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Absorption measurement of high-reflectance coated mirrors at 193 nm with a Shack–Hartmann wavefront sensor

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Abstract. All dielectric high-reflectance (HR) mirror coatings consisting of AlF_3/LaF_3/oxide layers were deposited on deep-ultraviolet-grade fused silica and CaF_2. A novel technique was employed to measure the absorption of these mirrors during irradiation by a 193-nm ArF excimer laser source. The method involves the application of a photothermal measurement technique. The setup uses a Shack–Hartmann wavefront sensor to measure wavefront deformation caused by the heating of the coating by the ArF beam. Laser calorimetric measurements of absorption were used to calibrate the wavefront sensor. The new test setup was used to investigate HR mirror coatings both before and after exposure to high average power ArF laser beams. HR mirror samples were irradiated by a 193-nm kilohertz laser source for either 500 million or 18.6 billion pulses. The differences between wavefront distortion measured inside the beam footprint compared to measured outside the beam footprint can be explained by compaction of the coating in the area heated by the ArF laser. Interesting wavefront-distortion results from testing mirrors with either fused silica or CaF_2 substrates can be explained by considering the figure of merit of these materials for excimer-laser mirror substrates. *© 2012 Society of Photo-Optical Instrumentation Engineers (SPIE).* [DOI: 10.1117/1.OE.51.12.121803]

Subject terms: 193 nm; high-reflectance mirror; fluoride; oxide; wavefront distortion; photothermal absorption measurement; Shack–Hartmann wavefront sensor; excimer laser coatings.

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1 Introduction

Since the early days of high-energy lasers (HELs), thermally induced wavefront distortion caused by absorption in optical elements such as mirrors and windows has been a serious problem. This laser-induced distortion limits the performance of the HEL itself, as well as the optical systems in which these lasers are used. Long before catastrophic damage has occurred, wavefront distortion usually ruins the performance of an optical component and leads to system performance problems. This phenomenon has been long studied, including papers presented in the laser damage symposium over 30 years ago.[1,2]

The challenge of designing and producing optical elements with low-absorption and high-power handling capability began with the advent of the high-power infrared chemical lasers and continued with the development of high-power CO_2 lasers used for materials processing. The performance of the optics and the lasers go hand in hand with each other. This history clearly illustrates how the development of lasers is critically dependent on the production of the optics. Once the substrate materials, optical polishing, and thin film coatings are improved, the laser power increases until the optical performance can no longer withstand the increased power and the cycle continues. This is certainly the case in the recent development of excimer lasers used for photolithography.

Over the last 10 years, the need for shorter wavelengths and higher output powers has driven the development of line-narrowed ArF excimer lasers with average powers up to 100 watts. The very few materials that can be used for substrates and thin film coatings in the deep ultraviolet (DUV) at 193 nm are limited, because most materials absorb in the DUV. The fact that these optics must withstand high DUV fluences for many billions of pulses without damage has only added to the challenge for the optics manufacturers.

One part of the key to success is the ability to measure the absorption of the materials at 193 nm. For mirrors, this means accurately measuring the absorption of the thin film coatings. There are many methods for measuring absorption including laser calorimetry,[3,4] photothermal deflection,[5,6] photothermal interferometric technique,[7] thermal imaging,[8] thermal lens,[9] and transient optical absorption.[10] A novel method using a Shack–Hartmann wavefront sensor (SHWS) to measure the distortion caused by the absorption in low-loss CaF_2 and high-purity fused silica was presented at SPIE Laser Damage Symposium in Boulder, Co., in 2008.[11] This method, to measure the absorption of high-reflector coatings for excimer-laser mirrors, has been adapted and results are presented on the employment of this technique in production both for quality control of new mirrors and for analysis of mirrors after use in lasers for many billions of pulses.

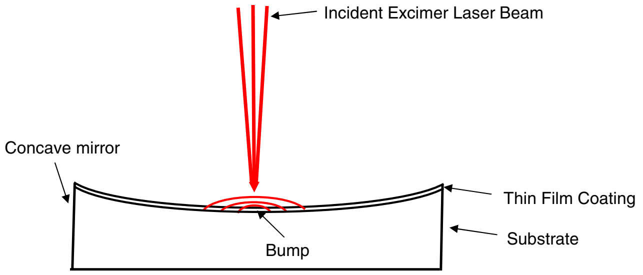

Fig. 1 Thermal distortion needs to be minimized for high performance.

