An X-ray detector based on a Pt-MgZnO-Pt structure

Xiaocheng Guo1,a, Haoning Wang1,b, Hao Long1,c*, Jing Yang2,d*

1 Hubei Key Laboratory of Intelligent Wireless Communications, School of Electronic and Information Engineering, South-Central University for Nationalities, Wuhan 430074, People’s Republic of China

2 Cancer Center, Union Hospital, Tongji Medical College, Huazhong University of Science and Technology, Wuhan 430023, People’s Republic of China

aemail: 2020110187@mail.scuec.edu.cn, bemail: whn@mail.scuec.edu.cn

c*Corresponding authors: cemail: longhao_1118@163.com, demail: jingy200@hust.edu.cn

Abstract. Direct-conversion X-ray detectors made of wide-bandgap semiconductors have many advantages and important applications. In this study, a direct-conversion X-ray detector based on Pt-MgZnO-Pt structure was fabricated by radio frequency magnetron sputtering. The X-ray detection characteristics of the device were studied. The optimized device with a structure of Pt-MgZnO-Pt exhibited a responsivity of 72.6 nC Gy air$^{-1}$ cm$^{-2}$, response time of ~0.1 s (rise)/~0.4 s (fall), and a high SNR of 206 at a bias of 30 V under a dose rate of 100 mGy air/s. The experimental results demonstrate the good application prospects of the wide-bandgap MgZnO-based X-ray detectors.

1. Introduction

X-ray applications have been extensively used in plentiful momentous fields in recent years, such as medical diagnosis, safety systems, nuclear reaction monitoring, astronomy research and spacecraft navigation. Therefore, the precise detection of X-rays has put forward higher performance requirements. In this context, scintillator-based X-ray detectors with high sensitivity and fast response time are manufactured and widely used[1-4]. However, the application of the scintillator-based detector is limited by their low resolution because of the lateral diffusion of the scintillation light in the conversion layer[5]. Recently, due to the rise of semiconductor materials, more and more researchers become interested in X-ray detectors based on semiconductors, which could convert X-rays directly into electrical signals. Semiconductor direct-conversion X-ray detectors have many outstanding properties such as high energy, spatial resolution, low cost and suitable for large-scale production[6]. The current research is predominantly focused on narrow-bandgap semiconductors, such as Si, Se, Ge and CdZnTe[7, 8]. However, these detectors are used primarily for low-energy X-ray detection, and there are numerous fatal flaws, such as poor radiation resistance and short service life. Recently, perovskite has attracted considerable attention due to its high power conversion efficiency in solar cell applications. Many researchers have commenced studying perovskite-based light-emitting diodes and photodetectors including X-ray detectors[9, 10]. However, X-ray detectors based on perovskite still face the challenge of stability issues, which greatly limits their use for practical applications[11, 12].

Wide-bandgap oxide semiconductors, including ZnO, MgZnO, Ga$_2$O$_3$, TiO$_2$, NiO, have attracted much attention of researchers for their great physical and chemical properties, such as low intrinsic
carrier concentrations, large resistivity, insensitivity to solar, good radiation resistance, and environmental friendliness[13-18]. The development of direct-conversion X-ray detectors based on wide-bandgap oxides becomes an ideal choice. ZnO is a remarkable and widely used member of wide-bandgap oxide semiconductors. It has a large bandgap of 3.37 eV and has good stability and low cost. By alloying ZnO with MgO, the bandgap of MgZnO can be adjusted from 3.37 eV to 7.8 eV[19, 20]. Recently, ZnO-MgO alloys have be used for X-ray and ultraviolet (UV) photodetectors due to their widely adjustable bandgap, large resistivity, and high radiation resistance[21]. S. Han, et al. have obtained mix-phase MgZnO film on sapphire substrates to fabricate MgZnO detectors, which showed a high $I_{uv}/I_{dark}$ ratio of 864 at 40 V bias voltage under deep UV (230 nm)[22]. In the existing research, there are few reports exploring the performance of MgZnO X-ray detectors.

