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Impact of laser wavelength on pulsed laser deposition method-deployed gallium nitride (GaN) thin films

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ABSTRACT

Pulse laser deposition (PLD) was used to produce and deposit nanofilms of gallium nitride (GaN) with a nanocrystalline structure on quartz substrates. The pulsed laser wavelength effect on the structural and optical properties of these films was studied. The PLD process achieved with the constant parameters includes a temperature of 300°C, a frequency of 3 Hz, a number of pulses of 250, and a voltage of 900 V, but with the three various wavelengths of the pulsed laser, it includes 1.064, 0.532, and 0.355 µm. The influence of varying deposition conditions, such as laser wavelengths (1.064, 0.532, and 0.355 µm), on the optical and structural characteristics of these nanofilms was examined. The characteristics were examined utilizing several analytical techniques, including X-ray diffraction (XRD) and UV-Visible spectrophotometry (UV-VIS). The XRD results indicated that the crystalline structure of the deposited films enhances with an increase in laser wavelength, laser energy, substrate temperature, and the use of a target with an equal material ratio, as evidenced by the heightened peak intensity. The ultraviolet-visible (UV-vis) was used to characterize and study these nanostructure thin films. According to the UV-vis measurements, the refractive index achieved the same values when wavelengths of the pulsed laser, 1.064 µm and 0.532 µm, were used. As the wavelength of the pulsed laser increases, the values of absorption also increase, while the optical energy gap values decrease, but the transmission values exhibit random behavior with this increase. So, from these results, we can find the two various laser wavelengths (1.064 µm and 0.532 µm) achieved very similar results except for the transmission results, which means that using any one of them in the deposition process will achieve the same results for the desired application.

Keywords: *Gallium nitride (GaN), Laser wavelengths, Pulsed laser deposition, Quartz substrates, Thin films*

1. INTRODUCTION

Gallium nitride (GaN) is a highly durable, mechanically stable wide bandgap semiconductor material. Power devices utilizing GaN have superior breakdown strength, accelerated switching speed, enhanced thermal conductivity, and reduced on-resistance, thereby dramatically surpassing silicon-based devices and establishing GaN as the second most favored material in the semiconductor industry after silicon [1, 2]. GaN is a great choice for high-power applications and high-temperature operation because it has electron mobility that is equivalent to silicon but with a bandgap that is three times bigger [3, 4]. Also, there is a growing interest in the use of GaN as a mechanical material [5, 6]. In addition, GaN offers a great deal of promise for use in other devices (electronics and photonics) such as high-electron mobility, inhomogeneous bipolar junction transistors, and field-effect transistors [7, 8]. These devices include high-quality optoelectronic devices like light-emitting diodes [9, 10].

There are widely techniques that can be used to prepare GaN films, including molecular beam epitaxy (MBE) and chemical vapor deposition (CVD), but these techniques are very complex and expensive. Pulsed Laser Deposition (PLD) represents the simple technique to deposit GaN films [11, 12]. PLD is a thin-film deposition method that using a high-energy pulsed laser to deposit the surface of a solid target inside a vacuum chamber, subsequently condensing the vapor onto a selected substrate to create a nanofilm with a thickness of several micrometers [13, 14] Pulsed laser deposition (PLD) is commonly used to deposit nanofilms of oxides [7, 8]. This technique is performed in a super vacuum and in the presence of a background gas, unlike other techniques [15, 16]. Common laser wavelengths PLD users select are Nd:YAG (1.064 µm, 0.532 µm and 0.355 µm).

In his study, a high purity GaN nanostructure has been deposited using a simple and cheap method (PLD), and the optical analysis for fabricating optoelectronics and sensing devices is studied.

2. EXPERIMENTAL SECTION

GaN nanostructured thin film was synthesized and placed onto quartz substrates via the PLD method [17, 18].

The substrates of quartz were cleaned prior to the process of the deposition. These methods of cleaning are crucial for the removal of contaminants and the prints of fingers from the quartz substrates. The quartz is submerged in a water solution and soap for 10 minutes while being manually cleaned. Subsequently, clean it using water multiple times, then immerse it in the laboratory ethanol for 5 minutes. Ultimately, dry it with heated air.

