Laser-manufactured mirrors for geometrical output coupling of intracavity-generated high harmonics

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Abstract: We demonstrate micro structuring of fused-silica laser mirror substrates by Inverse Laser Drilling. Slits of a width down to ~80 µm and circular holes with diameters down to ~50 µm have been structured into quarter-inch thick substrates. Except for chipping, the surface areas around these openings have not been irreversibly affected by the manufacturing process. The micro structured mirrors can be used for geometrical output coupling of coherent EUV radiation from cavity-enhanced high harmonic generation.

OCIS codes: (220.4610) Optical fabrication; (070.5753) Resonators; (140.7240) UV, EUV, and X-ray lasers; (190.4360) Nonlinear optics, devices.

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1. Motivation

An established method for the generation of coherent radiation in the extreme ultraviolet frequency range (EUV) is high harmonic generation (HHG) of ultrafast laser radiation in gases. The required peak intensity for this nonlinear process is >10^{13} W/cm^2, preferably at a long Rayleigh length to maximize the interaction volume with the gas and to improve the phase-matching. At repetition rates of several tens of MHz the peak power of the driving radiation can be increased by the use of an enhancement resonator. This allows for achieving the required peak intensities at these high repetition rates with moderate focusing and with moderate driving laser powers in the order of magnitude of 10 W [1,2].

However, the output coupling of the high harmonics from the resonator has been difficult because no dichroic mirrors exist for wavelengths in the extreme-ultraviolet spectral region. Different methods of output coupling have been proposed and investigated: Gohle et al. and Jones et al. used a sapphire plate placed at Brewster's angle to separate the harmonics from the fundamental beam [1,2]. Yost et al. demonstrated output coupling by using a small-period diffraction grating etched into the surface of a dielectric resonator mirror [3]. Pronin et al. proposed an anti-reflection-coated grazing-incidence plate [4], and Pupeza et al. proposed a wedge-on-mirror output coupler [5], these two being advancements of the Brewster plate technique. A different method is geometrical output coupling through an opening in the cavity mirror following the gas jet [6–8]. Geometrical output coupling has a number of features which can be advantageous for some applications: there is no additional optical element in the resonator and therefore no additional dispersion, nonlinearity, bandwidth limitation or polarization discrimination for the fundamental radiation. The harmonics are not angularly dispersed. The output coupling efficiency can still be large for very high harmonic orders [7].

Different methods of geometrical output coupling have been proposed: Cavity-enhanced non-collinear HHG with two circulating pulses that overlap at a focus at some angle allows for the geometrical separation of the harmonic beam by output coupling through a gap between two cavity mirrors (or a hole or slit therein), as proposed by Moll et al. [6] with first experiments having been carried out by Ozawa et al. [9]. Moll et al. also discuss the possibility of using transverse eigenmodes of the resonator which exhibit a zero of the electric field on the optical axis, e.g. a $GH_{1,0}$ (Gauss-Hermite) mode. This allows for a reduction of the losses induced by a hole in a cavity mirror compared to using the fundamental transverse mode. However, the field distribution is disadvantageous for HHG, a drawback which might be overcome by using phase masks on the focusing mirrors [6]. Weitenberg et al. demonstrated the possibility of using a quasi-imaging resonator, i.e. a degenerate resonator with an obstacle in the beam path, where transverse modes are combined to avoid that obstacle and therefore experience small losses ("slit mode" avoiding a slit or "hole mode" avoiding a hole in a cavity mirror) [10,11]. Such a mode combination has been recently demonstrated to be well-suited for HHG by Pupeza et al. [8]. Another possibility is using a resonator for a circulating Bessel-Gauss beam, as described by Putnam et al. [12]. The conceptually simplest way to realize a geometrical output coupling is to use the resonator's fundamental transverse mode ($GH_{0,0}$) and a small hole on the resonator axis. Because the intensity is highest on the axis, the hole has to be very small in order to achieve small resonator losses and therefore a reasonable power enhancement [7].

