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SHORT WAVELENGTH OPTICS FOR FUTURE FREE ELECTRON LASERS*

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I. Overview

Although much free-electron laser work is directed toward achieving sufficient single-pass gain to be useful for research purposes, the availability of mirrors of high reflectance for the vacuum ultraviolet and soft x-ray regime would make resonant cavities a possibility. In addition, as in ordinary synchrotron radiation work, mirrors are required for the construction of realistic experiments and for beam manipulation purposes such as folding and extraction. The Working Group discussed a number of approaches to reflecting optics for free electron lasers, which are summarized here, and described in some detail in the following sections.

Figure 1 summarizes the Working Group's collective understanding of the present availability of normal incidence reflecting optics, together with an estimation of what might become available in the near term (~5 years). For the visible and near U.V. down to about 1500A, multilayer interference coatings can achieve normal incidence reflectance values as high as 95%. In addition, aluminum mirrors over-

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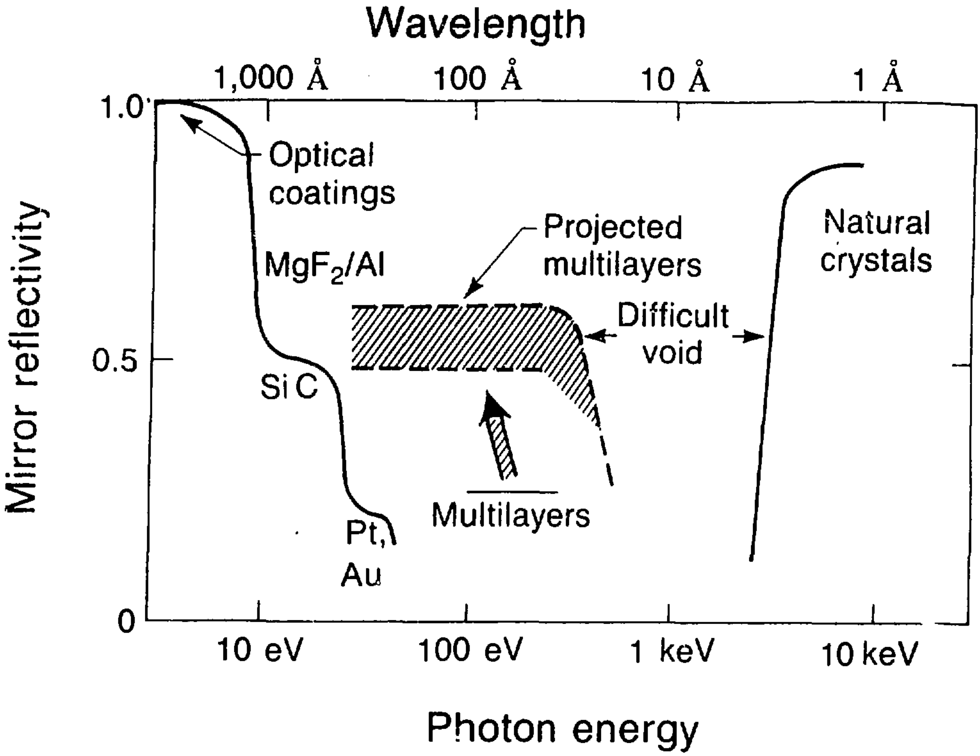

Figure 1. Summary of present and projected normal incidence mirror capabilities with presently understood technologies.

coated with magnesium flouride (MgF_2) achieve reflectances of 80-90% in this same spectral region[1]. For wavelengths shorter than 1500Å achievable reflectances drop dramatically for coated and multicoated surfaces. For wavelengths between 1200Å (10 eV) and 500Å (25 eV), single surface reflectances approaching 50% can be achieved with silicon carbide, a material that additionally has high thermal loading attributes. For still shorter wavelengths, extending to perhaps 300Å (40 eV) noble metal mirrors of gold, platinum, and iridium have conventionally been used, but achieve reflectances of no better than 25%; at shorter wavelengths their reflectance falls precipitously, as λ^4.