The target is prepared from the ultra-purity GaN material from Aldrich Company (United States) when the GaN material is subjected to a pressure of 15 tons to create a disk with a diameter of 2 cm and a height of 1 cm [19, 20].

The parameters that used to prepare GaN thin films through PLD technique includes temperature 300°C, frequency 3Hz, number of pulses 250 pulse, voltage 900V and use three different laser wavelength 1064 nm, 532 nm and 355 nm (Table 1). These parameters were selected according to their relevance to the objectives of the study, which include the deposition of thin films with a good quality as possible, as it was found that the best parameters used to achieve this are the parameters mentioned above, with the use of three different wavelengths to study their effect during the deposition process.

The objective of employing various parameters during the deposition process is to identify the optimal circumstances that yield high-quality thin films for application in gas sensor devices.

Utilizing a ultra-violet visible (UV-vis) spectrophotometer (Shimadzu UV-Vis 1800, Japan) within the wavelength range of 200–1000 nm, we determined the optical properties of the thin film through the transmittance (T) spectrum, which includes optical absorption (A) and optical energy band gap (E_g).

The structural properties of the thin films were examined utilizing the X-Ray Diffractometer (XRD) technique (Aeris benchtop, XRD System / Malvern Panalytical Company,

Table 1. The practical parameters in the deposition process

The Practical Parameters	Value
Wavelengths of Laser	1.064, 0.532, and 0.355 µm
Substrate Type	Porous silicon and quartz
Substrate Temperature	300°C
Laser Pulse Duration	7 ns
Shoot Frequency	3 Hz
Pulse Number	250 pulses
Pressure	bar

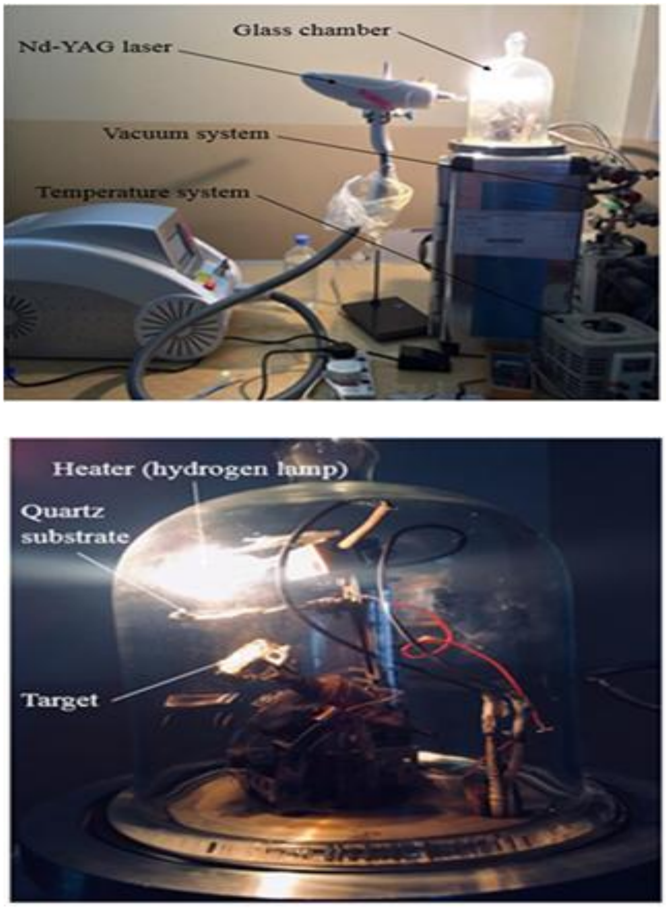

Figure 1. Experimental work setup

Netherlands) with a Cu-K wavelength ($\lambda = 1.54$ Å), a maximum 2θ range of $-4° < 2\theta \leq 142°$ (employing a scanning detector and full active length), direct optical position sensing (DOPS3) with precise lifetime positioning accuracy, and a scan speed of 2.17°/s. Refer to Figure 1 (b). The scanning angle 2θ varied from 30° to 75° in increments of 5 degrees. The analysis was conducted at Al-Hora company in Baghdad.

