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Enhanced Ultraviolet Photodetector Based on Mg-Doped ZnO Nanorods Films

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Abstract

Magnesium-doped zinc oxide (ZnO: Mg) nanorods and nanotubes films were prepared by hydrothermal method deposited on glass substrates. X-ray diffraction (XRD), scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDX), photoluminescence (PL), and optical absorption spectroscopy (UV) were performed to characterize the prepared films. X-ray diffraction analysis showed a decrease in the lattice parameters of Mg doped ZnO NRs. The Photoluminescence of the undoped and Mg-doped ZnO NRs displayed a near band edge. At 10 V bias, the metal-semiconductor-metal (MSM) ultraviolet (UV) photodetector performance of the Mg-doped ZnO prepared for various Mg concentrations of 0.0, 0.02, and 0.06 was investigated under radiation of 40μW/cm² at the wavelengths of 365 and 385 nm UV light. The responsivity, detectivity and quantum efficiency of Mg-doped based on MSM detector were 0.118A/W, 1.0579*10¹² and 40.05157 under UV of wavelength 365nm respectively.

Keywords: pure ZnO nanorods Mg doping ZnO nanorods, MSM Photodetector, hydrothermal method, nanotubes.

الخلاصة

تم تحضير أغشية أكسيد الزنك النقي والمشوب بالمغنيسيوم بتركيزات مختلفة على قواعد زجاجية بالطريقة الحرارية المائية. تم إجراء فحوصات حيود الأشعة السينية (XRD)، المسح المجهر الإلكتروني (SEM)، التحليل الطيفي للأشعة السينية (EDX)، الفلومينيسcence (PL)، والامتصاص البصري (UV) لوصف واختبار الأغشية المحضرة. أظهرت تحليلات حيود الأشعة السينية (XRD) بان جميع النماذج متعددة البلورات ذات تركيب سداسي بالاتجاه (002). تم دراسة تأثير تشويب المغنيسيوم على الخصائص التركيبية والكهربائية والبصرية لأغشية أكسيد الزنك ZnO، ولوحظ التحسن الواضح على التبلور وكذلك زيادة في فجوة الطاقة البصرية. عند تحيز 10 V، تم فحص أداء كاشف (MSM) للزنك أكسيدات النقي والمشوب بالمغنيسيوم بتركيزات مختلفة من 0.0 و 0.02 و 0.06 تحت أشعة فوق البنفسجية ذات شدة 40 μW / سم² عند أطوال موجية 365 و 385 نانومتر. كانت الاستجابة والكفاءة والكفاءة الكمية لكاشف الزنك المشوب بالمغنيسيوم بتركيز 0.06 A / W، 1.0579 * 10¹²، و 40.05157 عند الطول الموجي 365 nm للأشعة فوق البنفسجية على التوالي.

Introduction

Zinc oxide is a semiconductor material composed of zinc ions (Zn²⁺) and oxygen ions (O₂), represented by ZnO. It has special electrical, optical and electrical properties. It also has a large energy gap and a relatively large exciton binding energy of about 3.37eV and 60 MeV respectively and with room temperature [1, 2]. Different doping in ZnO (i.e., Mg) can adjust bandgap to make UV photodetectors in different

UV region. There were several reports focused on efforts to control the properties of ZnO by adding the transition metal impurities such as Al [3, 4], Cu [5], Co [6], In [7], and Mg [3, 8] Doping work to control and adjust the band gap, which greatly contribute to increase the efficiency and power, and in many applications, such as Light-emitting diodes [9], laser diodes



[10], solar cells, transparent electrodes, thin film transistors and optical sensors [3, 11].

The ZnO sensing mechanism is highly correlated with surface reactions hence; oxygen absorption, grain size, defects, and other parameters have an important effect in controlling the sensing response.

In the present study, systematic Mg doping (0, 0.02, 0.06) in ZnO NPs was achieved and is analyzed by XRD, SEM, and UV-Vis spectroscopy and it has discussed the fabrication of Metal Semiconductor Metal (MSM) UV photodetector that importance of this study is the amplification of (MSM) UV detectors performance using ZnO nanorods arrays, resulting in a low-cost, time-saving fabrication process. It has also discussed the performance of (MSM) UV detectors fabricated using Mg-doped ZnO nanorods and nanotubes that produce the highest responsivity, quantum efficiency and others parameters compared with pure ZnO.

