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Effective characteristic matrix of ultrathin multilayer structures

A.J. HAIJA, W. LARRY FREEMAN, THOMAS ROARTY

Department of Physics, Indiana University of Pennsylvania, Indiana, PA 15705 - 1087, U.S.A.

This paper presents the calculation of the reflectivity, transmissivity and optical constants of ultrathin Cu-Ni multilayer stacks using the characteristic effective medium approximation (CEMA) introduced in an earlier communication. Each of the Cu-Ni multilayer stacks has an identity period of 100 Å, Cu – 45 Å and Ni – 55 Å. Calculations of the reflectivity and transmissivity are executed via the characteristic matrix technique employed in three ways. In the first the characteristic matrix of the Cu-Ni bilayer is calculated and then raised to a power equal to the number of layers in the stack following the characteristic matrix technique. The second is based on the calculation of the characteristic matrix of the bilayer identity period using the effective complex index of refraction of the identity period as derived according to the CEMA. The third is based on the equivalent characteristic matrix of the whole stack represented by one characteristic matrix, also using the CEMA; in this method the equivalent optical constants of the layered structure are also calculated. All calculations are in the visible and for normal incidence. A comparison between results of the first two methods of calculations shows that they are almost identical. However, displaying values using the equivalent effective matrix of the whole structure shows definite quantitative differences throughout the whole studied spectral range. The difference becomes rather noticeable when the number of layers is greater than or equal to six amounting to a minimum total thickness of 600 Å for the layered stack as a whole. This establishes a quantitative criterion to the limit beyond which the CEMA cannot be applied.

Keywords: multilayers, superlattices, bilayer stacks.

1. Introduction

This article follows several papers [1–3] that have addressed the derivation and application of new expressions for the optical constants (index of refraction and extinction coefficient) of the identity period in a multilayer superlattice stack [1] and the implications of these formulae for the values of the optical properties of such structures. The objective of this communication is two-fold, the first is to assert the validity of the derived expressions established and labeled by Haija as the characteristic effective medium approximation (CEMA) and the second is to establish a decisive criterion for the upper limit of the CEMA when employing it for a superlattice stack. In the first objective, not only will one have new corrected expressions for the effective optical constants of a bilayer identity period in contrast to the geometric effective approximation, labeled for convenience as GEMA, but to provide expressions that will accordingly simplify and correct calculations of several optical properties of multilayer structures such as reflectivity, transmissivity, absorptivity, absorption coefficient, skin depth, optical conductivity and the dielectric functions, once the optical constants of the composite layers are determined. As the optical constants of single layers of Cu and Ni layers are available in the literature [4, 5], the two materials, layered in a stack, become an appropriate pick for such a test study. Therefore, several Cu-Ni ultrathin stacks of 100 Å periodicity (Cu – 45 Å and Ni – 55 Å) are the subject of the present study. To limit the scope of this study, we present calculations of only the reflectivity and transmissivity of several Cu-Ni layered stacks and their effective optical constants. Calculations of the reflectivity and transmissivity are done using a computer program that has been developed and frequently employed in different investigations by the first author and colleagues [6–8]. Calculations of the reflectivity and transmissivity are executed via the characteristic matrix technique [9], employed in three ways. In the first the characteristic matrix of one Cu-Ni bilayer is calculated; and then the characteristic matrix of a stack of m bilayer units is obtained from a direct multiplication of this matrix into itself m times. In this method no approximation is involved. The second is based on the calculation of the characteristic matrix of the bilayer identity period using the effective complex index of refraction of the identity period as derived according to the CEMA [1]. The third is based on the equivalent characteristic matrix of the whole stack represented by one characteristic matrix, also using the CEMA [1]; in this third method the equivalent optical constants of the layered structure are also calculated. All calculations are for normal incidence and are carried out in the visible range of the spectrum.

