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Diffraction Gratings for High-Intensity Laser Applications

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Laser safety: Tools and Training

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Diffraction Gratings for High-Intensity Laser Applications

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Introduction

The scattering of light into wavelength-dependent discrete directions (orders) by a device exhibiting a periodic modulation of a physical attribute on a spatial scale similar to the wavelength of light has been the subject of study for over 200 years. Such a device is called a diffraction grating. Practical applications of diffraction gratings, mainly for spectroscopy, have been around for over 100 years. The importance of diffraction gratings in spectroscopy for the measurement of myriad properties of matter can hardly be overestimated. Since the advent of coherent light sources (lasers) in the 1960's, applications of diffraction gratings in spectroscopy have further exploded. Lasers have opened a vast application space for gratings, and apace, gratings have enabled entirely new classes of laser systems.

Excellent reviews of the history, fundamental properties, applications and manufacturing techniques of diffraction gratings up to the time of their publication can be found in the books by Hutley (1) and more recently Loewen and Popov (2). The limited scope of this chapter can hardly do justice to such a comprehensive subject, so the focus here will be narrowly limited to characteristics required for gratings suitable for high-power laser applications, and methods to fabricate them. A particular area of emphasis will be on maximally-efficient large-aperture gratings for short-pulse laser generation.

Light of a wavelength λ incident at angle θ_i on a diffraction grating that has modulation with a period p is diffracted in a direction θ_m according to the *grating equation*:

$$\sin \theta_m = \sin \theta_i + m \lambda / p \quad (1)$$

where m is the grating order. Figure 1 illustrates the geometry of diffraction of light of a single wavelength into various orders from a grating:

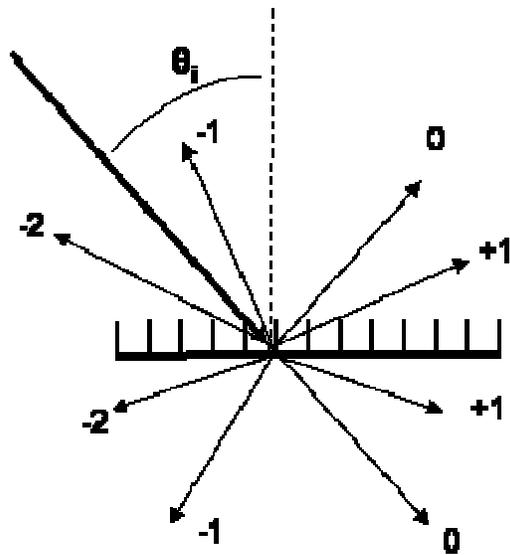


Figure 1. Orders diffracted from a grating.

Gratings can operate in transmission, reflection, or both. The modulation can be in the form of periodic variations in the density or refractive index throughout the bulk of a material. These are ‘volume gratings’. Or, the modulation can be physical corrugations of the surface of a material. These are ‘surface-relief gratings’.

The use of gratings for high-power laser applications requires that, other than redirecting the incident light according to the grating equation, the grating does nothing else to distort the beam, and also, that the beam do nothing to the grating, such as heating or damage. These restrictions turn out to be quite serious, and severely limit potential candidates for the intended application, as is discussed below.

Of particular note appropriate for this book dealing in the main with laser safety issues, is the fact that diffraction gratings can direct intense beams in several directions at once, directions that are counterintuitive to researchers used to dealing with normal reflective and refractive optics. This makes them more dangerous in general than normal optics in high-power laser applications, and so must be treated with the appropriate attention to safety, with beam blocking setups in both reflection and transmission to safely block unused orders.

Types of Gratings and their Uses.

Volume Gratings

Dichromated Gelatin (DCG) gratings (cf 3) are perhaps the best-known variety of volume grating. These are made by casting a many-micron thick film of gelatin sensitized with a photoreactive dichromate salt onto a glass plate, exposing this plate to a holographically generated interference pattern, and using wet-chemistry to change the refractive index of the exposed regions in the gelatin. The resulting film is hygroscopic, and must be hermetically encapsulated by a cover plate sealed at the edges, to retain the

index modulation. DCG gratings are typically used in transmission, but can be used in reflection by double-passing the light through the film with a reflective surface on the backside plate. They can have very high efficiency and low polarization-dependent losses, and can be made quite large. They are used in spectroscopic and display applications, but in general have poor diffracted wavefront flatness that must be corrected by post-polishing or adaptive optics in a laser application. Also, the organic gelatin material has a very low laser-induced damage threshold in comparison with some transparent inorganic dielectric materials.

PhotoThermal Refractive (PTR) gratings (4) are a relatively recent technology finding increasing use in demanding laser applications. PTR glass is a multicomponent glass doped with small amounts of photosensitive constituents, that undergoes a very small reduction in refractive index upon exposure to UV light below about 330 nm followed by a heat treatment. Interference lithography, discussed in a later section, can be used to generate the grating pattern in this glass. PTR glass has laser damage resistance to IR light quite similar to BK7 or other multicomponent glasses, and is therefore quite useful for many high-power laser applications. (e.g. 5,6). At present, the glass, and the interference lithography platforms capable of recording into it, are limited to a few mm in aperture. These gratings also will not be useful for kilowatt-class high average-power laser systems as the bulk material will heat and distort due to low-level absorption by some of the glass constituents.

Fiber Bragg Gratings (7) are a related type of grating whereby an index modulation is induced longitudinally in the core of an optical fiber by exposing the fiber to interference from UV light. The fiber can be a germanium-doped fused silica exposed to 244 nm light, or even pure fused silica exposed with 193 nm at high temperature in the presence of hydrogen. FBG structures were an area of intense current research for optical telecommunications applications several years ago, and are currently being developed for short-pulse lasers that require dispersion control.

