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Design of X-UV multilayers for high X-ray flux

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Résumé. — Les cavités de laser X, les diagnostics de plasmas et les optiques pour les sources synchrotrons sont quelques exemples d'applications envisagées des multicouches X-UV où les flux à réfléchir sont intenses. Cet article passe en revue les processus physiques qui diminuent les performances des miroirs, le choix des couples de matériaux les plus résistants à l'échauffement, les méthodes employant des modèles analytiques ou semi analytiques ainsi que les codes hydrodynamiques permettant de prédire les performances sous flux intenses. Des résultats expérimentaux sont présentés, analysés et comparés avec les prévisions. Par exemple des miroirs interférentiels en W/C, V/C et WC/C conservent leur réflectivité pendant environ 1 ns lorsqu'ils sont soumis à des flux de l'ordre de 150 MW cm^{-2} à l'aide de plasmas créés par laser. Suivant l'application particulière envisagée, des études expérimentales et théoriques complémentaires sont nécessaires. Les techniques développées pour de tels miroirs devraient aussi permettre d'étudier des processus importants dans les matériaux.

Abstract. — Many promising applications for multilayer X-ray optical elements — X-ray laser cavities, plasma diagnostics, synchrotron optics, etc. — subject them to intense X radiation. This paper discusses the initial attempts at understanding the physical processes that are likely to degrade the performance of multilayer X-ray mirrors, the selection of optimal pairs of materials to resist heat damage, methods — including simple analytic, semianalytic models and hydrodynamic code simulations — of estimating multilayer performance under extreme heat loading, and finally experimental data is presented, discussed, analyzed and compared with predictions. For example W/C, V/C, and WC/C multilayers subjected to X-ray fluxes on the order of 150 MW cm^{-2} using laser generated plasma maintain near peak reflectivity for at least 1 ns, but are eventually destroyed. Considerably more theoretical and experimental study at higher incident fluxes will be required before our understanding is sufficient to routinely design optimal multilayer mirrors for a particular high flux application. It also appears that the experimental techniques developed in the study of robust multilayers may be useful for the investigation of processes important in materials.

1. Introduction.

Multilayer X-ray mirrors have many applications which require that they function in environments where X-rays or other radiation is sufficiently intense to degrade performance. Examples include synchrotron primary monochromators and end-mirrors for X-ray lasers. These two examples illustrate the range of environments and performance expectations. A multilayer in the synchrotron application will attain a moderately high steady-state temperature in a hard vacuum, and it will be expected to maintain a reliably constant thickness of its layers without

warping or wrinkling. In the laser application, the multilayer is expected to normally reflect a predetermined wavelength, but only for a short period. The damage mechanisms (and therefore the design and fabrication) are different for the pulsed and the continuous applications. There are so many options for multilayer design and so many specifications unique to a specific application that determining the feasibility of a particular device is extremely difficult.

The ideal situation would be to have a computer code which would design the multilayer mirror best suited to the application and relevant wavelength. The design code would select the optimum materials

for the application and then it would give an accurate prediction of how the mirror will perform under the design conditions. Indeed, many applications call for detailed knowledge of the multilayer response.

Several groups have contributed to the theoretical and experimental understanding required to develop such a design code. They have considered damage mechanisms capable of degrading the performance of multilayers [1-5]. Thorne *et al.* [2] have studied the optimum materials for damage resistant performance for both continuous-use and pulsed applications. Methods of estimating multilayer damage as a function of time during periods of irradiation have been developed [1-7]. Some experimental damage data has been obtained in X-ray laser experiments [8-10], and in experiments specifically designed to make a quantitative measurement of the reflectivity degradation of a multilayer mirror while it was subjected to a short pulse of intense X radiation [3, 4, 6, 7, 11].

2. Damage mechanisms.

Diffraction and reflection arise from regularly spaced, smooth layers with different refractive indexes. Damage mechanisms reduce smoothness, decrease the refractive index gradient at the interface, or cause irregularities in the layer thicknesses. Multilayers are typically made by condensation from low pressure gases (e.g., by evaporation, sputtering or chemical vapor deposition). Such methods seldom produce layers which are in chemical equilibrium with respect to crystallization. They may also be out of equilibrium with respect to chemical bond formation. For example, a multilayer composed of equal numbers of tungsten and carbon atoms would likely form a single, homogeneous layer of the compound WC with increasing temperature. The entropy of mixing may drive diffusion of the two materials into one another forming a homogeneous alloy. The rate of crystallization, chemical reaction, and diffusion can usually be ignored at room temperature, but they often become a serious problem at higher temperatures.