3. DISCUSSION AND RESULTS

3.1. X-Ray Diffraction (XRD) of GaN and In_2O_3 Thin Films

Figure 2 shows the effect of the pulsed laser wavelength on the X-ray diffraction patterns of GaN nanofilms prepared and dropped on a PSi substrate with energy density 900 mJ/cm^3 and substrate temperature 300°C. XRD patterns were exhibited in the range of 2theta = 30–65° due to no diffraction picks for GaN above 2theta = 65°. The XRD pattern contains two prominent peaks appearing at 2theta = 33.6° and 63.3° corresponding to the (002) and (103) planes, which match with the hexagonal crystal of GaN. Also, the patterns of the XRD show the intensity of these peaks is high at 1064 nm laser wavelength, which indicates an improvement in the crystalline structure of the film.

The effect of laser wavelength on the XRD patterns of In_2O_3 thin films deposited on the PSi substrate with energy density 900 mJ/cm^3 and substrate temperature 300°C are shown in Figure 2. XRD patterns displayed in the range of 2theta = 30–65° because of no diffraction peaks for In_2O_3 above 2theta = 65°. X-ray diffraction patterns have three clear peaks appearing at 2θ = 31.2°, 34.67° and 62.5° corresponding to (222), (400) and (622) planes, which match with the cubic crystallographic of In_2O_3. In addition to that, XRD patterns show increased peak intensity when using a laser wavelength of 1064 nm through the deposition process; it implies enhanced control over the crystalline structure, leads to better surface shape and film quality, and, according to previous studies [21–23].

Table 2 explains the values of structural parameters that were obtained from XRD patterns, which are average crystal size (D), full width at half maximum (ß), and angles corresponding to each peak (2θ) for both films.

3.2. UV-Visible spectrophotometer (UV-VIS)

GaN nanostructured thin films were formed utilizing the PLD approach on quartz substrates at different wavelengths of the pulsed laser: 1.064 µm, 0.532 µm, and 0.355 µm. The deposited films were examined and evaluated using a UV-Vis spectrophotometer. The optical characteristics of GaN thin films were ascertained using transmission measurements within the wavelength range of 200 to 1000 nm.

Figure 3 illustrates the optical transmission of GaN nanostructured thin films. This figure indicates that the optical transmission of these films exhibits random behavior with varying laser wavelengths. The high value of T% appear with the laser wavelength 532nm which equal

Table 2. XRD characteristics of GaN/PSi and In_2O_3/PSi prepared at wavelengths of pulsed laser: a) 1.064 µm, b) 0.532 µm, and c) 0.355 µm

Material	Laser λ (nm)	2θ (Degree)	2θ (Rad)	hkl	ß (Degree)	ß (Rad)	D (nm)	Average D (nm)
GaN/PSi Thin film	1064	33.6	0.58	(002)	0.51	0.009	16.3	21.9
		63.3	1.10	(103)	0.11	0.002	27.5	
	532	33.6	0.58	(002)	0.57	0.010	13.93	17.4
		63.3	1.10	(103)	0.11	0.002	20.88	
	355	33.6	0.58	(002)	1.95	0.034	4.30	16.55
		63.3	1.10	(103)	0.11	0.002	28.8	

