Optical and structural study of low dose alpha irradiated zinc oxide (ZnO) thin film

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In this paper, the effect of low-dose alpha particles irradiation on the optical and structural properties of different thicknesses ZnO thin films was investigated. Preferentially oriented (002) ZnO thin films were prepared using radio frequency (RF) magnetron sputtering where Si (100) and glass were chosen as substrates. ZnO films with c-axis-oriented Würtzite structure are obtained at room temperature. The effect of the irradiation was tested on ZnO films prepared with two different thickness of 300 nm and 700 nm representing thin and thick film, respectively. The irradiation was realized using a 5-MeV alpha particles beam emitted from an Americium-241 alpha sources (Am-241). The films were irradiated to different radiation doses (1.5 to 25 Gy). The obtained results showed that the PL intensity was found to increase with increasing of the radiation dose in both thin and thick ZnO films. This finding allows improving the scintillation output signal of the material. Moreover, the X-ray diffraction patterns did not show any changes in the grain size. However, the band gap extracted by UV transmittance spectrum was found to be slightly decreased with increasing the radiation dose.

(Received September 2, 2018; accepted June 14, 2019)

Keywords: ZnO film, Magnetron sputtering, Alpha irradiation, Optical characterization

1. Introduction

Zinc oxide (ZnO) is a direct wide-band gap material of 3.37 eV, with an exciton binding energy around 60 meV at room temperature [1] [2]. For this reason, ZnO is considered to have high-quality electronic properties compared to classical semiconductors as Si and Ga [2]. As, with its components being exuberantly existing in nature, inexpensive, nontoxic, very environmentally stable, highly conductive and transparent as well, and high sensitivity in the ultraviolet energy range (UV). ZnO is thus useful as transparent electrode in solar cells [2] crystal display (LCD) [3], and light emitting diode (LED) [4]. Moreover, ZnO can be used in thin film transistors (TFT) [5] [6] , gas sensor [7], laser diode and UV photodetector [8].

ZnO has also several applications in photovoltaic where it can act as electron transport material (ETM) in dye-sensitized solar cells (DSC) and hybrid solar cells (HSC) [9]. It can also applied as antireflection coating in inorganic solar cells [10] [11], or as optical spacer in polymer solar cells [12] [13] [14] [15].

The growing uses of conductive transparent films for optoelectronic device such as solar cells and displays have increased the interest in developing various techniques of film preparation. ZnO could be technologically considered as one of the important scintillation materials. It has attracted great attention due to its interesting optical and electrical properties. ZnO is a promising material due to its availability and low coast. However, the stability of ZnO thin films under high irradiation conditions and at high temperature is still a matter of research [16].

Interaction of ionizing radiation with matter, especially alpha particles, is very important theoretically and practically. Heavy charged particles, such as alpha particle, interact with the orbital electrons of the absorber atoms through coulomb forces [17]. Interactions of the particle with nuclei are also possible. These interactions could produce changes in the microstructural properties of the material [18], which could also affect the optical [19] and other physical properties of the material [20] [21] [22]. Prabukanthan et al [23] observed that the intensity and FWHM of the (002) diffraction peak in the case of ZnO thin films irradiated with 100 MeV O^{7+} ions are comparable to those of the pristine film.

For AgGaS2 single crystals grown irradiated with Ag9+ ions (120 MeV) with various ion fluencies, there was no change in the FWHM of the (1 1 2) peak but the intensity was slightly decreased at LNT irradiation [24].

Because alpha particles are relatively heavy and have a positive charge, they interact strongly with matter, and produce a large number of ions per unit length of their path. As a result, they are not very penetrating. Alpha particles with energy of 5 MeV will only travel about 14 μm in Zinc oxide (calculated by SRIM software).

Numerous researches have recently been investigated the influence of the irradiation with gamma and alpha rays on thin films prepared using different metal oxides and polymers, in order to study the possibility of using them as

radiation detectors [25] [26] [27] [28]. In addition, Physical properties of ZnO are strongly dependant on the growth parameters and the post deposition treatments [29].

The aim of this work is to study effect of Alpha irradiation on optical and structural properties of ZnO films deposited on both Si and glass substrates, and for thin and thick films corresponding to 300 nm and 700 nm respectively.