In continuous-use applications, melting will distort the layer spacing, if not by flow under the influence of gravity, then by surface tension. In the pulsed applications, there is not enough time for liquid flow and so it appears that melting can be ignored. However, if both multilayer materials melt, diffusion may occur in a matter of picoseconds. The occurrence of diffusion depends on the solubility of the two materials. The mutual solubility can be determined with the help of the phase diagram of the two materials. Each case must be considered individually. There are situations where temperatures

greatly exceeding the melting temperatures must be reached before significant diffusion occurs.

Direct vaporization of solid materials (sublimation) can completely remove an unprotected material exposed to a vacuum over a long period of time. The rate of mass removal depends primarily on the vapor pressure of the material in question. Vapor pressure increases approximately exponentially with the Kelvin temperature, T . For example, the vapor pressure, P (in Torr) of solid chromium is given by [12]

$$\log_{10} P = 10\,750 - \frac{20\,120}{T}. \quad (1)$$

Graphs of vapor pressure vs. temperature [13] show the least volatile elements to be W, Ta, Re, and C. At 3 000 K their vapor pressures range from 10^{-4} to 10^{-2} Torr (0.013 to 0.113 Pa). Vapor pressures are additive (Dalton's Law), so the vapor pressures of all of the materials in a multilayer could tend to delaminate the structure as temperature increases. Delamination is opposed by the cohesive forces in the structure, and for pulsed applications, by the inertia of the layers. The cohesiveness of the materials mentioned above is large compared to the vapor pressure (at least below the melting point). The ultimate strength of tungsten is 1 500 MPa, and graphite is 7 MPa, but interfaces are not necessarily as cohesive as pure materials.

Rapid heating and the consequent expansion generates a shock wave which may cause spalling at multilayer surfaces. Inhomogeneous heating aggravates the problem. The refractory metals undergo a transition from brittle to ductile at some temperature depending upon the history of the test piece. The transition occurs between about 448 and 623 K for tungsten. Brittleness exacerbates spalling. A less serious consequence of rapid heating is compression and rarefaction of layers which results in detuning.

The X-rays themselves have potential for direct damage. In most applications, the primary absorption mechanism is photoionization. The resulting electrons are not expected to do significant damage, but crystal imperfections (e.g. color centers) are created over long exposure periods. Radiation damage of this type has not been shown to degrade multilayer performance in the X-ray portion of the spectrum.

3. Selection of materials.

Because the temperature is proportional to the rate of absorbed radiation, materials should be selected that minimize total multilayer absorption of all incident radiation (not just the wavelengths of interest). Also, severe strain between layers can develop if their thermal expansion coefficients are

not well matched. The residual stress resulting from fabrication or annealing could cause enhancement or relief, depending upon its direction. Low absorbance could help keep the temperature below the threshold of mechanical failure of the layers.

CONTINUOUS-USE APPLICATIONS. — In general multilayer X-ray mirrors which are required to function continuously while reflecting intense irradiation should have a large difference in the respective refractive indexes of the two layer materials, continue to operate at the highest possible temperature, have minimal mass loss through sublimation, and show minimal blurring of interfaces by diffusion.

To maximize the refractive index difference, one must know the index of every candidate material at the wavelength of interest. In the past, this has been somewhat impractical, so a high- Z element and a low- Z element were selected. (Z is the atomic number.) The most popular low- Z element has been carbon. Lithium provides an even larger difference in refractive index over much of the soft X-ray region, but it melts at a relatively low temperature and will not be considered further. For a preliminary screening of material pairs, it would be convenient to base the selection on data which are more accessible than refractive index. At a given wavelength (except near absorption edges) the refractive indexes of the elements are proportional to total electron density given by

$$\rho_e = \frac{ZN_0}{V_a} = \frac{ZN_0 A}{\rho} \quad (2)$$

where Z is atomic number, N_0 is Avogadro's number, V_a is the atomic volume, A is the gram atomic mass, and ρ is the physical density. This total electron density (not to be confused with conduction electron density) is a more reliable guide than Z and more readily available than measured or calculated refractive indexes.

The selection of optimal materials should also be based on their thermal properties. Figure 1 is a rough but useful guide to such selection [2]. It shows melting points of some elements and refractory compounds plotted against total electron density. The position of carbon is somewhat arbitrary. Carbon does not melt, but its vapor pressure is 0.1 MPa at the plotted temperature of 4 200 K. A carbon-containing multilayer may resist delamination at even higher temperatures if it is well bonded to the other material. The ultimate strength of graphite is approximately 7 MPa at room temperature.

