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Single and Double-Layer Antireflection Coatings on Silicon

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Abstract

In this study, the effect of single and double-layer thin film coatings on the reflectance spectrum of silicon surfaces has been investigated. Thin film coatings have been prepared by vacuum evaporation of the materials with suitable refractive index. Reflectance measurements in the visible and near infrared have shown that the coated samples acquire antireflection properties. Thickness and wavelength dependence of the antireflection properties of SiO, CeO_2 and ZnS single-layer coatings with optical thickness of one quarter wavelength has been studied.

In double-layer coatings, a thin film of SiO, CeO_2 and ZnS with optical thickness of quarter wavelength were used as inner layer while a thin film of MgF_2 with equal optical thickness was used as outer layer. It was shown that double-layer coatings can produce a significantly broader low reflectance region than does the single-layer coating. The results have been discussed by considering the zero reflectance conditions for the refractive index and the optical thickness of layers.

1. Introduction

Silicon is a semiconductor optical material with relatively high refractive index. It is used in infrared devices as windows, lenses and transmission filters [1-4]. The most important application of silicon in the visible region is photovoltaic solar cells [5-7]. Conversion of solar energy into other energy forms is more effective if the reflectance of light- receiving surface of solar device is minimal in the solar spectrum range [1,5,8]. Efficiency of a solar cell and its lifetime can be raised by coating the light sensitive surface of the cell with an antireflection coating [5,7,9]. This coating reduces the reflectance of the light incident on the cell surface and also protects it from radiations and atmospheric effects.

The high reflection index of silicon causes important reflection loss from its surface, even in thin film form. Therefore, its surface should be coated with an antireflection coating to reduce the reflectance or to increase the transmittance. The principle of the single and multilayer antireflection coatings is based on the destructive interference of light reflected from the interfaces of the coating layers. Theory and design of the antireflection coatings for optical and infrared optical materials may be found, for example, in Refs. [1,5,8,10,11].

The aim of this study is to prepare single and double-layer thin film coatings on silicon surface and to investigate their antireflection properties by spectrophotometric reflectance measurements.

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2. Zero Reflectance Conditions for Antireflection Coatings

A multilayer coating consisting of a thin film stack is shown in Figure 1 [1]. The refractive indices of the surrounding medium and the substrate are $n_o$and n_s, respectively. The geometrical thicknesses d_i and refractive indices n_i of the layers and the Fresnel amplitude reflection coefficients r_i at the interfaces are also shown in the figure. If the materials are all assumed to be nonabsorbing, the refractive indices are real numbers. The substrate and thin film materials are assumed to be both nonabsorbing and nondispersive in the first approximation.

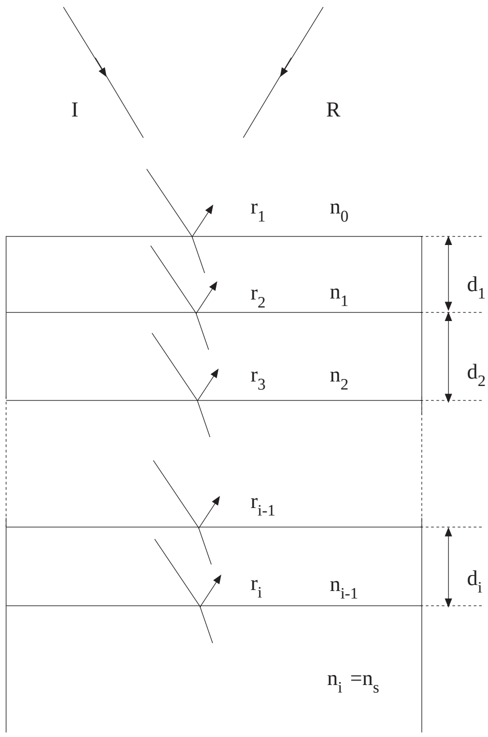

Figure 1. Optical model of a multilayer coating and the notation used for the refractive index, thickness of the layers and the amplitude reflection coefficients at the interfaces.