Surface-Relief Transmission Gratings

Low-cost transmission gratings made by embossing surface relief patterns from master gratings onto plastic substrates, or polymer films on glass substrates, are available from several commercial suppliers. These can exhibit very good efficiency and optical quality, but in general are not suitable for intense laser applications due to damage to the films or thermal distortion the beam, by low-level absorption of the laser light. Gratings etched into bulk fused silica (8-10) have demonstrated the ability to withstand very high peak power laser pulses at both IR and UV light. High-efficiency transmission gratings used for beam steering, focusing and wavelength discrimination (8,9) require periods on the order of the laser wavelength and rather large incidence angles, so that only the 0 and -1 orders exist, and also require deep grating structures with height-to-width aspect ratios $H/W \sim 4$. These features require ion-beam milling to transfer-etch the pattern from the photoresist mask to the bulk material. Gratings of this type 470x420 mm in aperture have been demonstrated recently for a high-energy beam-steering and focusing application (9). Low-efficiency beam sampling transmission gratings that diffract and

focus a very small but precisely known fraction of the total beam energy to an energy diagnostic have been fabricated at 400x400 mm aperture by wet-etching the mask pattern into the bulk fused silica using a weak hydrofluoric acid solution. (10). This is possible due to the very low aspect ratios of the features required for low-efficiency gratings, typically $H/W \sim 0.01$. It is possible to create multifunction, multi-scale low-aspect ratio grating structures by wet etching, that simultaneously perform wavelength discrimination and beam sampling duties (11).

General limitations of transmission gratings in high-power laser systems:

- High average-power lasers: PPM absorption in multicomponent-glass transmissive optics can very quickly cause thermal distortion and lack of focusability of the beam in multi-Kilowatt lasers (12). A probable exception would be high-purity fused silica surface-relief gratings.
- Intense nanosecond pulsed lasers: Again, high-purity fused silica is acceptable in many cases, but coherent addition of the electric fields generated by more than one propagating order in the bulk of the glass can cause laser damage. This is true even if the grating is on the output face of the optic, as it is typically not possible to totally suppress back reflections of both 0, -1 or possibly higher orders.
- Intense short-pulse lasers: Very intense laser beams operating at 10's of picoseconds or shorter can cause dielectric breakdown even in atmospheric gases, and so are propagated in vacuum. Needless to say, all-reflective optics are required for beam manipulation in this case.

Surface Relief Reflection Gratings

This is probably the largest class of gratings in terms of numbers and application space, and certainly the most important for intense short-pulse lasers. As with transmission gratings, relatively low-cost, high-quality gratings replicated in epoxy or other polymeric films from holographically written or mechanically ruled master gratings and overcoated with a reflective metal layer, are readily available from commercial sources. However, to obtain the highest possible diffraction efficiency, wavefront quality, and thermal loading performance, the large-aperture short-pulse laser community for years has demanded high line-density gratings produced by laser interference lithography (holographic gratings). The most intense short-pulse lasers require very large aperture gratings to reduce the power density on the grating surface to below the optical damage level. The first Petawatt-class laser system, built at LLNL in the mid 1990's (13), used internally-produced gold-overcoated holographic master gratings 94 cm in diameter. These are still the largest monolithic gratings made, and now several laser systems around the world employ gratings of the same design to do basic high energy-density and nuclear fusion-related science (14,15).

At the same time the Nova Petawatt was being built, a new class of reflective diffraction grating was being developed, also at LLNL. Multilayer dielectric (MLD) gratings (16,17) combine a multilayer dielectric high-reflector stack with a grating etched into the top layer or top several layers, to create a grating having theoretically 99.9% diffraction

efficiency into the -1 order with no absorption of the light and therefore much higher laser damage thresholds. Consequently, just as MLD high-reflectors have supplanted metallic mirrors for high power laser systems, MLD gratings have supplanted gold-overcoated gratings for most, but not all, short-pulse laser applications. The remainder of this treatise will briefly review short pulse generation, then describe in detail the design and manufacture of large-aperture holographic gratings.

Plane Gratings for Laser Pulse Compression.

The technique of chirped-pulse amplification (CPA) (18) uses one or more gratings with other optics in a ‘stretcher’ to temporally disperse a low-energy, broadband, short-pulse beam by a factor $\sim 10^3$. The stretched beam is then amplified by conventional gain media without undergoing nonlinear self-focusing. Gains of $\sim 10^6$ are possible. The amplified, stretched pulse is then sent through a ‘compressor’ typically containing 2-4 gratings that undo the temporal dispersion of the stretcher to create an intense pulse of nearly the initial pulse duration. A schematic of CPA is shown in Figure 2. The requirements of the compressor gratings in particular are quite demanding. Typical compressor designs employ 4 grating bounces, so the overall throughput is (grating efficiency)⁴. The beam size and intensity is in large part limited by the size and damage threshold of the compressor gratings. Therefore, maximizing the efficiency and damage threshold of the gratings has an enormous impact on energy delivered to target, as well as the cost of the laser system.

