Carbon buffer layers for smoothing substrates of EUV and X-ray multilayer mirrors

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ABSTRACT

Smoothing of surfaces by thin film deposition is facilitated by methods which release hyperthermal particles on the substrate. One of these techniques is pulsed laser deposition (PLD), with high kinetic particle energies of up to several 100 eV. The concrete energy distribution of the particles can be widely influenced by the laser power density. We investigated the deposition of carbon layers by PLD on numerous substrates with rms-roughnesses between 0.15 and 0.75 nm using different laser power densities and film thicknesses. It turns out that a better smoothing can be obtained with higher laser power densities, whereby diamond-like carbon films are created. With typical thicknesses of $d_C = 100$ nm, the rms-roughness is reduced from 0.75 nm to 0.55 nm and from 0.32 nm to 0.18 nm. Accordingly by applying smoothing carbon buffer layers, the EUV reflectance of Mo/Si multilayers on rough substrates is increased from typically 60 % to > 65 % on substrates with initial roughnesses of 0.75 nm.

Keywords:

1. INTRODUCTION

The production of high-reflection EUV and X-ray mirrors requires extremely smooth substrate surfaces for the deposition of dedicated nanometer multilayers. In order to achieve EUV reflectances in the range of 70 % ($\lambda = 13.5$ nm), a surface finish with rms-roughnesses of 0.1-0.2 nm is needed. For such substrates meeting the specifications for surface finish and figure at the same time is very complicated and often unaccomplished. More modest requirements particularly for the micro-roughness of the substrate surfaces are very desirable.

The first option to obtain high reflectances also on rougher substrates is to apply deposition techniques which are less sensitive to substrate roughness. Spiller et al. have shown that ion beam sputter deposition is a powerful method that can smooth out substrate roughness [1]. EUV reflectances of up to 66.8 % were obtained on substrates with rms-roughnesses of 0.39 nm [2].

The second option is to use smoothing layers between the rough substrate surface and the optical film [3, 4]. In this case the high-frequency roughness of the underlying substrate is smoothed by the appropriate deposition of smoothing layers. One material with a high potential to smooth substrate surfaces is carbon [3, 4]. Independently on the concrete deposition method, extremely smooth diamond-like carbon films can be deposited, if the energy of the condensing particles on the substrate surface is > 50 eV [5]. It was already shown that the reflectance on EUV and X-ray multilayer mirrors can be increased by applying such carbon buffer layers [6, 7]. For the EUV spectral range the enhancement of the reflectance amounts to 0.7 % on substrates with surface roughnesses of 0.55 nm using carbon layers with thicknesses of approximately 15 nm [7].

In this paper we will focus on the second approach to increase the reflectance on rough substrates. For the deposition of carbon smoothing layers we use PLD [8]. This method is known to deliver particles with high kinetic energies as

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necessary to obtain smooth layers. We will report about atomic force microscopy (AFM) investigations on carbon layers, which were deposited on substrates of various surface roughnesses. We will show how carbon layer thickness and deposition parameters will affect the resulting roughness. Furthermore different carbon layers were coated with Mo/Si multilayers by magnetron sputter deposition (MSD). The corresponding EUV reflectance was measured and the values are discussed in connection with the AFM investigations and with measurements on super-polished substrates.

2. LAYER GROWTH MODELS

The surface evolution of thin films can be described by two competing effects: Stochastic surface roughening due to random shot noise of the deposition process and smoothing due to surface relaxation [9, 2]. The general description of the surface topography with heights $h_i(f)$ for the growth of layer i on the surface of layer (i-1) depending on the frequency f of the roughness is given by

$h_i(f) = d_i(f) + a_i(f) \cdot h_{i-1}(f)$ [9, 2]

with $d_i(f)$ as the intrinsic thickness distribution of the growing layer i, $a_i(f)$ as replication factor from surface (i-1) to surface i.

The relaxation mechanism strongly depends on the deposition technique and its parameters used. Herring [10] has identified several relaxation mechanisms characterized by different integer values n, which have an impact on the replication factor

$a_i(f) = \exp\text{-}(\nu_i d_i q_s^n)$,

with $q_s = 2\pi f$, d_i = film thickness and ν_i as a measure for the distance over which particles can relax [2]. This formula indicates that lower frequencies f can be easier damped with low numbers of n and with higher film thicknesses. Since n = 1 holds for viscous flow, n = 2 for condensation and re-evaporation, n = 3 for bulk diffusion and n = 4 for surface diffusion [10], the pulsed laser deposition technique with n = 1 or n = 2 should be a very attractive candidate for an effective smoothing of surface roughnesses. Figure 1 shows the replication factor for different values for d_i and ν_i with n = 1 and n = 2. It indicates that the easiest way to decrease the surface roughness is to increase the layer thickness of the coating.