1.1 *Thermally Induced Distortion*

When the excimer laser beam strikes the mirror surface, a small amount is absorbed in the thin film coating structure as shown in Fig. 1. This generated heat then propagates into the mirror surface and by thermal expansion distorts the surface outwardly. The height of the localized distortion (bump) is inversely proportional to the thermal conductivity (κ) since the higher this value, the faster heat diffuses into the substrate. In the normal application of the mirror, the concave surface focuses the impinging excimer laser light. Thermally induced convex surface distortion (bump) causes defocus and thus degrades optical performance. Advantages to this technique are that the actual optics (not just witness samples) can be measured directly and completed in a reasonable time frame.

2 Measurement Technique

It is highly desirable to measure the absorption of optical components for high-power lasers on the actual optic rather than on witness samples from the coating run, since the actual mirror must perform in the laser system. It has been shown that absorption can be measured by measuring the thermally induced wavefront distortion caused by the absorption of the laser radiation with an SHWS. A pump-probe technique is used with an ArF excimer laser as the pump beam and a red diode laser as the probe beam that measures the effect of the heating of the surface of the mirror. The repetition rate of the laser was 500 Hz, with pulse duration of 5 ns with a fluence level of 32 $\mathrm{mJ/cm^2}$. The rectangular beam size on the sample was 10×1 mm.

Since the distortion of the surface is proportional to the absorbed power, the magnitude of the distortion is directly proportional to the absorption. The setup for measuring excimer-laser mirrors is shown in Fig. 2. To minimize the oxygen absorption of 193-nm laser, the enclosure of the test setup was purged with the N_2 gas. The oxygen concentration was maintained below 100 ppm. The output power from the ArF excimer laser is focused by a lens onto the surface of the mirror. A collimated red diode laser beam overlaps the 193-nm pump beam and is reflected into the SHWS which measures the wavefront distortion induced by the heating of the localized spot on the surface. An uncoated $\mathrm{CaF_2}$ window has been inserted in the pump beam to reflect a small amount of the incident power into a detector for calibration. A shutter controls the irradiation time. Since a typical measurement takes only about three minutes, many measurements can be taken on a single sample and averaged to increase data reliability.

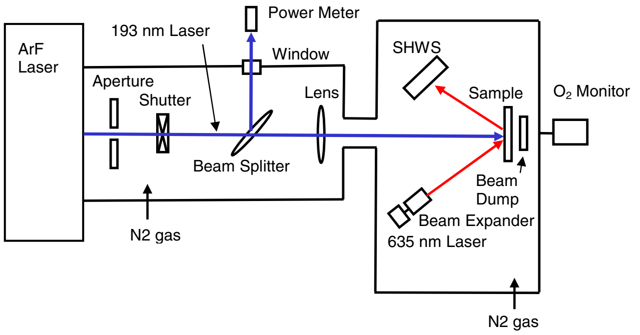

Fig. 2 The photothermal absorption test setup with a Shack-Hartmann wavefront sensor.

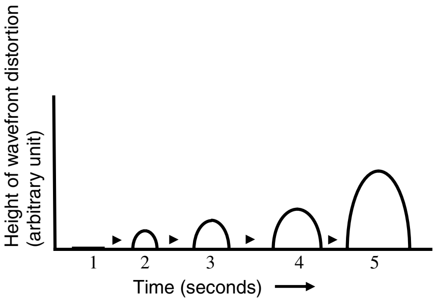

Fig. 3 The height of wavefront distortion increases with the time of laser irradiation.

The sequential procedure for measuring absorption of a 193-nm HR coated mirror is as follows: The 193-nm laser irradiates the sample for five seconds with the shutter open. The height of the bump (wavefront distortion) increases as the laser irradiation time increases (cf. Fig. 3). The sample is cooled down for 10 s with the shutter closed. This three-step process is then repeated 10 times resulting in 10 sets of wavefront-distortion data which are automatically acquired during the heating by a computer. A root mean square of the 10 data sets represents the final wavefront distortion for a given mirror. The measured wavefront signal was normalized by the laser power measured *in situ*.

