Study of point defects in ZnO thin films irradiated with alpha particles

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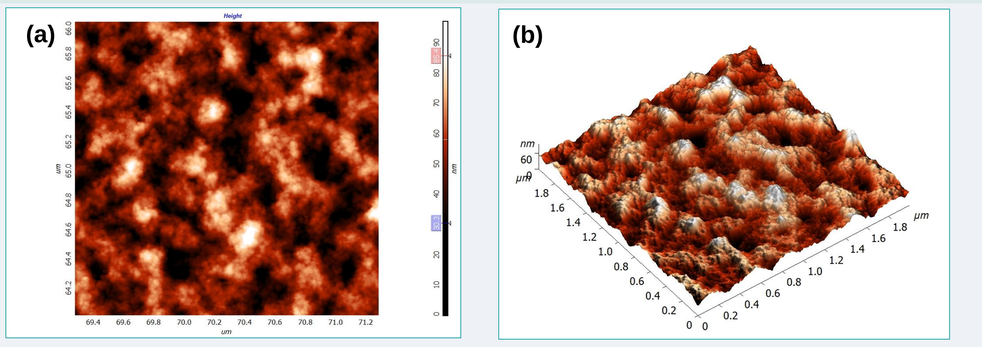
Outline

- At laboratory setup scale, the irradiation with high energy alpha particles has been used to study the effects on ZnO thin films for devices operating in space environment. The targeted materials are relevant for advanced nanoelectronics, optoelectronics, transparent electronics, UV high performance photodetectors, solar cells, sensors, that will potentially operate in harsh radiation conditions.
- The effect of alpha particles irradiation on the morphology, structure and luminescence emission of ZnO thin films has been investigated by x-ray diffraction (XRD), atomic force microscopy (AFM) and photoluminescence (PL), aiming to evaluate the types of defects generated by radiation as a function of the exposure time.

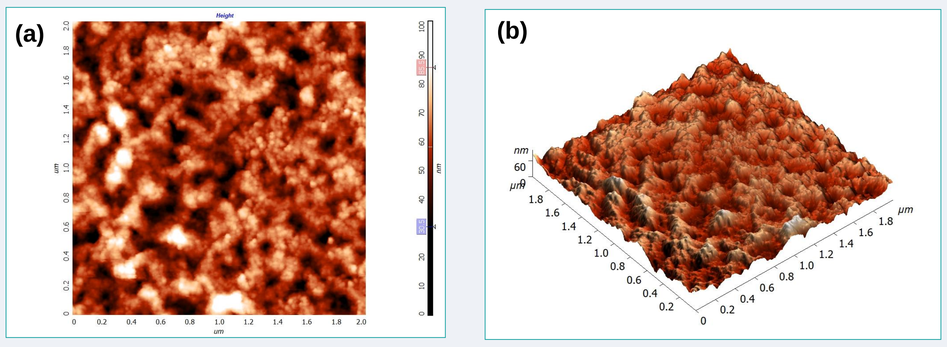
Experimental

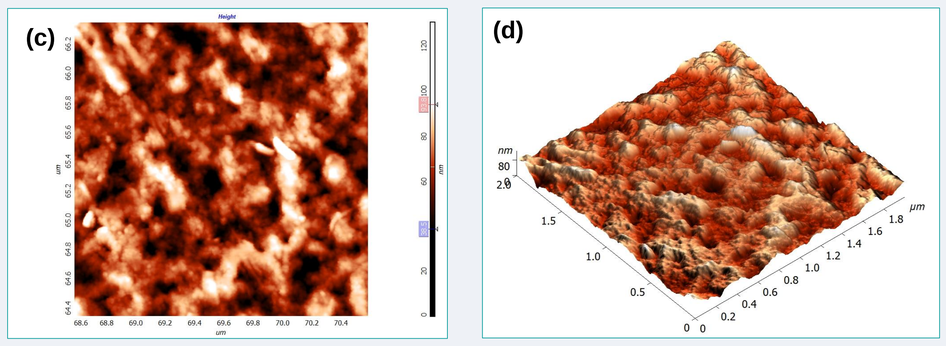
- ZnO films with wurtzite type structure, thickness of about 100 nm, and average crystallite size of 21 nm have been synthesized by sol-gel method. The optical transmission of the films in the visible region is 88% and the band gap energy, calculated from the absorption spectra is 3.24 eV [1].
- The films were irradiated with alpha particles at a doze of 5.3 kGy/h, the energy of 3 MeV, and the irradiation time was varied from 100 s (~11 μC) to 500 s (~54 μC), 1000 s (~104 μC) and 8 h (~2100 μC).
- The Irradiations were performed at the U120 Cyclotron facility at the National Institute for R&D in Physics and Nuclear Engineering-“Horia Hulubei” (IFIN-HH), Bucharest, Romania.