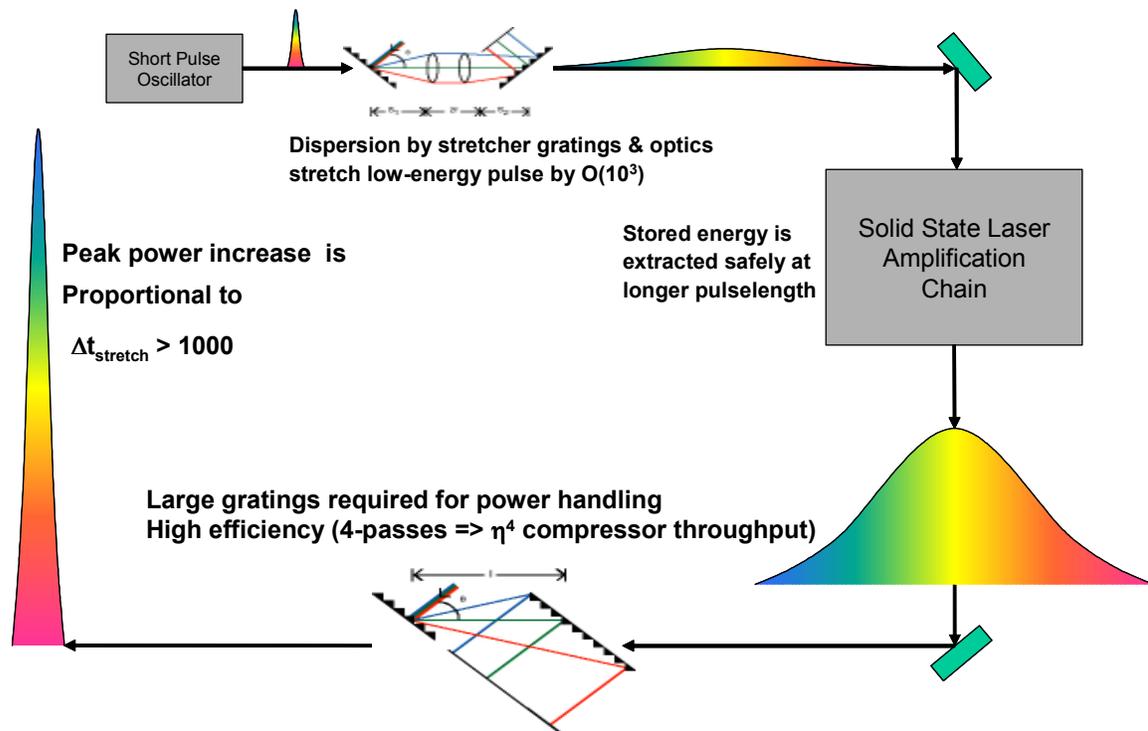


Figure 2. Schematic of CPA for short-pulse laser generation.

Design Considerations

High-energy Petawatt-class lasers being built today operate at 1053 nm, using Nd:glass as the laser medium, and are designed to produce pulses from a few hundred fs to several ps in duration. These pulse durations require a high diffraction-efficiency bandpass less than 20 nm, a condition easily met by MLD gratings. To maximize diffraction efficiency and peak-power handling ability, MLD grating designs have evolved to high-line densities (1700-1800 l/mm) operating at high near-Littrow incidence angles, comprised of hafnia/silica layers with the top grating layer made of silica. The Littrow angle is defined as that in which the -1 order diffraction angle is the same as the incident angle. Hafnia (HfO_2) and silica (SiO_2) are the materials of choice based on extensive experience for high-intensity nanosecond-pulse MLD high-reflectors (19). SiO_2 , because it is a high bandgap material with a high intrinsic laser damage threshold (20) and is also amenable to deposition and subsequent processing incurred during grating manufacture, is the material of choice for the grating layer.

We use an in-house grating design code based on (21) in conjunction with commercially available thin-film codes to design MLD gratings. Commercial grating design software (22) is available as well, and offers good agreement with results of our in-house code, although the electric fields generated in the stack and grating are not readily viewable by it.

Typically, the stack is comprised of 20-60 layers. The main criterion is that it be a high reflector at the use wavelength and angle range, and that it have a relatively thick SiO_2 capping layer. The large number of layers gives the designer a great deal of flexibility to incorporate other features into the stack, such as antireflection properties at the holographic exposure wavelength and angle (17), and etch-stop layers for precise grating depth control (23). Grating design codes are used iteratively with the thin film design codes to optimize the etched depth and duty cycle (ratio of grating linewidth to period), shape and layer thicknesses for the application of interest. Of particular importance for large-aperture gratings is a design that is robust to small duty cycle and depth errors inevitable in large-scale manufacturing. Lamellar (vertical sidewall) grating profiles are relatively easy to produce with high-contrast photoresist masks and collimated ion-beam etching, and exhibit high laser damage thresholds. The following discussions pertain to this type of structure.

Figure 3 shows a MLD grating design with a plot of expected diffraction efficiency as functions of the grating depth and duty cycle. Notice that a ridge of constant >99%

