

Basic optical properties: effective optical constants, skin depth, absorption coefficients and optical conductivity of an ultrathin Ag-SiO superlattice identity period

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Several optical properties and optical constants of a thin Ag-SiO bilayer of 110 Å, 60 Å – Ag and 50 Å – SiO were calculated. This bilayer constituted the identity period of several ultrathin Ag-SiO superlattice stacks studied earlier. The calculations included the effective optical constants, the skin depth, the absorption coefficients and the optical conductivity of the bilayer unit. The calculations have relied on previous measurements of the basic optical properties of the individual Ag and SiO thin layers. The original measurements that included the reflectivity, transmissivity, and absorptivity of those layers in the UV and visible regions were carried out by the near normal incidence method. All studied samples relevant to this work, the single thin layers and superlattices, had originally been deposited on quartz substrates and prepared in a vacuum of $\sim 10^{-6}$ torr. Analysis of the present results revealed interesting features of an Ag-SiO bilayer that was incorporated in the fabrication of ultrathin Ag-SiO superlattices of whose optical properties were measured and documented in a previous communication.

1. Introduction

The study of various structural, electrical, and optical properties of multilayers and superlattices continues to be one of interest to numerous investigators [1]–[8]. Papers [1]–[8] serve only as examples of concerned researchers in this field. This interest stems from the genuine electronic, electrical and optical properties that may result from the drastically miniaturized band structure, which superlattices usually acquire upon layering. The basic arguments and characteristics that relate to the band structure of superlattices have been exposed widely in literature since the early

seventies [9]–[12]. Therefore, the argument presented here will be limited only to the portions necessary for the understanding and follow up of the objectives and findings of the present article. As a matter of fact, those very same samples, Ag-SiO superlattice stacks, whose identity period is reexamined here from an optical computational vantage, had been pursued in previous efforts by the first author and other colleagues. Those efforts involved the optical and electrical properties of Ag-SiO superlattice stacks; the corresponding treatments and results had been reported earlier in several communications [13]–[15]. In particular, full details of the preparation of those samples, theoretical discussions, measurements, and results can be found in [14] and [15]. While paper [14] will be most useful for learning about the physical dimensions, conditions of preparation, and fabrication of the samples, paper [15] will be most relevant to the objectives of the present work.

In this work, we calculate several optical constants and optical properties of experimentally prepared thin Ag-SiO bilayers. These quantities include the effective optical constants, skin depth, absorption coefficients and optical conductivity of those Ag-SiO bilayers. Such quantities, despite their essential significance, were unfortunately overlooked in the relevant earlier investigations. The present calculations also include the values of the designated optical quantities for an Ag thin film of 60 Å thickness and an SiO thin film of 50 Å thickness. The single film calculations were carried out because these two thin films, being layered upon each other, constituted the basic identity period of each of the Ag-SiO multilayer structures [14], and whose optical properties could be tailored from predetermined properties of the superlattice basic identity period.

As a first step towards extracting the values for the above optical constants and properties, we employed some of the results and calculations obtained and documented in [15]. In other words, all of the quantities that are the subject of this communication are solely and in whole computed from the originally measured basic optical properties: reflectivity R , transmissivity T , and absorptivity A , and from the optical constants n and κ (n is the index of refraction and κ is the extinction coefficient) of the Ag and SiO constituent layers. Therefore, there should be no surprise in the present paper not containing any experimental or measurement sections of its own, simply because such essential and complementary parts did exist fully in the last two references.

1.1. Single thin layers

The starting point in the handling of the data was the computation of the absorption coefficients α for an Ag (60 Å thick) and an SiO (50 Å thick) single thin films in the UV and visible. The covered wavelength ranges were, of course, constrained to the ranges of the raw data, 2400–7500 Å for the Ag thin film [15], and 3000–7500 Å for the SiO thin film [16]. For an easy reference in the forthcoming analysis in Section 2 the raw data, limited only to the optical constants n and κ for the Ag and the SiO thin films available from [15] and [16], are sketched in Fig. 1a, b.

