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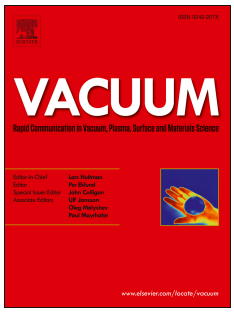


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# Comparative study of radiation tolerance of GaN and $Ga_2O_3$ polymorphs

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The mechanisms of ion-induced defect formation and physical characteristics promoting radiation tolerance of wide and ultra-wide bandgap semiconductors are not well-studied and understood. In contrast to gallium nitride (GaN), gallium oxide ($Ga_2O_3$) can be crystallized in several polymorphs having different crystal structures and physical properties. In the preset paper, the damage buildup in wurtzite GaN as well as in corundum ($\alpha$-) and monoclinic ($\beta$-) $Ga_2O_3$ polymorphs bombarded at room temperature with 40 keV $P^+$ ions is studied by Rutherford backscattering/channeling spectrometry. We demonstrate that ion-beam-induced damage formation in $Ga_2O_3$ is different from that observed in GaN and dramatically depends on the polymorph type. Both $Ga_2O_3$ polymorphs cannot be rendered amorphous and exhibit considerably higher damage saturation at ~90% of the full amorphization as compared to that of GaN. Intriguing enough the metastable $\alpha$-$Ga_2O_3$ demonstrates considerably higher radiation resistance as compared to the most thermodynamically stable $\beta$-$Ga_2O_3$ polymorph. Furthermore, our results indicate that the sample surface and dynamic annealing play a significant role in the ion-induced damage formation processes in all Ga-based compounds studied.

**Highlights**

Kinetics of ion irradiation damage accumulation in $\alpha$- and $\beta$-$Ga_2O_3$ and GaN is studied
$\alpha$-$Ga_2O_3$ is more susceptible to radiation damage than GaN
$\alpha$-$Ga_2O_3$ is considerably higher radiation resistant then the stable $\beta$-$Ga_2O_3$
Mechanisms of radiation damage formation in the $\alpha$- and $\beta$-$Ga_2O_3$ are different

## 1 Introduction

Gallium nitride (GaN) and gallium oxide ($Ga_2O_3$) are semiconductors with wide bandgaps of 3.4 and 4.5-5.3 eV, respectively. Research interest in these materials has been stimulated by their unique properties utilized in a number of promising applications [1-8]. Indeed, GaN has become one of the major semiconductors in electronic device technology after Si, GaAs, and SiC, and still expands its presence on the market primarily for optoelectronic devices able to work in the blue and UV spectral ranges [1-4]. In contrast, research on $Ga_2O_3$ is still not in its maturity [5-8]. It can be crystallized in different polymorphs such as $\alpha$-, $\beta$-, $\gamma$-, $\delta$- and $\kappa(\varepsilon)$-$Ga_2O_3$. Among these polymorphs $\beta$-$Ga_2O_3$ is the only thermodynamically stable phase at ambient conditions. It has been successfully grown in large sizes by Edge-defined Film-fed Growth (EFG) technique and other bulk methods. Properties of this polymorph have been extensively studied up to now [6-10]. In addition, due to its ultra-wide bandgap (4.5 – 4.9 eV) and extremely high break-down field value (~8 MV/cm), which is larger than those of III-nitrides (3.3 MV/cm for GaN) and silicon carbide

(2.5 MV/cm for 4H-SiC), gallium oxide is considered as the most promising candidate for the next generation power electronics as well as for UV photodetectors and sensors [5, 6].On the other hand, the α-$Ga_2O_3$ phase has even wider bandgap ($E_g$ = 5.3 eV) [5]. Thus, it can potentially possess even a higher breakdown electrical field value, and given its similar structure to other wide bandgap materials such as AlN, it should be possible to produce functional heterostructures or tune bandgap through alloying [7]. Despite this phase is metastable, the temperature of its transition to the thermodynamically stable $\beta$-phase exceeds 650°C [5], which gives a reason to believe that it is possible to create a new generation of power devices on its base [7].