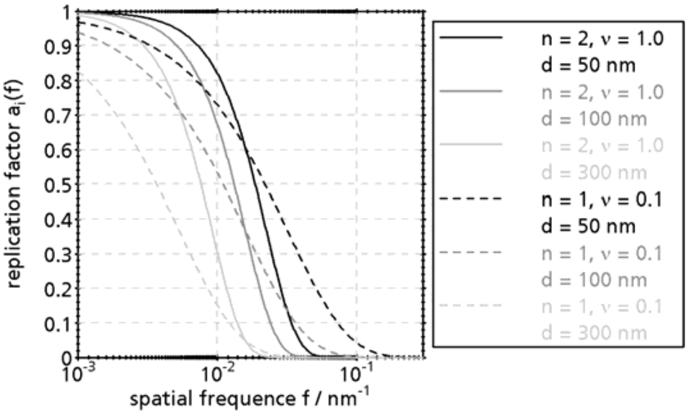

Fig. 1: Replication factor $a_i(f)$ for n = 1, ν = 0.1 and n = 2, ν = 1.0. The best replication suppression can be obtained with thick smoothing layers, large values ν and for n = 1.

3. EXPERIMENTAL SETUP

3.1. PULSED LASER DEPOSITION (PLD)

The interaction of a focused pulsed laser beam with solid state materials inside a vacuum chamber results in the emission of strongly directed plasma, the so-called plasma plume. The plasma consists of a mixture of atoms, ions, electrons and clusters. The kinetic energies of the ablated particles are typically distributed as shown in Figure 2. Deviating from conventional deposition techniques like thermal or electron beam evaporation, where particle energies in the order of 0.1 eV occur, PLD is characterized by a significant amount of particles with higher kinetic energies. Using lasers with pulse durations in the nanosecond range, the kinetic energy of the ions in the plasma can reach values of up to 2000 eV with typical mean energies between 100-400 eV. Higher mean energies are observed for longer laser wavelengths [11].These hyperthermal particles are necessary to obtain a layer growth mechanism which follows the forth zone of the structure zone model of Guenther [12]. In this zone, the layer formation is characterized by smooth vitreous-amorphous films.

One of the main drawbacks of static PLD is that due to the strong alignment of the plasma plume, the coating uniformity necessary for optical elements can only be reached on small areas of appr. $10 \times 10\ \mathrm{mm}^2$. In order to overcome this problem, an arrangement was developed that enables the deposition of large areas by PLD [13] (LA-PLD, Fig. 3). Using this technique, uniform smoothing layers can be deposited on flat or curved substrates with diameters of up to 150 mm.

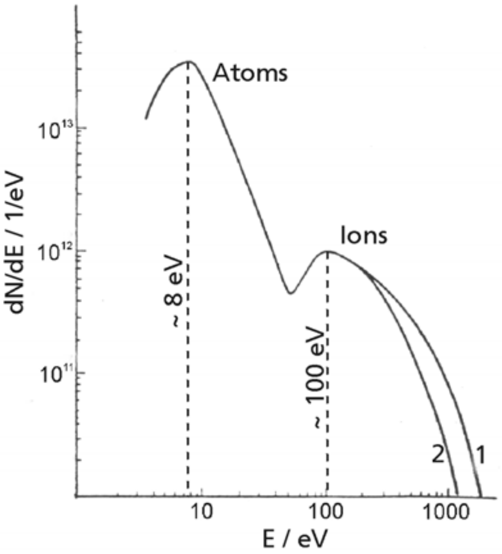

Fig. 2: Energy distribution of particles (atoms, ions) generated by the PLD of a PbTe plasma for two different laser power densities (1: 10^9 W/cm^2, 2: $4\mathrm{x}10^8$ W/cm^2) [11]. The most probable ion energy is ~100 eV, average energies are ranging from 150 to 300 eV.

