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Combining two-photon-polymerization with UV-lithography for laser particle acceleration targets

T Abel[1], R Dörr[2], W Krüger[1], R Horstmann[2], G Schaumann[1], M Roth[1] and H F Schlaak[2]

[1] Institut für Kernphysik, Technische Universität Darmstadt, Schlossgartenstr. 9, 64289 Darmstadt, DE
[2] Institut für Elektromechanische Konstruktionen, Technische Universität Darmstadt, Merckstr. 25, 64283 Darmstadt, DE

E-mail: `torsten.abel@physik.tu-darmstadt.de`

Abstract. In the field of laser driven particle acceleration, many experiments with plain foil targets have been performed to investigate ion acceleration mechanisms such as TNSA and BOA. A more complex target geometry, for which the front and rear surface can be structured in a deterministic way, will lead to a better understanding of e.g. the actual source size, laser contrast related effect to the coupling efficiency of laser energy into the target of specifically shaped target surfaces or means to reduce the particle beam divergence such as ballistic focusing. A promising technology to manufacture complex 3D freeform targets is the two-photon-polymerization. With resolutions better than 200 nm, it is possible to create small targets with microstructured surfaces. We implemented the combination of two-photon-polymerization and UV-lithography to facilitate the handling of such small structures.

1. Introduction

So far, in the field of laser driven ion acceleration, a large variety of experiments have been performed with flat foil targets to investigate the fundamental processes involved [1, 2, 3, 4]. To expand the range of applications for the laser-generated particle beams (e.g. proton therapy [5], x-ray Thomson scattering [6]), the interest in manufacturing more complex, freeform targets increases. Simulations show that the highest ion energies are achieved with target sizes that are in the order of few times the laser spot size [7, 3]. It should be noted that targets in the size equal to the laser focus diameter are difficult to strike due to the pointing stability of the laser system. Given the fact that the diameter of the laserspot can be as small as several micrometers, it is challenging to manufacture freeform targets of similar size by using classic micro machining processes.

But not only the total size is of interest. We aim for small targets with a substructure like the sinusoidal pattern on the concave side of the hemispherical shape shown in figure 1. Flat foil targets with such a structure can be used to determine interesting parameters like ion sources size and divergence. In the same way targets like the conceptual design in figure 1 could be used to explore these parameters in combination with focusing effects. The outer diameter should be in the order of the incident lasers focus size. The whole target should be bigger than one times the laser focus due to typical pointing stabilities of current high power laser systems. The wavelength of the sinusoidal microstructure should be sufficiently small in order to observe several periods across the source size. However, those targets can be created by two-photon-polymerization. The idea using two-photon-absorption to manufacture arbitrary 3D geometries dates back to the 90s [8]. Since that time it was used for many applications in different fields of research [9, 10].

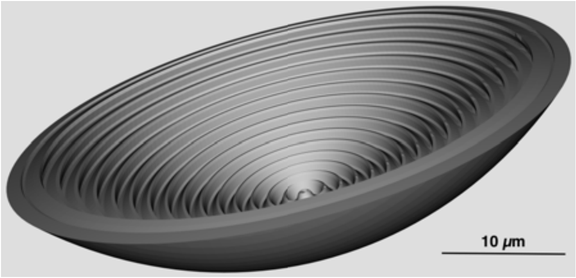

Figure 1. Conceptual design of an hemisphere target with inner sinusoidal microstructure. The outer diameter should be in the order of several times the incident lasers focus size. Due to the pointing stability of the laser the whole target should be bigger than one times the laser focus. The wavelength of the sinusoidal microstructure has to be smaller than the laser spot size.

The bare target can not be handled because of the small size. To attach the small targets to different standardized holders, as used in the respective experimental facilities, some kind of handle or frame is needed. Due to the fact that the expenditure of time for manufacturing strongly correlates with the target volume, the two-photon-polymerization is not suitable for large geometries. We managed to combine classic UV-lithography with the two-photon-polymerization, to shorten the manufacturing time.