The main electronics applications of $Ga_2O_3$-based devices involve high-power grids and switching systems with a low power loss and truly solar-blind deep ultraviolet (UV) photodetectors [5, 6]. A lot of these devices are expected to operate in radiation harsh environment, for example in avionic and space systems, in military systems, space communication, ozone-layer monitoring, robotic inspection systems used near reactor cores or in accident response etc., where they could be exposed to energetic particles. In addition, ion implantation is a well-established powerful technique for materials modification and, in particular, for selective area doping in electronic device processing. However, the ion irradiation of semiconductors is always accompanied by formation of radiation defects which can dramatically affect properties of the irradiated layer. Therefore, a deep understanding of the ion-irradiation-induced processes of defects formation and their evolution is urgently needed for development of new electronic devices and expanding their functionality.

A large number of works have already been devoted to the studies of defect accumulation during the implantation of accelerated heavy and medium-mass ions into GaN [11-18]. In its turn, the data on ion-irradiation-induced defect formation in gallium oxide are limited and all the main results concern only the most stable $\beta$-phase [19-23]. Initial study on the phase stability and strain accumulation in mixed $\alpha/\kappa(\varepsilon)$-$Ga_2O_3$ phase was recently performed [24]; however systematic investigation of irradiation-induced effects in metastable $Ga_2O_3$ polymorphs is missing. At the same time, data on both polytypes of gallium oxide are urgently required.

The aim of the present paper is to study the process of ion-beam induced defect formation in the two main polymorphs of gallium oxide ($\alpha$- and $\beta$-$Ga_2O_3$) and to reveal the features of this process by comparing the defect accumulation in $Ga_2O_3$ and GaN under room temperature (RT) 40 keV P ion bombardment.

**Table 1.** Irradiation parameters employed in the present study ($n_{at}$ is the target atomic density, $R_{PD}$ – depth position of maximum of vacancy generation, $R_P$ – projected ion range as obtained by the SRIM code [27] simulations). Ion fluences, fluxes and their dose equivalents in DPA are also presented.

| Target | $n_{at}$, $10^{22}$cm$^{-3}$ | $R_{PD}$, nm | $R_P$, nm | Fluence range $10^{14}$ cm$^{-2}$ | Dose range, DPA | Flux $10^{12}$ cm$^{-2}\times$s$^{-1}$ | Dose rate $10^{-3}$DPA$\times$s$^{-1}$ | 1 DPA $10^{14}$ cm$^{-2}$ |
|---|---|---|---|---|---|---|---|---|
| $\alpha$-$Ga_2O_3$ | 10.3 | 17 | 28 | 0.9-43.3 | 0.15-6.9 | 1.5 | 2.4 | 6.3 |
| $\beta$-$Ga_2O_3$ | 9.45 | 17 | 31 | 0.9-11.1 | 0.15-1.8 | 1.5 | 2.4 | 6.3 |
| GaN | 8.85 | 17 | 31 | 5-150 | 1.0-29 | 1.9 | 3.6 | 5.0 |

## 2 Experimental

Three types of semiconducting samples were used in the present study. Two kinds of research grade epilayers grown at Ioffe Institute, St. Petersburg, Russian Federation, namely MOVPE wurtzite [0001] GaN [25] and corundum-like [0001] $\alpha$-$Ga_2O_3$ grown by halide vapor phase epitaxy

(HVPE) [26] both deposited on *c*-plane sapphire substrates, and commercial monoclinic [-201] $\beta$-$Ga_2O_3$ single crystals grown by EFG technique (Novel Crystals Technology). The samples were irradiated by 40 keV $P^+$ ions in a wide range of doses employing 500 kV HVEE implanter. All the implantations were carried out at RT and 7° off the channeling directions. In order to compare the irradiation effects in all materials studied, the ion doses were normalized to the average number of displacements per target atom (DPA) taken at the maximum of the nuclear energy loss profile $n_v$: DPA = $n_v \times \Phi / n_{at}$, where $n_{at}$ is the atomic concentration and $\Phi$ is the ion fluence. The corresponding $n_v$ values were calculated using the TRIM code (version SRIM 2013) [27] with an effective atomic threshold displacement energy equal to 25 eV for all Ga, O, and N sub-lattices. The irradiation parameters are summarized in the Table 1. Note, the ion fluxes were close to each other for all the implants thus, making possible to neglect the dose-rate effects [12, 13, and 23].