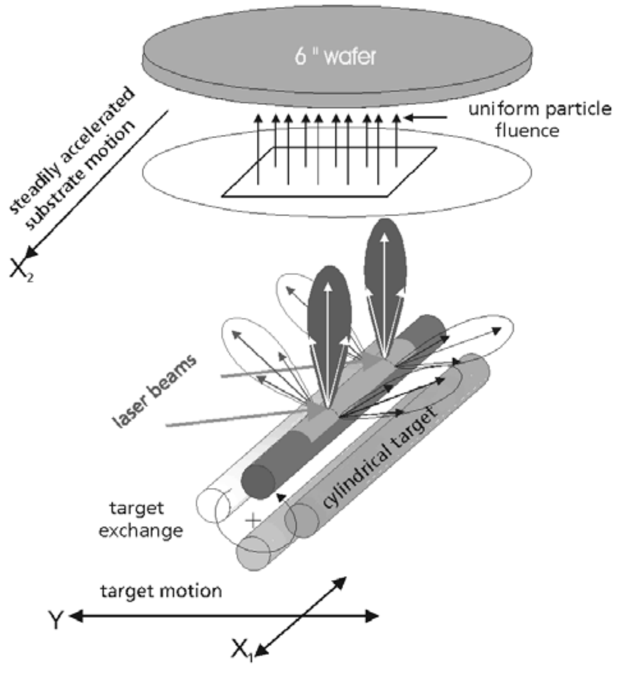

Fig. 3: Schematic view of the target-substrate arrangement for pulsed laser deposition (PLD) on large substrates [13]. The uniformity in x-direction can be controlled by the motion of the substrate, the uniformity in y-direction is obtained by pivoting the plasma plume due to the sinusoidal target motion in y-direction.

Based on the energy distribution of the particles generated by the PLD process, the smoothening of rough surfaces is considered by deep penetration of high energy particles into the formed film. Therefore impingements of these high energetic particles with particles of the formed film result in fill up of voids and higher mobility of particles at the surface (Fig. 4). The produced film can be described as a dense, diamond like carbon film with a smooth surface.

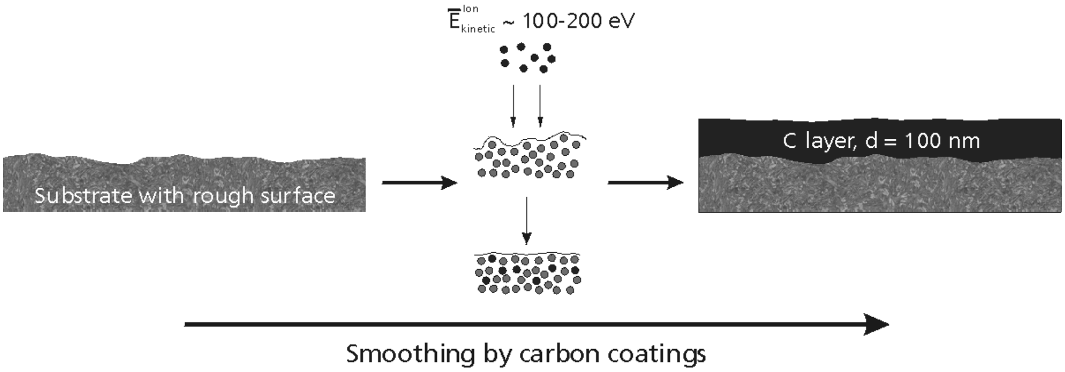

Fig. 4: Schematic model representation of the smoothing of rough surfaces by carbon films coated by PLD.

3.2. MAGNETRON SPUTTER DEPOSITION (MSD)

The deposition of Mo/Si/C multilayers was performed by magnetron sputter deposition. The set up, arrangement and the deposition have already been described elsewhere [14]. It is important to point out that there is an automatic transfer system between the MSD vacuum chamber and the PLD vacuum chamber. Therefore it is not necessary to take out samples and exposure at air.

3.3. CHARACTERIZATION TECHNIQUES

In order to quantify the effect of surface smoothening by the deposition of carbon films onto the substrate surface, the rms-roughness was measured by the AFM Nanoscope III using the tapping mode. The EUV reflectance was determined for these samples using a new EUV reflectometer [15]. The reflectance values measured for synchrotron calibrated samples using this stand-alone laboratory EUV reflectometer with a laser produced plasma source show a reproducibility of 0.5%. The EUV-reflectometry was performed with a near normal incidence beam of $\alpha=5°$.