In this study, the MgZnO thin film was deposited on (0001) oriented substrate using radio frequency (RF) magnetron sputtering. A metal-semiconductor-mental (MSM) structured X-ray direct-conversion detector based on the MgZnO film was fabricated. The response of the detector to hard X-ray irradiation was demonstrated and analyzed.

2. Materials and Methods

The MgZnO thin film was deposited on (0001) oriented $c$-planed sapphire substrate using RF magnetron sputtering system. A ceramic ZnO:MgO (94/6 wt.%) target was used for the deposition of the films. Prior to deposition, the first step in this process was to ultrasonically clean the sapphire substrates sequentially in acetone, absolute ethanol and deionized for 20 min. Then the substrate was dried with a nitrogen gun. After they were loaded into the deposition chamber, pre-sputtering for 10 min to remove impurities and contaminants from the target surface. We filled the vacuum chamber with a mixed gas containing argon and oxygen (the flow ratio is 5:1). Usually, the pressure of the sputtering chamber was maintained at about 1.4 Pa. Then, the MgZnO film was prepared with a sputtering power of 90 W and a sputtering time of 120 min. Finally, Pt planar interdigital electrodes were deposited on the MgZnO film for 1.5 min by magnetron sputtering at room temperature. The width of the interdigital electrodes and the instance between adjacent finger electrodes were both 0.5 mm. The schematics of the fabricated Pt-MgZnO-Pt MSM X-ray detector and Pt planar interdigital electrodes are shown in Fig. 1 (a) and (b), respectively. The effective detection area is rectangular and the size is $1.2 \times 1.0$ cm$^2$.

![Fig. 1.(a) The stereo schematic of the MgZnO direct-conversion X-ray detector with MSM structure; (b) the plan sketch of the Pt interdigital electrodes.](image)

The current-voltage ($I$-$V$) characteristics measurements of the MSM detector in forward and reverse bias mode were carried out by varying the applied voltages using Keithley 2400 Source Measure Unit (SMU) Instrument. To stabilize the dark current, the detector was placed in dark for more than 24 hours before measuring the dark $I$-$V$ characteristic. Then, the transient responses of the detector were measured on a clinical linear accelerator (Varian Trilogy) with a tungsten target and an accelerating voltage of 6 MV.
3. Results & Discussion

The analysis of absorption spectral is helpful in studying the optical bandgap of the MgZnO material. Fig. 2(a) displays the optical transmittance spectra of the MgZnO film. As seen, the MgZnO film exhibits a transmittance of over 80% in the wavelength regions of 360 to 750 nm. The direct bandgap ($E_g$) can be determined by extrapolating the curve ($\alpha h\nu$) versus $h\nu$ to zero absorption (Tauc’s plots, as illustrated in Fig. 2(b)) and can be expressed as[23]

$$ah\nu = B(h\nu - E_g)^{1/2}$$  \hspace{1cm} (1)

Where $\alpha$ is the absorption coefficient, $h\nu$ is the incident photon energy, B is a constant. Then the optical bandgap of the device was calculated to be 3.60 eV. The combined effect of the Burstein-Moss effect and the high energy level resulting from the increase of the surrounding electron energy caused by Mg doping widen the bandgap of MgZnO[24, 25].

Fig. 2. (a) Transmittance spectra of MgZnO film in the MSM device. (b) The ($\alpha h\nu$)$^2$ - ($h\nu$) curves of MgZnO film.

Fig. 3 $I-V$ characteristics of the fabricated Pt-MgZnO-Pt detector in dark and under X-ray irradiation.
The bandgap widening shields most of infrared, visible, and part of UV light which may interfere with the X-ray detection. This positive effect can reduce the useless noise and inaccuracy in practical applications. The Mg contents in Mg$_x$Zn$_{1-x}$O could be calculated from optical bandgap by the following empirical formula:

$$E_g = 3.373 + 1.046x + 0.87x^2$$  \hspace{1cm} (2)

The $x$ value in Mg$_x$Zn$_{1-x}$O fabricated in this study was calculated to be 0.19.

