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Optical properties of ZnO/MgF_2 bilayer thin films prepared by PVD technique

Malek A.H. Muhi

Nanotechnology and Advanced Materials Research Center, University of Technology, Baghdad, Iraq

E-mail: malik_muhi@yahoo.com

Abstract

Zinc Oxide (ZnO) is considered as one of the best materials already used as a window layer in solar cells due to its antireflective capability. The ZnO/MgF_2 bilayer thin film is more efficient as antireflective coating. In this work, ZnO and ZnO/MgF_2 thin films were deposited on glass substrate using pulsed laser deposition and thermal evaporation deposition methods. The optical measurements indicated that ZnO thin layer has an energy gap of (3.02 eV) while ZnO/MgF_2 bilayer gives rise to an increase in the energy gap. ZnO/MgF_2 bilayer shows a high energy gap (3.77 eV) with low reflectance (1.1-10 %) and refractive index (1.9) leading to high transmittance, this bilayer could be a good candidate optical material to improve the performance of solar cell window.

Key words
Antireflection coating, thin film, ZnO, MgF_2.

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الخصائص البصرية للاغشية الرقيقة ثنائية الطبقة أوكسيد الزنك / فلوريد المغنسيوم المحضرة بتقنية الترسيب الفيزيائي بالبخار

مالك عبدالحسن محي

مركز بحوث النانوتكنولوجي والمواد المتقدمة، الجامعة التكنولوجية، بغداد، العراق

الخلاصة

يعتبر اوكسيد الزنك (ZnO) واحد من افضل المواد المستخدمة بفاعلية كنافذة على الخلايا الشمسية. وقد ثبت ان الغشاء ثنائي الطبقات المكون من اوكسيد الزنك وفلوريد الغنيسيوم (ZnO/MgF_2) هو اكثر كفاءة كطلاء مضاد للانعكاس من غشاء اوكسيد الزنك احادي الطبقة. لقد تم في هذا البحث ترسيب اغشية رقيقة من اوكسيد الزنك واخرى من طبقة مزدوجة من اوكسيد الزنك وفلوريد المغنيسيوم باستخدام تقنيتي الترسيب بالليزر النبضي والترسيب بالتبخير الحراري. من خلال الفحوصات البصرية وجد ان فجوة الطاقة لغشاء اوكسيد الزنك هي (3.02 eV) في حين وجد انها تساوي (3.77 eV) للغشاء المزدوج من اوكسيد الزنك وفلوريد الغنيسيوم. كذلك وجد ان انعكاسية الغشاء مزدوج الطبقة منخفضة جدا، حيث تتراوح بين (% 10-1.1) وان معامل الانكسار له هو (1.9)، مما ادى الى ان تكون نفاذيته عالية جدا. هذه النتائج اثبتت ان الغشاء مزدوج الطبقات من اوكسيد الزنك وفلوريد الغنيسيوم هو الاكثر ملائمة لاستخدامه كنافذة على الخلايا الشمية لتحسين ادائها.

Introduction

During the recent past years, antireflective coatings (ARC) of various materials have attracted increasing attention as a great importance to get low reflectance solar cell. Various coatings such as ZnS, MgF_2, ZnO have been used in this application. The importance of antireflective coating comes during in solar cell fabrication process which adds a significant improvement in solar cell efficiency [1, 2]. Zinc oxide (ZnO) is a metal oxide of the II–VI compound semiconductor group with n-type conductivity and has band gap energy of 3.37 eV for bulk material at room temperature; the high band gap of ZnO makes it transparent to visible light [3]. Due to the significant properties ZnO is a promising compound for electronic or optoelectronic such as solar cells (anti-reflecting coating and transparent conducting oxides), gas sensors, liquid crystal displays, heat mirrors, surface acoustic wave devices etc [4-6].

In the present work, ZnO and MgF_2 have been chosen to prepare a bilayer antireflective coating. ZnO/MgF_2 bilayer antireflection coating has been investigated because monolayer antireflective coating is not suitable to cover a broad range of the solar spectrum. One of the basic principles indicates that materials with higher refractive index are preferred for the bottom layer while lower refractive index for the upper-layer of a double-layer antireflection coating [7]. Furthermore, the single-layer antireflective coating is achieved lowest reflection for a single wavelength of incident radiation; it must retain [8]:

(a) Square root of the refractive indices of the materials constrained the coating equal to the refractive index of the ARC.