The absorption coefficient α is defined as

$$\alpha = \frac{4\pi n\kappa}{\lambda_0} \quad (1)$$

where λ_0 is the wavelength in vacuum. Concurrently, the skin depth δ is defined as

$$\delta = \frac{1}{\alpha}. \quad (2)$$

This quantity was also calculated for both the Ag and the SiO thin films. Although δ for the single thin layers and the bilayer identity period was calculated, a display of its values, tabular or graphical, is not included. The display of α was seen sufficient and indicative enough of the implied behaviour of the skin depth δ .

The third quantity to be calculated was the optical conductivity

$$\sigma = 2n\kappa\omega \quad (3)$$

where ω is the angular frequency of radiation, n and κ are again the optical constants of the Ag and the SiO thin films. The significance of σ comes from the fact that its knowledge could offer a great deal of understanding of results relevant to the other two optical properties α and δ . Moreover, its knowledge could also offer a reasonable correlation with the d.c. electrical conductivity of these films.

The calculated values of the absorption coefficient, and optical conductivity for the designated Ag and SiO thin films are sketched in Figs. 2 and 4, respectively. Analysis of these values will be reserved for the next section, where all results will be described.

1.2. Ag-SiO bilayers

The three previous quantities, absorption coefficient and optical conductivity for the prescribed bilayers were also calculated. However, as a prerequisite for these calculations, the values of the effective indices of refraction and extinction coefficients had to be provided.

The corresponding optical constants $n(\lambda)$ and $\kappa(\lambda)$ for the Ag-SiO bilayer were calculated from the effective medium method [17] which assumes that when two layers, Ag and SiO, of thicknesses d_1 and d_2 and optical constants n_1 , κ_1 and n_2 , κ_2 , respectively, are superimposed upon each other to form a multilayer system, then each basic identity period in this multilayer system consists of one bilayer (Ag + SiO) with an average effective index of refraction n_{eff} and an average effective optical constant κ_{eff} defined by:

$$n_{\text{eff}} = \frac{n_1 d_1 + n_2 d_2}{d}, \quad (4a)$$

$$\kappa_{\text{eff}} = \frac{\kappa_1 d_1 + \kappa_2 d_2}{d} \quad (4b)$$

where d is the total thickness of the bilayer identity period, *i.e.*, $d = d_1 + d_2$. The method adopted in the present investigation is direct, simple and fits well for normal incidence optical measurements which is the case for the present investigation.

Again, the calculated values of the absorption coefficient and optical conductivity for the constructed Ag-SiO bilayer unit are presented in Figs. 5 and 6. Analysis of these values will also be deferred to the next section, where these results, combined with those of the single layers, will be subject to analytical comparison

2. Results

In this section, all the calculated quantities are presented graphically (Figs. 1–6). Figure 1, which may seem a redundant documentation of previous results [15], [16], is sketched here due to its significance as an entry basis to the whole calculations and to their subsequent analysis. Before we embark on the result description, sorting out these graphs into two sets may be helpful. One set (Figs. 2 and 3) consists of those associated with the Ag and SiO single thin films with the specifications as defined in Section 1. The second set (Figs. 4–6), however, is devoted to the optical quantities as derived and calculated for the Ag-SiO bilayer combination.

Figure 2a, b shows the spectral dependence of the absorption coefficient α of the Ag and SiO thin films, respectively. Again, although for the sake of completeness, the skin depth $\delta = 1/\alpha$ was calculated [18], it was deemed unnecessary to have it displayed in a separate sketch since the displays of δ for both the Ag and SiO thin layers are in essence the reciprocal of those of absorption coefficient α of the same Ag and SiO layers, respectively. As it shows, Fig. 2a demonstrated a sharp minimum for the absorption coefficient at a wavelength of about 3000 Å with a slight structural dependence below 4000 Å wavelengths and an almost smooth decline for wavelengths above 5000 Å. Since the optical properties of the thin Ag coating was among the objectives of our previously documented study [15], comments on its results that are imported here should be helpful in supporting the calculated optical properties of the Ag-SiO superlattice period, of which this Ag layer is a constituent. In this regard, we can say that the calculated values of the absorption coefficient agreed in the orders of magnitude and in features with the results of HAGEMANN *et al.* [19]. For the SiO thin layer the absorption coefficient declined from its highest value at 3000 Å to practically zero at about 3800 Å explaining the considerable absorption of SiO in the UV and its near zero absorption in the visible.