The required sizes of the holes and the slits depend on the beam shape and size on the output coupling mirror. For a circulating $GH_{0,0}$ mode the hole radius must be only a few percent of the Gaussian beam radius in order to allow for an enhancement of a few hundred [7,11]. For a circulating slit mode in a quasi-imaging resonator the slit width should be in the order of magnitude of 10% of the Gaussian beam diameter, the slit mode's radius perpendicular to the slit being a factor of ~2.4 larger than the Gaussian radius (determined as the distance from the beam's centroid to the outer intensity maximum's 1/e^2 intensity level) [10]. For both geometries the sharpness of the edges is important: only the clear aperture is exploited for output coupling of high harmonics, and any vertical bulge or edge chips will increase the round-trip losses. Moll et al. used pierced mirrors for their experiments which were manufactured by a combination of mechanical drilling and laser machining with the polishing process afterwards. The hole diameters were between 100 µm and 300 µm [6].

In this paper we report manufacturing of "slit mirrors" and "hole mirrors" by Inverse Laser Drilling [13]. With this method we are able to drill holes down to a diameter of ~50 µm and slits down to a width of ~80 µm into uncoated standard fused-silica 6.35 mm thick mirror substrates. All holes and most of the slits are undercut. The mirrors are designed for enhancement resonators with an angle of incidence of a few degrees on the output-coupling mirrors, which is typical for ring resonators for cavity-assisted HHG [1–3,7,8]. The harmonic radiation is incident on the mirror under the same angle, and the undercut (a cone with 5.5° opening angle) represents a free aperture for the harmonics. The mirrors are curved. It allows for using the focusing mirror succeeding the focus in a bow-tie ring resonator as the output-coupling mirror. Using a plane hole or slit mirror or other output-coupling methods instead would require an additional optical element between the focus and the successive focusing mirror [1–3]. Thus using a focusing mirror can be considered an advantage because it simplifies the resonator design and avoids higher intensity on this element compared to the focusing mirror.

2. Method

One difficulty of drilling into dielectric materials by conventional laser ablation is that drilling dust fills the bore hole and has to be removed. In addition to that, the aspect ratio is limited by beam characteristics and geometry. However, dielectric materials like glasses are transparent for laser radiation of common wavelengths. In this case it is possible to drill inversely by propagating the laser radiation through the material and focusing it onto the opposite surface.

In our patented [13] process we focus the laser beam through the top surface onto the bottom surface of the bulk (Fig. 1). The beam is deflected by scanning mirrors in x- and y-direction, this way the desired geometry's surface layer is ablated. The sample is placed on a translation stage which allows for positioning in z-direction. This way the desired geometry can be ablated or "drilled" layer by layer. Drilling dust is removed by gravity.

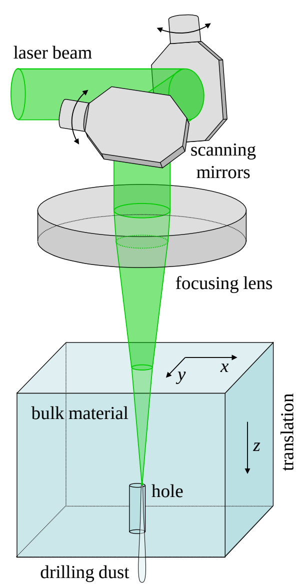

Fig. 1. Scheme of Inverse Laser Drilling.

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The beam quality of the lasers we used for the application is nearly diffraction limited. To preserve the beam quality and to get an undisturbed focus the sample's top surface needs to be polished. For the same reason the bulk needs to be optically homogeneous. The samples we used are examined both with interferometry and with a polarizer-analyzer-assembly. As expected the mirror substrates are of sufficient quality.

Three different lasers are used in our experiments: A commercially available frequency-doubled and q-switched diode-pumped slab oscillator (QSSO, Edgewave CX6II-DE) and a diode-pumped q-switched and frequency-doubled rod oscillator (QSRO, designed and assembled at Fraunhofer ILT) both generate pulses in the nanosecond regime. For shorter pulses we used a diode-seeded regenerative amplifier (DSRA, also developed at Fraunhofer ILT). The DSRA's pulse duration is variable between 250 ps and 1.5 ns [14]. In addition to that it is, unlike commercially available picosecond lasers, freely triggerable, which is very useful for our application because it is easier to control the position of each laser pulse on the target compared to free running lasers. The laser specifications have been measured at the position of the work piece and are summarized in Table 1.