In the region beyond 300A (50Å to 300Å), appreciable normal incidence reflectance values (say R > 10%) have yet to be demonstrated. The best hope appears to lie in the development of specially fabricated multilayer reflectors based on new material combinations. A few recent measurements in the 40Å to 100Å region, have demonstrated that peak reflectivities of 10% to 20% can be obtained. Computations indicate that reflectances of up to 50% may be achieved in this difficult spectral region. Recent normal incidence measurements reported by Underwood and Barbee[2], and by Henry et al[3]., support this conjecture. Figure 2 summarizes the normal incidence reflectance calculations of Rosenbluth[4]. Note that these calculations are idealized in several ways. Material compatabilities were not a subject of that study. Substantial experimental efforts are required by materials scientists to determine the effects of material interpenetration, long term interface stability, and the control of surface roughness. At the present time, achievable d-spacings limit realistic opportunities to wavelengths approaching 20Å (E < 600 eV). For wavelengths between 20Å and 5Å there is presently no capability for normal incidence reflectance. For wavelengths less than 5Å, high peak reflectance is achievable with natural crystals[5].

In addition to questions of fabrication, one must also consider the practical but widespread problems of mirror degradation by radiation damage, surface contamination and thermal distortion. There is substantial evidence that all of these problems exist and are impediments to progress not only in the field of FEL research, but to

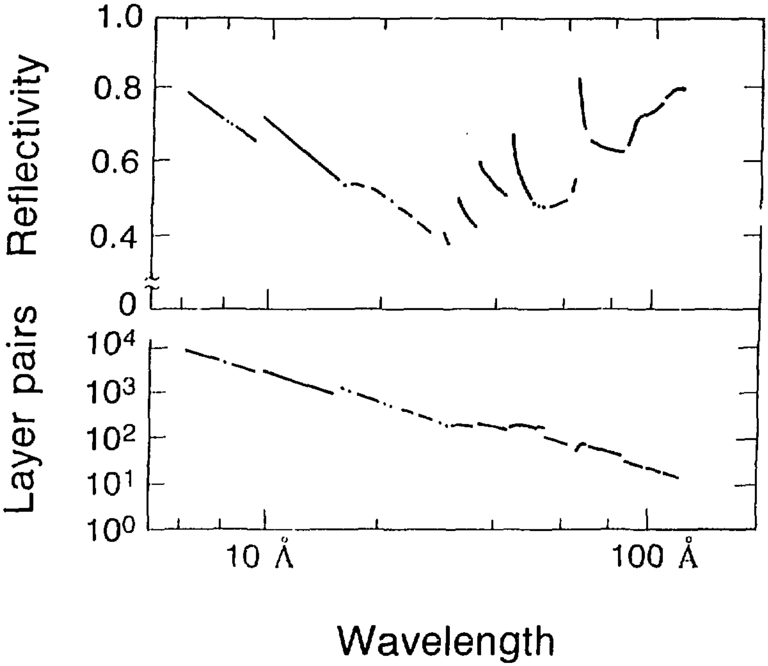

Figure 2. Calculated normal incidence peak reflectivity from idealized multilayer coatings of optimum material combinations. Required number of layer pairs, N, is also shown. Calculations were performed by A. Rosenbluth[4] of the University of Rochester. Much work remains for materials scientists who must determine which combinations and d-spacings are suitable for stable interface formulation.

the broader area of synchrotron radiation usage. Further experimentation is clearly required.

Diffractive optics were also reviewed by the working group. Although these structures are not seen as having a primary role in cavity definition, they will serve a major role in subsequent utilization of FEL radiation, as focusing and diffractive elements. In addition, they may play an important role as pulse extracters (switch out devices). It is necessary that diffraction efficiencies, which currently approach only 10%, be addressed by directing community efforts towards phase structures (rather than opaque structures), which would achieve efficiencies approaching 40%. Additional topics reviewed in this working group were those of substrate preparation, roughness and scattering, glancing angle performance, spectral selectivity, polarization effects, etc., all of which are described to some degree in the following sections.

In summary, optical capabilities were reviewed for the VUV and soft X-ray regions of the spectrum - regions in which it is presently difficult to work. However, it was the group's consensus that because of pressures from several scientific and technological disciplines, this region of the spectrum will experience rapid growth in the coming years. The field is ripe for significant progress. With additional, well directed support this "last frontier" spectral region will yield significant new opportunities across the scientific spectrum. New or intensified efforts are required to develop broader multilayer capabilities, including very low Z materials, as well as new experimental efforts to better understand thermal loading limits, surface degradation, and counter measures which will permit applications to proceed across the scientific and technological spectrum.