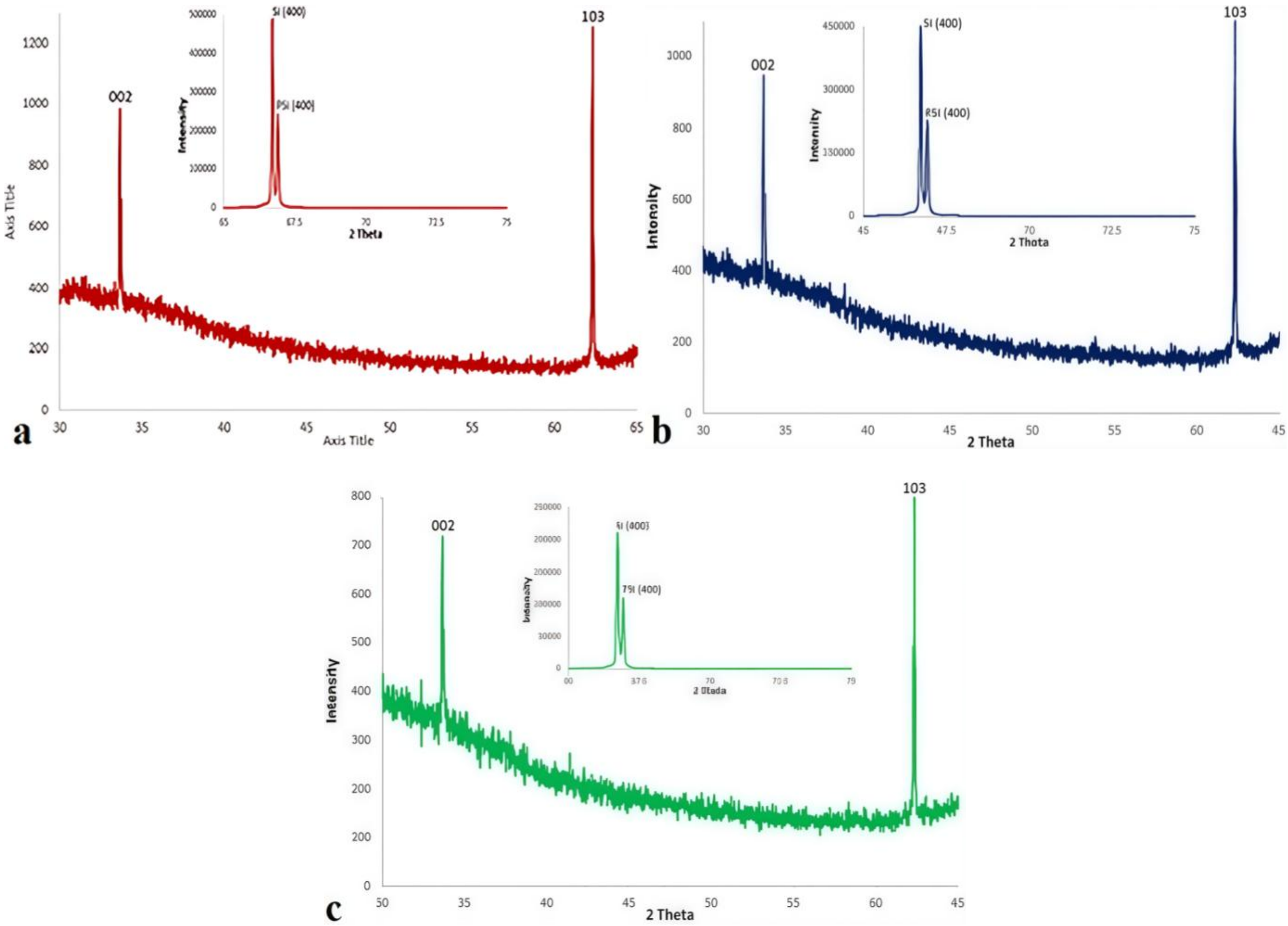

Figure 2. XRD pattern of GaN/PSi prepared at wavelengths of pulsed laser: a) 1.064 µm, b) 0.532 µm, and c) 0.355 µm

to 74%, while the low value of T% appear with the laser wavelength 1064nm which equal to 58% and the laser wavelength 355nm appear T% value equal to 71%. This random behavior can only be explained by the instability of the optical transmission values of the films with the wavelength of the laser used, which means that using wavelengths higher than 1064 nm will not necessarily give lower transmission values, or using a wavelength lower than 532 nm will not give higher transmission values, as was proven when using a deposition laser wavelength of 355 nm, where the transmission value was lower according to previous studies [24, 25].

Figure 4 shows the optical absorption of these films, where it can be noted that the absorption of these films decreases when the laser wavelength rises, with these values being about 6.8, 6.2, and 2.9 at laser wavelengths of 1064 nm, 532 nm, and 355 nm, respectively, according to previous studies [26–28].

