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Magnetic Field and Temperature Dependent Measurements of Hall Coefficient in Thermal Evaporated Tin-Doped Cadmium Oxide Thin Films

O.A. Hamadi[1], **N.J. Shakir**[2], **F.H. Mohammed**[3]

[1]Department of Physics, College of Education, The Islamic University, Baghdad, Iraq
[2]School of Applied Sciences, University of Technology, Baghdad, Iraq
[3]Department of Physics, College of Science, Almustansiriya University, Baghdad, Iraq

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Abstract. CdO:Sn thin films are deposited onto glass substrates by thermal evaporation under vacuum. The studied films are polycrystalline and have an NaCl structure. The Hall effect is studied for films with different thickness as substrates are maintained at different temperatures. The temperature dependence of the Hall mobility is also investigated.

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1 Introduction

The semiconducting cadmium oxides attract a great attention due to their electrical and optical properties. Films prepared from pure and metal-doped CdO have been widely studied for extensive optoelectronic applications in transparent conducting oxides (TCO), solar cells, smart windows, optical communications, flat panel displays, photo-transistors, as well as other types of applications like IR heat mirror, gas sensors, low-emissive windows, thin-film resistors, *etc.* [1-5].

Generally, CdO films are transparent in visible and NIR spectral regions and have n-type semiconducting properties with an electrical resistivity of 10^{-2}–10^{-4} Ω cm [1,6] and a band gap in-between 2.2 eV and 2.7 eV [7–9]. The procedure of preparation and the kind of the metal dopant are responsible for those different values of the conductivity and the variety of the optical properties. The conduction of pure CdO is attributed to its native defects of Oxygen vacancies and Cadmium interstitials. Therefore, the conductivity of CdO films

can be controlled by those native defects through the procedure of film preparation including doping with metallic ions. Moreover, these electrical properties can be improved by reducing the effect of depletion regions formed on grain boundaries that can be done by employing various treatments with Hydrogen including low-temperature post-annealing [10-12]. The present study focuses on the improvement of the dc-electrical conduction properties and the variation in the optical absorption properties of CdO thin films because of low-temperature post-annealing in Hydrogen atmosphere at different durations.

The cadmium oxide (CdO) is an important semiconductor material for the development of various technologies of solid-state devices, (panel display, opto-electronic components, thermally insulating glass, *etc*.) [1-3]. Some electrical properties of this oxide have been investigated by several authors [4-9]. The experimental data concerning the Hall effect of CdO are rather poor. A variety of methods has been used to prepare thin films of cadmium oxide, such as thermal evaporation, oxidation of cadmium films, spray pyrolysis, metalorganic chemical deposition, plasma-induced bonding, *etc*. [5-9]. It was experimentally found that the electronic transport and optical properties of CdO thin films strongly depend on the preparation method and deposition conditions [7-11]. On the other hand, we remark that in the above-mentioned papers on the electronic transport in CdO and CdO:Sn thin films, the experimental data were discussed with respect to a relative, small number of samples prepared under different conditions. Consequently, it is difficult to compare the results presented by different authors. It was experimentally established that the stable structure of films can be obtained if, after preparation, they are submitted to a heat treatment [12,13].

In this paper, the Hall coefficient in CdO:Sn thin films is investigated as a function of deposition conditions, magnetic induction and film temperature.

2 Experiment

CdO:Sn thin films were deposited onto glass substrates by physical vapour deposition under vacuum of high purity CdO and Sn. The experimental conditions were settled for obtaining films of a uniform thickness on large areas of the substrate surface. The samples under study were prepared using the following deposition parameters: the source evaporator-substrate distance was $D = 50$ mm, the source temperature was $T_\nu = 1100$ K; the deposition rate was $r_d = 10$ μm/s; the substrate temperature $T_S = 300$–500 K. The source temperature was monitored by a Pt/Pt-Rh thermocouple. A special holder for substrates was made of Aluminium and permits to prepare four samples simultaneously. A chromel-alumel thermocouple, which monitored substrate temperature, was attached to the front surface of the substrate.

The film thickness was measured by using multiple-beam Fizeau's fringe method [7] at reflection of monochromatic light, $\lambda = 550$ nm. The X-ray

diffraction (XRD) patterns of the films were recorded with a DRON-2 diffractometer operating with CuKα radiation ($\lambda = 1.5418$ μm). For measurement of the Hall coefficient and its temperature dependence a special arrangement has been used [12]. The Indium thin film electrodes were used. The Hall voltage was determined by a standard DC potentiometric method. Two Keithley instruments (Model 6517 electrometer and a Model 487 picoampermeter) were used for measurements. The Hall coefficient was calculated from the following expression [14]:

$$R = \frac{U_H d}{BI}\,, \tag{1}$$

where U_H is the Hall voltage, d represents the film thickness, B is the magnetic induction and I denotes the intensity of the total current which passes through the film.