The $I-V$ characteristics of the MgZnO MSM detector under the bias from $-30$ to $30$ V was measured in dark and under X-ray irradiation with a dose rate of 100 mGy$_{air}$/s are shown in Fig. 3. The dark current is not more than 3 nA even at 30 V bias. During the measurement, the current was found to significantly increase under X-ray irradiation. This phenomenon indicates that a certain high carrier concentration has been generated in the device.

The low dark current has a great effect on improving the signal-to-noise ratio (SNR) of detectors. The SNR was calculated by means of the following formula[26]:

$$\text{SNR} = \frac{I_{\text{signal}}}{I_{\text{noise}}} = \frac{\bar{I}_{\text{light}} - \bar{I}_{\text{dark}}}{\sqrt{\frac{1}{N} \sum_{i=1}^{N} (\bar{I}_{\text{light}} - \bar{I}_{\text{light}})^2}}$$  \hspace{1cm} (3)

where $\bar{I}_{\text{light}}$ and $\bar{I}_{\text{dark}}$ represent the average $I_{\text{light}}$ and average $I_{\text{dark}}$, respectively. And the denominator represents the standard deviation of the photocurrent. The calculated SNR of the device under X-ray irradiation at a dose rate of 100 mGy$_{air}$/s was 206. The responsivity is used for evaluating the sensitivity of a photodetector and is defined as following[26]:

$$R = \frac{\bar{I}_{\text{light}} - \bar{I}_{\text{dark}}}{D \cdot S}$$  \hspace{1cm} (4)

where $R$ refers to the responsivity, $D$ represents the dose rate, and $S$ refers to the photosensitive region area. The $R$ of the MgZnO MSM detector was calculated to be 72.6 nC Gy$_{air}^{-1}$ cm$^{-2}$.

The transient responses of the prepared Pt-MgZnO-Pt X-ray detector at a bias of 30 V with and without irradiation of X-ray is shown in Fig. 4. We define the rise time ($t_{\text{rise}}$) and fall time ($t_{\text{fall}}$) of the X-ray detector as the total time required for the output to rise from 10% to 90% of the pulse peak and to fall from 90% to 10%, respectively. Once irradiation with X-ray, the photocurrent will increase rapidly. When the X-ray excitation is turned off, the photocurrent quickly returns to the initial current value due to the trapped carriers[27]. Fig. 4 shows the $I-t$ curve measured at 30 V, and the photoresponse speed of the MgZnO-based MSM X-ray detector is estimated to be $\sim$0.1 s (rise)/$\sim$0.4 s (fall). The device presents a fast response speed.

![Fig. 4 The transient response characteristics of X-ray detector based on MgZnO at 30 V under X-ray irradiation with a dose rate of 100 mGy$_{air}$/s.](image-url)
4. Conclusions
In summary, a direct-conversion X-ray detector based on a Pt-MgZnO-Pt MSM structure was fabricated by RF magnetron sputtering. The $I-V$ characteristics under dark conditions and the transient response characteristics under hard X-ray irradiation confirmed that the detector showed good responses to X-ray with a responsivity of 72.6 nC Gy$_{air}^{-1}$ cm$^{-2}$, response time of $\sim$0.1 s (rise)/$\sim$0.4 s (fall), and a high SNR of 206 at 30 V under a dose rate of 100 mGy$_{air}$/s. We firmly believe that in the near future, by optimizing the material properties and detector structures, a greater breakthrough can be made in improving the performance of X-ray detectors based on wide-bandgap semiconductor.

Acknowledgments
This work was financially supported by the National Natural Science Foundation of China (Grant No. 12075322).

