Photothermal measurement of absorption and wavefront deformations in UV optics

K. Mann, A. Bayer, U. Leinhos, M. Schöneck, B. Schäfer

Laser-Laboratorium Göttingen e.V., Hans-Adolf-Krebs-Weg 1, D-37077 Göttingen (Germany),

Email: kmann@llg-ev.de

Abstract: A measurement system for quantitative registration of transient and irreversible lens effects in DUV optics induced by absorbed UV laser radiation was developed. It is based on a strongly improved Hartmann-Shack wavefront sensor with an extreme sensitivity of $\lambda/10,000$ rms @ 193nm, accomplishing precise on-line monitoring of wavefront deformations of a collimated test laser beam transmitted through the laser-irradiated site of a sample. Caused by the temperature dependence of the refractive index as well as thermal expansion, the initially plane wavefront of the test laser is distorted into a convex or concave lens, dependent on sign and magnitude of index change and expansion. The observed wavefront distortion yields a quantitative measure of the absorption losses in the sample. Results for fused silica and CaF$_2$ are presented.

Keywords: Hartmann-Shack wavefront sensor, thermal lens, DUV absorption

1. Introduction

Ever increasing demands on the resolution in microlithography call for a comprehensive information about the imaging quality of critical components under process conditions. Along with aging mechanisms as color center formation or compaction the unavoidable absorption within the bulk and on the surfaces raises the temperature, which in the case of inhomogeneous irradiation leads to local changes of the refractive index and thus results in the development of a thermal lens. In order to estimate the influence of thermal lensing on optical quality and perhaps consider it during the design stage of lithography objectives, the knowledge of material absorption at operation wavelength is highly demanded. Unfortunately, the most commonly used techniques in this field, i.e. calorimetry [1] and conventional transmission measurements suffer from several problems which can be avoided by a direct inspection of the laser induced wavefront deformation. The advantages of the photo-thermal approach compared to calorimetry are the fast measurement, the more versatile sample dimensions, better spatial resolution and the direct measurement of optically relevant quantities. Above that, the dynamics of the process can be evaluated. Compared to a transmission measurement the photo-thermal method is more sensitive, gives more stable values for low absorption and in particular, contrary to transmission there is no influence of scattering. Furthermore the effects of laser induced densification on the transmitted wavefront can be investigated [2].

Here we present a setup for quantitative determination of laser induced wavefront deformations and absorption in DUV optics. Following a brief section on theoretical foundations of Hartmann-Shack
wavefront sensing and thermal lensing the experimental setup is presented, and results for UV grade fused silica and CaF$_2$ are discussed.

2. General
2.1 Hartmann-Shack wavefront sensor
The Hartmann principle [3]-[5] is based on an orthogonal or hexagonal array of lenses (Hartmann-Shack) or pinholes (Hartmann), which divides the incoming beam into a large number of sub-rays. The total irradiance $I_{ij}$ and the position of the individual spots are monitored with a position sensitive detector placed at a distance $f$ behind the array. The displacement of the spot centroid $x'$ with respect to a plane wave reference position $x''$ measures the direction of the time averaged transverse Poynting vector and thus the wavefront gradient $\beta_{ij}$, averaged over a sub-aperture $(i,j)$, according to

$$
\begin{bmatrix}
\beta_{x} \\
\beta_{y}
\end{bmatrix}
= \frac{1}{f} \begin{bmatrix}
\frac{\partial w}{\partial x} \\
\frac{\partial w}{\partial y}
\end{bmatrix}_{ij}.
$$

(1)

A series expansion of the wavefront $w$ in e.g. Zernike polynomials $P_l$ with expansion coefficients $c_l$, i.e.:

$$w(x, y) = \sum_{l=0}^{N} c_l P_l(x, y)$$

(2)

in combination with a familiar least square approach:

$$
\sum_{i,j} \left[ \left( \sum_{l} c_l \frac{\partial P_l}{\partial x}(x_i, y_j) - \beta_{x, exp}^{ij} \right)^2 + \left( \sum_{l} c_l \frac{\partial P_l}{\partial y}(x_i, y_j) - \beta_{y, exp}^{ij} \right)^2 \right] = \min
$$

(3)

then leads to a linear system of equations for $c_l$, which can efficiently be solved by standard techniques of linear theory.

