Study of Nonlinear Refraction and Nonlinear Absorption Coefficients of Different CdS Film Thickness by diode laser

Dawood O. Altaify* Muhammad A. Hazaa**

Received 15 June, 2009
Accepted 16 April, 2012

Abstract:
In the present work, different thicknesses of CdS film were prepared by chemical bath deposition. Z-Scan technique was used to study the nonlinear refractive index and nonlinear absorption coefficients. Linear optical testing were done such as transmission test, and thickness of films were done by the interference fringes (Michelson interferometer). Z-scan experiment was performed at 650nm using CW diode laser and at 532nm wavelength. The results show the effect of self-focusing and defocusing that corresponds with nonlinear refraction $n_2$. The effect of two-photon absorption was also studied, which correspond to the nonlinear absorption coefficient $B$.

Keywords: CdS film, nonlinear refraction, nonlinear absorption

Introduction:
CdS, is an important compound semiconductor has a typical wide band gap of (2.4 eV) at room temperature [1,2]. The CdS compound semiconductors exhibit excellent electrical, chemical and optical properties which make them one of the promising candidates in the field of photovoltaic energy conversion. Direct band gap thin film cadmium sulfide has been the subject of intensive research because of its intermediate band gap, high absorption coefficient, reasonable conversion efficiency and stability. Also it used in light amplifiers, radiation detectors, thin film transistor and diodes, piezoelectric transducers, laser materials and other optical devices [3,4]. The interest in the nonlinear optical properties of semiconductors have become a subject of intensive research for their extraordinary properties compared to their bulk counterparts. The Z-scan method provides a sensitive and straightforward method for the determination of the nonlinear refractive index and the nonlinear absorption coefficient. The simplicity of both the experimental set-up and the data analysis has allowed the technique to become widely used by many research groups [5].

Synthesis of Semiconductor
Chemically Deposition

The process of thin film deposition involves the deposition of material atom-by-atom, molecule-by-molecule, ion-by-ion or cluster-of-species-by-cluster-of-species condensation. This methodology is applied extensively in the manufacture of photovoltaic cells and is being used in optical coating, microelectronics, surface science engineering and other technologies. The technique of chemically depositing thin films has the advantage of being a low cost and applicable to the production of large-area devices. Various aspects of chemically deposited thin films have been reported. The effect of solar radiation and ultrasonic-cation on the various properties of thin films has been

* Institute of Laser for Postgraduate studies, University of Baghdad, Baghdad, IRAQ
** Department of physics, University of Baghdad, Baghdad, IRAQ
reported. Moreover, the effect of varying growth parameters, such as deposition rate, bath composition and bath temperature, on the various properties of thin films have also been reported by several workers. Normally, for obtaining (CdS) thin films by Chemical bath deposition -CBD- in aqueous solution ammonia is used as a base to adjust the [pH] value of solution, and a ligand to control the precipitation of chalcogenides and hydroxides [3,4].

Optical Nonlinearity
The basic optical properties involved in the light-matter interaction are absorption, which is defined by the absorption coefficient $\alpha$, and refraction, which is defined by the index of refraction $n$. When the material is irradiated, the energy of the absorbed photons makes it possible for the transition from the ground state to the excited state. This gives rise to what we call linear absorption. The further excitation may be possible due to the abundance of incoming photons; this gives rise to what we call nonlinear absorption. The absorption of the material $(\alpha)$ is intensity dependant given by [6,7]:

$$\alpha = \alpha_0 + \beta I .... (1)$$

where, $\beta$: the nonlinear absorption coefficient related to the intensity, $\alpha_0$: linear absorption coefficient.

There is also a change in the refractive index when a material is placed in a strong electric field. In fact, the index of refraction becomes dependant on the intensity of the electric field. At high intensity, the refractive index is given by [6,7]:

$$n = n_o + n_2 I .... (2)$$

where, $n_2$: the nonlinear refractive coefficient related to the fluence, $n_o$: linear refractive index. The coefficients $n$, $\alpha$ are related to the intensity of the laser.

The Z-Scan Technique
The Z-scan technique is a simple and popular experimental technique to measure intensity dependent nonlinear susceptibilities of materials. In this method, the sample is translated in the Z-direction along the axis of a focused Gaussian beam, and the far field intensity is measured as function of sample position. Analysis of the transmitted intensity versus sample position Z-scan curve, predicated on a local response, gives the real and imaginary parts of the third order susceptibility. In this technique the optical effects can be measured by translating a sample in and out of the focal region of an incident laser beam. Consequently increases and decreases in the maximum intensity incident of the sample produces wavefront distortions created by nonlinear optical effects in the sample being observed. The z-scan is obtained by moving the sample along a well-defined, focused laser beam, by varying the aperture in front of the detector, one makes the z-scan transmittance more or less sensitive to either the real or imaginary parts of the nonlinear response of the material, i.e., nonlinear refractive index and nonlinear absorption, respectively. The z-scan method is an experimental way to obtain information of nonlinear refractive index and nonlinear absorption properties of materials. With nonlinearity, the intensity dependent response of the material, which can be used to obtain an optical limiting device, either by nonlinear refraction or absorption. The nonlinear refractive index is calculated from the peak to valley difference of the normalized transmittance by the following formula [7]:

$$n_2 = \Delta \Phi_0 / \Phi_0 \text{, eff, k } .... (3)$$
where, $\Delta \Phi_0$ :- nonlinear phase shift, $k = 2\pi / \lambda$, $\lambda$ :- is the wavelength of the beam. $L_{\text{eff}}$ :- the effective length of the sample.

