OPTICAL INVESTIGATIONS AND OPTICAL CONSTANTS OF NANO SILVER OXIDE PREPARED BY PLD METHOD

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ABSTRACT:*High purity Ag_2O (silver Oxide) thin films with high transparent conductive properties will be deposited successfully by reactive pulsed laser deposition method. The Optical property shows a high transparency reached to about (90) % and found to decreases sharply with decreasing of laser fluency. The optical energy band gap of the deposited film at the optimum prepared condition is around 3.46eV. The prepared samples were analyzed using the UV-visible and photoluminescence. The results give an indication that the deposited films crystallize to be more regular in the distribution of atoms, and the energy band gap approaches 3.47 eV when the laser fluency decrease. The optical investigations and constant of the energy band gap, the refractive index, and the extinction coefficient were also elaborated.*

Keywords: Silver Oxide, Optical properties, Optical constants, PLD technique.

I. INTRODUCTION

One of the most important p-type semiconductors with a direct band gap around 1.4 eV is the Silver oxide (Ag_2O) [21-23] thin films of this material have some unique physical properties that have attracted much interest.[1–4]. They have been used widely in the batteries electric pole and in the photography [5]. And in the SERS (surface-enhanced Raman spectroscopy) and in a surface-plasmon (SP) source. Many methods can be used to prepare and deposit the AgOx, such as the (CBD) chem.-bath dep. [3, 6], thermal evaporation and (RMS) reactive magnetron sputtering, exposing the films of silver to atomic oxygen environment, and Pulsed Laser Deposition techniques [7, 8].

The reactive Pulsed Laser Deposition using a pure silver targets while feeding it with oxygen gas as a catalyst to increase the interaction between pure silver atoms and gas to obtained and controlling to the oxygen composition ratio and to allowed to use a low energy of lasers for deposition [9,10]. The optical properties of AgOx have been studied previously [11], the correlation between the microstructural features and the process parameters during deposition has not yet been well established. In this work, we studied the optical properties of Deposited AgOx thin films on the glass substrate. The additional analysis by X-ray diffraction results [12, 13].

In this paper, we present the PLD as a technique to deposite Ag_2O over substrate of glass. The research continued to study and analyze the effect of Laser fluincy on the growth mechanism of the material and, thereafter, transmission, reflection, bandgap and refractive index are presented and discussed deeply.

II. MATERIAL AND METHODS

Undoped Ag_2O thin films were prepared and deposited on a cleaned slide of glass as a substrate using tattoo removal Nd: YAG laser. The pulse duration of the Q-switched Nd: YAG laser is 7 ns (FWHM) with wavelength= 1.064 nm, the laser beam was focused through a lens with a focal length=10 cm spotted on a silver target (99.999% provided from Fluka com.). The targets spin at a rate of one cycle per minute. The energy density of a pulse laser at the target surface was maintained within the range 43 and 57 mJ/cm^2. All prepared thin films were created by 50 laser shots at the temperature of the substrate is 150 C^o. The thicknesses of the deposited thin film were tested using the scanning optical reflectometer. The optical properties such as the transmittance of the prepared films were investigated at spectral range (300–1000) nm using UV-VIS Shimatzu double beam spectrophotometer.

The value of the incident energy of photon was studded as a function of (λ) depending on the formula [14-16]

$E_g(eV)=1240/\lambda(nm)$ (1)

whereλ is wavelength. The energy gaps are dependence on the value of the absorption coefficient (α) and the excitation of the transition could be described depending on the Tauc formulas [17, 18]

$(\alpha h\nu)=B\,(h\nu–E_g)r$ (2)

where the B is a constant inversely proportional to amorphous, r is a constant = 2, 3, etc… depending on the raw used materials and the type of the optical transition $\alpha h\nu$. If the straight part of the plot of the $(\alpha h\nu)^{1/r}$ against ($h\nu$) is extrapolated to $(\alpha h\nu)^{1/r}$ =0, the intercept gives the energy gap value. The value of the absorption coefficient (α) for all wavelengths was examined by [19, 20]

$\alpha=2.303(A/t)$ (3)

where$A$ is absorbance and t is transmittance.

with A is the absorption and t is the thickness of nanophotonic films. Equation (4) relates the absorption coefficient (α) with the excitation ratio (K) [21, 22]:

$$K = \alpha\,\lambda / 4\pi \quad (4)$$

All other constants such as refractive index (n), the ε_r and ε_i which they relate to the real and imaginary parts of the dielectric constants, the conductivity (σ) are used to calculated from equation (5) [23, 24]:

$$n = \{[4R / (R-1)]-K^2\}^{1/2} – [(R+1) / (R-1) \quad (5)$$

Note that R represents the reflectance which can be found from (R = 1-T-A), or it can be obtained from equation (6) [25, 26]:

$$n=n_s\left(\frac{1+\sqrt{R}}{1-\sqrt{R}}\right)^{1/2} \quad (6)$$

where n_s denotes to the substrate refractive index. It is an important to mention that the refractive index of the glass substrate is considered to be 1.51, [27, 28]:

III. RESULT AND DISCUSSION

UV-VIS optical spectra for the transmission of the thin film deposited and preparedunder different laser fluency conditions are present. The optical properties of deposited Ag_2O_3 at two different laser fluence could be shown in Figure (1). In general, a decrease in the optical transmission as a function of wavelength could be recognized with increasing with laser fluence due to the increase in laser ablation efficiency and the amount of the ablated material and finally this effect on the thickness of thin films and the distribution of ablated materials on the substrates. Implying that the deposited films at the low value of laser energy have a much smaller size. The exciton peak shift with the particle size values is due to the quantum confinement attributed to inducing the energy band gap variation. The spectral characteristic of the semiconductor has been shown to vary with the effects of quantization. The values of energy band gap (ΔE) vary with the radius values of the particles (d).

$$\Delta E = \left(\frac{h}{2me^*}\right)\left(\frac{\pi^2}{d^2}\right) \quad (7)$$

where ΔE (energy shift or optical band gap shift with respects to bulk band gap value (3.35 ev) and d is the size of the particle, h the constant of Planck's and me^* is the electron reduced mass thus with the decrease in the value of the particle size.

Indeed the increase in the value of laser fluency means transfer more of the laser energy and indicate to ablating the larger amount of the raw materials. It was observed that the increase of the Fluency of the laser produce the plume of the plasma become denser and could become more intense and this gives an indication that the large particles will be produced due to the two fact, the first fact is due to the longer time of growth and the other fact as a result of the high probability of clustering. The alteration of the $(\alpha h\nu)^2$ value with the values of the photon energy (hν) is present in figure (2), the value of the optical band gap (E_g) of Ag_2O_3 NPs are calculated from the extrapolating of linear part of $(\alpha h\nu)^2$ as a function of photon energy (hν) plot on x-axis. The values of the optical band gap (Eg) are found to be varied from (3.47-3.45) eV with the effect of laser fluency. These values of the optical energy band gap have as lightly redshift as the laser fluency will be increased (increase in particle size), this result agrees with other work [29, 30].

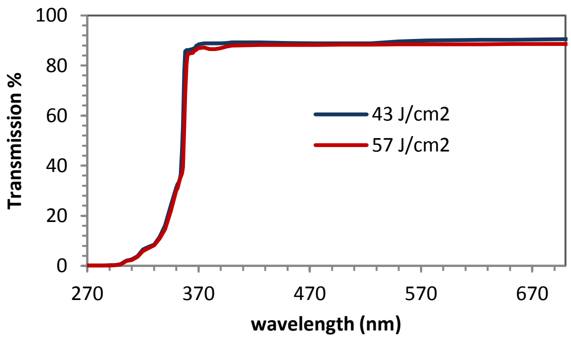

Fig 1 optical transmission spectra of Ag_2O_3 at two different laser fluency

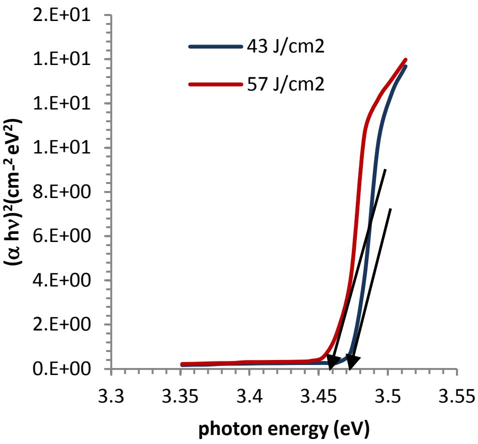

Figure (2), the optical band gap (Eg) of Ag2O3

The reflectance (R %) of Ag_2O_3 NPs was testing by using the double-beam UV-vis instrument. Fig. 3 was present the reflectance spectra of the Ag_2O_3 NPs films as a function of (λ) range of 300-700 nm. It's clearly observed that optical reflection increased with the increasing of the Laser fluency and this increasing as a result due to the fact that when the beam of the light effects on the thin layer of the film the light beam is slightly reflected and then transmitted, depending on the energy of the incident photon, the thickness of the films and the size of the deposit particles since the deposited material consists of the charged particles, the bounds and the loose electrons, and the ionic elements, etc.