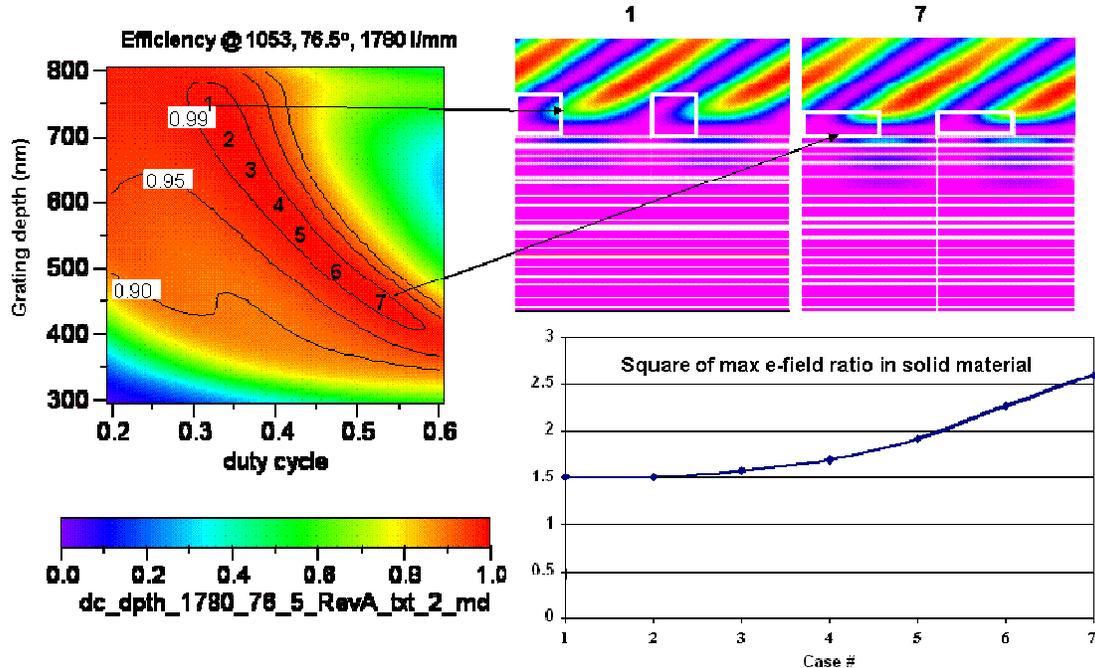


Figure 3. Left: Contour map of -1 order diffraction efficiency as function of grating depth and duty cycle for a 1780 l/mm MLD grating/ Right: outline of grating structure showing layers and grating shape for conditions 1 and 7 both exhibiting >00% diffraction efficiency, superimposed with the electric field distribution for each case.

diffraction efficiency is possible for a combination of grating height and duty cycle values. Examination of the electric field distribution generated on these gratings by the incident beam shows standing waves caused by interference from the incident and high-efficiency diffracted wave. These areas of high e-field penetrate into the back side of the grating ridges. It is here that damage to the grating due to multiphoton ionization and avalanche breakdown occurs (20). Analysis shows that the maximum e-field in the solid material is lowest for tall, thin grating lines so these should have the highest damage threshold (24). This has recently been experimentally confirmed (25). Typical laser damage thresholds for 1780 l/mm MLD gratings at 1053 nm, 10 ps, $\sim 75^\circ$ incidence angle, exhibiting >97% diffraction efficiency, are ~ 5 J/cm (measured for the normal incidence beam). This is more than 5 times the damage threshold of a similar holographic gold-overcoated photoresist grating that has slightly lower efficiency (24,25).

MLD gratings are not the answer for all short-pulse applications, however. Ultrashort-pulse high-intensity Ti:Sapphire lasers operating below 50 fs typically require a high-efficiency bandpass of up to 100 nm, at a central wavelength of ~ 800 nm. Designers of these lasers demand low-line density gratings to better manage dispersion and provide more alignment tolerance. Figure 4 compares diffraction efficiency –vs- wavelength for optimized MLD and gold gratings at 1480 l/mm, centered at 800 nm. Not only does the MLD design not cover the desired bandpass, but exhibits very narrow-band resonance regions where the interaction of the impedance in the grating and underlying stack couples light into one or more of the layers, acting as a waveguide. Figure 5 shows the electric field distribution at one of these resonance conditions. These types of

resonances seem to be ubiquitous, regardless of design details of the MLD stack. Although this resonance feature may be useful in some waveguiding applications, it is disastrous for a wide-bandwidth damage-resistant grating. Therefore, gold gratings remain viable for these applications.