The coefficients of nonlinear absorption can be easily calculated from such transmittance curves. The total transmittance is given by [7]:-

$$T(z) = \sum_{m=0}^{\infty} \left[ \frac{\beta L_{\text{eff}}}{1 + (Z/Z_0)^2} \right]^m \quad \ldots(4)$$

where, $Z$: - is the sample position at the minimum transmittance, $z_0$: the diffraction length, $m$: - integer $(0, 1 \ldots \infty)$. $T(z)$: - the minimum transmittance. The two terms in the summation are generally sufficient to determine the nonlinear absorption coefficient $\beta$.

**Materials and Methods:**

**Sample Preparation**

CdS films were prepared from cadmium sulfate and thiourea by chemical bath deposition in alkaline solution. Different bath concentrations were used in different composition. The deposition of CdS films is achieved from dilute solutions. Sulphide ions are released in the bath by the hydrolysis of thiourea, in the presence of $OH^-$ ions. Cd2+ ions are complexed with one or more of the complex agents like NH3 [directly added as NH3 (aq)], or NH4Cl. This ensures slow release of Cd2+ ions in the solution [6, 8, 9].

$$\text{Cd(NO}_3\text{)}_2 + H_2O + \text{NaOH} \rightarrow \text{NaNO}_3 + H_2O + \text{Cd}^{2+}$$

$$(\text{NH}_2)_2\text{CS} + \text{OH}^- \rightarrow \text{CH}_3\text{N}_2 + H_2O + \text{HS}^-$$

$HS^- + \text{OH}^- \rightarrow H_2O + S^{2-}$

$\text{Cd}^{2+} + S^{2-} \rightarrow \text{CdS}$

**Sample Testing**

**Transmission and Thickness Measurement**

The CdS thin film was tested using UV-visible spectrophotometer for measuring the transmission.

The thickness of CdS film was measured by Michelson interferometer. The film thickness $t$ can be determined by the following formula [10]:

$$t = \frac{\Delta x \times \lambda}{x \times 2\pi} \quad \ldots(5)$$

where, $x$: - the fringe width, $\Delta x$: - the difference in fringes width, $\lambda$: - the wavelength of laser.

**The Z-Scan System**

Z-scan measurements were performed in two parts, closed and open aperture. Each part was employed at 650 nm and at 532nm. Figure (1) shows the set-up of the Z-scan system.
Fig. (1) z-scan system

Where,
1: Diode laser, 2: Lens, 3: Sample, 4: Aperture, 5: Detector, 6: beamsplitter

**Results and Discussion:**

**Transmission and Thickness Measurements**

The optical transmissions of the CdS film were analyzed using UV-VIS spectrophotometer. Fig. (2) shows the transmission spectrum of the CdS thin film and the substrate. At lower wavelength, the transmission is limited by the absorption in the CdS film. The layers thickness were measured by Michelson interferometer method. The layers thickness were found to be (200μm, 300μm, and 500μm) film thickness is equal to (2 μm, 2.5 μm, and 6 μm) respectively.

**Nonlinear Optical Properties Results**

z-scan measurements were done at 650 nm and 532 nm including, the closed-aperture and open-aperture Z-Scan.

**Nonlinear Refractive Index**

Figs. (3,4) show the closed-aperture Z-scan results to investigate the nonlinear refractive index at different layer thickness. A valley followed by a peak is the hallmark of a positive $n_2$. The peak to valley profile displayed in the figures, demonstrates the sample exhibited a self-focusing effect, i.e., it has a positive nonlinearity. The external self-focusing arising from the Kerr effect in CdS, which appears in the peak and valley transmittance of each of Z-scan trace. The closed-aperture Z-scan defines variable transmittance values, which is used to determine the nonlinear phase shift $\Delta \Phi$ using equation (3) and the nonlinear refractive index. This can be shown in table (1).
Table (1) Nonlinear refractive index and nonlinear phase shift at two wavelengths

<table>
<thead>
<tr>
<th>Wavelength λ</th>
<th>Thickness µm</th>
<th>T&lt;sub&gt;max&lt;/sub&gt;</th>
<th>T&lt;sub&gt;min&lt;/sub&gt;</th>
<th>ΔΦ</th>
<th>n&lt;sup&gt;2&lt;/sup&gt; cm&lt;sup&gt;2&lt;/sup&gt;/MW</th>
</tr>
</thead>
<tbody>
<tr>
<td>650nm</td>
<td>200</td>
<td>0.12</td>
<td>0.065</td>
<td>1.375</td>
<td>1.144</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>0.11</td>
<td>0.058</td>
<td>1.3</td>
<td>1.6224</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>0.06</td>
<td>0.04</td>
<td>0.5</td>
<td>1.04</td>
</tr>
<tr>
<td>532nm</td>
<td>200</td>
<td>0.26</td>
<td>0.16</td>
<td>2.5</td>
<td>1.7024</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>0.14</td>
<td>0.05</td>
<td>2.25</td>
<td>2.29824</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>0.04</td>
<td>0.02</td>
<td>0.5</td>
<td>0.8512</td>
</tr>
</tbody>
</table>