The reflectance R of a multilayer thin film system is defined as the ratio of the intensity of the reflected beam from the system to that of the incident beam. With this in mind, computation of the reflectance of such a multilayer thin film system will be based on the summation rule for the Fresnel reflection coefficients [1,8].

A single-layer coating is the simplest, both theoretically and experimentally, and is widely used in practice. The necessary and sufficient conditions for a single-layer coating to produce zero reflectance are [1] :

$$n_1 = (n_o n_s)^{1/2}, n_1 < n_s \tag{2.1}$$

and

$$\varphi_1 = (2m-1)\pi/2, m = 1, 2, 3, ... \tag{2.2}$$

where φ_1=2πn$_1 d_1/\lambda$ is the phase thickness of the coating and λ is the wavelength of the incident light. In practice m is usually chosen to be one, and the optical thickness $n_1 d_1$ is then one quarter wavelength. This antireflection coating is known as a quarter-wave coating.

The reflectance of a quarter-wave coating is equal to zero at the wavelength corresponding to the optical thickness of quarter wavelength, if Eq. (2.1) is satisfied. If Eq. (2.1) is not satisfied, then the reflectance will indicate a minimum at the same wavelength. The position of the reflectance minimum of a surface coated with a quarter-wave coating depends on the optical thickness of the coating and shifts towards longer wavelengths as the optical thickness increases.

The effectiveness of a single-layer antireflection coating is limited by the available materials with suitable index. Moreover, zero or minimum reflectance can be obtained at only one wavelength and the reflectance increases rapidly on both side of the zero or minimum position. These difficulties can be largely overcome by using coatings with two or more layers.

The necessary and sufficient index condition for a double- layer coating with equal optical thickness($n_1d_1 = n_2d_2$) to give zero reflectance is [1]:

$$n_1/n_2 = (n_o/n_s)^{1/2} \tag{2.3}$$

or

$$n_1n_2 = n_on_s \tag{2.4}$$

The properties of the reflectance spectra of double-layer antireflection coatings with equal optical thickness depend on the satisfied index condition. When the optical thickness of each layer is one quarter wavelength(i.e., $n_1d_1 = n_2d_2= \lambda/4$) the reflectance spectrum corresponding to Eq. (2.3) has only one minimum at which reflectance is zero. If the index condition of Eq. (2.4) is fulfilled, two distinct minima and a weak central maximum between them appear on the reflectance spectrum. The position of this maximum corresponds to the optical thicknesses of one quarter wavelength of two layers [1]. Double-layer antireflection coatings with equal optical thickness of one quarter wavelength are known as quarter-quarter coatings.

3. Material and Method

Thin film coatings were deposited onto polished silicon sheets of 0.35 mm thickness by thermal evaporation of the selected materials in an Edwards 12 E3 high vacuum coating unit. MgF_2, SiO, CeO_2 and ZnS were chosen as coating materials.

The evaporation source was a tungsten conical basket filament for SiO and CeO_2, and a molybdenum strip for MgF_2 and ZnS. The thickness of the coating layer was controlled by the mass of the material to be eavaporated. The mass of the coating material was estimated from the Knudsen's cosine law about the variation of the deposition rate by assuming the evaporation source is a point source for conical basket filament, or a directed surface source for molybdenum strip [12]. An analytical balance sensitive to 10^{-4} g was used to measure the mass of the material evaporated.

The reflectance spectra of the samples were recorded by Shimadzu UV-160A spectrophotometer which has a reflectance attachment with 5° angle of incidence. Its bandwidth, measurement step and wavelength accuracy is 2 nm, 0.1 nm and ± 0.5 nm, respectively.