2. Experimental procedure

Zinc oxide films have been elaborated by RF magnetron sputtering using a PLASSYS setup equipped with an embedded microbalance to control the deposition thickness. The quality of the prepared films is sensitive to deposition parameters. These parameters have been optimized in order to get highly oriented ZnO films. The ZnO films were deposited on 100 silicon wafers and glass substrates. 15 cm diameter zinc oxide target of 99.99% purity was used. The cathode to substrate-holder distance was 6 cm. The deposition chamber was pumped down to a base pressure of 2×10-7 Torr by a turbo-molecular pump prior to the introduction of argon gas. The RF power and the pressure were 600 W and 1.5 mTorr respectively [30]. The deposition rate (1.23nm/s) at RF power 600 W and morphology were obtained from a scanning electron microscope (SEM) type TSCAN Vega\\XMU (Czech Republic) operated at 30 kV. Atomic composition and stoichiometry of the ZnO films were determined by EDX. In addition, All the deposition conditions are summarized in previous work [30].

The crystallographic properties of the films were analyzed by X-ray Diffraction (XRD) using the CuKα (with λ = 1.5405 A°) radiation. The optical characteristics were investigated using a UV-vis Shimadzu UV-310PC Spectrophotometer where the transmittance of films was measured. PL measurements were used to characterize the ZnO thin films. The experimental set-up consists of UV excitation source which is a 325 nm He/Cd laser and grating monochromator with 1200 groves/mm provided with a cooled photomultiplier tube PMT. Synchronous detection technique was applied by chopping the laser beam and employing a lock-in amplifier to process the electrical output of the PMT. The excitation beam was guided and focused on the tested sample using a set of optical devices.

The effect of alpha beam irradiation was examined by exposing the ZnO thin films to different doses: 1.5, 2.8, 5.5, and 25 Gy by using Am-241 alpha source with activity of 9.0 kBq measured by an alpha-spectrometry (OasisTM) Oxford Alpha Spectroscopy Integrated Systems (Version 3.5) Detector Type ICP 500-100-19EM)] USA 1993-1994. Optical transmittance and photoluminescence spectroscopy have been used to study the physical changes induced by the alpha irradiation.

3. Results and discussions

3.1. Structural and morphological properties

Fig. 1 shows the XRD pattern of three 300 nm thickness ZnO/ Si samples: non irradiated, irradiated to a 1.5 Gy dose and irradiated to 25 Gy dose. In all the cases, one strong peak at 2θ ~ 34.4° is observed. It can be attributed to the (002) peak of the hexagonal ZnO Würtzite phase [31]. All the ZnO films are highly textured, with the c-axis perpendicular to the substrate surface.

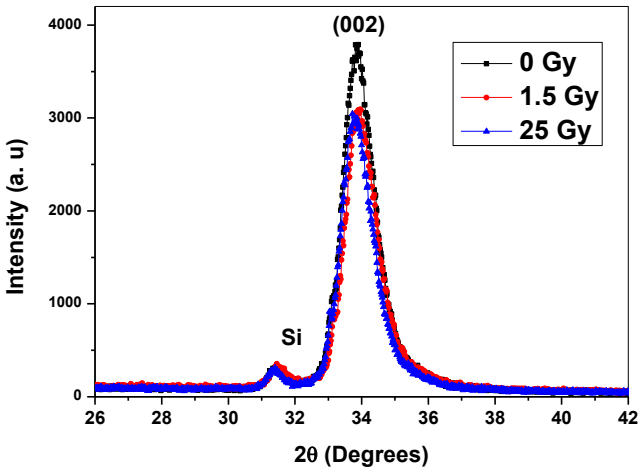

Fig. 1. XRD patterns of ZnO film exposed to 0, 1.5 and 25 Gy alpha particles doses

The mean grain size d is obtained by the Scherrer formula [30], d = 0.94 λ/(B cosθ), where λ,θ and B are the X-ray wavelength (1.54 A˚), the Bragg diffraction angle, and the peak width at half maximum of the peak (FWHM) respectively. The grain size was a 17 nm for non-irradiated film. No significant changes were noted for the irradiated films in comparison with the non-irradiated films [24]. Moreover, the peak intensity is observed to increase and the grain size is the highest at for ZnO film non-irradiated. The obtained results could be explained by the low alpha particles doses used in our work. Prabukanthan et al [23] observed that the intensity and FWHM of the (002) diffraction peak in the case of RT irradiated ZnO thin films are comparable to those of the pristine film.

Y-K. Moon et al [32] have observed that the grain size of the ZnO thin films decreased from 14 nm to 10 nm with increasing proton irradiation dose by using a high- energy 6.1-MeV beam at a dose of 1012 - 1014 protons- cm-2. They

have reported that only the grain size was affected, while no changes were noted in the direction of orientation [32].

