



Fast X-ray detectors based on bulk β -Ga₂O₃ (Fe)

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ABSTRACT

(010) EFG-grown Fe-doped β -Ga₂O₃ was tested as a low-noise X-ray detector with Ti/Au electrodes vertical structure. Its performance at low, high and no applied voltages was examined. The fabricated detector showed high X-ray detection performance manifested in its signal's short fall and rise time (< 0.3 s) in all operation modes, showing two orders of magnitude decrease in response time of β -Ga₂O₃ X-ray detectors. The same temporal response was exhibited by a tested Au/Ni/ β -Ga₂O₃/Ti/Au device. The detector's signal is also characterized by excellent linear relation with X-ray tube current and high signal-to-noise ratio (SNR) optimized at -5 V ($> 10^3$). Moreover, the X-ray-induced current signal exhibits high stability. Sub-band UV photocurrent signal showed a significantly slower response compared to X-ray-induced conductivity signal. Possible charge transport mechanisms involving ion migration are suggested and discussed. In this study, Fe doping is shown to significantly improve X-ray detection performance of Ga₂O₃, consolidating the applicability of Ga₂O₃ as a next-generation X-ray detector functioning with low power, high SNR and linearity, and significantly improved transient characteristics.

Introduction

β -Ga₂O₃ possesses multiple superior material properties including high thermal stability, ultra-wide bandgap, high device breakdown voltage, n-conductivity controllability, excellent radiation hardness and fabrication versatility [1–5]. Such characteristics are currently paving the road for high-performance devices based on β -Ga₂O₃ in power electronics [2], UV photodetection [3], gas detection, indirect radiation detection [6–8] and neutron detection [9]. Moreover, β -Ga₂O₃'s relatively high density

(6.44 gm/cm³) in addition to its high radiation resistance attracts strong attention as a candidate X-ray detector with exceptionally high performance in harsh environments. Recently, β -Ga₂O₃ Schottky barrier diode (SBD) as well as amorphous Ga₂O₃ (a-Ga₂O₃) flexible metal–semiconductor–metal (MSM) X-ray detectors have shown high X-ray responsivity and linearity [10–12]. SBD suffered from long response time ($\tau_{r1, d1} > 10$ s) under -10 V reverse bias which was significantly shortened in the absence of bias voltage. MSM as well as UV sensors [3] based on β -Ga₂O₃ have long transient characteristics as

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well, and photo-persistent current (PPC) is often encountered. This problem should be solved for successful X-ray or photodetector implementation. In this work, we report on the successful development of a fast, stable, high signal-to-noise ratio (SNR) and linear X-ray detector based on Fe-doped β -Ga₂O₃ bulk single crystal operating in low and high electric fields as well as self-powered mode.

Experimental

(010) EFG Fe-doped β -Ga₂O₃ with 5×5 mm² surface area and 0.5 mm thickness was used as the active detector material. 4×4 mm² Ti/Au (50/50 nm) contacts were deposited on each side after cleaning with acetone. For better contact formation, the device was annealed for 10 min at 400 °C in air. More details on device fabrication and its electrical and optical characterization can be found in our previous report [13]. The X-ray-induced conductivity experimental setup is shown in Fig. 1a. The X-ray tube (Cu, 45 kV, 40 mA) was equipped in a X-ray diffractometer (Empyrean, PANalytical), and no optics were used in front of the beam other than a mask and a Soller slit which were used to define the beam geometry. A Keithley 6487 source measure unit (SMU) was used to apply the external electric field and measure the induced current. The temporal X-ray-induced conductivity was measured in 0.3 s steps. Another Au (50 nm)/Ni (40 nm)/ β -Ga₂O₃/Ti (50 nm)/Au (50 nm) device was fabricated using the same procedures followed for the primarily studied device. We used UV LED (365 nm) for photoconductivity transient measurements. Time of flight–secondary ion mass spectroscopy (TOF–SIMS) analyses were conducted using a TOF SIMS V (ION TOF, Inc. Chestnut Ridge, NY) instrument. The surface was sputtered with 10 keV Cs⁺ beam at 30 nA for 20 scans over $550 \mu\text{m} \times 550 \mu\text{m}$ area, which only sputtered away a few nanometers of β -Ga₂O₃ (Fe), mainly surface contaminations. Analysis beam was 25 keV Bi₃⁺ ion at 0.15 pA current.