2. Experimental Setup

With the two-photon-polymerization it it possible to manufacture small 3D freeform targets in the range of some µm [11]. But without some kind of holder it is nearly impossible to handle such small structures. It is too time-consuming to expose holding structures in the scale of some mm^3 with the two-photon-polymerization, so we use UV-lithography to create the comparatively large holding structures.

2.1. Substrate preparation

One of the most common substrates used with photoresist is Si-wafer, but in principle all kinds of flat homogeneous substrates are feasible. As we need a sacrificial layer to lift the exposed structures from the substrate, we used two distinct substrates with different metallic sacrificial layers. The first combination was a *FR-4* substrate with 35 µm copper on top, the second 18 µm aluminium on glass. *FR-4* is a glass-reinforced epoxy laminate. The substrate with copper on top is a standard source material for printed circuit boards and commercially available. Aluminium works well as sacrificial layer in combination with the *AZ 125 nXT* photo resist because it is solvable in the developer (*AZ 726MIF*) we used for this resist.

We use the same photoresist for the two-photon-polymerization and the UV-lithography. Most of the resists are designed to be used with an extensive UV light source. Traditionally, mercury vapour lamps have been used in lithography and many available photoresists are optimized for wavelengths at the $g-$, $h-$ and $i-$spectral lines from Hg. The g-line has a wavelength of 435 nm, the h-line is 404 nm and the i-line is 365 nm. These wavelengths fit well to a frequency doubled Titanium:Sapphire laser. We used two different types of photoresists with different characteristics (*AZ 125nXT* and *SU-8 2025*). The *SU-8 2015* resist is an epoxy based resist with a proportion of solids of 68.55 % and a density of 1.219 g/ml. The ingredients of the AZ resist are not known. We choose negative resists to minimise the volume to be exposed and the exposure time. With this approach the thickness of the photo resist limits the target extension in one dimension. With both resists we could easily achieve film thicknesses thicker than 100 µm. To prepare the resist for the exposure regardless of which process, UV-lithography or two-photon-polymerization, a plane, thin layer of resist spread on a substrate is needed. We used spin coating to obtain such a layer. Spincoating means to spread the resist by rotating the substrate. The combination of the acceleration, the speed of rotation and the viscosity of the resist determines the resist thickness. To achieve a 100 µm thick layer of *AZ 125nXT* resist on a 20 mm × 20 mm rectangular glass substrate, we used the routine shown in table 1.

step	revolution per s	duration in s
1	300	10,0
2	1000	1,2
3	800	12,0
4	2000	1,5

Table 1. Programm steps to spincoat 100 um *AZ 125nXT* photo resist on a 20 mm rectangular glass substrate.

2.2. Exposure

The standard setup for UV-lithography consists of the photoresist, a mask and the UV-lightsource. The mask covers all the parts of the resist which should not be exposed. In the case of a negative resist, like in our setup, that means the covered parts will be removed by the development process after the exposure. With that method it is possible to fabricate so called 2.5D structures. 2.5D means the 2D pattern from the mask is extruded in the third dimension. The whole wafer area will be exposed in one step, thereby allowing the creation of multiple target frames in one step.

Instead of an extensive mask and UV-light source, which are used in conventional lithography, a focused laser with about 800 nm center wavelength comes into operation for the two-photon-polymerization. To expose the resist a nonlinear optical process, the two-photon-absorption is used. In lieu of one photon with 400 nm, two photons with 800 nm are absorbed, which results in the same energy transfer, as can be seen in Fig. 2.

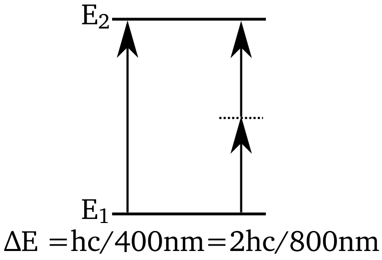

Figure 2. The simultaneous absorption of two photons with 800 nm is equal to the same energy transfer with the absorption of one photon at 400 nm.