The magnetic induction (produced by an electromagnet) was measured by a teslameter with Hall sonde. The temperature dependence of the electrical conductivity, σ, was studied using surface-type cells. The optical bandgap of CdO:Sn thin films was determined from the optical absorption spectra. The absorption coefficient, α, was calculated from the expression [15]

$$\alpha = \frac{1}{d} \ln \frac{(1-R)^2}{T}\,, \tag{2}$$

where d is the film thickness, and R is the reflection coefficient and T represents the transmission coefficient.

The reflection and the transmission spectra (in the spectral range from 400 nm to 1800 nm) were recorded using a Zeiss, Jena spectrophotometer and an ETA-STO spectrometer.

3 Results and Discussion

A linear dependence of the voltage, U_H, as a function of magnetic induction, B, has been observed for films with different thickness, deposited onto the substrates maintained at different temperatures during the film growth (Figures 1–3). It is known that in polycrystalline thin films the electronic transport mechanism is influenced by the intercrystalline boundaries. This mechanism is based upon the consideration that the crystallite boundaries have an inherent charge region due to the interface. Consequently, energy band bending occurs, and potential barriers to the transport of charge carriers result. For investigated samples, the activation energy of electrical conduction, E_a, calculated from the temperature dependence of the electrical conductivity can vary from 0.30 to 0.70 eV. These values strongly differ from energy gap, E_g, of CdO crystals (the values of E_g reported by different authors for cubic CdO crystals are, usually, ranged between 2.70 and 3.20 eV [2,4,10]).

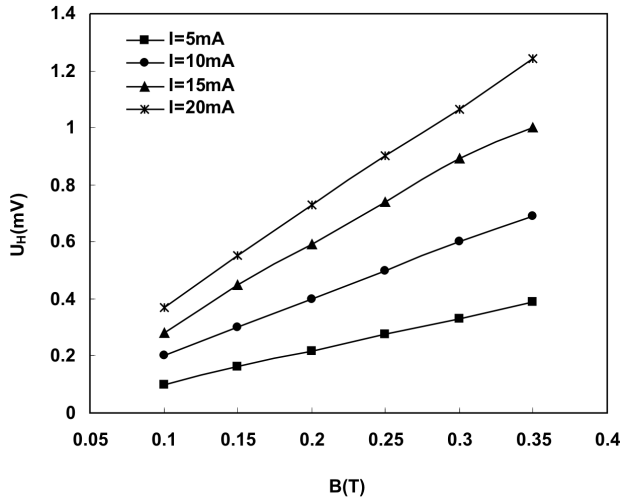

Figure 1. Hall voltage *vs.* the magnetic induction for sample A21, $d = 0.22$ μm and $T_S = 473$ K.

Clearly, that the conduction mechanism in the investigated samples can be explained by applying the models developed for the semiconducting discrete (polycrystalline) structure [14,19]. Seto's model [1,20] could explain the electronic transport mechanism in ZnTe, Sb_2O_3 and Sb_2S_3 polycrystalline films. On the

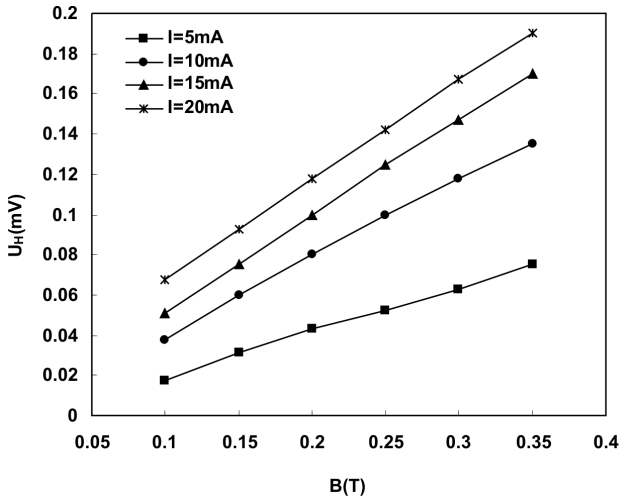

Figure 2. Hall voltage *vs.* the magnetic induction for sample A23, $d = 0.68$ μm and $T_S = 473$ K.