Results and discussion

A small detector thickness is preferred from the standpoint of charge collection efficiency and detector fabrication. Thus, an ideal candidate X-ray

sensing material should have high attenuation coefficient. Figure 1b shows the linear attenuation coefficient (for incoherent scattering [14]) of Ga₂O₃, compared with other semiconductor X-ray sensing materials. Other than lead compounds, the linear attenuation coefficient of Ga₂O₃ is among the highest exhibited by these currently studied semiconductor X-ray detector materials. Particularly, gallium compounds have early L and K edges improving their attenuation relative to those compared materials around these energies.

The current–voltage (I–V) behavior was studied in our previous report and is presented here in Fig. 1c [13]. Au/Ti/ β -Ga₂O₃ (Fe)/Ti/Au showed a near-Ohmic behavior and demonstrated a high resistivity close to $9.1 \times 10^{13} \Omega \text{ cm}$, which would lead to an enhanced SNR and thus motivated our efforts to develop Fe-doped β -Ga₂O₃-based radiation detectors.

Self-powered operation

Upon X-ray illumination (45 kV, 40 mA), the device exhibited X-ray-induced conductivity at no external applied voltage with the current reaching -21 pA as shown in Fig. 1d. For such no bias operation, the dark transient current was $-0.15 (\pm 0.05)$ pA, which is close to the SMU resolution. (The testing box cover was open compromising the noise level.) Assuming that a built-in voltage is responsible for the no-bias device operation leaking 0.15 pA, the calculated SNR is 139. More importantly, the rise and fall time of the signal is less than 0.3 s. This operation is expected to be in the photovoltaic mode, where no electrons originating from traps contribute to the signal (no detrapping). To gain insight into the underlying reason for the built-in voltage, we consider three different scenarios. First, it could indicate that the near-Ohmic (no turn-on voltage between -10 and 10 V) behavior exhibited by the device is indicative of Schottky contact formation. However, the current does not exceed -10 pA under an applied voltage of -100 V. Thus, a Schottky contact formation is highly unlikely to be responsible for the built-in voltage. The second possibility is that the built-in voltage was triggered by electron–hole formation from X-ray interaction (photoelectric and Compton scattering) near the irradiated surface. Charge carriers get trapped near the surface by deep-level defects that are potentially abundant near the surface, leading to a built-in electric field. The third possibility is that Fe

