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Structural, morphological, electrical and optical properties of amorphous $In_xAl_{1-x}N$ thin films for photovoltaic applications

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A B S T R A C T

The indium and aluminum nitride ($In_xAl_{1-x}N$) semiconductor material was postulated in 2008 as an excellent candidate to produce solar cells. In this research, amorphous layers of $In_xAl_{1-x}N$ ($0.55 \le x \le 0.60$) were synthesized using the RF sputtering magnetron technique, and then studied for their possible use in photovoltaic applications. Physical, structural, morphological, optical, and electrical properties were studied. The amorphousness was checked by XRD difractograms. Samples have very smooth surfaces, with very low roughness values, according to AFM and SEM techniques.

Electrical properties of the InAlN films were studied using Van Der Pauw technique and Hall Effect parameters. The volumetric carrier concentration decreases when InN fraction mole increases. The mobility values range between 6×10^{-2} and $5 \times 10^{-1}\,cm^2\,V^{-1}\,s^{-1}$, which are lower than those obtained for crystalline and polycrystalline samples. The obtained band gap values could be used in both the absorbing layer and the window layer in solar cells (1.9 eV–2.3 eV). Interestingly, this figures of Eg are very close to the previously obtained values for polycrystalline samples. The optical absorption coefficients were high compared to the materials currently used in solar cells (~$10^5\,cm^{-1}$). This implies the possibility of using thinner layers in photovoltaic devices based on $In_xAl_{1-x}N$.

1. Introduction

III-nitrides are interesting materials [1], and the Indium and Aluminum nitride ($In_xAl_{1-x}N$) ternary alloy arouses increasing interest in the industry due to its potential applications in optoelectronic and photovoltaic devices [2]. Sputtering technique is very promising for developing InAlN-type thin layers for future applications to solar cells, due to its low cost and high industrial scalability. In addition, latest researches have shown that this technique has a great advantage over epitaxial techniques, due to high concentrations of In can be managed without showing segregation [3]. Therefore, some research groups worldwide have obtained InAlN through RF Magnetron Sputtering, Asians standing out. In the first years (1989–2008), studies were oriented to possible applications in laser devices. In more recent times, it has been directed towards the possible application in multi-junction solar cells.

In 2009, Dong et al. [4] grew $In_xAl_{1-x}N$ layers on sapphire, Si (111), and glass substrates using AlN as buffer layer. They used a single InAI target, and claim that it is possible to grow this material with crystalline structure, suitable electrical properties, and at low cost through RF Sputtering. That same year, Jiang et al. [5] studied the optical properties of $In_{1-x}Al_xN$ (x = 0, 0.05, 0.25, 0.4 y 1) layers as a function of temperature. In 2011, the this group [6] used similar growth conditions, obtaining $In_{1-x}Al_xN$ ($0 \le x \le 0.53$) layers.

In 2010, He et al. [7, 8] analyzed the structural, morphological and optical properties of 200 nm layers type $In_xAl_{1-x}N$ ($0.7 \le x \le 1$) on quartz glass substrates. They showed that when substrate temperature exceeds 500 °C there is In segregation. It was deduced that carrier density is between $10^{20}\,cm^{-3}$ and $10^{21}\,cm^{-3}$.

In 2012 Liu et al. [9] developed $In_xAl_{1-x}N$ (x = 0.72) layers on substrates of p-type Si(001), and sapphire. They used a single InAI

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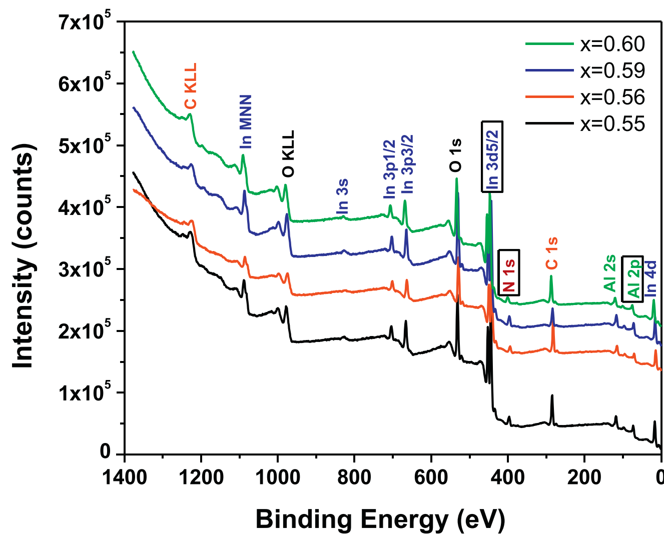

Fig. 1. XPS Survey spectrum for $In_xAl_{1-x}N$ layers, deposited on glass substrate for different Indium molar fractions "x". The spectral intensity has been shifted to improve the observation.

target at a power of 100 W for 120 min, obtaining a thickness of 90 nm. Using the Hall Effect, they determined n-type conductivity, with a carrier concentration of $4.5x10^{20}cm^{-3}$. Similarly, Lu [10] et al. obtained $In_xAl_{1-x}N$ (x = 0.76) using Si (111) and sapphire (0001) substrates. They implemented an Al-rich bilayer buffer InAlN/AlN type, highlighting the fact that this configuration significantly improves the structural quality of the film. The band gap Energy reported was 1.2 eV. N-type conductivity was determined by Hall Effect, with a carrier concentration of 10^{19} cm $^{-3}$.