Results

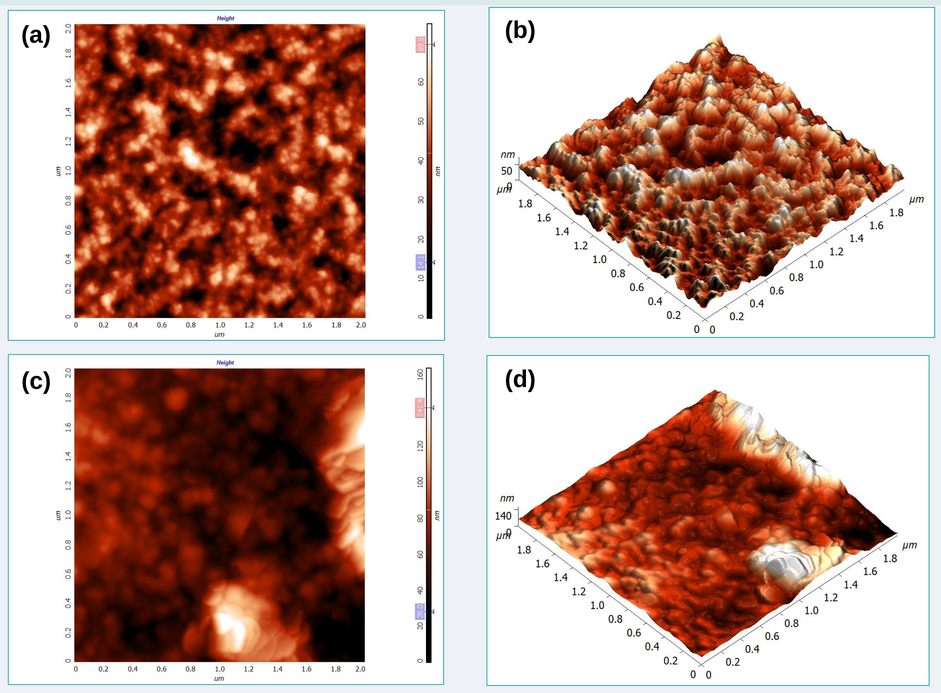


Surface morphology of ZnO films, showing an uniform distribution of the grains size and roughness. Atomic force microscopy images (a) 2D and (b) 3D.