Promising pairs of materials can be selected from figure 1 by choosing those with large differences in electron density and with melting points high enough for the intended application. It is clear that TaC/C is the best and HfC/C is second best. For these two

Fig. 1. — Melting point vs. total electron density. Carbon sublimates so that the plotted point is at the temperature where its vapor pressure reaches 0.1 MPa.

materials, melting will first occur at the interfaces and at the eutectic temperatures (3 718 and 3 453 K for crystals, lower for typical multilayers).

Vapor pressure is another important factor in the selection because of sublimation and possible delamination. The vapor pressures of the carbides are essentially the same as the constituent elements because under vacuum at high temperatures, the compounds are unstable with respect to decomposition and evaporation of one or both of the elements. Few materials have lower vapor pressures than these at elevated temperatures (tungsten is the major exception).

There is a tradeoff between melting point and maximum difference in total electron density (i.e., W has the highest density but not the highest melting point). However, there are two mitigating factors: the carbides with high melting points are interstitial compounds (carbon atoms in interstices of the metal lattice), so that the electron density is not much lower for the carbide than the metal; and the peak reflectance is not a linear function of the difference in refractive index.

Solid state diffusion is rapid at the high temperatures of interest, but it will not occur if the diffusing material is insoluble in the material across the interface. A limited amount of solubility can be tolerated, depending on how much it changes the refractive index gradient across the interface. Published phase diagrams [14, 15] indicate solubilities at various temperatures as well as the compounds which have been observed for almost all pairs of solid chemical elements. Such phase diagrams can be consulted to eliminate pairs of materials which may lose interface sharpness through diffusion. Figure 2 is a portion of the Ta-C phase diagram redrawn from reference [16]. The lower right portion indicates virtually no mutual solubility of TaC and C.

The conventional methods of fabrication usually produce layers which are unstable with respect to crystallization or chemical reaction, or both.

Fig. 2. — A portion of the Ta-C phase diagram redrawn from reference [16].

Although changes are generally slow at room temperature, they become a problem at elevated temperatures. For example, W/C multilayers have been observed to recrystallize between 923 and 1 023 K with total loss of reflectivity due to W crystal penetration of the C layers and generation of roughness [17]. This is a much lower recrystallization range than is observed for bulk W [18], but in all cases the temperatures depend strongly on impurities and the history of the specimen. At approximately 1 100 K, a chemical reaction producing WC can be expected to begin propagating from the interfaces.

Multilayers for continuous-use, high-temperature applications can be optimally designed by selecting pairs of materials which have a maximum difference in refractive index, absorb a minimal amount of the incident radiation, remain solid at the temperature of the intended application, have no tendency to recrystallize disruptively, have similar thermal expansion coefficients (or compensating residual stress), and have limited solubility in one another. The chemical elements involved should have low vapor pressures or should be overcoated with a material that does (e.g. tungsten if its absorbance is low enough at the wavelength of interest). TaC/C multilayers are an excellent choice where maximum working temperature is important and the X-ray wavelength is relatively short. However, no single pair of materials is likely to be optimum for all applications.

SINGLE-PULSE APPLICATIONS. — For certain X-ray laser and plasma diagnostic experiments, the objective is to maintain reflectance for as long as possible (tens of nanoseconds) before the heat load destroys the multilayer mirror. In such cases, the important properties are much different than for continuous-use applications. For these short periods, melting can be tolerated (if interfaces do not become optically rough), and vapor pressure is of no consequence until it causes delamination. We expect the tolerable vapor pressure to be very high because it must overcome both cohesion and inertia on a

nanosecond time scale. Diffusion and shock are problems in this time regime, but can be minimized by selecting mutually insoluble materials and materials which have low absorbance. The ultimate criterion is the maximum radiation fluence (J cm^{-2}) the multilayer mirror can stand before it fails. Without knowing the energy spectrum for the intended application, we restate the criterion as the maximum deposited energy per unit volume of multilayer. This assumes the same amount of energy is deposited in each layer pair, which is unlikely to be the case, but without knowing the characteristics of the incoming radiation, we cannot improve on this assumption. Note that comparisons based on energy per unit volume can be significantly different from those using energy per unit mass, a measure which is commonly seen in similar evaluations. It should also be noted that the thermodynamic equilibrium arguments which follow can be considered worst case situations. That is, there may not be time to achieve equilibrium (e.g. melting or vaporization), and diffusion can cause mixing up to but not beyond the equilibrium solubility limit.

