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TITLE: MULTIFACET, METAL MIRROR DESIGN FOR EXTREME-ULTRAVIOLET AND
SOFT X-RAY FREE-ELECTRON LASER RESONATORS

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MULTIFACET, METAL MIRROR DESIGN FOR EXTREME-ULTRAVIOLET AND SOFT X-RAY FREE-ELECTRON LASER RESONATORS*

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Resonator mirrors with reflectance greater than ~40% are required if free-electron laser (FEL) oscillators are to be extended to wavelengths below 100 nm. The principle of total-external reflection has been used to design multifacet mirrors of several metal films that surpass this requirement between 10 and 100 nm. These reflectors have high reflectance over a relatively broad band which suits well the inherent FEL wavelength tunability. The practical limitations to achieving and maintaining high reflectance include oxide- and carbon-contamination, scattering, and surface-plasmon absorption.

Key words: aluminum films; carbon contamination; extreme-ultraviolet reflectors; free-electron lasers; metal reflectors; multifacet reflectors; oxide contamination; rhodium films; silicon carbide; silicon films; soft x-ray reflectors.

1. Introduction

The recent successful operation of several free-electron laser (FEL) oscillators in the visible and near-infrared spectral ranges [1,2] has encouraged researchers to examine the possible short-wavelength limits for these devices [3,4]. One of the major technological constraints presently blocking their extension into the extreme-ultraviolet (XUV) is the inherently low reflectance of available resonator mirrors, of the order of 10 to 20%. Unless this limitation is overcome, very high values of single-pass optical gain will be needed. For example, if each resonator mirror were to have a reflectance of only 25%, the small-signal gain would have to exceed 1600%

* Work was performed under the auspices of the U. S. Department of Energy.

just to begin oscillation, a value which may be difficult to achieve. Development of resonator mirrors with reflectance $\geq 40\%$ appears to be a prerequisite for future operation of FEL oscillators at wavelengths below 100 nm.

2. Present Status of Reflectors for the XUV

There are four types of normal-incidence reflectors under development for the XUV. First, smooth surfaces of chemically vapor-deposited (CVD), single-crystal, silicon carbide (SiC) have been produced and used successfully in synchrotron radiation beam lines. The highest measured values of the normal-incidence reflectance for SiC have varied between 40 to 50% for wavelengths between 60 and 220 nm [5,6]. Below 60 nm, SiC reflectance drops rapidly to less than 10%, as shown in figure 1.

Simple metallic films represent a second type of reflector, and the reflectance properties of two examples, platinum and gold, are compared in figure 1. Only for wavelengths longer than 250 nm does their reflectance exceed our minimum 40% requirement. The notable exception are aluminum films freshly deposited on smooth substrates in ultra-high vacuum (10^{-9} to 10^{-10} torr). With care, the normal-incidence reflectance can exceed 40% for wavelengths as short as 80 nm [7-9]. However, even in high vacuum the reflectance gradually decreases with time as an oxide forms on the surface. Overcoating with a layer of MgF_2 does prevent the oxidation, but high reflectance ($\geq 80\%$) is then limited to wavelengths longer than 120 nm. We will revisit the use of Al films in following sections.

The third class of reflector includes multilayer thin-film structures which operate on the principle of standing-wave interference of multiple reflections from the film interfaces. Since all dielectric materials are absorbing for wavelengths shorter than 110 nm, alternating metal layers having differing absorption coefficients are used. For the soft X-ray and XUV spectral regions from 10 nm to 110 nm, this technology generally has yielded reflectances less than 50% for near-normal incidence over a bandwidth limited to $\leq 10\%$ [10]. The status of normal-incidence reflectors as of 1983 was summarized by Attwood, et al. [11], which is reproduced here as figure 2.

Multifacet reflectors, the fourth type of XUV reflector and the subject

of this paper, involve multiple reflections from a series of metal mirrors. These make use of the principle of total **external** reflectance (TER) which is possible only when the refractive index is less than unity. We have determined that semi-TER ($R < 100\%$) is possible for a few metals over certain spectral ranges in the XUV. Surprisingly high values of retroreflectance (redirection of the optical beam by 180°) are possible, especially with S-plane reflections at large angles of incidence. In addition, the high reflectance extends over a relatively broad range, a feature that well suits the inherent tunability of FEL oscillators.

In the following sections, we review the potential for the multifacet, vacuum-deposited metal films, the risks due to practical fabrication limitations and environmental contamination, and how they would be used in a ring-resonator configuration.

