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Transparent high-surface-work-function Al-doped CdO electrodes obtained by rf magnetron sputtering with oxygen flow

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ABSTRACT

The surface work function of transparent conducting oxides is a critical parameter influencing device efficiency by controlling charge transport across interfaces. In this study, Al-doped CdO films were deposited on glass substrates by rf magnetron sputtering with and without oxygen flow. For Al-doped CdO films deposited with (without) oxygen flow, we measure the high (low) surface work function close to 5.4 (4.6) eV. Our results suggest a method for fabricating Al-doped CdO electrodes with large, tunable work functions that could be relevant in designing electrodes for improving the performance of optoelectronic and electronic devices.

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1. Introduction

Transparent conducting oxide (TCO) is most widely used in fabricating optoelectronic and electronic devices. Indium tin oxide (ITO) is one of the most promising oxide materials because of its potential applications, such as light-emitting diodes (LEDs), solar cells, and thin-film transistors (TFTs). However, the surface work function (SWF = 4.7 eV) of ITO is lower than that of Au, and so ITO is not as effective a hole-injecting metal [1]. The ability to controllably tune the SWF of TCO thin films is essential for optimizing the efficiency of optoelectronic devices. The hole injection ability at the TCO/organic interface plays a critical role in key device parameters such as operating voltage. An increased SWF of the TCO surface is required to lower the barrier for hole injection [2]. It is also a complicated function of both bulk and surface properties and depends on material processing [3]. On the other hand, CdO thin films have high transparency in the visible region of the electromagnetic spectrum and show degenerate n-type conductivity mainly due to oxygen vacancies V_O^{2+} and high carrier concentration contributed by shallow donors resulting from self non-stoichiometry [4]. Further, CdO is an important semiconductor with a band gap of 2.2 eV [5,6]. CdO has shown promising results in solar cell application [7–9]. Unfortunately not much work has been done on CdO due to its toxic nature. Although, there are reservations to the application of CdO material due to environmental concerns, CdO films have shown a transmittance of over 80% in the visible and near infrared region of the spectrum [10,11]. Jaramillo and Ramanathan [3] pointed out that the work function is sensitive to the oxygen process pressure, thus suggesting an approach for tuning the work function of TCO materials. Therefore, it is expected that such an oxygen effect also occurs equally in the case of the CdO films, since most n-type dopants also very sensitively react with oxygen during growth. In this study, we used the Kelvin probe (KP) to examine the SWF of the Al-doped CdO (AlCdO) films deposited on glass substrates by rf magnetron sputtering with and without the O_2 gas flow. It is shown the sensitivity of the work function to oxygen content provides an opportunity to tune the SWF. We found that the high work function can be achieved in an air atmosphere condition (O_2:N_2 = 1:4). Therefore, an O_2/N_2 gas ratio of 0 (0 sccm/10 sccm) or 0.25 (2.5 sccm/10 sccm) was chosen in this study.

2. Experimental details

AlCdO films were grown by an rf magnetron sputtering system using two magnetrons, a high-purity CdO target (rf power was fixed at 30 W), and an Al target (rf power was fixed at 60 W). The target size was 2 inches and the target-substrate distance was 65 mm. Targets were used in conjunction with Ar/N_2 as an ambient gas for sputtering. The flow of Ar and N_2 was 70 and 10 sccm (sccm denotes standard cubic centimeter per minute), respectively. The sputtering pressure was fixed at 5×10^{-3} Torr. The substrate temperature was fixed at 300 °C. The sputtering time was 30 min. For depositing

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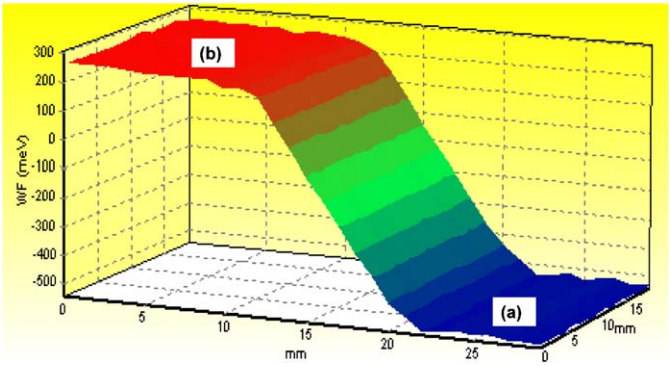

Fig. 1. (a) WFD image of the probe and AlCdO films deposited without the O_2 gas flow. (b) WFD image of the probe and AlCdO films deposited with the O_2 gas flow.

AlCdO films with an O_2 gas flow, the flow of O_2 into the chamber was fixed at 2.5 sccm during growth. The AlCdO thickness, as estimated from the field emission scanning electron microscope, was about 220 nm. The Van der Pauw-Hall measurements were performed at room temperature to obtain conductivity of the AlCdO films. The optical properties were examined using an ultraviolet-visible spectrophotometer. The work function of the AlCdO films was examined with the SKP5050 Scanning Kelvin probe (KP technology). The theory and operation of the vibrating KP method for measuring work function have been well documented [12,13]. Briefly, the KP surface is held parallel to the grounded sample to form a paralle-plate capacitor, while a small backing potential is applied between the two surfaces. The probe vibrates back and forth with respect to the sample, changing the capacitance and in turn inducing a sinusoidally varying current. When the capacitance reaches a null point, the measured contact potential difference equals the work function difference between the probe and sample. X-ray photoelectron spectroscopy (XPS) was employed to examine the chemical bonding states at the AlCdO surfaces. To identify the band bending, XPS is used to study the surface Fermi level position within the band gap of AlCdO. The binding energy scale was calibrated using the position of the Au 4f peak, measured on a clean gold foil in electrical contact with the substrate. XPS was also employed to identify the atomic concentration ratio. No N1s XPS signal was detected for AlCdO films deposited with or without the O_2 gas flow. The atomic % of Al, Cd and O for AlCdO films deposited without (with) the O_2 gas flow is 1.6 (1.5), 51.6 (49.5) and 46.8 (49.0)%, respectively.