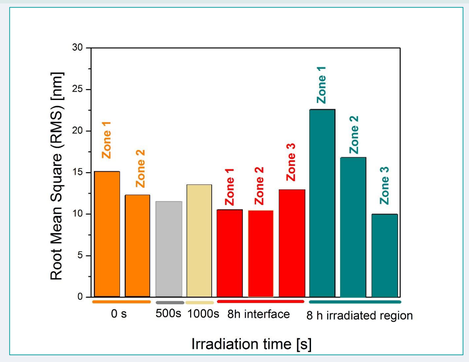




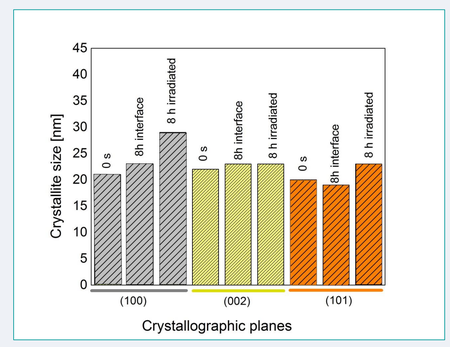
Changes of the ZnO films surface morphology after irradiation with 3 MeV alpha particles for (a), (b) 500 s, (c), (d) 1000 s. Atomic force microscopy 2D and corresponding 3D images showing surface modification: roughness decreases after short time irradiation (500 s) and increases after long irradiation (1000 s). This could indicate a slight recrystallization of the Zn- terminated surface caused by the accumulated heat generated by irradiation.



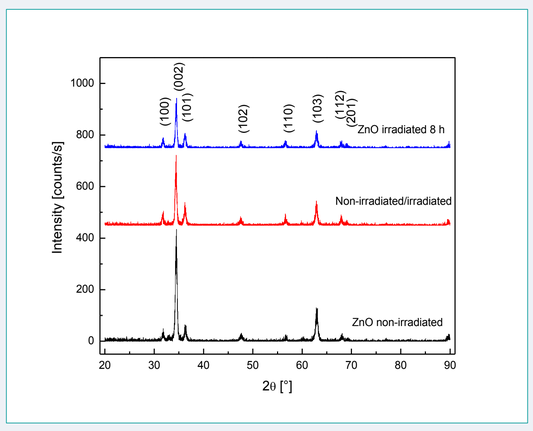
Atomic force microscopy 2D and corresponding 3D images of ZnO films surface irradiated 8 h: (a), (b), irradiated/non-irradiated interface region, (c), (d) 8 h irradiated region. The surface roughness decreases but still remains uniform at the interface between non-irradiated and irradiated regions, while accumulation of grain leads to increased and non uniform roughness in the irradiated region.



Variation of surface roughness after 8 h irradiation with 3 MeV alpha particles.



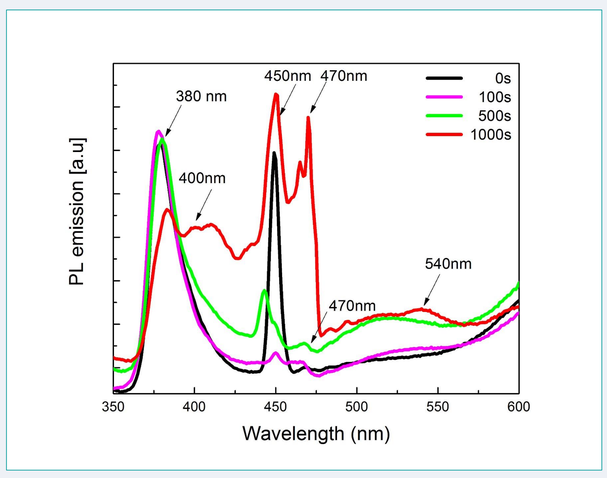
Variation of crystallite size after 8 h irradiation with 3 MeV alpha particles.



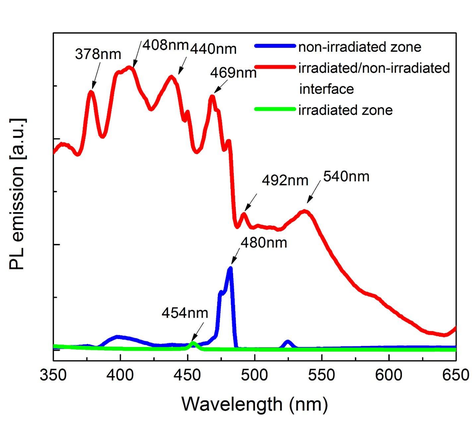
X-ray diffraction patterns of the ZnO films. The peaks correspond to hexagonal ZnO structure, with (002) texture. The (002) peak position shifted slightly from 34.45 to 34.37 and 34.43 in the non-irradiated, transition and irradiated regions, suggesting the presence of in-plane residual stress.