Although some of the optical properties, the optical constants in particular, of thin films of noble metals (copper, silver, and gold) have been reported in literature [20]–[23], an important consideration urged us to measure the optical properties of our, as prepared, Ag single thin film (60 Å thick). Thin films of thicknesses within this range are extremely sensitive to preparation conditions, and the optical constants as

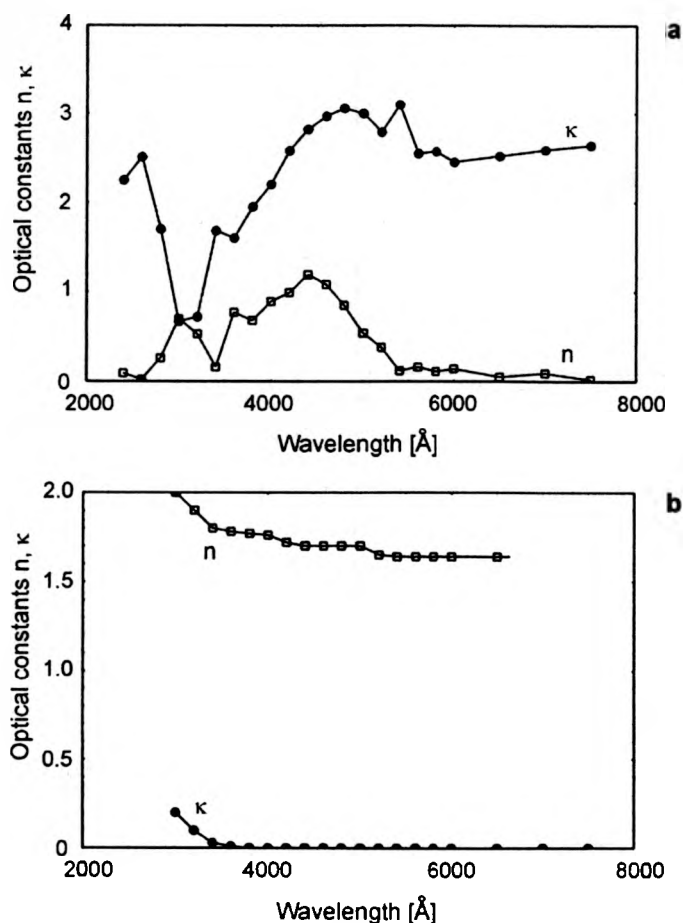


Fig. 1. Spectral dependence of the optical constants in the UV and visible for single thin layers of: Ag, 60 \AA (a) and SiO₂, 50 \AA (b) thicknesses [13], [14].

reported by different workers in the field vary from one to another. Since this Ag film became a primary layer in each identity period in our prepared and confirmed superlattices, preparation conditions, naturally departing from our preparation conditions, could result in unmatched values of the film's optical constants that would then propagate to all subsequent calculations carried out for the bilayer unit. One of course has to bear in mind that it is widely believed that an Ag layer, 60 \AA thick, exhibits a discontinuous structure, and its optical constants are of effective values. Despite this fact, our experimentally measured and cited values for the optical constants of Ag [15] are taken as input values of the Ag layer in the calculation of those of the superlattice period, simply since the Ag layer is one of the constituent primary layers in the presently discussed superlattice period. Since the present two composite layers are the very same layers that formed the X-ray experimentally confirmed parent superlattice stacks, the effective optical constants and optical

