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High-efficiency, dielectric multilayer gratings optimized for manufacturability and laser damage threshold

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ABSTRACT

Ultrashort pulse, high-intensity lasers offer new opportunities for the study of light-matter interaction and for inertial confinement fusion. A 100 Terawatt laser operating at 400 fs and 1.053 μm is operational at LLNL, and a 1000 Terawatt (Petawatt) laser will come online in early 1996. These lasers use large-aperture (40 cm and 94 cm diameter, respectively) diffraction gratings to compress the amplified laser pulse. At present, holographically produced, gold overcoated photoresist gratings are used: these gratings represent the fuse in the laser chain. Higher laser damage thresholds and higher diffraction efficiencies are theoretically possible with multilayer dielectric gratings (MDG's). A number of design parameters regarding both the multilayer stack and the etched grating structure can be optimized to maximize the laser damage threshold and also improve the processing latitude for the interference lithography and reactive ion etching steps used during manufacture of these gratings. This paper presents model predictions for the behavior of hafnia/silica MDG's both during processing and in operation, and presents experimental data on the diffraction efficiency and short-pulse laser damage threshold for optimized witness gratings.

Keywords: diffraction gratings, multilayers, laser damage, pulse compression, ultrashort pulse lasers

1. INTRODUCTION

The technique of chirped pulse amplification (CPA) applied to laser systems^{1,2} has led in recent years to an explosion of activity involving ultrashort pulse, ultrahigh peak power lasers³. The basic design of a CPA laser system is shown in Figure 1. A broadband laser pulse of a few nanojoules at 1 ps or less is expanded in time by a factor of several thousand by the dispersion created by a diffraction grating pair. The pulse is then amplified by conventional means with a gain of up to 10^{12} , and recompressed with a second grating pair to nearly its original pulselength. CPA lasers have been fielded using a variety of solid-state laser materials such as Ti:sapphire^{4,5}, alexandrite^{6,7}, Cr:LiSAF⁸⁻¹⁰, and Nd:glass^{11,12}. Applications of CPA lasers include harmonic generation¹³, laser dentistry¹⁴, x-ray generation^{15,16} and laser fusion research¹⁷. We have recently fielded the world's highest peak-power CPA laser system based on Nd:glass¹⁸, with an output of 120 TW with a pulselength of 395 fs. A larger CPA laser under construction on a beamline of LLNL's Nova laser will provide 1 Petawatt of peak power.

Figure 1. CPA laser system schematic

High-power CPA laser systems presently use gold overcoated diffraction gratings to perform the pulselength manipulations in the laser chain. Pulse compression gratings in particular must have a high threshold for optical damage, and must be as efficient as possible as well, since, in a typical four-pass compressor, the beam diffracts from a grating surface four times, giving an overall compressor efficiency of η^4 for a nominal grating efficiency η . Our gold overcoated gratings^{19,20} exhibit diffraction efficiencies of about 94% and have laser damage thresholds of about 400 mJ/cm² for 1053 nm, 300 fs pulses²¹. Multilayer dielectric gratings have potentially two to three times the laser damage threshold as gold overcoated gratings. Furthermore, the theoretical diffraction efficiency of a MDG exceeds 99%, since there is no fundamental limit imposed by absorption.

2. THEORY AND DESIGN

The overall reflectivity of a MDG is determined by the multilayer stack; the ratio of light reflected in the 0 order to that diffracted in the -1 order is governed by the details of the grating formed in the top layer. As is the case for metallic gratings, interference lithography is used to pattern the gratings; but instead of overcoating the photoresist grating with a metal layer, the resist grating is used as a mask for subsequent etching processes. We have made MDG's based on ZnS/ThF₄ multilayers that exhibit efficiencies in excess of 96%.²² These gratings were made by a procedure using interference lithography with metallic transfer etch layers and absorbing layers between the underlying layers and the photoresist.²³ In order to substantially reduce the number of processing steps (and thereby reduce cost) and not expose the multilayer to metallic or absorbing coating materials, we have been exploring the possibility of patterning the photoresist directly on the top layer of the multilayer and etching through this mask only.

