Need for Abrupt Transitions in Index of Refraction in the Soft X-ray Spectral Region

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ABSTRACT

Gradual transitions in index of refraction from layer to layer, as in rugate coatings, can be advantageous. When absorption or the k-value is negligible, it can be shown that the first order reflection band is not significantly changed from when the transition between layers is "square" even to the case where it is sinusoidal. In most visible spectrum cases, the difference between the indices of the layers is almost exclusively in the real part of the refractive index, and it is that difference which causes Fresnel reflection at the interfaces. In the EUV/Soft X-ray region, the real parts of the indices are near unity, and therefore more layer pairs are required to achieve high reflection because of the low reflection at each interface. In these cases where one of the materials has a relatively high k-value, it can be shown that slow transitions in index from layer to layer are detrimental to the reflection desired. Even when interface roughness and interlayer diffusion are ignored per se, it can be shown that the layers with the highest k-values need to be as thin as possible to reduce absorption losses. This leads to sharp transitions between layers because the most absorbing material components are compressed into the smallest thicknesses practical.

INTRODUCTION

Having not worked in the EUV/Soft X-ray region before, it was surprising to learn of the real concerns expressed for obtaining the sharpest practical demarcation between layers. Barbee et al. [1] reported on Mo-Si multilayer mirrors for the EUV, and Windt [2] later reported improved data on the optical constants of these and other materials. Stearns et al. [3] compared multilayers of Mo-Si, Ru-Si, and Rh-Si where the later two pairs had extensive intermixing of layers. Although Figure 7 of the paper by Yamamoto and Namioka[4] shows that Ru and Rh have more favorable indices of refraction than Mo, the Stearns et al. [3] report shows why Mo-Si is the favored combination for practical physical reasons. Yulin et al. [5] demonstrate that there tends to be diffusion or intermixing even between layers of Si and Mo, and this causes the boundaries to be inhomogeneous in index of refraction (both n and k). With dielectric materials in the visible and near regions, this might be considered an asset rather than a liability. Some layer structures have been purposely "blended" to gain structural integrity and avoid interface problems of adhesion and interface contamination. From an optical viewpoint, rugate structures have been popular because of their potential to reduce or eliminate higher order reflection bands. The whole field of rugate coatings is built upon these inhomogeneous layer transitions. We will first discuss this aspect of the benefits of the inhomogeneity. The differences found in the EUV/Soft X-ray region and the need for homogeneity and sharp index transitions will then be discussed.

CONVENTIONAL AND RUGATE THIN FILMS

A conventional "quarter wave optical thickness" (QWOT) reflector stack of dielectric materials where H is 2.35 and L is 1.46 and the k-values equal 0 might have a design of: (.5L 1H .5L)6. If we were to imagine that such materials were available for the 12.4 nm spectral region, we could produce the "Fantasy X-Ray Mirror" shown on a wavelength scale in Figure 1. This is a good reflector at 12.4 nm with only 13 layers. Figure 1 also shows that there would be additional reflection bands at all of the odd harmonics of 12.4 nm or 3, 5, 7, etc., times the fundamental frequency, or at wavelengths of 4.133, 2.48, 1.77, 1.38, 1.13 nm, etc.

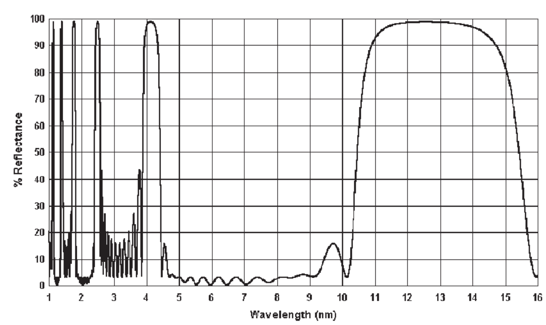

Figure 1: A "Fantasy X-Ray Mirror" for 12.4 nm with only 13 layers.