References
[1] Chen, Q., Wu, J., Ou, X., Huang, B., Almutlaq, J., Zhumekevnov, A. A., Guan, X., Han, S., Liang, L., Yi, Z., Li, J., Xie, X., Wang, Y., Li, Y., Fan, D., Teh, D. B. L., All, A. H., Mohammed, O. F., Bakr, O. M., Wu, T., Bettinelli, M., Yang, H., Huang, W., and Liu, X. (2018) All-inorganic perovskite nanocrystal scintillators. Nature, 561(7721): 88-93.
[2] Zhang, Q., Yan, J., Deng, B., Zhang, J., Lv, J., Wen, X., and Gao, K. (2017) An ultrafast X-ray scintillating detector made of ZnO(Ga). J. Instrum., 12(12): 12033.
[3] Shrestha, S., Fischer, R., Matt, G. J., Feldner, P., Michel, T., Osvet, A., Levchuk, I., Merle, B., Golkar, S., Chen, H., Tedde, S. F., Schmidt, O., Hock, R., Rührig, M., Göken, M., Heiss, W., Anton, G., and Brabec, C. J. (2017) High-performance direct conversion X-ray detectors based on sintered hybrid lead triiodide perovskite wafers. Nat. Photon., 11(7): 436-440.
[4] Pan, W., Wu, H., Luo, J., Deng, Z., Ge, C., Chen, C., Jiang, X., Yin, W.-J., Niu, G., Zhu, L., Yin, L., Zhou, Y., Xie, Q., Ke, X., Sui, M., and Tang, J. (2017) Cs$_2$AgBiBr$_6$ single-crystal X-ray detectors with a low detection limit. Nat. Photon., 11(11): 726-732.
[5] Heiss, W. and Brabec, C. (2016) Perovskites target X-ray detection. Nat. Photon., 10(5): 288-289.
[6] Owens, A. and Peacock, A. (2004) Compound semiconductor radiation detectors. Nucl. Instrum. Methods. Phys. Res. B, 531(1-2): 18-37.
[7] Guazzoni, C. (2010) The first 25 years of silicon drift detectors: A personal view. Nucl. Instrum. Methods. Phys. Res. B, 624(2): 247-254.
[8] Kasap, S., Frey, J. B., Belev, G., Tousignant, O., Mani, H., Laperriere, L., Reznik, A., and Rowlands, J. A. (2009) Amorphous selenium and its alloys from early xeroradiography to high resolution X-ray image detectors and ultrasensitive imaging tubes. Phys. Status Solidi B Basic Res., 246(8): 1794-1805.
[9] Lu, H., Liu, Y., Ahlawat, P., Mishra, A., Tress, W. R., Eickemeyer, F. T., Yang, Y., Fu, F., Wang, Z., Avalos, C. E., Carlsem, B. I., Agarwalla, A., Zhang, X., Li, X., Zhan, Y., Zakeeruddin, S. M., Emsley, L., Rothlisberger, U., Zheng, L., Hagfeldt, A., and Grätzel, M. (2020) Vapor-assisted deposition of highly efficient, stable black-phase FAPbI$_3$ perovskite solar cells. Science, 370(6512): 74.
[10] Lin, K., Xing, J., Quan, L. N., de Arquer, F. P. G., Gong, X., Lu, J., Xie, L., Zhao, W., Zhang, D., Yan, C., Li, W., Liu, X., Lu, Y., Kirman, J., Sargent, E. H., Xiong, Q., and Wei, Z. (2018) Perovskite light-emitting diodes with external quantum efficiency exceeding 20 per cent. Nature, 562(7726): 245-246.
[11] Li, X., Meng, C., Huang, B., Yang, D., Xu, X., and Zeng, H. (2020) All-perovskite integrated X-ray detector with ultra-high sensitivity. Adv. Opt. Mater., 8(12): 2000273.
[12] Kim, Y. C., Kim, K. H., Son, D.-Y., Jeong, D.-N., Seo, J.-Y., Choi, Y. S., Han, I. T., Lee, S. Y., and Park, N.-G. (2017) Printable organometallic perovskite enables large-area, low-dose X-ray imaging. Nature, 550(7674): 87-91.
[13] Li, M., Zhao, M., Jiang, D., Li, Q., Shan, C., Zhou, X., Duan, Y., Wang, N., and Sun, J. (2020) Optimizing the performance of ZnO/Au/MgZnO/SiO2 sandwich structured UV photodetectors by surface plasmons in Ag nanoparticles. Appl. Phys. A, 126(4): 310.