For a simultaneous absorption of two photons, a high photon density is needed because of the quadratic dependence of two-photon absorption on the intensity of the incident laser beam [12]. Two-photon-absorption is related to $\chi^{(3)}_{\text{imag}}$, the imaginary part of the third order susceptibility which is a material depended nonlinear optical property. Simultaneous absorption means in this case, that the first absorbed photon creates a virtual state and the second photon must be absorbed during the lifetime of this state, which is approximately 10^{-15} s [13]. Therefore, this phenomenon only takes place when the light intensity is high enough. This condition can be found only in a small volume around the point of maximum intensity within the focus region, both laterally and along the direction of laser propagation. This small volumen is marked in figure 3.

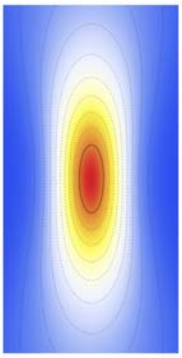

Figure 3. Image of an focused beam with iso-intensity lines. The solid contour marks the voxel.

Such a 3D volumetric pixel in which the exposure gets triggered is named a *voxel*. To create a 3D freeform by two-photon-polymerization, the resist has to be exposed point by point, respectively voxel by voxel. By increasing the voxel size, which leads to a reduction of the resolution, it is to a certain extend possible to reduce the process time.

In our setup we move the substrate with three high precision linear stages. Within one layer, we achieve 80 nm bidirectional accuracy over a travel range of 160 mm in X-direction and 50 mm in Y-direction. To reach a small voxel size, we use an objective with a numerical aperture of 0.85 and a working distance of 1.1 mm to focus the Ti:Sapphire laser into the resist. With our setup, we aim to achieve a radial resolution in the order of 200 nm. Along the laser propagation, the extension of the exposed volume pixel is typically larger, as compared to the radial dimension, and depends e.g. on the focusing geometry, exposure time, and laser intensity [14].

3. Multi component target manufacturing process

The first step of creating a target in a frame, made from photoresist, is to create a CAD model of both structures. In the following, two different frame designs for the same hemi-cone target are presented. We manufactured two different designs, using different resists depending on ongoing applications based on the final structures. The further application will be discussed elsewhere. The shown hemi-cone structure was chosen for demonstration purposes. To this end the structure in the frame is rotated by 90° and not hollow. The current orientation, as depicted in figure 4, eases the successive characterization with respect to the measurement of relevant geometrical features.

The target-frame combinations are designed to be exposed with the two previously described techniques and different resolutions. In figure 4, an example design for a fractionally manufactured target in a frame is shown. The hemi-cone target in the middle (red) and the two retaining bars (yellow), which connect the target and the ring in the frame, were manufactured with two-photon-polymerization. The frame (green), which is designed as a 2.5D object, was exposed by conventional UV-lithography.

The dimensions of the frame are displayed in figure 5. The frame was manufactured out of a 100 µm thick layer of *AZ125nXT*. The resist was spincoated on a glass substrate and prebaked for 40 min at 140 °C. A 13 µm aluminium layer between glass and resist worked as sacrificial layer.

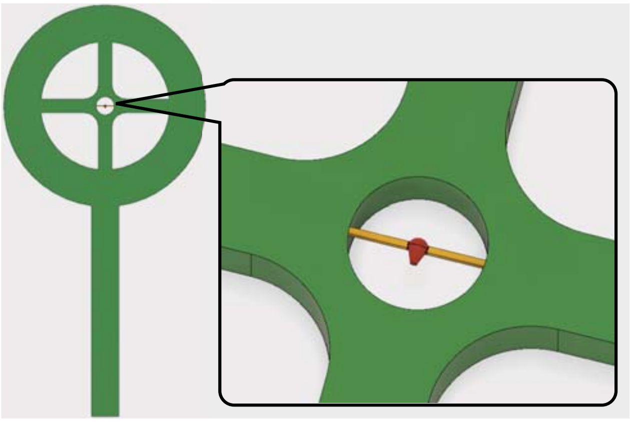

Figure 4. CAD model of a multi part target in a handling frame. The target (red) and the retaining bars (yellow) are exposed via two-photon-polymerization with different resolutions to be time efficient. The frame (green) is 2.5D and should be manufactured with UV-lithography.