4. Results and Discussions

The refractive index n_s of silicon in the region of 400–1100 nm is between 3.5–4.0 [5]. Therefore, when the surrounding medium is air (n_o=1.0), the refractive index of the coating material to be used for a quarter-wave antireflection coating should have a value between 1.87–2.0, according to the index condition in Eq. (2.1). SiO (n_1=1.85), CeO_2(n_1=2.2) and ZnS (n_1=2.3) were selected as coating materials, because their refractive indices are rather close to this index range. The silicon surfaces were coated with thin films of these materials with optical thickness of one quarter wavelength to obtain a minimum reflectance at a wavelength in the range of 400-1100 nm. The indicated wavelength region is important because of its being the sensitivity region for solar cell applications of silicon.

If the MgF_2(n_1=1.38) is selected as outside layer for silicon surface (n_s=3.5), the index condition in Eq. (2.4) for a quarter-quarter antireflection coating requires an index value of n_2=2.54 for the inside layer, in the case of surrounding medium is air (n_o=1.0). SiO (n_2=1.85), CeO_2(n_2=2.2) and ZnS (n_2=2.3) were selected as the coating materials with suitable index. Silicon surfaces were first coated with one of these materials as the inner layer, and then with MgF_2 as the outer layer.

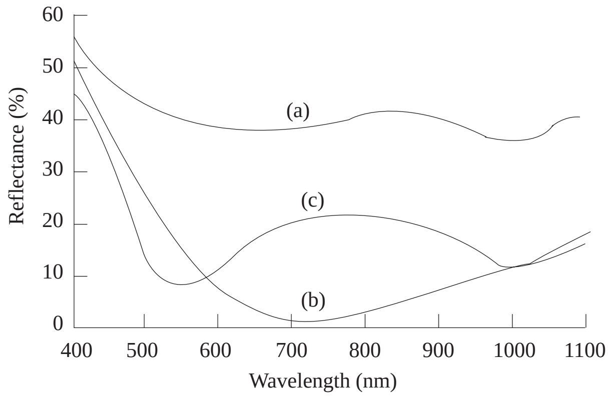

Figure 2. Reflectance spectra of (a) an uncoated silicon surface, (b) a SiO single-layer coating with quarter wavelength optical thickness and (c) a MgF_2+SiO double-layer coating with equal optical thickness of one-quarter wavelength.

Figures 2–4 show the comparison of the measured reflectance spectra of single- and double-layer coatings with that of the uncoated silicon surface. As seen from the (a) curves in Figures 2–4, the reflectance of the uncoated silicon surface is rather high and varies between 56.8% and 37.5% in the considered spectral region. If spectra (a) are compared with spectra (b), it is seen that the reflectance of all the quarter-wave coatings of SiO, CeO_2 and ZnS first decreases rapidly and then increases again after passing a minimum. However, the reflectances of quarter-wave coatings decreased at all wavelengths with respect to that of the uncoated surface. The positions of the reflectance minima give the optical thicknesses of these quarter-wave coatings as 710/4 nm for SiO, 809/4 nm for CeO_2 and 611/4 nm for ZnS. The minimum reflectances are below 5% for all the coatings. The position of the minimum can be shifted to the desired wavelength by varying the thickness of the coating. But, in practice there are limitations due to the dispersion of the refractive indices and absorption in the coating materials.

The reflectance spectrum of MgF_2+SiO quarter-quarter coating on silicon in Figure 2(c) has two minima and a weak maximum between these minima. These observed properties are characteristics theoretically expected from double-layer antireflection coatings with equal optical thickness of quarter wavelength. The minimum reflectances at 540 nm and 1021 nm are 8.9% and 12.6%, respectively. The reflectance of the central maximum at 802 nm is 22.7%. If spectrum (c) is compared with (b), it is seen that this quarter-quarter coating consisting of MgF_2+SiO produces a broader low reflectance region than does the SiO quarter-wave coating. The optical thicknesses of the inner and outer layer of the coating were estimated as $n_1d_1 = n_2d_2$= 802/4 nm, from the position of the maximum in spectrum (c).

