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Title: Reflectance enhancement with sub-quarterwave multilayers of highly absorbing materials

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Abstract

A new theory of multilayers with enhanced normal reflectance has been developed based on the superposition of a few layers of various different radiation absorbing materials. Every layer in the multilayer had a sub-quarterwave optical thickness. The theory was developed for materials with small refractive index differences, although it is also valid in some cases for materials with large refractive index differences. Reflectance enhancements were obtained in a very broad band and over a wide range of incidence angles. The theory is particularly suited to design multilayers with enhanced reflectance in the extreme ultraviolet (EUV) for wavelengths above 50 nm. In this spectral region the reflectance of single layers of all materials is relatively low and standard multilayers are not possible because of the high absorption of materials.

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1. Introduction

All conventional coatings in the extreme ultraviolet (EUV) spectral region are burdened with low normal incidence reflectance. For wavelengths shorter than ~50 nm, multilayer coatings with enhanced EUV reflectance over that of single layers have been prepared over the last two decades. At wavelengths greater than ~50 nm all conventional materials strongly absorb radiation and standard multilayer coatings cannot be utilized because radiation is mostly absorbed in the outermost layer. Consequently, optical components for the EUV above 50 nm are coated with a single layer, except where a protective coating is needed, such as MgF_2- and LiF-protected Al films.

Design and fabrication of multilayer coatings for the EUV and soft x-rays has been mostly limited to periodic or quasi-periodic structures of bilayers consisting of two different materials. One material with low absorption is called the spacer, and the other material with a high refractive index contrast to the spacer, is commonly referred to as the scattering material. Other configurations have been developed by Boher *et al.*[1] and by Larruquert and Keski-Kuha[2]. Boher *et al.*[1] designed multilayer coatings with periodic structures by alternating three different materials. These multilayers were made of repeated trilayers of W/Rh/C, with reflectance values slightly higher than Rh/C or W/C multilayers in the spectral region 1-11 nm. Larruquert and Keski-Kuha[2] designed and prepared multilayer coatings with three layers of three different materials. These multilayers had an innermost opaque layer of Al, a spacer layer of MgF_2, and an outermost layer of a stable high reflectance material. They had high reflectance in the 50-200 nm spectral region.

In the present paper we investigate a new concept of multilayer coatings with reflectance enhancement: multilayers constructed by superposition of sub-quarterwave films of highly absorbing materials. Even though the mathematical derivations are valid in any spectral range, they are particularly suited for the EUV where transparent materials are not available. In Section 2 we show the mathematical derivations that give us material selection rules, optimum layer thickness, and optimized multilayer reflectance. Finally, in Section 3, we provide examples of reflectance enhancement with multilayers developed with the current theory.

2. Multilayer coatings with highly absorbing materials

2.1 A thin layer over an opaque substrate

In Fig. 1a a diagram of a simple multilayer containing a thin non-opaque film over an opaque substrate is shown. Both materials are assumed to be absorbing. The aim is to design multilayer coatings with reflectance higher than that of the individual materials. The radiation is assumed to impinge on the film from an incidence medium that may absorb radiation in general. Complex refractive indices for the two materials and the incidence medium are represented by N_1, N_2, and N_{Inc}, with $N_j=n_j+ik_j$, j=1,2, and $N_{Inc}=n_{Inc}+ik_{Inc}$. The amplitude reflectance of the bilayer is given by:

$$r = \frac{r_{Inc,1} + r_{1,2} \exp\left(4\pi i x N_1 \cos\theta_1 / \lambda\right)}{1 + r_{Inc,1} r_{1,2} \exp(4\pi i x N_1 \cos\theta_1 / \lambda)} \qquad (1)$$

where $r_{Inc,1}$ and $r_{1,2}$ are the well-known Fresnel reflectance coefficients between the incidence medium and medium 1, and between media 1 & 2, respectively, and λ is the vacuum wavelength. In the following we will always refer to normal-incident radiation, i.e. $\theta_1 = 0^0$.

Our goal is to obtain a mathematical relationship between N_{Inc}, N_1, and N_2 in order to achieve combination of materials providing a reflectance increase for a bilayer consisting of a thin film over an opaque substrate. Eq. (1) is difficult to handle in practice when we want to obtain general trends of the reflectance as a function of N_{Inc}, $N_1=N$, and $N_2=N+\Delta N$ with $N=n+ik$ and $\Delta N=\Delta n+i\Delta k$. Therefore, we will limit calculations to the particular case of $|\Delta N| << |N|$, which implies $|\Delta n|, |\Delta k| << |N|$. In Section 3 we will analyze practical examples where the following derivations are still valid for large $|\Delta N|$ values.

Under the above assumption $r_{1,2}$ can be expanded to first order in ΔN as:

$$r_{1,2} = \frac{N-(N+\Delta N)}{N+(N+\Delta N)} = -\frac{\Delta N}{2N} + O(\Delta N^2) \qquad (2)$$

Eq. (1) can now be approximated to first order in ΔN by:

$$r = r_{Inc,1} - \left(1 - r_{Inc,1}{}^2\right)\frac{\Delta N}{2N}\exp\left(\frac{4\pi i N x}{\lambda}\right) \qquad (3)$$

and the reflectance is given by the square modulus of Eq. (3), making use of the Fresnel coefficients:

$$R = R_{Inc,1} - \frac{4}{\left|N_{Inc} + N\right|^4} \mathrm{Re}\left[\frac{z}{N} \Delta N \exp\left(\frac{4\pi i N x}{\lambda}\right)\right] \qquad (4)$$

where Re[ ] stands for the real part of the expression in brackets. A convenient complex number z was used in Eq. 4, defined by:

$$z = N_{Inc} N\left(N_{Inc} *^2 - N *^2\right) \qquad (5)$$

The asterisk refers to the complex conjugate. Reflectance as a function of the outermost film thickness will take a maximum value when its first derivative with respect to the film thickness is zero and the second derivative is negative. The first derivative of reflectance is given by:

$$\frac{dR}{dx} = \frac{16\pi}{\lambda\left|N_{Inc} + N\right|^4} \exp\left(-\frac{4\pi k x}{\lambda}\right) \mathrm{Im}\left[z \Delta N \exp(i\varphi)\right]\Big|_{x = x_{ext}} = 0 \qquad (6)$$

where Im[ ] stands for the imaginary part of the expression in brackets, and x_{ext} stands for the film thickness at which R has an extreme value. In Eq. (6) we introduced the convenient parameter φ, which is related to x by:

$$\varphi = \frac{4\pi n x}{\lambda} \qquad (7)$$

Solutions φ_{ext} of Eq. (6) are the following:

$$tan\varphi_{ext} = -\frac{\Delta n A + \Delta k B}{\Delta n B - \Delta k A} \qquad (8)$$

$$A = \left(n_{Inc}{}^2 + k_{Inc}{}^2 + n^2 + k^2\right)\left(n_{Inc} k - k_{Inc} n\right)$$

$$B = \left(n_{Inc}{}^2 + k_{Inc}{}^2 - n^2 - k^2\right)\left(n_{Inc} n + k_{Inc} k\right)$$