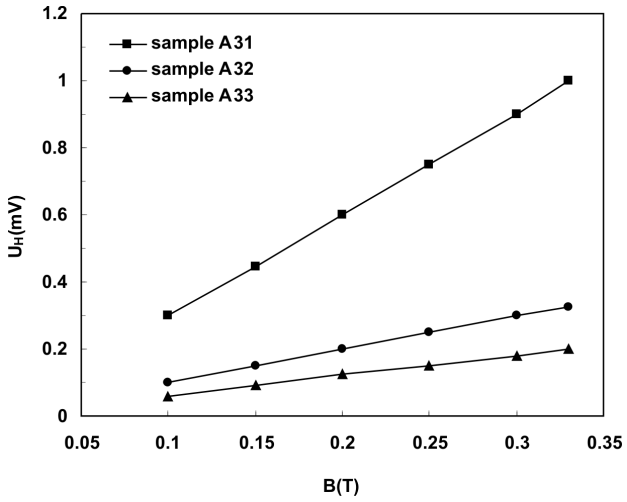

Figure 3. Hall voltage *vs.* the magnetic induction: sample A31 ($d = 0.19$ μm), sample A32 ($d = 0.46$ μm), sample A33 ($d = 0.62$ μm), $T_S = 523$ K and $I = 12.5$ mA.

other hand, the linear dependence of the Hall voltage, U_H, as a function of magnetic induction, B, (Figures 1–3) shows that in the CdO:Sn thin films, the boundary domains little influence on the carrier transport in magnetic field.

We consider that the presence of tin crystalline (metallic) precipitate in the intercrystallite domains determines an important decrease of the electrical resistivity of these domains. The crystallite size and film thickness can also influence the Hall coefficient. One of the well-known models accounting for the Hall coefficient in the semiconducting thin films is that developed by Amith [21]. According to this model, the effective Hall coefficient, R_H, is related to the Hall coefficient intracrystalline domains R_{Hb} through the expression

$$R_H = R_{Hb}\eta\Big(\frac{\lambda}{d}\Big), \tag{3}$$

where $\eta(\lambda/d)$ is a function depending on the ratio λ/d (λ denotes the mean scattering length and d is the film thickness).

For smaller values of λ/d ($\lambda/d < 2$), the function $\eta(\lambda/d) \cong 1$ and R_H is little influenced by the film thickness and mean scattering length. For bulk CdO crystals the mean scattering length has lower values ($\lambda \cong$ 10–15 nm). Figure 4 shows that the Hall coefficient does not depend on the magnetic induction B. This behavior is justified, because the condition $\mu^2 B^2 \ll 1$ (where μ is the carrier mobility) is true for the investigated samples ($\mu = 1.2 \times 10^{-5}$–7.5×10^{-5} m^2/(Vs)).

The temperature dependence of the Hall coefficient is presented in Figures 5–

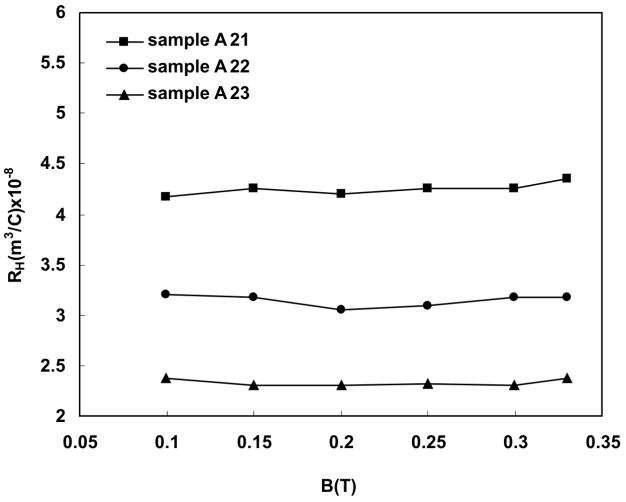

Figure 4. Hall coefficient *vs*. the magnetic induction: sample A21 ($d = 0.22$ μm), sample A22 ($d = 0.51$ μm), sample A23 ($d = 0.68$ μm), $T_S = 523$ K and $I = 12.5$ mA.

6. An exponential increase of the Hall coefficient with increasing temperature is observed in all investigated temperature ranges. Clearly, the extrinsic conduction prevails in studied samples. Impurity atoms (tin atoms) introduce energy levels within the energy gap and act as donors. The activation energy of the impurities, E_d, calculated from the relation $\ln(R_H T^{3/2}) = f(10^3/T)$, generally ranged between 0.020 eV and 0.070 eV.

In Figure (7), the dependence of the Hall mobility on the temperature is illustrated for three studied samples. The Hall mobility μH has been estimated by means of the expression

$$\mu_H = \frac{|R_H|}{\rho}\,, \tag{4}$$

where R_H is the Hall coefficient and ρ denotes the electrical resistivity.

The carrier mobility due to lattice vibrations can be expressed by [16]

$$\mu_L = A_L m_{\text{eff}}^{-5/2} T^{-3/2} \tag{5}$$

and the charged ionized centers affect mobility as follows [16]:

$$\mu_I = A_I m_{\text{eff}}^{-1/2} T^{-3/2} \tag{6}$$

where A_L and A_I are the characteristic parameters, m_{eff} denotes the scalar effective mass of the charge carriers and T is the absolute temperature.