Table 1. Laser specifications

	QSSO	***QSRO***	***DSRA***
wavelength	532 nm	532 nm	1,064 nm
max. power	15 W	1.2 W	2.2 W
max. pulse energy	15 mJ	1 mJ	1 mJ
max. repetition rate	100 kHz	2 kHz	10 kHz
pulse duration	~6 ns	>12 ns	250 ps - 1.5 ns
beam quality	$M^2 = 1.8$	$M^2 = 1.5$	$M^2 = 1.4$

3. Experiments

The following geometries have been structured successfully into 6.35 mm thick concave (radius of curvature 150 mm) uncoated fused-silica mirror substrates (Fig. 2):

- Eight slits into two substrates, widths from ~150 µm up to ~250 µm without undercut using the QSSO. The curved surfaces of the substrates have been of grinded quality and pointed downwards when being laser drilled. Afterwards they have been polished to optical quality.
- Eight slits into three substrates, widths from ~100 µm up to ~350 µm with a 5.5° undercut using the QSRO. Both substrate surfaces have been polished to optical quality before drilling with no surface treatment afterwards.
- Four slits into two substrates, widths from ~220 µm up to ~430 µm without undercut using the DSRA. Both substrate surfaces have been polished to optical quality before drilling with no surface treatment afterwards.
- Several holes with different diameters down to ~50 µm with a 5.5° undercut using the DSRA. Both substrate surfaces have been polished to optical quality before drilling with no surface treatment afterwards.

The diameter of all substrates is 25 mm. The slits' length is 4 mm. They are shaped as elongated holes instead of rectangles to avoid any additional stress induced by sharp corners in the bulk. For drilling the slits with undercut we positioned the substrates with the plane surface pointing downwards. This required propagation of the laser beam through the curved surface, which caused no additional problems to the ablation process although the optical axes of the laser beam and the curved substrate have in general not been parallel to each other.

The lasers have been set to pulse energies of about 250 µJ (QSSO, QSRO) and 90 µJ (DSRA). The spot radii have been set to 14 µm, 9 µm and 10 µm, respectively. The pulse duration of the DSRA has been set to ~1 ns. Brief tests have shown that best results in terms of process stability seem to be achievable with this pulse duration. However, those examinations have not been sufficient yet.

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All structures have been examined with a microscope (Keyence VHX-2000), a polarizer-analyzer-assembly and a white light interferometer (Zygolot NV6k-0135).

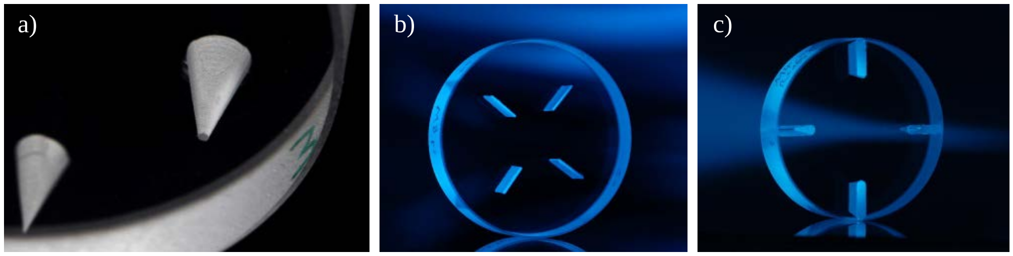

Fig. 2. Laser-manufactured mirror substrates with 25 mm diameter: circular holes with under-cut (a), slits without undercut (b), slits with undercut (c).

4. Results

Slits

Microscope examinations reveal shell breaks (chipping) at all laser-structured edges (Fig. 3). The chipped area can be called "lost area" because in general it is adding losses to the enhancement in the cavity without participating to the output coupling. To quantify the amount of the chipping and to make an easy comparison of the amount of lost area depending on the laser parameters the distances between the slit's edge and the outer limit of the chips on each side of the slit have been measured with the microscope for each slit. The arithmetic means and the standard deviations of this "lost width" are given in Table 2. While judging the results the polishing of the substrates structured with the QSSO after the drilling process has to be taken into account: chipping is less strong when drilling into grinded surfaces and in addition to that polishing after drilling reduces the chipped area. Best results concerning the size of the chippings have been achieved with the DSRA.