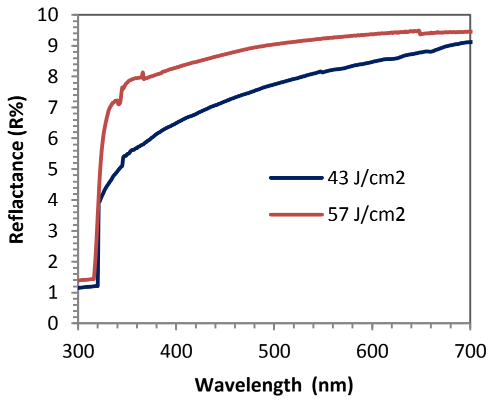

Figure (3), the reflectance of Ag_2O_3

The value of (n) was calculated using the reflactance spectrum values in the range of 300-700 nm. There is a tangible change in the refractive index (n) value in the visible range; according to estimates and found to be about 1.24 – 1.31 at 330 nm as present in Fig. 4 and is tabulated in Table 1. The increasing in the particle size values results in overall causes decreasing of the refractive index; this due to the result of decreasing the transmission value as the thickness increases. The value of the (n) changes a bit after 310 nm to 700 nm wavelength. It could be showed that the n decreases with the laser fluence, this behavior may be attributed to a decline of the film thickness as present in table 1, which resulting in low reflection, the elevated value of the refractive index is good for an optoelectronic device.

The extinction coefficient K is a very important physical parameter related to microscopic atomic interactions. The extinction coefficient is shown in Fig. 5 as a function of photon energy, has high value at higher Laser fluency.

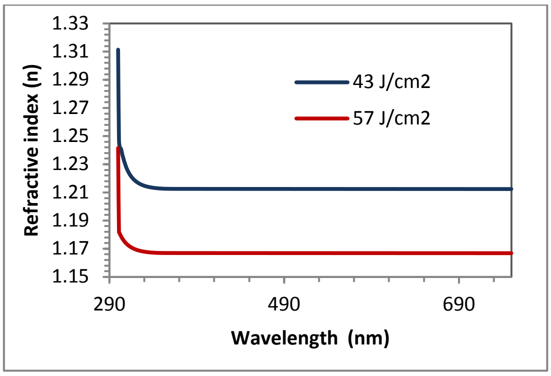

Figure (4), the refractive index of Ag_2O_3

It shows that the extinction coefficient is high in the region of the high energy photon, and it will decrease with decreasing of the photon energy. The stronger absorbing medium present high extinction coefficient [49]. The loss of the energy of the electromagnetic radiation out of the medium is examined using the extinction coefficient value of a special substance. The value of extinction coefficient is inversely associated with the value of the spectra of the transmittance [50], therefore the low value of T% gives a high value of extinction coefficient.

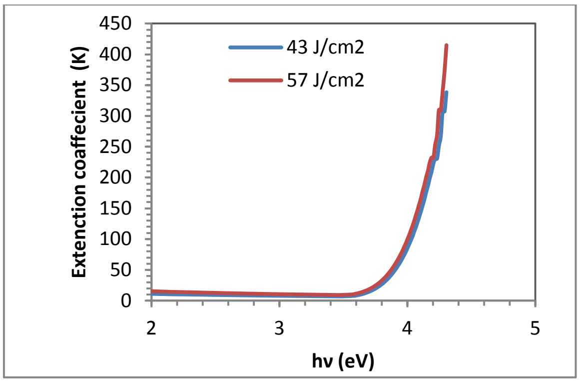

Figure (5), the extinction coefficient of Ag_2O_3

Table 1: The energy band gaps, refractive index and optical transmittance of Ag_2O_3nanostructures.

Laser Fluency (J/cm^2)	Film Thecknes (nm)	E_g measured (eV)	*n* measured	*T* %
43	340	3.47	1.31	90
57	325	3.45	1.24	87

The photoluminescence of the deposited Ag_2O_3nanofilms are grown on the glass substrate for 150 °C are presented in Fig. 6. One emission band at 355 nm (3.47 eV) and 362 nm (3.45 eV) for different laser fluency could be observed. The peak 355 nm is the more intense than the 362 nm. It could be shown that the photoluminescence spectra have a strong UV emission zone, relating to high crystallization with ideal energy gap compared with the experimental data. Moreover, the relative intensity of the peaks also depends on the corresponding irradiative recombination efficiency. The obtained read shift was related to the reduction in the deposited material grain size. Thus, the photoluminescence spectra of Ag_2O_3 prove that the material produced has sufficient quality for use in research of the optoelectronic devices.