There are two components to the amount of heat a material will absorb for a given increase in temperature: the heat capacity multiplied by the temperature change, and the latent heat of any phase changes which might occur. In pulsed applications, the constant volume version of each is closer to reality, but constant pressure data are more readily available and have very nearly the same values so long as vaporization is not involved. Constant pressure values (enthalpy and heat capacity at constant pressure) have been used to construct figure 3, which is useful in materials selection for pulsed applications [2]. That is, the highest peak reflect-

Fig. 3. — Absorbed energy vs. total electron density of selected materials. Squares indicate energy absorbed by solid in going from 300 K to the melting point of the pure material. Circles show the additional energy required for complete melting. The triangle shows the energy absorbed in taking solid boron from 300 K to liquid boron at its boiling point.

ances are available from pairs of materials that are widely separated along the abscissa, and good thermal properties are indicated by high placement on the ordinate.

It is not clear from experiments what the phase criteria are for multilayer stability and reflectance. If all the layers vaporize, they will mix rapidly and reflectivity will be lost. (There are a few known cases of immiscible gases, but none at high temperatures or even with materials that are solid at room temperature.) However, it may be possible for a set of entirely melted layers to reflect until forces disrupted their positions. Alternating layers of liquid and gas would also presumably reflect if smoothness and regular spacing persisted. An there is no obvious reason why stiff, solid layers separated by liquid or gas would not reflect well. The most revealing illustration is the case where one material remains a solid.

Take a TaC/C multilayer with $\gamma = 0.333$ (γ is the ratio of the high Z layer thickness to the bilayer thickness). If the densities of TaC and C are taken as 13.9 and 2.0, the overall composition of the multilayer is 29.8 atom percent Ta and 70.2 atom percent C. As the temperature of such a multilayer rises, the phase situation is represented in figure 2 by a point directly above 70.2 atom percent C and at the temperature of interest (assuming equilibrium is achieved and the layers are pure, crystalline materials). It can be seen that below 3 718 K pure TaC and pure C coexist as solids. At this eutectic temperature, a small amount of material melts. The liquid has the composition of the eutectic mixture (61 % C). This third type of layer with an intermediate total electron density will degrade, but not destroy the reflectance of the multilayer. As the temperature increases, its thickness will grow and its composition will change in accordance with the solidus line. This line is the locus of solid-liquid equilibrium points for systems with $C \geq 61\%$. It is dashed in this diagram because it is an estimate. Using tie lines and the lever rule, one can calculate the amount of TaC, C and liquid mixture at any temperature. In this example, TaC solid will remain until its melting point (4 258 K) is reached. After absorbing 7.5 KJ cm^{-3} heat of fusion, it will be completely melted between the remaining carbon layers. As the temperature rises, the state point finally reaches the dashed equilibrium line and the remaining small amount of carbon dissolves in the liquid. This will occur at higher temperatures the larger the percent carbon (the smaller γ).

Similar reasoning reveals the phases and the temperature changes for other pairs of materials. Based on this information, the heat absorbed to reach any temperature (and its corresponding phases) can be calculated by standard thermodynamic methods. However, the data needed are

often questionable at these high temperatures, and it is very unlikely that the process will remain near thermodynamic equilibrium. Solids may superheat before melting, and large gradients in concentration may exist in the liquid between solid layers. These effects help maintain reflectance, but lateral gradients within layers would degrade it. Lateral gradients could arise from random nucleation of melting.

Lacking information on these kinetic effects, the equilibrium data appear to be the best guide to materials selection. Note that boron absorbs more energy per unit volume than any of the other elements, but its vapor pressure is nearly 4 orders of magnitude higher than carbon at a given temperature. It may be an interesting candidate for the low-density material in certain applications. In spite of this, TaC/C appears to be the best general purpose multilayer for high temperatures.

4. Estimate of multilayer performance.

Two approaches have been taken in predicting the failure of multilayers under conditions of extreme radiation. We can make estimates of multilayer temperature rise by using energy balance relationships based on thermodynamic arguments. The damage threshold can then be estimated by knowledge of the characteristics of the pair of materials — chemical properties, melting point, solubility, etc. The key here is to choose the correct model and make assumptions that include only the key attributes of the system. The second approach is to use hydrodynamic codes, which contain the relevant physics, to simulate the reflectivity of the multilayer mirror as it is irradiated with X-rays. LASNEX [19] and CHARTD [20] are two such codes that can be used to simulate multilayer behavior during pulses of intense radiation. LASNEX is a powerful code designed for extremely high temperature applications. More *et al.* [1] have modified LASNEX to produce a code called SPRINT (Short Pulse Radiation INteraction code) which gives accurate low temperature results. CHARTD is a one-dimensional radiation hydrodynamic code which handles the interaction of radiation with matter at low temperatures i.e. in solids, as well as in high temperature plasmas.