From the reconstructed wavefront the global parameters peak-valley deformation $w_{pv}$ and rms deformation $w_{rms}$

$$w_{pv} = \max_{ij} w_{ij} - \min_{ij} w_{ij} \quad \text{and} \quad w_{rms} = \sqrt{\frac{\sum_{i,j} (w_{ij} - \bar{w})^2}{N}}$$

(4)

can be determined to quantify photo-thermal lensing.

2.2 Thermal lensing
Consider a cylindrical specimen $G$ irradiated by a circular laser beam traveling in $z$-direction as shown in fig. 1. The transient temperature change $\delta T(r,t)$ is obtained by the solution of the heat equation with appropriate boundary conditions on $\partial G$:

$$
\begin{align*}
c_p \rho \frac{\partial \delta T(r,t)}{\partial t} + \nabla \cdot (\kappa \nabla \delta T(r,t)) + \mu \cdot I_p(r,t) &= 0 \quad r \in G \\
n \cdot (\kappa \nabla \delta T(r,t) + \beta \delta T(r,t) n - \beta \gamma_p(r,t) e_z) &= 0 \quad r \in \partial G.
\end{align*}
$$

(5)

In eq. (5) $\mu$ and $\beta$ are the bulk and surface absorption coefficients respectively, $I_p$ denotes the laser power density distribution, $\chi$ the coefficient of heat transfer, $\lambda$ the thermal conductivity, $c_p$ the specific heat, $\rho$ the density and $n$ the outward directed surface normal.
Given certain initial and irradiation conditions, sample dimensions and material parameters, eq. (5) can be solved numerically for any instant of time. However, usually $\mu_l, \beta << 1$ holds and from linearity the solution $\delta T(r,z,t)$ can be expressed as:

$$\delta T(r,z,t) = P \left[ \mu V(r,z,t) + \beta S(r,z,t) \right],$$

where $P$ is the average laser power. In (6) the volume and surface terms $V(r,z,t)$ resp. $S(r,z,t)$ depend on specimen dimension, laser profile as well as on $\lambda, c, \rho$ and $\kappa$, but are independent of $P, \mu$ and $\beta$.

As a consequence of the inhomogeneous temperature distribution, the refractive index varies locally according to

$$n(\delta T(r,z,t)) = n_0 + \frac{\partial n}{\partial T} \delta T(r,z,t)$$  \hspace{1cm} (7a)$$

and the elongation of the sample $\delta l(r)$ writes:

$$\delta l(r,t) = \int_0^l \frac{\partial l_t}{\partial z}(r,z,t) \, dz.$$  \hspace{1cm} (7b)$$

According to eq. (7a, b ) the wavefront of a well collimated probe beam, traveling parallel to the specimen z-axis picks up a wavefront deformation $\delta w(r,t)$:

$$\delta w(r,t) = (n_0 - 1) \int_0^l \frac{\partial n}{\partial T}(r,z,t) \, dz + \frac{\partial n}{\partial T} \int_0^l \delta T(r,z,t) \, dz \quad \sim \quad P \left[ \mu V'(r,t) + \beta S'(r,t) \right].$$  \hspace{1cm} (9)$$

As in eq. (6), the form factors $V'$ and $S'$ are independent of $P, \mu$ and $\beta$.

3. Experimental

3.1 Setup

Figure 2, Setup for measurement of the laser induced photo-thermal wavefront deformation
The setup for photo-thermal measurements is shown in Figure 2. It consists of a nitrogen purged chamber, which contains all optically relevant components. The collimated ArF laser ($\lambda=193\text{nm}$, pulse length $\sim 15\text{ns}$, repetition rate $150\text{Hz}$) irradiates the samples collinearly, and the probe beam, a $639\text{nm}$ fiber coupled diode laser, meets the specimen at $8^\circ$. For homogeneous illumination the probe laser is expanded 25-times and, after passing the sample demagnified again to fit the detector area of the Hartmann-Shack sensor. Both camera- and instant power data are fed into a standard PC for online data processing. For defined irradiation conditions the excimer laser beam is expanded and confined to a round flattop profile of $\varnothing 7\text{mm}$ by a circular aperture. Furthermore, the beam line is provided with a PC controlled shutter in order to achieve variable heating and cooling intervals. The Hartmann-Shack sensor consists of a 12bit CCD camera with 1280-1024 pixels. The microlens plate was a $0.3\text{mm}$ pitch quartz array with $f=40\text{mm}$ plan-convex lenslets placed at a distance $d=50\text{mm}$ in front of the detector. For noise reduction up to 256 frames per record were sampled and the oxygen pressure was purged below $100\text{ppm}$ before starting the measurement.