Implantation-produced disorder was measured by Rutherford backscattering (RBS/C) spectrometry in a channeling mode with 0.7 MeV $^4He^{2+}$ ions backscattered to 103° from the incident beam direction. The effective number of scattering centers (referred to below as "relative disorder") was deduced from the RBS/C spectra using one of the conventional algorithms as described in Ref. 28.

## 3 Experimental results

### 3.1 General observations

Typical experimental RBS/C spectra are plotted in Fig. 1 to illustrate the ion-beam induced damage formation in the three gallium-containing compounds. It is seen from figure 1 that for virgin (unimplanted) samples the backscattered ion yield collected while probing He ions come to the surface in a channeling direction is fairly low. Hence, all the as-grown samples are of reasonably good crystalline quality where probing ions are able to go deep along the lattice channels. The peak at channel 290 corresponds to the backscattering from Ga atoms at the sample surface. Further we analyze the damage only in the Ga sublattice due to much higher sensitivity of RBS technique to heavy elements as compared to light ones. The number of backscattered ions increases after bombardment with 40 keV P ions due to formation of various defects in the target. Interestingly, for all compounds, the disorder-depth profiles look more or less similar at low DPA values, despite that the different doses are needed to obtain a measurable change in the backscattered yield. Indeed, at low DPA the surface Ga peak starts to grow up and dominates the damage in all the compounds, while the second peak appears in the crystal bulk for higher doses. However, with a dose increase, the defect accumulation exhibits drastically different behavior in the three compounds as demonstrated in Secs. 3.2-3.4.

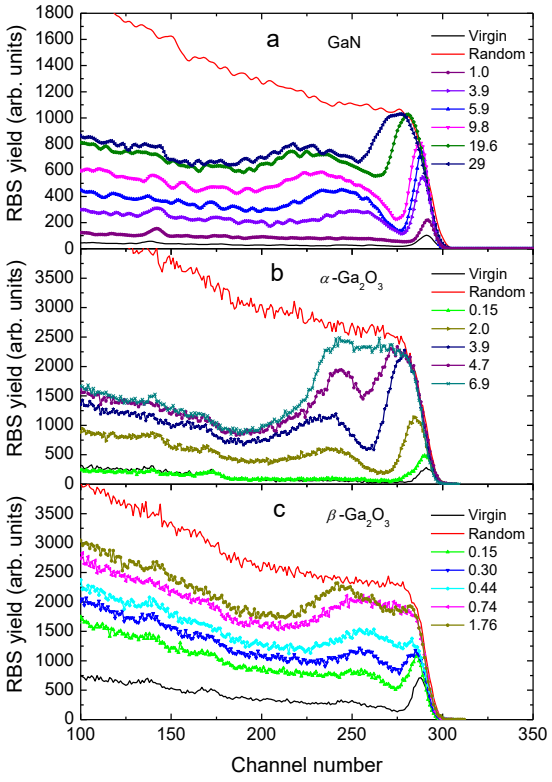


**Fig. 1.** RBS/C spectra of (a) GaN, (b) α-$Ga_2O_3$, and (c) $\beta$-$Ga_2O_3$ implanted with 40 keV P ions to different doses as indicated in the legends in the units of DPA. See Table 1 the column entitled "Fluence equivalent to 1 DPA" to get values of ion fluences in ions/cm$^2$.

The selected depth distributions of relative disorder created in Ga compounds, as deduced from the RBS/C spectra, are shown in Fig.2. The ion doses are chosen in such a way to obtain a disorder level at bulk maxima of ~ 0.1-0.15 from the complete amorphization. These doses are low enough to prevent accumulation of a high defect concentration in the materials and, accordingly, the possible disorder saturation does not affect the damage formation at this stage. The TRIM calculated vacancy generation functions $n_v$ are also plotted in Fig. 2. It should be noted that the generation functions in GaN and $\beta$-$Ga_2O_3$ coincide each other with a difference less than 5%, so only one of them is shown for simplicity.