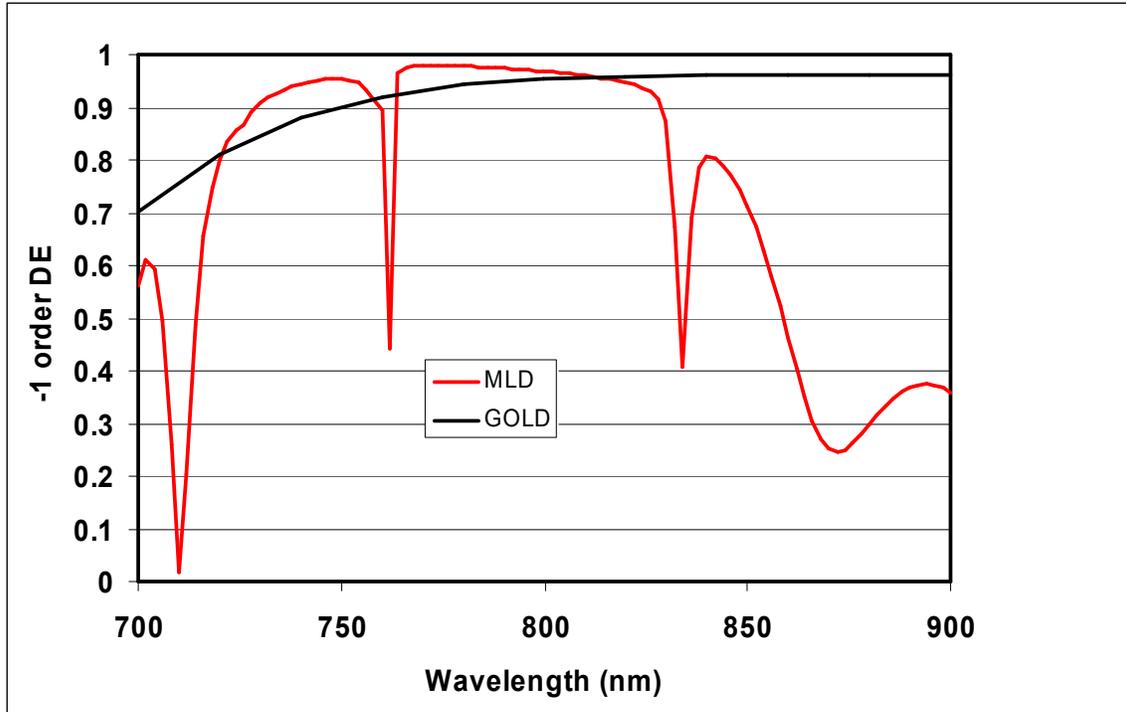


Figure 4. Comparison of diffraction efficiencies of gold and MLD gratings at 1480 lines/mm and 36 deg incidence angle. Notice the drop in efficiency of the MLD grating at 710, 762 and 834 nm. Here, the efficiency is coupled into the stack at the -1 or -2 transmitted order.

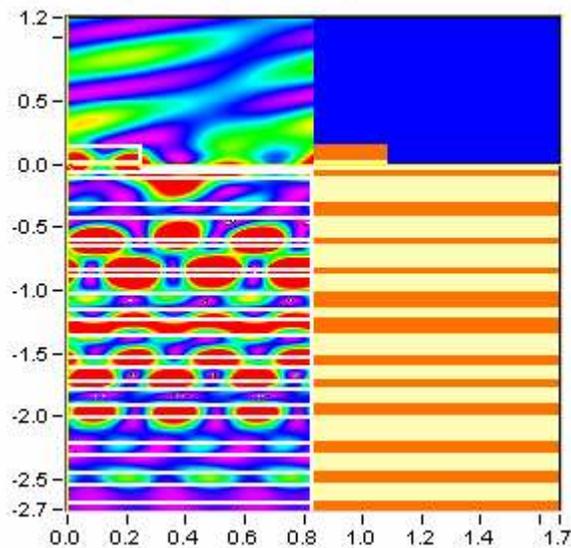


Figure 5. Electric field distribution in the multilayer stack of a MLD grating for a resonance condition of Figure 4, on left. On right is a pictorial representation of the MLD stack. Dimensions are in microns.

Gold-overcoated gratings operate best in TM polarization, in contrast to MLD gratings which require TE polarization. They also require a quantitatively different shape to be efficient. We use a design code based on a different convergence algorithm (26) to design metallic gratings. Sinusoidal modulations, relatively shallow with respect to the laser wavelength, work very nicely. Certain low-contrast photoresists generate reasonably sinusoidal profiles when processed holographically, but the efficiency is sensitive to the modulation depth, which is difficult to control over a large aperture exposure format in a thick resist layer. Therefore, we design gold gratings of the shape shown in Figure 6, whereby the modulation depth is fixed by the thickness of the photoresist layer (27). Exposure nonuniformities in this design translate into linewidth variations, to which the diffraction efficiency is relatively insensitive. These gold gratings can exhibit high diffraction efficiency for a very large bandwidth in the near infrared (28).

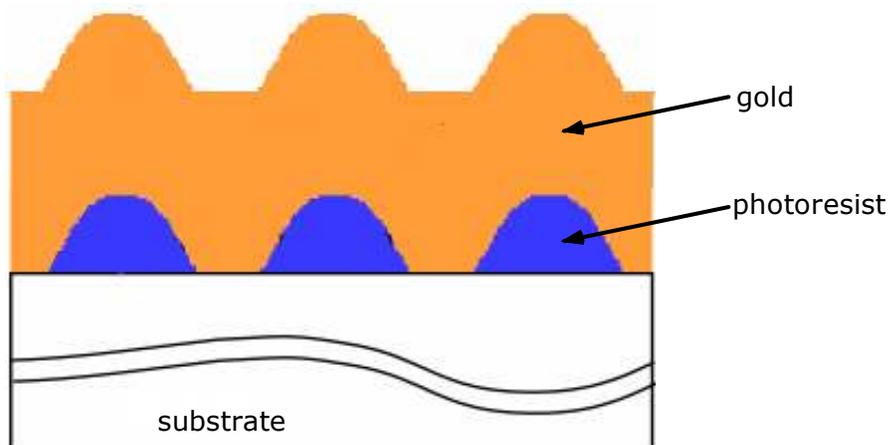


Figure 6. Optimized profiles for gold-overcoated reflection gratings.

Manufacture of large-aperture holographic plane gratings:

A process flowchart for manufacture of MLD gratings is shown in Figure 7. Dielectric oxide materials are deposited according to the design by a variety of methods. Electron beam deposition is a common method that gives very high laser damage threshold coatings for nanosecond-scale pulses. These coatings are typically somewhat porous, and so can undergo stress transformations during processing and operation in vacuum as they lose adsorbed water, even to the point of crazing (cracking) under sufficient tensile stress. Ion-assisted e-beam deposition uses an ion beam during deposition to densify the coating. This can drastically reduce the stress evolution of the film during processing and use. Ion-beam sputtering can produce very dense films with very low scatter, but at present this technique is limited to apertures on the order of 60 cm, too small for the largest grating applications.