Materials and Methods

Experimental

The hydrothermal method was used to synthesize pure and Mg-doped ZnO nanorods and nanotubes arrays as reported by several researchers [3, 12]. First, ultrasonic cleaner technology was used to clean the glass substrates in stages, starting with ethanol, acetone, and propanol for 10 minutes per solvent, then cleaning with deionized water and drying it.

Zinc acetate dehydrates 0.5 M was first dissolved in the ethanol with stirring at 60°C for (2-3) h until a clear and transparent homogeneous solution was formed. A homogeneous seed layer was obtained using a spin coating technique by taken by dropper of the prepared solution and pours over the center of a substrate, the rate of speed was 2500 for 30 seconds, this process was repeated several times on hotpot temperatures about (140-150) °C for 10 min per cycle to remove the solvent, then the samples were finally annealed at (350-400) °C in the furnace for 2 h to getting the ZnO seed layer.

After the formation of a seed layer, ZnO nanorods were formed through the hydrothermal method. The substrates with a seed layer were putting verticality inside Teflon vessels filled with the aqueous solution of 0.05M zinc nitrate hexahydrate $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 0.05 M Hexamethylenetetramine $\text{C}_6\text{H}_{12}\text{N}_4$, sealed and heated at (110-120) °C for 3 h. Then, the substrate removed from the Teflon vessel, cleaned with distilled H_2O and dried. Next, the Mg-doped ZnO nanorods and nanotubes were grown on the seed-layer-coated glass substrate using aqueous solutions of 0.05M zinc nitrate hexahydrate, 0.05M Hexamethylenetetramine and concentrations (the atoms' ratio of Mg to Zn) Mg/Zn (0.02, 0.06) magnesium nitrate $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$. These reagents were dissolved and reacted in Teflon vessel with a temperature of (110-120) °C for 3h.

Following, the substrate was rinsed with distilled water and all the samples were annealed for 2h in furans at 350°C. Then aluminum Al contacts were deposited on the nanorods or nanotubes films by using a thermal evaporator technique to produce a metal–semiconductor–metal (MSM)-type UV photoconductive detector. The deposition of metal contacts was performed at a low pressure of 9.5×10^{-5} Pa. The separation between the metal contacts was fixed at 0.05 mm for all samples using a metal mask as showed in Table1.

Table 1: Represent as name and ratio Mg-doped ZnO for samples used in this paper

samples	element	ratio atoms Mg/ atoms Zn doped ZnO
A	pure ZnO	0
M1	Mg	0.02
M2	Mg	0.06

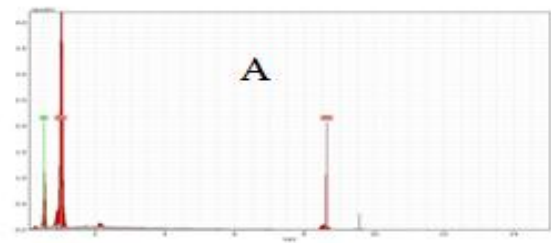
Results and discussions

Structural studies

Pure and Mg-doped ZnO nanorods and nanotubes have a polycrystalline hexagonal wurtzite structures as shown in Figure1, it is noted that the intensity of diffraction peaks of the ZnO deposition appears at positions 31.2954,

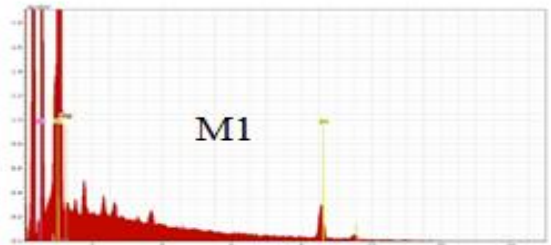
Spectrum: Acquisition

Element	Series	unn.	C norm.	C Atom.	C Error (1 Sigma)
		[wt.%]	[wt.%]	[at.%]	[wt.%]
Zinc	L-series	72.94	75.20	42.60	4.69
Oxygen	K-series	24.05	24.80	57.40	4.06
Total:		97.00	100.00	100.00	