LLNL's 100 TW and Petawatt laser systems have been designed to use gratings with a line density of 1480 lines/mm, for which the Littrow angle is 52°. A standard quarterwave stack for use at 1053 nm and 52° exhibits substantial reflectivity from the third harmonic at 413 nm and 17.8°, which is the wavelength of our exposure laser and the angle for which a line density of 1480 lines/mm can be obtained in the photoresist grating mask, respectively. High substrate reflectivity during exposure of the photoresist film results in standing waves in the film due to interference effects. These lead to corrugations in the resist grating mask lines with a resulting loss of contrast and lack of line height control. To overcome this problem, absorbing underlayers must be applied. These are incompatible with our goals of manufacturing simplicity and high laser damage.

An alternative solution is to tailor the properties of the multilayer structure to give high reflectivity at use conditions and high transmissivity at the exposure conditions. A design based on alternating quarter-wave layers of HfO₂ and half-wave layers of SiO₂ at a design wavelength of 830 nm at normal incidence (the HLL design) exhibits these properties. Figure 2 shows the spectral transmission properties of a 10 pair HLL multilayer at exposure and use conditions. At exposure conditions, the transmission of the stack equals that of the bare substrate. This allows for facile patterning of a high-aspect ratio photoresist grating mask directly on the top layer of the stack.

Figure 2. Transmission spectra of HLL multilayer stack at exposure conditions of 413 nm, 18°, and use conditions of 1053 nm, 52°.

A grating can be etched into either SiO₂ or HfO₂ as the top layer to give high diffraction efficiency. The choice of material determines the optimum grating shape and aspect ratio. A grating design code based on the multilayer modal method^{19,24}

which can calculate diffraction efficiencies for arbitrary grating shapes and multilayer properties, has been used to determine optimum designs for both HfO_2 and SiO_2 gratings atop the HLL multilayer. These designs have been optimized for robustness with respect to etch depth variability.

Figures 3 and 4 compares efficiency contour plots of two MDG designs, which differ in the material composing the top grating layer. The plot of Figure 3 shows calculated efficiency as a function of the grating depth (y-axis) and total top layer thickness (x-axis) for a duty cycle (ratio of grating line width to period) of 0.35. Similar solutions exist for duty cycles up to about 0.5. The results show that high efficiencies can be obtained with relatively shallow etches (180-250 nm), which is fortunate since HfO_2 is difficult to etch. Partial etches into the top layer are also required for high efficiency, but as the figure shows, for an optimized total layer thickness (x-axis fixed at 350 nm, for example) the efficiency is insensitive to etch depth variations. Figure 4 shows the calculated efficiency as a function of the etch depth (x-axis) and duty cycle (y-axis) for a MDG with SiO_2 as the top layer. In this case, we assume a complete etch through the top SiO_2 layer, and rely on the fact that the HfO_2 layer underlying this is an effective etch stop for the SiO_2 etch chemistry²⁵. Thus, this design is robust to etch rate variability as well. Significantly deeper etches (700-800 nm) are required for high efficiency, but SiO_2 is relatively easy to etch, with etch rate selectivity relative to the resist mask significantly above unity. Also, high efficiency conditions are insensitive to duty cycle. The ultimate choice of a grating design will depend on manufacturing issues and, more importantly, on laser damage threshold.

Figure 3. Grating efficiency as function of grating depth and thickness of remaining top layer, for HLL multilayer with grating etched partway into top HfO_2 layer. Grating duty cycle of 0.35 assumed.

Figure 4. Grating efficiency as a function of grating depth and duty cycle for HLL multilayer with grating etched completely through top SiO_2 layer.