We will now determine the condition for the extreme value to be a maximum by imposing that the second derivative of R at $x=x_{ext}$ be negative:

$$\frac{d^2R}{dx^2} = -4\left(\frac{4\pi}{\lambda}\right)^2 \frac{n}{\left|N_{Inc}+N\right|^4} \exp\left(-\frac{4\pi k x_{max}}{\lambda}\right) \frac{|z\Delta N|^2}{\mathrm{Im}(z\Delta N)} sin\varphi_{max} < 0 \qquad (9)$$

The value of φ_{ext} satisfying Eq. (9) was referred to as φ_{max}. $\tan^{-1}[-\mathrm{Im}(z\Delta N)/\mathrm{Re}(z\Delta N)]$ is a multi-valued function: if φ_0 is one solution of Eq. (8), then any number differing from φ_0 by an integer number times π is also a solution of Eq. (8). The solutions of Eq. (8) also satisfying Eq. (9) will be maxima and those not satisfying Eq. (9) will be minima. Since we are dealing with absorbing films, we will limit φ to values between 0 and π. This corresponds to a sub-quarterwave outermost film. We will now determine the condition for a sub-quarterwave film over an opaque substrate that will produce a reflectance enhancement. When condition (9) is not satisfied the sub-quarterwave film over an opaque substrate corresponds to a minimum. This case will not be analyzed in the present work.

Condition (9) can only be satisfied for φ_{max} ranging between 0 and π if the following condition:

$$\mathrm{Im}(z\Delta N) > 0 \qquad (10)$$

is satisfied. Expression (10) gives us the necessary condition when searching for materials with optical constants which will provide an increase in reflectance by means of depositing a sub-quarterwave film over an opaque substrate. Using definition (5), condition (10) turns into the following refractive index relationship:

$$\Delta nA + \Delta kB > 0 \qquad (11)$$

A and B are defined in Eq. (8). For the particular case where the incidence medium is vacuum, simplified values are obtained for A and B:

$$A_v = \left(1 + n^2 + k^2\right)k \qquad B_v = \left(1 - n^2 - k^2\right)n \qquad (12)$$

For instance, since A_v is always positive, a reflectance increase can be obtained with a bilayer where *n* increases from the film with the optimum thickness to the substrate, for a constant *k* value, and for vacuum as the incidence medium.

When condition (9), or the equivalents (10) and (11), are satisfied, the reflectance maximum is obtained using Eqs. (4) and (8):

$$R_{max} = R_{Inc,1} + 4n\frac{|N_{Inc}||N_{Inc} - N|}{|N||N_{Inc} + N|^3} \, |\Delta N| \exp\left(-\frac{4\pi k x_{max}}{\lambda}\right) \qquad (13)$$

Since the right hand term of Eq. (13) is positive, provided that condition (11) is satisfied, R_{max}>$R_{Inc,1}$. Therefore, reflectance of the above multilayer is higher than reflectance at the interface between the incidence medium and an opaque film of material with refractive index N_1=N. We will now show that R_{max}> $R_{Inc,2}$, where $R_{Inc,2}$ is the reflectance obtained at an interface between the incidence medium and the opaque substrate. The first derivative of the multilayer reflectance with respect to the thin film thickness is obtained from Eq. (4) and can be expressed as:

$$\frac{dR}{dx} = \frac{16\pi}{\lambda|N_{Inc}+N|^4}|z\Delta N| sin(\varphi_{max}-\varphi)\exp\left(-\frac{4\pi kx}{\lambda}\right) \qquad (14)$$

dR/dx is positive for any film thickness in the interval [0, x_{max}], and therefore, reflectance monotonically grows from 0 to x_{max}. Hence, R_{max}>R(x=0)=$R_{Inc,2}$. In short, the reflectance of the multilayer is higher than the reflectance at the interface between material with refractive index N_{Inc} and each material with either refractive indices N_1 or N_2. This is an important result which shows that for any incidence medium, if we have two materials X and Y with a small difference in their refractive indices, there is a way to construct a bilayer with a higher reflectance compared to the reflectance of each material independently. If condition (10) is not satisfied with a bilayer X-on-Y, it will be automatically satisfied with a bilayer Y-on-X, because Δn and Δk will have opposite signs.

2.2 Multilayers with two layers over an opaque substrate

The reflectance increase obtained with one film over an opaque substrate leads us to wonder whether the addition of a third material with a small refractive index difference may provide an additional reflectance increase under a certain refractive index selection rule. Fig. 1b shows a sketch of a multilayer consisting of 2 layers with thicknesses x_1 and x_2, over an opaque substrate. The refractive index of the outermost layer, middle layer, and substrate are expressed by: N_1=N, N_2=N_1+ΔN_1, N_3=N_2+ΔN_2, respectively. The three refractive indices are assumed to differ by small amounts: $|\Delta N_1|, |\Delta N_2| << |N|$, which implies $|\Delta n_1|, |\Delta k_1|, |\Delta n_2|, |\Delta k_2| << |N|$. The amplitude reflectance of the multilayer to first order in ΔN_1 and ΔN_2 is obtained in a similar manner to the case of a single layer discussed in Section 2.1:

$$r = r_{Inc,1} - \frac{1 - r_{Inc,1}^{\ \ 2}}{2N}\left\{\Delta N_1 \exp\left(\frac{4\pi i N x_1}{\lambda}\right) + \Delta N_2 \exp\left[\frac{4\pi i N(x_1 + x_2)}{\lambda}\right]\right\} \qquad (15)$$