(b) Optical thickness of ARC equal to one quarter of the wavelength.

Therefore, the single layer antireflective coating able to be non-reflective only at single wavelength, it is normally at the middle of the visible spectrum. This problem has solved by using the double layer antireflective coating to cover over the whole visible spectrum are more effective.

Experimental part

Bilayer thin film has been deposited by Physical Vapor Deposition (PVD) method, resistor heating thermal evaporation system has been used to deposit MgF_2 layer and pulsed laser deposition system has been used to deposit ZnO thin layer. Both systems were under high vacuum by rotary and diffusion pumps, Penning and Pirani gauges were used to monitor vacuum in these systems.

Glass slides were used as a substrate for thin films. Before deposition, the substrates were cleaned by deionized water and ethanol. Then, these substrates were dried for about 10 minutes. Both powders ZnO and MgF_2 with the purity approximately 99.999 % were used as sources materials.

The preparation of thin film was divided in two sections:

First layer: ZnO thin films were grown on glass slide substrates using a pulsed laser deposition technique. This film was deposited in vacuum environment about of 1×10^{-5} mbar. The Q-switched Nd:YAG laser was used to deposit with the parameters of laser as number of pulses 200 pulses, wavelength $\lambda = 1064$ nm, energy 700 mJ, repetition rate of 2 Hz and the pulse duration ~8 ns. Furthermore, the distance between the substrates and the target plate was 30 mm, and the substrates heat was maintained at 400 ^{o}C within deposition process.

Second layer: MgF_2 was grown as second layer on ZnO thin film using thermal evaporation technique. Under high vacuum, MgF_2 powder was heated to reach temperature greater than the melting point of this powder by applying current on ends of tungsten boat which was 180 mm away from substrates.

The optical properties of the thin films such as reflectance, transmittance, energy band gap and refractive index were calculated. The transmittance was measured by Metertech Inc. SP-8001 UV-VIS Spectrophotometer in UV–VIS wavelengths ranged from 300 to 1100 nm. The reflectance was measured using Avalight-DH-S-BAL diffused reflectance spectrometer in UV–VIS wavelengths ranged from 250 to 900 nm.

Results and discussion

The optical properties for ZnO thin film and ZnO/MgF_2 thin film deposited on glass substrates were analyzed, the reflectance, absorbance and transmittance have been studied. Additional, The optical energy gap and optical constants have been calculated. Fig.1 shows the reflectance measurements in the range of wavelengths 250–900 nm, both films have lowest reflectance in ultraviolet region. In general, it will be seen that all figures of the optical properties for both tested thin films have the same behavior themselves carves in range 640-1100 nm. However, there are noticed constant different values in y-axis.

In Fig. 1, reflection spectrum of ZnO film shows the minimum reflection of 3 % at 308 nm while reflection spectrum of ZnO/MgF_2 coating shows the minimum reflection of 1.1 % at 301 nm within the wide range of wavelengths (250-900) nm. In additional, the ZnO/MgF_2 reflectance is 10 % within the visible wavelength range. However, there is clear reduction in reflectivity by depositing a second layer.