3. Requirements for Total-External Reflectance Mirrors

We first describe the fundamentals of total external reflection. When light travels across the boundary from one medium of refractive index n_1 into a second medium of index n_2 in which the velocity is **greater**, total reflection occurs when the angle of incidence exceeds the critical angle θ_c defined by $\sin\theta_c = n_2/n_1$. If the first medium is vacuum for which $n_1 = 1.0$, TER can occur only if $n_2 < 1.0$. For metals, there are many spectral regions where the real part n of the refractive index $N = n + ik$ is less than unity. This is less common for dielectrics, however. A second requirement for total (100%) reflection is that the extinction coefficient k of the media be absolutely zero (which is never attained). Still, if the absorption is small ($k \ll 1.0$ while $n_2/n_1 < 1.0$), a very large reflection can occur. Finally, higher values of reflectance are achieved with S-polarized light rather than with P-polarization. Figure 3 visualizes the conditions for TER.

Soviet researcher A. V. Vinogradov and colleagues [11-13] were the first to analyze the potential for high-retroreflectance in the soft X-ray regime using multiple, grazing-incidence reflections beyond the critical angle. They considered a single, cylindrical mirror configuration of a variety of metals and used optical constants derived from the theoretical tables of Henke et al. [14] For use as resonator mirrors, however, we suggest a multifacet, flat

mirror configuration as indicated in figure 4. Flat mirrors will cause much less astigmatism which is essential in an FEL resonator to obtain maximum intensity within the magnetic undulator. Also, flats are easiest to fabricate with the required surface figure. (The final facet should be an off-axis paraboloid for collimating the beam.)

4. Examples for the XUV: Aluminum and Silicon Retroreflectors

We have scrutinized the available tabulations of optical constants [14-16] for the metallic elements and various compounds, searching for wavelengths for which both k and n are sufficiently less than unity. In the XUV spectra' region ≥ 25 nm, we initially identified three candidates: Al, crystalline Si, and Mg. The spectral dependence of the optical constants of Al is shown in figure 5 for reference. Vacuum-deposited films of Al are especially attractive since they have been characterized by a number of researchers over the last 35 years. Also, since the films are transparent in this region, interference effects with the substrate, e.g. SiC, can enhance the reflectance slightly. Less information (but still adequate) is available for crystalline Si films in the XUV. Practical use of Mg films is doubtful since an absorbing oxide forms rapidly at the surface.

The reflecting properties of a smooth, uncontaminated Al film on SiC at the wavelength of 50 nm are shown in figure 6 as a function of the number of mirror facets. Optical constants from the critical review and tabulation of Smith et al. [8,9] were used in the computations. According to the analysis of Goldstein et al. [17] for an FEL conservatively designed for 50 nm, the predicted 60% retroreflectance indicated in figure 6 is above the level needed to reach laser oscillation. Interestingly, only six facets are needed to achieve this value. Figure 7 shows the calculated retroreflectance over the entire spectral range (25 to 130 nm) where TER is operative for various numbers of mirror facets. It is obvious that the reflectance of a multifaceted aluminum mirror greatly exceeds that of a single, normal-incidence Al mirror over most of this region.

Examination of the optical constants of crystalline-Si films [18] reveals that TER occurs over the same 25- to 80-nm spectral range as for Al. Figure 8 illustrates the spectral characteristics of the retroreflectance of multifacet reflectors of this element.

5. Example for Soft X-Ray Wavelengths - Rhodium Retroreflector

In the soft X-ray region below 14 nm, a variety of metal coatings may be useful as multifacet retromirrors in laser resonators. These and the approximate wavelengths for which maximum reflectance enhancement occurs include Rh (12 nm), Ru (12 nm), Os (12 nm), Mo (12 nm), Ag (10 nm), Au (10 nm), In (7.5 nm), La (6.5 nm), and Ba (5.5 nm) [14,19]. The calculated retroreflectance (60 to 65%) of a multifaceted rhodium reflector at 12 nm (figure 9) is very encouraging as is the 30% spectral band (10 to 13.5 nm) over which the reflectance exceeds 40%. A corresponding plot of reflectance versus number of mirror facets at 12 nm is shown in figure 10. Rhodium films are especially interesting since the available tabulation of optical constants [19] is for films deposited in modest vacuum (10^{-5} torr). From this fact we expect that oxide growth may not be serious problem.

6. Risks of Contamination and Surface Roughness

Unless great care is taken, the TER enhancement in retroreflectance of a multifacet mirrors will not be realized. The hazards to be avoided are surface contamination by oxide and carbon films and surface roughness which represent potential losses by absorption or scattering. We now discuss the magnitudes of these hazards and the conditions necessary to minimize their effects.