II. Non-Interference Reflective Optics

The subject of single surface reflecting optics for VUV and soft x-ray radiation has been extensively reviewed from the viewpoint of synchrotron-radiation applications by several authors[6,7]. A few of these reviews are listed in the references, and others are contained as references therein. Two recent workshops, the LBL workshop on Synchrotron Radiation Vacuum and the Mirror-Darkening Problem (Lawrence Berkeley Laboratory, 25 - 26 July 1983, R. Avery, Ed.) and the SSRL New-Rings Workshop (Stanford, July 27 through 29, 1983, A. Bienenstock, Ed.) have not yet issued reports. In both workshops reflective optics for use with new state-of-the-art wiggler and undulator sources were discussed. The SSRL Workshop included a working group entitled "Novel Optics For Synchrotron Radiation," (chaired by V. Rehn). Because of the considerable background literature available in this area, we include only short, partly tutorial paragraphs on the problems which we see in free-electron laser applications, although most of these problems are generic to VUV and X-ray mirrors at storage ring facilities. "Conventional" mirrors for application in the VUV and X-ray spectrum are generally made by grinding a suitable substrate material to the required optical figure and then polishing it to make the final corrections to this figure and to bring the reflecting surface to a desired degree of smoothness. This is then coated by a film of highly reflective material (usually a heavy metal) which provides the best reflectivity available for the spectral range of interest. The reason for this separation of function is that the polishing process (or in some cases the single-point diamond turning process) used to produce the optical figure and smoothness is highly material sensitive. For the FEL, state-of-the-art performance is required in reflectivity, optical figure, smoothness (low scatter), recoatability and power handling. Many of these properties are enhanced by proper selection of substrate material and reflective coating. Note that many of the substrate properties needed for "conventional" mirrors and synthetic multilayer optics are the same, and will not be separately discussed under that heading.

Substrate Materials. The criteria for substrate selection have been outlined above. Insofar as conventional optical grinding and polishing techniques are concerned, by far the most experience has been obtained with optical glasses and vitreous materials such as fused silica, in particular proprietary low expansion varieties such as "ULE", "Cer-Vit" and "Zerodur"[8]. The state-of-the-art optical figure achievable for a small (~ 10 cm dia.) mirror is typically $\lambda/200$, where the test wavelength is typically visible at 6328Å or 5770A. By this it is meant that macroscopic departures from the derived planar or spherical surfaces can be held to 30Å over a distance 10 cm, for 90% of the area. The smoothness obtainable with glassy materials is about 3A rms.

It is more difficult to achieve figure accuracy on surfaces other than spheres or planes, or with materials other than vitreous materials. Notable exceptions are: single-crystal silicon, which can be polished plane to a very smooth surface (again, about 3Å rms.), silicon carbide which has been chemically vapor deposited, and electrodeless nickel. The last material is the only metal which has been polished to a smoothness comparable with that obtained on glasses. Desired figure on metalic reflectors can be generated more quickly and less expensivly by using the diamond turning process, and then given a final polish to remove turning marks. However, experiments to date using this process have not met expectations, although much effort has been expended. The microscopic smoothness obtainable with metals such as copper, molybdenum and aluminum is 4 to 10 times worse than the 3Å achieved in state-of-the-art work. Another serious problem with metals is their dimensional instability (secular instability), which causes them to change their geometrical figure over a period of time. These materials questions have been studied extensively[9] by x-ray astronomers for projects such as HEAO and AXAF, with the result that metal mirrors have been abandoned in favor of vitreous ones for all but the lowest resolution x-ray telescopes.

In FEL applications, distortions due to temperature gradients must also be avoided. It is clear that if the thermal load on the mirror due to beam heating causes it to bulge by 30Å or more, the optical figure has been severely degraded. A useful figure of merit for gauging the potential distortion of a mirror substrate material is given by κ/α, where κ is the thermal conductivity and α is the thermal expansion coefficient; high values of κ/α indicate reduced distortion. With a value $\kappa/\alpha = 3.5 \times 10^5$ W/cm, silicon carbide is a good material. For comparison, κ/α values for copper and fused silica are 2.4×10^5 and 3.75×10^4 W/cm, respectively.

Because the exposure of mirrors to synchrotron radiation leads to the formation of a carbonaceous coating that reduces reflectance, mirrors, especially those for use between 25 and 45Å, should be stripped of the reflective coating and recoated periodically. To avoid repolishing, the mirror substrate material should be chemically inert to allow such a process without degradation of the surface smoothness or optical figure. Silicon carbide is again a good choice, for it resists all acids, and all but strong, hot alkali. Fused silica can be stripped several times if it is done with care using a minimum exposure to strong acids such aqua regia.