In 2014, Besleaga et al. [11] reported $In_xAl_{1-x}N$ ($0 \le x \le 1$) layers on glass, and other flexibles substrates. In order to vary the concentration they only modified the deposit atmosphere composition, maintaining a single InAl target, with the substrate at room temperature.

Because of the $In_xAl_{1-x}N$ alloy has potential applications in solar cells, a research of the properties in its amorphous state has significant importance. In the present work the RF Magnetron Sputtering technique was used to deposit amorphous InAlN films. The physical, structural, morphological, optical, and electrical properties of the semiconductor alloy InAlN were studied. The objective is to describe their behavior when they are deposited on glass substrates, with a view to their future application in thin film solar cells.

2. Methodology

Thin films of $In_xAl_{1-x}N$ were grown on glass substrates using the reactive RF Magnetron Sputtering technique. Water and soap were first used, followed by drying, and finally acetone and alcohol to clean the samples. After cleaning the samples, they were placed inside the magnetron sputtering chamber, which was subjected to a vacuum of 10^{-5} Torr. A mixture of gas Ar and N_2 was injected into the chamber at a pressure ratio of 2:3, respectively. A 50%–50% In—Al target, 99.99% purity, and 1 in. diameter was used. The target-substrate distance was 5 cm. The working power had range between 40 W and 48 W. Morphological analysis was performed using the JEOL JSM-5300 scanning electron microscope (SEM), at a magnification of 35,000×. Surface topography and roughness were studied using the XE-70 Park System atomic force microscope (AFM). PHI 548 X-ray photoelectron spectroscopy technique (XPS) was used to obtain information about the atomic bonds and stoichiometry present in the semiconductor alloy $In_xAl_{1-x}N$. Structural investigations of the films were carried out using the Panalitycal X'pert Pro MRD high resolution diffractometer (HDXRD). Optical properties were characterized by means of the Ultraviolet/Visible Spectroscopy technique (UV-VIS), using a CINTRA 202 equipment. Electrical properties were evaluated using the Van der Pauw technique of Hall Effect, in an ECOPIA HMS 5000 equipment.

3. Results and discussion

3.1. X-ray photoelectron spectroscopy (XPS)

The X-ray photoelectron spectroscopy (XPS) technique was used to obtain information about the atomic bonds and stoichiometry present in the semiconductor alloy $In_xAl_{1-x}N$. For this study, the peak of a particular element is defined as the data output in counts versus the binding energy of each characteristic transition in the full survey spectrum. A subpeak is defined as one or more components of a peak, which are mathematically generated to represent different chemical states of each element. The baselines were established using an integral model (Shirley). As reference binding energy, the primary peak C1s was set at 284.5 eV [12]. Binding energies of the other sub peaks were adjusted according to the difference between the measured position and defined position of the reference peak.

Fig. 1 shows the complete survey spectrum for all samples with different Indium molar fractions "x". The spectrum shows peaks of considerable intensity located at 531.4, 444.5, 396.6, 284.5, and 73.7 eV; corresponding to O1s, $In3d_{5/2}$, N1 s, C1s and Al2p binding energies, respectively. Oxygen and carbon peaks are probably due to contamination of the sample.

The estimated molar fractions of Indium (Composition of In) in the samples of $In_xAl_{1-x}N$ were determined by the equation:

$$C_i = \frac{A_i/S_i}{\sum A_i/S_i} \tag{1}$$

Where S is the sensitivity factor, A is the integral of each peak, and C is the atomic concentration of the element in question.

Fig. 2 shows the high-resolution spectra for the $In3d_{5/2}$, N1 s, and Al2p signals for the $In_{0.59}Al_{0.41}N$ semiconductor alloy. The peak of the N1 s signal shown in Fig. 2.a was deconvolved in two Gaussian functions. Sub peaks were centered on two binding energies, 396.4 and 399.2 eV, attributed to the N—In and N—Al bonds, respectively [13, 14]. Fig. 2.b shows the $In3d_{5/2}$ XPS spectrum, which was adjusted by three components. The binding energies 445.3, 444.3, and 443.3 eV, were assigned to the In—O, In—N, and In—In bonds, respectively, according to previous research reports [14].

All Al2p peaks were adjusted as one or more pairs of divided spin-orbit sub peaks, and with a separation of 0.4 eV between the $Al2p_{3/2}$ and $Al2p_{1/2}$ components [15]. The area ratio of components $2p_{3/2}$ and $2p_{1/2}$ was set as 2:1, and the binding energy of the Al2p peak is reported as the weighted pair average, which is given by the equation:

$$E_{Al_2p} = \frac{A_{Al_2p^{3/2}} \times E_{Al_2p^{3/2}} + A_{Al_2p^{1/2}} \times E_{Al_2p^{1/2}}}{A_{Al_2p^{3/2}} \times E_{Al_2p^{1/2}}} \tag{2}$$

Where A is the integrated area of the sub peak and E is its link energy in eV.

Fig. 2.c shows the peak corresponding to the Al2p level, which was deconvolved in three Gaussian functions. Two of the sub peaks were

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