There is another potential advantage to the HLL design used to permit transparent exposure conditions during manufacture of the gratings. Calculations of the steady-state electric field distribution in and above the MDG have been performed for use conditions with both conventional quarterwave stacks and the HLL design. Figure 5 compares the results of the calculations. In the standard quarterwave design, the electric field maxima are situated at the interfaces in the multilayer stack and at the edges of the grating. In the HLL design, the maxima are shifted into the low-index SiO_2 layers inside the stack, and into the air space between the grating ridges above the stack. Therefore, the HLL design has theoretically a higher probability of laser damage resistance.

Figure 5. Calculated electric field distribution in and above multilayer dielectric grating structures. Left: quarterwave stack with high-efficiency grating, showing field maxima at layer interfaces and at corner of grating ridge; right: HLL stack with field maxima concentrated in the low index layers and in the air space between the gratings.

3. GRATING MANUFACTURE

In-house evaporation systems were used to make HLL multilayer designs on 5 cm fused silica or BK7 substrates with top SiO_2 or HfO_2 layers adjusted according to calculations to give maximum diffraction efficiency. Photoresist films 500-700 nm thick were applied by spin coating, and after softbaking the films grating patterns were exposed using interference lithography with the large exposure facility used to pattern our gold-overcoated gratings^{19,20}. The resist gratings were developed using in-situ monitoring²⁶ to ensure clearance of the resist to the substrate in the grating grooves. The samples were then submitted without further processing to reactive ion etching at remote facilities. Etches in SiO_2 overlayers were performed by Rochester Photonics Corp. on a Plasmatherm RIE system using $\text{CHF}_3/\text{Ar}/\text{O}_2$ chemistry. Etches in HfO_2 overlayers were performed at Hughes Aerospace Malibu Research Center using a Veeco Microtech ion beam etcher with $\text{CCl}_2\text{F}_2/\text{Ar}$ chemistry. After etching was complete, the resist mask was stripped by plasma ashing or acetone rinse, and the samples subjected to efficiency measurements, SEM analysis, and laser damage testing.

4. RESULTS AND DISCUSSION

4.1 Diffraction efficiency

High-efficiency gratings were produced with both HfO_2 and SiO_2 etched overlayer designs. Figure 6 shows a scanning electron micrograph of a witness MDG etched into HfO_2 which exhibits a diffraction efficiency of $95 \pm 1\%$ for TE-polarized 1053 nm light at 53.5° . This grating has an etch depth of approximately 200 nm and a duty cycle of about 40%. Another grating etched to the same depth but with a duty cycle of 50-55% exhibited a slightly lower efficiency of 89%. Yet another, with an etch depth of 250 nm and a duty cycle of 50-55%, measured 86%. In all cases the remainder of the incident light can be accounted for to greater than 99% in the zero-order specular reflection. The sensitivity of the efficiency of these gratings to groove shape and depth is greater than theoretical predictions suggest, so a close examination of the grating and the underlying multilayer stack was undertaken along with sensitivity analysis calculations. It was noted that the SiO_2 layers in the multilayer stack, designed to be half-wave at 830 nm or 294 nm thick, were actually about 265 nm thick. A calculation of grating performance with this error in the stack shifts the maximum efficiency plateau shown in Figure 3 to the right (i.e. deeper etches and a thicker final HfO_2 layer and required). The present gratings were made in the design space on the left shoulder of the high-efficiency plateau of Figure 3, where efficiency can fall off rapidly for small shape changes.