Coating Materials. The selection of coating material is based on reflectance in the desired spectral range, quality (smoothness, adherence and continuity) of films obtainable, and resistance to degradation in use. For use in the 400-1200Å spectral range (10 to 30 eV), uncoated silicon carbide has the highest reflectance and is superior in all other respects. Below 400Å the most common choices are gold and platinum, although nickel, silica, aluminum oxide and Zerodur (a proprietary product of Schott Optical Glasses) have been used. Typically Pt and Au show $0.1 < R < 0.2$ in the range 300 to 500Å, while other materials do no better. Below $\lambda = 300$Å, the reflectance of materials decreases proportional to λ^4, reaching a value near $R \sim 10^{-6}$ near 10Å. The best hope for normal-incidence mirrors in this range are the multilayer coatings discussed in a later section.

Film quality is usually best over a limited range of thickness, and is strongly influenced by the substrate surface. For example, thicknesses greater than 2000Å often result in increased optical scattering, while thicknesses less than 100Å are frequently characterized by discontinuous or "pinholed" films. Diffusion barriers are often used, especially on metal mirrors. A chromium or nickel film of 100-200Å thickness is sometimes used as a barrier to provide a bond between the substrate and the film, and is particularly effective in the case of gold, which becomes rough without it. Platinum films are more stable against interdiffusion than gold, and are often smoother, especially in thin coatings. Platinum and gold have both been successfully deposited on silicon carbide without a diffusion barrier, although Au/Pt/SiC proves to be smoother than Au/SiC. Deposition techniques can also affect coating quality. Careful thermal depositions using an electron beam in ultrahigh vacuum have produced reliable high-quality coatings. Sputter deposition can also give excellent results. Degradation in use is the subject of a later section of this report.

Glancing-Incidence Reflection Optics. As previously noted, no material yields high reflectance at normal incidence in the wavelength region below 300Å ($E > 40$ eV). However, since the real part of the refractive index of most materials becomes less than unity in this region, it is possible to utilize the phenomenon of "total external reflection" by utilizing glancing angles of incidence ($\theta = 90°$ - optical angle of incidence). High reflectance values ($R > 0.5$) can be obtained if θ is less than the critical angle θ_c for that material, where $\theta_c = \sqrt{2\delta}$, $n = 1- \delta$ is the real part of the refractive index, and where the absorption index β is not too large. Detailed discussions of total external reflectance are found in the references cited, but as a rough guide, $\delta \sim \lambda^2 Z$. Thus for platinum or gold mirrors to be used at wavelengths down to 12Å, $\theta = 3°$ is a typical incidence angle. Figure 3 describes the glancing-incidence reflectance of gold mirrors as a function of photon energy. (Provided by G. Williams and M. Howells of Brookhaven). As the reflectance is quite large for $\theta < \theta_c$, the possibility of obtaining large beam deflections through multiple reflections is apparent, although many such deflections would be required, and thus might prove impractical.

Scattering of the XUV Radiation by Optical Surfaces. Scattering is particularly strong when the incident wavelength is of the same

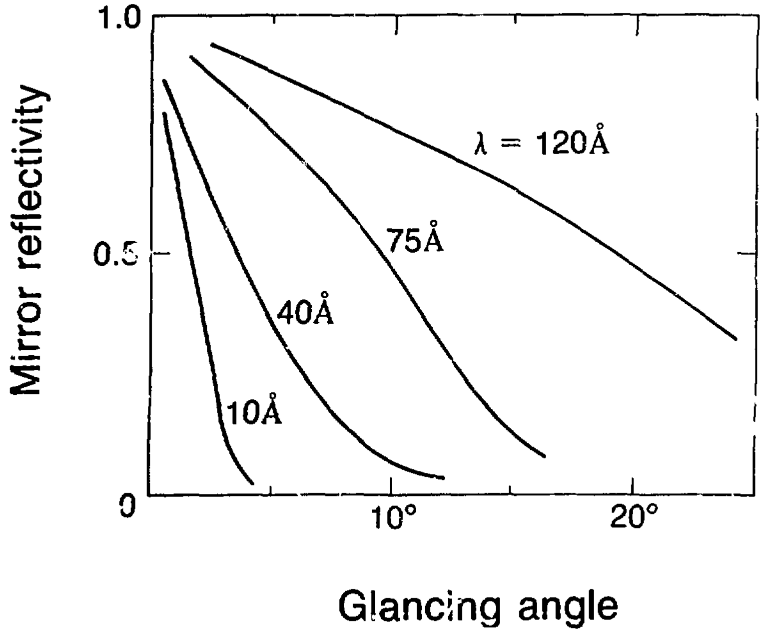

Figure 3. Grazing incidence reflection from a single gold surface, as a function of photon energy.