The next section outlines a brief account of three formulations for calculating the reflectivity and transmissivity of the targeted stacks; all of these formulations are based on the well known characteristic matrix technique. The rational is to enhance the use of the characteristic matrix technique for ultrathin multilayer stacks through the CEMA [1]. With the values of effective optical constants of the identity period in a superlattice stack as calculated according to the CEMA [1], several other optical properties of the multilayer stack beside the basic optical properties (reflectivity, transmissivity, and absorptivity) can be calculated. The input parameters for these calculations are the thickness of the individual composite Cu and Ni layers, values of the indices of refraction and extinction coefficients of the Cu and Ni layers as reported in literature [4, 5], wavelengths of the covered spectral range, and the number of layers in the stack.

As will be demonstrated in the results section, the criterion for applying the CEMA is found to be tied not only to the thickness of each individual layer being less than or equal to 100 Å for metallic thin films, but to the total number of layers m in the layer structure as well. The condition presents itself in the product of the number of the identity periods m and the thickness of the identity period itself denoted here by h, *i.e.*, mh becomes a quantitative measure of the total thickness for any particular superlattice stack. Section 3 presents the computational results of reflectivity and transmissivity for several multilayer stacks displayed for contrast with the reflectivity and transmissivity of a single bilayer system and the individual composite Cu and Ni layers as well. The effective optical constants of these stacks are also presented. The discussion and conclusions are left for Section 4.

2. Theoretical review

The essential theory of this study is based on the representation of a thin film by a 2×2 characteristic matrix [9]. Denoting the alternate individual layers that form a bilayer by the subscript j ($j = 1, 2$), and assuming that each of the two layers is of a non-magnetic dielectric material, the matrix for each single layer is

$$M_j(h_j) = \begin{bmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{bmatrix}, \quad j = 1, 2 \tag{1}$$

where

$$m_{11} = \cos\beta_j, \quad m_{12} = -\frac{i}{n_j}\sin\beta_j, \quad m_{21} = -in_j\sin\beta_j, \quad m_{22} = \cos\beta_j \tag{2}$$

In the case of normal incidence $\beta_j = (2\pi/\lambda)h_j n_j$. In Eqs. (2), $\hat{n}_j = n_j + i\kappa_j$ is the index of refraction, which in general is complex, and i is the imaginary number $\sqrt{-1}$.

In the characteristic matrix technique, the effective characteristic matrix $\hat{M}(h)$ of two layers of thicknesses h_1 and h_2 and complex indices of refraction $\hat{n}_1$ and $\hat{n}_2$, combined in one bilayer of total thickness $h = h_1 + h_2$ (Fig. 1) is determined from the product of the two characteristic matrices $\hat{M}_1(h_1)$ and $\hat{M}_2(h_2)$; that is,

$$\hat{M}(h) = \hat{M}_1(h_1)\,\hat{M}_2(h_2) \tag{3}$$

or

$$\hat{M}(h) = \begin{bmatrix} \cos\beta_1 & -\frac{i}{n_1}\sin\beta_1 \\ -in_1\sin\beta_1 & \cos\beta_1 \end{bmatrix} \begin{bmatrix} \cos\beta_2 & -\frac{i}{n_2}\sin\beta_2 \\ -in_2\sin\beta_2 & \cos\beta_2 \end{bmatrix} \tag{4}$$

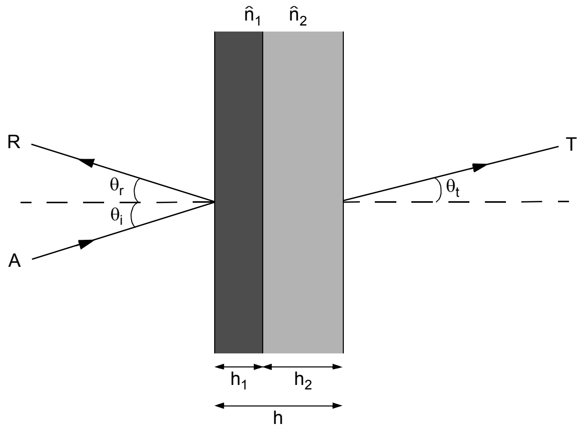


Fig. 1. Schematic of a bilayer that consists of two constituent layers of thicknesses h_1, h_2 and of complex indices of refraction $\hat{n}_1$ and $\hat{n}_2$, respectively.