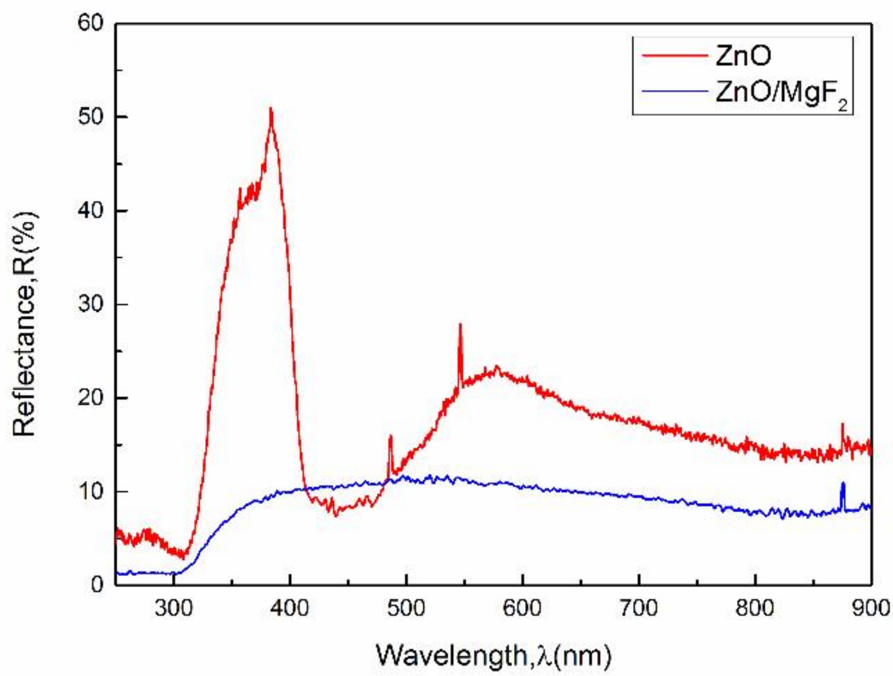

Fig. 1: Reflectance of ZnO and ZnO/MgF_2 thin films.

The results of transmission spectrum were consistent with the results of reflectivity, as shown in Fig. 2. The spectrum of ZnO/MgF_2 coating film is no drop of the total transmittance in the any reign of wavelength range (350-1100) nm but the spectrum of ZnO coating is gradual slope drop in visible wavelength toward ultraviolet. The transmittance spectrum of ZnO/MgF_2 coating in the all wavelength region $\lambda=$ (350-1100) nm is greater than (85%). This average value of transmittance is acceptable comparing with others [9], and it may be due to depend mainly on three factors [9], i.e., (1) oxygen deficiency; because thin film was deposited by pulsed-laser deposition with poor transparencies. (2) surface roughness; scattering ray from surface must be reduction the transmission depending on the grain size, and (3) impurity centers. In this work, the loss of light in ZnO thin film is mainly due to the surface roughness and the oxygen vacancies.

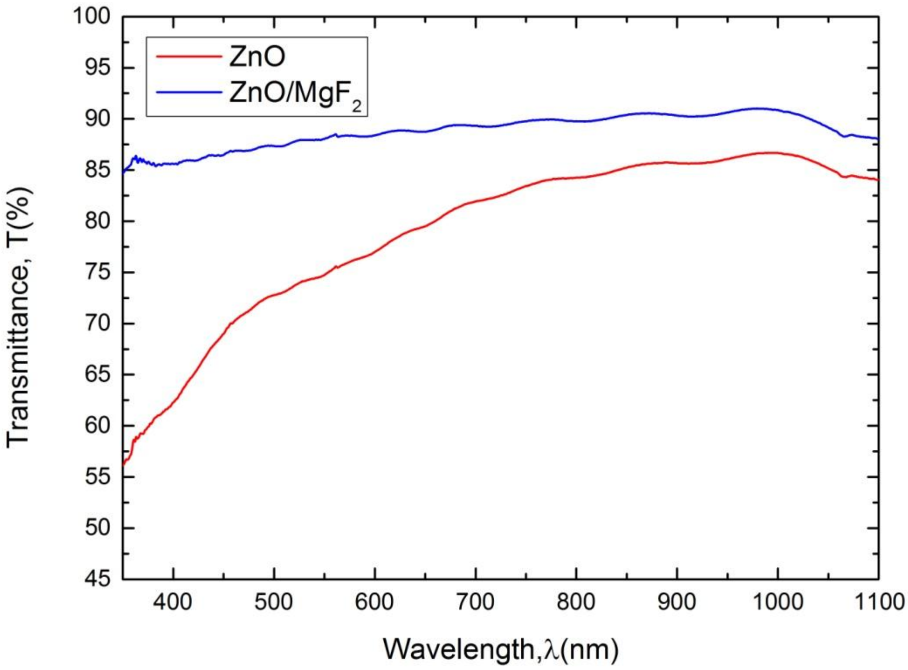

Fig. 2: Transmittance of ZnO and ZnO/MgF$_2$ thin films.

The optical band gaps of the both films were evaluated from Eq. (1) [10]:

$$(\alpha h\nu) = A(h\nu - E_g)^m \qquad (1)$$

where α is the absorption coefficient, A is proportionality constant and E_g is optical energy band gap. m equal to ½ for direct band gap material. The absorption coefficient was calculated using the following expression [10, 11]:

$$\alpha = \frac{1}{t}\ln(T) \qquad (2)$$

where (t) is the thickness of the film and (T) is the transmittance.