3.2. Optical properties

3.2.1. UV study

Fig. 2 (a) represents the optical transmittance of ZnO films deposited at 600 W of sputter power as a function of wavelength. The optical transmittance could provide useful indication about the optical band gap of the semiconductor [33].

Fig. 2 (a) shows that the average transmittance of samples deposited on glass in the visible and near infrared range is varied from 92% (non-irradiated film) to 80% (25 Gy dose irradiated film) for ZnO film of 300 nm thickness.

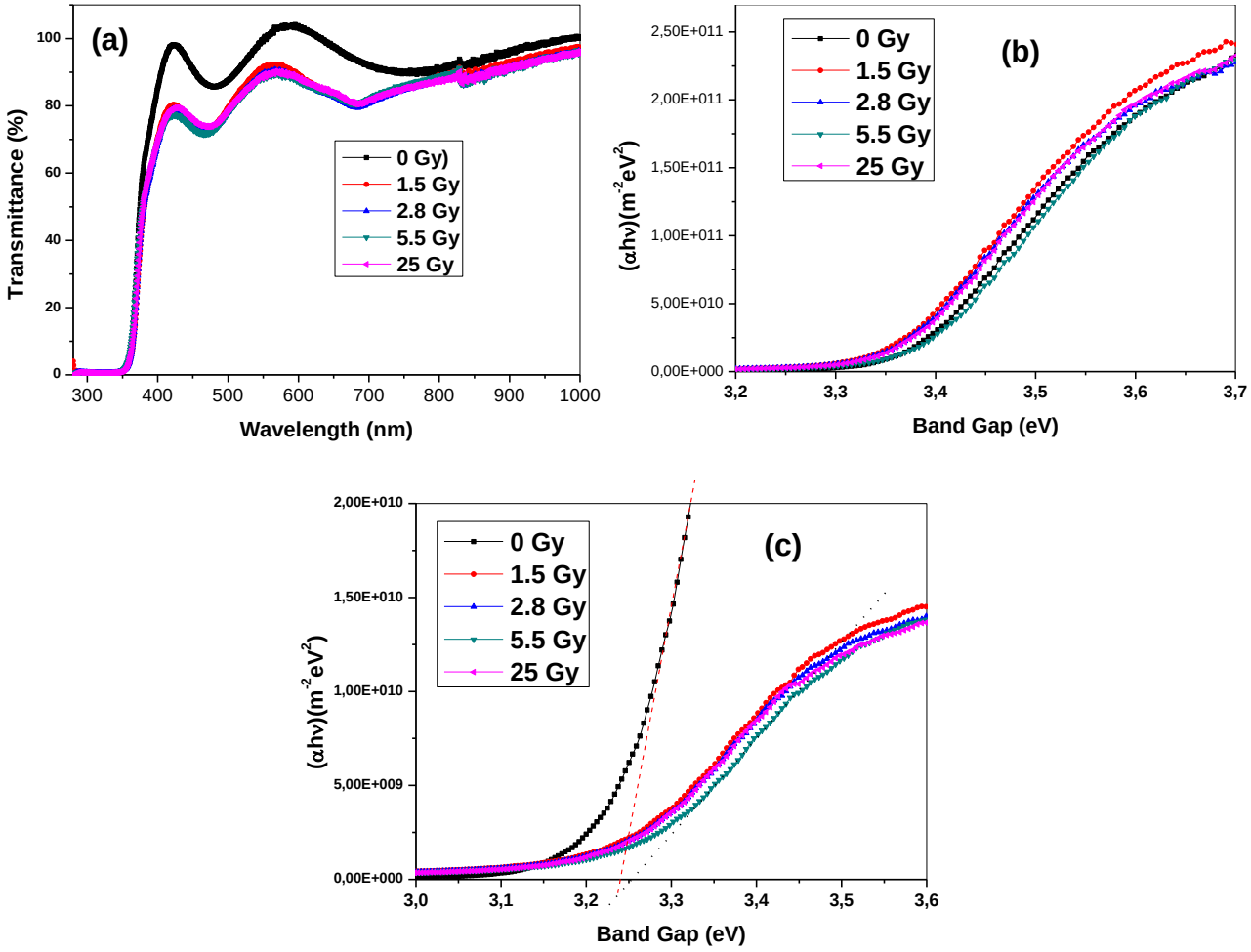

Fig. 2. (a) The transmittance spectra, (b) the corresponding optical band gap of ZnO 300 nm and (c) Optical band gap of ZnO for film at 700 nm

Fig. 2 (b) shows the slightly decrease of the optical band gap from 3.37 eV for the non-irradiated film to 3.36 eV for the 25 Gy dose irradiated film.