Figure 2 shows this same result plotted on a wavenumber scale (waves per centimeter), which is linear in frequency and shows a more symmetrical result. These are representative of homogeneous layers where the transitions in index from one material to the next are sharp or like square waves in an index versus thickness profile as seen in Figure 3. If the transitions were sinusoidal from one index to the next, as simulated in Figure 4, we would obtain a somewhat different result.

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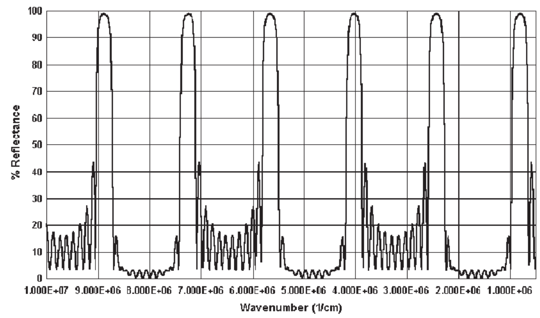

Figure 2: Same “Fantasy X-Ray Mirror” as Figure 1 shown on a wavenumber scale.

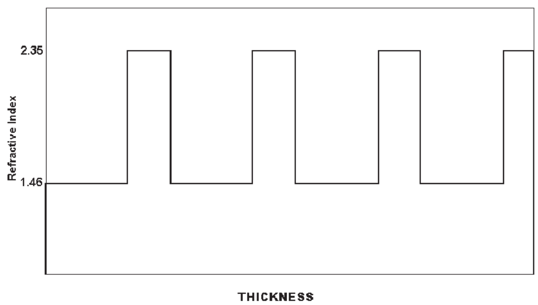

Figure 3: Index versus thickness profile of a typical homogeneous layer stack.

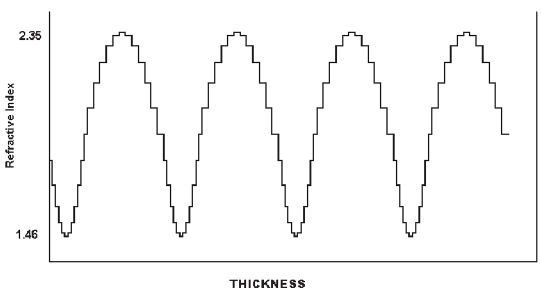

Figure 4: Index versus thickness of a rugate structure.

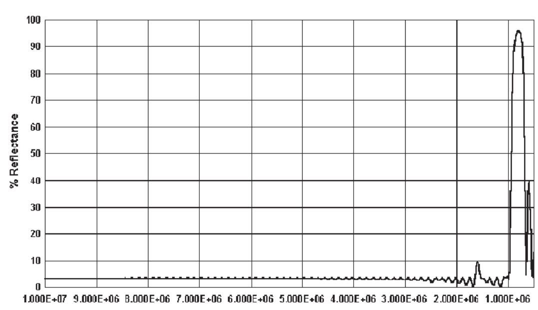

Figure 5: Reflectance of a rugate, same period as Figure 2.

Figure 5 shows that such a “rugate” index profile produces essentially the same fundamental reflection at the wavelength where the peak to valley spacing in the index values are one QWOT apart. However, the higher harmonics of this are not present (except for a small artifact due to imperfections in the simulation near the second harmonic which we will ignore).

Figure 6 shows a plot of the reflectance amplitude and its phase as a function of film stack thickness as described by Apfel [6] for the conventional homogeneous QWOT stack.

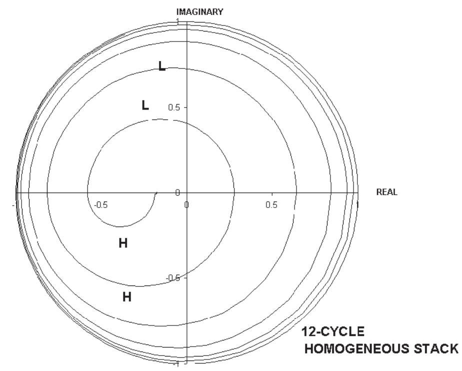

Figure 6: Reflectance amplitude diagram at the design wavelength of the homogeneous coating as in Figures 1 and 2.