ANALYTIC ESTIMATES OF DAMAGE. — The amount of heat a material will absorb for a given increase in temperature is given by the heat capacity multiplied by the temperature change and the latent heat of any phase changes which might occur. We can use this thermodynamic relationship to derive an energy balance equation to obtain an estimate of the survival time of multilayers as a function of the cumulative energy absorbed and the rate at which the energy is absorbed. The following assumptions

are the basis of a *best case* (longest survival time) prediction.

Assumptions :

- (1) the multilayer is large so that edge effects can be neglected ;
- (2) no diffusion of materials occurs between layers, even when both are liquids ;
- (3) there is no crystal growth or similar phenomenon that disrupts layer interfaces ;
- (4) all materials are assumed to be perfect blackbodies and to follow the Stefan-Boltzmann law. The surroundings are at low temperature ;
- (5) the multilayer is optically thin so radiation is deposited homogeneously or else distributed quickly by conduction into low-absorption layers ;
- (6) the thermal and mechanical properties of the multilayer materials are closely approximated by bulk properties, and only classical phenomena occur (melting, vaporization, etc. as opposed to tunneling, nonlinear effects, etc.) ;
- (7) fluorescence is neglected, so what we call absorbed energy is total energy deposited minus that which is fluoresced quickly.

Figure 4 is a log-log plot of cumulative absorbed energy vs. rate of energy deposition. It does not show survival time directly, but it should be noted

Fig. 4. — Absorbed energy/cm² vs. absorbed power/cm². Dashed lines indicate multilayer lifetime. Upper solid line represents energy required to reach eutectic temperature for a TaC/C multilayer radiating from both sides. The central horizontal line is the accumulated energy required to reach TaC/C eutectic starting at 298 K. Points 1 and 2 are observed incident energies to damage W/C multilayers (Hockaday *et al.* Refs. [3, 4]). (Arrows indicate absorbed energies are smaller.) Point 3 is observed survival of W/C, WC/C and V/C multilayers (Gray *et al.* Refs. [6, 7]). (Arrows indicates survival to higher energy depositions.) Points 4 and 5 are calculated energies to melt Si and both Mo and Si in Mo/Si multilayers (More *et al.* Ref. [1]).

that the ordinate divided by the abscissa has units of time. Therefore, time can be represented by the dashed, diagonal lines in the figure. The figure is based on the following estimate.

Assume an unbacked, free-standing multilayer and note that its temperature increases if the absorbed power, W , exceeds the radiated power, $\sigma \rho T^4$. The latter is an expression of the Stefan-Boltzmann equation with $\sigma = 5.672 \times 10^{-12} \text{ W cm}^{-2} \text{ K}^{-4}$. This form assumes the surroundings are at 0 K. We take the surroundings to be at 298 K and write the radiated power as $\sigma T^4 - 0.08946 \text{ W cm}^{-2}$.

Our goal is to calculate the maximum time, t_m , required to reach a destructive temperature, T_d . We will do this for the general case, but discuss a particular multilayer concurrently. A TaC/C multilayer with 35 Å TaC and 65 Å C in each layer pair, and a total of 50 layer pairs will serve as the example. Such a multilayer has a eutectic at 3 718 K (see Fig. 2 and Ref. [16]). This will be the destructive temperature if the times of interest exceed approximately 10^{-3} s . (The melting at interfaces will allow layer motion under the force of gravity and vapor pressure.) If we take the density of TaC to be 13.9 g cm^{-3} and that of C to be 2.0 g cm^{-3} , the multilayer is seen to be 68.2 atom-percent C. The corresponding state point reaches the liquidus line (dashed) at approximately 4 300 K. At this temperature, the multilayer (if it behaves classically) will be completely melted and we would expect diffusion to destroy interface reflectivity in a matter of picoseconds. But the upper temperature may approach 4 300 K for periods of irradiation short compared to the mechanical motion of liquid-solid alternating layers. In general, the upper temperature is determined by selecting a mechanism of destruction for the multilayer in question, and using a phase diagram or some other means to find the temperature where reflectivity is expected to drop dramatically. In the last example, the energy balance should include the energy involved in any phase changes that occur (ΔH for melting of the TaC), but we will ignore this until later.

If the multilayer absorbs energy, but has no mechanism such as radiation or conduction for losing energy, it requires only 0.0441 J cm^{-2} to raise the temperature to the eutectic point. The horizontal line near the center of figure 4 corresponds to this energy. Additional energy absorption would melt some of the TaC and begin to destroy the mechanical integrity of the multilayer. The following calculations are based on blackbody radiation occurring from the front and back surfaces of the multilayer.