Spectrum: Acquisition

Element	Series	unn.	C norm.	C Atom.	C Error (1 Sigma)
		[wt.%]	[wt.%]	[at.%]	[wt.%]
Oxygen	K-series	20.98	26.41	59.08	3.05
Zinc	L-series	57.90	72.90	39.90	3.69
Magnesium	K-series	0.55	0.69	1.02	0.07
Total:		79.43	100.00	100.00	



Spectrum: Acquisition

Element	Series	unn.	C norm.	C Atom.	C Error (1 Sigma)
		[wt.%]	[wt.%]	[at.%]	[wt.%]
Oxygen	K-series	22.53	28.50	61.32	8.03
Zinc	L-series	55.61	70.35	37.04	4.46
Magnesium	K-series	0.92	1.16	1.64	0.29
Total:		79.06	100.00	100.00	

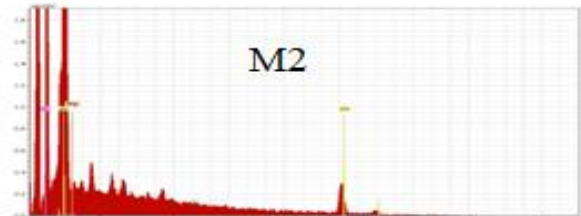


Figure 3: EDX spectrum of ZnO and Mg-doped ZnO (A, M1 and M2)

Optical properties

The optical absorption spectrum of undoped and Mg-doped ZnO nanorods and nanotubes as shown in Figure 4a at room temperature; it showed that all samples have sharp absorption edges in the UV region. The absorption band edge was observed at 390 nm, 382.5 nm, and 379 nm for samples A, M1, and M2, respectively, because of the band-to-band transition and Mg-doped ZnO shows that the absorption edge shifted to a short wavelength with an increase in the doping ratio due to quantum-size effects. The optical bandgap of ZnO was calculated using the fundamental relation [13-15]

$$(Ah\nu) = B(h\nu - E_g)^n \quad (4)$$

Where A , ν , h , and E_g represent the absorbance, frequency, Planck's constant and optical energy gap respectively, $n = 1/2$ for direct allowed transition and B is the proportionality

constant. The optical energy gap of the A, M1, and M2 was observed equal to 3.23, 3.275, and 3.296 eV, respectively [3] as shown in Figure 4b, agreed with the theoretical band gap value of 3.37 eV. However, it was noted that with Mg doping an increasing in the energy gap of pure ZnO with a blue shift that can be attributed to that the new ion (Mg^{+2}) energy level formed in the energy gap, thereby increasing the ZnO band gap.[3, 16]

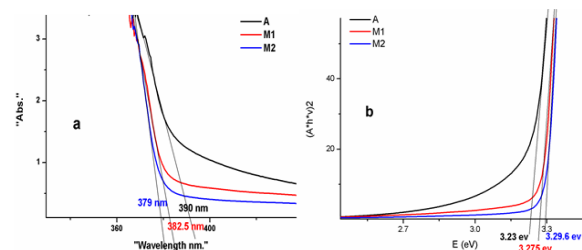


Figure 4: Absorption spectra and b) the optical energy gap of pure ZnO and Mg-doped ZnO nanorods deposited on the glass substrate

Photoluminescence spectra

The photoluminescence (PL) spectra for pure and Mg-doped ZnO nanorods samples with excitation at a wavelength of 325 nm show that Figure 5, That is showing the photoluminescence (PL) spectra of samples A, M1 and M2 at peaks 389.8, 382.5 and 378.3 nm at sharp UV emission due to the recombination of free-exciton across which was called the near band edge emission (NBE). A minor broad visible emission, which is called deep level emission (DLE) that range of wavelength 450–550 nm [3, 17], due to the transition between defect levels as well as extrinsic impurities [18]. The near band edge emission (NBE) of samples A, M1 and M2 were detected at UV peaks 387.8, 381.66, and 377.45 respectively with excitation a wavelength of 325 nm. The increase in the Mg composition of the samples leading to widening of the bandgap and blue shifted the NBE emission. [19]. Thus, Mg doping should be responsible for such a blue shifted. PL spectra showed an increase in DLE upon doping which was due to the increase in defect states [20], the effect of oxygen may be one of the defects in the composition of materials according to researches [3, 21] and shows its obvious effect in Figure 5, the defect green emission occurred around 543nm, 541nm and 538nm for samples A, M1 and M2 respectively, which was due to the presence of O_i according to the previous reports by [22].

