Within the last two decades micron- and submicron patterning of material surfaces by laser-induced ablation, etching, deposition, and surface modification has been extensively investigated [1]. Here, patterning was performed by “direct writing” where the laser light is just focused onto the substrate, by projection of the laser light via a mask, by employing a direct-contact mask, or by the interference of laser beams. Another technique employs a SNOM-type (scanning near field optical microscope) setup. Here, the laser light is coupled into the tip of a solid or hollow fiber. By positioning the substrate within the near field of the fiber tip, one can produce patterns with widths that are not limited by optical diffraction. This technique has been employed for nanolithography [2], ablation [3], material etching [4] and for local reduction of oxides [5]. Its main disadvantage is the low throughput that can be achieved in a fabrication process, even when bundles of fibers are used.

Presently, we are investigating a new method for single step maskless patterning. It permits one to produce on a substrate surface thousands or millions of single submicron features with a single or a few laser shots. The technique employs a regular two-dimensional (2D) lattice of microspheres. Such lattices are formed by well-known self-assembly processes, e.g. from colloidal suspensions. In contrast to earlier investigations [6] we used such 2D lattices not as lithographic masks for consecutive processes, but as an array of microlenses on a transparent support. The microspheres focus the incident laser radiation onto the substrate, albeit with significant (spherical) aberration. The
experimental arrangement employed is schematically shown in Fig. 1. According to geometrical optics, the “focal plane” of microspheres, i.e., the distance between their centers where both principal planes are positioned, and their foci, $f$, is given by

$$f = \frac{r}{2} \frac{n^2 - 1}{n}$$  \hspace{1cm} (1)$$

where $r$ is the radius and $n$ the refractive index of the microspheres. A white light image generated by a-SiO$_2$ microspheres at a distance $z = f$ is shown in Fig. 2a. In this case, the microscope picture reveals the hexagonally close-packed structure of the lattice formed by the microspheres. The distance between intensity maxima is equal to the diameter of microspheres which was $d = 2r = 3 \pm 0.15 \mu m$. With $n(a-SiO_2) = 1.50$, we find for the distance of the focal plane $z = 2.25 \mu m$. In reality, spherical aberration shifts the (diffraction) focus towards the center of the sphere, i.e., the distance between their centers $z_{(a)} = \frac{3}{2} \pm 0.15 \mu m$. With $n(\text{a-SiO}_2) = 1.50$, we find for the distance of the focal plane $z_{(a)} = 2.25 \pm 0.15 \mu m$. With $n(\text{a-SiO}_2) = 1.50$, we find for the distance of the focal plane $z_{(a)} = 2.25 \pm 0.15 \mu m$.

The microscopic mechanisms of photoablation of polymers have been discussed in detail in [1]. For PI and 302 nm laser radiation with pulse lengths between 10 ns $\leq t_\lambda \leq 1$ s we found that ablation can be quantitatively described by purely thermal mechanisms [12].

### Interference Subpatterns

If we now vary the distance $z$ by an amount $\epsilon$, so that the total distance between the centers of the microspheres and the substrate surface is given by $z = f + \epsilon$, we can generate different interference patterns in the Fresnel region, where one has a similar situation, was found to increase with decreasing particle size [1,11].

The process studied in most detail is the ablation of polymers by means of excimer-laser radiation. Fig. 3 shows an atomic force microscope (AFM) picture of a small part of a polyimide (PI) foil patterned with single-shot 248 nm KrF-laser radiation. The fluence was $\phi = 50 \text{ mJ/cm}^2$ and the pulse length at full width half maximum (FWHM) $t_\lambda = 28$ ns.

With these laser parameters, we observe strong ablation of the polymer foil within the focal region of the microspheres. The holes shown in the figure have a FWHM diameter of $500 \pm 100 \mu m$ and a depth of $250 \pm 50 \mu m$. Their mean distance is equal to the mean diameter of the microspheres. The arrangement of these holes clearly reveals the hexagonal lattice structure of the microspheres. By using microspheres of various diameters, the distance between holes can be varied by at least two orders of magnitude. Similar experiments have been performed with other materials. However, in contrast to most other polymers as, e.g., polyethylene-terephthalate (PET), PI does not melt, but directly sublimes into gaseous product species.

The changes in the spatial frequency of the hexagonal pattern can be qualitatively understood along the following lines. Spheres create waves, which first converge, and then diverge behind the foci. These waves interfere in the region where

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**Fig. 1**: Schematic picture of the setup employed in the experiments. The transparent microspheres of diameter $d = 2r$ focus the incident laser radiation onto the substrate surface [7].

**Fig. 2**: White-light microscope pictures generated by a-SiO$_2$ microspheres of diameter $d = 3 \pm 0.15 \mu m$.

a) $z = f$; in this case the distance between the intensity maxima is equal to the diameter of spheres. b) $z = f + \epsilon$. The distance between interference maxima is about $800 \text{ nm}$. At the right side of the figures the boundary of the array of spheres can be seen [8].
The focal plane of waves is given by $R = \frac{d^2}{4\pi m}$. The wavelength employed is $\lambda = 0.248 \, \mu m$ and the Rayleigh length $z_R = 3\lambda$. This corresponds to a beam waist $w_0(1/e$ field at $z = f) = \sqrt{\frac{\pi \lambda}{2}} = 0.242 \, \mu m$. a) Intensity distribution within the focal plane $z = f$. b) Same as a) but at $z = f + \varepsilon$. At that position, the radius of curvature of waves is given by $R(z) = \varepsilon + z^2/\varepsilon = d^2/4\varepsilon; \varepsilon = 9.01 \, \mu m$. Though about 50 adjacent beams were used in the numerical summation, only the nearest neighbors significantly contribute to the interference pattern as the beam waist $w (1/e$ field at $z = f + \varepsilon) = 2.94 \, \mu m$.

Here, all factors that are identical for all beams have been omitted. $R = R(\varepsilon)$ is the radius of curvature of beams and $A$ the amplitude. $\rho_j$ describes the center of beam number $j$. The calculated intensity distribution is shown in Fig. 5. Consider, e.g., the interference of 3 adjacent beams with spacing $d$. Interference will be constructive if the phases of beams differ by $2\pi m$, where $m$ is an integer. For the points 1 and 2 in Fig. 5b, this results in $R = \frac{d^2}{4\pi m}$ which was used in the calculations with $m = 1$. The primary maxima which are also observed within the focal plane, are indicated by a "0" and secondary (additional) maxima by "1" and "2". Both, primary and secondary maxima have similar intensities, as they are created by about the same number of interfering beams.

While the calculated intensity distributions shown in Figs. 5a,b qualitatively describe the pictures in Figs. 2a,b and the arrangement of generated patterns in Figs. 3 and 4 quite well, a direct, quantitative comparison is not possible. This has several reasons: The differences in wavelengths employed, the omission of aberration phenomena in the theoretical considerations, the size distribution of microspheres, the uncertainties in the material and experimental parameters, etc. The interference patterns before and behind the focal point may differ significantly. Arrays of spheres can be considered as a diffraction grating. The field distribution in the Fresnel region is quite complicated and is governed by the parameter $d^2/\lambda$ (Talbot effect [13]).

In summary the technique described in this contribution can be employed for large-area surface patterning of substrates. In preliminary experiments we have demonstrated that the same technique can be used for laser-induced chemical etching, deposition, and surface modification. In contrast to "direct writing", thousands or millions of single submicron features can be produced with a single or a few laser shots.

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References