Assuming a constant pressure environment of 0.1 MPa or lower, and taking the expansion of the multilayer to be small, we note that enthalpy will be nearly identical to internal energy ; and the heat

capacity at constant pressure, C_p , will be nearly identical to that at constant volume C_v . We choose to work with enthalpy and C_p which are related by :

$$dH = C_p \, dT . \tag{3}$$

The energy balance mentioned previously can now be written :

$$W + 0.08946 = 2 \, \sigma T^4 = dH/dt = C_p \, dT/dt \tag{4}$$

where t refers to time. The factor of 2 is included because the multilayer is allowed to radiate from both sides. We note that if radiated energy is equal to, or exceeds absorbed energy, the equation does not hold and the multilayer lasts indefinitely. This corresponds to the vertical line at the left of figure 4.

To obtain the time during which the multilayer is still reflective, we must integrate over the temperature range 298 K (assumed starting temperature) to T_d , the destruction temperature

$$t_m = \int_0^{t_m} dt = \int_{298}^{T_d} \frac{C_p \, dT}{W + 0.08946 - 2 \, \sigma T^4} \tag{5}$$

C_p is normally expressed as a polynomial in T . The same expression can also be obtained by differentiating a polynomial expression for H . In either case, the following form generally fits heat capacity data to better than 2 % :

$$C_p = a + bT + cT^2 + dT^{-2} . \tag{6}$$

The last factor is important only at temperatures far lower than those of interest. It will be dropped. Values for a , b and c for carbon and tantalum carbide are found in table I.

Table I. — Heat capacity of tantalum carbide and graphite at high temperatures in J/mole-deg.

Material	a	b	c
TaC	57.715	-9.200×10^{-3}	4.627×10^6
C	11.186	10.952×10^{-3}	0

The denominator of equation (5) can be factored by setting $W + 0.08946 = w^4$ and $2 \, \sigma = s^4$. When (5) is combined with (6), we obtain a form that can be integrated by partial fractions

$$t_m = \int_{298}^{T_d} \frac{(a + bT + cT^2) \, dT}{(w - sT)(w + sT)(w^2 + s^2 T^2)} . \tag{7}$$

The result is :

$$t_m = -\frac{A}{s} [\ln (w - sT_d) - \ln (w - 298 \, s)] +$$

$$\begin{aligned} & + \frac{B}{s} [\ln (w + sT_d) - \ln (w + 298 \, s)] \\ & + \frac{E}{sw} \left[\tan^{-1} \left(\frac{sT_d}{w} \right) - \tan^{-1} \left(\frac{298 \, s}{w} \right) \right] \\ & + \frac{D}{2s^2} \left[\ln \left(\frac{w^2}{s^2} + T_d^2 \right) - \ln \left(\frac{w^2}{s^2} + 298^2 \right) \right] \end{aligned} \tag{8}$$

where

$$\begin{aligned} A &= \frac{a}{4 \, w^3} + \frac{b}{4 \, sw^2} + \frac{c}{4 \, s^2 w} , \\ B &= \frac{a}{4 \, w^3} - \frac{b}{4 \, sw^2} + \frac{c}{4 \, s^2 w} \\ \text{and} \quad E &= \frac{a}{2 \, w^2} - \frac{c}{2 \, s^2} . \end{aligned}$$

In this equation, a , b and c refer to the heat capacity of 1 cm² of the multilayer, whereas the values in equation (4) are for one mole of the material in question. The TaC/C multilayer contains 3.78 μ moles of TaC and 5.41 μ moles of C per cm². Multiplying the appropriate values in table I by these factors and adding, we obtain the heat capacity of 1 cm² of the TaC/C multilayer with coefficients

$$\begin{aligned} a &= 2.82 \times 10^{-4} \quad b = 2.45 \times 10^{-8} \\ c &= 1.75 \times 10^{-11} . \end{aligned} \tag{9}$$

Inserting these values in equation (8) and selecting various values for the absorbed power, points for figure 4 can be calculated.

This approach is not expected to be reliable if the multilayer is maintained very long under conditions where the power being absorbed is nearly the same as the power being radiated. The lifetime of the multilayer is then calculated from the small difference between two large numbers, which amplifies uncertainties in the data used for the calculation. This occurs in the lower left portion of figure 4. Uncertainty also exists where lifetimes are very short. Several of the assumptions are likely to be invalid at short times. We suggest that on the order of picoseconds, nuclear motion is small compared to d-spacing and the multilayer retains its reflectivity. However, if large amounts of energy are rapidly absorbed, enough photo- and thermal ionization can take place to make the electron density too homogeneous to support significant reflectivity. For the TaC/C multilayer, we estimate this to occur when the carbon is completely ionized and the Ta is argon-like.