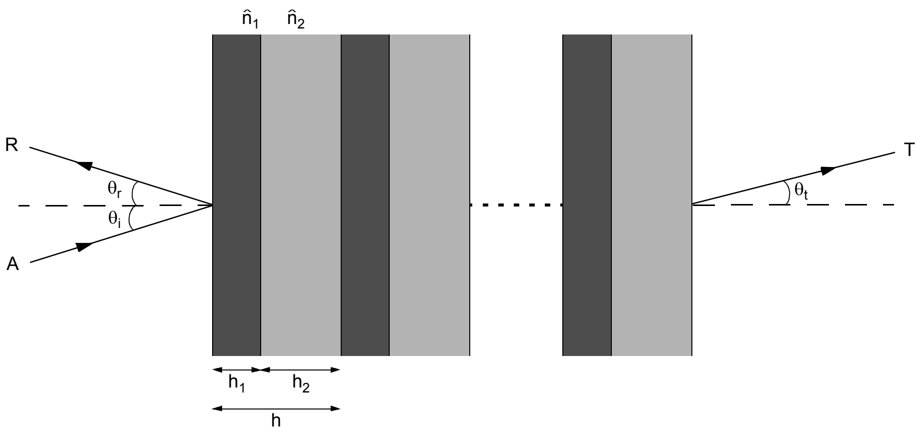


Fig. 2. Sketch of a side view of a multilayer structure of an arbitrary number of basic identity bilayers, each of which consists of two constituent layers of complex indices of refraction $\hat{n}_1$ and $\hat{n}_2$.

For an ultrathin bilayer superlattice stack in which h_1 and h_2 are assumed to be so small compared to the incident wavelength, terms of the order $(h_1 h_2/\lambda^2) \to 0$, and hence they are dropped. Consequently, the characteristic matrix of the bilayer unit in the normal incidence mode, Eq. (4) reduces to the following form [1]:

$$\hat{M}_{\mathrm{eff}}(h) = \begin{bmatrix} 1 & -i\dfrac{2\pi}{\lambda}h \\ -i\dfrac{2\pi}{\lambda}\hat{n}_{\mathrm{eff}}^2 h & 1 \end{bmatrix} \tag{5}$$

where in the above matrix, the complex effective index of refraction of the bilayer is $\hat{n}_{\mathrm{eff}} = n_{\mathrm{eff}} + i\kappa_{\mathrm{eff}}$. By the derivation of the reference 1, the real and imaginary parts of the effective optical constants of the bilayer are inferred to be

$$n_{\mathrm{eff}} = \sqrt{\frac{n_1^2 h_1 + n_2^2 h_2}{h}} \tag{6a}$$

$$\kappa_{\mathrm{eff}} = \sqrt{\frac{\kappa_1^2 h_1 + \kappa_2^2 h_2}{h}} \tag{6b}$$

Simple algebra shows that the two expressions in (6) can be generalized for a thin trilayer or multilayer system as follows:

$$n_{\mathrm{eff}} = \sqrt{\frac{\sum_i^s n_i^2 h_i}{h}} \tag{7a}$$

$$\kappa_{\mathrm{eff}} = \sqrt{\frac{\sum_i^s \kappa_i^2 h_i}{h}} \tag{7b}$$

where $i = 1, 2, .., s$, designates the number of composites of the identity period in the superlattice stack and the total thickness would then be $h = \sum_i^s h_i$.

As was asserted in the introduction, in the present work expressions (6) are applied to one bilayer identity period whose repetition *m* times makes up a periodic Cu-Ni layer system of *m* identity periods.

Finally, the characteristic matrix of a layer stack (Fig. 2) comprised of *m* bilayers as constructed from (2) becomes

$$\hat{M}(mh) = \begin{bmatrix} 1 & -i\dfrac{2\pi}{\lambda}mh \\ -i\dfrac{2\pi}{\lambda}\hat{n}_{\mathrm{eff}}^2 mh & 1 \end{bmatrix} \tag{8}$$

with $m(h/\lambda)^2 \ll 1$. A comparison between Eqs. (8) and (5) shows that the two are very similar except for the thickness *h* of the bilayer identity period as represented by (5) and the thickness *mh* of a layer stack consisting of *m* bilayers each of thickness *h*. The analogy, however, suggests that the designated layer stack is of a complex effective index of refraction equal to that of its basic bilayer unit. This is the basis of the calculations executed in the third method for calculating the reflectivity and transmissivity of the stack as a whole.