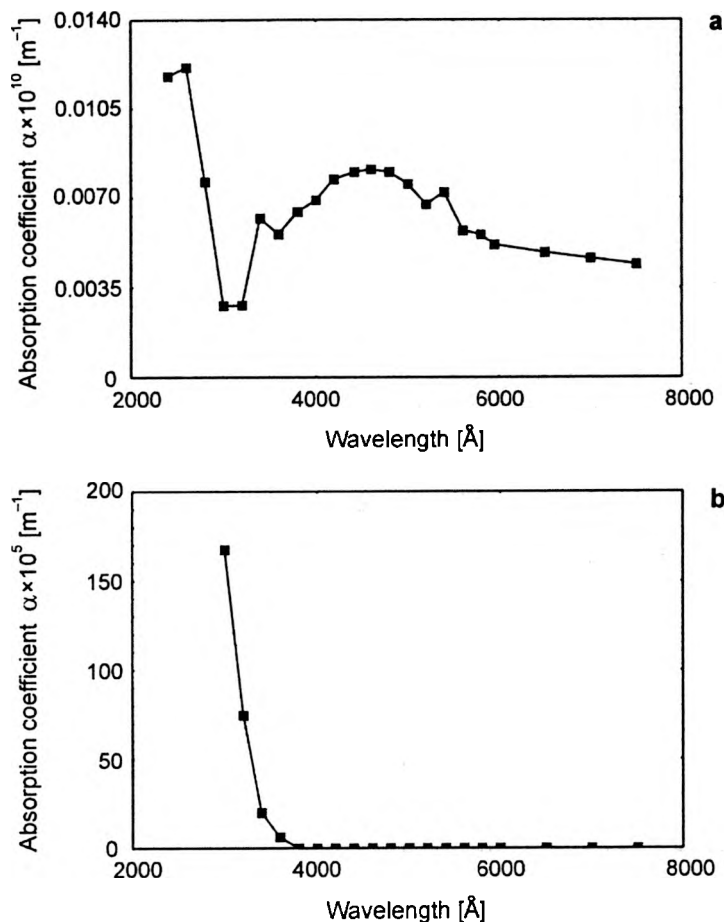


Fig. 2. Spectral dependence of the absorption coefficient in the UV and visible for single thin layers of: Ag, 60 Å (a) and SiO, 50 Å (b) thicknesses.

properties of the bilayer unit, should then inherit the properties of the Ag layer itself as a fundamental layer constituent. In other words, the present work does not single out the Ag thin coating itself as a targeted study, but incorporates its properties as prepared and as we previously measured to explore the output influence it could have on the bilayer itself. These calculations in our opinion could be considered as being more realistic in pertinence to the real multilayer stacks to which this study is relevant.

In Figure 3a, b, the spectral dependence of the optical conductivity $\sigma(\lambda)$ is plotted for the Ag and SiO single thin films, respectively. It is of course needless to say that, aside from the huge difference of the orders of magnitude of σ for the Ag and the SiO thin layers as displayed in Fig. 3 from the values of the absorption coefficient α displayed in Fig. 2 also for the same Ag and SiO single thin layers, Fig. 3 seems to be a replica of Fig 2. Regarding the orders of magnitude of σ , however, the figure shows