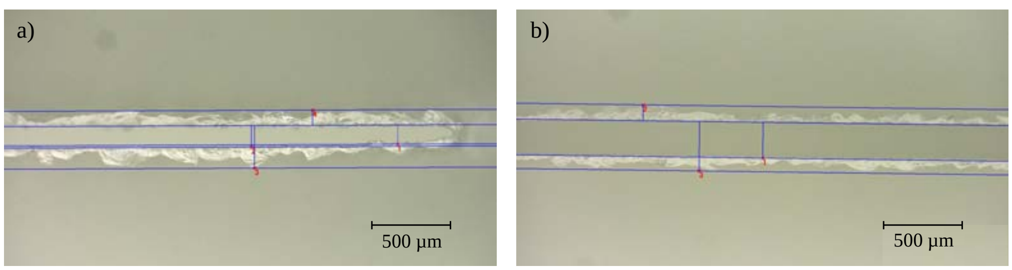

Fig. 3. a) Microscopy of a 120 µm wide slit structured with the QSRO. The distance between the outer limits of the shell breaks is ~350 µm. b) Microscopy of a 220 µm wide slit structured with the DSRA. The distance between the outer limits of the shell breaks is ~400 µm.

Table 2. Distance between the slit's edge and the outer limits of edge chips

	arithmetic mean	**standard dev.**
QSSO	98 µm	13 µm
QSRO	164 µm	29 µm
DSRA	87 µm	18 µm

To determine whether the manufacturing process induced stress into the substrates, they have been examined with the polarizer-analyzer-assembly. Inner stress is detected especially at the ends of the slits. It has been removed by tempering (Fig. 4).

#193855 - $15.00 USD Received 20 Sep 2013; revised 18 Oct 2013; accepted 19 Oct 2013; published 30 Oct 2013

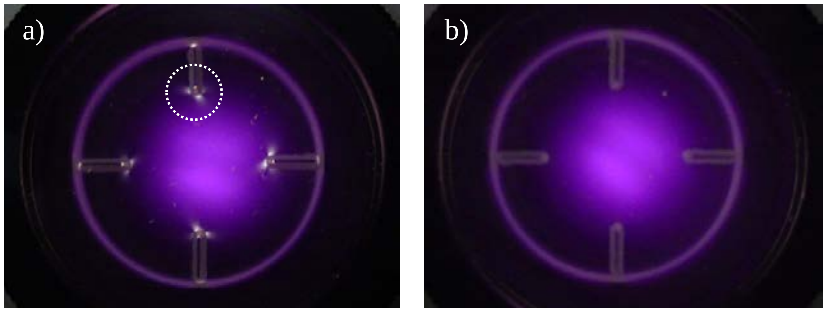

Fig. 4. Substrates between crossed polarizers. Stress in the substrate after laser processing is revealed (a, marked by the white dashed circle), after tempering is not detectable any more (b).

After the manufacturing process the substrates' surface profiles have been examined with the white light interferometer right after processing, after tempering and after polishing. The influence on the surface is depicted in Fig. 5. The false color representations on top show the surfaces' profiles around the slits: for one substrate right after processing without any further treatment (a) and after tempering (b), and for a different substrate after polishing (c). The surface profile along the section view indicated by the lines is depicted underneath the false color representations. The sections views have been taken roughly perpendicular to the slits approximately in their middle. Because the substrates are curved a sphere with the proper radius is subtracted from the data, thus the measurement is supposed to show a plane surface with just the process-induced deviations. This proper radius of curvature has been determined with the white light interferometer in the middle of the substrates.

After drilling the mirror surface is bent upwards throughout an area of about > 1 mm around the slits by more than 100 nm in the peak. The affected area is in the order of magnitude of the laser spot size in a typical enhancement cavity.

Polishing rounds off the slits' edges downwards. The affected area and the depth of deformation are in the same order of magnitude as they are without polishing.

