Enhancement of Photoluminescence Characteristics of Zinc Oxide Nanostructures Using Inclined Pulsed Laser Irradiation

Ali Reza Bagheri, Shaheed Ibrahimi

Department of Physics, Faculty of Science, Ferdowsi University of Mashhad, Mashhad, IRAN

Abstract
Zinc oxide (ZnO) thin films were deposited on silicon substrates using thermal evaporation method. These films were annealed at 400°C for 15 minutes to support the adhesion of these films to the substrates as well as to enhance the structural characteristics of these films. The ZnO/Si structures were irradiated with pulses from a Q-switched Nd:YAG laser operating at 1064nm, 300µs, and 10 repetition rate at 45° incident angle with respect to the normal on the surface of sample. The photoluminescence (PL) spectra of these samples were recorded, analyzed and compared to introduce the enhancement as a result of laser irradiation.

Keywords: Zinc oxide; Laser irradiation; Photoluminescence; Inclined irradiation
Received: 22 October 2024; **Revised:** 25 November; **Accepted:** 16 December; **Published:** 1 January 2025

1. Introduction

Zinc oxide (ZnO) is a versatile material with unique properties, making it suitable for various applications. It is a wide-bandgap semiconductor with high chemical stability, high electrochemical coupling coefficient, and broad range of radiation absorption [1-5]. ZnO can be prepared using several methods, including wet chemical method, solid-state reaction method, and vapor phase deposition method [6-8]. ZnO finds applications in transparent conductive oxides, UV absorbers, varistors, chemical sensors, surface acoustic wave devices, spintronics, and biomedical applications [9-14].

Irradiating semiconductors with laser beams can modify their properties, leading to enhanced spectroscopic and structural characteristics [15]. The laser beam interacts with the semiconductor material, causing localized heating and rapid cooling [16,17]. This process can induce various effects, such as improved crystallinity, formation of nanostructures, enhanced optical and electrical properties, and doping and defect engineering [18-21].

These modifications can improve the performance of semiconductor devices in applications such as photovoltaics, light-emitting diodes, lasers, and sensors [22-24].

ZnO/Si multilayer structures offer several advantages in photonics and optoelectronics due to the unique properties of each material. ZnO is a wide-bandgap semiconductor with high optical transparency and efficient UV emission [25,26]. Silicon is a well-established semiconductor with excellent electronic properties and low cost [27,28]. Combining these materials in a multilayer structure can lead to enhanced light extraction efficiency in LEDs, improved UV detection in photodetectors, integration of optical and electronic functionalities on a single chip, and development of novel photonic devices [29-32].

Specifically, ZnO/Si structures can be used in UV LEDs, UV photodetectors, optical waveguides, and solar cells. Overall, ZnO/Si multilayer structures hold great promise for future developments in photonics and optoelectronics [33-36].

In this work, the effects of inclined irradiation of ZnO/Si multilayer structures with Nd:YAG laser pulses on the photoluminescence (PL) characteristics of these structures were studied.

2. Experimental Part

Silicon wafers were cut into desired sizes (2.5x2.5cm), and then cleaned thoroughly using the RCA cleaning procedure to remove organic contaminants and metallic impurities. This involves sequential cleaning in solutions of ammonium hydroxide (NH_4OH), hydrochloric acid (HCl), hydrogen peroxide (H_2O_2), and deionized water (H_2O). These substrates were dried and rinsed with deionized water and dried under a nitrogen (N2) stream.

Thermal evaporation method was used to deposit ZnO thin films on the Si substrates. The cleaned silicon substrates were loaded into thermal evaporation system. This system consists of a vacuum chamber, a resistive heating source (usually a tungsten boat or basket), and a rotating substrate holder. The source material, which is high-purity ZnO powder, was placed in the heating source. The chamber was evacuated to a high vacuum (typically around 10^{-6} Torr) to minimize contamination and ensure a long mean free path for the evaporated ZnO molecules. The source material is heated by passing a high current through the heating source. The ZnO will evaporate and deposit onto the silicon substrates. The deposition rate and film thickness are controlled by adjusting the source temperature and deposition time. A quartz crystal microbalance can be used to monitor the deposition rate and thickness. The substrate temperature can be controlled during deposition. Higher substrate temperatures can improve the crystallinity of the ZnO films. Annealing the ZnO/Si samples in air or oxygen at high temperatures can improve the crystallinity and optical properties of the films.

The x-ray diffraction (XRD) is a powerful technique for determining the crystalline structure, crystallite size, and preferred orientation of thin films. It involves irradiating the sample with X-rays and analyzing the diffraction pattern. The ZnO/Si sample is mounted in the XRD instrument, irradiated with X-rays (typically Cu Kα radiation). The intensity of the diffracted X-rays is measured as a function of the diffraction angle (2θ). The obtained diffraction pattern is then compared with standard JCPDS (Joint Committee on Powder Diffraction Standards) data to identify the crystalline phases present in the film. The crystallite size is determined using the Scherrer's equation to estimate the average crystallite size from the broadening of the diffraction peaks. Analysis of the relative intensities of the diffraction peaks is performed to determine if the film has a preferred orientation.