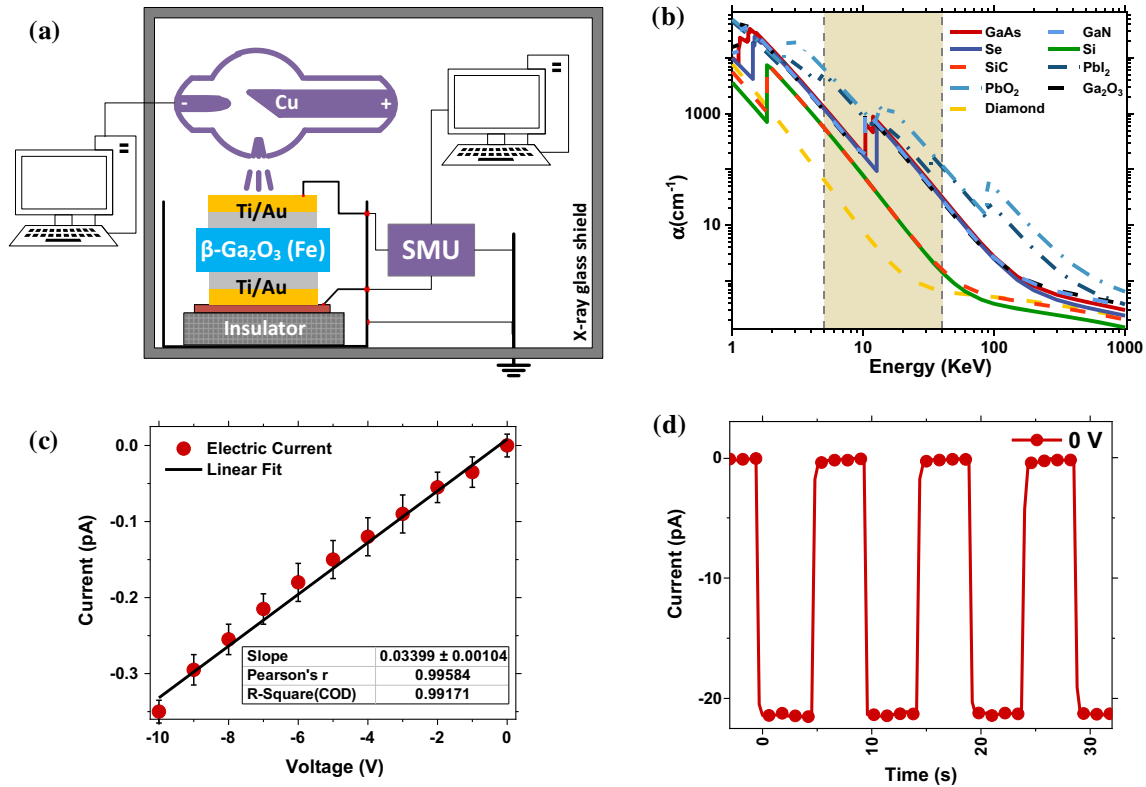


Figure 1 a X-ray-induced current measurement schematic setup, b linear attenuation coefficient from 1 keV to 1 MeV for Ga₂O₃ compared to a number of current and candidate X-ray sensing

materials. The highlighted region denotes energy range used for this study, c I–V curve of Fe-doped β-Ga₂O₃ along b-axis, d self-powered mode transient response of the device (period = 10 s).

dopants might have a nonuniform distribution throughout the thickness of the β-Ga₂O₃ crystal, leading to built-in voltage. In that case, the built-in voltage arises from the difference in electrons/holes concentrations through the thickness due to nonuniformity of Fe distribution. TOF-SIMS was used to obtain the information of Fe distribution through the thickness of β-Ga₂O₃ (Fe), as shown in Fig. 2, which revealed a nonuniform distribution of Fe doping. This is consistent with the assumption that the nonuniform distribution of Fe in EFG-grown β-Ga₂O₃ (Fe) is the underlying reason for the built-in voltage that allowed the X-ray detector operation under the absence of external applied voltage. From a sensor device viewpoint, the significant SNR and fast timing response show the potential of Fe-doped Ga₂O₃ as a promising X-ray detector for low-power operation.

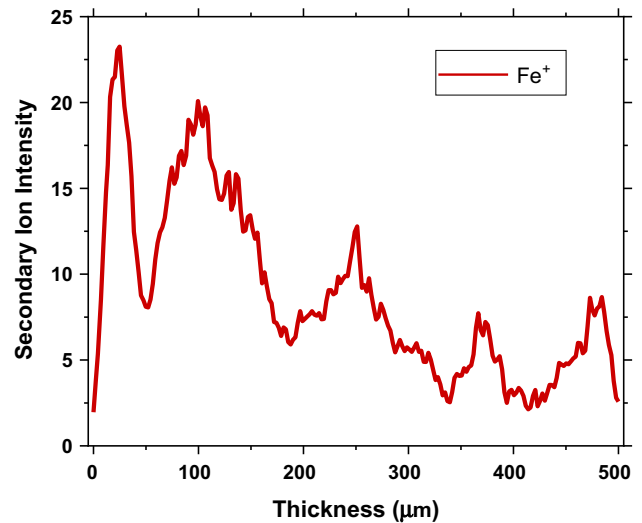


Figure 2 Fe⁺ secondary ion intensity through the β-Ga₂O₃ (Fe) thickness obtained by TOF-SIMS.

Temporal response

The device was tested under the low applied voltage of 5 V and -5 V, respectively. The transient response for 10-s X-ray (45 kV and 40 mA) period is shown in Fig. 3. The device showed no experimental lag in its response ($\tau_r, \tau_f < 0.3$ s) in the presence of low electric field similar to the case when it is absent. Compared to the same positive applied field, the signal was higher under the negative applied field.

To further examine the fast response time stability, the transient operations under -10 V, -20 V, -50 V and -100 V were examined. The results, shown in Fig. 4a, illustrate that the response time is not deteriorated under high electric fields as well, and it remains below 0.3 s.