Figure 7 shows this same type of “circle diagram” for the rugate design; the differences are subtle, but this will be a useful visualization tool in the sections below. Another small secondary effect of the rugate structure is that a few more index cycles are needed to achieve the same peak reflectance as the homogeneous structure. This can be observed in Figures 6 and 7 where the reflectance vector from the origin does not push toward the outer limiting circle as rapidly with each period or cycle for the rugate as with the homogeneous structure. Another view of this behavior is shown in Figure 8, where the reflectance as a function of increasing film thickness is plotted for both the homogeneous and rugate designs of the previous figures.

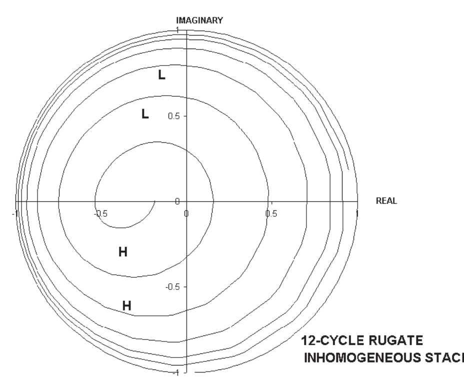

Figure 7: Reflectance amplitude diagram of the rugate coating as in Figure 5.

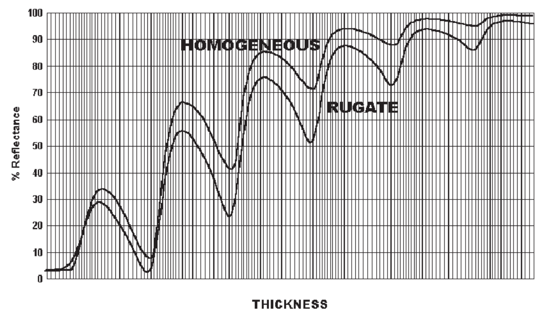

Figure 8: Reflectance versus thickness for the homogeneous and rugate designs of Figures 2 and 5.

If this same rugate behavior were applicable to the EUV/Soft X-ray region, one could produce a good reflector for the fundamental wavelength without concern for sharp transitions in index of refraction at the interfaces. However, as we will show in the next section, this is not the case with the real materials that are available in the EUV/Soft X-ray spectral region.

THE EUV/SOFT X-RAY SITUATION

Yamamoto and Namioka [4] have provided an extensive discussion of the design of multilayer reflector coatings for the soft-x-ray region at 12.4 nm. They show the n and k values for various materials (B, Be, Br, C, Ca, Ce, Nb, Mo, P, Pr, Rb, Rh, Ru, Sr, S, Sc, Si, Y, and Zr) and give selection criteria for such coating materials. These criteria are that the materials for a 2-material coating should: 1) have the smallest possible k, and 2) have the greatest difference in n-values. Si and Mo seem to represent a practical choice with n, k values of 1.0319, 0.0015 and 0.9324, 0.010 respectively. Because the differences in the indices are small compared to those in the visible region, many more layers are required to produce a mirror with high reflection. Because the k-values are relatively high, the absorption in the structure is much more of an issue than with most visible dielectric mirrors. Carniglia and Apfel [7] provided an extensive discussion of the limitations of peak reflection in the presence of some absorption in the coating materials and how to minimize these effects.

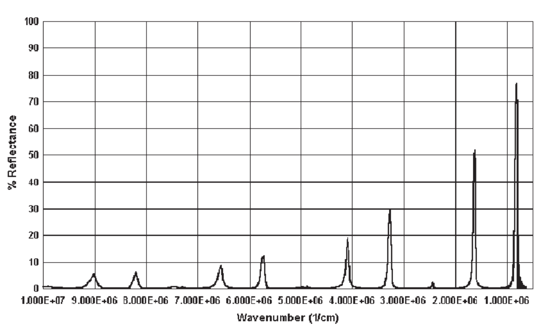

Figure 9: Spectral reflectance of a Mo and Si design for 12.4 nm in the same format as Figure 2.