This technique utilizes the measurement of "golden" samples with known absorption (laser calorimetry) to normalize the wavefront signal. The sample mirror's SHWS values are compared to two golden reference mirrors A and B, and its absorption is determined by linear interpolation as shown in Fig. 4. The same coatings and substrates of the golden samples as those of the samples were used to achieve a reliable calibration of photothermal technique to later calorimetric measurements. At Laser-Laboratorium Göttingen e.V. (Germany) the method of laser absorption calorimetry (ISO 11551) was applied to determine the absolute absorption of two golden samples at a wavelength of 193 nm and a repetition rate of 200 Hz. In order to remove surface contaminants the samples were irradiated for 360,000 pulses each at a fluence of around of 10 $\mathrm{J/cm^2}$. Each test consisted of a series of 110 single measurements with an irradiation time of 45 s. In the graph of absorption versus number of measurement, a constant level of absorption was found after 30 measurements.

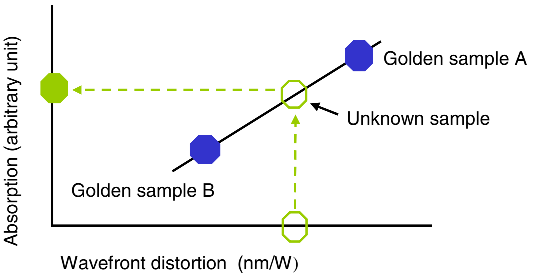

Fig. 4 Absorption of unknown sample is determined by linear interpolation.

3 Sample Preparation

The samples were all dielectric HR (193-nm) mirrors consisting of (AlF_3/LaF_3/Oxide) thin film layers deposited on DUV-grade fused silica and CaF_2 substrate. The substrate size was 38 mm in diameter and 10 mm in thickness. Dielectric oxide films were necessary because previous work[12] had shown that their intrinsic compressive stress could compensate for the high tensile stress of the fluoride layers and thus minimize the overall stress of the multilayer stack. Otherwise, the highly tensile stress in the films leads to increased scattering losses through stress reduction via material cracking, crazing, or delamination.

The HR mirror coatings were deposited by PVD (thermal boat evaporation) for fluoride and by e-beam evaporation for oxide layers. The coating chamber was pumped by oil-free, dry roughing pumps and by cryo-pumps to attain high vacuum base pressures in the low 10^{-8} Torr range. During deposition thickness control for each layer was determined by optical monitoring, and deposition rates were controlled by quartz crystal monitoring. Figure 5 shows data for a high-reflectance (98 ∼ 98.3% at 193 nm) coating on a high-purity fused silica substrate at zero degree angle of incidence.

4 Absorption Measurement Results

For low absorption at DUV wavelengths, the best materials for thin film coatings are fluorides, which can cause tensile stress to build up, especially on fused silica substrates. The reason is the thermal expansion mismatch between the coating materials and the substrate which is heated during deposition. This stress can be highly detrimental to the lifetime of the coating when exposed to high-rep-rate, high-peak-power pulses of 193-nm laser light.

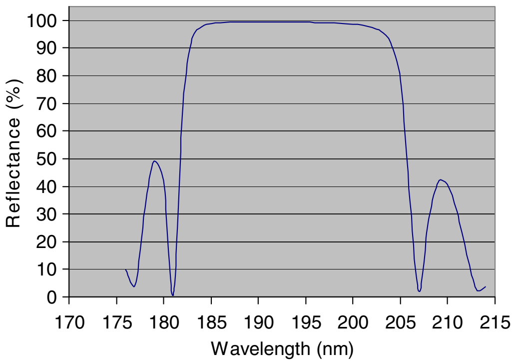

Fig. 5 Reflectance of 193-nm HR coated mirror on fused silica.

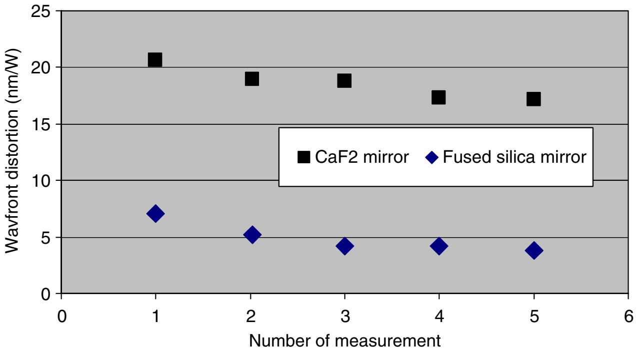

Fig. 6 Wavefront distortion of a 193-nm HR coating on fused silica and CaF_2 substrates.