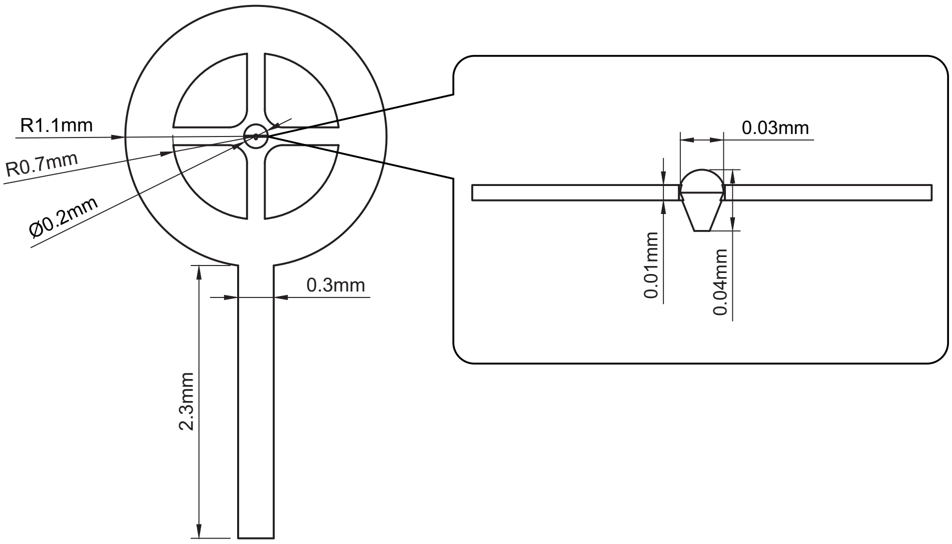

Figure 5. Dimensions of the designed target and its frame as shown in figure 4.

The sample was exposed with a light dose of $5.76\,\mathrm{J/cm^2}$. As the mask-aligner we used for the exposure does not have any filters available, we used the g-,h-and i-line of the Hg-spectrum.

For the two-photon-polymerization we used a Titanium:sapphire laser with a central wavelength of 800 nm, a repetition rate of 80 MHz and typically a pulswidth of 100 fs. Depending on the resolution and processing time, we can provide an average laser power in the range of 5 mW to 50 mW with a movement speed of $0.1\,\mathrm{mm\,s^{-1}}$ to $10\,\mathrm{mm\,s^{-1}}$.

Since the target shown in red (figure 4) is the most important part, a high line resolution of 200 nm was used. This high geometrical resolution was achieved by a movement speed of 0.3 mm s^{-1} and 20 mW laser power. The combination of low speed and high number of lines results in a very time consuming process. For the retaining bars, the resolution is of less interest, since they just have to connect the target to the frame. The aim of the design must be to reduce the diameter of these bars to find the thinnest bars which withstand the forces during development, to further reduce the process time.

Once the liquid is dried and the bars do only need to support the target itself, structural integrity is not an issue any more, as the weight of the target does not impose any significant force on the interconnection bars. Right now, the bars are 10 µm to make sure they are strong enough. Future designs will further reduce this part. The bars were exposed with a line resolution of 500 nm and a movement speed of 1 mm s^{-1} and a laser power of 50 mW to speed up the process.

The result of the combined exposure with two-photon-polymerization and UV-lithography is shown in figure 6.