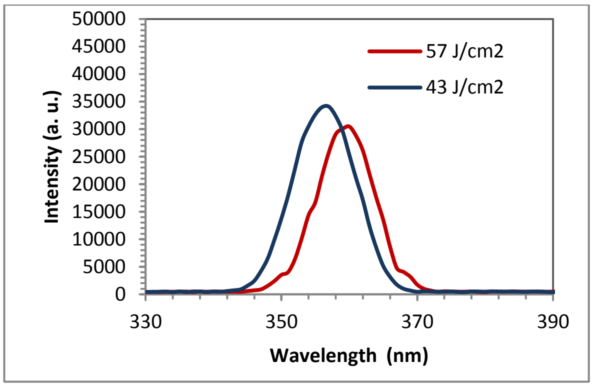

Figure (6), the photoluminescence of Ag_2O_3

3. CONCLUSION

Ag_2O_3 nanostructures have been prepared using PLD technique. It was found an approximate match between the value of the energy band gap that calculated by UV and PL. The maximum value of the transmission was found to be about 90%. The value of the refractive index showed the highest value of 1.24, while 1.31 that is appropriate for optoelectronic applications.

REFERENCE

1. A. H. Hammad, M. S. Abdel-Wahab, A. Alshahrie“*Structural and Morphological Properties of Sputtered Silver xideThin Films: Thr Effect of Thin Film Thickness*” ,Digest Journal of Nanomaterials and Biostructures 11(4): 1245-1252 (2016)
2.W. Wu , C.C. Tseng , C. Li, C.K. Chang, J.H. Hsieh*"Characterization of Cu_2O and Cu_2O-Ag_2O thin films synthesized byplasma oxidation"*.Vacuum 118 (2015) 147-151
3. L.A.A. Pettersson , P.G. Snyder." *Preparation and characterization of oxidized silver thin films*" Thin Solid Films 270 (1995) 69-72
4. T.C. Kaspar, T.C. Droubay, S.A. Chambers“*Atomic oxygen flux determined by mixed-phase Ag/Ag_2O deposition*”, Thin Solid Films 519 (2010) 635–640
5. C.C. Tseng, J.H. Hsieh, W. Wu" *Microstructural analysis and optoelectrical properties of Cu_2O, Cu_2O–Ag,and Cu_2O/Ag_2O multilayered nanocomposite thin films*". Thin Solid Films 519 (2011) 5169–5173
6. Esben Lund, AugustinasGaleckas, Alexander Azarov, Edouard V. Monakhov, Bengt G. Svensson**“***Photoluminescence of reactively sputtered Ag_2O films***”**, Thin Solid Films 536 (2013) 156–159
7. Yang You, Long Wan, Shiying Zhang, DifaXu."*Effect of different doping methods on microstructure and photo-catalyticactivity of Ag_2O–TiO2 nanofibers*,**"** Materials Research Bulletin 45 (2010) 1850–1854
8. K.Lalitha, J. K.Reddy,M. V. P. Sharma, V. D.Kumari,M.Subrahmanyam“*Continuous hydrogen production activity over finelydispersed Ag_2O/TiO_2*

*catalysts from methanol:watermixturesunder solar irradiation: A structure–activity correlation"*International Journal of hydrogen energy 35:3991-4001 (2010).

9. Jonathan Derouin, Rachael G. Farber, Stacy L. Heslop, Daniel R. Killelea*"Formation of surface oxides and Ag_2O thin films with atomic oxygen on Ag(111)"*Surface Science 641 (2015) L1–L4

10. X.Gao, S.Wang, J. Li, Y.Zheng, R. Zhang, P. Zhou, Y.Yang, L.Chen"*Study of structure and optical properties of silver oxide films byellipsometry, XRD and XPS methods*" Thin Solid Films 455 –456 (2004) 438–442

11. F.X. Bocka, T.M. Christensenb, S.B. Riversc, L.D. Doucettea, R.J. Lada*"Growth and structure of silver and silver oxide thin films on sapphire,"*Thin Solid Films 468 (2004) 57– 64.

12. S.B. Rivers a, G. Bernhardt b, M.W. Wright c, D.J. Frankel b, M.M. Steeves c, R.J. Lad" *Structure, conductivity, and optical absorption of $Ag_{2-x}O$ films*" Thin Solid Films 515 (2007) 8684–8688

13. G.Xiaoyong, Z.Mengke, Z.Zengyuan, C. Chao, M.Jiaomin, L.Jingxiao" *Effects of hydrogen annealing on the microstructure and optical propertiesof single-phased Ag_2O film deposited using direct-current reactivemagnetron sputtering*"Thin Solid Films 519 (2011) 6620–6623

14. M. A. Fakhri,E. T. Salim, U. Hashim, A. W. Abdulwahhab, Z. T. Salim" *Annealing temperature effect on structural and morphologicalproperties of nano photonic $LiNbO_3$*" J Mater Sci: Mater Electron28: 16728–16735 (2017).