One possible way to manage the stress and increase the lifetimes of coated mirrors is to use a CaF_2 substrate which more closely matches the thermal expansion coefficient of the coating materials. Comparison tests between fused silica and CaF_2 substrates showed that the lifetime with the same HR coating on CaF_2 substrates had predicted double (100 Bp) the lifetime on fused silica. Since this appears to be a real benefit for HR coatings on CaF_2, mirrors were produced for testing. The results of the SHWS absorption test comparing the same coating on fused silica and CaF_2 showed an interesting result. This is shown in Fig. 6, where the normalized wavefront signal for a CaF_2 mirror was four times larger than the signal for a fused silica reference mirror, although absorption (1.0%) of the CaF_2 mirror was a little lower by ∼10% than that (1.09%) of the fused silica mirror.

This was at first unexpected, since the absorption of the coating on both of the mirrors was assumed to be equal. Laser calorimetry measurements were then performed on the mirrors to confirm the absorption values. The results showed that the absorption was very nearly the same for both mirrors, as mentioned above. Therefore the explanation for the factor-of-four difference in optical distortion of the surface as measured by the SHWS must be explained by the differences in the properties of the substrate materials. The wavefront distortion decreased over the number of measurements due to the laser conditioning effect.

Since the performance of the optical train for HELs is so dependent on minimizing the wavefront distortion from the many mirrors in the system, figures of merit for substrate materials were developed over 30 years ago. These studies focused on materials like Si, SiC, Cu, Mo, and even diamond, but these materials do not make good choices for DUV mirrors that do not require water cooling.

However, the simple figure of merit for mirrors (FOM_M) used for uncooled mirrors in high-power CO_2 mirrors can readily explain the higher wavefront distortion observed for the CaF_2 substrates.[13] As shown in Eq. (1), the FOM_M is proportional to the thermal conductivity (κ).[14] The faster heat can be conducted away from the coating, the less the localized increase in temperature.

$$\mathrm{FOM_M} \propto \frac{\kappa}{\alpha}. \tag{1}$$

The FOM_M is proportional to the inverse of the coefficient of linear expansion (α), and the absorption of the mirror surface. For HR coated DUV mirrors, the absorption of

Table 1 Physical parameters of fused silica and CaF_2, and figure of merit for mirrors.

Parameter	Fused silica	CaF_2	FOM_M ratio Fused silica/CaF_2
Thermal conductivity κ (W/mK)	1.4	10.3	—
Thermal expansion coefficient α (1/K)	5.5×10^{-7}	187×10^{-7}	—
Figure of merit (FOM_M)	0.25	0.06	4.62

the surface is essentially that of the coating, since almost all of the light is reflected by the coating and the substrate materials have very low losses compared to the coatings. Clearly, materials with high coefficients of linear expansion will cause larger distortions of the surface for an equal amount of temperature rise; thus their figures of merit will be lower. Wavefront distortion is inversely proportional to the figure of merit as shown in the Eq. (2).

$$\text{wavefront distortion} \propto \frac{1}{\mathrm{FOM_M}} \propto \frac{\alpha}{\kappa}. \tag{2}$$

The figures of merit for fused silica and CaF_2 can be calculated using the values in Table 1, where it can be seen that CaF_2 has a much higher thermal conductivity than fused silica, but a 34-times larger linear-expansion coefficient than fused silica.

Assuming that the absorption of the coatings on both substrates are the same, then the ratio of the two figures of merit shows that fused silica is 4.6 times better compared to CaF_2. The large coefficient of thermal expansion of CaF_2 dominates the FOM_M and predicts that the thermally induced distortion of the ArF-laser heated mirror surface with this substrate material will be about 4.6 times higher than for the fused silica substrates. This simple figure of merit clearly explains the four-times-higher wavefront distortion measured for the HR coated mirrors on the CaF_2 substrate.