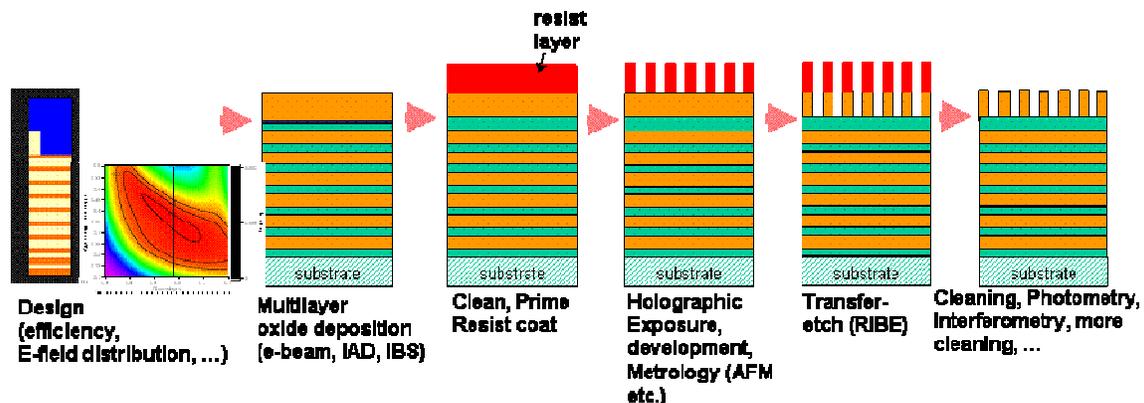


Figure 7. Process flowchart for MLD grating manufacture.

After coating of the stack, the grating substrate is cleaned and primed to facilitate adhesion of the photosensitive film. The priming agents, developed for the semiconductor processing industry, replace surface hydroxyl groups on the SiO_2 surface with organic groups; otherwise these surface hydroxyls will react with the base-sensitive photosensitive film and detach it during the development step.

The photosensitive film is typically a positive photoresist formulated for sensitivity to I-line (365 nm) or G-line (436 nm) wavelengths, developed again for the semiconductor industry. A positive photoresist is one that undergoes a photochemical reaction that renders it soluble in a base solution in areas where it has been illuminated with NUV light, but remains as a solid film where not exposed. Meniscus coating (29) has become the method of choice for depositing precise submicron-thick photoresist films from liquid solution onto the surface of large, heavy optics.

Exposure of the resist film is done on a large interferometer as shown in Figure 8. The largest commercially-available class-4 high coherence-length Ar-ion (351 nm) or Kr-ion (413 nm) lasers are used. We use fused-silica aspheric lenses 1.1 m in diameter as shown in Figure 8 to generate the two interfering plane waves for our largest format exposures. Alternatively, two large off-axis parabolas can be used in place of the folding

mirror/lens combination of this figure. However, the mirror/lens combination results in better diffracted wavefronts and is certainly easier to align and collimate.

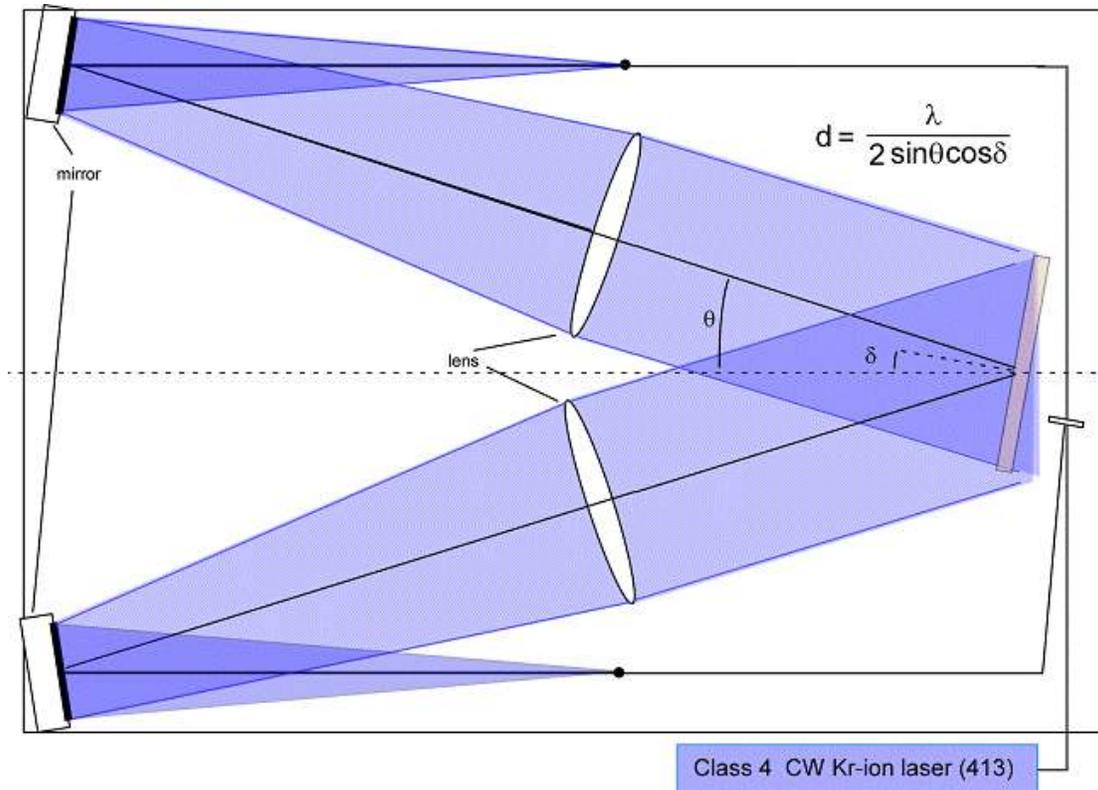


Figure 8. Large-format grating exposure setup.

Recently, a writing process has been developed that uses an interferometrically-controlled XY stage to move a substrate under two small interfering beams and pattern a grating by raster scanning (30). Gratings at 1740 lines/mm approaching 1 meter in dimension have been demonstrated. This technology has the potential to revolutionize the patterning of gratings to arbitrary scales, as well as to write variable line densities, or chirp, across a grating substrate (31).