Photoluminescence (PL) spectroscopy is used to study the optical properties and defect states in the ZnO thin films. It involves exciting the sample with a laser and analyzing the emitted light. The ZnO/Si sample is excited with a laser of appropriate wavelength (e.g., a He-Cd laser with a wavelength of 325 nm). Collect the emitted light using a spectrometer. The near-band-edge (NBE) emission is observed in the UV region, which is due to the recombination of free excitons. Similarly, the defect-related emissions is observed in the visible region, such as green emission is attributed to oxygen vacancies, and yellow emission is attributed to zinc vacancies or interstitial oxygen. The peak positions and intensities are analyzed to gain information about the defect types and concentrations in the ZnO films.

By combining these experimental and characterization techniques, you can gain a comprehensive understanding of the structural and optical properties of ZnO thin films deposited on silicon substrates using thermal evaporation. This knowledge is crucial for optimizing the deposition process and tailoring the properties of the films for various applications in optoelectronics and photonics.

Figure (1) shows the principle of irradiation the samples prepared in this work with a Q-switched Nd:YAG laser pulses at 45° with respect to the normal on the sample's surface. The laser is operated at 1064nm, 300μs pulse duration, and 10 repetition rate.

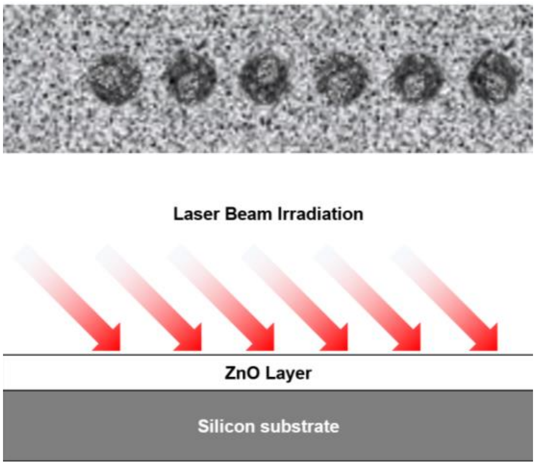

Fig. (1) Principle of inclined irradiation of the ZnO/Si structure with laser pulses (upper photo shows the irradiation spots on ZnO layer)

3. Results and Discussion

The XRD pattern shows a dominant peak at approximately 34.4°, corresponding to the (002) plane of hexagonal ZnO, indicating a strong c-axis preferred orientation. A smaller peak around 36.2° suggests the presence of the (101) plane. This confirms the crystalline nature of the ZnO thin film with a dominant orientation along the c-axis.

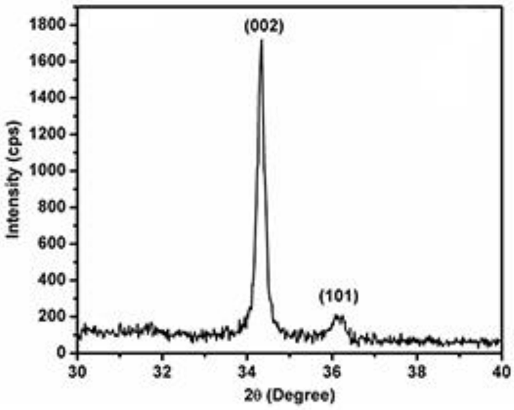

Fig. (2) XRD pattern of ZnO layer prepared in this work

The PL spectra of ZnO thin films irradiated with different laser power densities show a clear trend. As the laser power density increases, the intensity of the near-band-edge (NBE) emission peak around 380 nm increases significantly. This indicates that higher laser power densities can enhance the radiative recombination processes in the ZnO films, leading to stronger UV emission. Additionally, the intensity of the defect-related emission peaks in the visible region (around 500-700 nm) also increases with increasing laser power density. This suggests that laser irradiation can introduce or activate defects in the ZnO films, which can act as radiative recombination centers.

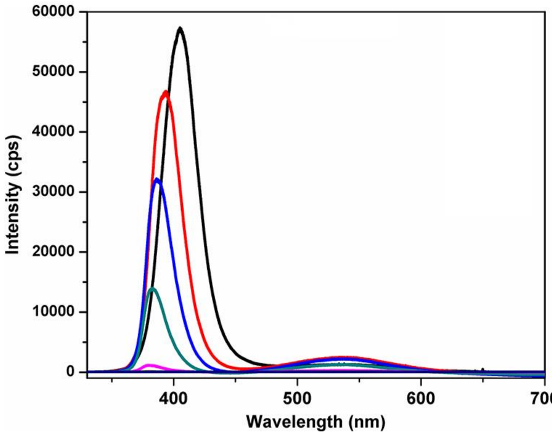

Fig. (2) PL spectra of ZnO/Si samples irradiated with different laser power densities (Black: 80 W/cm^2, Red: 60 W/cm^2, Blue: 40 W/cm^2, Green: 20 W/cm^2, Pink: not irradiated)

4. Conclusion

In conclusions, ZnO/Si structures were irradiated with pulses from a Q-switched Nd:YAG laser operating at 1064nm, 300µs, and 10 repetition rate at 45° incident angle with respect to the normal on the surface of sample. The photoluminescence (PL) spectra of the irradiated samples showed a redshift in the PL peak with increasing laser power density, which highlights a reasonable enhancement in the spectroscopic characteristics of such structures.