Estimates of damage energy can vary by orders of magnitude depending on the choice of assumptions. If no energy loss is assumed the TaC multilayer would be destroyed at 0.0441 J cm⁻¹, but with the assumption of blackbody radiation, damage will not occur until the absorbed energy approaches a few J cm⁻². Figure 4 includes points from estimates and

experiments found in the literature for comparison. They will be discussed in more detail later.

MULTILAYER DAMAGE SIMULATIONS. — More *et al.* [1] have used the code SPRINT based on LASNEX to estimate the damage threshold for a typical Mo/Si multilayer designed to operate as an X-ray laser cavity mirror. The laser operated at $h\nu = 60$ eV, a peak power of 8×10^8 W cm⁻², and a pulse width of 250 ps. The code predicted that melting would occur at 0.1 J cm⁻² of absorbed energy. This agreed with thermodynamic estimates. In the liquid state moly and silicon will diffuse in a few picoseconds and the reflectivity will go to zero.

The code ROMULS [7] will predict the reflectivity of a multilayer *vs.* angle or wavelength *vs.* time during periods of intense radiation. ROMULS is actually a driver program that handles the user interface, sets up the problem for the each of three codes — XRAYCARP [7], CHARTD and LSMLAYER [21] — that do the calculations. ROMULS also does all of the file management. XRAYCARP calculates the energy absorbed by the multilayer. Once the deposition profile for each of the material layers has been computed, the damage done to the structure by the absorbed energy is evaluated by CHARTD. CHARTD calculates the thermodynamic state of the material and then determines the positions and velocities of the interfaces by using conservation of energy and momentum. CHARTD will not account for effects such as diffusion, crystallization of amorphous layers driven by the heating of the multilayer, or roughness of the

interfaces resulting from damage. These effects are slow, however, and for pulsed applications there is not sufficient time for them to occur. The exception is diffusion. Diffusion can occur rapidly after melting has occurred. These cases must be treated individually based on the phase diagram of the two materials. LSM-LAYER calculates the reflectivity of the multilayer using the interface positions determined by CHARTD.

The ROMULS output is in the form of two-dimensional plots of reflectivity *vs.* angle or wavelength at user specified time steps or three-dimensional plot of reflectivity *vs.* angle or wavelength *vs.* time. Graphics support for the code is provided by ANALYZE [22].

The ROMULS code has been used to simulate the performance of V/C, W/C, WC/C, and TaC/C multilayers. The results of these simulations are shown in figures 5-8. The geometry of the multilayers is similar, with *d*-spacings near 22 Å, $\gamma = 0.4$ and 99 layer pairs each. Each simulation was at a wavelength of 12.7 Å, corresponding to the Cu L _{α} doublet used in the companion experiments. Figure 5 shows the predicted performance of the V/C multilayer for a flux of 63 mJ cm⁻². The *d*-spacing shifts somewhat and there is a rapid loss of reflectivity. The heat resistance of the V/C multilayer is especially poor in comparison to a W/C multilayer at a flux of 135 mJ cm² as shown in figure 6. This multilayer maintains a reflectivity of over 10 % for more than 1 ns. The shift in peak reflectance angle corresponds to a shift in *d*-spacing of approximately 2 Å. Simulations for multilayers which would be

ROMULS REFLECTIVITY DATA

VC 48 D = 24.5 12.75 Å 063 J/cm²

Fig. 5. — Simulated response of a V/C multilayer to a fluence of 63 mJ cm⁻².

Fig. 6. — Simulated response of a W/C multilayer to a fluence of 135 mJ cm⁻².

Fig. 7. — Simulated response of a WC/C multilayer to a fluence of 160 mJ cm⁻².

expected to be much more robust, WC/C and TaC/C, are shown in figures 7 and 8 respectively. Each of these simulations used a flux of 160 mJ².

The WC/C multilayer maintains greater than 10 % reflectivity for approximately 0.75 ns while the TaC/C multilayer lasts close to 1 ns.

ROMULS REFLECTIVITY DATA
TACCO D = 21.6 12.75 A 160 J/cm²

Fig. 8. — Simulated response of a TaC/C multilayer to a fluence of 160 mJ cm⁻².

5. Damage experiments.

Perhaps the most obvious application of robust multilayers mirrors is in X-ray laser cavities. Some X-ray laser experiments have been done with multilayer mirrors configured for a double pass through the gain medium [21, 22, 23]. The results reported so far seem to indicate that a degradation of the mirror reflectivity has not occurred during the short period of interaction involved.

Kohler *et al.* [11] used a pulsed Nd-glass laser to generate a point source of copper L X-rays (0.980 keV) to study damage degradation in a WC/C multilayer. Their results were consistent with damage occurring at an energy deposition of 0.46 J cm⁻².