The plot $(\alpha h\nu)^2$ versus the photon energy ($h\nu$) were evaluated the direct band gap (Eg) of the ZnO/MgF$_2$ and ZnO thin films which were found to be 3.77 eV and 3.05 eV respectively as shown in Fig. 3. This result, of ZnO thin film, is good agreement with previously reported data of ZnO thin film [12, 13].

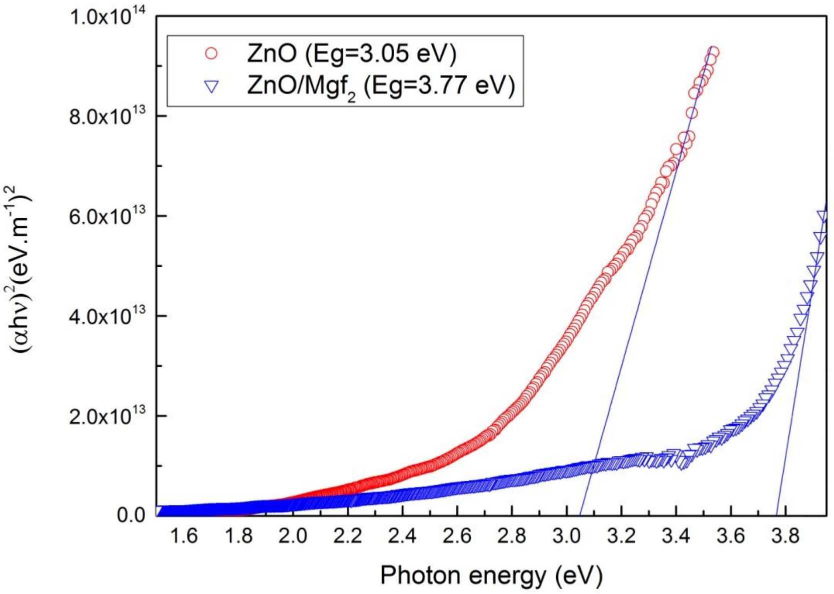

Fig. 3: Energy band gap of ZnO and ZnO/MgF$_2$ thin films.

Fig. 4 shows the refractive index of both Films, and the films have high refractive index (2.5) and (1.95) at wavelength (350 nm) for ZnO and ZnO/MgF$_2$ receptively. The refractive indexes of the both films were evaluated from Eq. (3) [14]:

$$n = \left(\frac{1+R}{1-R}\right) - \sqrt{\frac{4R}{(1-R)^2} - k^2} \quad (3)$$

$$k = \frac{\alpha\lambda}{4\pi} \quad (4)$$

where, R is the reflectance of the film and λ is the wavelength of the incident beam. then the both values decrease to meet them at wavelength 640 nm and reach a value (1.9).

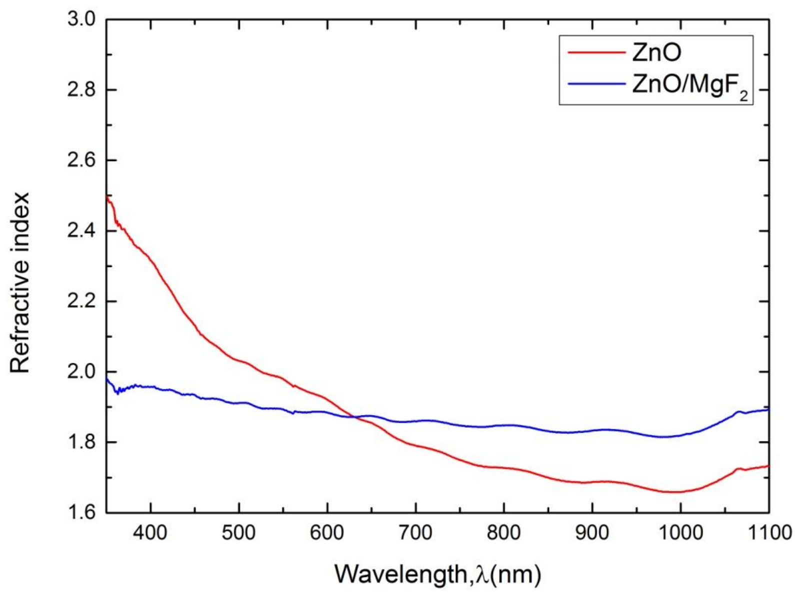

Fig. 4: Refractive index as a function of wavelength for ZnO and ZnO/MgF$_2$ thin films.

