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Structural and optical characterization of In$_x$Ga$_{1-x}$N nano-structured grown by chemical vapor deposition

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Nitrides of group III have generated important applications in optoelectronic devices. Principally InGaN is a novel alloy for the development of solid-state lighting and photovoltaic systems, since it is possible to control its bandgap from 3.4 eV to 0.7 eV by simply varying the indium concentration. However during the growth of InGaN inherent defects are obtained in the material, degrading its optical properties. In this work, the effect of the indium concentration is studied. The results of the optical and structural characterization of a series of In$_x$Ga$_{1-x}$N films (0 $\leq$ x $\leq$ 0.3) deposited by chemical vapor deposition (CVD) are reported.

Keywords: InGaN; semiconductor; luminescence and optoelectronics.

Los nitruros del grupo III han generado aplicaciones importantes en los dispositivos optoelectronicos. Principalmente el InGaN ha mostrado ser una aleación novedosa para el desarrollo de la iluminación de estado sólido y sistemas fotovoltaicos, ya que es posible controlar el ancho de su banda prohibida desde 3.4 eV a 0.7 eV con solo variar la concentración de indio. Sin embargo durante el crecimiento de las películas de InGaN aparecen defectos en el material debido a las diferencias entre los átomos indio y galio. En este trabajo se estudia el efecto de la concentración de indio en las propiedades del InGaN. Se reportan los resultados de las caracterizaciones ópticas y estructurales de las películas de In$_x$Ga$_{1-x}$N (0 $\leq$ x $\leq$ 0.3) depositadas por vapores químicos (CVD).

Descriptores: InGaN; semiconductor; luminiscencia.

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1. Introduction

In the development field of new materials, the compound semiconductors continue being an area of great interest and rapid expansion [1]. The ternary semiconductor InGaN is an important alloy for the development of lighting emitting devices, photovoltaic systems and power electronic, due to the capacity to control the band gap ($E_g$), which varies according to the indium concentration in a range of energies from 0.7 eV (InN) to 3.4 eV (GaN) [2]. Recently some attempts to grow high-quality low-cost InGaN have been done. One of the techniques that more likely fulfill the requirements is the chemical vapor deposition (CVD). This technique has reduced the cost of the synthesis maintaining an acceptable level in the optoelectronic properties of InGaN. However, the inherent mismatch between the lattice parameters of the substrate (sapphire, SiC, AsGa, Si, LiGaO) [3,4] and the InGaN phase, plus the indium incorporation (0 $\leq$ x $\leq$ 1) limits the growth of the material and degrade the optical and electronic InGaN properties. [5,6,8] In this work spectroscopy UVVIS and photoluminescence (PL) have been used to study the optical properties of InGaN films grown by CVD [9]. Scanning electron microscopy and X-ray diffraction were used to characterize the morphology and structure of the InGaN films.

2. Experimental

The synthesis of In$_x$Ga$_{1-x}$N multilayer films with an indium composition of 0 $\leq$ x $\leq$ 0.3 deposited on sapphire at temperature of 900°C were grown by CVD. These films use the layers of aluminium nitride (AlN) and gallium nitride (GaN) as buffer and nucleation layer, respectively. The Fig. 1 shows the schematic diagram of the multilayer structure.

The absorption measurements were made by two different techniques: transmission and diffuse reflectance. The absorption spectra were obtained with an AVANTES spectrometer (AvaSpec 256) in the wavelength range from 180 nm to 1100 nm. The diffuse reflectance was carried out in a UV-visible spectrometer Cary 300. All measurements of absorption were realized at room temperature. The PL measurements were obtained using two different light sources. The first, using a He-Cd laser (74 Series omnichrome - $\lambda$=325 nm). The luminescence of the sample is collimated through a spectrometer (SPECTRAPRO 500i) where the
signal is quantified. In the second PL measurement, a UV-visible spectrometer, Hitachi Digilab F4500, with xenon lamp as excitation source was utilized. The XRD characterization was carried out in a powder diffractometer (Philips X’pert). The surface of the InGaN films was studied in a SEM Jeol 5300.