Figure 6. SEM of HLL MDG etched into HfO_2 top layer. 95% diffraction efficiency (3-31-1)

A SEM of a SiO_2 etched MDG exhibiting 94% diffraction efficiency is shown in Figure 7. This design called for etching completely through the SiO_2 layer to the etch-stop HfO_2 layer underneath, and so was underetched. Another grating which was

Figure 7. SEM of HLL MDG etched into SiO_2 top layer. 94% diffraction efficiency (9-20-5)

slightly (10%) overetched is shown in Figure 8. This grating exhibited an efficiency of 92% and suffered material deposition, apparently from sputtering of the HfO_2 underlayer, on the grating sidewalls. A measurement of diffracted and reflected light from this grating indicated 2-3% scatter loss. Another grating overetched approximately 3x showed the SiO_2 grating ridges completely encapsulated by columnar growths of this material, and the 110 nm thick HfO_2 underlayer about two-thirds removed. Etching of a HfO_2 single layer covered by photoresist grating grooves under identical etch conditions did not result in deposition of material on the resist grating sidewalls. Therefore, a physical sputtering process involving a nonvolatile Hf-compound is not considered likely. Apparently, a chemically coordinated Hf-Si compound is likely formed, with a considerable volume expansion. Further research on the etch characteristics of $\text{SiO}_2/\text{HfO}_2$ multilayers is continuing.

Figure 8. SEM of HLL MDG etched through SiO_2 top layer, showing Hf-compound deposits sputtered onto grating sidewalls. 92% diffraction efficiency (9-20-3)

4.2 Short-pulse Laser Damage Measurements

Laser damage measurements were performed on the above-described MDG witness samples at 1053 nm and 300 fs pulselength, using a single-grating compressor CPA laser system described elsewhere.^{21,27} A spot size of 0.5 mm was used, and the first occurrence of visible alteration of the surface under 100X Nomarski magnification was defined as laser damage. A standard s-on-1 test protocol (no conditioning) was performed, and up to 600 shots at a 10 Hz rep rate were performed at a given fluence.

Incipient damage to MDG structures is characterized typically by the ablation of small grooves orthogonal to the grating grooves which extend partway across the top of the grating ridge and are spaced more-or-less periodically along the length of the grooves. Figure 9 shows an SEM of incipient damage of a HfO₂-etched MDG at a fluence of 0.21 J/cm². At a fluence of 1.25 J/cm², the grating was almost completely destroyed after 5 shots, but the multilayer structure underneath remained intact after 600 shots. Earlier, a multilayer stack from the same coating run but without a grating exhibited a damage threshold of about 0.76 J/cm². It is important to note, however, that HfO₂-etched gratings made By Hughes Aerospace for use at 825 nm light exhibited short-pulse laser damage thresholds of about 0.55 J/cm², and optical damage thresholds as high as 1.3 J/cm² under these conditions have been measured for HfO₂/SiO₂ multilayer coatings from commercial vendors²⁸.

Figure 9. Incipient damage on HfO₂-etched HLL multilayer grating at 0.21 J/cm², 1053 nm, 300 fs pulselength. The damage is confined to strips of material ablated along the top of the grating ridges, perpendicular to the grating direction. The damage features are about 100-200 nm long.

Incipient damage of the SiO₂-etched grating shown in Figure 7, occurred at 0.51 J/cm², and massive damage occurred after a few shots at 0.75 J/cm². In this case, massive damage extended deep into the multilayer structure.

The damage results are summarized and compared with typical damage thresholds for gold-overcoated gratings in Table 1. The damage threshold of the HfO₂-etched grating is disappointing, but may be influenced by the etching process. It is known that etching with a significant sputtering component can result in preferential loss of oxygen and a subsequent reduction in stoichiometry of oxide films, resulting in reduced optical damage threshold. Further experiments are planned involving oxygen post-treatment of etched HfO₂ gratings in the hope of improving the damage resistance. The damage threshold of the SiO₂-etched grating is about 25% higher than that of our gold standard gratings, but further improvement is required before such gratings will be made on a large scale for pulse compression. Damage thresholds for longer nanosecond-scale pulselengths have not been measured, but are expected to be significantly higher than for gold gratings under these conditions.

Table 1

Laser damage thresholds at 1053 nm, TE polarization, 520, 300 fs pulselength for HLL multilayer dielectric stacks with and without gratings etched into top layer, and gold gratings. Gratings are 1480 lines/mm. Units are J/cm².