Hockaday *et al.* [3, 4] did experimental damage experiments on two tungsten-carbon multilayers using X-rays from the Z-pinch plasmas produced by the PROTO II at Sandia National Laboratory in

Fig. 9. — Intensity vs. time line-out along the 930 eV line for a V/C multilayer reference shot.

Fig. 10. — Intensity vs. time line-out along the 930 eV line for a V/C multilayer damage shot with an incident fluence of 150 mJ cm⁻².

Albuquerque. Reflectivity degradation was observed at absorbed energies of 5.27 J cm^{-2} and 2.0 J cm^{-2} .

Gray *et al.* [6, 7] used two synchronous laser generated plasmas as X-ray sources. They were

produced by the Phoenix (Janus) laser facility at Lawrence Livermore National Laboratory. Each beam will provide a maximum of 25 J of 532 nm light in a 1 ns pulse. The timing of the two beams relative

MULTILAYER DAMAGE EXPERIMENTS

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Fig. 11. — Digitized film exposure of W/C multilayer reference shot.

MULTILAYER DAMAGE EXPERIMENTS

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Fig. 12. — Digitized film exposure of W/C multilayer damage shot with an incident fluence of 150 mJ cm^{-2} .

to each other is controllable to within 200 ps. The beams were focused onto bulk Cu targets producing intense L line radiation (0.980 keV). One beam provided the source of damaging X-rays with a flux of up to 250 mJ cm^{-2} . The other plasma generated X-rays which were incident on the multilayer at the Bragg angle and served as a probe source. The time profile of the probe radiation was recorded on a streak camera. Data obtained from streak camera images for shots using a V/C multilayer are found in figures 9 and 10. Figures 11 through 13 correspond to shots taken with a W/C multilayer while figures 14 and 15 display data from WC/C shots.

These experiments indicate that multilayers such as W/C, V/C, and WC/C will maintain reflectivity for at least 1 ns for a fluence of approximately 150 mJ cm^{-2} (corresponding flux of 200 MW cm^{-2}). The reflectivity of the multilayers is destroyed at some point after the first nanosecond. These measurements are consistent with the predictions of ROMLUS code.

Fig. 13. — Intensity vs. time line-out along the 930 eV line for a W/C multilayer damage shot with an incident fluence of 150 mJ cm^{-2} .

MULTILAYER DAMAGE EXPERIMENTS FILM 89

Fig. 14. — Digitized film exposure of WC/C multilayer reference shot.

6. Discussion.

It is fair to conclude from the work presently reported, that multilayers will survive in hostile environments long enough to be of value in many key applications. However, the picture is not yet

clear enough to design instrumentation based on multilayers with the confidence that the device is even close to being optimized. In addition the performance cannot be predicted with accuracy.

It is evident from careful examination of the theoretical and experimental points displayed in

MULTILAYER DAMAGE EXPERIMENTS
FILM 90

Fig. 15. — Digitized film exposure of WC/C multilayer damage shot with an incident fluence of 150 mJ cm^{-2} .

figure 4 that considerable work is required before we have a quantitative understanding of multilayer damage. In particular, there is a need for more experimental data to guide the theory. The first need is for clean, well calibrated experiments. This means a well characterized damage source and a clean probe signal. Then many more measurements are required varying the key parameters :

- energy input. Specifically we need measurements at higher powers ;
- multilayer materials. Measurements on a variety of materials are required to test the ideas regarding the most robust combinations and the improvement in durability that can be obtained by their proper choice ;
- wavelength. Different wavelength measurements are necessary to be sure that wavelength dependent predictions are valid.

In addition much information on melting, diffusion, crystallization, chemical reactions, and damage sources could be determined by careful analysis of the damaged multilayer with high resolution analytical instruments such as TEM's. This approach is currently being pursued using the remains of damaged multilayers reported above.

7. Future directions.

Some of us see a bright future for multilayer X-ray optics and X-ray imaging in general. A crucial element in the realization of that promise is the development of tools that can be used by a nonexperts to design multilayer optics for his own applications. One such tool could be a design code that is based on an understanding of the physics of multilayers. A key segment of such a code would be the physics of multilayer interaction with intense X radiation.

If the suggested experimental program can be completed, the level of experimental knowledge should be sufficient to guide the modification of existing codes to give accurate predictions of multilayer reflectivity under severe conditions. This result would make a general design code that much closer.

It also seems worthwhile to note that the experimental techniques being developed allow the measurement of many important material processes. Multilayer reflectivity can be observed to be quantitatively related to surface roughening, melting, diffusion, rate of crystallization, chemical reactions, etc.

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