![Figure 3](image)

**Figure 3**, Recorded spot diagram (left) and reconstructed wavefront (right) of a quartz cylinder $\varnothing 25\text{mm} \times 45\text{mm}$, irradiated at $193\text{nm}$ with an intensity of $\sim 100\text{mW/cm}^2$. The arrows indicate the relative amount of spot displacement.

A typical spot pattern from the Hartmann-Shack sensor is shown in Figure 3 on the left. The arrows indicate the local displacement with respect to the unirradiated position of the spots during heating of a quartz sample with $100\text{mW/cm}^2$. The right picture shows the reconstructed wavefront exhibiting a peak-valley deformation of $2\text{nm}$. For a spherical surface this would correspond to a defocus term of approximately $5\text{km}$.

### 3.2 Calibration

![Figure 4](image)

**Figure 4**, left: Calibration of the photo-thermal measurement setup using quartz sample ($d=25\text{mm}$, $l_c=45\text{mm}$) with central bore (= dia. of laser beam) and resistor chain; right: peak-valley and rms wavefront deformation plotted vs. electrical power. Solid lines represent the linear fit to the measured data.

In order to gain absolute values for the absorption the setup was calibrated by measuring the photo-thermal signal of a resistor heated probe as sketched in Figure 4 on the left. A probe with a central
bore corresponding to the laser beam diameter and of same material and dimensions as the samples under test was heated by a resistor chain glued within the bore. Thus, assuming that heat production is approximately constant in z-direction, the temperature distribution in the outer zone will be the same as in the laser heated case, provided the surface absorption can be neglected. Assuming further, that elastic constraints are small, the rms or peak-valley deformations of a wavefront extrapolated into the obscured area can be plotted over the absorbed power, and from the coefficients a and b of a linear fit one obtains a calibration factor that permits the absolute determination of the absorption coefficient $k$ (base 10) according to:

$$k = \frac{w_{\text{rms}} - b}{a \cdot P \cdot l_c} \log(e) \quad (10)$$

The estimated fit parameters of $a_{\text{rms}}=140\text{nm/W}$ and $b_{\text{rms}}=0.02\text{nm}$ indicate a detection limit of ~20…50pm for the wavefront rms corresponding to 0.12…0.35mW absorbed power.

Alternatively a calibration is possible from results of numerical simulations. According to eq. (9) the wavefront rms value can be calculated for various sample geometries, any set of material coefficients as well as arbitrary laser parameters. Then, as a consequence of linearity, a straight line $w_{\text{rms}} = a \cdot k + b$ and therefore the $k$-factor for a given laser fluence can be determined from $w_{\text{rms}}$.

4. Result

4.1 Absorption of fused silica at 193nm

On the left of Figure 5 the rms deformation is plotted vs. fluence for three quartz samples of equal size taken from different charges of the same vendor. The samples exhibit non-linear absorption which shows up in the parabolic increase of the deformation with laser fluence. The resulting absorption coefficient $k$ is shown on the right, ranging from $1 \times 10^{-4}$ to $5 \times 10^{-4}$/cm. The coefficients of linear absorption given by the intercept with the ordinate are even lower, e.g. $5 \times 10^{-5}$ extrapolated for probe 3. As has been shown earlier using laser calorimetry [1], the slope of the straight lines in Figure 5 (right) depends on the pulse duration due to a two-step excitation process.

![Figure 5](image-url)

**Figure 5**, (left): Measured $w_{\text{rms}}$ data as a function of laser fluence (@ 150Hz, $\lambda=193\text{nm}$, average over 5 measurements) for three uncoated quartz substrates of equal size. Right: Corresponding absorption coefficients $k$ (calculated with calibration factors from resistor heated sample). The slopes indicate the presence of two-photon non-linear absorption.