3. Results

This study targets the calculation of the optical properties (reflectivity, transmissivity and absorptivity) and the optical constants (effective indices of refraction and effective extinction coefficients). The results that pertain to the optical properties are presented in three sets. The first is the calculation of the reflectivity, transmissivity and absorptivity of the targeted systems using the characteristic matrix incorporating the corresponding optical constants $n(\lambda)$ and $\kappa(\lambda)$, for each of the constituent Cu and Ni layers of the multilayer structure. The thicknesses of these layers are 45 Å and 55 Å, respectively, and their optical constants are adopted from the literature [8, 9].

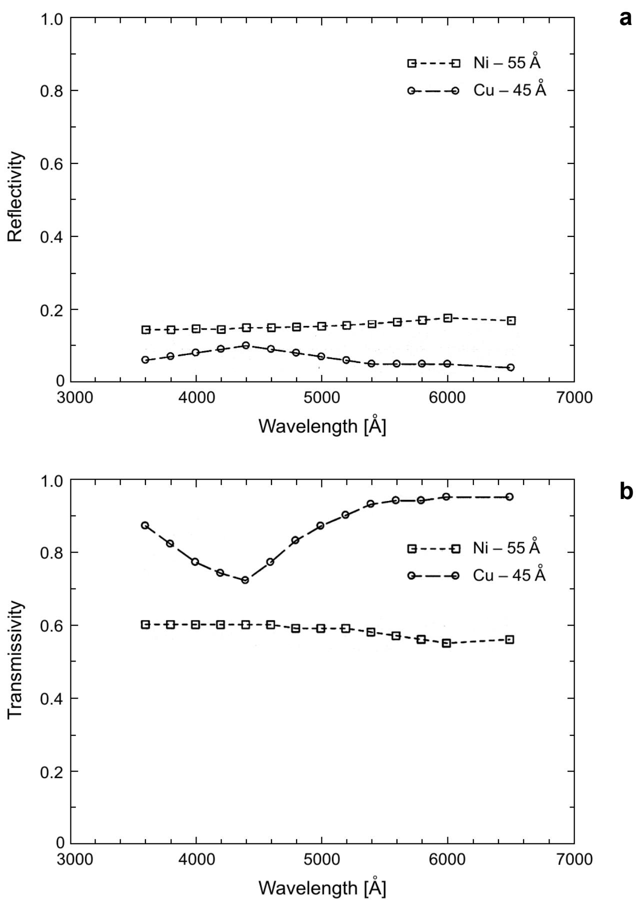
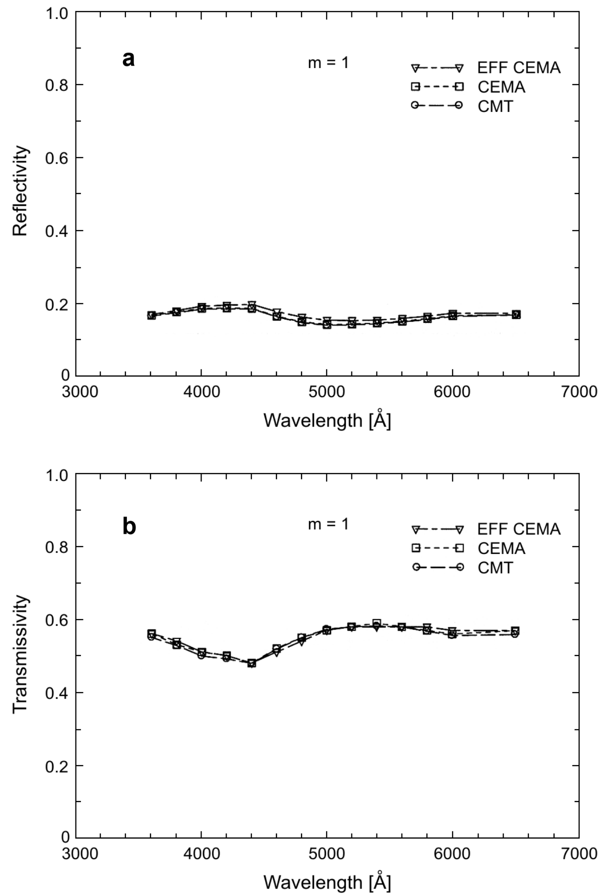
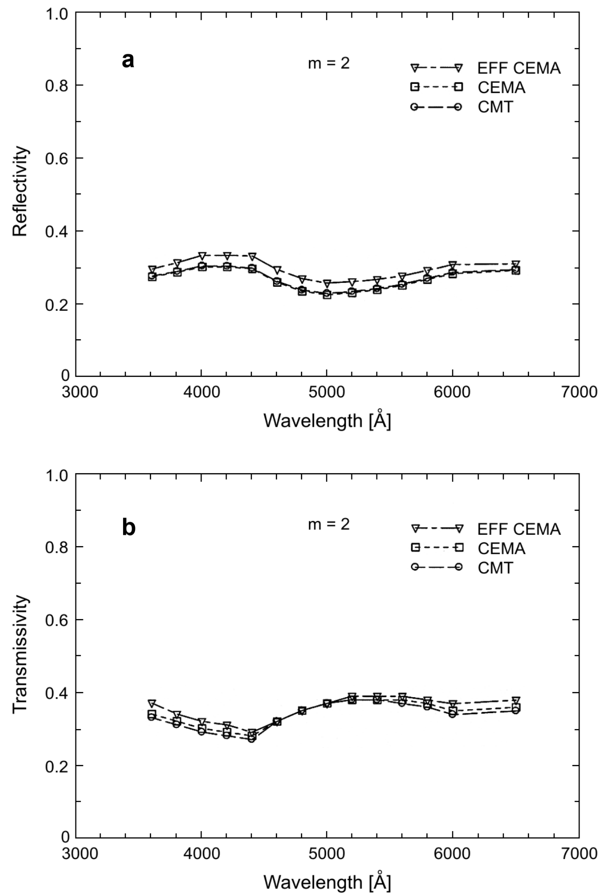