	HLL+HfO ₂ multilayer	HLL+HfO ₂ grating	HLL+SiO ₂ grating	Gold Grating*
Incipient damage	0.75	0.21	0.51	0.42
Massive damage	-	1.25	0.75	-

*TM polarization

5. CONCLUSIONS

High-efficiency multilayer dielectric grating designs which allow patterning of photoresist etch masks directly onto the topmost oxide layer, and which are robust to etch depth variations, have been demonstrated theoretically and fabricated on 5-cm diameter witness samples. The multilayer design calls for half-wave SiO₂ layers alternating with quarter-wave HfO₂ layers, both defined for 830 nm and normal incidence, to give high reflectivity at 1053 nm, 520 use conditions and high transmission at 413 nm. 180 exposure conditions. Photoresist gratings have been patterned on top of these multilayer structures by interference lithography, and these patterns have been reactively ion-etched into the top oxide layer. Gratings in HfO₂ top layers have exhibited diffraction efficiencies of up to 95%, and gratings etched in SiO₂ top layers up to 94%. With respect to our gold-overcoated photoresist gratings, the laser damage threshold of one SiO₂-etched grating is about 25% higher, while that of the HfO₂-etched grating tested was about a factor of 2 lower. The laser damage thresholds of the etched gratings may be influenced by preferential sputtering of oxygen from the grating layer. Use of a HfO₂ layer as an etch stop for etching completely through the SiO₂ grating layer leads to Hf-based deposits on the grating sidewalls which contribute to scatter losses. Further experimentation with etch-stop chemistry and materials is ongoing.

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7. REFERENCES

1. D. Strickland and G. Mourou, "Compression of amplified chirped optical pulses", Opt. Commun. **56**, 219-221 (1985)
2. P. Maine, D. Strickland, P. Bado, M. Pessot, and G. Mourou, "Generation of ultrahigh peak power pulses by chirped pulse amplification" IEEE J. Quantum Electron. **24**, 398-403 (1988)
3. M.D. Perry and G. Mourou, "Terawatt to petawatt subpicosecond lasers", Science, **264**, 917-924 (1994)
4. J. Kmetec, J.J. Macklin and J.F. Young, "0.5-TW, 125-fs Ti:sapphire laser", Opt Lett. **16**, 1001-1003 (1991)
5. A. Sullivan, H. Hamster, H.C. Kapteyn, S. Gordon, W. White, H. Nathel, R. Blair and R. W. Falcone, "Multiterawatt, 100 fs laser", Opt. Lett., **16**, 1406-1408 (1991)
6. M. Pessot, J. Squire, P. Bado, G. Mourou and D. Harter, "Chirped-pulse amplification of 300fs pulses in an Alexandrite regenerative amplifier" IEEE J. Quantum Electron. **25**, 61-66 (1989)
7. M. Pessot, J. Squire, G. Mourou and D. Harter, "Chirped-pulse amplification of 100-fsec pulses" Opt. Lett., **14**, 797-799 (1989)
8. T. Ditmire and M.D. Perry, "Terawatt Cr-LiSrAlF₆ laser system", Opt. Lett. **18**, 426-428 (1993)
9. T. Ditmire, H. Nguyen and M.D. Perry, "Design and performance of a multiterawatt Cr-LiSrAlF₆ laser system". J. Opt. Soc. Am. B **11**, 580-590 (1994)