The results from a calibration based on numerical simulations of eq. (6-8) yield qualitatively the same behaviour, the absolute values, however, are about 20-40% smaller. Considering the small magnitude in the observed wavefront-rms values, the unknown surface absorption, uncertainties in material parameters as well as – in the case of resistor heated calibration probes - errors introduced by
wavefront extrapolation into the obscured region, these results are quite satisfactory. Further improvements of signal-to-noise ratio, e.g. by an increase of the repetition rate up to 1-2kHz, as well as more accurate numerical calculations in combination with an additional calibration procedure based on a reference “gray” sample will certainly yield a better consistence of future results.

4.2 First measurements of laser induced wavefront distortion in CaF$_2$

Figure 6 shows laser induced wavefront deformation in CaF$_2$ obtained at 248nm irradiation wavelength for two samples of different lengths. In contrast to fused silica, which always exhibits a positive lens effect, a sign reversal was observed for CaF$_2$. This is due to the fact that, other than in quartz, the change of refractive index with temperature is negative. Although the thermal expansion is almost 40 times higher compared to quartz, $dn/dT$ normally overcompensates the specimen expansion, leading to a negative thermal lens effect (Figure 6, left).

![Figure 6](image)

**Figure 6**, Photothermal measurement of CaF$_2$ samples (left: $l_s$=20mm, right: $l_s$=70mm) using an irradiation wavelength of 248nm and 50mW/cm$^2$ power density. A sign reversal of the thermal lens is observed.

However, no such sign reversal was observed for another CaF$_2$ sample of different dimension (Figure 6, right). A comparison with finite difference (FD) calculations performed for different geometries and absorption coefficients indicates that a strong surface absorption could be responsible for this positive lens effect. Unfortunately, the large dependence of the photo-thermal signal on surface absorption renders calibration much more cumbersome compared to fused silica. Instead of resistor heating, different techniques based on reference samples, thickness series and numerical calculations must be considered. These attempts will be evaluated in the near future.

5. Conclusion

A novel photo-thermal measurement technique was developed, allowing to monitor thermal lens as well as compaction effects in optical materials. Using a high-sensitivity Hartmann-Shack sensor this multiple beam technique can detect wavefront distortions in real-time with a resolution of about $\lambda/10,000$, corresponding to < 100pm. The quantitative evaluation of the thermal lens data accomplishes a fast assessment of both absorptance properties and imaging quality of the optics under test. An absolute calibration of the measured data is possible by comparison with signals from a resistor heated specimen as well as from numerical simulation. The advantages of the novel setup in comparison to calorimetry are the much faster measurement (no equilibrium, only wavefront gradients), the more flexible sample dimensions and the possibility to investigate the dynamics in imaging quality. In comparison to transmission measurement the setup gives more stable values, and there is no bias from scatter losses. Thus, the new method may be regarded an alternative to both techniques.
References

[1] Görling, C., Leinhos, U., Mann, K., "Comparative studies of absorptance behaviour of alkaline-earth fluorides at 193 nm and 157 nm.", Appl. Phys. B 74, 259 (2002)

[2] Borelli, N.F., Smith, C., Allan, D.C., Seward II, T.P., “Densification of fused silica under 193-nm excitation”, J. Opt. Soc. Am. B, 14 (7), 1606-1625, (1997)

[3] Neal, D.R., Alford, W.J., Gruetzner, J.K., Warren, M.E., “Amplitude and phase beam characterization using a two-dimensional wavefront sensor”, Proc. SPIE 2870, 72 (1996)

[4] Schäfer, B., Mann, K., “Investigation of the propagation characteristics of excimer lasers using a Hartmann-Shack sensor”, Rev. Sci. Instrum. 71, 2663 (2000)

[5] Schäfer, B., Mann, K., “Determination of beam parameters and coherence properties of laser radiation by use of an extended Hartmann-Shack wave-front sensor”, Appl. Optics, 41 No. 15, 2809-2817 (2002)

[6] Born/Wolf, [Principles of Optics], Cambridge University Press, 7th Ed., Sect. 15.4, 823pp (2001)