Thus, the intensity reflectance is given by:

$$R = R_{Inc,1} - \frac{4}{\left|N_{Inc} + N\right|^4}\,\mathrm{Re}\left\{\frac{z}{N}\left(\Delta N_1 \exp\left[\frac{4\pi i N x_1}{\lambda}\right] + \Delta N_2 \exp\left[\frac{4\pi i N(x_1 + x_2)}{\lambda}\right]\right)\right\} \quad (16)$$

Optimum film thicknesses x_1 and x_2 are obtained when the multilayer reflectance takes a maximum value with respect to both variables. Proceeding with a similar method used to determine the maximum reflectance with a single film, we can obtain the refractive index conditions for maximum reflectance of the 3-layer. Further details of the calculation are given in Appendix A. Similar to the case for one film, among the different solution pairs (φ_{ext1}, φ_{ext2}) for which the two partial derivatives of reflectance given by Eq. (16) are zero, we are interested in the lowest positive φ_{ext1} and φ_{ext2}. In other words, we limit φ_{ext1} and φ_{ext2} to values between 0 and π which correspond to sub-quarterwave films. The conditions for the reflectance to have a maximum at φ_{ext1} and φ_{ext2} are the following:

$$\Delta n_1 A + \Delta k_1 B > 0 \qquad (17-1)$$

$$\Delta n_1 \Delta k_2 < \Delta n_2 \Delta k_1 \qquad (17-2)$$

We refer to them as φ_{max1} and φ_{max2} and they are related to x_1 and x_2, respectively, through Eq. (7). A and B are as defined in Eq. (8) and are simplified to A_v and B_v given by Eq. (12) when the incidence medium is vacuum. Condition (17-1) is the same that condition (11). The additional condition (17-2) depends only on the refractive index increments and not on the incidence medium or on the material refractive indices. Film thicknesses at the maximum are given by:

$$tan\varphi_{max1} = -\frac{\Delta n_1 A + \Delta k_1 B}{\Delta n_1 B - \Delta k_1 A}; \qquad (18-1)$$

$$tan\varphi_{max2} = \frac{\Delta n_2 \Delta k_1 - \Delta k_2 \Delta n_1}{\Delta n_1 \Delta n_2 + \Delta k_1 \Delta k_2}; \qquad (18-2)$$

and the reflectance at the maximum is given by:

$$R_{max} = R_{Inc,1} + 4n\frac{|N_{Inc}||N_{Inc} - N|}{|N||N_{Inc} + N|^3} \times \\ \times \left\{ |\Delta N_1| \exp\left(-\frac{4\pi k x_{max1}}{\lambda} \right) + |\Delta N_2| \exp\left[-\frac{4\pi k \left(x_{max1} + x_{max2} \right)}{\lambda} \right] \right\} \qquad (19)$$

Details of the derivations of Eqs. (17) to (19) are given in Appendix A.

Assuming we have the same incidence medium, the reflectance of the 3-layer is higher than that of any individual layer with material refractive indices of N_1, N_2, and N_3. The reflectance of the 3-layer is also higher than a sub-quarterwave multilayer of a thin film of material No. 1 over an opaque substrate of material No. 2, and also higher than a sub-quarterwave multilayer of a thin film of material No. 2 over an opaque substrate of material No. 3. In addition, it is also true that reflectance of the 3-layer is higher than sub-quarterwave multilayers of a thin film of material No. 1 over an opaque substrate of material No. 3 at least for the particular case where $Im(z\Delta N_2)>0$ and conditions (17-1) and (17-2) are satisfied. Indications to demonstrate the above are given in Appendix A. Consequently, with the adequate material selection given by Eqs. (17-1) and (17-2) we can construct 3-layer coatings with higher reflectance than single or 2-layer coatings.

2.3 Multilayers with three or more layers over an opaque substrate

We can generalize the previous results to any number *m* of thin films over a substrate of *m*+1 different materials whose refractive index differ by a small amount. We express the refractive index difference of adjacent layers by $\Delta N_i = N_{i+1} - N_i$, i=1 to *m*, starting with the outermost film. Assuming $|\Delta N_i| << |N_1|$, i=1 to m, the conditions for the reflectance to have an m-dimensional maximum at point (φ_{max1}, φ_{max2},...,$\varphi_{max,m}$), with $0 \le \varphi_{max1}, \varphi_{max2}, ..., \varphi_{max,m} < \pi$, are the following:

$$\Delta n_1 A + \Delta k_1 B > 0 \qquad (20-1)$$
$$\Delta n_1 \Delta k_2 < \Delta n_2 \Delta k_1 \qquad (20-2)$$
$$\Delta n_2 \Delta k_3 < \Delta n_3 \Delta k_2 \qquad (20-3)$$
$$...$$
$$\Delta n_{m-1} \Delta k_m < \Delta n_m \Delta k_{m-1} \qquad (20-m)$$

The above are the conditions required for the *m*+1 materials to combine in a multilayer with *m* sub-quarterwave films over an opaque substrate with a reflectance enhancement. A and B are as defined in Eq. (8), and are simplified to A_v and B_v given by Eq. (12) when the incidence medium is vacuum. The above conditions (20-1) to (20-m) give us selection rules for the materials to be used in the multilayer. Optimum film thickness are given by:

$$tan\varphi_{max,1} = -\frac{\Delta n_1 A + \Delta k_1 B}{\Delta n_1 B - \Delta k_1 A}; \qquad (21-1)$$

$$tan\varphi_{max2} = \frac{\Delta n_2 \Delta k_1 - \Delta k_2 \Delta n_1}{\Delta n_1 \Delta n_2 + \Delta k_1 \Delta k_2}; \qquad (21-2)$$

$$tan\varphi_{max3} = \frac{\Delta n_3 \Delta k_2 - \Delta k_3 \Delta n_2}{\Delta n_2 \Delta n_3 + \Delta k_2 \Delta k_3}; \qquad (21-3)$$

...

$$tan\varphi_{max,m} = \frac{\Delta n_m \Delta k_{m-1} - \Delta k_m \Delta n_{m-1}}{\Delta n_{m-1} \Delta n_m + \Delta k_{m-1} \Delta k_m}; \qquad (21-m)$$

and the reflectance at the m-dimensional maximum is given by:

$$R_{max} = R_{Inc,1} + 4n\frac{|N_{Inc}||N_{Inc} - N|}{|N||N_{Inc} + N|^3}\left\{ |\Delta N_1| \exp\left(-\frac{4\pi k x_{max1}}{\lambda}\right) + \right.$$

$$\left. + |\Delta N_2| \exp\left[-\frac{4\pi k\left(x_{max1} + x_{max2}\right)}{\lambda}\right] + ... + |\Delta N_m| \exp\left[-\frac{4\pi k \sum_{i=1}^{m} x_{max,i}}{\lambda}\right] \right\} \qquad (22)$$

All the above derivations of Sections 2.1, 2.2, and 2.3 were obtained assuming that the refractive index of each individual film and of the substrate differ by a small amount. This assumption may seem very restrictive, however, we will show examples in Section 3 that reflectance enhancement through sub-quarterwave layers can also be obtained for large refractive index differences.