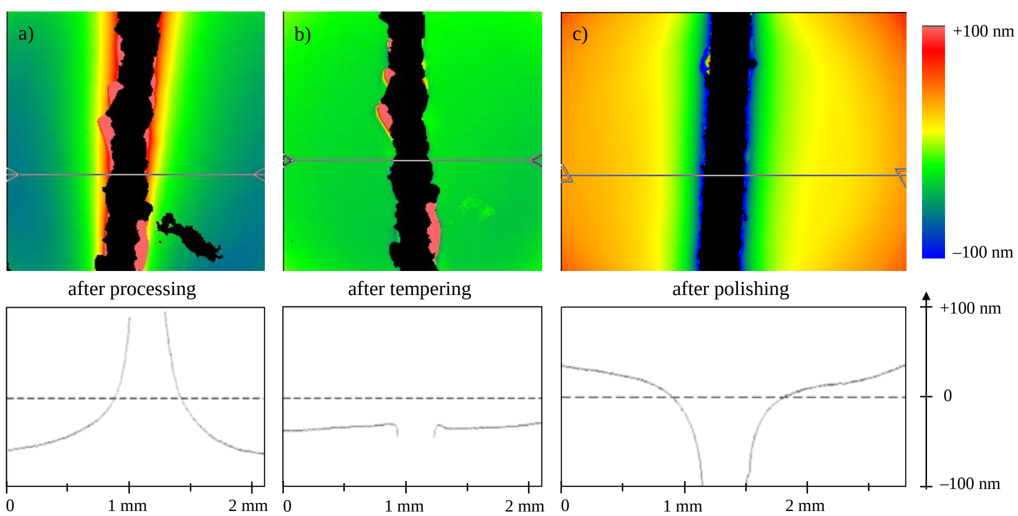

Fig. 5. Surface profiles around slits after processing (a), after tempering (b) and after polishing (c). See text for further explanations.

If, however, the stress is relieved by tempering, the surface re-flattens (see Fig. 5, which depicts the same slit before (a) and after tempering (b)). The phase error, which is added to a laser beam reflected off this surface, is significantly below $\lambda/10$. Some small areas of the substrates have been spoiled by the tempering process due to burned dust. However, the deposi-

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tion of dust particles can be avoided by conducting the tempering process in a clean room environment.

Holes

The hole mirrors structured in this work were used in a cavity with a circulating Gaussian beam [7]. In this case the hole's center is located on the optical axis where the intensity of the Gaussian beam is highest. Thus it is very important for this application to keep the lost area as small as possible for a reasonable enhancement. Since the results with the slits showed that the lost area is smallest with the DSRA, the holes have only been drilled with this laser.

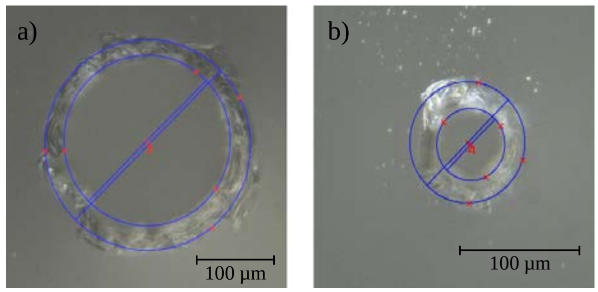

Fig. 6. Laser-drilled holes with different diameters. a) ~160 µm inner diameter and ~200 µm outer diameter. b) ~60 µm inner diameter and ~100 µm outer diameter.

Quantification of the chippings' extent has been done differently than for the slits: one circle has been approximately fitted to the free aperture (inner diameter), and one circle to the lost area (outer diameter) as depicted in Fig. 6. Although this method does not provide an exact quantification of the chipping, it was determined that in almost all cases the outer diameter is about 40-55 µm larger than the inner diameter. This is independent of the inner diameter's magnitude. The size of the lost area relatively to the hole's area decreases with increasing free aperture. Therefore, a resonator design with a large mode area on the output coupling mirror is useful [15].

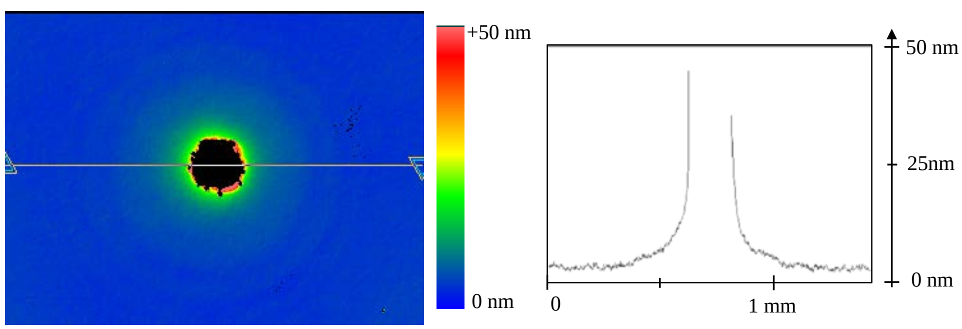

Fig. 7. Surface profile of area around the circular drilling. No further treatment has been done after processing. The height of the deformation is about one fourth compared to the slits and the affected area is smaller.