Fig. 3. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) for single layers of Cu (45 Å) and Ni (55 Å) using the CMT.



▲ Fig. 4. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) for one Cu-Ni bilayer identity period displaying the CMT, the CEMA and the EFF CEMA.



▲ Fig. 5. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) in the UV and visible for a 2 Cu-Ni bilayer system displaying the CMT, the CEMA, and the EFF CEMA.

The second set is the calculation of the effective optical constants (effective indices of refraction and effective extinction coefficients) of the identity period Cu + Ni using the CEMA as developed by HAIJA [1] using Eq. (6). The obtained effective optical constants are then incorporated in calculating the reflectivity, transmissivity and absorptivity of these same structures. The third is the calculation of the reflectivity, transmissivity and absorptivity of these structures using the effective optical constants of the whole structure treated as if it were one effective layer of a characteristic matrix as expressed in Eq. (8) and designated as EFF CEMA. The notion EFF CEMA is used to make a distinction between calculations made via the third method and those made via the second method. In both methods the characteristic effective medium approximation CEMA is used. However, in the second method, the characteristic matrix of the Cu-Ni bilayer (Eq. (5)) is calculated on the basis of the CEMA (Eq. (6)), and then this characteristic matrix is raised to a power equal to the number of layers

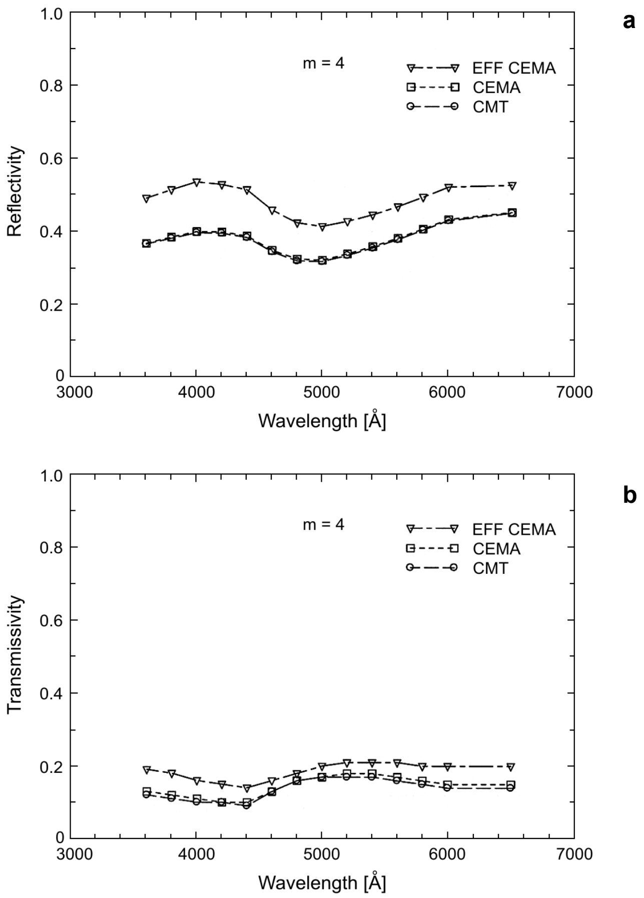


Fig. 6. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) in the UV and visible for a 4 Cu-Ni bilayer system displaying the CMT, the CEMA, and the EFF CEMA.

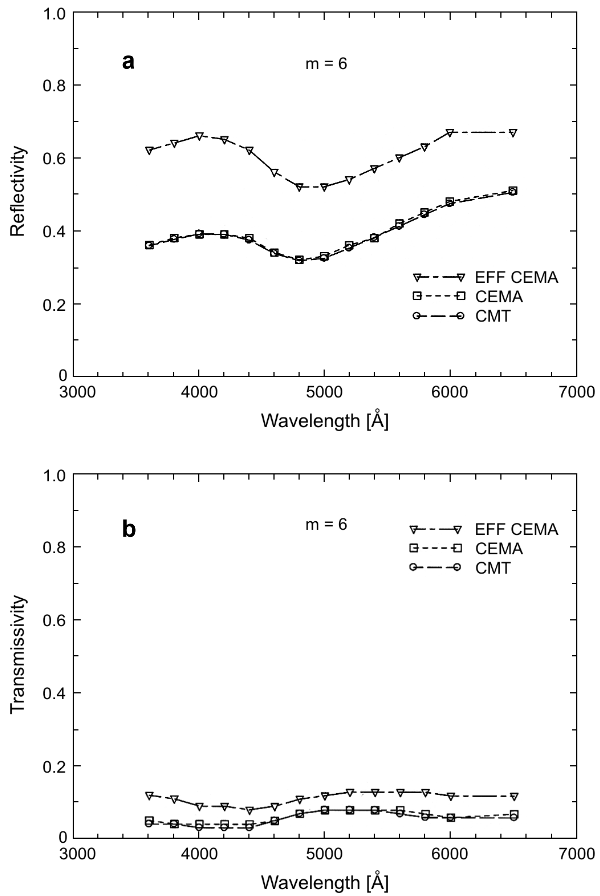


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Fig. 7. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) in the UV and visible for a 6 Cu-Ni bilayer system displaying the CMT, the CEMA, and the EFF CEMA.

