

Investigations of the drift mobility of carriers and density of states in nanocrystalline CdS thin films

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ABSTRACT

Nanocrystalline Cadmium Sulfide (nc-CdS) thin films have been prepared on well-cleaned glass substrate at room temperature (300 K) by thermal evaporation technique using inert gas condensation (IGC) method. X-ray diffraction (XRD) analysis reveals that the films crystallize in hexagonal structure with preferred orientation along [002] direction. Scanning electron microscope (SEM) and Transmission electron microscope (TEM) studies reveal that grains are spherical in shape and uniformly distributed over the glass substrates. The optical band gap of the film is estimated from the transmittance spectra. Electrical parameters such as Hall coefficient, carrier type, carrier concentration, resistivity and mobility are determined using Hall measurements at 300 K. Transit time and mobility are estimated from Time of Flight (TOF) transient photocurrent technique in gap cell configuration. The measured values of electron drift mobility from TOF and Hall measurements are of the same order. Constant Photocurrent Method in ac-mode (ac-CPM) is used to measure the absorption spectra in low absorption region. By applying derivative method, we have converted the measured absorption data into a density of states (DOS) distribution in the lower part of the energy gap. The value of Urbach energy, steepness parameter and density of defect states have been calculated from the absorption and DOS spectra.

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1. Introduction

Thin films of nanocrystalline semiconductors hold great promise in wide range of applications due to their unique electronic and optical properties that can be tuned by varying the size and composition of the nanoparticles [1,2]. Among these, nc-CdS thin films offer large number of applications in solid state technologies such as a window layer in solar cells, Schottky diodes, optical filters, photo detectors etc. [3,4]. A broad variety of well-established deposition techniques including vacuum evaporation [5], spray pyrolysis [6], sputtering [7], and chemical bath deposition [8] have been used to prepare CdS thin films. The vacuum evaporation is known to be the suitable technique for the preparation of nc-CdS thin films [9,10].

In this paper, we have investigated the key quantities such as carrier type, carrier mobility and density of defect states, which directly affect material potential for application in optoelectronic and photovoltaic devices [11]. We first measure the carrier type and electron mobility using Hall measurements and then compared with the electron mobility measurements by Time of Flight

(TOF) technique. For thin film semiconductors, TOF is an established technique that is used for accurate determination of mobility of charge carriers, which in turn may lead to insight about the transport mechanism and structural properties of the materials. In general, the samples used in TOF have two types of electrodes configuration which are sandwiched electrodes configuration (thin film is sandwiched between the electrodes) and coplanar or gap cell electrodes configuration. In sandwiched electrodes configuration, excess carriers of one polarity are allowed to transit through the sample, so it is possible to measure electrons and holes drift mobility separately with this configuration. For using sandwiched electrodes configuration samples thickness should be of the order of few micrometers [12]. On the other hand, there is a bipolar measurement of the transient photocurrent due to electrons and holes in coplanar electrodes configuration. When analyzing gap cell transient photocurrents, the different drift mobility of electrons and holes must be taken into account [13]. Keeping in mind that CdS thin films are used as thin window layers in solar cells, we have performed TOF transient photocurrent in gap cell configuration.

The electrical and optical properties of materials and performance of solar cells depend upon the optical absorption and density of states (DOS) in the sub-gap. For the accurate measurement of the sub-gap absorption and density of defect states, we

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utilized the Constant Photocurrent method (CPM) which have high sensitivity in low absorption region $\alpha d < 1$, here ' α ' is the absorption coefficient and ' d ' the film thickness [14]. There are a number of techniques which are widely used for the measurement of absorption coefficient and density of defect states such as Photothermal Deflection Spectroscopy (PDS) [15], Dual Beam Photoconductivity (DBP) [16], Electron Spin Resonance (ESR) [17] and CPM. The CPM technique has the advantage as compared to other techniques of being insensitive with the surface defects and able to measure both charged and neutral defect densities below the Fermi level [18]. This technique can be configured in two modes which are continuous mode (dc-CPM) [19] or the periodic mode (ac-CPM). In the present work, we have used ac-CPM because of its more sensitivity than dc-CPM [20].