Oxide contamination. In general, the metal films must be deposited and maintained in high vacuum to minimize the formation of oxide layers that absorb strongly at all wavelengths below 150 nm. This is evident for aluminum in figure 11 in which Smith et al. [9] have summarized the normal-incidence reflectance for a variety of Al surface conditions. In particular, our computations of the potential degradation of an Al retroreflector (figure 12) show that very little oxide formation can be allowed before the reflectance drops below 40%. Several researchers have determined that ultra-high vacuum (10^{-9} to 10^{-10} torr) and fast deposition rates are necessary to preclude oxide contamination [20,21]. At these vacuum pressures, H. E. Bennett [21] was not able to detect (via ellipsometry) the growth of an oxide layer on Al films over the period of two days. Oxide growth on Si and Rh is expected be much less of a threat.

Carbon film contamination. Surface contamination by carbon compounds, which are normally present in all environments, must be prevented to minimize optical absorption. As shown in figure 13, a carbon layer of only a few angstroms thickness will seriously degrade the TER condition required for multifaceted Al retromirrors. We have also calculated the potential for degraded performance of a multifaceted Rh mirror at 12 nm due to growth of a carbon epifilm (see figure 14). From comparison of figures 13 and 14, we conclude that carbon contamination should have less serious consequences near 10 nm than near 50 nm. The presence of high-energy photons such as present in synchrotrons, has been observed to greatly increase carbon film growth (~10 nm) on optical surfaces [22]. Synchrotron users have been able to control the rate of carbon accumulation on optical surfaces by use of ultra-high vacuums (10^{-10} to 10^{-11} torr). Of course, a superior solution is to eliminate all carbonaceous compounds from the vacuum environment. Recently, an rf plasma discharge of O_2 or H_2O vapor has been used to effectively remove all traces of such contamination on metallic optical gratings [23,24].

Scattering losses. The third requirement is that the reflecting films be deposited on smooth substrates to minimize scattering losses. For wavelengths much greater than the rms surface roughness σ , that is $\lambda \gg \sigma$, the effect of scattering on the specular reflectance, $R(\theta)$, at some angle of incidence, θ , is approximately [25-27]

$$R = R_0(\theta) \exp[-(4\pi\sigma \cos\theta/\lambda)^2], \quad (1)$$

where R_0 is the reflectance for a perfectly smooth surface. For XUV wavelengths ≥ 30 nm, mirror substrates with rms surface roughness of ≤ 5 Å are sufficiently smooth to limit scattering losses to $\leq 4\%$ for normal incidence. When used at 80° incidence, the losses should decrease to $\leq 0.1\%$ according to equation (1).

Although the derivation of equation (1) should make its application at soft X-ray wavelengths less valid, we use it here for estimation purposes for relatively smooth surfaces at 12 nm (120 Å). For example, incidence at 80° on Rh films with 5 Å rms surface roughness would result in 0.8% scattering loss per reflector facet. The net drop in retroreflectance (9 facets) would be about 5% (from 65% to 60%).

Surface-plasmon absorption. Polished substrates with the above smooth surfaces are attained in practice on flat and spherical substrates of fused silica, glass, Si, and occasionally SiC. For large angles of incidence, such as 80° , and relatively long wavelengths ≥ 30 nm, still rougher surfaces, e.g. with σ equal to 10 to 15 Å, could be tolerated. However, at wavelengths near a surface plasmon resonance, surface roughness can produce absorption loss within the metal. For Al, such a resonance occurs at 10.54 eV (118 nm) [28], and the effect on reflectance is shown in figure 15. This motivates a tighter limit on the tolerable roughness when using Al reflectors in the vicinity of this wavelength.

7. Practical Implementation

It is obvious that the practical application of multifacet metal reflectors will require deposition of the films in ultra-high vacuum with the reflector substrates mounted in place in the laser resonator. This is convenient for an FEL oscillator in which the reflectors are usually mounted in vacuum anyway. With this provision, it would be possible to periodically overcoat the metallic films as required to offset the effects of any gradual deterioration that may occur. Of course, the total thickness of the films must not become too great or else surface roughness will become a problem. It is conceivable, too, that an ion gun mounted in the vacuum chamber could be used to periodically sputter away aged films and then evaporate fresh layers. Certainly, initial removal of carbonaceous compounds from the vacuum environment by use of a rf plasma discharge, cited above, will also be beneficial.