4. EXPERIMENTAL RESULTS

The two main parameters which influence the smoothing behavior are the film thickness and the relaxation mode. The first parameter can be directly changed, whereas the second parameter can be indirectly influenced by changing the energy distribution of the plasma particles only. This can be accomplished either by varying laser pulse energy or changing of laser wavelength respectively laser pulse duration. The first one is a very easy option, hence we restrict to this, otherwise larger setup changes would be necessary.

In order to investigate the correlation of surface roughness and smoothing film thickness, carbon layers with gradient thicknesses have been deposited. This ensures that identical deposition parameters are used for different thicknesses and that continuous thickness values are available. After the deposition of these carbon gradients, a half of the gradient is covered and a uniform Mo/Si multilayer mirror is coated onto the other half (Fig. 5). Afterwards AFM and EUV reflectance measurements are performed on the marked positions of the gradient.

Fig. 5: Schematic view of the layer design and the measurement positions (dots at 0, ±20 mm and ±40 mm) for AFM and EUV reflectometry.

4.1. CARBON LAYERS ON SUPER-POLISHED SUBSTRATES

As stated, the growth of thin films is characterized by two competing processes: surface roughening and surface smoothing. Therefore the use of coatings as smoothing layers raises the question how super-polished surfaces of silicon wafers or glass substrates are affected by the coatings. The deposition of carbon layers on perfectly smooth surfaces would always result in a rougher surface after the deposition. However, even super-polished surfaces exhibit a certain remaining roughness. Substrates currently known to have highest surface finish exhibit rms-roughnesses of 0.1 nm. On these substrates EUV reflectances of Mo/Si multilayers of up to 70 % can be reached using tiny barrier layers between the Mo and Si layers [14,16].

In order to qualify the carbon smoothing layers deposited by PLD, experiments where carbon layers with a gradient thickness had been coated on smooth silicon wafers were performed. The typical rms surface roughness of these wafers is in the range of 0.15-0.18 nm. Afterwards the gradient was coated by a uniform Mo/Si multilayer and the EUV reflectance was measured. Initial experiments showed that the EUV reflectance of mirrors with carbon layers between wafer and Mo/Si multilayer is lower than for those without the carbon layer. In these initial experiments the substrates were taken out of the deposition chamber between the two depositions (C with PLD, Mo/Si with MSD). An increase of surface roughness with time was detected by AFM measurements immediately after vacuum removal of the samples (Fig. 6).

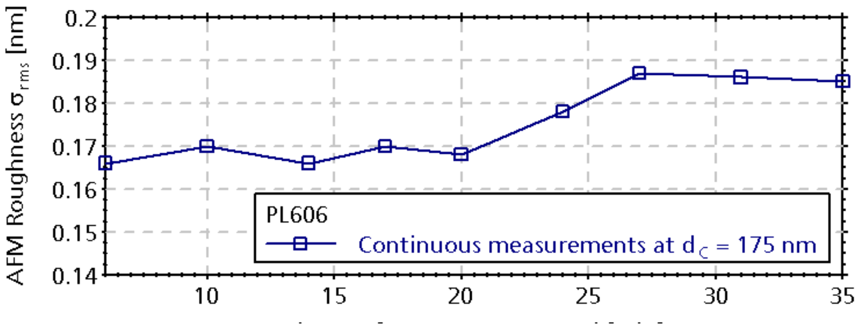

Fig. 6: Surface roughness increases directly after atmospheric contact.

A second series of depositions was carried out, whereby the atmospheric contact of the samples between the deposition of carbon and the Mo/Si multilayer was avoided. These experiments showed systematically higher EUV reflectances than for the previous mirrors and the absolute values are comparable with results on pure super-polished silicon wafers. The measured EUV reflectances are in the range between 69.5 and 70 % (Fig. 7). As a consequence of these results it can be stated that carbon smoothing layers prepared by PLD do not deteriorate the final surface if they are not exposed to air. The exact mechanism why the surface roughens under atmospheric conditions is unknown up to now. Maybe non-reversible water adsorption is responsible for this effect.