The (c) curve in Figure 3 shows the reflectance spectrum of the MgF_2+CeO_2quarter-quarter coating on silicon. Similarly, this double-layer coating has two minima at 601 nm and 997 nm and a weak maximum between them at 808 nm. The minimum reflectances at these wavelengths are 4.9% and 6.9%, respectively. The reflectance value at the weak maximum is 11.7%. The optical thicknesses of the layers, estimated from the position of the maximum, are $n_1d_1 = n_2d_2$=808/4 nm. The second maximum, observed near 450 nm, has been evaluated as an interference maximum. Two reflectance minima in spectrum (c) are higher than the minimum reflectance in spectrum (b). However, it is clear that a MgF_2+CeO_2 quarter-quarter coating has a broader low reflectance band with respect to that of a CeO_2 quarter-wave coating.

Furthermore, the reflectance minima in spectrum (c) of Figure 3, obtained for MgF_2+CeO_2 quarter-quarter coating, are lower than that of the MgF_2+SiO quarter-quarter coating in Figure 2. Lower-or zero-reflectance may not be the result of more successful matching of the optical thicknesses of the coating layers, since the working principle of single and multilayer antireflection coatings are based on the destructive interference of beams reflected from the interfaces of the layers. On the other hand, the destructive interference condition is related with the optical path difference between the light beams reflected from the interface of layers. That is, the optical thickness condition for an antireflection coating is only determinative for desctructive interference or minimum reflectance, while the index condition is determinative for the value of the minimum. The lower reflectance minima observed in the spectrum of MgF_2+CeO_2 quarter-quarter coating indicates that the refractive index of CeO_2more closely satisfies condition Eq. (2.4), making CeO_2a more suitable material than SiO.

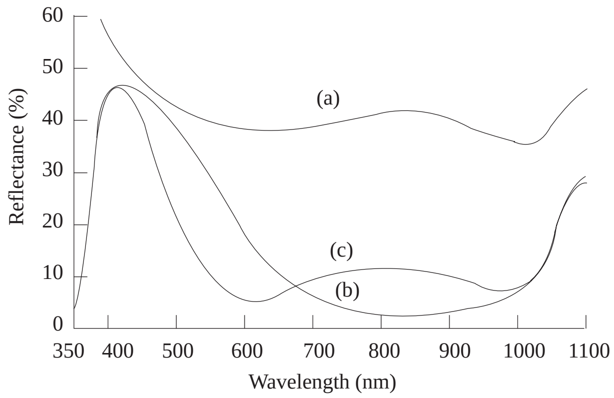

Figure 3. Reflectance spectra of (a) an uncoated silicon surface, (b) a CeO_2 single-layer coating with quarter wavelength optical thickness and (c) a MgF_2+CeO_2 double-layer coating with equal optical thickness of one-quarter wavelength.

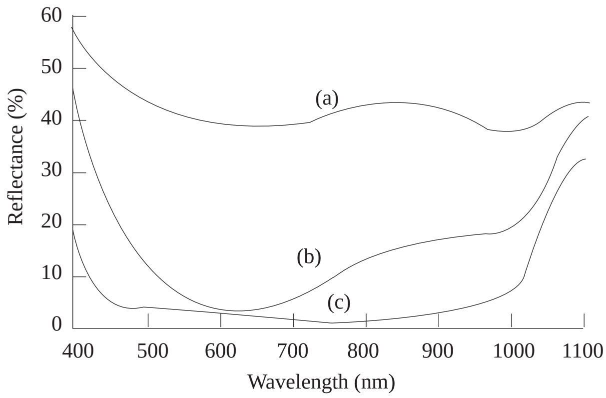

Figure 4. Reflectance spectra of (a) an uncoated silicon surface, (b) a ZnS single-layer coating with quarter wavelength optical thickness and (c) a MgF_2+ZnS double-layer coating with equal optical thickness of one-quarter wavelength.