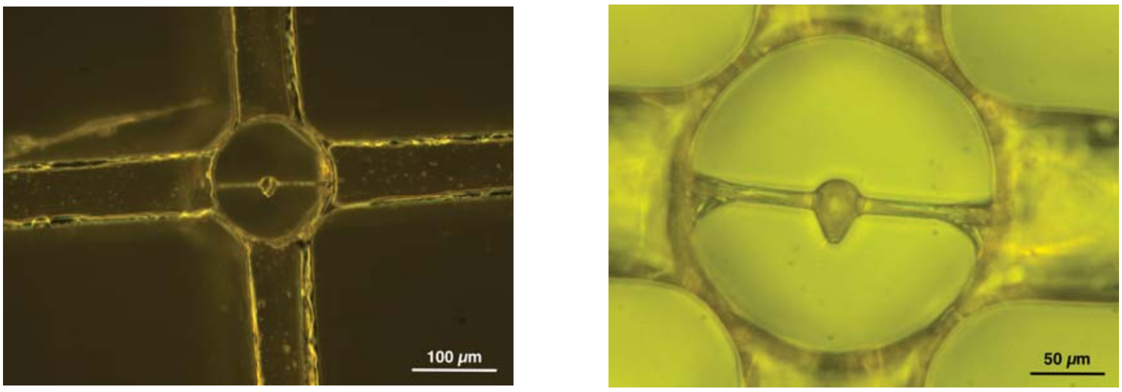

Figure 6. Manufactured target, following the design shown in figure 4. The hemi-cone target and the retaining bars were created with two-photon-polymerization with the target structure having a higher resolution. The handling frame was manufactured with conventional UV-lithography.

For a second application, we prepared 80 µm of *SU-8* on a *FR-4* substrate with 36 µm copper on top. To make sure that the UV exposed structure overlaps with the target stilts, alignment marks were needed, which were created by partially removing the sacrifical layer. The mark consists of a circle in the middle of a cross as can be seen in figure 7. The circle has a diameter of 250 µm.

After spincoating, a first prebake of 3 min at 105 °C was done, before the target was build via two-photon-polymerization. After the two-photon-polymerization, a second bake of 180 min at 105 °C minimized the solvent content. In contrast to the exposure of the first frame design, here the large holding structure was exposed with 800 mJ/cm^2 light with only one wavelength of 365 nm corresponding to the Hg-i-line. After the exposure, a postbake at 95 °C for 60 min followed. We used Propylene glycol methyl ether acetate (PGMEA) for 10 minutes to develop the structure. After the development process, some resist was left around the target. To remove all the remains, the samples were plasma etched. The etching unit has a power of 1000 W and was used for 5 min at 25 °C. The process gases were 1000 sccm of O_2, 50 sccm of CF_4 and 50 sccm of N_2.

4. Conclusion

We successfully combined the two-photon-polymerization with the UV-lithography in the field of targetry. With the combination of these techniques, we made it possible to produce and

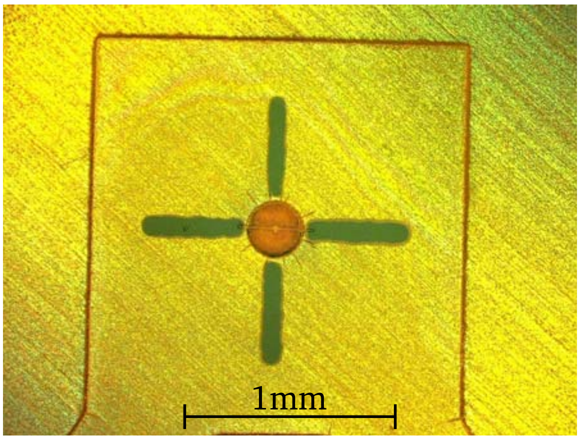

Figure 7. The target frame, made of SU-8 photo resist, has a hole of 250 µm in the middle to enable a free-standing target. In the background, the Cu sacrificial layer is visible. The cross visible through the photoresist serves as an alignment mark. It was prepared by removing the sacrificial layer.

handle micro structured targets in the size of only a few micrometers, safely held by a larger supporting structure. With the two-photon-polymerization exposure and successive processing, one can fabricate structures in true 3D with sufficiently high resolution to be used for many applications in the realm of targetry. With UV-lithography, it is possible to expose target handling frames in the size of several mm^2 within minutes. Using both lithographic processes for one target is a promising method for high number production of complex micro targets as this approach combines the batch processing at medium resolution during conventional mask exposure lithography with the ultra high resolution and true free form exposure with the two-photon-polymerization.

5. Acknowledgement

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