To study whether the contact has a role in the device fast signal temporal behavior or not, Au/Ni/ β -Ga₂O₃ (Fe)/Ti/Au device was fabricated and the signal exhibited the same fast response as shown in Fig. 5a. We emphasize that the I–V behavior of this device was near-Ohmic as well, which is the same behavior exhibited by the Au/Ti/ β -Ga₂O₃ (Fe)/Ti/Au device.

Iron is expected to form Fe_{Ga} deep level around 0.8 eV below conduction band minimum (CBM) [15–17]. The fact that the signal rise and fall time is so fast, compared to undoped Ga₂O₃-based X-ray and photo-detectors, may either indicate that this deep-level band contributes to conduction or that it traps charge carriers without an effective followed detrapping process.

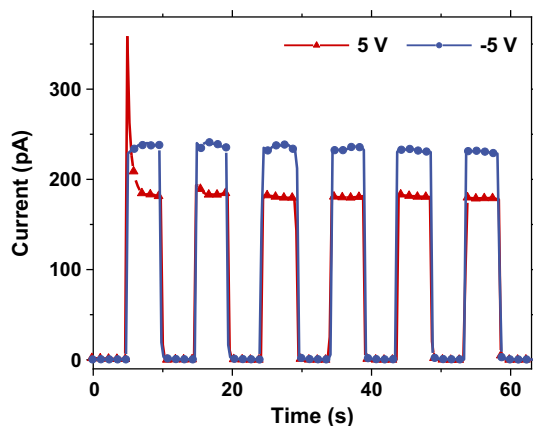


Figure 3 Low-voltage transient response of the device (period = 10 s). The negative current was flipped for illustration.

Signal-to-noise ratio

Considering that the device can be operated under zero, low and high applied voltages, the preferable operating regime should be the one that produces the highest SNR. Furthermore, a high SNR is critical if the operation of the device would be extended to pulse reading mode for energy-resolved X-ray detection. In our treatment, we define SNR as the ratio between the net induced current and the dark current ($\text{SNR} = \frac{I_{\text{x-ray induced}} - I_{\text{dark}}}{I_{\text{dark}}}$; $I_{\text{dark}} = \frac{3.2 \times V_{\text{applied}}}{9.1 \times 10^{13}}$; 3.2 is the detector area to length ratio in cm. However, it should be noted that in practical detector operation, sources of signal error include instrumental sources and statistical sources arising from the statistical nature of X-ray interaction with electrons. As shown in the inset of Fig. 4b, the SNR for operating voltages between -5 and -50 V stays above 800 and decreases for the higher applied voltages. This indicates that the increase in intrinsic dark current is higher than the increase in the induced current. More importantly, the SNR stays above 1000 for applied voltages between -5 and -20 V, and it is further optimized at -5 V exceeding 1200. These results clearly show that the device possesses better characteristics at low-voltage operation compared to high-voltage region.

To gain more insight about the charge carriers transport properties, the single-carrier Hecht equation fitting was used to estimate the drift length of the effective charge carriers, as shown in Fig. 4b. It should be noted that although single charge carrier is assumed here, this assumption is only made in order to gain a quantitative estimate for the transport properties of charge carriers, and the contribution from both electrons and holes should not be overlooked in highly accurate treatments as both types of charge carriers could contribute to the signal. In fact, the mobility–lifetime product ($\mu\tau$) factor calculated from this rough treatment was $2.28 \times 10^{-5} \text{ cm}^2/\text{V}$, which yields $45.6 \mu\text{m}/456 \mu\text{m}$ carrier drift length for 10 V/100 V, respectively. These values are higher than that obtained from electron beam-induced current (EBIC) for electrons and holes [22–25], suggesting that they both contribute to the signal.