After development of the exposed pattern using a base solution, the grating undergoes a series of characterization steps. There are more than 600 km of grating lines on a 800x400 mm 1740 l/mm grating, and all must be within a narrow width and height range for the finished part to be acceptable. We do full-aperture photometry of the photoresist grating at a use angle and wavelength chosen to give high sensitivity to the -1 order diffraction efficiency. The efficiency map generated is used to provide coordinates for submicron-scale examination of the grating lines by an atomic force microscope. Thus areas out of the expected bounds of diffraction efficiency can be examined in detail. The resist coating can be stripped off and the part re-processed if necessary. If deemed acceptable, it then undergoes ion-beam etching to transfer the pattern in resist to the underlying oxide layer(s). Our ion mill uses a rail transport system to scan a vertically

mounted grating back and forth in front of an RF generated ion beam 1 meter long by 3 cm wide, using a reactive gas $\text{CHF}_3/\text{Ar}/\text{O}_2$ blend to selectively etch SiO_2 with respect to the resist mask. It can process parts up to 2x1 meter in dimension.

After wet-chemical stripping of the remaining photoresist mask, the grating is characterized for diffraction efficiency and diffracted wavefront. Then it undergoes several cleaning steps to remove monolayers of processing residues that can dramatically reduce the laser damage threshold. A representative final product microstructure is shown in Figure 9.

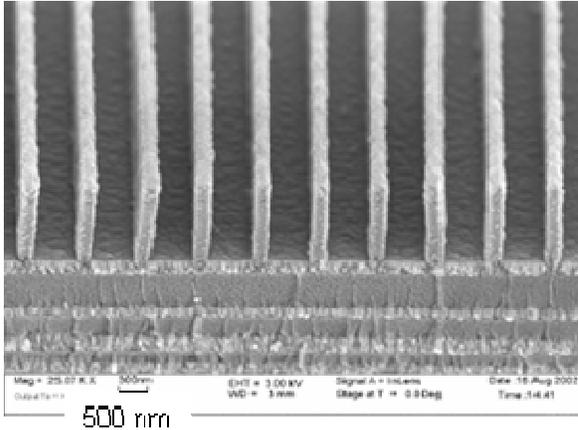


Figure 9. Scanning electron micrograph of finished 1780 l/mm witness grating.

Gold-overcoated gratings are manufactured in very similar fashion. The bare substrate is cleaned, primed, coated with resist, exposed, developed and characterized as above. Then, it is gold-overcoated by electron-beam evaporation. The amount of gold deposited is optimized to increase the duty cycle and laser damage resistance to the extent possible with this type of grating.

Grating Performance

To date we have fabricated >80 MLD gratings at 1740 l/mm, the largest being 807x417 mm in aperture. These represent over 12 square meters of grating area, with average diffraction efficiency over 96%. Most have been made for pulse compression at 1053 nm at various incidence angles for high-energy Petawatt laser systems being fabricated worldwide (reviewed in 32) although several were made for the Petawatt Field Synthesizer pump laser (33) that will operate at 1030 nm, 59° . Figure 9 shows efficiency statistics for these gratings.

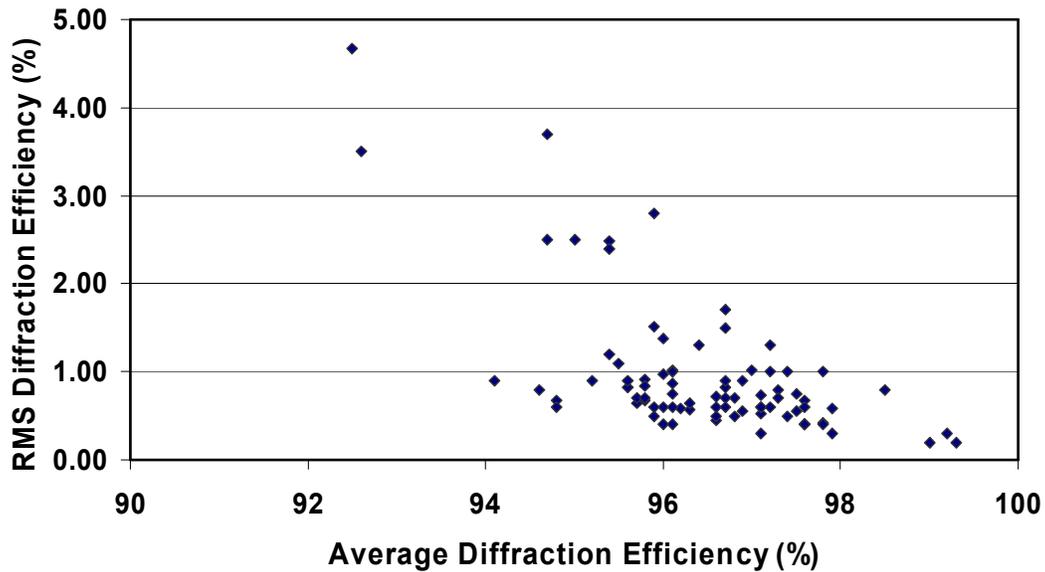


Figure 10. Average and RMS diffraction efficiency of ~80 1740 l/mm MLD gratings representing >12 m² of grating area.