Films	Pos.[°2Th.]	Height [cps]	FWHM [°]	Crystallite size [nm]	σ (GPa)
Non-irradiated ZnO	(100) 31.78	289	0.41	21	0.19
	(002) 34.45	4992	0.40	22	
	(101) 36.21	582	0.44	20	
Non-irradiated/ irradiated interface zone	(100) 31.64	441	0.38	23	0.08
	(002) 34.37	2888	0.38	23	
	(101) 36.23	772	0.47	19	
Irradiated ZnO	(100) 31.80	399	0.30	29	0.11
	(002) 34.43	2024	0.38	23	
	(101) 36.19	638	0.37	23	

Table. *The XRD parameters of non-irradiated ZnO films, non-irradiated/ irradiated interface region and 8 h irradiated ZnO films: 2θ (crystallographic planes), peaks height, FWHM and the resulted crystallite size and residual stress, σ.*



Photoluminescence emission of ZnO films irradiated with 3 MeV alpha particles for: 100 s, 500 s, and 1000 s.



Photoluminescence emission of ZnO films irradiated for 8 h.

- *PL spectra of the non-irradiated films exhibit emission peaks situated at 3.26 eV, 2.75 eV and a broad peak, with low intensity, in the region 2.60-2.25 eV. While the PL emission in the UV region is determined by free excitons band to band transitions, the emission peak in the green-orange region is related to radiative transitions involving native point defects in ZnO [2-4].*
- *PL emission after irradiation for 100 s: increased intensity and shift of the peak at 3.28 eV, decreased intensity of the peak at 2.75 nm. Low intensity and broad emission in the region 2.59 – 2.23 eV.*
- *PL emission after irradiation 500 s: 3.26 eV peak preserves the intensity, while the intensities of the peaks at 2.80 eV, 2.65 eV, 2.40 eV increase.*
- *PL emission after irradiation 1000 s: new peaks appear at 3.10 eV, 2.64 eV which suggest the formation of Zn interstitials (ZnI) defects and increase the intensity of the peak at 2.30 eV associated with oxygen vacancies (OVs).*
- *PL emission after 8 h irradiation: several peaks (3.23 eV, 3.10 eV, 3.02 eV, 2.75 eV, 2.62 eV, 2.64 eV, 2.30 eV) observed at the interface between non-irradiated and irradiated regions and almost quenced PL emission in the irradiated region.*

Conclusions

- The irradiation of ZnO films with 3 MeV, alpha particles at a dose of 5.3 kGy/h generates displacement defects such as ZnI only after longer irradiation time (more than 1000 s), while the OVs are produced even after a short exposure time (100s, 500 s).
- PL spectra of ZnO films show that intensity of the PL peak situated at 3.26 eV, corresponding to near band edge transition decreases after irradiation for 1000 s, while the PL emission in the region 3.10-2.30 eV increases with increasing the exposure time from 500 to 1000 s.
- New emission peaks appear at 3.10 eV and 2.64 eV in the PL spectrum of the films irradiated for 1000 s related to ZI formation.
- XRD data evidenced the increase of (002) oriented crystallites size from 21 nm to 29 nm after 8 h irradiation. The lattice constants remain unchanged, the residual stress is reduced at the interface and in the irradiated area.
- AFM 2D and 3D images reveal modification of the ZnO films surface roughness after irradiation. The films irradiated 8 h show a transition region at the interface between the irradiated and non-irradiated zone with R_{rms} = 11-15 nm and inside the irradiated area, R_{rms} = 23 nm in the localized zones containing clusters or large grains.
- The grains growth and the clusters formation on the ZnO films surface could be attributed to thermal annealing effect resulted by energy transfer from the incident particles, due to inelastic collisions. This effect could also account for the slight crystallite size growth and residual stress relaxation observed in the films volume in our study.

References

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Acknowledgement

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