Table 1 shows the characteristic temporal response and SNR of β -Ga₂O₃ (Fe) X-ray detector compared to previously reported Ga₂O₃-based X-ray detectors as well as other X-ray detectors based on wide bandgap

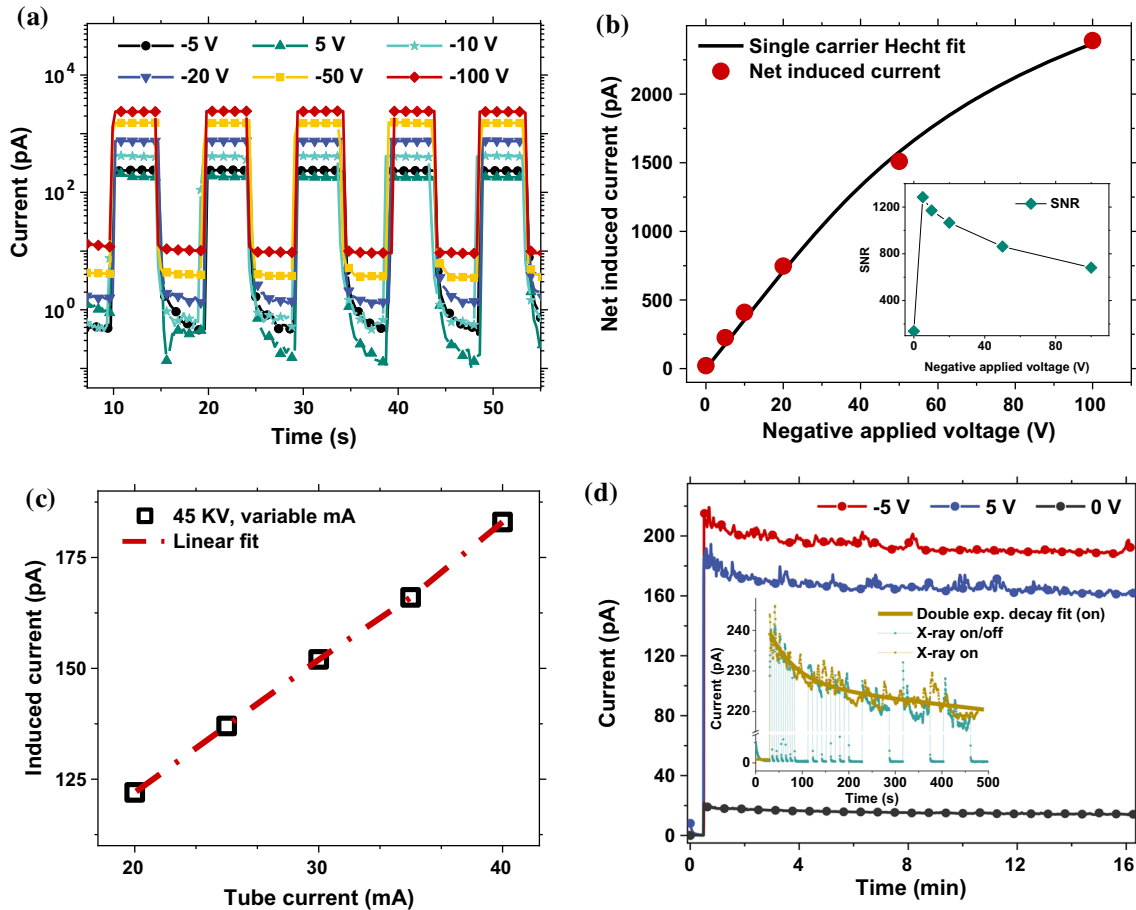


Figure 4 a Transient response of the Au/Ti/ β -Ga₂O₃ (Fe)/Ti/Au device under low and high applied voltages (period = 10 s). b Single charge carrier Hecht fitting [18–21]. The inset shows SNR at tested applied voltages (45 kV, 40 mA). c Response current linearity. d Stability test for 15 min. The inset shows

double exponential decay fitting for 15-min stability test under -5 V applied voltage with X-ray tube operating at 45kV and 40 mA, compared to ON/OFF testing under the same applied voltage and tube parameters. The negative current was flipped for illustration.