3. Examples of sub-quarterwave multilayers

In this Section we provide several examples of sub-quarterwave multilayers to show the validity of the theory developed in Section 2.

3.1 Multilayers with high reflectance at 83.4 nm

The OI 83.4 nm is an important spectral line for atmospheric physics. Observations at this and nearby wavelengths are sometimes difficult due to the low intensity of the sources observed. At this wavelength the reflectance of standard coatings is moderately low and the high absorption of all materials prevents the use of standard multilayer coatings. Hence, an increase in the reflectance of optical coatings at this wavelength is highly desirable.

In order to construct a sub-quarterwave multilayer with enhanced reflectance at 83.4 nm we will start with a simple multilayer of one thin film over an opaque substrate. Coatings of ion-beam-deposited (IBD) SiC are among those with highest reflectance at 83.4 nm. The optical constants of IBD SiC are shown in Table 1, and were obtained from Larruquert and Keski-Kuha data[3]. Let the incidence medium be vacuum and let the thin film consist of SiC. We will look for a substrate material with adequate refractive index. We apply these data to Eq. (11) in order to obtain the conditions for Δn and Δk to achieve an increase in reflectance. Eq. (11) results in values of $3.26\Delta n - 0.50\Delta k>0$. A list of optical constants for various materials is given in Table 1. The optical constants were obtained from Blumenstock and Keski-Kuha[4] (IBD B_4C), Larruquert and Keski-Kuha[5] (IBD C), the compilation by Palik[6] (Al_2O_3) and Larruquert and Keski-Kuha[7] (evaporated MgF_2). The above inequality is satisfied for multilayers of SiC on B_4C, on C, on Al_2O_3 or on MgF_2.

We will now address the more complex case of a multilayer of several thin films over an opaque substrate. In the theory developed in Section 2, the optimum thickness and reflectance calculation were referred to the optical constants of the outermost film N_1=N. If we had referred the derivations to any material with refractive index N_i, i=2 to m+1, then all the Eqs. in Sections 2.2 and 2.3 would have been formally identical, only replacing N_1=N with N_i of i-th film. The reason for that is that all the refractive indices are assumed to be close to each other and the equations were developed to first order in ΔN_i, i=1 to m. However, in the current example of a thin SiC film over an opaque substrate, where optical constant difference is large, the above is not true.

We determined that the use of the optical constants of the material for which film thickness was calculated (instead of that of the outermost film) gave a better reflectance and thickness agreement with the exact calculations. This result can be incorporated in the theory by making a simple modification. The optimum thickness calculation for film *i*, obtained from Eqs. (21), was related to the parameter φ through Eq. (7). This equation is now modified to:

$$\varphi_i = \frac{4\pi n_i x_i}{\lambda}, \quad i = 1 \quad to \quad m \qquad (23)$$

where *n* is replaced by n_i. Modification given by Eq. (23) is of second order in ΔN_i, i=1 to m, so that all Eqs. given in Section 2 are not changed to first order in ΔN_i, but are more accurate when the refractive index differences are large. This modification will be used in the calculations below.

We calculated all the multilayers with enhanced reflectance based on the materials given in Table 1 with SiC as the outermost layer. In order to test the current theory, the optimum thickness and reflectance of sub-quarterwave multilayers were obtained both by using the current theory and by exact calculation of the multilayer reflectance. The exact calculation was performed by a search of the optimum film thicknesses by trial and error. Both sets of thickness data and optimum reflectance for the different possible multilayers are given in Table 2. Any multilayer obtained by selecting materials in the order given by SiC/ B_4C/ C/ Al_2O_3/ MgF_2 starting with the outermost layer satisfies conditions (20-1) to (20-m) and gives a reflectance enhancement over a multilayer where one or more of the materials are eliminated. Any change in the order will prevent some of the conditions (20) to be satisfied.

The material selection rule given by Eq. (20) was always successful in determining the combination and order of materials to obtain a multilayer with enhanced reflectance. Furthermore, film thickness and reflectance values calculated with current theory are remarkably accurate for multilayers of SiC on B_4C, and provides the right order of magnitude in all the multilayers shown in Table 2, even though the refractive index differences were very large.

From Table 2, a multilayer with one (two, three, four) layers over the substrate results in a reflectance increase of 0.029 (0.043, 0.044, 0.047, respectively) over the intrinsic reflectance of SiC. Due to the high radiation absorption of all the materials used, the reflectance increase saturates for a multilayer with about 4-5 layers.

The reflectance increase may appear modest. Nevertheless, since at this wavelength neither materials with high reflectance nor standard multilayers are available, any reflectance increase is welcome, particularly for astrophysical applications. Furthermore, optical systems with multiple reflections may benefit from such a reflectance enhancement at every reflection.

In Fig. 2, we present the reflectance as a function of wavelength for multilayers consisting of one to five layers. Remarkably, the increase in reflectance, that was optimized at 83.4 nm, extends over a wide spectral region.

In order to observe the intrinsic multilayer bandwidth, we set the refractive indices of the 4 materials to be constant over the entire spectrum and calculated the reflectance versus wavelength of a multilayer SiC/B_4C/C/Al_2O_3 optimized at 83.4 nm. The refractive index for each material was set equal to its value at 83.4 nm over the entire spectrum investigated. Thus, we eliminated any effects of the wavelength-dependent refractive index, which may mask the intrinsic multilayer bandwidth. As shown in Fig. 3, the reflectance smoothly decreased below 83.4 nm towards the IBD SiC reflectance. Above 83.4 nm the reflectance decreased extremely slowly and maintained a level greater than the IBD SiC reflectance at 83.4 nm up to about 180 nm.

The reflectance for non-polarized radiation versus the angle of incidence of multilayer SiC/B_4C/C/$Al_2O_3MgF_2$ is shown in Fig. 4. Similarly to the dependence on wavelength, the reflectance increase of a multilayer over single layers is obtained for a wide range of incidence angles.