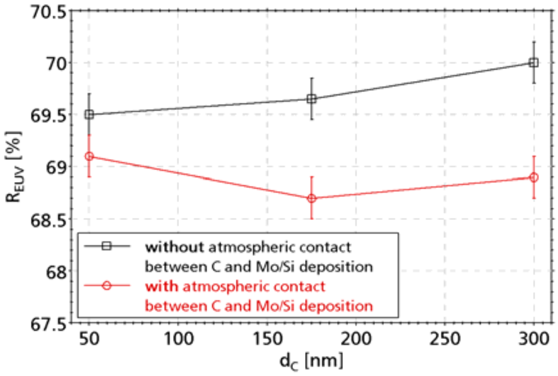

Fig. 7: EUV reflectance of Mo/Si multilayers deposited on super-polished silicon wafers with carbon smoothing layers. The two different curves show the values obtained with (circles) and without (squares) atmospheric contact between the carbon and Mo/Si multilayer deposition.

4.2. CARBON LAYERS ON ROUGH SUBSTRATE SURFACES

Initial coating experiments were using glass substrates with rms-roughnesses in the range of 0.31-0.33 nm. The deposition of carbon films with thicknesses of 100 nm was performed using a laser beam energy of 530 mJ. The characterization of the surface topography by AFM shows that PLD carbon layers offer the potential to effectively smooth high-special frequency surface roughness. The rms-roughness is decreased by the carbon coatings to values of appr. 0.18 nm from appr. 0.32 nm prior to the deposition (Fig. 8).

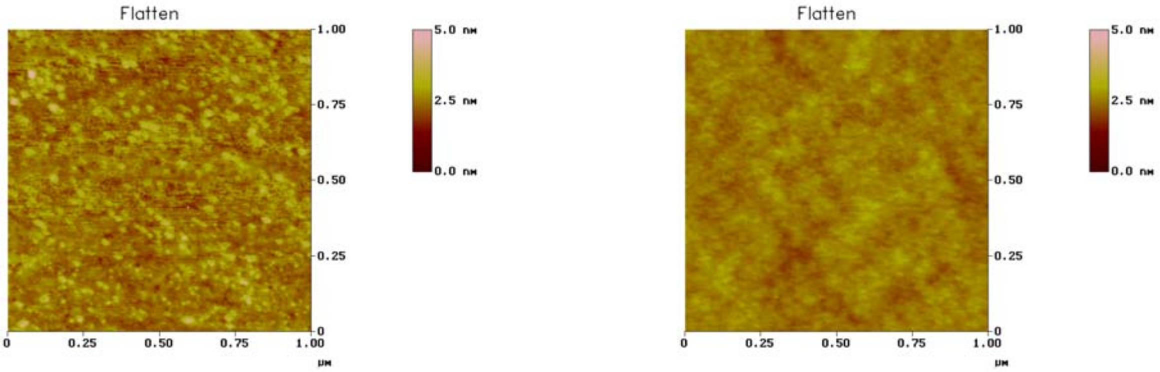

Fig. 8: AFM picture of the uncoated substrate (left) with an rms surface roughness of 0.32 nm and after PLD of a carbon film, $d_C = 100$ nm, (right) with an rms surface roughness of approximately 0.18 nm.

The successful smoothing of substrates with medium roughnesses (0.32 nm) encouraged experiments on substrates with higher surface roughnesses. The following systematic investigations were performed using surfaces with rms-roughnesses of appr. 0.75 nm (type 7315 substrates). The surfaces were produced by MSD of aluminum on smooth silicon wafers.

The results of the laser pulse energy variation show that higher energies have a higher potential to smooth surface roughnesses. Carbon layers with different thicknesses showed consistently a lower roughness when they were coated using higher laser pulse energies (Fig. 9). Obviously the parameter n of the relaxation mechanism tends to decrease for increasing laser energies.

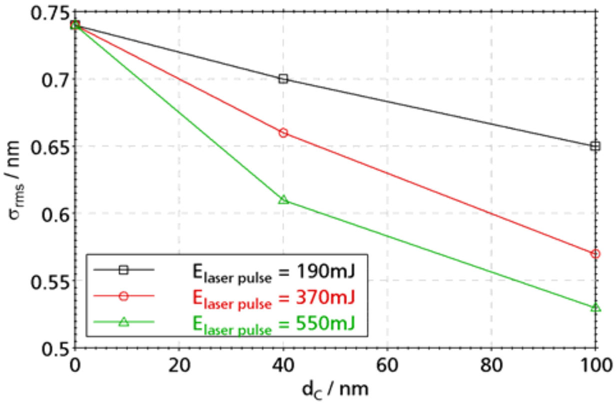

Fig. 9: AFM surface roughness of carbon single layers with various thicknesses deposited by PLD with different laser pulse energies. The best smoothing effect can be obtained for thick films which were deposited with high laser beam energies. The substrate with AFM roughnesses of 0.74-0.75 nm consists of silicon wafers coated with aluminum.