For the 700 nm ZnO film, the optical band gap also decreases when the irradiation dose increases as shown in Fig. 2 (c). The value of the optical band gap could be related to the nanoscopic size of crystallites [34] [35] .

3.2.2. PL study

Najiba et al. [35] found that the value of the band gap was about 3.25 eV for un-irradiated ZnO films and 3.2 eV after 0.662 MeV Gamma rays irradiation for 21 days. The band gap decrease shows a slight ‘red shift’ in the optical spectra caused by irradiation. This decrease could be related to the increase in the energy width of the band tails of localized states. Similar results have been reported by other researches [36] [37] [38]. While, J.C. Nappé et al [39] did

not note any new defect created by the irradiation for AlN under Swift Heavy Ion Irradiations.

3.3. Photoluminescence study

Fig. 3 (a-b) shows room temperature PL spectra of the 300 nm and 700 nm thickness ZnO films. ZnO exhibits two emission Bands: the first is around λ = 384 nm (~3.23 eV), which is attributed to band-to-band emission in the UV region. Peaks at about 420, 436 nm could be related to Zn vacancy and interstitial respectively (blue region) [40], and a broad peak at about 550 nm which could be related to oxygen vacancies or interstitials (green region). The optical band gap of ZnO is affected by O_2 vacancies and Zn interstitial atoms as well as the grain size [1].

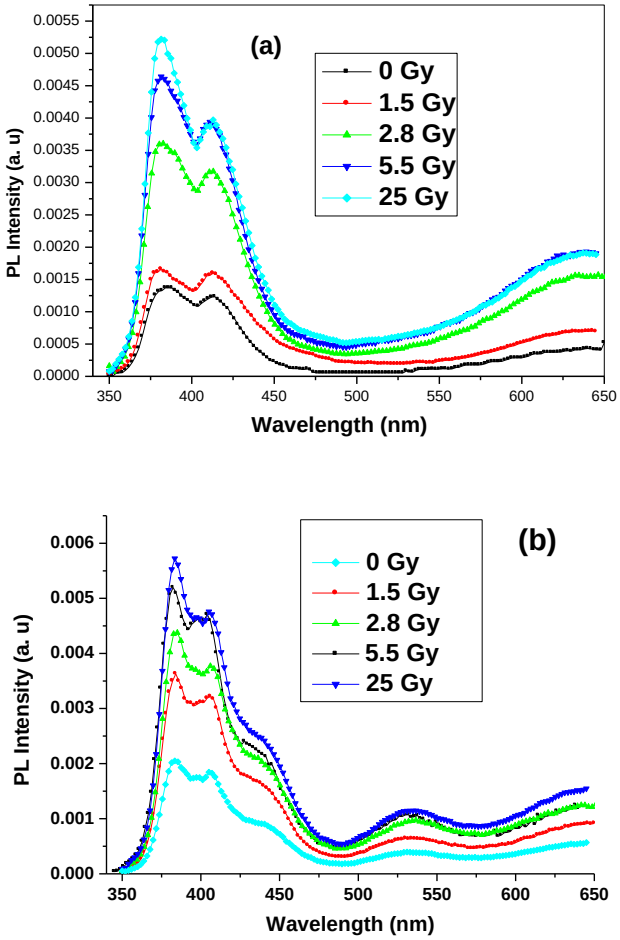

Fig. 3. PL spectra obtained from as deposited and irradiated ZnO thin films (a) at 300nm thickness and (b) at 700nm thickness

The intensity of the PL spectra (387 and 420nm) was increased with increasing the alpha irradiation dose for both 300 nm and 700 nm thickness ZnO film (Fig 3). Increasing of intensity can be indicating to the degradation of quality and increasing the defect in the film. The FWHM increases and the related emissions of the Zn, O_2 vacancy and interstitial are more clearly appeared for higher alpha dose. It could be attributed due to the decreasing of the film crystallinity i.e. the intensity of UV emission could be considered as an indicator to the crystallinity of the ZnO film, and the higher crystallinity possesses the higher intensity of UV emission [41] [42], only in thin film as it is not the case for thicker film [32]. In previous work has been investigated the applicability of using the radio-luminescence of Si-doped Gallium Nitride GaN(Si) for the detection of alpha particles [43].