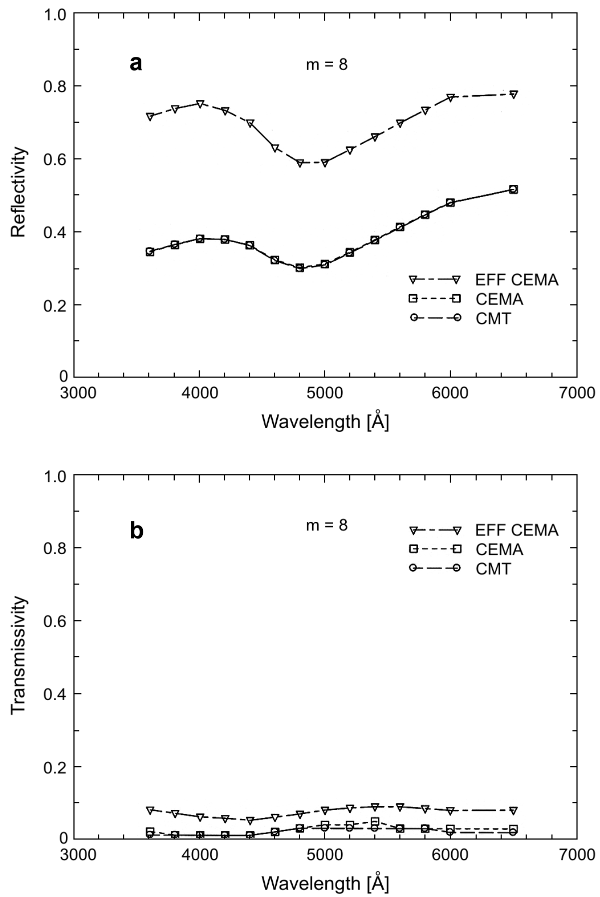

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Fig. 8. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) in the UV and visible for an 8 Cu-Ni bilayer system displaying the CMT, the CEMA, and the EFF CEMA.

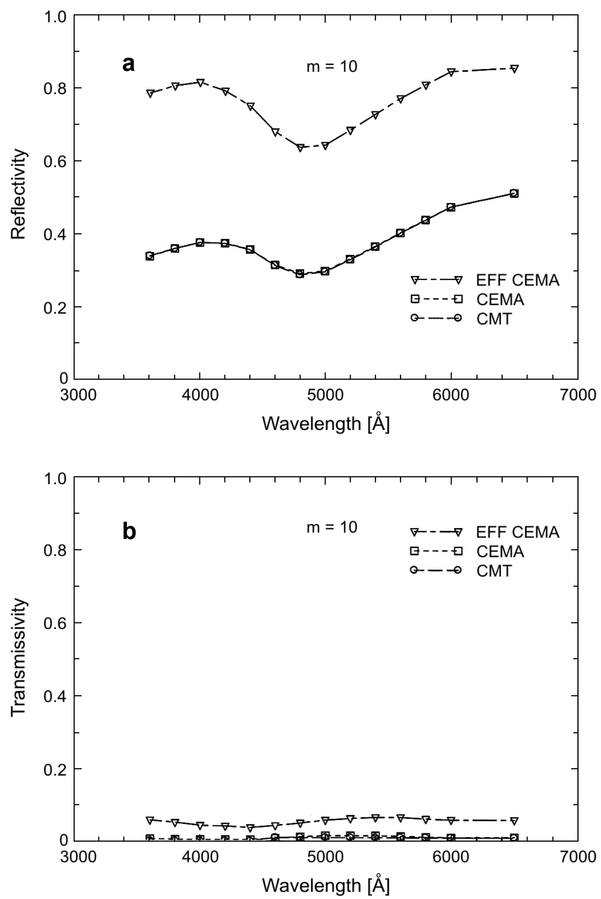

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Fig. 9. Spectral dependence of the reflectivity (**a**) and the transmissivity (**b**) in the UV and visible for an 10 Cu-Ni bilayer system using the CMT, the CEMA and the EFF CEMA.