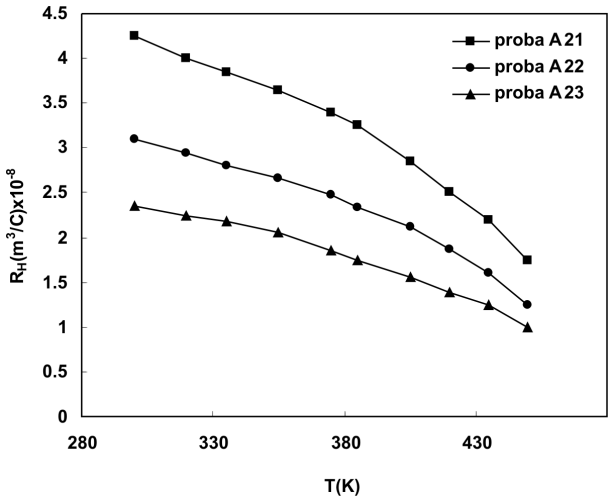

Figure 5. Temperature dependence of the Hall coefficient for films with various thickness: sample A21 ($d = 0.22$ μm), sample A22 ($d = 0.51$ μm), sample A23 ($d = 0.68$ μm), $T_S = 473$ K and $B = 0.22$ T.

In the polycrystalline thin films, free carriers are scattered by the crystallite boundary surface in addition to the scattering mechanisms observed in the re-

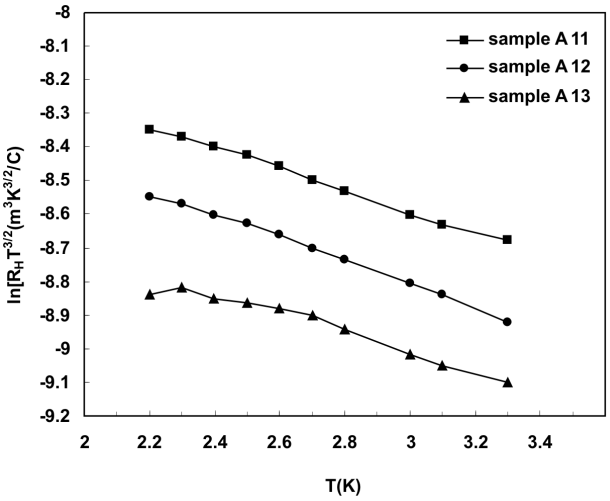

Figure 6. Temperature dependence of the Hall coefficient for films with various thickness: sample A11 ($d = 0.17$ μm), sample A12 ($d = 0.39$ μm), sample A13 ($d = 0.57$ μm), $T_S = 293$ K and $B = 0.22$ T.

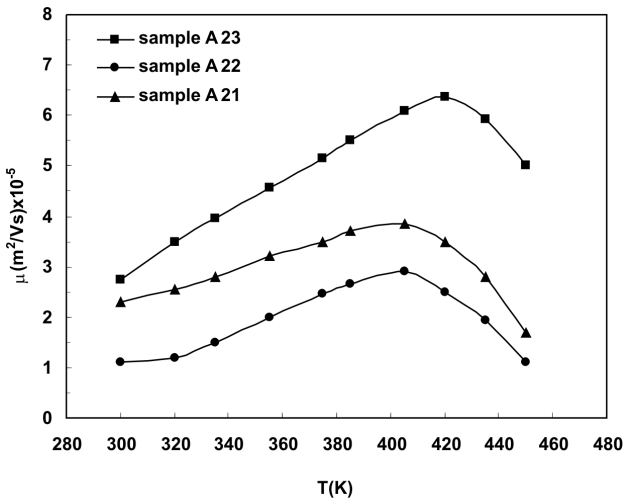

Figure 7. Temperature dependence of the Hall mobility for films with various thickness: sample A23 ($d = 0.68$ μm), sample A22 ($d = 0.51$ μm), sample A21 ($d = 0.22$ μm), $T_S = 473$ K.

spective bulk materials. Figure 7 shows temperature dependence of Hall mobility for three studied samples. It can be observed that these dependences present two parts. We suppose that in the first parts the carrier scattering by ionized impurities (tin donors) prevails. According to Eq. (6), in this case an increase of carrier mobility with temperature is observed. The decrease of mobility in higher temperature is caused by lattice scattering of charge carriers. Also, the large value of the carrier concentration determines a decrease of the mobility [16].

4 Conclusion

From the results obtained in this work, we can conclude that the Hall voltage linearly depends on the magnetic induction. In the studied temperature range (300–500 K), an exponential increase of the Hall coefficient with increasing temperature is observed. An important characteristic of absorption spectra for polycrystalline semiconducting films is an additional maximum (compared to single crystals) for photon energies less than the band gap. At higher photon energy the absorption spectra are little influenced by the polycrystalline structure of the films.

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