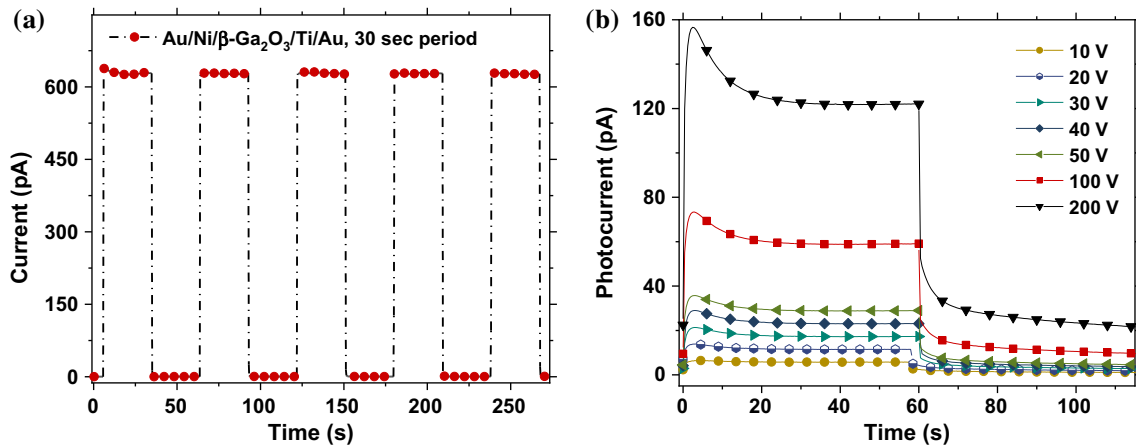


Figure 5 a X-ray-induced (45 kV and 40 mA) conductivity transient response for Au/Ni/ β -Ga₂O₃/Ti/Au device under 10 V (period = 1 min), b sub-band UV transient photoconductivity at different applied voltages (period = 2 min).

Table 1 Comparison of temporal response and SNR ratio of X-ray detectors based on wide bandgap semiconductors, showing the significant improvement in timing characteristics of Fe-doped β -Ga₂O₃-based X-ray detector compared to unintentionally doped β -Ga₂O₃ and amorphous Ga₂O₃, without compromising SNR

Material	Preparation method	Rise time (s)	Decay time (s)	SNR	References
a-Ga ₂ O ₃	RF sputtering (PO ₂ 1.4 × 10 ⁻³ Pa)	15.5	1.1	–	[12]
	RF sputtering (PO ₂ 1.0 × 10 ⁻³ Pa)	50.1	3.3	> 10 ⁴	
Unintentionally doped β -Ga ₂ O ₃	EFG	18.3 (– 15 V) < 0.02 (0 V)	20.9 (– 15 V) < 0.02 (0 V)	> 800 (– 15 V) –	[10]
	β -Ga ₂ O ₃ (Fe)	EFG	< 0.3	> 10 ³ (low voltages) > 10 ² (0 V and high voltage)	This work
GaN	MOCVD	15	–	< 30	[26]
		< 0.02 (0 V)	< 0.02 (0 V)	< 20	[27]
GaN (Fe)	HVPE	28	2	180	[28]
Zn ₂ SiO ₄	RF sputtering and annealing	< 1	< 1	> 10 ³	[29]
		~ 30	~ 8	480	[30]
ZnO	RF sputtering	–	~ 0.1	< 5	[31]
	RF sputtering	0.2	0.2	300	[32]

semiconductors. It can be seen that β -Ga₂O₃ (Fe) X-ray response showed approximately two orders of magnitude transient response improvement compared to previously reported Ga₂O₃ X-ray detectors, without compromising SNR. Compared to other wide bandgap X-ray detectors, β -Ga₂O₃ (Fe) shows outstanding compromise between the transient response and SNR, especially under low applied voltages, which can be achieved using 5 V batteries.

Linearity

The X-ray tube power was varied by changing the tube current at constant tube voltage under – 5 V applied voltage in order to test the device response linearity. As shown in Fig. 4c, the X-ray-induced current response is highly linear with the current of the X-ray tube. Such linear response encourages further investigation on operating the device in the pulse mode (potentially low-power X-ray spectrometer) as well as in the current mode (X-ray counter as in regular imagers with very high dynamic range when operated at high voltage).

Stability

Continuous X-ray (40 kV, 40 mA)-induced conductivity measurements were conducted for 15 min at 5 V, – 5 V and 0 V to assess the device response stability. As shown in Fig. 4d, the device shows very

high stability at tested operating voltages, all with the same fast rise time. However, a closer look to the signal time behavior reveals small exponential decay within the first minute that stabilizes after that. The inset of Fig. 4d shows a double exponential decay fitting for the continuous X-ray time-dependent response (– 5 V, 45 KV, 40 mA), resulting in $\tau_1 = 57.7$ s and $\tau_2 > 10^6$ s (stability). Two possible scenarios can be responsible for this behavior.