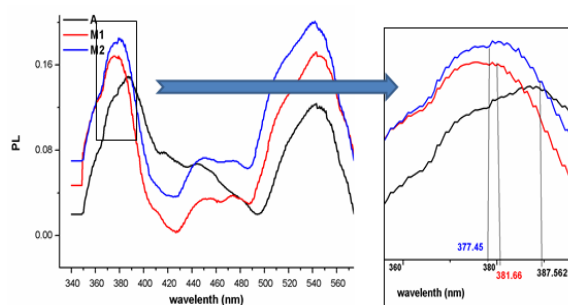


Figure 5: PL spectra of undoped ZnO and Mg doped ZnO nanotubes and nanorods with concentration M1 and M2 deposited on the glass substrate

Device Fabrication Process

The UV photodetector device with a dimension of 1 cm x 1 cm for the structure of Al/ZnO/Al was fabricated by the MSM. The Al electrode was deposited onto samples A, M1, M2 and the annealed films using a mask through thermal evaporation technique.

Characteristic of the UV detector

The current-voltage (I-V) curves of the fabricated MSM under dark and UV (365, 385) nm with power density of $40 \mu\text{W}/\text{cm}^2$ were measured for all samples. The results can be shown in Figure 6; it showed a linear response of the photocurrent density curve; that can be attributed to an ohmic contact between the ZnO film and Al electrodes. The highest sensitivity or higher ratio between the electrical resistance under the action of light and the dark state can be obtained when the conductivity between the metal and the ZnO is a ohmic connection [3, 23]. The responsivity (R), quantum efficiency (η), detectivity (D) and photosensitivity (S) can be calculated by the Equations (5 to 8):