2. Experimental details

For the fabrication of thin films, CdS powder (STREM Chemicals) was used as source material. Thin films were deposited on glass substrate by thermal vacuum evaporation [vacuum coating unit HINDHIVAC, MODEL: VS-65D] using the inert gas condensation (IGC) technique. The substrate cleaning plays an important role in the preparation of thin films. Initially the corning 7059 glass substrates were cleaned with soap solution then with acetone followed by ultrasonic bath with distilled water and methanol. These well cleaned glass substrates were placed on substrate holder inside the chamber. The substrate holder temperature was room temperature (300 K) during deposition of films. CdS powder was placed in the Molybdenum boat positioned at the center of the vacuum chamber. Before deposition, the chamber was evacuated to a base pressure of 2×10^{-5} mbar, and then backfilled with Argon gas atmosphere at pressure of 2×10^{-2} mbar through specially designed inlet tube having a jet diameter of 0.5 mm. The jet was kept adjacent to the boat pointing towards the glass substrate and flow of Argon gas was controlled by needle valve. After passing high current, the material in the boat was turned into molten state and vapors of the material were formed. The vapors come in contact with Argon gas which act as carrier gas and transport the sublimated vapors to the substrate which deposited on the surface of substrate in the form of thin film. The films were kept in deposition chamber in dark for 24 h before taking measurements to attain thermodynamic equilibrium. The structural parameters of the sample were evaluated by X-ray diffraction using a Spinner 3064 XPERT-PRO X-ray diffractometer ($\text{CuK}\alpha = 1.54056 \text{ \AA}$) at a scanning speed of $0.02^\circ \text{ s}^{-1}$ in the 2θ range from 10° to 80° . Optical absorption and transmission measurement were carried out in an UV/VIS/NIR computer controlled spectrophotometer (Perkin Elmer LAMBDA 750) in the transmission range from 400 nm to 2500 nm at room temperature (300 K). FE-SEM graphs were taken with SU-8000 (MODEL: HI-0876-0003) operated at an accelerated voltage of 15 kV. Thin layer of platinum (Pt) was sputtered on all the samples before FE-SEM analysis. The thickness (d) of thin film was measured with the help of 'ImageJ 1.48 v' microscope software using side-view SEM image of sample. TEM micrograph was studied using Transmission Electron microscope Hitachi H7500. In order to perform Hall measurements, Maltese cross-like Indium (In) electrodes were deposited on thin film by vapor deposition (diagonally separated by 0.25 cm each).

The experimental setup for TOF is shown in Fig. 1. In TOF and CPM, coplanar cell configuration was used (separation between electrodes is $w=0.08$ cm). We used a pulsed Nd-YAG laser having wavelength (λ) ~ 1064 nm, pulse width ~ 10 ns and power ~ 10 mJ as a light source. The sample was kept in a special type of metallic sample holder to improve the signal-to-noise ratio because the electrical signal is usually very small [21]. The current

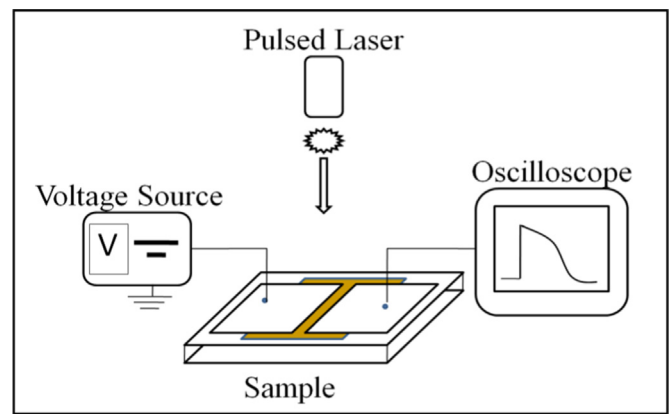


Fig. 1. Experimental setup of the Time of Flight (TOF) transient photocurrent measurement.

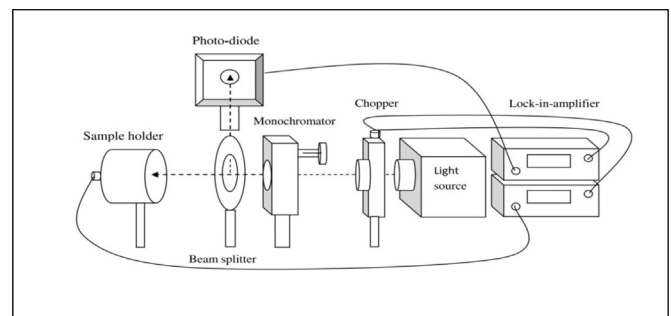


Fig. 2. Experimental setup for ac-CPM.

produced by the laser pulse in the presence of applied electric field was recorded using a Digital Phosphor Oscilloscope (Tektronix DPO 4054; interface software Tekvisa). ac-CPM setup used is shown in Fig. 2. Halogen lamp was used as a light source. The focused light from the light source was allowed to pass through chopper (AMETEK Model 197) chopped at 80 Hz, monochromator and beam splitter (20% reflection) before falling on the sample as shown by dotted line in setup. The intensity of light was varied by changing the power supply knob of the light source. A dc bias voltage of 2 V was applied across the sample. The ac-current and light intensity were monitored by Lock-In-Amplifiers (EG & G DSO 7265).

3. Results and discussion

3.1. Structural, optical and thickness measurement

XRD pattern of nc-CdS thin film deposited on glass substrate at 300 K is shown in Fig. 3. The nc-CdS thin film consists of diffraction peaks at 2θ values of 24.76° , 26.36° , 28.13° and 47.76° respectively, which are identified to be originated from (100), (002), (101) and (103) reflections (JCPDS card No. 75-1545). These diffraction planes correspond to hexagonal phase of CdS crystal. The presence of large number of peaks indicates polycrystalline nature of nc-CdS thin films [22]. The lattice parameters ' a ' and ' c ' of unit cell are calculated by using relation [23]:

$$\frac{1}{d_{hkl}^2} = \frac{4(h^2 + h k + k^2)}{3a^2} + \frac{l^2}{c^2} \quad (1)$$

The lattice parameters $a = 2 \cdot d_{100}/\sqrt{3}$ and $c = 2 \cdot d_{002}$ are obtained from the positions of the peaks (1 0 0) and (0 0 2).

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