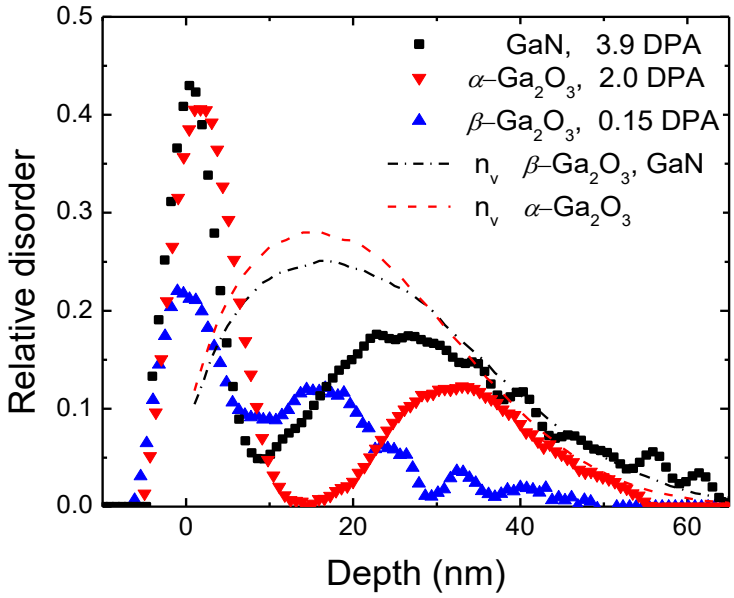


**Fig.2.** Depth profiles of relative disorder in GaN, α- and $\beta$-$Ga_2O_3$ implanted at RT with 40 keV P ions to doses indicated. Profiles of ion-generated lattice vacancies (with arbitrary vertical scale) predicted by TRIM simulations for gallium compounds are shown by the dashed lines. Note that TRIM predicted vacancy distribution in GaN is similar to that in $\beta$-$Ga_2O_3$.

Furthermore, Fig. 2 confirms that the main features of disorder depth distributions are similar for all the cases under consideration. Indeed, the disorder profiles are bimodal over depth with two distinct peaks situated at the target surface and in the bulk. Thus, hereinafter we refer them as a surface disordered layer (SDL) and bulk defect peak (BDP). In order to study the behavior of these two peaks with the ion dose, we have performed deconvolution of the damage-depth distributions using the approximation considering defect distribution at the surface as a flat disordered/amorphous layer, and their concentration in the bulk follows a Gaussian distribution. The corresponding procedure is described in detail in [16, 29]. Figs. 3 *a*, and *b* illustrate dose dependences of the SDL width and the relative disorder in the BDP maximum, respectively. The bulk peak positions are drawn in Fig.4. Ion-induced damage accumulation in each compound is discussed in detail separately in the next sections.

### 3.2 GaN

The near-surface damage buildup in GaN proceeds as layer-by-layer amorphization originating at the sample surface, which is a nucleation site for amorphization (see Fig. 3a). The maximum of the bulk disorder profile appears at a depth corresponding to the maximum of elastic energy losses for relatively low ion fluences [12, 14] and shifts deeper into the target with increasing ion dose, as seen in Fig. 4. In addition, the relative concentration of stable damage in the bulk peak exhibits saturation at ~40% (black squares in Fig. 3 b). Qualitatively similar damage buildup behavior, with layer by-layer surface amorphization and disorder saturation in the bulk, has also been observed in GaN bombarded at RT with keV light and intermediate mass ions [11, 12]. Such a damage buildup behavior is characteristic of GaN bombarded at RT when the chemical effects of implanted species are negligible [11, 13-15, 30]. Disorder in the sample bulk accumulates primarily as interstitial-based planar defects parallel to the (0001)-planes [4, 11, 12, and 31]. Some explanations of physical reasons of the observed behavior will be given in the next sections.