Figure 9 (for comparison with Figure 2) shows the reflectance spectrum of a design for 12.4 nm using homogeneous Mo and Si layers. Figure 10 shows the spectral comparison of the "Fantasy" design of Figure 1 with the more realistic Mo/Si design of Figure 9. Figure 11 shows the reflectance amplitude circle diagram of the Mo/Si film stack designed by the Yamamoto and Namioka[4] method. The dots indicate the transitions from one homogeneous layer to the next.

The k-value equal to 0.010 for Mo illustrates the crux of the problem that makes the soft-x-ray region situation different from that of the visible region. This is actually one of the lower k materials at 12.4 nm, but it is a large value by the standards of dielectric coatings in the visible region. Because of the low n-values, a substrate of any of these materials would appear to be (almost) a black mirror in reflection (low Fresnel reflection, <0.3%R), if we could see at a wavelength of 12.4 nm. They would, of course, transmit the radiation variously as a function of their k-values and thickness. Thin films of these materials absorb radiation and reduce the reflections coming from underlying layers.

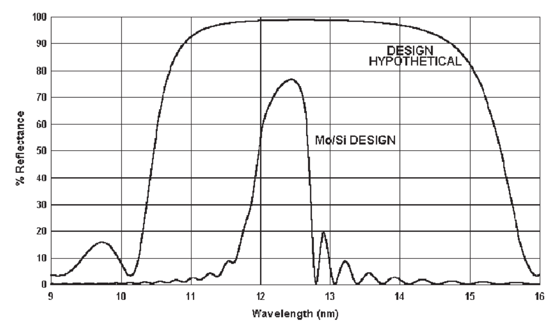

Figure 10: Spectral reflectance of a Mo and Si design for 12.4 nm compared to "Fantasy" design of Figure 1.

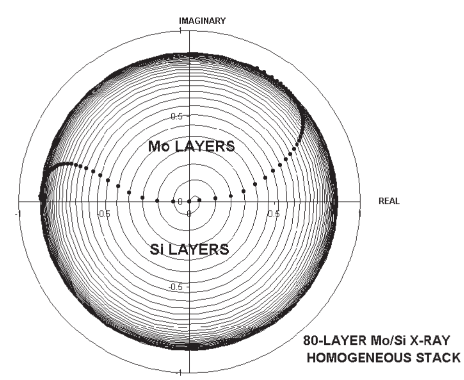

Figure 11: Reflectance amplitude plot of a Mo and Si design for 12.4 nm as in Figure 9.

Macleod [8] shows that the energy absorbed (I_{abs}) in a film is proportional to:

$$I_{abs} \propto n\, k\, \epsilon^2 \Delta t. \qquad (1)$$

Here ϵ is the electric field at that point in the film and Δt is the incremental thickness to which these values of n, k, and ϵ are being applied. Because the n-values here are near unity, the primary effects are caused by the product $k\epsilon^2$. Since k is not under the control of the designer, once the materials are specified, the primary design effort is to minimize absorption (and thereby maximize reflection) by making the ϵ-field as low as practical where the k-values are high.

A computer automatic thin film optimization program such as *FilmStar* [9] deals with these factors indirectly by maximizing the reflection as the thickness of the individual homogeneous index layers are adjusted. Inhomogeneous layers have been simulated by a series of thinner homogeneous layers of various indices as illustrated in Figure 12. When the thickness of the layers of such a structure is optimized for the X-ray mirror case, the index profile of Figure 3 is the result. Here, the lowest absorption layers become as thick as possible, the intermediate k-value layers become vanishingly thin, and the highest absorption layer is only as thick as necessary to maximize the reflectance at the design wavelength. The fact that the higher k layers (Mo) are proportionately thinner can be seen also in the reflectance amplitude diagram of Figure 11. The automatic design program solves this problem empirically without specific reference to Eqn. 1, but only by the fact that those factors are intrinsically accounted for by its analysis of the reflection of a coating design. By this result, we can say that these X-ray mirror designs "want" to have sharp transitions between the two layer materials with low and higher k-values. This implies that the rugate transitions cannot produce as high a reflection as the sharp transitions.