Certainly, this also shows one disadvantage of the SHWS test for measuring absorption, since it requires a golden reference sample of each type of substrate material with the same geometry. This must be weighed against the advantage that the absorption can be measured on the actual coated mirror rather than only on a witness sample. As a tool for quality control, this relative test is advantageous, since it eliminates variations in the measurement from changes in the test setup over time. The absorption of mirrors from new coating lots can be easily measured relative to the golden reference mirrors which have known values. This SHWS technique has been used as a new process-control tool in production, and this is illustrated in the statistical-process-control chart of the absorption in Fig. 7. The chart shows that the process is well within the control limits, with a favorable slight downward trend in absorption over time. This new test also allows new coating designs to be evaluated for absorption relative to the production coating design in an effort to lower the absorption and optimize the performance of the mirrors. The stability of the golden reference samples was presumed to be considerably high since the absorption remained constant and did not increase after 110 measurements in the first measurement with the 193-nm laser calorimeter. This was confirmed by remeasuring the absorption four years later.

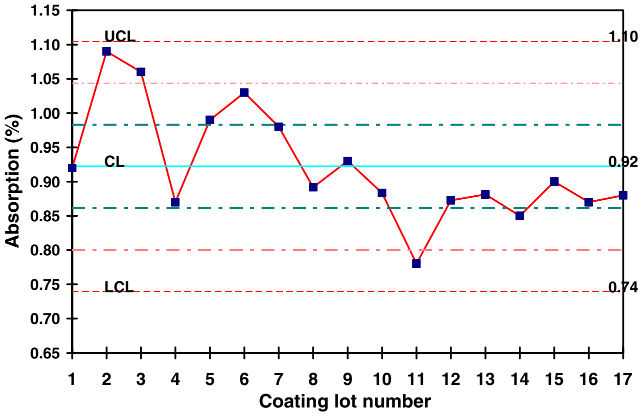

Fig. 7 Statistical-process-control chart of absorption in 193-nm HR mirrors in production.

The spatial resolution of this absorption method is good, since the pump beam is focused to a small area on the mirror, and this allowed the change of absorption of the HR coatings to be measured on mirrors after exposure to 18.6 billions of pulses. The repetition rate of ArF 193-nm laser was several kilohertz and the fluence was $5.1\ \mathrm{mJ/cm^2}$. Previously it was shown that the HR coatings are compacted by the high fluence pulses, and this physical change is beneficial for these fluoride-based coatings.[15] There is a clear visible "footprint" where the laser beam has compacted the coating in Fig. 8. The evaporated films contain point defects such as vacancies and micropores, as mentioned earlier. It is known that the thermally activated diffusion is facilitated by the presence of point defects in the solid-state film. Compaction is likely to occur by the thermally activated diffusion process during the laser irradiation. The activation energy for diffusion is provided as the laser beam irradiation generates heat through absorption in the thin film coating layers.

By measuring absorption starting from the edge of the mirror and through the beam footprint, the change in the absorption of the coatings has been measured as shown in

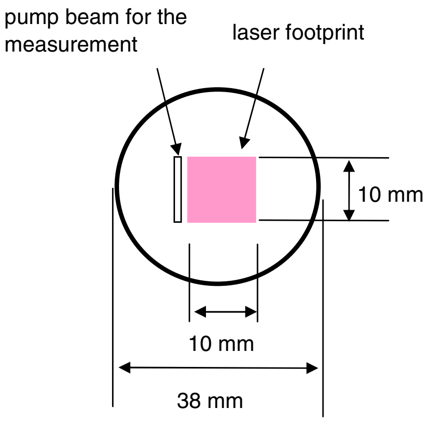

Fig. 8 The 193-nm HR coated mirror on fused silica and the laser footprint.

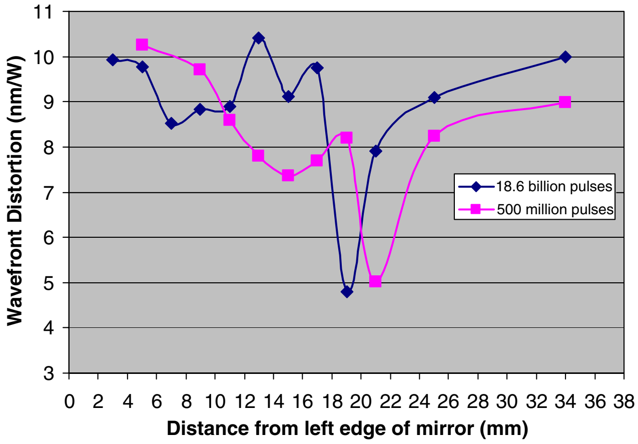

Fig. 9 Wavefront distortion of 193-nm HR coated fused silica mirrors with an exposure of 500 million pulses and 18.6 billion pulses of 193-nm laser irradiation.