$$R = \frac{I_{ph} - I_{dark}}{P_{opt}} \quad (5)$$

$$\eta = R \frac{hc}{e\lambda} \quad (6)$$

$$D = \frac{\lambda \eta e}{hc} \left(\frac{RA}{4kT} \right)^{1/2} \quad (7)$$

$$S = \frac{I_{ph} - I_{dark}}{I_{dark}} * 100\% \quad (8)$$

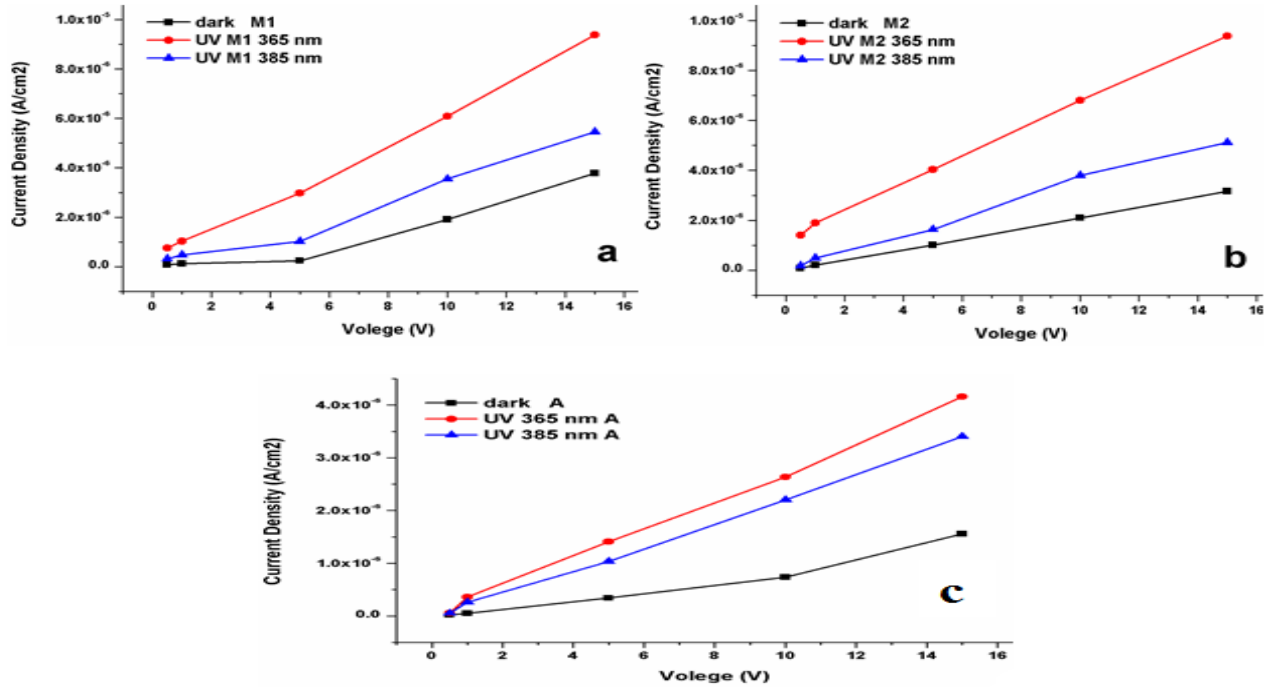


Figure 6: I-V characteristics for MSM ZnO and Mg-doped ZnO in dark and under UV 365 and 385 nm for samples a) M1 (0.02) b) M2 (0.06) c) A (0%)

Where I_{ph} is the photocurrent, I_{dark} is the dark current and P_{opt} is the optical power of the UV source, c , e and λ are represented blank constant, light speed, electron charge and wavelength used, respectively. According to the relations (5-8), the results can be found in Table3, at a wavelength of 365, 385 nm is as a function applied voltage.

Table 3: Parameters of MSM ZnO and Mg doped ZnO photodetector as a voltage of at UV wavelength 365 nm and 385 nm

Par.	V	365 nm		
		A	M1	M2
η	1	2.6242	7.74393	14.3516
	5	9.10533	23.30504	25.78499
	10	16.19009	35.54741	40.05157
S%	1	617.33333	746.6124	806.73039
	5	314.69148	1132.73421	300.95923
	10	259.56383	218.62517	224.44021
D	1	1.42027E11	2.68313E11	3.79689E11
	5	4.22366E11	1.282E12	6.95082E11
	10	7.2337 E11	9.83711E11	1.05797E12
Gain	1	7.17333	8.46612	9.0673
	5	4.14691	12.32734	4.00959
	10	3.59564	3.18625	3.2444

385 nm

η	1	1.70336	2.84456	2.27002
	5	5.60443	6.34055	5.06918
	10	11.84292	13.1947	13.70282
S%	1	422.66667	289.27921	134.59379
	5	204.3095	325.067	62.40883
	10	200.2726	85.5972	80.99512
D	1	9.72411E10	1.03959E11	6.33468E10
	5	2.74216E11	3.67902E11	1.44137E11
	10	5.58133E11	3.85148E11	3.81796E11
Gain	1	5.22667	3.89279	2.34594
	5	3.0431	4.25067	1.62409
	10	3.00273	1.85597	1.80995

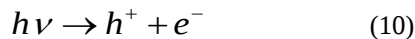
The photoswitching characteristic of the UV detector

Figure7 shows the photo response of UV photodetector at a wavelength of 365 and 385 nm. The ON/OFF switching UV time duration characteristics was 2/5 min for the samples A, M1, and M2, respectively, with the intensity of $40 \mu W/cm^2$ at 10V applied bias. According to [2, 24], the photodetector sensitivity was closely related to oxygen adsorption and desorption processes. The oxygen molecules tend to absorb the free electrons at the nanorod surfaces to produce oxygenated ions adsorbents, in the case

of darkness, i.e., without ultraviolet radiation, as shown by following equation



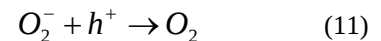
Where is O_2 , e^- and O_2^- represent are oxygen molecule, a free electron and O_2^- adsorbed oxygen ion respectively. Before UV illumination applied, a low current is generated because a barrier is created at near the surface by the adsorbed oxygen ions. When UV light is irradiated onto the nanorods or nanotubes, an electron-hole pairs are generated at the surface according to the equation: [25]



where $h\nu$, h^+ and e^- are represented the photon energy of UV light, generated hole in the valence band and generated electron in the conduction band respectively.