The values of n<sub>2</sub>, corresponds to the change in film thickness, and the wavelengths used. As a comparison, the value of n<sub>2</sub> is at maximum value at thickness of 300µm. This behavior is more enhanced at 532.

**Nonlinear Absorption Coefficient**

In order, two wavelengths were considered 650 nm and 532 nm. Figs. (5,6) show the open-aperture Z-scan to investigate the nonlinear absorption coefficient at 650 nm at different layer thickness. The transmittance is sensitive to the nonlinear absorption as a function of input intensities. The change in transmittance is caused by two-photon or multiphoton absorption in the samples as it travels through the beam waist [11,12]. A symmetric valley is contributed to the positive nonlinear absorption coefficient β, indicating the two-photon absorption; this property is more enhanced at 532 nm. The open-aperture Z-scan defines variable transmittance values, which used to determine absorption coefficient. This can be shown in table (2).

Table (2) Nonlinear absorption coefficients at two wavelengths

<table>
<thead>
<tr>
<th>Wavelength λ</th>
<th>Thickness µm</th>
<th>T&lt;sub&gt;min&lt;/sub&gt;</th>
<th>β (cm/GW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>650nm</td>
<td>200</td>
<td>0.1</td>
<td>9.473</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>0.11</td>
<td>10.072</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>0.09</td>
<td>8.857</td>
</tr>
<tr>
<td>532nm</td>
<td>200</td>
<td>5.4</td>
<td>194.184</td>
</tr>
<tr>
<td></td>
<td>300</td>
<td>5</td>
<td>181.616</td>
</tr>
<tr>
<td></td>
<td>500</td>
<td>2.8</td>
<td>110.767</td>
</tr>
</tbody>
</table>

The value of β depends on the change in film thickness, the wavelengths used. The nonlinear effect is more enhanced at 532 nm, which in turn leading to the increase in nonlinear effect at the focal point. This was attributed to the higher intensity in the transmission process.
**Conclusions:**
In the present work, the conclusions that can be extracted from the experimental results are summarized as follow:-

The nonlinear optical properties of CdS nanoparticles are independent of the pulse energy (Kerr nonlinearity), this property is more enhanced at 532 nm with the same results (but at less effect) at 650 nm. This was attributed to enhance the self-focusing and the two-photon absorption.

---

**Fig. (2)** The optical transmission of CdS films deposited onto glass substrate

**Fig. (3):** Closed-aperture Z-Scan at 650nm
Fig. (4): Closed-aperture Z-Scan at 532nm

Fig. (5) Open-aperture Z-Scan at 650nm

Fig. (6) Open-aperture Z-Scan at 532nm
References:
8. Ezema F. I. and Okeke C. E., 2003 ”Chemical Bath Deposition Of Beryllium Sulphide (BeS) Thin Film And Its Applications” AOIJ, 9; 1-18.
دراسة معامل الأنكسار ومعامل الأمتصاص اللاخطيين لغشاء مختلف السمك من كبريتيد الكادميوم باستعمال ليزر الدايد

داود عبيد الطيفي

محمد عيال هزاع

معهد الليزر للدراسات العليا / جامعة بغداد – العراق

كلية العلوم للبنات / قسم الفيزياء / جامعة بغداد – العراق

الخلاصة:
في هذا الجزء العملي، تم تحضير أنموذج من كبريتيد الكادميوم على شكل أفلام مختلفة السمك. استخدمت تقنية المسح على المحور الثالث (Z-Scan) لدراسة الخواص البصرية اللاخطية مثل معاملات الأنكسار والامتصاص اللاخطية. تم تحضير العينة باستخدام تقنية الترسيب الكيميائي الحراري، ثم أجريت فحوصات بصورية خطية مثل فحص النفاذية وتم قياس سمك الافلام بطريقة تداخل الأهداب (Michelson interferometer). تم أجراء تجربة المسح على المحور الثالث باستعمال ليزر مستمر بطولين موجيين عند (650) نانومتر و (532) نانومتر. أظهرت النتائج تأثير ظاهرة التبؤر الذاتي للمادة عند الطاقات العالية المرتبطة بمعامل الأنكسار اللاخطي (n2). فضلا عن ذلك تم دراسة تأثير عملية الامتصاص الثاني أو المتعدد الفوتون (B) للمادة الذي يرتبط بمعامل الأمتصاص اللاخطي.

معيد الليزر للدراسات العليا / جامعة بغداد – العراق

كلية العلوم للبنات / قسم الفيزياء / جامعة بغداد – العراق