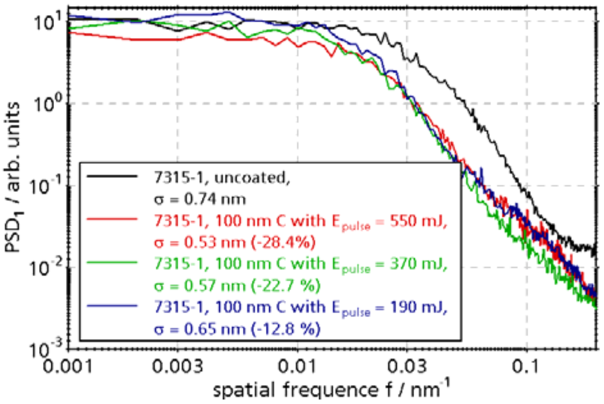

Fig. 10: Analysis of the power spectral densities of the AFM-images. The graphs show a preferential smoothing of high frequencies for which no dependence on laser power energy is detected. But the lower frequencies are better smoothed out at higher laser pulse energies.

The decrease of surface roughness for carbon layers produced by different laser energies was also examined by power spectral density treatment of the AFM-images. In all cases, a remarkable reduction over the whole frequency band can be observed. In particular, the lower frequencies are better smoothed out at higher laser pulse energies, whereas the higher frequencies do not show a distinct dependence on laser pulse energy. The roughness decrease obtained with carbon layers is an indication that reflectance of the optical multilayer can be increased by applying smoothing layers. In order to quantify the EUV reflectance, Mo/Si multilayers were deposited on carbon gradients. The carbon gradients themselves show a decreasing roughness with increasing thickness. However the roughness can only be decreased down to a saturation level. For the substrates with σ_{rms} = 0.75 nm, this level has a value of σ_{rms} = 0.4 nm and is already reached for carbon layer thicknesses d_c of 175 nm. (Fig. 11).

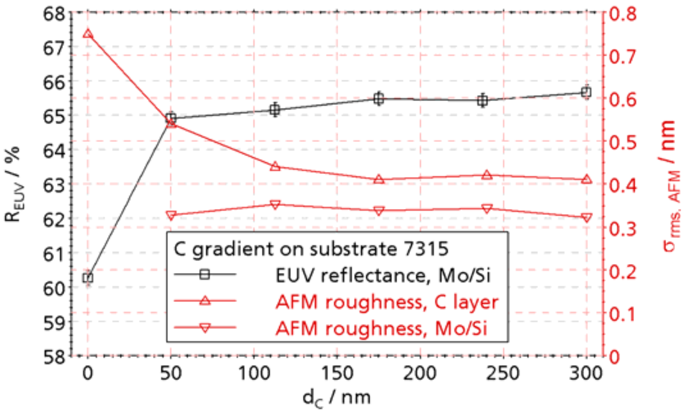

Figure 11: AFM surface roughness of carbon single layers with a graded layer thickness and resulting EUV reflectance of Mo/Si multilayers coated onto the carbon smoothing layer.

5. SUMMARY AND CONCLUSIONS

Carbon layers prepared by pulsed laser deposition effectively smooth surface roughnesses with rms values of up to 1 nm. It was shown that the carbon layers do not introduce additional roughness: A super-polished substrate remains smooth after carbon deposition. Investigations with rough substrate surfaces ($\sigma_{rms} = 0.75$ nm) show that the smoothing capability of carbon films increases with increasing laser power densities and with higher film thicknesses. Typical values of rms surface roughnesses before and after carbon deposition are: 0.32 nm → 0.18 nm and 0.75 nm → 0.41 nm. In connection with the roughness reduction, a remarkable increase of EUV reflectance can be obtained with carbon smoothing layers. An EUV reflectance gain of approximately 5 % was measured on rough substrates ($\sigma_{rms} = 0.75$ nm) by applying carbon smoothing layers.

Further reduction of surface roughness can be expected from higher laser pulse energies. Additionally the choice of alternative materials or material combinations could result in an improved smoothing behavior especially for lower roughness frequencies. Another open question is the internal stress of the smoothing layers. This will be the focus of future work.

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