order as geometrical sizes encountered. This is particularly a problem in the XUV where wavelengths range from 10 to 1,000Å. All XUV optical systems are limited to some degree by scattering from micro-roughness. Although a considerable theoretical effort has dealt with this problem over many years, the situation is still unsatisfactory in the XUV spectral range. A major reason for this deficiency is the difficulty in experimentally characterizing the microroughness topography of high-quality optical surfaces, and the unavailability of intense, collimated sources, optics, etc. With increased activities in this region the issue of surface roughness should receive renewed experimental attention.

Polarization. For normally incident radiation and for radiation incident at extreme grazing incidence ($< 5°$), there is no polarization effect on the incident beam by the mirror. However, at incidence angles at or near the pseudo-Brewster angle, generally between 60° and 80°, the dependence of reflectance on the state of polarization is strong. Thus, reflectance ratios R_s/R_p of the order of 10 can be achieved in the XUV near the psuedo-Brewster angle, and glancing-incidence optics can be used as polarizers or analysers. The effectiveness of such devices is, however, limited by scattering. For shorter wavelengths, towards the soft x-ray region, this same phenonemon occurs for angles near 45°. New crossed undulator techniques, such as that of Kim[10], may make it possible to achieve arbitrarily polarized radiation in certain regions of the spectrum.

III Multilayer Technology

The recent development of atomic layering techniques has led to several significant achievements in the field of XUV optics. Of particular interest is the demonstration of normal incidence x-ray mirrors[2,3] at several hundred eV, and the understanding that with sufficient support and extension of material synthesis techniques, similar capabilities might soon extend through much of the VUV and soft X-ray regions of the spectrum. The use of multilayer coatings, well defined and stable to only a few atomic monolayers, has now been demonstrated as a means to achieve specified spectrally sensitive reflection and transmission characteristics with various substrates for photon energies from 100 eV to 20 KeV, and looks promising for new achievements in the 10 to 100 eV spectral range if apropriate efforts are made.

The application of multilayer technology has been made possible by the development of atomic monolayer synthesis techniques[11,12]. At the same time, predictive capabilities have been enhanced by improved understanding of the complex refractive index for many materials[13]. Experimental data now exists for multilayers of several material combinations in the wavelength region of 45Å to 100Å, at angles of incidence near the surface normal. Although relatively few measurements have been made, peak reflectivities generally approach 20%, and are often 65 to 90% of calculated values. Stabilities of these materials may be quite high but have not been studied in detail. Tungsten-carbon multilayers are stable at 500°C in vacuum for over 20 hours and have been utilized in high flux-short time experiments with success. Primary degradation mechanisms which must be evaluated for all candidate layer parts include layer interdiffusion, chemical reactions, layer agglomeration, delamination, formation of contamination layers, and substrate/multilayer distortion.

It is expected that progress in the next few years will result in normal incidence multilayer optics for wavelengths extending from 1000Å to 20Å. In the 40Å - 20Å, (just below the carbon K-edge) synthesis techniques must be developed which utilize other materials. For $100Å < \lambda < 1200Å$, uncertainty in the optical constant data limits present predictive capabilities, but it is generally anticipated that reflectivities of 50% will be achieved with the development of optimized material combinations. A summary of computational results due to Rosenbluth[4] is presented in Figure 2, which shows peak reflectivity, and the number of required layer pairs, for computationally optimized material combinations. These choices may be compromised by material interface incompatibilities.

Transmission multilayer structures are now under development. Substrate flatness, stability and absorption characteristics are of primary interest. Intensive work on window supported transmission substrates for scientific and user applications is necessary. It is likely that considerations similar to detection window design criteria will be applicable, although with far greater emphasis on surface quality.

In summary, although absorption will in all cases cause the spectral resolution of normal incidence XUV mirrors to be limited, it now appears that with sufficient effort and resources applied to these problems it may be possible in the coming years to synthesize multilayer optics of high spectral resolution and high peak reflectivity or transmissivity, in the difficult 50 to 1200Å spectral range.