The slow dependence of multilayer reflectance with wavelength results in a very high tolerance for layer thickness and for possible errors in the optical constants of materials. As an example, let us calculate the thickness tolerance for the multilayer 7.6 nm SiC/ 3.2 nm B_4C/ 4.4 nm C/ 2.6 nm Al_2O_3/ opaque MgF_2 optimized at 83.4 nm at normal incidence. In order to lower the multilayer reflectance at 83.4 nm with 0.01, it would be necessary to increase (decrease) simultaneously every layer thickness by 40% (25%) with respect to the thickness for the optimized multilayer.

3.2 Multilayers with high reflectance at 53.6 nm

The He I 53.6-nm spectral line is an important line for astrophysical observations. However, the normal reflectance of the available coatings is low and a reflectance increase is desirable. Standard coatings at 53.6 nm are Ir, Pt and B_4C, among others. In the present calculation we will also make use of Mo and C_{60} fullerenes, because their optical constants are adequate. The optical constants of these materials are shown in Table 3. Optical constants were obtained from Blumenstock and Keski-Kuha[4] (IBD B_4C), Larruquert and Keski-Kuha[3] (IBD Mo), the compilation by Palik[8] (Ir) and Méndez *et al.*[9] (C_{60}). Table 4 compares the reflectance of different optimized multilayers with that of single layers; in this table only exact thickness and reflectance values are provided. The order in which materials are superimposed on the multilayer is given by Eqs. (20-1) to (20-m) and it is IBD B_4C on IBD Mo on Ir on C_{60}. Any change in the order will be detrimental to reflectance. A multilayer with one (two, three) layer over an opaque substrate results in a reflectance increase of 0.042 (0.049, 0.054) compared to the material with the highest single-layer reflectance of the above (Mo). Since Mo oxidizes over time, with an important reflectance decrease, comparison of multilayer reflectance with that of single Ir films is more informative. The reflectance increase of the multilayer over single Ir films is 0.060 (0.067, 0.072), a relative increase of 26% (29%, 31%).

4. Discussion

We have developed a mathematical theory and material selection rules to construct sub-quarterwave multilayers to obtain the highest possible normal incidence reflectance by combining highly absorbing materials. Even though the derivations were made under the assumption that the refractive index difference among the materials was small the equations were found to be qualitatively and even quantitatively valid for rather large differences. Hence, the theory presented here may find applications for a wider material spectrum that can be expected from the approximations used. The theory presented provides a method of determining the correct combination of materials to achieve a reflectance increase. Film thickness and reflectance may have to be checked by exact calculation in the most usual case where refractive index differences are large. However, thickness and reflectance values calculated with current theory give the right order of magnitude for large refractive index differences. The high tolerance of these multilayers helps the use of the approximate calculations in a real multilayer design.

Eqs. (21-1) to (21-m) predict further reflectance increases by adding new layers to the multilayer as long as the material refractive indices of the different layers satisfy Eqs. (20-1) to (20-m). However, the examples shown in Sections 3.1 and 3.2 suggest that the reflectance increase saturates at approximately four to five layers. This is caused by the high absorption of the materials: radiation intensity decreases as it penetrates into deeper layers. Therefore, the high absorption of materials limits the number of useful layers and the reflectance enhancement of sub-quarterwave multilayers.

The applicability of current multilayers is mostly suitable in the EUV spectral region ~50-125 nm. This is because outside this interval high normal reflectance is regularly obtained by use of highly reflective materials (above 115 nm) or by multilayer coatings (both below ~50 nm and above ~125 nm). In any event, the derivations are applicable all over the spectrum.

In section 2 we have shown that a reflectance increase will occur but we have not addressed the compatibility among materials and multilayer stability over time. The model presented assumes smooth and abrupt interfaces between the adjacent layers of different materials, with no interdiffusion through the interface. In practice, actual multilayers may not be ideal and therefore, the particular examples of Section 3 might give rise to unstable multilayers.

The multilayers proposed in Sections 3.1 and 3.2 have outermost layers of SiC and B_4C. These materials oxidize slightly under normal atmospheric conditions, resulting in a limited reflectance decrease over time. In spite of this, SiC and B_4C are standard coatings for the EUV. Slight oxidation of outermost thin films of SiC or B_4C would probably result in a limited multilayer reflectance decrease over time. The multilayers that have been proposed in Section 3 should be understood as examples of what can be expected of sub-quarterwave multilayers. Real reflectance enhancements and stability over time will have to be experimentally determined for a given set of materials.

Conclusions

A theory of sub-quarterwave multilayers with highly absorbing materials has been developed. The theory provides the selection rules for multilayer construction, along with layer thickness and multilayer reflectance. Even though the theory was developed under the assumption that the refractive index of all materials in the multilayer are close, examples are shown in which the material selection rules are still valid for rather large refractive index differences and the calculated layer thickness and reflectance give the correct order of magnitude.

The theory states that when two materials with close refractive indices are selected, a multilayer can always be designed with higher reflectance than that of the individual materials by the superposition of a thin film of one of the materials on top of an opaque substrate of the other material.

Reflectance enhancements with sub-quarterwave multilayers were optimized at a given wavelength and were shown to extend over a wide spectral region and over a wide angle of incidence range. Since the materials used in the multilayer strongly absorb radiation, the reflectance enhancement due to the addition of new layers saturates at a small number of layers, on the order of four-five.

The theory developed is particularly suited for the design of multilayers for the EUV in the spectral range of ~50-125 nm.

Acknowledgments

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Appendix A.