10. P. Beaud, M. Richardson, E. Miesak and B.T. Chai, "8-TW 90-fs Cr:LiSAF laser", *Opt. Lett.* **18** 1550-1552 (1993)
11. M.D. Perry, F.G. Patterson and J. Weston, "Spectral shaping in chirped-pulse amplification", *Opt. Lett.*, **15**, 381-383 (1991)
12. C. Rouyer, E. Mazataud, I. Allais, A. Pierre, S. Seznec, C. Sauteret, A. Migus and G. Mourou, "generation of 50-TW femtosecond pulses in a Ti:Sapphire/Nd:glass chain", *Opt. Lett.*, **18**, 214-216 (1993)
13. A. Lhuillier, T. Auguste, Ph. Balcou and B. Carre, "High-order harmonics - A coherent source in the XUV range", *J. Nonlinear Optics & Materials*, **4** 647-665 (1995)
14. B.C. Stuart, M.D. Perry, L. DaSilva, D. Matthews and J. Neev, U.S. Patent Pending (1995)
15. J.D. Kmetec, C.L. Gordon III, J.J. Macklin, B.E. Lemoff, G.S. Brown, S.E. Harris, "MeV X-ray generation with a femtosecond laser", *Phys. Rev. Lett.*, **68**, 1527-1530 (1992)
16. J.C. Kiefer, J.P. Matte, H. Pepin, M. Chaker, Y. Beaudoin, T.W. Johnston, C.Y. Chien, S. Coe, G. Mourou and J. Dubai, "Electron distribution anisotropy in laser-produced plasmas from X-ray line polarization measurements", *Phys. Rev. Lett.* **68** 480-483 (1992)
17. M. Tabak, J. Hammer, M.E. Glinsky, W.L. Kruer, S.C. Wilks, J. Woodworth, E.M. Campbell, M.D. Perry and R.J. Mason, "Ignition and high gain with ultrapowerful lasers", *Phys. of Plasmas* **1** 1626-1634 (1994)
18. J. Miller, B.C. Stuart, J.A. Britten, R.D. Boyd, H. Nguyen, G. Tietbohl, W. Olson and M.D. Perry, in preparation (1995)
19. R.D. Boyd, J.A. Britten, D.E. Decker, B.W. Shore, B.C. Stuart, M.D. Perry, and L. Li, "High efficiency metallic diffraction gratings for laser applications", *Appl. Optics* **34**, 1697-1706 (1995)
20. J.A. Britten, M.D. Perry, B.W. Shore and R.D. Boyd, "A universal grating design for pulse stretching and compression in the 800 to 1100 nm range" *Opt. Lett.* (submitted) (1995)
21. B.C. Stuart, M.D. Feit, S. Herman, A.M. Rubinchik, B.W. Shore and M.D. Perry, "Optical ablation by high-power short-pulse lasers", *J. Opt. Soc. Amer. B*, (in press) (1995)
22. M.D. Perry, R.D. Boyd, J.A. Britten, D. Decker, B.W. Shore, C. Shannon, E. Shults, and L. Li, "High efficiency multilayer dielectric diffraction gratings" *Opt. Lett.* **20**, 940-942 (1995)
23. L.M. Holbrock, R. Withrington and H. Garvin, "Method for fabrication of low-efficiency diffraction gratings and product obtained thereby", U.S. Pat. 4,828,356 (May 9, 1989)
24. L. Li, "Multilayer modal method for diffraction gratings of arbitrary profile, depth and permittivity", *J. Opt. Soc. Am. A*, **10**, 2581-2591 (1993)
25. K.K. Shih, T.C. Chieu and D.B. Dove, "Hafnium dioxide etch-stop layer for phase-shifting masks", *J. Vac. Sci. Technol. B*, **11**, 2130-2131 (1993)
26. J.A. Britten, R.D. Boyd and B.W. Shore, "In-situ endpoint detection during development of submicron grating structures in photoresist", *Opt. Eng.* **34** 474-479 (1995)
27. B.C. Stuart, M.D. Feit, A.M. Rubinchik, B.W. Shore and M.D. Perry, "Laser-induced damage in dielectrics with nanosecond to subpicosecond pulses" *Phys. Rev. Lett.* **74**, 2248-2251, (1995)
28. B.C. Stuart, personal communication (1995)

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