Of special interest are gratings we have made that can be used in an all-reflective grating interferometer proposed for measuring gravity waves (34). These require maximal diffraction efficiency in the Littrow order at 1064 nm and must be capable of high average-power operation with no wavefront deformation. We have produced gratings 200x100 mm in aperture that exhibit >99% average diffraction efficiency at 1064 nm at the Littrow angle (67.8°), that have withstood ~2 KW/cm² with no observable change in beam quality (35). Recently, we have fabricated a similar grating 470x430 mm in aperture that has an average diffraction efficiency of 99.1% (Figure 11). Others have also recently demonstrated MLD gratings with >99% diffraction efficiency (36), but the optic of Figure 12 is by orders of magnitude the largest such grating produced.

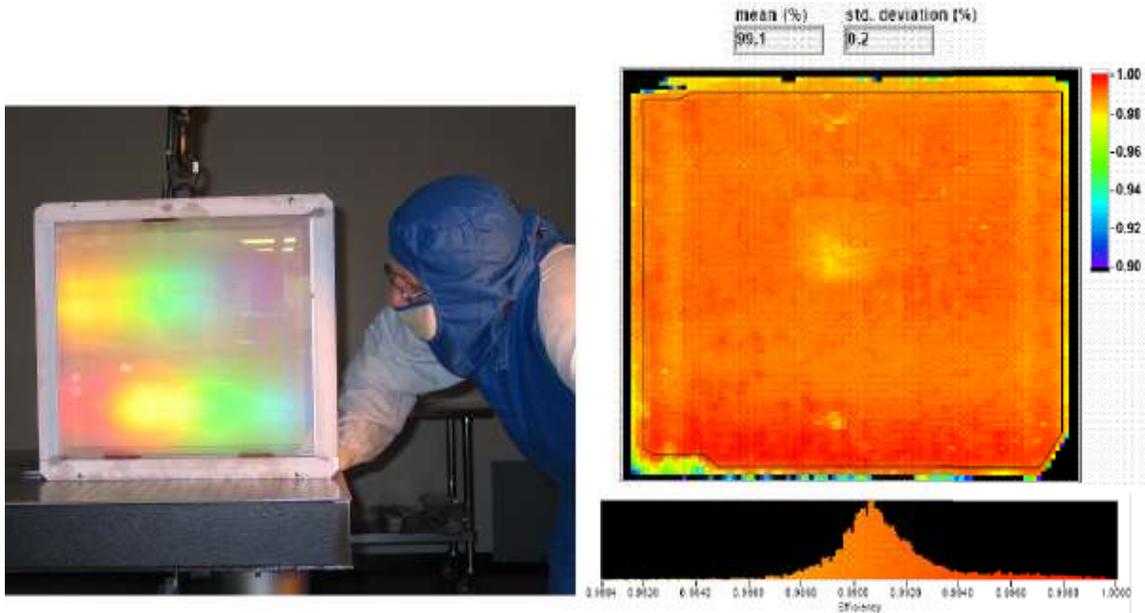


Figure 11. Photograph and diffraction efficiency map of a 470x430 mm, 1740 l/mm MLD grating exhibiting >99% average diffraction efficiency at the Littrow angle at 1064 nm, TE Polarization.

The diffracted wavefront of gratings can be measured interferometrically at the Littrow angle with the grating oriented so that its surface normal is rotated clockwise and then counterclockwise with respect to the incident beam (CW and CCW, respectively). The CW and CCW datasets can be subtracted and scaled by $\frac{1}{2}$ to cancel the surface wavefront and produce the holographic wavefront. This provides a measure of the errors associated with grating line curvature, chirp, etc. Similarly, the CW and CCW datasets can be added and scaled to cancel the holographic component and return the surface wavefront which is of course related to the flatness of the part. A collection of all wavefronts for a representative grating 470x430 mm in aperture is shown in Figure 12. To our knowledge these gratings have the flattest diffracted wavefront available at this time.

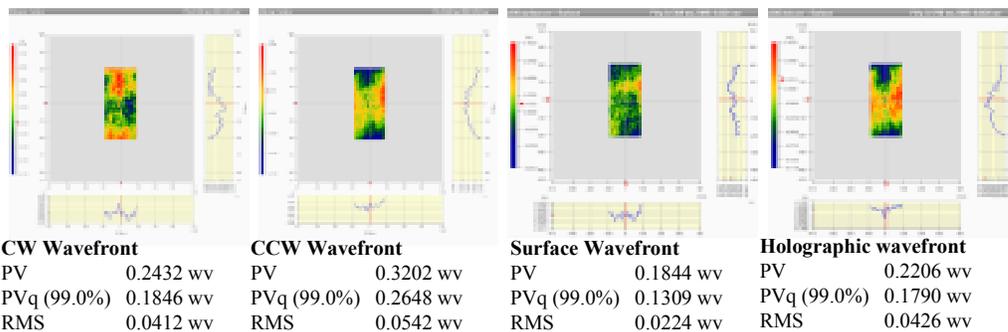


Figure 12. CW, CCW holographic and surface wavefronts of a 470x430 mm 1740 l/mm MLD grating measured at 1053 nm at the Littrow angle 66.7°.

Conclusion

Intense, short-pulse lasers using large-aperture MLD gratings are just coming online at this writing. These instruments will generate new data on the property of matter at extreme energy densities, and further research into nuclear fusion for energy as well as defense applications. New ultrashort-pulse lasers utilizing very large gold overcoated gratings are being built now as well, to probe the behavior of matter at even shorter time scales. At LLNL we are beginning to build short-pulse capability on the NIF laser. NIF ARC will utilize 4 beamlines with 32 gratings to deliver picosecond pulses for x-ray diagnostic generation and research into fast-ignition fusion. This is a burgeoning technological field that promises to yield very exciting science in the coming years.

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