We will calculate the optimum thickness for the highest possible reflectance of a multilayer containing two thin films over an opaque substrate. We have adopted the nomenclature from Section 2.2 and Fig. 1b, with the constraints: $|\Delta N_1|$, $|\Delta N_2| << |N|$. The reflectance of the multilayer was given in Eq. (16), and is reproduced here:

$$R = R_{Inc,1} - \frac{4}{|N_{Inc}+N|^4}\operatorname{Re}\left\{\frac{z}{N}\left(\Delta N_1 \exp\left[\frac{4\pi i N x_1}{\lambda}\right] + \Delta N_2 \exp\left[\frac{4\pi i N(x_1+x_2)}{\lambda}\right]\right)\right\} \quad (A \;\; 1)$$

At this point we want to obtain film thicknesses x_1 and x_2 for which reflectance takes a maximum. The conditions to have a maximum at point ($x_{max\ 1}$, $x_{max\ 2}$) are:

$$\left.\frac{\partial R}{\partial x_1}\right|_{(x_{max1},\, x_{max2})} = 0 \qquad (A \;\; 2-1)$$

$$\left.\frac{\partial R}{\partial x_2}\right|_{(x_{max1},\, x_{max2})} = 0 \qquad (A \;\; 2-2)$$

$$\left.\frac{\partial^2 R}{\partial x_2{}^2}\right|_{(x_{max1},\, x_{max2})} < 0 \qquad (A \;\; 2-3)$$

$$\left.\left[\frac{\partial^2 R}{\partial x_1{}^2}\frac{\partial^2 R}{\partial x_2{}^2} - \left(\frac{\partial^2 R}{\partial x_1 \partial x_2}\right)^2\right]\right|_{(x_{max1},\, x_{max2})} > 0 \qquad (A \;\; 2-4)$$

Conditions (A 2-1 to A 2-4) lead to:

$$\mathrm{Im}\left[z\Delta N_2 \exp\{i(\varphi_{max1} + \varphi_{max2})\}\right] = 0 \qquad (A \quad 3-1)$$

$$\mathrm{Im}\left[z\Delta N_1 \exp(i\varphi_{max1})\right] = 0 \qquad (A \quad 3-2)$$

$$\frac{n|z|^2|\Delta N_2|^2}{\mathrm{Im}(z\Delta N_1)\mathrm{Im}(u)} sin\varphi_{max1} sin\varphi_{max2} > 0 \quad (A \quad 3-3)$$

$$\frac{n^2|z|^4|\Delta N_1|^2|\Delta N_2|^2}{\mathrm{Im}^2(z\Delta N_1)\mathrm{Im}(u)} sin\varphi_{max1} sin^2\varphi_{max2} > 0 \quad (A \quad 3-4)$$

where

$$\varphi_{max1} = \frac{4\pi n x_{max1}}{\lambda} \qquad (A \quad 4)$$

$$\varphi_{max2} = \frac{4\pi n x_{max2}}{\lambda}$$

From Eqs. (A 3-1) and (A 3-2) we get:

$$tan\varphi_{max1} = -\frac{\mathrm{Im}(z\Delta N_1)}{\mathrm{Re}(z\Delta N_1)}; \qquad (A \quad 5-1)$$

$$tan\varphi_{max2} = -\frac{\mathrm{Im}(\Delta N_2 / \Delta N_1)}{\mathrm{Re}(\Delta N_2 / \Delta N_1)}; \qquad (A \quad 5-2)$$

where z is a complex number defined in Eq. (5). From Eqs. (A 5-1) and (A 5-2) it is straightforward to obtain Eqs. (18-1) and (18-2). Similar to Section 2.1, we will limit ourselves to sub-quarterwave films, i.e., $0 \leq \varphi_{max\,1}$, $\varphi_{max\,2} < \pi$. Therefore, the inequalities (A 3-3) and (A 3-4) yield:

$$\mathrm{Im}(z\Delta N_1) > 0 \qquad (A \quad 6-1)$$

$$\mathrm{Im}(\Delta N_2 / \Delta N_1) < 0 \qquad (A \quad 6-2)$$

From the definition of z, inequalities (A 6-1) and (A 6-2) turn into:

$$\Delta n_1 A + \Delta k_1 B > 0 \qquad (A \quad 7-1)$$

$$\Delta n_1 \Delta k_2 < \Delta n_2 \Delta k_1 \qquad (A \quad 7-2)$$

where A and B are as defined in Eq. (8). The reflectance at the maximum is obtained from Eqs. (A1) and (A5):

$$R_{max} = R_{Inc,1} + 4n \frac{|N_{Inc}||N_{Inc} - N|}{|N||N_{Inc} + N|^3} \times \\ \times \left\{ |\Delta N_1| \exp\left(-\frac{4\pi k x_{max1}}{\lambda} \right) + |\Delta N_2| \exp\left[-\frac{4\pi k \left(x_{max1} + x_{max2} \right)}{\lambda} \right] \right\} \quad (A \quad 8)$$

Since the right hand term of (A 8) is positive, the multilayer reflectance is always higher than the intrinsic reflectance of material No. 1 in the same incidence medium. Comparing Eqs. (13) and (A 8) we determine that the reflectance of the 3-layer is higher than a multilayer of layer No.1 over an opaque substrate of material No. 2. Since we are at a local maximum of the function R(x_1, x_2), the reflectance will decrease in any direction starting at ($x_{max\,1}$, $x_{max\,2}$). Partial derivatives of reflectance are given by:

$$\frac{\partial R}{\partial x_2}\left(x_1, x_2\right) = -\left(\frac{4\pi}{\lambda}\right) \frac{4}{|N_{Inc} + N|^4} \exp\left[-\frac{4\pi k \left(x_1 + x_2\right)}{\lambda} \right] \frac{\left| \frac{\Delta N_2}{\Delta N_1} \right|^2}{\mathrm{Im}\left(\frac{\Delta N_2}{\Delta N_1} \right)} \times \\ \times sin\left(\varphi_{max1} + \varphi_{max2}\right) sin\left(\varphi_1 - \varphi_{max1} + \varphi_2 - \varphi_{max2}\right) \qquad (A \quad 9-1)$$

$$\frac{\partial R}{\partial x_1}\left(x_1, x_2\right) = -\left(\frac{4\pi}{\lambda}\right) \frac{4}{|N_{Inc} + N|^4} \exp\left(-\frac{4\pi k x_1}{\lambda} \right) \left| z \Delta N_1 \right| sin\left(\varphi_1 - \varphi_{max1}\right) + \\ + \frac{\partial R}{\partial x_2} \qquad (A \quad 9-2)$$

In order to demonstrate that the reflectance of the 3-layer is higher than that of a multilayer of layer No.2 over an opaque substrate of material No. 3 we calculated the directional derivative of the 3-layer reflectance along a straight line starting at point P_1 and terminating at P_2. P_1 corresponds to the maximum for the multilayer of layer No.2 over an opaque substrate of material No. 3. $P_1=(x_1=0,\ x_2=x_{max\ 1}+x_{max\ 2})$ when $\varphi_{max\ 1}+\varphi_{max\ 2}<\pi$; otherwise $P_1=(x_1=0,\ x_2=x_{max\ 1}+x_{max\ 2}-\lambda/4n)$. $P_2=(x_1=x_{max\ 1},\ x_2=x_{max\ 2})$ corresponds to the maximum for the 3-layer multilayer. It is straightforward to notice that the directional derivative is positive from P_1 to P_2 and the reflectance continuously increases from P_1 to P_2 and approaches zero at the point P_2. This demonstrates that reflectance of the 3-layer is higher than a multilayer of layer No.2 over an opaque substrate of material No. 3.