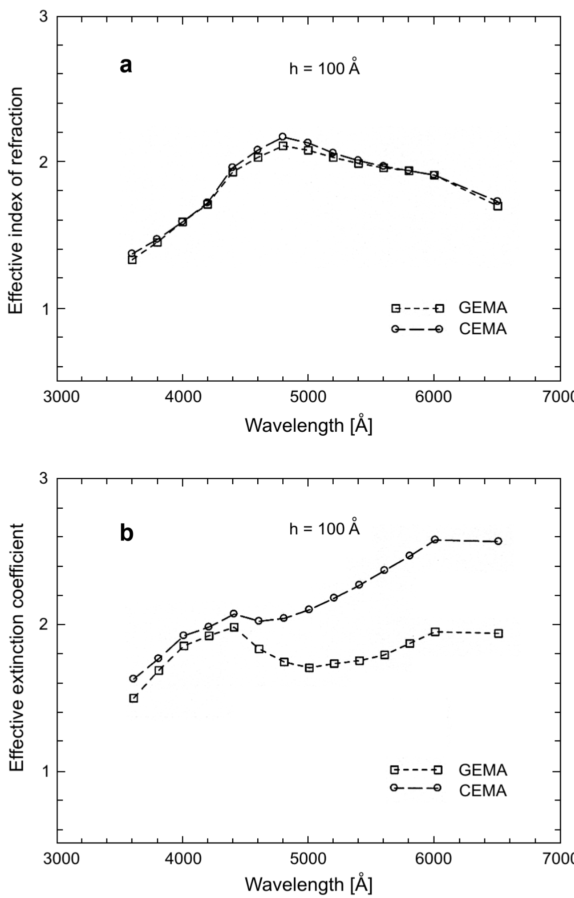

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Fig. 10. Spectral dependence of the effective index of refraction (**a**) and the effective extinction coefficient (**b**) for one Cu-Ni bilayer using the CEMA and the GEMA.

in the stack, while in the third method, the whole stack is treated as one effective layer characterized with one characteristic matrix (Eq. (8)); this effective layer would then be of a total thickness equal to mh, and of a complex index of refraction calculated via the CEMA (Eq. (7)). This third set of calculations is necessary so that one would be able to establish an upper limit on the total thicknesses of the whole structure upon treating it as one effective layer using of the CEMA, a conclusion that is established and elaborated on in Section 4. These calculations for the reflectivity and transmissivity are presented in Figs. 5–9. Figure 3 displays the reflectivity and transmissivity of single Cu and Ni layers applying the characterictic matrix technique (CMT) through the RTA computer program referred to in [6–8]. Figure 3 is depicted to generate the results as computed by this program and contrast them with those published by Shklyarvskii and Pallick. Figure 4 displays the reflectivity and transmissivity of one bilayer identity period. To avoid redundancy, the absorptivity is not sketched. The part that pertains to the optical constants resides in calculating the effective optical constants of the bilayer using both the characteristic effective medium approximation (CEMA) developed in Eq. (6) and the geometric effective medium approximation (GEMA). The results of this part are displayed in Fig. 10.

As Figures 4–9 show, in the CEMA and the CMT calculations for stacks comprised of a number of bilayer identity periods $m = 1, 2, 4, 6, 8$ and 10, the reflectivity and transmissivity are almost identical. Therefore, the CEMA stands as a valid substitute to the ordinary CMT calculations that are carried out without any approximation. The diversion of the CEMA occurs when it is applied to the stack in whole treated as if it were one composite effective layer. The consistency is quite acceptable when the number of layers m is less than or equal to 6. In the studied layer stacks this amounts to a total thickness of 600 Å. It is also worth noting that we found that if the identity period is made thinner, the limit of m beyond which the significant diversion of the EFF CEMA from both the CEMA and the CMT increases to 12 bilayers. Therefore, the criterion for the total thickness is dependent on both the periodicity h of the stack and the number of the identity periods m in the stack such that mh is the proper criterion.

4. Conclusions

As presented in this study, several optical properties such as the reflectivity and transmissivity of several Ni–Cu superlattice stacks are calculated on three bases, one is through multiplication of the characteristic matrices of the individual composite layers, another is through multiplication of the characteristic matrices of the bilayer identity periods in the superlattice using the CEMA and the third is through the effective characteristic matrix of the whole structure treated as if it were one effective layer of effective optical constants, also using the CEMA. In addition, the effective optical constants and dielectric constants, real and imaginary parts of these structures are also calculated.

The values of the reflectivity and transmissivity of the superlattice stacks, calculated according to the first two methods, are practically identical. The third method produces results consistent with the first two methods when the number of layers is less than or equal to 6. However, the divergence becomes significant and increases as the number of layers increases beyond 6. Noting that the periodicity of the handled superlattice stacks was 100 Å, the upper limit on total thickness of the whole stack becomes 600 Å, beyond which the CEMA seems to be invalid. In cases when the periodicity is cut in half, the number of layers in a designated superlattice stack may be increased to 12. In such cases the approximation mh/λ is no longer much less than 1, and the CEMA ceases to be valid. Therefore, the legitimacy of the approximation is established and its use may be generalized for any type of layered materials and periodicity h of superlattices as long as the stated approximation is applicable.

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