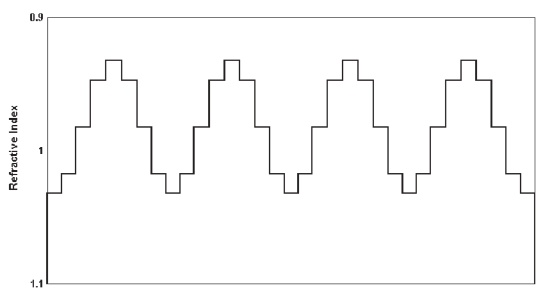

Figure 12: Index profile of rugate used for start.

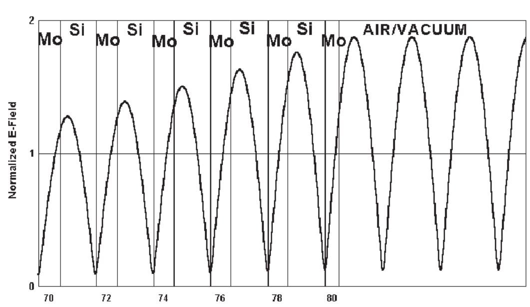

Figure 13: Electric field versus thickness in last few layers of coating from Figures 9-11.

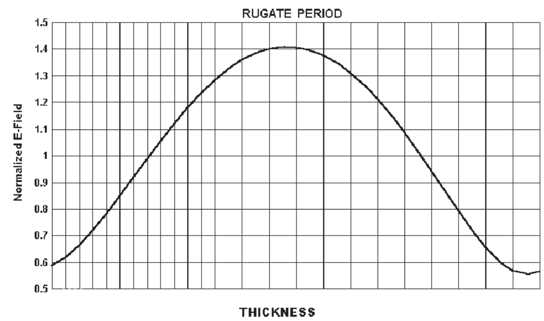

Figure 14: Electric field versus thickness for a rugate period.

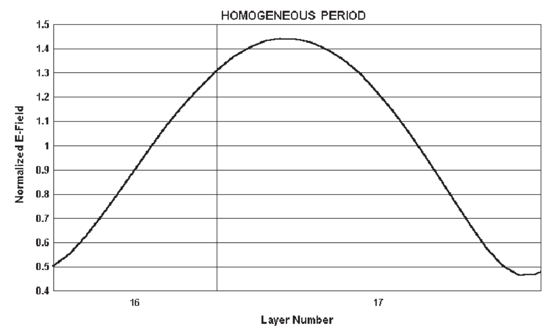

Figure 15: Electric field versus thickness for a homogeneous layer period (layer pair).

The relative electric field in the last few layers of the coating illustrated in Figures 9-11 is shown in Figure 13. The greater absorption of the rugate versus the homogeneous layers can be further illustrated by integrating Eqn. 1 over a period (or cycle) of the rugate film and compare that to a period of the homogeneous layer film. Figure 4 shows the index profile of 24 index steps used for this integration (summation). The electric field versus film thickness is calculated for this rugate period and shown in Figure 14. The same is shown for the homogeneous layer period in Figure 15.

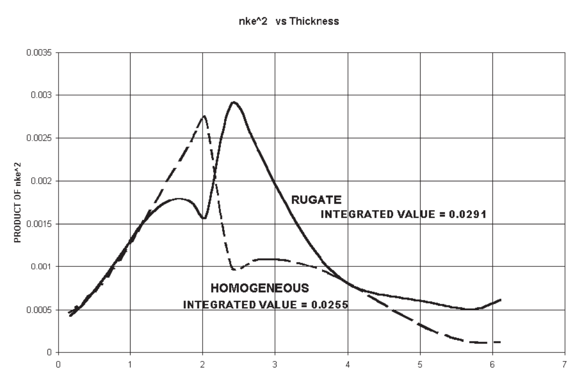

Figure 16: Product of absorption factors versus thickness for rugate and homogeneous periods.