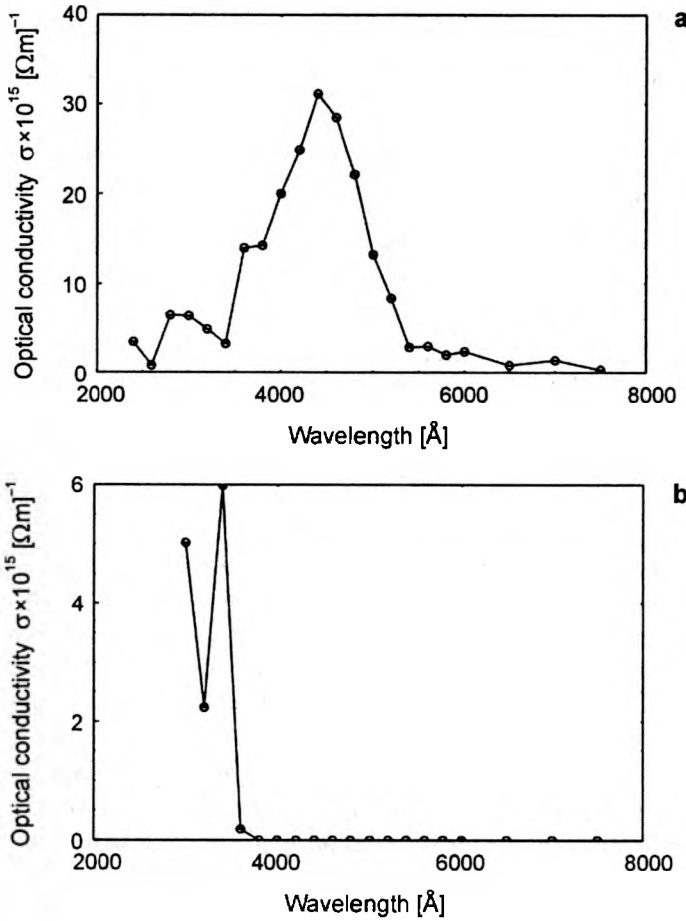


Fig. 3. Spectral dependence of the optical conductivity in the UV and visible for single thin layers of: Ag, 60 \AA (a) and SiO, 50 \AA (b) thicknesses.

that the optical conductivity of Ag was of $\sim 10^{16} (\Omega\text{m})^{-1}$. This is about one order of magnitude less than its value for bulk silver [18]. Such a change from the bulk characteristic value is expected due to the partial dielectric property which thin metallic films are known to exhibit [21], [24], [25]. For the SiO thin layer, σ changed from zero to about $5 \times 10^{15} (\Omega\text{m})^{-1}$. The finite value which σ for SiO had was in the UV, and that was almost zero in the whole visible spectrum. Again, such values are consistent with values known for typical dielectrics. This is in consistence with the values already established for these materials.

In order to obtain the corresponding wavelength dependence of the quantities α , δ and σ for the designated bilayer, its effective optical constants n_{eff} and κ_{eff} were calculated using Eqs. (4). The obtained values of these optical constants are plotted in Fig. 4. As the figure shows there is a noticeable dispersion in the bilayer optical constants in the wavelength range 3000–5800 \AA . The dispersion, although inherited

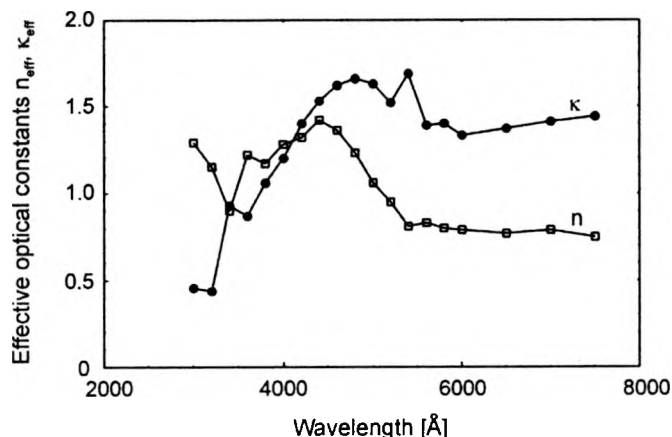


Fig. 4. Effective optical constants of an Ag-SiO bilayer consisting of two distinct layers – Ag, 60 Å and SiO, 50 Å thicknesses.

in part from both of the Ag and the SiO thin layer composites (Fig. 1a, b), is obviously distinct from either one. This supports the concept of the role of layering of thin films that not only it induces a mere superposition of the optical properties but creates new and possibly profound properties of its own. Such an effect should look more obvious in the graphical display of the other rather revealing bilayer optical properties α , and $\sigma(\lambda)$ which are sketched in Figs. 5 and 6, respectively.

In Figure 5 the spectral dependence of the absorption coefficient α of the Ag-SiO bilayer is shown. Here, two main characteristics of this α vs. λ curve deserve comment. First, the magnitudes of α for this bilayer are close but slightly lower than their orders of magnitude for the Ag thin layer. The second concerns the structural features of the

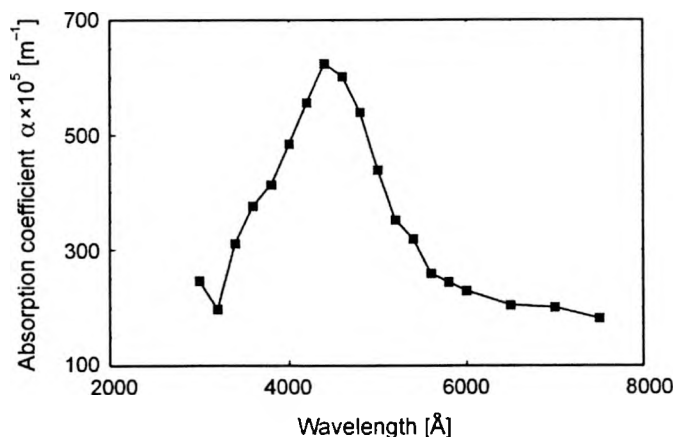


Fig. 5. Spectral dependence of the absorption coefficient in the UV and visible for the Ag-SiO bilayer.