IV Diffraction by Perfect Crystals

During the past few decades the possibility of growing perfect crystals has greatly stimulated the field of multi-KeV X-ray optics. Both experimental techniques and theoretical methods have progressed to a high degree of sophistication. A recent development of particular interest here is diffraction at normal incidence[5]. Despite the scarcity of available experimental data, it is safe to assert that in the near future normal incidence diffraction will play an important role in high angle diffraction studies for spectroscopy, interferometry and other X-ray optical techniques. More directly related to the spirit of this meeting is the possibility of constructing efficient reflectors to be used with a potential multi-KeV X-ray free electron laser. These crystals would exhibit both high reflectivity (>90%) and extreme wavelength selectivity ($\frac{\lambda}{\Delta\lambda}$ 10^5). Thus the same crystal would serve as both reflector and monochromator in an FEL of sufficient gain at extremely short wavelengths (few A).

V. Diffractive Optics

Diffractive optics are sure to play a very important role in the scientific exploitation of XUV radiation, but more likely in the area of beamline optics and applications than as primary elements in defining an FEL optical cavity. An area potentially of direct FEL interest is that of using gratings for pulse extraction. On the other hand, coherent radiation sources such as FEL's may play an important role in the fabrication of diffractive optics of high spatial periodicity.

At the present time, high resolution diffractive optics for the XUV, such as transmission gratings and Fresnel zoneplates, are primarily fabricated as free-standing gold structures[14]. Because this type of construction allows the optical properties of the material to be either that of vacuum or of a perfect absorber, diffraction efficiencies are low, not exceeding 10%. With a concerted effort phase objects could be made with efficiencies approaching 40%. Higher efficiencies might be realizable further into the future utilizing microscopic blazing techniques. Present fabrication techniques utilize both photon interference and electron beams as initial pattern-writing techniques in appropriate resists. These are then followed by elaborate replication, etching and plating techniques to form free standing microstructures of desired scale and aspect ratio. Spatial periods of less than 1000Å have been achieved by several groups, but are not generally available. Commercial availability is even more limited, not extending to periods below 3000Å.

With only absorptive transmissive gratings available, not only is efficiency low, but difficulties may arise because of high thermal loading. Reflection gratings, on the other hand, may handle power loading when cooled, but would not be appropriate for cavity pulse extraction, and will suffer from carbon contamination, etc., as do other reflective optics.

VI. Optics Degradation

In addition to the issue of producing high performance optics for XUV FELs, there is also the important issue of maintaining peak performance in a potentially hostile environment. This environment consists of not only intense radiation capable of direct degradation through such mechanisms as thermal distortion and site dislocation, but also indirect processes such as photon induced desorption and adsorption which can seriously impair optical surface or coating properties. The present state of knowledge in this field is primitive, suggesting the need for significant experimental efforts to identify damage mechanisms, effected atomic and molecular species, and appropriate counter-measures to protect available optics.

Specifically, the problem posed for successful operation of a storage ring FEL is that of maintaining high reflectance cavity mirrors, for a sufficiently long period of time, in an intense radiation environment. The problems one can expect depend strongly on the wavelength region. For instance, in the near UV where oxide multilayer coatings are used for high reflectance, the mirror degradation observed at Orsay[15] appears attributable to chemical decomposition of the materials producing photodesorbtion of oxygen. Deeper into the UV where bare metal reflectors presently provide the best technology, mirrors which are more stable can be built. The major problem here appears to be thermal control: optical elements must not be allowed to heat to the point where the metallic surface spontaneously roughens or distorts. Multilayer structures, which are likely to play an important role throughout the VUV and soft X-ray spectral regions, appear to be at least as damage resistant as single film mirrors. Nonetheless, they too are likely to be affected by thermal loading problems, and perhaps by photo-induced diffusion among layers. As for crystalline Bragg reflectors, it is known[16] that some crystals such as silicon and indium antimonide are relatively resistant to intense synchrotron radiation, while others, such as beryl and quartz, can undergo transitions to an amorphous state in a matter of hours. The FEL radiation will be spectrally far more intense, but perhaps more controllable, since it falls in a very narrow spectral window where high reflectivity is possible, and thus where absorption may be controllable by interference (Bormann) effects.

In addition to the high flux direct degradation mechanisms, optic surfaces must also be protected from surface contamination due to residual gases in the vicinity. It is well known that carbon deposition on the surface of synchrotron radiation optics reduces reflectivity just above the carbon edge ($20Å < \lambda < 44Å$). Some counter-measures were discussed at the LBL Vacuum workshop discussed earlier. It is evident that in addition to efforts to fabricate appropriate high reflectance optics for the XUV, it is equally important that experiments be undertaken to understand, quantify, and design counter-measures to optical degradation mechanisms in the potentially hostile environment of storage ring driven free electron lasers.

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