Fig. 4 shows the variation of the PL intensity as a function of Alpha irradiation dose for 300 nm and 700 nm thicknesses ZnO films.

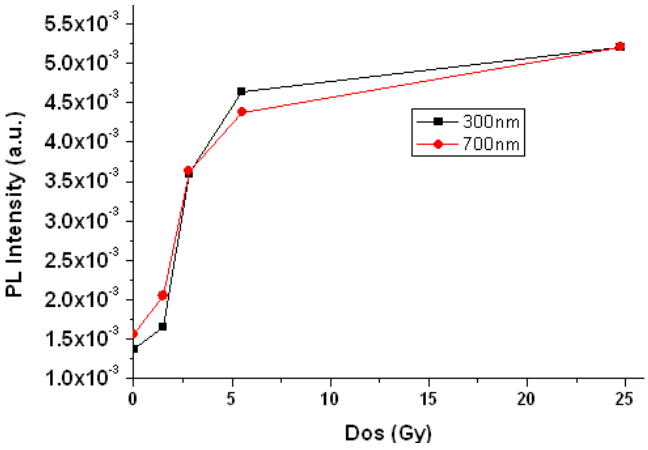

Fig. 4. PL intensity as a function of dose Alpha irradiation for ZnO film deposited at 300 nm and 700nm

Fig. 4 shows a quasi-linear relation between the PL intensity and the irradiation dose up to dose around 5.5 Gy where PL intensity increased threefold. Afterward the PL intensity starts to reach the saturation. These results showed that the PL intensity has increased with increasing the irradiation dose leading to improve the scintillation output signal of the material and thus improving the scintillation properties of the ZnO films. This could be of great importance for various optoelectronic applications

4. Conclusions

The effect of alpha particles irradiation on the structural and optical properties of ZnO thin films, prepared by RF-magnetron sputtering, has been investigated. The XRD results did not show any significant changes in the grain size after alpha irradiation. The optical band gap of the ZnO films deposited on glass has been extracted from UV transmittance spectra. It was found that the band gap has slightly decreased after alpha irradiation for both thickness 300 nm and 700 nm. Finally, it is noteworthy to mention that

optical parameters of the films (PL intensity) are modified by alpha irradiation which allowed to improve the scintillation intensity (even at small radiation doses), whereas the structure of the film has not witnessed any significant modification in term of grain size or preferred orientation.

Acknowledgements

The authors gratefully acknowledge Prof. I. Othman (Director General of AECS) and Prof. M.S. Al-Masri (Head of Protection and Safety Department), for their continuous encouragement and support.