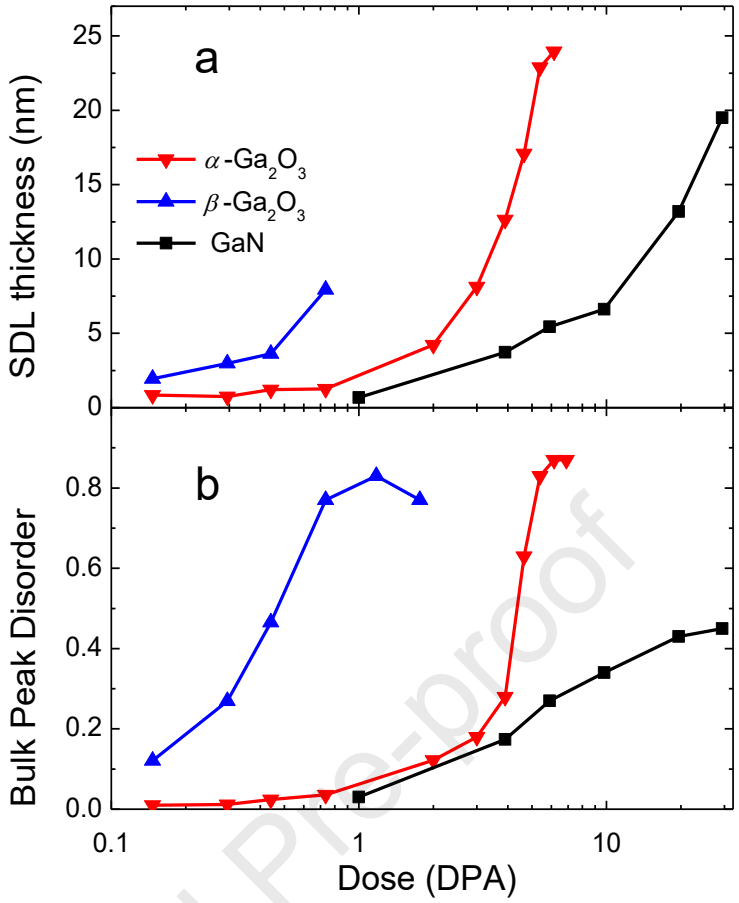


**Fig.3.** Dose dependence of (a) the thickness of the surface disordered layer and (b) of the maximum of the bulk defect peak in GaN as well as $\alpha$– and $\beta$-$Ga_2O_3$ implanted at RT with 40 keV P ions.

### 3.3 $\alpha$-$Ga_2O_3$

Similar to GaN, a well-resolved SDL peak is observed in the relative disorder depth profiles in $\alpha$-$Ga_2O_3$ (see Figs. 1 and 2). However, according to Figs. 2 and 3a, the doses needed to achieve the same SDL thickness in $\alpha$-$Ga_2O_3$ are 5-10 times lower than those in GaN. Interestingly, BDP in $\alpha$-$Ga_2O_3$ appears at a depth, which is much larger than the depth corresponding to the maximum of nuclear energy loss of stopping ions. To a certain extent, this is surprising since the primary displacements generation maxima are located at approximately the same depth for all three semiconductor materials (see Fig.2). The maximum of BDP in $\alpha$-$Ga_2O_3$ originates close to the maximum of the implanted ions distribution (~28 nm) and slightly shifts deeper towards the crystal bulk with increasing ion dose. Thus, the damage formation in this gallium oxide polymorph could be promoted by the implanted impurity. Figs. 1 and 3 clearly show that the disorder in both the surface damaged layer and in the bulk does not reach a complete amorphization level. Instead, it saturates at ~90% of full amorphization. Hence, ion-induced damage in the $\alpha$-$Ga_2O_3$ is formed more efficiently as compared to that in GaN. However, the doses needed to create a measurable amount of radiation defects is still above 1 DPA, which suggests efficient dynamic annealing processes in this polymorph at RT.