We determined the optical band gap, Eg, as a function of photon energy by plotting the curve of $(\alpha h\nu)^2$ against (hv) [5, 13, 21, 29] as illustrated in Figure 5. The computed band gap energy (Eg) is approximately 4.7 eV, 4.8 eV, and 5.1 eV for laser wavelengths of 1064 nm, 532 nm, and 355 nm, respectively, indicating that as the laser wavelength increases, the band gap energy decreases, and vice versa. The energy band gap values, corresponding to the variation in laser wavelength, range from 4.7 eV to 5.1 eV, indicating that the laser wavelength has a minimal impact on the energy band gap values. In this instance, the elevated laser wavelengths (1064 nm and 532 nm) yielded remarkably similar readings, consistent with prior research [12, 30–32].

Likewise, mathematically, from the values of transmission and absorption, and depending on the relationship $R + T + A = 1$ [25, 33–35], the optical reflectance (R) of these nanostructure films can be calculated. Finally, as a function of the wavelength and from the transmittance values in the range of 200–1000 nm, it can calculate the values of refractive index (n) at various laser wavelengths. As shown in Figure 6, it can be noted the films deposited by laser wavelengths 1064 nm and 532 nm achieved the same value on refractive index (2.58), while the film deposited by laser wavelength 355 nm achieved a refractive index value (2.26). This indicates two points: The first point is that the wavelength of the laser in the case of depositing a GaN film has almost no effect on the reflectivity values; the second point is that to determine slightly different values, widely different wavelengths must be used, according to previous study [1, 10, 36, 37].