First, the poor heat conduction of β -Ga₂O₃ may have caused a slight increase in the sample temperature due to device operation such as X-ray material interaction and charge flow in the device when the bias voltage is applied. This temperature increase deteriorates the transport properties of charge carriers, as illustrated by previous EBIC studies, which would lead to a decrease in the induced current [22–25]. In this scenario, the total drop in the current should significantly differ at different operating voltages, with higher drop at higher voltages, and/or higher X-ray power. Moreover, the detector under continuous operation should suffer from continuous induced current drop, which is not the case in our study. Thus, we exclude that the poor thermal conductivity was the reason behind this effect.

The second scenario can be triggered by space charge accumulation on the surface near the electrodes, causing an internal electric field opposite to the external applied electric field and leading to slight

polarization effect. In further analysis of this behavior, ionic conductivity of defects (e.g., oxygen vacancies) should be considered.

Comparing the time-dependent evolution of the current under continuous and ON/OFF X-ray illumination under the same measurement conditions, we observed memory effect of the signal, as shown in Fig. 4d inset. In other words, this decay effect is independent of the X-ray illumination state after the initial X-ray pulse, which consolidates the second scenario involving the polarization effect. After the space charge is formed on the semiconductor surface near the electrodes, they stay there as long as the electric field is applied; thus, the electric field is deteriorated and remains this way even in the absence of charge carrier generation. This behavior could be related to ionic migration of oxygen vacancies (ionic conductivity).

Temporal UV photoconductivity under different applied voltages

To investigate the UV photocurrent transient response, the sub-band gap excitation with 365 nm LED (3.4 eV) was used since the band gap of $\beta\text{-Ga}_2\text{O}_3$ (Fe) is 4.45 eV [13]. The UV photoconductivity results are shown in Fig. 5b. 10–90% rise/fall time improved from 2 s/16 s at 5 V to 1 s/10 s at 200 V, and the photocurrent had a linear dependency with applied voltage (unlike the X-ray-induced current voltage dependence that follows Hecht model). The enhanced transient and charge carrier transport properties of the X-ray-induced current (compared to sub-band UV photocurrent) are probably caused by the enhanced charge carrier transport properties for charge carriers excited above the deep acceptor level of Fe_{Ga} , often referred as E_2 (~ 0.8 eV below CBM) [16, 17] in the case of X-ray-induced response.

Conclusion

Fe-doped $\beta\text{-Ga}_2\text{O}_3$ was investigated as a direct X-ray detection material motivated by its high resistivity and ultra-low leakage current. $\beta\text{-Ga}_2\text{O}_3$ (Fe) with Ti/Au electrodes demonstrated high SNR under three operation modes including low applied voltage (> 1200 at -5 V), high applied voltage (> 650 at -100 V) and self-powered operation mode (> 100). We also observed high linearity between X-ray-

induced photocurrent and X-ray tube current. More importantly, the signal's rise and fall time was below 0.3 s, which offers excellent measurement time resolution. This fast response was independent of the applied voltage and was not electrode dependent, as indicated by the fact that the response did not deteriorate upon testing a Au/Ni/ $\beta\text{-Ga}_2\text{O}_3$ (Fe)/Ti/Au device. The device showed high stability upon continuous illumination for 15 min with an exponential decay behavior ($\tau = 57.7$ s) which stabilizes shortly after that. The sub-band photocurrent measurements showed significantly slower transient response. Further investigations of the device response to UV light, especially in the energy range between E_2 and CBM band (3.65 to 4.45 eV), are necessary to evaluate the device performance as solar-blind deep UV sensor. Analysis of $\beta\text{-Ga}_2\text{O}_3$ (Fe) X-ray detector characteristics including charge carrier transport suggests that both holes and electrons contribute to the signal formation and that a slight polarization effect takes place. The results demonstrate the great potential of $\beta\text{-Ga}_2\text{O}_3$ (Fe) as a radiation-resistant X-ray detector with excellent temporal response for a wide range of applications.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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