The O_2 molecules which desorbs from the surface are produced by adsorbed oxygen ions combine with the holes under applied a bias voltage, the unpaired electrons are collected at the anode; thereby an increase in conductivity

takes place with the decrease in the width of the depletion layer, this process is given by the equation:



At the same time, desorption of O_2^- ions on the nanorods surface leaves photo generated electrons in the conduction band, increasing ZnO conductivity and contributing to the photocurrent [25, 26]. After the UV exposure, the O_2 molecules get reabsorbed on the surface until equilibrium is restored to repeat the process again onto the nanorod surface, making decrease the conductivity of the sensor. When the nanorods or and nanotubes array, the surface area increases, enabling UV light to be absorbed more effectively and to generate more electron-hole pairs.

It is to be noted that voltage was chosen for getting the highest and best in the different responsivity samples of Mg-doped and undoped at a wavelength of 365 and 385 nm as illustrated in Table3.

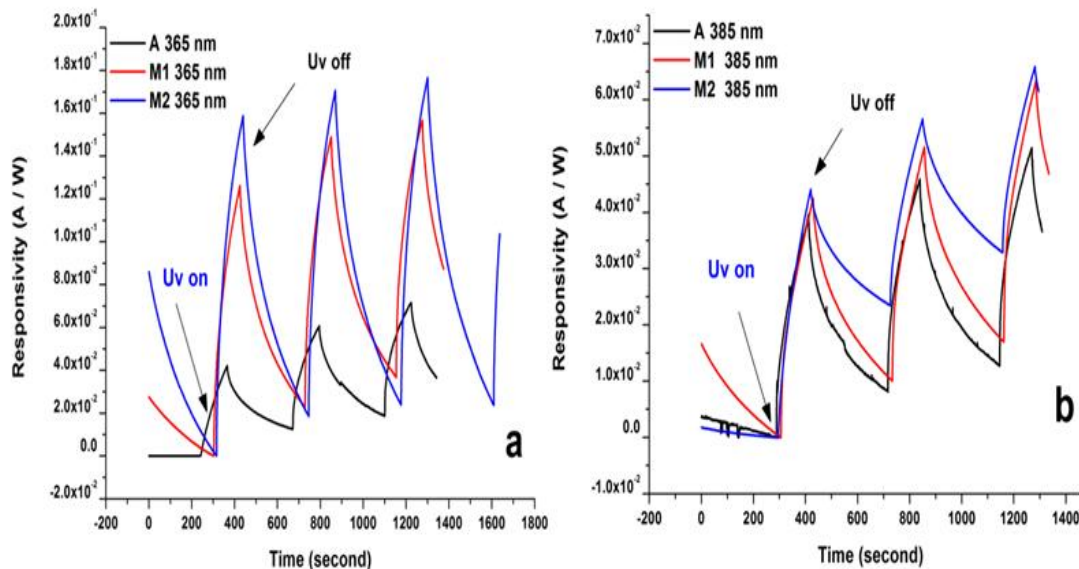


Figure 7: Responsivity with a time of MSM Mg-doped ZnO photodetectors are compared with MSM ZnO photodetector at a) UV 365nm b) UV 385nm

Responsivity and quantum efficiency via wavelength

The responsivity is defined as the generated photocurrent per unit power of incident light on the effective area of the photodetector and external quantum efficiency (η) is nothing but the number of electron-hole pairs generated due to the one absorbed a photon of energy $h\nu$. The responsivity and quantum efficiency of a device have been at 1V applied bias which calculated

by equations 5 and 6 as a function of wavelength [27]

It is observed that the responsivity of devices Mg-doped ZnO samples M1 and M2 shows a higher and shifted at low wavelength, these were a high responsivity in the UV region and prominently does not show a photo response to visible region, which is credited to the wide band gap energy of ZnO so that no charge carrier is excited at low energy visible photons.