Fig. 9. In the center of the compacted beam footprint, the absorption has dropped to one-half (~5 nm/W) of the value in the surrounding area, which is another example of how laser compaction of the coating has a beneficial effect on the performance of the mirrors. It appears that the compaction and reduction of the absorption occurs relatively quickly in the center of the beam, since there is no further reduction in the mirror with 18.6 billion pulses compared to the mirror with only 500 million pulses. This saturation of the absorption happens once the coating reaches 100% density and cannot compact any further.

5 Conclusions

High (~4 times) wavefront distortion of 193-nm CaF_2 mirrors compared to the fused silica mirrors was explained by a 4.6-times-high figure of merit of the CaF_2. Two indications of coating compaction in the 193-nm mirrors with 500 million and 18.6 billion pulses were demonstrated. The wavefront distortion (absorption) in the center of the beam footprint dropped to half the value of the outer portions of the footprint. Reflectance curves inside the beam footprints shifted toward shorter wavelengths as compared to outside the beam footprint.[15] Once coating compaction occurred, around 500 million pulses, it did not continue as the number of pulses increased to 18.6 billion. Coating compaction was attributed to thermally activated molecular diffusion by heat from laser irradiation. The coating compaction may beneficially increase coated-mirror lifetime. The photothermal absorption test with SHWS has been successfully employed for testing 193-nm mirror coatings and also in determining the statistical process control in the coating production.

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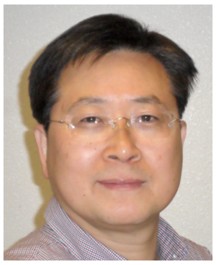
References

1. H. E. Bennett and P. C. Archibald, "Optical requirements for laser mirrors," *Damage in Laser Materials: 1974*, Nat. Bur. Stand. (U.S.) Spec. Pub. 414 (1974).
2. C. A. Klein, "High energy laser optical train performance," *Damage in Laser Materials: 1979*, Nat. Bur. Stand. (U.S.) Spec. Publ. 568 (1979).
3. A. Hordvik, "Measurement techniques for small absorption coefficients: recent advances," *Appl. Opt.* **16**(11), 2827–2833 (1977).
4. S.-Z. Jin and J.-F. Tang, "Measurement of weak absorption in optical thin films," *Appl. Phys.* **26**(12), 2407–2409 (1987).
5. B. Li and E. Welsch, "Configuration optimization and sensitivity comparison among thermal lens, photothermal deflection, and interference detection technique," *Proc. SPIE* **3578**, 594–603 (1998).
6. C. Mühlig et al., "Characterization of perform raw materials for high power fiber lasers using LID absorption measurement technique," *Proc. SPIE* **7842**, 78421M (2010).
7. J. F. Power, "Pulsed mode thermal lens effect detection in the near field via thermally induced probe beam spatial phase modulation," *Appl. Opt.* **29**(1), 52–62 (1990).
8. V. Draggoo et al., "Optical coating absorption measurement for high power systems," *Proc. SPIE* **622**, 186–190 (1986).
9. M. Liu, "Measuring surface deformation of optical components with surface thermal lens technique," *Proc. SPIE* **7132**, 71320T (2008).
10. S. Martin and E. Welsch, "Optical measurement of UV absorption in dielectric coatings," *Proc. SPIE* **4347**, 93–101 (2001).
11. K. Mann et al., "Photo-thermal measurement of absorption and wavefront deformations in fused silica," *Proc. SPIE* **7132**, 71321F (2008).
12. J. Earl Rudsill, A. Duparre, and S. Schröder, "Determination of scattering losses in ArF* excimer laser all-dielectric mirrors for 193 nm microlithography application," *Proc. SPIE* **5647**, 9–22 (2005).
13. G. H. Sherman, "CO_2 optics: absorption's dominant role," in *Electro-Optical Systems Design* pp. 50–56 (June 1982).
14. C. A. Klein, "High-power laser faceplate materials: figures of merit for optical distortion," *Proc. SPIE* **3151**, 138–149 (1997).
15. B. Cho, J. Rudisill, and E. Danielewicz, "193 nm laser induced spectral shift in HR coated mirrors," *Proc. SPIE* **7504**, 750409 (2009).



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