Figure 16 shows the product of nkϵ^2 versus thickness for these two cases. It can be seen that the area under the rugate curve is greater than that of the homogeneous period, and therefore, there would be more absorption and less reflection in the rugate case. The relative integrated values are 0.0291 for the rugate and 0.0255 for the homogeneous design.

Figure 17 shows the limiting reflectance of rugate and homogeneous layer designs, and it illustrates the problem caused by a lack of sharp interfaces between layers in this spectral region. It is not really a case of the Fresnel reflection being diminished by the lack of sharpness, but that the distributed absorption's interaction with the electric field is less favorable in the rugate (non-sharp) case.

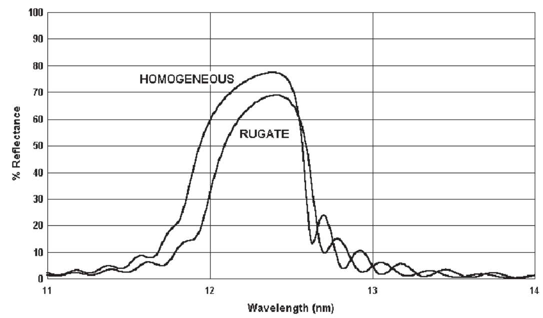

Figure 17: Limiting spectral reflectance of rugate and homogeneous designs with Mo and Si at 12.4 nm.

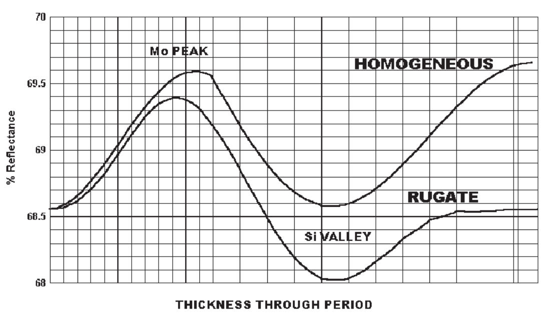

Figure 18: Reflectance versus thickness for a rugate and homogeneous period at the reflectance limit for the rugate design.

An additional view of this is provided in Figure 18. These illustrate the reflectance versus thickness for a period in the film where the rugate design has lost its ability to further increase the reflectance. It can be seen that the reflectance at the end of the period has returned to the same value as the starting reflectance. The fact that the rugate reflectance passes through a peak in the first portion of the cycle points to the fact that the last layer of the coating should be terminated at this peak for maximum reflectance. The homogeneous design first peaks near this same thickness at the end of the Mo layer. The reflectance of the Si layer then goes through a minimum and on to a peak which is higher than where the homogeneous period started. Therefore, the reflectance of this homogeneous design can benefit from additional periods before it too reaches a point-of-diminishing-returns as the rugate design did earlier, but at a higher reflectance in this case.

CONCLUSIONS

We have shown that smooth or sinusoidal transitions between high and low index regions in a period of a multi-period thin film with no absorption can have various advantages and insignificant disadvantages. However, when significant absorption must be dealt with, as in the EUV/Soft X-ray region, we have shown why sharper transitions in index of refraction, like square waves, are beneficial. The maximum peak reflectance of a design is limited to a lower value, if the transitions are not essentially "sharp"; and more periods (layer pairs) are needed to achieve a maximum reflectance, which is less than the limit when the layers are homogeneous and have sharp transitions.

The concept that the sharpness of the interface materially effects the Fresnel reflection at the interface is potentially misleading. It is the integrated absorption which includes the product of n, k, and the electric field squared that causes the difference between gradual and sharp transitions.

ACKNOWLEDGEMENTS

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