References

[1] B. Abdallah, S. Al-Khawaja. Acta Physica Polonica A **128**(3), 283 (2015).
[2] Z. Wang, J. Chu, H. Zhu, Z. Sun, Y. Chen, S. Huang, Solid State Electronics **53**(11), 1149 (2009).
[3] B. Oh, M. Jeong, M. TH, W. Lee, J. Myoung, J. Hwang, Journal of Applied Physics **99**(12), 124505 (2006).
[4] S. Pearton, W. Lim, J. S.Wright, L. Tien, H. Kim, D. Norton, Journal of Electronic Materials **37**, 1426 (2008).
[5] R. Hoffman, B. Norris, J. Wager, Applied Physics Letters **82**(5), 733 (2003).
[6] J. Zhu, H.Chen, G. Saraf, Z. Duan, Y. Lu, S. T. Hsu, Journal of Electronic Materials **123**, 701 (2008).
[7] V. R. Shinde, T. P. Gujar, C. D. Lokhande, Sensors and Actuators B: Chemical **123**(2), 701 (2007).
[8] P. K. Basu, P. Bhattacharyya, N. Saha, H. Saha, S. Basu, Sensors and Actuators B: Chemical **133**(2), 357 (2008).
[9] I. Gonzalez-Valls, M. Lira-Cantu, Energy and Environmental Science **2**(1), 19 (2009).
[10] Y.-J. Lee, D. S. Ruby, D. W. Peters, B. B. McKenzie, J. W. P. Hsu, Nano Letters **8**(5), 1501 (2008).
[11] J. Y. Chen, K. W. Sun, Solar Energy Materials and Solar Cells **94**(5), 930 (2010).
[12] F. C. Krebs, S. A. Gevorgyan, J. Alstrup, Journal of Materials Chemistry **19**(30), 5442 (2009).
[13] F. C. Krebs, Solar Energy Materials and Solar Cells **92**(7), 685 (2008).
[14] S. Tomoki, U. Tokiyoshi, H. Yuuki, F. Akihiko, Y. Katsumi, Journal of Physics D: Applied Physics **37**(6), 847 (2004).
[15] C.-H. Hsieh, Y.-J. Cheng, P.-J. Li, C.-H. Chen, M. Dubosc, R.-M. Liang, C.-S. Hsu, Journal of the American Chemical Society **132**(13), 4887 (2010).
[16] J. J. Schneider, R. C. Hoffmann, J. Engstler, O. Soffke, W. Jaegermann, A. Issanin, A. Klyszcz, Advanced Materials **20**(18), 3383 (2008).
[17] G. F. Knoll, Radiation, detection, and measurement, USA, John Wiley & Sons, (2010).
[18] P. Prabukanthan, K. Asokan, D. K. Avasthi, R. Dhanasekaran, Materials Science in Semiconductor Processing **10**(6), 252 (2007).
[19] P. Prabukanthan, R. Lakshmi, G. Harichandran, T. Tatarchuk, New Journal of Chemistry **42**(14), 11642 (2018).
[20] A. M. Ibrahim, L. I. Soliman, Radiation Physics and Chemistry **53**(5), 469 (1998).
[21] E. Atanassova, A. Paskaleva, R. Konakova, D. Spassov, V. F. Mitin, Microelectronics Journal **32**(7), 553 (2001).
[22] R. L. Clough, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **185**(1), 1 (2001).
[23] P. Prabukanthan, G. Harichandran, Materials Science in Semiconductor Processing **16**(1), 193 (2013).
[24] P. Prabukanthan, K. Asokan, D. Kanjilal, R. Dhanasekaran, Semicond. Sci. Technol. **23**(125042), 1 (2008).
[25] E. Colby, G. Lum, T. Plettner, J. Spencer, IEEE Transactions on Nuclear Science **49**(6), 2857 (2002).
[26] K. Arshak, A. Arshak, S. Zleetni, O. Korostynska, IEEE Transactions on Nuclear Science **51**(5), 2250 (2004).
[27] K. I. Arshak, O. Korostynska, J. Molloy, J. Harris, IEEE Sensors Journal **6**(3), 656 (2006).
[28] K. Arshak, O. Korostynska, Materials Science and Engineering B **133**(1), 1 (2006).
[29] S. Ambily, C. S. Menon, Thin Solid Films **347**(1), 284 (1999).
[30] S. Al-Khawaja, B. Abdallah, S. Abou Shaker, M. Kakhia, Composite Interfaces **22**(3), 221 (2015).
[31] C. Messaoudi, D. Sayah, M. Abd-Lefdil, Physica Status Solidi A **151**(1), 93 (1995).
[32] Y.-K. Moon, D.-Y. Moon, S. Lee, J.-W. Park, Journal of Korean Physical Society **54**(3), 1059 (2009).
[33] B. Abdallah, A. K. Jazmati, R. Refaai, Materials Research **20**, 607 (2017).
[34] G. G. Valle, P. Hammer, S. H. Pulcinelli, C. V. Santilli, Journal of the European Ceramic Society **24**(6), 1009 (2004).
[35] N. A. A. Hamdani, R. D. Al-Alawy, S. J. Hassan, IOSR Journal of Computer Engineering (IOSR-JCE) **16**(1), 11 (2014).
[36] S. L. Sharma, T. K. Maity, Bull. Mater. Sci. **34**(1), 61 (2011).
[37] T. J. Alwan, Turk J. Phys. **36**, 377 (2012).

[38] T. K. Maity, S. L. Sharma, Indian Journal of Pure and Applied Physics **49**, 606 (2011).
[39] J. C. Nappé, P. Grosseau, M. Benabdesselam, M. Beauvy, B. Guilhot, Proceedings of the 11th ECERS Conference, Cracovie, Poland, (2009).
[40] K. Y. Wu, Q. Q. Fang, W. N. Wang, C. Zhou, W. J. Huang, J. G. Li, Q. R. Lv, Y. M. Liu, Q. P. Zhang, H. M. Zhang, Journal of Applied Physics **108**(6), 063530 (2010).
[41] Y. G. Wang, S. P. Lau, H. W. Lee, S. F. Yu, B. K. Tay, Journal of Applied Physics **94**(1), 354 (2003).
[42] C. Ting, Nanorods. Rijeka: InTech Open, 2012, p. 33.
[43] A. Ismail, A., R. Shweikani, B. Jerby, Optoelectron. Adv. Mat. **16**(1-2), 47 (2016).

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