The surface areas around the holes have also been examined with the white light interferometer. At each hole the edge is also bent upwards, but the affected area is smaller compared to the substrates with slits, and the height of the bulge is only about one fourth compared to the slits (see Fig. 7). Hence, we neither tempered nor polished the substrates with the circular holes. It is expected that the surface also bends down at the edge during the polishing process, and we also did not want to risk tempering the substrates. The expected improvement of the surface by relieving the stress would have been too small compared to the risk of damaging the mirrors through burning dust particles.

5. Discussion

The radiation in an enhancement resonator impinges on a mirror many times depending on the enhancement, and the sensitivity to aberrations is therefore also enhanced. Hence, we expect that the deformation of the mirror surface around the holes or slits in the mirrors has a possibly deleterious effect on the resonator enhancement, even if the height of the deformation is small compared to the wavelength. We performed simulations in order to evaluate the influence of the deformation.

We examine the two situations of a circulating Gaussian beam in a resonator with a hole mirror and of a circulating slit mode in a quasi-imaging resonator with a slit mirror. The surface deformation is assumed to be exponentially decaying with a varying height h and a transverse extension l (Fig. 8(a)), which approximates the experimental observation shown in Figs. 5 and 7. The size of the hole and the slit relative to the beam size a/w, the extension of the deformation relative to the beam size l/w and the reflectivity of the input coupling mirror R_1 of the enhancement resonator are chosen to represent reasonable experimental situations [7,10]. These chosen parameters yield for both cases the two situations of an enhancement of a few hundred and of about one hundred in absence of the deformation. The parameters are denoted in Fig. 8 together with some further details in the caption.

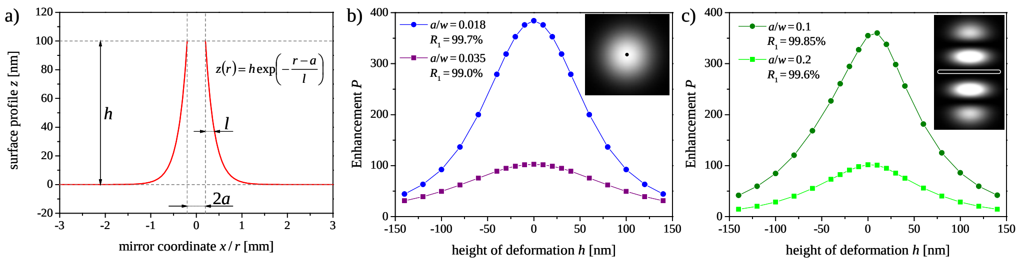

Fig. 8. Deformation of the mirror surface (with sphere subtracted) assumed for the calculations, with hole radius or half slit width a, and height h and transverse extension l of the deformation (a). Enhancement P (circulating power to incident power) as a function of the height h of the deformation in an enhancement resonator with circulating fundamental mode and hole mirror (b) and in a quasi-imaging resonator with a circulating slit mode and slit mirror (c). The inlays show the transverse mode at the position of the hole or slit mirror. The transverse extension of the deformation is assumed to be $l/w = 0.03$ for the hole and $l/w = 0.2$ for the slit. For a beam radius of $w = 2.7$ mm as in [7] and $w = 1$ mm as in [10] this corresponds to $l = 0.08$ mm (compare Fig. 7) and $l = 0.2$ mm (compare Fig. 5), respectively. For the simulation of the slit mode, a resonator loss factor of $R = 0.99955$ and an aperture with $A/w = 3.0$, as well as spherical aberration (0.5 mrad per focusing mirror) are included (see [11] for details). The spherical aberration leads to a slight asymmetry of the curves in (c).

For the Gaussian beam the enhancement P is calculated from the loss factor R which results from the incomplete overlap u of the normalized field with and without hole and deformation, i.e. $P = (1 - R_1)/(1 - (R \cdot R_1)^{½})^2$ with $R = U^2$ and $U = |u|^2$ [11]. For the slit mode the circulating field is modeled as a superposition of Gauss-Hermite modes $GH_{0,0}$, $GH_{4,0}$ and $GH_{8,0}$, which are coupled at the slit and the deformation [11].