We will now determine a sufficient condition for the 3-layer to achieve a higher reflectance compared to a sub-quarterwave film of material No. 1 over an opaque substrate of material No. 3. Let $P_3=(x_1=x_{max1}',x_2=0)$ be the point in the thickness space for which the latter multilayer has a maximum. x_{max1}' is then the sub-quarterwave film thickness of the outermost film for optimum reflectance. From Eqs. (A 9-1) and (A 9-2) we can obtain that the directional derivative of the 3-layer multilayer is positive along a straight line from P_3 to P_2, and reflectance continuously increases from P_3 to P_2, when the following condition is satisfied:

$$\mathrm{Im}(z\Delta N_2) > 0 \qquad (A\ \ 10)$$

(A10) is a sufficient condition, and it is added to conditions (A 6-1) and (A 6-2). As shown in Section 2.1, the reflectance of the 2-layer is higher than that of any individual material. In summary, the reflectance of the optimized 3-layer with the added condition (A10) is higher than any other possible combination of the 3 materials.

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Table 1. Optical constants of different materials at 83.4 nm

Material	n	k
SiC	0.58	1.07
B_4C	0.816	1.246
C	1.16	1.29
Al_2O_3	1.350	1.147
MgF_2	1.59	0.49

Table 2. Reflectance of sub-quarterwave multilayers at 83.4 nm. When two thickness and reflectance values are given, the first one is the exact value and the second one is the calculated value with current theory

Thickness (nm)									Reflectance	
IBD SiC		IBD B_4C		IBD C		Al_2O_3		MgF_2	Exact	Calculated
Opaque									0.363	
		Opaque							0.327	
				Opaque					0.267	
						Opaque			0.212	
								Opaque	0.085	
8.7	9.4	Opaque							0.383	0.383
10.6	12.5			Opaque					0.392	0.389
12.8	15.6					Opaque			0.388	0.382
17.4	22.7							Opaque	0.383	0.372
8.1	9.4	3.8	3.9	Opaque					0.396	0.396
8.1	9.4	6.0	6.7			Opaque			0.396	0.397
8.0	9.4	9.1	11.5					Opaque	0.399	0.395
10.3	12.5			6.3	6.0	Opaque			0.397	0.393
9.7	12.5			7.6	8.4			Opaque	0.406	0.399
11.7	15.6					6.5	6.5	Opaque	0.400	0.389
8.0	9.4	3.6	3.9	4.9	4.8	Opaque			0.400	0.400
7.6	9.4	3.3	3.9	6.5	7.1			Opaque	0.409	0.406
7.7	9.4	5.1	6.7			5.3	5.1	Opaque	0.408	0.403
9.6	12.5			5.1	6.0	3.1	2.7	Opaque	0.408	0.400
7.6	9.4	3.2	3.9	4.0	4.8	3.1	2.7	Opaque	0.411	0.408

Table 3. Optical constants of different materials at 53.6 nm

Material	n	k
B_4C	0.517	0.575
Mo	0.549	0.731
Ir	0.79	0.96
C_{60}	0.804	0.477

Table 4. Reflectance of sub-quarterwave multilayers at 53.6 nm

Thickness (nm)				Exact reflectance
IBD B_4C	IBD Mo	Ir	C_{60}	
Opaque				0.215
	Opaque			0.251
		Opaque		0.233
			Opaque	0.076
5.8		Opaque		0.295
3.0	3.8	Opaque		0.302
3.0	3.7	11.7	Opaque	0.307

Figure Captions

Fig. 1. Schematic diagram of the incident, reflected and transmitted rays for (a) one thin film on an opaque substrate, and (b) two thin films on an opaque substrate.

Fig. 2. Calculated reflectance of an opaque layer of IBD SiC, and of different multilayers optimized at 83.4 nm. From left to right the materials begin from the outermost layer to the opaque substrate, respectively.

Fig. 3. The intrinsic multilayer reflectance dependence upon wavelength. Refractive indices of the materials are set constant in the calculation throughout the spectral range shown, equal to their values at 83.4 nm. Materials and film thickness data starting with the outermost layer were those for the optimized multilayer at 83.4 nm: 8.0 nm SiC/ 3.7 nm B_4C/ 5.5 nm C/ opaque Al_2O_3. SiC film reflectance of 0.363 at 83.4 nm is displayed for reference.

Fig. 4. Calculated reflectance versus angle of incidence with respect to the normal for opaque IBD SiC and for the following multilayer that was optimized at 83.4 nm (starting with the outermost layer): 7.6 nm SiC/ 3.2 nm B_4C/ 4.4 nm C/ 2.6 nm Al_2O_3/ opaque MgF_2. The radiation is assumed to be unpolarized.