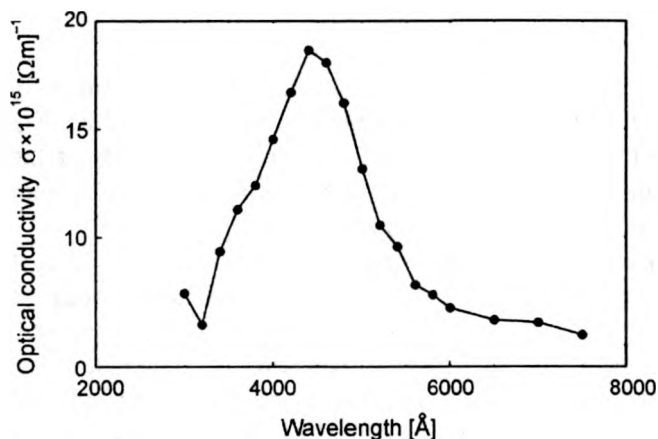


Fig. 6. Spectral dependence of the optical conductivity in the UV and visible for the Ag-SiO bilayer.

dependence of α on λ compared to those of the bilayer components, the Ag and SiO individual layers. In this regard, the wavelength dependence of α over the entire scanned spectral range for the bilayer was found smoother in its features than that for the Ag layer. This should be attributed to a modulating effect of the top SiO layer on the bilayer and consequently on any superlattice structured from those basic bilayer units as was the case of the study documented in [15].

Finally, the spectral distribution of the optical conductivity $\sigma(\lambda)$ for the Ag-SiO bilayer was calculated from the bilayer effective optical constants $n(\lambda)$ and $\kappa(\lambda)$. The results for the bilayer are plotted in Fig. 6. Looking into their values we can record the following remarks. First, the values of the optical conductivity for the bilayer in whole were little less than, but of the same order of magnitude as, those of the Ag thin film (Fig. 3a). However, for the SiO thin film the optical conductivity being almost zero over the whole wavelength range had values that were significant only in the UV and dropped to zero around 4000 Å. This character of the SiO layer affected noticeably the optical conductivity of the bilayer in lowering it by one order of magnitude in the wavelength range above 5800 Å. Most important was the sharp peak in the σ - λ curve around 4400 Å for both the bilayer and the Ag thin film. However, the σ - λ curve for the bilayer had a broader central width than that of the Ag thin film.

Regarding the first remark of no significant change in the values of σ for the Ag-SiO bilayer from those of the thin Ag film, may be interpreted in terms of the relaxation time τ of the conduction electrons in Ag-SiO bilayer as being close to the relaxation time τ in the defined Ag thin film. In other words, the layering effect was far too small to influence the optical conductivity of the system. For any observable change of σ upon layering, the number of layers has to be rather high. In this case, however, such samples tend to become more opaque, and measuring the transmittance then becomes a serious drawback [15].

3. Conclusions

As has been presented, several optical properties, the absorption coefficient, skin depth, optical conductivity, and the optical constants n and κ , of single thin films, Ag, 60 Å thick and SiO, 50 Å thick were calculated. The corresponding values of the previous optical properties and optical constants for an Ag-SiO bilayer fabricated from the just prescribed Ag and SiO layers were also calculated.

The previously defined quantities sketched for the bilayer showed distinct variations in their structural dependence, and to a less degree in magnitude, from the same quantities that corresponded to the single Ag and SiO thin layers. It seemed, however, as if one of the layers, SiO say, modulated the optical property in question of the other layer. This made each bilayer look as a medium being more uniform and homogeneous toward that optical property than either of the bilayer composites, the Ag or the SiO layers. This is quite anticipated from the effective medium method that was applied in this study.

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