References

[1] Peter M. Martin, **“Introduction to Surface Engineering and Functionally Engineered Materials”**, John Wiley & Sons, Inc. (NJ), 2011, p. 262-264.
[2] M. Ohring, **"The Materials Science of Thin Films"**, Academic Press, 1992, Ch. 4, p. 182.
[3] S.M. Sze, J. Appl. Phys., 38 (1967), p. 2951
[4] S. Pujar, G.K. Rao, and M.G. Mahesha, Opt. Mater., 157(Pt 1) (2024) 116141.
[5] M.A. Hameed and Z.M. Jabbar, Iraqi J. Appl. Phys., 12(4) (2016) 13-18.
[6] H. Lorentz et al., J. Vac. Sci. Technol. B, 9 (1991), p. 208

[7] S.W. Hseih et al., J. Appl. Phys., 76 (1994), p. 3645
[8] A.M. Hameed and M.A. Hameed, Iraqi J. Appl. Phys., 18(4) (2022) 9-14.
[9] J.G. Simmons, J. Phys. D: Appl. Phys., 4 (1971), p. 613
[10] K. Mensah-Darkwa et al., Physica B: Cond. Matter, 667 (2023) 415155.
[11] O.A. Hamadi, N.J. Shakir and F.H. Mohammed, Bulg. J. Phys., 37(4) (2010) 223-231.
[12] M.A. Lampert, Rep. Prog. Phys., 27 (1964), p. 329
[13] E.C. Paloura, J. Lagowski, H.C. Gatos, J. Appl. Phys., 69 (1991), p. 3995
[14] K. Tanabashi, K. Kobayashi, Jpn. J. Appl. Phys., 12 (1973), p. 641
[15] M.K. Khalaf et al., Iraqi J. Mater., 1(2) (2022) 57-66.
[16] R.D. Gould, J. Appl. Phys., 53 (1982), p. 3353
[17] R. Parasuraman and K. Rathnakannan, Memories – Mater. Devices Circuits Syst., 8 (2024) 100114.
[18] A.M. Hameed and M.A. Hameed, Emergent Materials, 6 (2022) 627-633.
[19] R.O. Ocaya et al., Heliyon, 9(5) (2023) e16269.
[20] O.A. Hammadi, M.K. Khalaf and F.J. Kadhim, Opt. Quantum Electron., 47(12) (2015) 3805-3813.
[21] R. Del Sole et al., Appl. Surf. Sci., 684 (2025) 161875.
[22] H.R. Ali, Iraqi J. Mater., 1(3) (2022) 107-116.
[23] K.T. Alan et al., Adv. Mater. Res., 47-50 (2008) 1498-1501.
[24] O.A. Hammadi, Photon. Sens., 5(2) (2015) 152-158.
[25] M.A. Hameed, S.H. Faisal, R.H. Turki, Iraqi J. Appl. Phys., 16(4) (2020) 25-30
[26] O.A. Hammadi, J. Optoelectron. Photon., 7(5) (2016) 21-24.
[27] M. Mohammadnezhad et al., Mater. Adv., 3(14) (2022) 5911-5921.
[28] O.A. Hammadi, Photonic Sensors, 6(4) (2016) 345-350.
[29] O.A. Hammadi, Optoelectron. Lett., 18 (2015) 111-116.
[30] X.-F. Zhang, P.-G. Wen, Y. Yan, Proc. SPIE 7995, 7th Inter. Conf. on Thin Film Physics and Applications, 79951M (February 17, 2011).
[31] O.A. Hammadi, Iraqi J. Phys., 2(3) (2003) 1-6.
[32] Y.A. Baydhon, Iraqi J. Mater., 1(3) (2022) 139-144.
[33] A.A. Mahmood et al., Indonesian J. Chem., 22(1) (2022) 205-211.
[34] L.P. Ward et al., Proc. of the Inst. of Mech. Eng., Part H: J. of Eng. in Medicine, 212(4) (1998) pp. 303-315.
[35] E. Camps et al., 16th IAEA Technical Meeting on Research using Small Fusion Devices, AIP Conf. Proc. 875, pp. 161-164.
[36] H. Kovaci et al., 13th Inter. Conf. on Plasma Surface Eng., Sept. 10-14, 2012, Germany.