8. Plans for Implementation

Los Alamos researchers Scott, Arendt, and Newnam have begun a project to implement the multifaceted, metal-mirror concept. Included in the research plans are ellipsometric measurements of the oxide- and carbon-contamination growth rates as a function of O_2 , H_2O , and CO partial pressures and reflectance measurements of single- and multiple-facet reflectors for vacuum environments from 10^{-8} to 10^{-10} torr. If the results are encouraging, the multifacet retroreflectors will eventually be used in an XUV FEL oscillator as shown in figure 16. Recent three-dimensional calculations of the single-pass, small-signal gain for a soft x-ray FEL at 12

nm have shown that the 60% net reflectance possible for Rh retromirrors should be sufficient to achieve laser oscillation. This is indeed an exciting prospect!

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Figure Captions

Figure 1. Reflectance of CVD-SiC at normal-incidence with comparison to platinum and gold films. After Rehn and Choyke [5].

Figure 2. Present and projected reflectance of normal-incidence mirror configurations as compiled by Attwood, et al. [11]. Recent data points are those for Mo/Si multilayer reflectors produced by Barbee, et al. [10].

Figure 3. Reflection of an optical beam from a vacuum-metal interface for angles larger than the critical angle $\theta_c = \sin^{-1}n/n_0$. If $k = 0$ when $n < 1$, total external reflection occurs; with k sufficiently small, very large reflection can occur.

Figure 4. Multifacet retroreflector composed of N flat metallic reflectors. Total-external reflection is possible only when the number of mirror facets N is large enough so that the angle of incidence $\theta = 90^\circ \times (N - 1)/N > \theta_c$.

Figure 5. Spectral dependence of the complex refractive index of aluminum films deposited in high vacuum (after Smith, et al. [8,9]). Semi-total external reflectance is possible from 25 to 80 nm where both n and k are simultaneously less than unity.

Figure 6. Calculated retroreflectance ($\theta=180^\circ$) and single-facet reflectance for S-polarized XUV radiation at 50 nm for multifacet aluminum films deposited on smooth SiC substrates in ultra-high vacuum. Calculations made use of the tabulation of optical constants by Smith, et al. [8,9]. Notably, only six mirror facets are needed to reach 60% reflectance with 64.4% being the maximum achievable.

Figure 7. Total retroreflectance ($\theta=180^\circ$) versus XUV wavelength for multifacet aluminum mirrors deposited on smooth SiC in ultra-high vacuum. Calculations were made for S-polarization using the tabulation of optical constants by Smith, et al. [8,9]. Below 90 nm, the reflectance attainable with multifacet mirrors is much greater than that for a single ($N=1$), normal-incidence Al mirror.

Figure 8. Total retroreflectance ($\theta=180^\circ$) versus XUV wavelength for multifacet, crystalline Si films deposited on smooth Si in ultra-high vacuum. Calculations were made for S-polarization using the optical constant compilation by Edwards [18]. Below 90 nm, the reflectance attainable with multifacet mirrors is much greater than that for a single ($N=1$), normal-incidence Si mirror.

Figure 9 Retroreflectance of multifaceted rhodium coatings on smooth SiC substrates in the soft X-ray region for S-polarized radiation. Optical constants for Rh films deposited in a moderate vacuum (10^{-5} torr) were used in the calculations because oxidation is not serious for Rh at ambient temperatures [19]. Retroreflectance greater than 60% is calculated at 12 nm.

Figure 10. Calculated retroreflectance and single-surface reflectance of rhodium films on smooth SiC for S-polarized radiation at 11.9 nm as a function of number (incidence angle) of mirror facets. Only seven mirror facets are needed to achieve 60% net reflectance with 64.9% maximum achievable. Optical constant tabulations by Lynch and Hunter [19] were used.

Figure 11. Reflectance of various types of aluminum surfaces at room temperature (after Smith, et al. [9]). Highest reflectance (curve a) is achieved with Al films deposited on smooth substrates in ultra-high vacuum (10^{-9} to 10^{-10} torr) with high deposition rates. The effect of light scattering and surface plasmon absorption (at 10.6 eV) is shown by curves b and c. Curve b is for Al on a clean microscope slide and curve c is for a uhv film with rms roughness of 18 Å [15].

Figure 12. Influence of oxide thickness on the calculated retroreflectance (S-polarization) of a nine-facet aluminum mirror at XUV wavelengths of 50 and 100 nm. Optical constant tabulations for Al by Smith, et al. [8,9] and Al_2O_3 by Hagemann et al. [15] were used.

Figure 13. Influence of carbon film contamination on the calculated retroreflectance (S-polarization) of a nine-facet, aluminum-coated mirror (smooth SiC substrate) at XUV wavelengths of 50 and 100 nm. Optical constant tabulations for Al by Smith, et al. [8,9] and C by

Hagemann, et al, [15] were used.