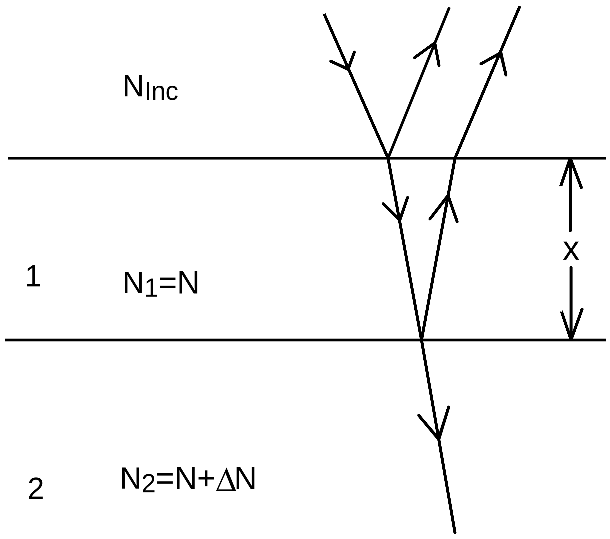

Fig. 1.a
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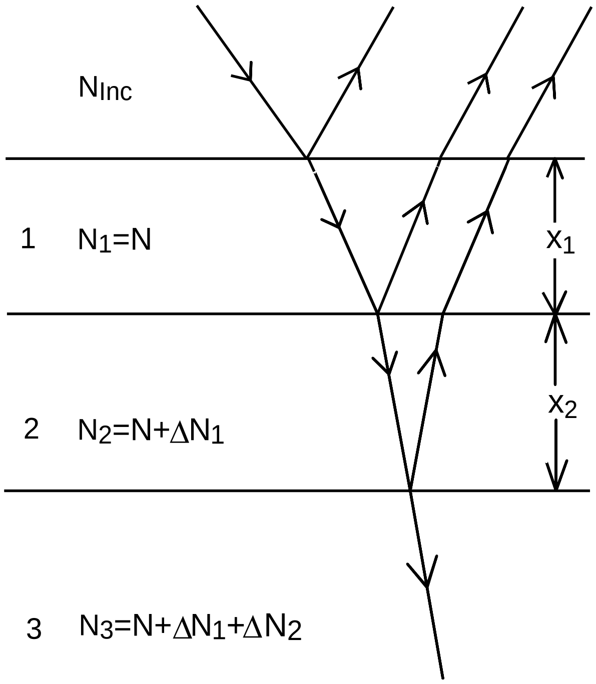

Fig. 1.b
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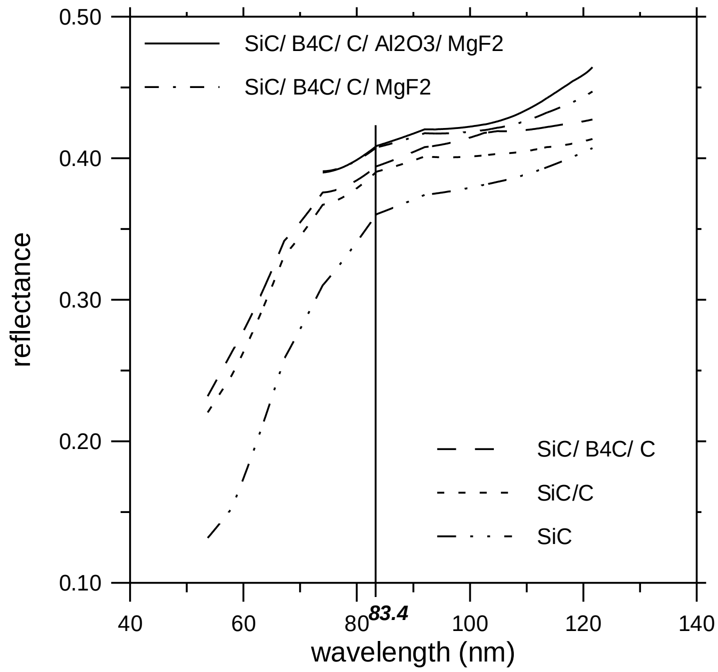

Fig. 2
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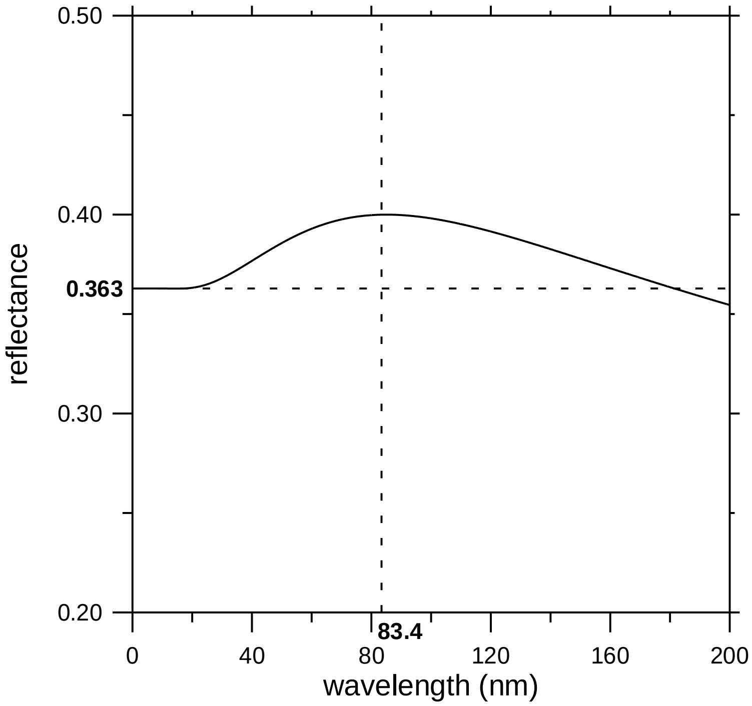

Fig. 3
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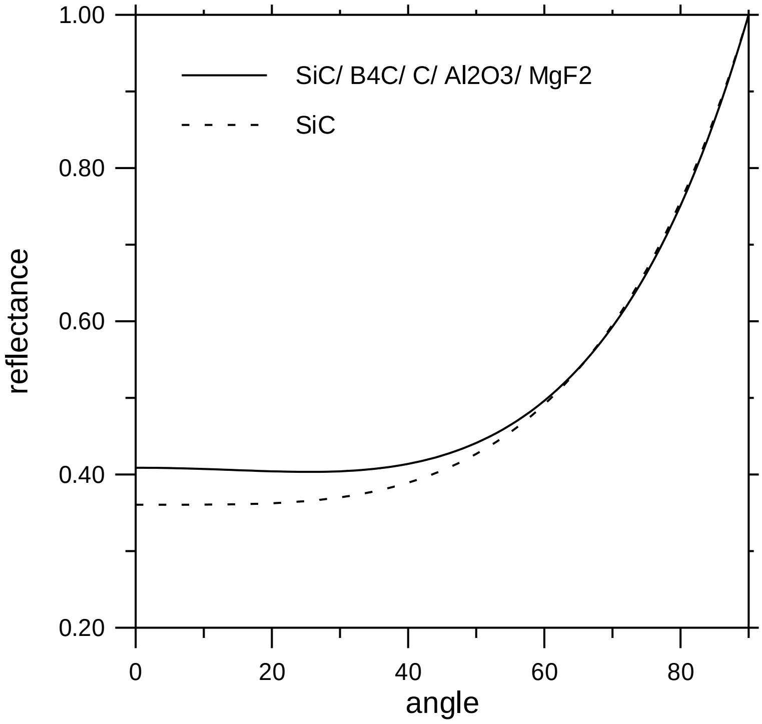

Fig. 4
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