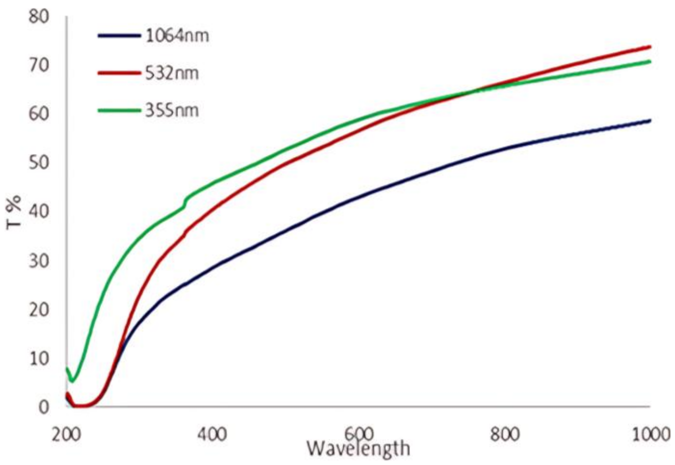

Figure 3. The optical transmission of GaN at different-laser wavelength

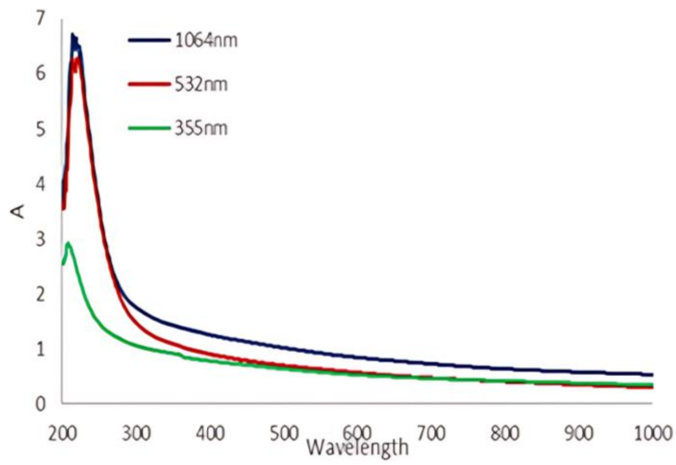

Figure 4. The optical absorption of GaN at different-laser wavelength

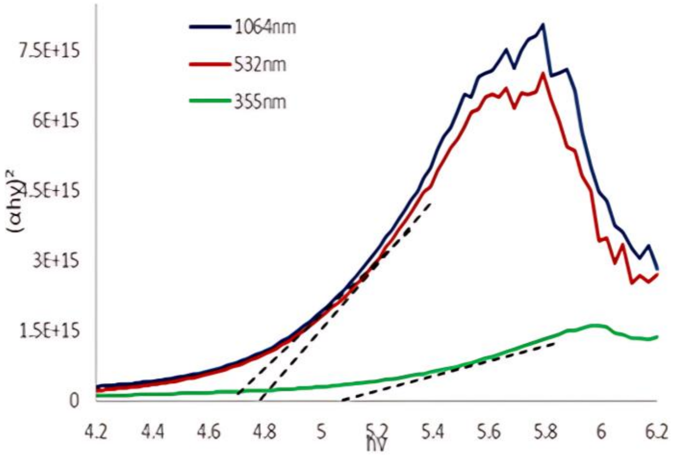

Figure 5. Optical energy gap (E_g) of GaN at various laser wavelength

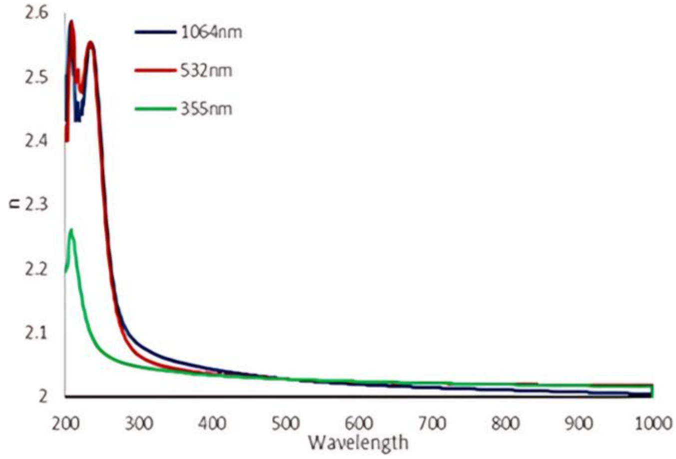

Figure 6. The refractive index of GaN at various laser wavelength

4. CONCLUSION

By using a Q-switched Nd:YAG laser at three different wavelengths, including 1064 nm, 532 nm, and 355 nm, nanostructure thin films of GaN were deposited on the quartz substrates. From the optical results presented in this paper, we note that the optical transmission of these films shows random behavior due to the effect of the laser wavelength, where the highest value was achieved with the laser wavelength of 523 nm and the smallest value was achieved with the laser wavelength of 1064 nm. While the absorption of these films increased with increasing laser wavelength and achieved close values at wavelengths (1064 nm and 532 nm). The energy gap values were close within the range of 4.7 eV to 5.1 eV, which means that the laser wavelength does not have a strong effect on the energy gap of the films. Finally, the effect of the laser wavelength on the reflectivity values was almost non-existent, as very close values were achieved with the three laser wavelengths used. So, it can be concluded from these results that using the laser wavelengths 1064 nm or 532 nm will have almost the same effect on the results related to the preparation of these thin films, which can be used in many applications, including waveguide and gas sensor systems. The effect of different deposition conditions, such as different pulsed laser wavelengths (1.064, 0.532, and 0.355 μm), on the structural and optical properties of the deposited nanofilms and structures has been studied. These properties were analyzed by using various analysis techniques, which include X-ray diffraction (XRD) and UV-Visible spectrophotometry (UV-VIS). The results of XRD presented that the deposited films' crystalline structure improves with increasing the laser wavelength to 1064 nm, which gives two best picks. The ultraviolet-visible (UV-vis) spectrophotometer was employed to characterize and analyze these nanostructured thin films. The UV-vis experiments indicated that the refractive index yielded identical values for laser wavelengths of 1064 nm and 532 nm. As the laser wavelength grows, absorption values rise and energy band gap values diminish, while transmission values display erratic behavior with this increase. The results indicate that the two distinct laser wavelengths (1064 nm and 532 nm) yielded comparable outcomes, except for the transmission results. Consequently, employing either wavelength in the deposition process will produce equivalent results for the intended application.

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