Optical Characteristics of GaN Thin Film Deposited by Pulsed Laser Ablation

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Abstract
We present optical data on GaN thin film samples grown by pulsed laser ablation, two different pulsed laser ablation powers. The bandgap of specimens with recognizable crystallinity has been deduced from the optical spectrum data. Longtails were observed below the gap band. In specimens are entirely dependent on the powers of the laser ablation process. The specimens with the highest laser power, 2000 mJ, show that a smaller near band edge emission peaked at 3.32 eV is observed up to room temperature. The maximum energy bandgap 3.62 is possibly observed at 1500 mJ laser energy.

Keywords: Gallium nitride; Pulsed Laser Ablation; energy bandgap; X-Ray Diffraction (XRD); Optical properties; Structural properties.

1-Introduction

Nanostructured Gallium nitride (GaN) materials have attracted significant attention recently due to the unique physical and optical characteristics. GaN is a semiconductor material with (3.4 eV) direct wide bandgap and 26 meV excitation binding energy [1-4].

GaN has been frequently studied due to its potential application in the areas of optoelectronic devices [5, 6]. Besides, short-wavelength electroluminescence applications, high electron mobility transistor (HEMT), and heterostructure field-effect transistor (HFET) based GaN is always a significant choice [7, 8]. Due to its distinguished lifespan and luminescence compared to conventional LEDs, GaN can also be used for detectors, short-wavelength optoelectronic devices, and UV or blue region emitters [9, 10].

GaN nanoparticles' purity is crucial for optoelectronic devices for applicability as active regions in the color-tunable light-emitting diodes and their direct wide bandgaps for laser diodes application.

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GaN-based nanoparticles have been synthesized using multiple methods [11-14]. However, most of these methods are established on temperature processes and expensive chemicals. In pulsed laser technique, the possibility of growing thin films into high vacuum level at low substrate temperatures and fast growth rate of the order of Å/pulse is relatively easy [15-18].

GaN results concerning GaN thin films' growth by pulsed laser deposition were reported by Vinegoni et al. [19]. This research group grew thin films with modest homogeneous diffuse granular structures over the entire tested surface. Parallelly, high-level crystallinity has been reported by Vispute et al. [20] on Al₂O₃ [0001] substrate surface.

To date, almost all the semiconductor III-nitrides available commercially is prepared by the heteroepitaxial growth using the epitaxy of the molecular beam, Pulsed Laser Deposition, MOCVD, HVPE [21-28]. Although high-quality Gallium nitride semiconductor layers can be prepared, grown, and deposited using the previously presented techniques, but, these methods of growing to involve Complex and sophisticated technologies that are expensive relatively and in setup are complicated [29-32].

In addition to that, some of the presented technologies used for deposition of the semiconductor III-nitrides nanostructures like metal-organic chemical vapor deposition and the hydride vapor phase epitaxy method include precursors of toxic and flammable [33-36]. Thereafter, scientific researchers need to pay special attention to chemical safety, chemical waste disposal and the cost spent on the equipment and maintenance, etc [37-40]. Therefore, due to the importance of this topic, it requires finding safe, simple, and inexpensive methods that are alternative and capable of growing thin and high-efficiency films as a successful alternative to previously used techniques such as pulsed laser deposition and pulsed laser ablation in liquid technology and this will be presented here as a new and safe technology

2-Experimental detail

In this experiment, The GaN target was immersed in 5ml ethanol and shot using Nd:YAG pulsed laser, as shown in Figure 1(a). Laser parameters were correctly indicated by wavelength (532nm) and frequency(4 Hz). Six samples were prepared for each flowing energy 1000 mJ, 1200 mJ, 1400 mJ, 1600 mJ, 1800 mJ, and 2000 mJ. each sample was irradiated with 500 pulses to saturate the liquid with nanomaterials. The focal length was 12 cm varying after every 100 pulses to keep the laser and the GaN surface interaction in the same manner. Figure 1(b) shows the liquid sample after the ablation method by Nd:YAG pulsed laser.
The GaN liquid samples were deposited on the quartz substrate using a drop-casting method, as shown in Figure 2. The quartz substrate was heated by hotplate at a temperature range from (70°C - 90°C). Nano-liquid was dropped slowly on the quartz substrate when it was reached the desired temperature; each drop on the quartz substrate was left drying and then followed by another drop (100 drops) to form the thin film GaN on the quartz substrate. All the processes were done in less than 12 hours to avoid oxidation. The quartz substrate was cleaned thoroughly before starting the dropping process by Alcohol. The nano-liquid should be adequately shaken before each drop to keep the GaN nanomaterials' distribution identical in all samples.

The quartz/nanoparticles (GaN) sample was tested in the XRD test, absorption, transmission, and analyzed to find the energy gap of GaN. All results are analyzed in the next section of results and discussions.

3-Results and Discussions
3.1. XRD
XRD patterns of GaN nanoparticles are presented in Fig.3. The characteristics peaks are divided into GaN and Sapphire. GAN thin films' XRD pattern was calculated in the 2θ=20-80 degree ranged by
using a Philips X-ray diffraction meter. In this result, it can be seen clearly that the film contain firm peaks of sapphire (100) (002) (102) (110) (112) (004) (0001) (0004) at about 32°, 34°, 48.2°, 58.2°, 68.8°, 72.8°, 42°, 72.5° respectively. GaN peaks are (001) and (002), both oriented at 36.8°. By using Scherrer’s Formula, $d = 0.9\lambda/(\beta \cos \theta)$, where $d$, is the diameter of the crystalline grain, $\beta$ are full width at half maximum (FWHM), $\lambda$, incident wavelength (0.154 nm) and $\theta$ is the reflection angle. The diffraction peak (0002) has a narrow FWHM magnitude of 0.12, and the high order GaN diffraction peak (0004) proves an excellent quality of the GaN thin films were grown on the sapphire substrate [41, 42].