15. M. A. Fakhri, U. Hashim, E. T. Salim, Z. T. Salim"*Preparation and charactrization of photonic $LiNbO_3$ generated from mixing of new raw materials using spry pyrolysis method*"Journal of Materials Science: Materials in Electronics 27 (12) (2016) 13105–13112.

16. E.T. Salim, M.A. Fakhri, H. Hasan"*Metal oxide nanoparticles suspension for optoelectronic device fabrication*"Int. J. Nanoelectronics and Materials 6(2013), 121-128.

17. E.T.Salim"*Optoelectronic properties of Fe_2O_3 /Si heterojunction prepared by rapid thermal oxidation method*"Indian Journal of Physics 87, (2013), 349-353.

18. M. A. Muhsien, E. T. Salem, I. R. Agool, H. H. Hamdan"*Gas sensing of Au/n-SnO_2/p-PSi/c-Si heterojunction devices prepared by rapid thermal oxidation*"Applied Nanoscience 4 (6) (2014) 719–732.

19. E. T Salim, M. S Al Wazny, M.A Fakhry"*Glancing Anglr Reactive Pulsed Laser Deposition (GRPLD) for Bi_2O_3/Si Heterostructure*" Modern Physics Letters B, 27(2013), 1350122-7- 1350122-1.

20. A. Kadhim, E. T. Salim, S. M. Fayadh,A. A. Al-Amiery,A. Amir H. Kadhum, and A. B. Mohamad"*Effect of Multipath Laser Shock Processing on Microhardness, Surface Roughness, and Wear Resistance of 2024-T3 Al Alloy*" The Scientific World Journal 2014 (2014), Article ID 490951, 6 pages http://dx.doi.org/10.1155/2014/490951

21. E. T. Salem, I. R. Agool, M. A. Hassan"*Construction of SnO_2/SiO_2/Si Hetrojunction and its Lineup using I–V AND C–V Measurements*" Int. J. Mod. Phys. B 25, 3863 (2011). DOI: http://dx.doi.org/10.1142/S0217979211102022.

22. I. R. Agool, E. T. Salem, M. A. Hassan" *Optical and electrical properties of SnO_2 Thin film prepared using RTO Method*" Int. J. Mod.Phys. B 25, 1081 (2011). DOI: http://dx.doi.org/10.1142/S0217979211058614

23. E. T. Salim"*Surface morphology and x-ray diffraction analysis for silicon nanocrystal based heterostructures*"Surf. Rev. Lett., 20 1350046 (2013) [6 pages]

24. E. T. Salim, R. A. Ismail, M. A. Fakhry, Y. Yusof"*Reactive PLD of ZnO thin film for optoelectronic application*"International Journal of Nanoelectronics and Materials, 9 (2016) 111-122.

25. M. A. Fakhri"*Annealing Effects on Opto-electronic Properties of Ag_2O Films growth using Thermal Evaporation Techniques*" Int. J. Nanoelectronics and Materials 9 (2016) 93-102.

26. A.H. Al-Hamdani, M. Qasim, K.S. Rida, A. Kadhim "*Enhancement of Solar Cell Performance Based On Porous Silicon*" Journal of Nanoscience and Technology, 2(2),(2016):73-75.

27. E. Yousif, A. A. Al-Amiery, A. Kadihum, A. A. H. Kadhum and A. B. Mohamad" *Photostabilizing Efficiency of PVC in the Presence of Schiff Bases as Photostabilizers*"Molecules2015, 20(11), 19886-19899

28. Z. T Salim, U Hashim, M.K MdArshad, M. A Fakhri, E. T Salim"*Zinc oxide flakes-corolla lobes like nano combined structure for SAW applications*"Materials Research Bulletin, 86 (2017),215-219.

29. M. A. Fakhri, Y. Al-Douri, U. Hashim, E. T. Salim"*XRD Analysis and Morphological Studies of Spin Coated $LiNbO_3$ Nano Photonic Crystal Prepared for Optical Waveguide Application*" Advanced Materials Research 1133 (2016) 457-461.

30. Z. T. Salim, U. Hashim, M.K.Md. Arshad, M. A. Fakhri, E. T. Salim"*Frequency-based detection of female Aedes mosquito using surface acoustic wave technology: Early prevention of dengue fever*"Microelectronic Engineering, 179(5) (2017), 83-90.