3. Results and discussions

Fig. 1 shows the noted work-function difference (WFD) images of AlCdO and probe by KP. Prior to scanning for AlCdO samples, the Au surface was scanned via the tip. We found that the Au work function is lower than that of the tip (50 ± 5 meV). Next, the AlCdO films deposited with and without the O_2 gas flow were placed together. The AlCdO films deposited without the O_2 gas flow are on the right-hand side and the AlCdO films deposited with the O_2 gas flow are on the left-hand side. Fig. 1 shows that the SWF of the AlCdO films deposited without the O_2 gas flow is lower than that of the probe (535 ± 10 meV) and the SWF of the AlCdO films deposited with the O_2 gas flow is higher than that of the probe (265 ± 10 meV). If the work function of Au is assumed to equal 5.1 eV [14], the SWF of the AlCdO films deposited without (with) the O_2 gas flow is calculated to be 4.615 ± 0.015 (5.415 ± 0.015) eV.

Fig. 2 shows the optical transmittance spectra of the AlCdO films deposited with and without the O_2 gas flow. The results of transmittance tests showed that all the samples were transparent in the visible-light region. As the wavelength of the incident light

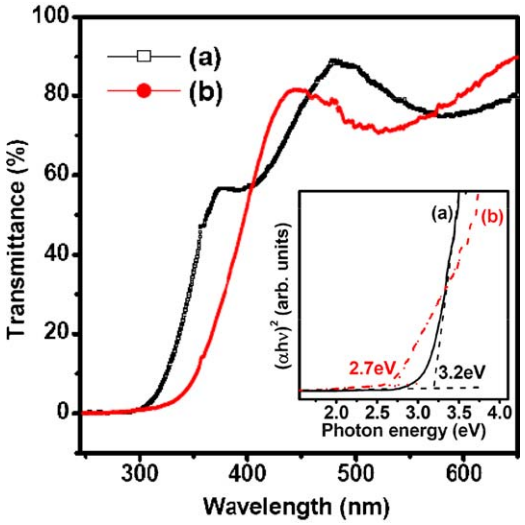

Fig. 2. Optical transmittance spectra of the AlCdO films deposited (a) without and (b) with the O_2 gas flow. Inset: square of the absorption coefficient as a function of photon energy for AlCdO films deposited (a) without and (b) with the O_2 gas flow.

reduced to the ultraviolet-light region, the transmittance dropped dramatically at the limit of optical absorption. We found that the transmittance of the AlCdO sample in the visible-light region was over 70%. The optical absorption coefficient α can be calculated from the relation presented in Ref. [15]. In the inset of Fig. 2, the relationship between $(\alpha h\nu)^2$ and $h\nu$ is plotted. h is the Planck constant and ν is the frequency of the incident photon. The optical band gap (E_g^0) is determined by the extrapolation method. E_g^0 of the AlCdO film deposited without (with) the O_2 gas flow is calculated to be 3.2 (2.7) eV. The difference in E_g^0 (shown in the inset of Fig. 2) is believed to be due to the Burstein–Moss effect [5].

According to the Hall measurements, we found that the electron concentration (n_e) and mobility of the AlCdO films deposited without (with) the O_2 gas flow were calculated to be 6.6×10^{20} (2.2×10^{20}) cm^{-3} and 45 (23) $cm^2\,V^{-1}\,s^{-1}$, respectively. It is found that n_e was higher than 10^{20} cm^{-3}, leading to the occurrence of the Burstein–Moss shift (BMS). The following well-known equation is derived [5]:

$$E_g^0 = E_g + \mathrm{BMS} = E_g + (3\pi^2 n_e)^{2/3} \left(\frac{h}{2\pi}\right)^2 \left(\frac{1}{2m^*}\right) \quad (1)$$

where m^* is the reduced effective mass and E_g is the band gap of undoped stoichiometric CdO. Assuming $m^* = 0.274\,m_0$ (m_0 is the rest electron mass) [5], BMS can be calculated to be 1.0 (0.5) eV for AlCdO films deposited without (with) the O_2 gas flow. Therefore, E_g equals 2.2 eV. E_g obtained in this work is similar to the reported value for CdO films by Ueda et al. [5], Bertram et al. [6], or Ocampo et al. [16]. On the other hand, Wolff [17] explained that when such a phenomenon competes with BMS for a semiconductor of $n_e \sim 10^{20}$ cm^{-3}, the exponent obtained from experimental value is not exactly equal to that in Eq. (1). As the carrier concentration increases, a bandgap-narrowing effect also occurs due to the effect of many-body carrier–ion interactions [18]. Zhao et al. [19] pointed out that this band shrinkage effect (BSE) begins to be competitive with the BMS at high carrier concentrations in the order of 10^{20} cm^{-3}. Thus, the band gap shift is determined by the combined effect of the Burstein–Moss and band shrinkage, the former leading to a widening of the band gap and the latter a narrowing ofthe band

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