3. Results and discussion

The absorption results obtained by diffuse reflectance (Fig. 2) were very different in comparison with the transmission measurements. The attenuation zone (including tails) varies in a region of energies from $\sim 2$ eV to $\sim 3.3$ eV (620 nm to 375 nm), which are near to the values of energies band gap expected in the In$_x$Ga$_{1-x}$N films according to the Vegard’s law. The origin of these absorption tails are attributed to the deformation of the crystalline lattice and to the existence of defects such as oxygen impurities and gallium/nitrogen vacancies [10].

Figure 3 shows the PL spectra of the In$_x$Ga$_{1-x}$N films. The samples with indium composition smaller than 20 atomic percent ($x<0.20$) showed peaks with a FWHM of $\sim 500$ meV whereas samples with higher indium composition ($x>0.20$) presented a broader peaks with a FWHM of $\sim 1$ eV. Therefore, as well as the composition is increased in the In$_x$Ga$_{1-x}$N phase the band gap energy is modified, showing a red-shift of the PL peak and also broader luminescence in the high indium samples. This behavior has its origin in the deformation of the In$_x$Ga$_{1-x}$N lattice (stress due indium incorporation and the formation of a wide range of different In$_x$Ga$_{1-x}$N crystals) and the existence of defects (oxygen impurities and gallium/nitrogen vacancies). Furthermore, in some parts of the spectra some modulations were observed due to the interference effect (Fabry-Perot) caused by internal reflections within the multilayer In$_x$Ga$_{1-x}$N films [11].

Figure 4a shows the XRD results of the In$_x$Ga$_{1-x}$N films. These diffractograms showed a hexagonal crystalline phase (wurzite) for the films. In$_x$Ga$_{1-x}$N (0002) and GaN (0002) planes are marked. The In$_x$Ga$_{1-x}$N crystalline phase was correlated with GaN phase located in the 2θ ($34.56^\circ$) position for the crystallographic plane (0002) according to the ICDD crystallographic letters [12]. In Fig. 4b is shown traces of
some impurities that appear in the \( \text{In}_x\text{Ga}_{1-x}\text{N} \) films, indium oxide (\( \text{In}_2\text{O}_3 \)), indium nitride (\( \text{InN} \)) and indium metallic (clusters). The indium oxide can be related with the emission in the 550 nm region (emission by an indirect transition of 2.09 eV reported by Novkovski) [7].

Figure 5 shows SEM images of the InGaN films. The surface morphology of the films does not follow a pattern of growth that has a relation with the indium composition. The growth mode of the \( \text{In}_x\text{Ga}_{1-x}\text{N} \) films appears to be the Volmer-Weber type. This growth mode is characterized by island formation due to nucleation crystals in diverse crystallographic directions. In this case, the crystals are the structures of columnar type which self-ensemble to form \( \text{In}_x\text{Ga}_{1-x}\text{N} \) islands.

4. Conclusions

A series of InGaN films deposited by CVD were characterized. It was found that the \( \text{In}_x\text{Ga}_{1-x}\text{N} \) films with indium composition, \( x \leq 0.20 \) present absorption and emission spectra that follow the Vegard’s law. \( \text{In}_x\text{Ga}_{1-x}\text{N} \) with higher content of indium \( (x \geq 0.20) \) showed a broad PL emission (FWHM \( \sim \) 1 eV) and large tails of absorption. In addition an extrinsic emission in the region of \( \sim \) 570 nm \( (\sim 2.17 \text{ eV}) \) was observed in this films. XRD showed the presence (traces) of undesirable phases such as \( \text{In}_2\text{O}_3 \), \( \text{InN} \) and metallic indium in the films. SEM analysis found the formation of \( \text{In}_x\text{Ga}_{1-x}\text{N} \) islands that affect the smoothness of the film surface.

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12. ICDD crystallographic letters: In (00-005-0642), In2O3 (00-006-0416), InN (00-050-1239), Al2O3 (00-010-0173), GaN (00-050-0792).