The results of these calculations are depicted in Fig. 8. It shows that for the considered situations the deformation of the mirror surface has a large impact on the performance of the enhancement resonator. The enhancement is already significantly smaller for a deformation of a few 10 nm and drops to a fraction at a deformation of 100 nm. This holds for both a surface bending upwards or downwards.

The untreated slit mirrors have a surface bulge of more than 100 nm which severely limits the enhancement. Through polishing the surface the slit's rim suffers about the same amount of deformation, this time with the surface bending downwards. The affected area is in the order of magnitude of 1 mm from the slit's rim (Fig. 5) which is significantly larger than the lost area caused by the chipping (Fig. 3). This means that it is possibly not advantageous to eliminate the chips by polishing off some 10 µm of the substrate, because the resulting deformation

#193855 - $15.00 USD Received 20 Sep 2013; revised 18 Oct 2013; accepted 19 Oct 2013; published 30 Oct 2013

can reduce the enhancement more than the chipping itself. Drilling of the circular holes causes less deformation to the surface. Because the bulge is only a few 10 nm high and the affected area is less expanded than at the slit mirrors, the influence on the enhancement is smaller. Still it cannot be neglected and presumably it is of advantage if those substrates are also tempered.

It is descriptive that the deformation for the case of the Gaussian beam causes significant losses reducing the enhancement, because the intensity is highest around the hole. The slit mode on the other hand has a small intensity around the slit. The influence of the deformation is comparably strong (Fig. 8). This holds even though in the calculation the deformation is assumed to be more expanded than for the hole; with $l/w = 0.2$ it is still small compared to the distance $x/w = 0.7$ of the first intensity maximum from the optical axis (inset of Fig. 8(c)). The reason is that the "slit mode" in a quasi-imaging resonator is more sensitive to aberrations which distort the phase-correct combination of the Gauss-Hermite modes.

6. Conclusion and outlook

A successful method for fabricating laser mirrors with small openings by Inverse Laser Drilling has been demonstrated. These mirrors can be used for geometrical output coupling of high harmonics from enhancement resonators. Slits down to a width of 80 µm with and without undercut and circular holes down to a diameter of 50 µm with undercut have been realized. Lasers in the ns and the sub-ns regime have been used, and the influence of the process on the substrates has been examined. The openings' edges are chipped and the surface is bent upwards which can cause losses and aberrations in an enhancement cavity as investigated by simulations. The amount of chipping depends on the type of laser used for the drilling. The surface bending is caused by stress within the material induced by the process and can be relieved by tempering.

Recently, the hole mirrors have been used in an enhancement cavity for successful generation and output coupling of high harmonics down to a wavelength of 11.45 nm [7]. The damage thresholds of the mirrors have not been investigated systematically. However, in the cavity described in [7] the mirrors withstood circulating powers of up to 8 kW at a pulse duration of 175 fs for several hours of operation. Together with the beam radius of 2.7 mm and the repetition rate of 78 MHz, this leads to intensities of 5.1 GW/cm^2 on the mirror. To the best of our knowledge this is the first demonstration of geometric output coupling of high harmonics. These holes have thus played a major role in demonstrating a new class of resonators for extreme nonlinear optics.

Most recently the slit mirrors have been used for output coupling of high harmonics from a quasi-imaging resonator with a circulating slit mode. Up to 11 µW of output coupled power contained in the 17th harmonic at 61.2 nm have been demonstrated [8]. As circulating power in enhancement cavities will be scaled in the future [15], further investigations of the mirrors concerning damage thresholds, thermal properties, degradation, coating quality at the openings' edges etc. will be necessary. If advantageous for the application, drilling into substrate materials other than fused silica will be examined. The focus on future works will be laid on reducing the chipping to minimize the lost area and to avoid local field enhancement at the sharp chip's edges. It is also possible to structure more complex geometries of openings into mirror surfaces than described in this paper if desired for any other application.

In general the Inverse Laser Drilling enables structuring of high aspect ratio geometries into dielectric materials, e.g. for manufacturing of photonic crystal fibers as described in [16]. In that work an array of holes with a diameter of ~600 µm has been drilled into an 80 mm long fiber preform.

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