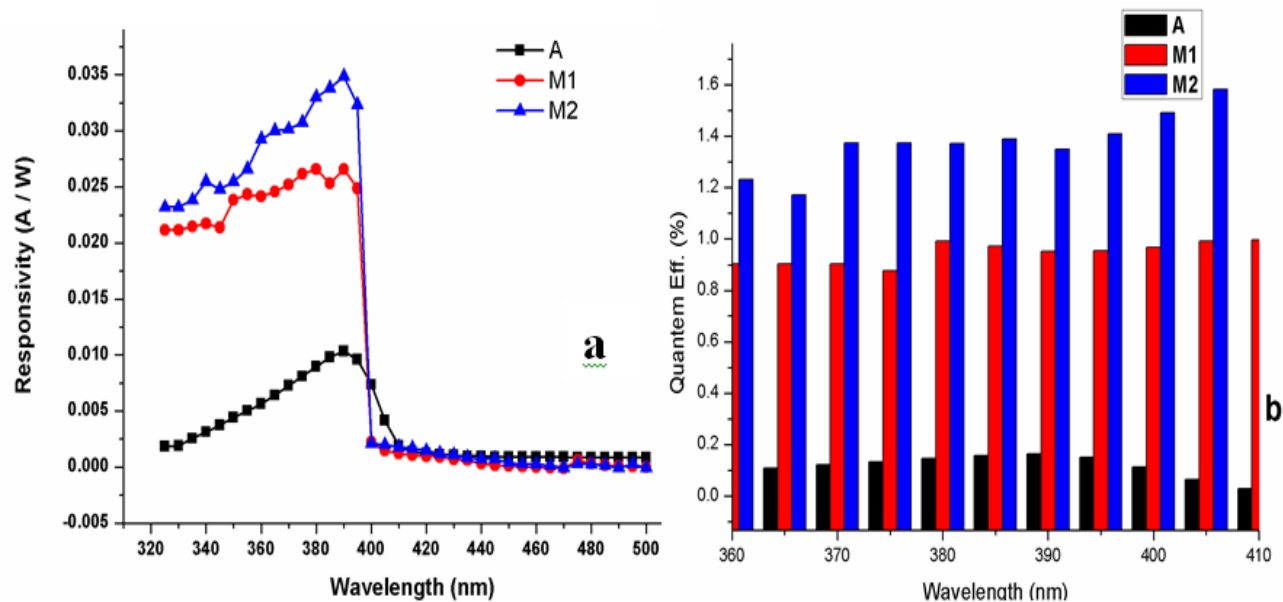


Figure 8: Responsivity and quantum efficiency as wavelength of MSM ZnO and MSM Mg-doped ZnO photodetectors

Conclusions

In this work, the structural, optical and electrical properties were measured for pure and Mg-doped nanorods and nanotubes of ZnO were successfully synthesized by hydrothermal. The MSM UV has been fabricated based on a pure and Mg-doped ZnO thin film. The photoresponse properties under the action of UV were studied at room temperature, the performance of the photodetector can be improved with respect to the photocurrent, the contrast ratio, dark current, and detectivity. The MSM UV based on an Mg-doped ZnO has shown enhanced responsivity and quantum efficiency and others parameters under a voltage 10 V applied shown that Table 4 that has compared to that pure ZnO at shifted low wavelength.

Table 4: Result parameters of devices are measured at 10 v applied biases of all samples

sample	UV 365 nm			
	R A/W	η	S %	D
A	0.047	16.190	259.56383	7.23×10^{11}
M1	0.1045	35.547	218.62517	9.81×10^{11}
M2	0.118	40.051	224.44021	1.05×10^{12}
sample	UV 385 nm			
	R A/W	η	S %	D
A	0.036	11.8429	200.2726	5.58×10^{11}
M1	0.0406	13.1947	85.5972	3.85×10^{11}
M2	0.0426	13.7028	80.99512	3.8×10^{11}

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