning the waveguide structures, combined with a large increase of the refractive index upon exposure (>0.005). Furthermore, a low optical loss at the communication wavelength of the electro-optical components is required and the material must be capable of forming thick film in order to fully embed the mounted electro-optical components on the board. The material must also withstand the chemical treatments in the PWB fabrication process.

In order to give consideration to the simplicity of direct laser writing of waveguides by means of two-photon 3D lithography, the concept of a fully integrated optical interconnect must also be kept simple (Fig. 7): the electrooptical components are directly attached to the board and embedded by a thick film consisting of the optical material that forms both, the core and the cladding of the waveguide. The core is directly inscribed in the material by scanning the laser focus through the thick film from the source (laser diode) to the detector (photo diode). The writing laser is incident from the top in Fig. 7 and focused as described in Fig. 6c. The embedded chips work at a wavelength of 850 nm and are mounted on their edge.



Fig. 7: Concept of PWB with integrated optical layer and embedded electro-optical components

The used boards for the demonstration of a working embedded optical interconnect between a laser- and a photodiode are either 5 cm or 15 cm long. These boards are provided by our project partner AT&S, the market leader in Europe of printed circuit board production. AT&S is developing innovative approaches to integrate an optical interconnection path (E/O-converter- optical waveguide - O/Econverter) completely into a multilayer printed circuit board. The demonstration boards are equipped with 850 nm VCSELs that are mounted upright on the die edge such that the emitting area of the VCSEL points towards a photodiode. A frame is attached to the boards, which enables the casting of thick ORMOCER® films on the board since a total embedding of the laser and photodiodes is required, and to avoid material flow through the drill holes which are used for electrical connections. The embedding film has to be thick enough to have a planar surface even above the diodes. Any distortions in the optical film cause problems to the laser writing of the waveguides. Since the laser is focused through the surface, the surface has to have properties that are comparable to an optically flat and smooth surface. The final film thickness was set to be in the range between 300 and 500 μ m.

The sample preparation comprises a rough cleaning of the board with lens cleaning paper, a rinse in isopropanol, and subsequent drying. The ORMOCER® is drop-cast on the board and soft-baked on a hotplate at approx. 100°C for 2-5 minutes. The elevated temperature reduces the viscosity of the material, and therefore supports the formation of a uniform thick film on the board. After thermal treatment of the sample, a UV flood exposure under Ar atmosphere is performed. This pre-curing of the ORMOCER® stabilizes the material mechanically. For this purpose, a low power sunlamp is used, and the sample is exposed to UV for up to 50s at a power of approx. 3 mW/cm². At this stage, the photocurrent of the photodiode is measured, when the laser diode is operated in order to define a reference prior to the waveguide fabrication. Subsequently, the lateral positions of the laser and the photodiodes on the substrate are registered, the waveguide bundle configuration is defined, and the surface along the waveguide path is mapped by monitoring the intensity of the back-reflected light of a He-Ne laser with a photo diode in a confocal setup. Input and output of the butt joint waveguide bundle is referenced to a point at the laser- and the photodiode top edge, respectively. For this purpose, the depth information of the sample is acquired at the location of the laser and photo diode chips. From the chip design, the exact position of the laser output and the sensitive area of the photo diode is known and the waveguide can be position precisely relative to the top edge of the chips, which is known from the confocal depth scans (Fig. 8).



Fig. 8: Referencing the waveguide depth relative to surface of optical film, top edge of laser diode chip and substrate position. The red curve indicates a scan at the position of the edge mounted laser diode. The two peaks represent the position of the polymer surface film and the top edge of the diode chip. The black line indicates a scan next to the diode chip. Here, the peaks show again the surface of the polymer film and the second peak indicates the position of the substrate. The shift of the two surface peaks indicates height variations of the optical film surface.

Taking into account the surface profile along the waveguide path and the coordinates of the start and end point of the waveguide bundle, the three axes stages move the sample such that the laser focus is scanned across the optical ORMOCER[®] layer in order to form a direct optical interconnect between the electro-optical components (Fig. 9).



Fig. 9: Sample surface registration and fabrication principle of directly written embedded waveguides. a) Principle of scanning the laser across the sample. b) Each interface gives a peak of back reflected light, and hence a depth profile of the sample (optical film thickness). c) Complete surface map and sample tilt. d) Transversal writing setup of the waveguides. The exclamation mark denotes the critical interface area between waveguide and diode chips, which must be planar for efficient coupling between diode chips and waveguide.

The feed rate of the axes is set to 20 mm/min for all waveguide samples. Thus, the writing of the waveguides takes approx. 7 to 40 minutes, depending on the waveguide bundle configuration (single line or bundle) and the length of the waveguides, respectively. After laser writing, the photocurrent is measured again and compared to the value before the inscription of the waveguides. Finally, the sample is thermally treated for 3 hours at 150°C to stabilize the complete system. At this state, the sample is ready for further processing such as lamination for terminating the PWB process (Fig. 10).



Fig. 10: 5x5 cm² PWB with attached frame, electro-optical components, and ORMOCER[®] film after the fabrication of the waveguides. At this stage, the PWB is ready for lamination.

4.3 Characterization of fabricated optical interconnects

The successful fabrication of a waveguide connection between laser- and photodiode was determined by the comparison of the light induced current of the photodiode before and after the waveguide writing. The total optical loss of the system (including coupling losses at both waveguide caps and propagation loss) is estimated in the range 11-20 dB, for a length of the waveguides of approx. 12 cm. The total damping of the waveguide is calculated to be approx. 7.8 dB. From cut back measurements, it is found that the average loss of the ORMOCER[®] waveguides is approx. 0.64 dB/cm. Details can be found in [14]. This value is higher than the pure material loss, and is related to waveguide imperfections such as wall roughness or optical layer imperfections along the path of the waveguide. Nevertheless, such a system is capable of transmitting data at a transfer rate of > 1Gbit/s with a bit error rate of 10^{-9} . Even higher data transfer rates of > 2 Gbit/s using this approach were reached and are described elsewhere [16].

5. Summary

The versatility of the two-photon 3D lithography with its inherent 3D capabilities is demonstrated by both, free standing 3D polymer objects and integrated working optical interconnects. The presented experimental setup is the same for both tasks; only the focusing optics has to be exchanged. Due to a modular setup, the exchange of the optics and the alignment can be done in rather short time. It is also shown that beside the experimental setup the material/photo initiator combination plays an important role for the final quality of the 3D structures.

In case of embedded optical interconnects a suitable inorganic-organic material was used that is capable of forming both, the core and the cladding of the embedded waveguide due to a rather large increase of the refractive index upon femtosecond laser irradiation. The setup was successfully adapted and the integration of working optical interconnects into a printed circuit board could be demonstrated. The enormous flexibility of this lithographic method allows an implementation of machine vision capabilities including sample registration and surface mapping that are prerequisites to make the waveguide alignment and coupling as part of the waveguide fabrication process. This method is considered very promising as a part of a largescale fabrication process of PWBs, which improves the board performance drastically in terms of the increasing demand of faster intra-board communication. The fruitful cooperation between the research organization Joanneum Research and the large PWB manufacturer AT&S brings the implementation of the described method into large scale industrial board fabrication processes closer.

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