**Fig.3. XRD pattern of GaN nanoparticles**

Figure 4 shows a 100 nm magnification TEM image, indicating that the nanoparticles' surface is relatively large particles ~ 100 nm mixed with ~50 nm in diameter particles. The nanoparticles appeared dispersed at the matrix, even though few Nanocluster structures appeared on the image and were associated with the XRD spectrum. This image represented all peaks that were recognized (reflection peaks) possibly indexed to hexagonal GaN structure.

**Fig. 4. TEM image of GaN nanoparticles**
The optical properties like the absorptions, absorption coefficient, and the transmission as a function of wavelengths in nanometer will be tested and calculated at the range the wavelengths of 300-1100 nm using a double beam UV-VIS spectrophotometer, and the value of the energy bandgap as a function of the optical energy.

The incident photon energy has been calculated using the formulae (1) [43-45]:

\[ E_g \text{ (eV)} = \frac{1.24}{\lambda \text{ (µm)}} \]  
(1)

Where \( E_g \) is the optical energy gap and the \( \lambda \) is the wavelength of the incident photon. The absorption coefficient was drawn as a function wavelengths, the following formula was used to estimates (2) [46-48]:

\[ (\alpha h\nu) = B (h\nu - E_g)^r \]  
(2)

where \((\alpha)\) is the calculated absorption coefficient value, \((h)\) is constant (Planck), \((\nu)\) the light speed, \((B)\) is constant and \((r)\) is a constant it values depends on the type of the material.

The optical band gap was deduced from the linear relation (extrapolation of straight line) of the curve between \((\alpha h\nu)^{1/2}\) and \((h\nu)\). Following eq. (3) has been used to estimate the absorption coefficient at a specific wavelength [49-53]:

\[ \alpha = 2.303 \left( \frac{A}{t} \right) \]  
(3)

\(A\) = absorptance and, \(t\) = thickness.

The sharp edge at the UV region insures the formation of the direct gaps of the prepared films. the following expression has been used to found it[54-59]:

\[ \alpha h\nu = A( h\nu - E_g)^{1/2} \]  
(2)

\(h\nu\): photon energy \(\alpha\): Absorption coefficient, and \(A\): constant. The energy gap \(E_g\) was obtained by extended the straight line of the \((\alpha h\nu) \) 2 versus \(h\nu\) plot with the energy of incident photons.

In Figure 5a, the 300 nm to 1100 absorption spectrum shows the absorption peak position at \(\sim320\) nm, which confirms GaN nanoparticles' size to be \(\sim100\) nm (Figure 4). The spectrum absorption experimental data have lower broadening, and higher absorption postulates a higher concentration of tetGaN NPs. Figure 5b shows the absorption coefficient at different laser energy. Each material has different absorption coefficients, and materials with higher value more readily absorb incident photons, which means they excite electrons from the valence band into the conduction band. The peak represents the maximum absorbed point of photons at that wavelength, and the lower values represent the poor absorption of photons at that wavelength. The sharp point at the absorption coefficient curve shows that the material has no enough energy to excite an electron from one level to another.
Figure 6a shows the transmission curve of GaN at different deposition energy, which varies between the maximum and the minimum value. In the visible range, the transmission value is found between 86 % to 93 %. This difference in proportions was due to the difference in the granular size of the Nano Claudel that was prepared using different laser energies, as the results showed that the transmission values were low when using high energies such as 2000 mJ and 1800 mJ in the ablation due to the large particle size and the random and irregular distribution of the eradicated granules inside the liquid, by reducing the laser energy, it was evident that the increase in the transmission values reached 1400 mJ, where the highest value of the transmission appeared, this is due to the small size of the particles and the uniform distribution of the Nano Claudel particles. Then the transmitter values start to gradually decrease again as a result of the effect of the weak laser energy on the randomly eliminated crystals and the weak distribution within the liquid.

Fig.6. a) transmission curve b) energy gap at different laser deposition energy
4-Conclusion

Growth high-quality thin-film GaN nanoparticles were performed using pulsed laser ablation. This paper presents the optical properties of GaN deposited on the quartz substrate and analyzed using XRD, TEM, and UV-Vis absorption tests. Firstly, the XRD test shows that the diffraction peak (0002) has a narrow FWHM magnitude of 0.12, and the high order GaN diffraction peak (0004) proves an excellent quality of the GaN thin films were grown on the sapphire substrate, secondly, the TEM test, the image appeared nanoparticles dispersed at the matrix the image represented all peaks that were recognized (reflection peaks) possibly indexed to hexagonal GaN structure and finally the UV-Vis absorption test shows the absorption peak position at \( \sim 320 \text{ nm} \), the absorption spectrum experimental data have lower broadening, and higher absorption postulates a higher concentration of GaN NPs. The sharp point at the absorption coefficient curve shows that the material has no enough energy to excite an electron from one level to another, the transmission curve of GaN in the visible range is found to have value between 50% to 95% and the energy gap value is 3.43 eV at 1200 mJ then went to a maximum value 3.6 eV at 1400 and back to 3.49 eV at 1000 mJ due to the core-shell phenomena.

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