[14] Qin, Y., Long, S., Dong, H., He, Q., Jian, G., Zhang, Y., Hou, X., Tan, P., Zhang, Z., Lv, H., Liu, Q., and Liu, M. (2019) Review of deep ultraviolet photodetector based on gallium oxide. Chin. Phys. B, 28(1): 126-142.

[15] Vikas, L. S., Vanaja, K. A., Subha, P. P., and Jayaraj, M. K. (2016) Fast UV sensing properties of n-ZnO nanorods/p-GaN heterojunction. Sens. Actuator A Phys., 242: 116-122.

[16] Chen, D., Taylor, K. P., Hall, Q., and Kaplan, J. M. (2016) The neuropeptides FLP-2 and PDF-1 act in concert to arouse caenorhabditis elegans locomotion. Genetics, 204(3): 1151-1159.

[17] Liu, G., Zhang, M., Zhang, D., Zhou, J., Meng, F., and Ruan, S. (2014) Ultrahigh responsivity UV detector based on TiO2/Pt-doped TiO2 multilayer nanoﬁlms. J. Alloys Compd., 616: 155-158.

[18] Coskun, C., Look, D. C., Farlow, G. C., and Sizelove, J. R. (2004) Radiation hardness of ZnO at low temperatures. Semicond Sci Technol., 19(6): 752-754.

[19] Chen, X. M., Ruan, K. B., Wu, G. H., and Bao, D. H. (2008) Tuning electrical properties of transparent p-NiO/n-MgZnO heterojunctions with band gap engineering of MgZnO. Appl. Phys. Lett., 93(11): 112112.

[20] Srikant, V. and Clarke, D. R. (1998) On the optical band gap of zinc oxide. J. Appl. Phys., 83(10): 5447-5451.

[21] Liu, J. S., Shan, C. X., Li, B. H., Zhang, Z. Z., Yang, C. L., Shen, D. Z., and Fan, X. W. (2010) High responsivity ultraviolet photodetector realized via a carrier-trapping process. Appl. Phys. Lett., 97(25): 251102-3.

[22] Han, S., Hu, S. R., Cao, F. J., Liu, W. J., Zeng, Y. X., Jia, F., Xu, W. Y., Liu, X. K., Zhu, D. L., and Lu, Y. M. (2018) High signal/noise ratio deep UV detector with maximum response at 230 nm based on mix-phase MgZnO deposited under high laser energy condition. Europhys. Lett., 124(1): 18006.

[23] Lu, Z., Kang, H., Zhong, Z., and Zhang, T. (2016) Structural, electrical and optical properties of transparent conductive titanium–gallium–zinc oxide films by magnetron sputtering. J. Mater. Sci.: Mater. Electron., 27(12): 13271-13279.

[24] Li, C., Meng, F. Y., Zhang, S., and Wang, J. Q. (2010) Effects of Mg content and B doping on structural, electrical and optical properties of Zn1-xMgxO thin films prepared by MOCVD. J. Cryst. Growth, 312(12-13): 1929-1934.

[25] Burstein, E. (1954) Anomalous optical absorption limit in InSb. Phys. Rev. Lett., 93(3): 632-633.

[26] Zhang, H., Dun, G., Feng, Q., Zhao, R., Liang, R., Gao, Z., Hirtz, T., Chen, M., Geng, X., Liu, M., Huang, Y., Zheng, X., Qin, K., Tan, X., Wang, X., Xie, D., Yang, Y., Tian, H., Zhou, Y., Padture, N. P., Wang, X., Hong, J., and Ren, T.-L. (2020) Encapsulated X-ray detector enabled by all-inorganic lead-free perovskite film with high sensitivity and low detection limit. IEEE Trans. Electron Devices, 67(8): 3191-3198.

[27] Dere, A., Tatortoglu, A., Alshehmi, A. G., Alghamdi, A. A., Eltantawy, F. F., Farooq, W. A., and Yakuphanoglu, F. J. P. B.-c. M. (2017) A functional material based photodiode for solar tracking systems. Phys. B (Amsterdam, Neth.), 520: 76-81.