The reflectance spectra of MgF_2+ZnS quarter-quarter coating is given by curve (c) in Figure 4. The reflectance of this coating first decreases rapidly, beginning from 20% at 400 nm, down to 3% at 450 nm, then decreases slowly to 0.9% at 720 nm. The reflectance then slowly increases through to 1000 nm, after which it rises rapidly.

As can be seen from the comparison of spectrum (c) in Figure 4 with spectra (c) of the other double-layer coatings in Figures 2 and 3, the double-layer coating consisting of MgF_2+ZnS is more efficient in that it exhibits a wider low reflectance region. It also has a lower minimum reflectance compared with spectra (c) of the other double-layer coatings. This can be attributed to the refractive index of ZnS, as it more closely meets the condition of Eq. (2.4). However, the expected maximum between the two minima in the reflectance spectrum of MgF_2+ZnS double-layer has disappeared. This case may be a result of a mismatch between the optical thicknesses of MgF_2 and ZnS layers.

5. Conclusion

Since the refractive index of silicon is relatively high, its surface reflects a high portion of the incident radiation throughout the spectral range between 400 and 1100 nm. The way to reduce this high reflectance is to coat the surface of silicon with at least a single layer of one-quarter optical thickness. SiO, CeO_2 and ZnS thin films have been studied in regard to their use as a quarter-wave coating. It was shown that the reflectance of the silicon surface in the studied spectral range can be reduced by coating its surface with a quarter-wave single layer coating of these materials. The position of the minimum in the reflectance spectra obtained for single-layer coatings can be shifted to the desired wavelength by changing the optical thickness of the coating. But, the minimum reflectance band obtained with these single-layer coatings is not sufficiently wide, as theoretically expected.

A more effective reflectance reduction in a broad spectral region can be obtained with quarter-quarter double-layer coatings, in which the inner layer is a thin film of SiO, CeO_2 or ZnS while outer layer is a thin film of MgF_2. Together the MgF_2+ZnS quarter-quarter coating exhibits a wider low reflectance region and also has a lower minimum reflectance compared with the other double-layer coatings. A mismatch between the optical thicknesses of the inner and outer layer of the coating may effect the properties of the low reflectance band.

References

[1] J. T. Cox and G. Hass, "Antireflection coatings for optical and infrared materials" in Physics of Thin Films, eds. G. Hass and R. E. Thun, vol. 2 (Academic Press, New York, 1968) p.239

[2] G. H. Sherman and P. D. Coleman, *Appl.Opt.*, **10**, (1971), 2675.

[3] J. Stone and L. W. Stulz, *Appl. Opt.*, **29**, (1990), 583.

[4] A. F. Peryeev, K. V. Gudkova, A. A. Poplavskii, R. S. Sokolova, E. I. Fadeeva, M. N. Cherepanova and Z. V. Shirokshina, *Sov. J. Opt. Technol.*, **60**, (1993), 82.

[5] M. M. Koltun, Selective Optical Surfaces for Solar Energy Converters, ed. D. P. Siddons, (Allerton Press Inc., New York, 1981).

[6] G. B. Abdullaev, M. Ya. Bakirov, and N. A. Safarov, *Geliotekhnika*, **29**, (1993), 76.

[7] G. B. Abdullaev, M. Ya. Bakirov, G. M. Akhmedov, N. A. Safarov and F. D. Safarova, *Applied Solar Energy*, **30**, (1994), 12.

[8] A. Musset and A. Thelen, "Multilayer antireflection coatings" in Progress in Optics, ed. E.Wolf , vol. VIII (North-Holland Publ.Co., Amsterdam, 1970) p.203

[9] J. Zhao, A. Wang and M. A. Green, *IEEE Trans. Electron Devices*, **41**, (1994), 1592.

[10] J. C. Monga, *Appl. Opt.*, **31**, (1992), 546.

[11] V. G. Dyskin, Z. S. Settarova and U. Kh. Gaziev, *Applied Solar Energy* , **29**, (1993), 85.

[12] L. Holland, Vacuum Deposition of Thin Films, (Dover Publications, Inc., New York, 1961) p.141