The relationship between absorption coefficient α (cm^{-1}) and wavelength shows in Fig. 5. It is noticed that absorption coefficient for ZnO/MgF_2 thin film has nearly constant with wide range wavelength while absorption coefficient of the ZnO thin film is decrease with increase wavelength. Also it can be noticed the value of absorption coefficient for bilyaer thin film is approximately (10^4 cm^{-1}). This may be related to decrease in grain size which causes the light scattering effect for its low surface roughness [15]. This may be explained as the variation of the carrier concentration with respect to the ion-implanted concentration and the increase of the ionized impurity scattering.

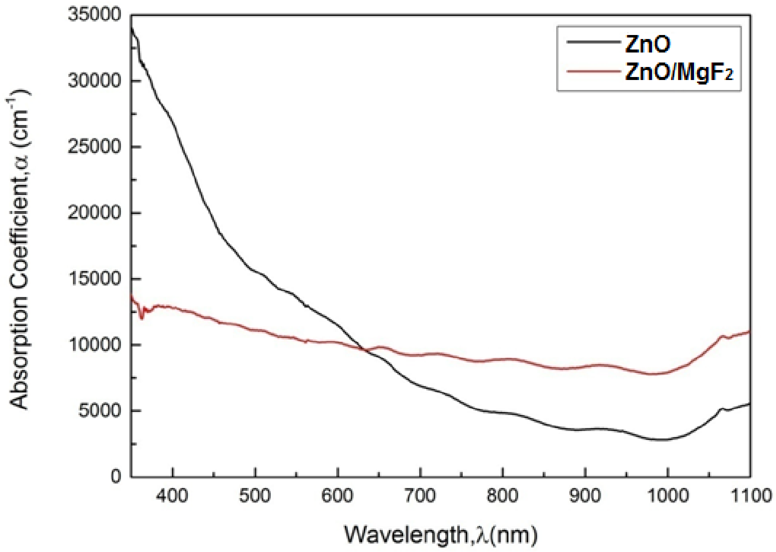

Fig. 5: Absorption confident as a function of wavelength for ZnO and ZnO/MgF_2 thin films.

Fig. 6 shows the extinction coefficient (K_0) as a function of wavelength for two thin films which they were determined from transmitted spectra. The extinction coefficient of ZnO thin film is decrease as the wavelength increases while the bilayer thin film is increases as the wavelength decrease, which is consistent with other reports [16].

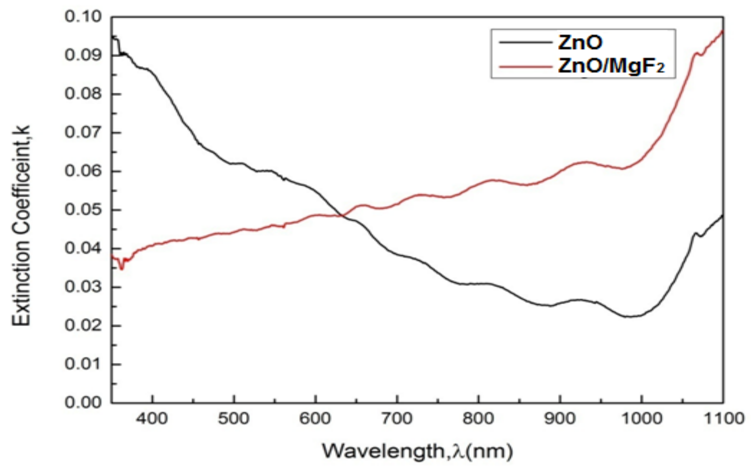

Fig. 6: Extinction coefficient as a function of wavelength for ZnO and ZnO/MgF_2 thin films.

Conclusions

ZnO and ZnO/MgF_2 thin films were successfully deposited into glass substrate using pulsed laser deposition and thermal evaporation deposition as window in solar cell application due to its low reflectance. Present ZnO has low energy gap (3.02 eV) while ZnO/MgF_2 results in an increase of the energy gap. The most promising ZnO/MgF_2 showing the highest energy gap (3.77 eV) with low reflectance (1.1-10%) and refractive index (1.9) leading to high transmittance which is very promising optical material to improve the performance of solar cell window.

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