Figure 14. Influence of carbon film contamination on the calculated retroreflectance (S-polarization) of a nine-facet, rhodium-coated mirror (smooth SiC substrate) at soft X-ray wavelengths of 10.5 and 11.9 nm. Optical constant tabulations for Rh by Lynch and Hunter [19] and C by Hagemann, et al. [15] were used.

Figure 15. Degradation of the normal-incidence reflectance of uvv aluminum coatings on rough surfaces caused by the surface plasmon absorption resonance at 10.6 eV. Use of a fire-polished microscope slide (Feuerbacher and Steinmann [20]) or a bowl-feed polished glass substrate minimized plasmon absorption (after Endriz and Spicer [28]).

Figure 16. A free-electron laser ring-resonator design for extreme-ultraviolet and soft x-ray wavelengths has two unique features:
1) intracavity grazing-incidence mirrors (hyperboloids) to expand the beam incident on 2) multifaceted retroreflectors (N-1 flats and one off-axis paraboloid for beam collimation). Below 100 nm the multifaceted mirrors exhibiting semi-total external reflectance, e.g. Al, Si, Rh, Ag, Ru, Os, are used to attain a retroreflectance $\geq 40\%$ in various XUV wavelength regions.

Fig. 1

CVD-Silicon Carbide Normal-Incidence Reflectance

Exceeds 40% from 60 to 220 nm

CVD-SiC data: V. Rehn and V. Jones, 1978.

Au data: H. Hagemann et al., 1974.

Pt data: W. Hunter et al., 1979; J. Weaver, 1975.

Fig. 1

● T. Barbee, et al., (Appl. Opt., 1985).

* Miyazaki and Sato, 1980.

Compiled by Optics Working Group (D. Attwood, ed.),

XUV FEL Conference (Brookhaven Nat'l. Lab., 1983)

Fig. 2

REFLECTANCE ENHANCEMENT IS DUE TO QUASI TOTAL EXTERNAL REFLECTION

$\sin \phi_c = n_m$ AT CRITICAL ANGLE

IF $\phi > \phi_c$ AND $k=0 \rightarrow R = 100\%$

$\phi > \phi_c$ AND $k \neq 0 \rightarrow R \ll 100\%$

$\phi > \phi_c$ AND $k \ll 1.0 \rightarrow R \text{ large}$

Fig. 4.

Retroreflection

Case 1 ($\theta = 90^\circ$)

Case 2 ($\theta = 45^\circ$)

$$\theta_0 = \left(\frac{N-1}{N}\right) \cdot 90^\circ$$

Case 3 ($\theta = 60^\circ$)

Case 4 ($\theta = 84^\circ$)

15 Flat Facets

Fig. 5

Fig. 1

60% RETROREFLECTANCE AT 50nm REQUIRES SIX OR MORE ALUMINUM MIRROR FACETS

CALCULATED RETROREFLECTANCE OF MULTIFACETED ALUMINUM MIRROR EXCEEDS 50% FOR XUV AND VUV WAVELENGTHS >40nm

198

CALCULATED RETROREFLECTANCE OF MULTIFACETED SILICON MIRROR EXCEEDS 65% AT 50 nm

CALCULATED RETROREFLECTANCE OF MULTIFACETED RHODIUM MIRROR EXCEEDS 60% AT 12nm

60% RETROREFLECTANCE AT 12nm REQUIRES SEVEN OR MORE RHODIUM MIRROR FACETS

Fig. 11

Aluminum Reflectance Depends on Environment

Figure from D. Smith, E. Shiles, and M. Inokuti,

Argonne Report ANL-83-24, 1983

CHM-VG-8340

SURFACE OXIDE LAYER DEGRADES XUV REFLECTANCE OF MULTIFACETED ALUMINUM RETROREFLECTOR

198

CARBON CONTAMINATION DEGRADES XUV REFLECTANCE OF MULTIFACETED ALUMINUM RETROREFLECTOR

**CARBON CONTAMINATION DEGRADES
SOFT X-RAY REFLECTANCE
OF MULTIFACETED RHODIUM RETROREFLECTOR**

Reflectance of Aluminum Films Decreases

Strongly with Surface Roughness in the

Vicinity of the Plasmon Absorption Resonance

Figure from J. G. Endriz and W. E. Spicer,

Phys. Rev. B 4, 4144 (1971).

MULTIFACET, ALL-METAL MIRRORS WITHSTAND HIGH INTENSITY IN RING RESONATOR