### 3.4 $\beta$-$Ga_2O_3$

It is seen from Fig. 3 that the damage accumulation in $\beta$-$Ga_2O_3$ is much more efficient as compared to that of both GaN and $\alpha$-$Ga_2O_3$. Indeed, a noticeable amount of stable radiation damage is formed in $\beta$-$Ga_2O_3$ already after 0.15 DPA, which is more than an order of magnitude lower than that in $\alpha$-$Ga_2O_3$ or GaN (2 and 3.9 DPA respectively). Thus, it can be concluded that the radiation resistance of $\beta$-$Ga_2O_3$ is about an order of magnitude lower than that of GaN and $\alpha$-$Ga_2O_3$. These findings may indicate the difference in the mechanisms of radiation damage formation in the two gallium oxide polymorphs. Moreover, it can be suggested that diffusion length of mobile point defects in *beta* polytype of gallium oxide is smaller than that in GaN and $\alpha$-$Ga_2O_3$.

Despite the efficient damage accumulation in the BDP of $\beta$-$Ga_2O_3$, the SDL growth rate is relatively low in this polymorph. Note that SDL to BDP height ratio is smallest in $\beta$-$Ga_2O_3$ as compared to that of GaN and $\alpha$-$Ga_2O_3$. The maximum of BDP formed in $\beta$-$Ga_2O_3$ corresponds well to the depth of maximum of ballistic energy loss profile of stopping P ions. Similarly to $\alpha$-phase, the bulk damage in $\beta$-$Ga_2O_3$ slightly shifts deeper to the bulk with the dose increase and saturates at ~0.8 of full amorphization (see Fig.3b).

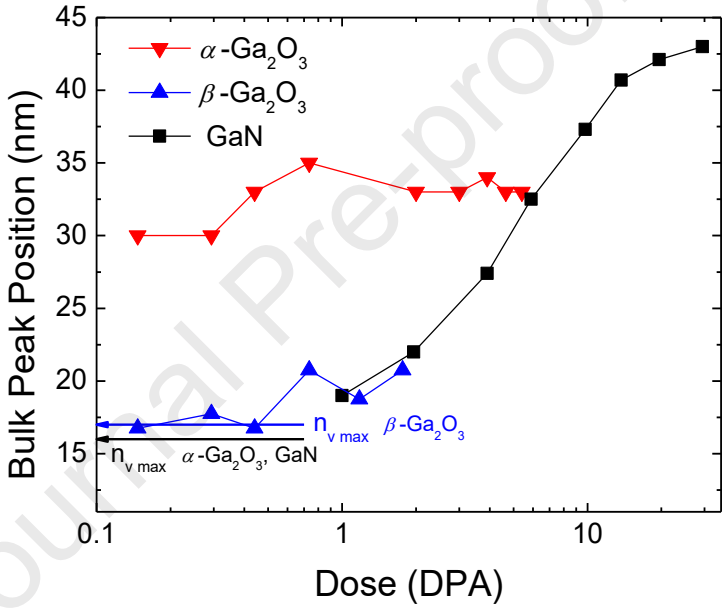


**Fig.4.** Bulk peak position in GaN and both $Ga_2O_3$polymorphs bombarded at RT by 40 keV P ions. The arrows indicate positions of maxima of primary defects generation functions in gallium compounds as calculated by the TRIM code [27] simulations.

## 4 Discussion

Despite the similarity mentioned in the Section 3.1, significant differences between defect formation in GaN and both polymorphs of $Ga_2O_3$ are clearly identified. The saturation level is usually about 0.3 - 0.4 for GaN [11-14], approximately 0.9 for $\alpha$-$Ga_2O_3$, and 0.8 (this study) or 0.9 [20] for [-201] $\beta$-$Ga_2O_3$, which values are quite close each other taking into account the experimental errors.

The behavior of the BDP observed in GaN, can be understood in the framework of the model proposed previously [17]. In this model the mechanism leading to the BDP shift and its saturation is related to the movement of the inner boundary of the surface disorder layer inward GaN bulk with increasing ion dose. This shift strongly affects the diffusion of mobile point detects (MPD) and their accumulation. It was experimentally established that the inner boundary of SDL is a very efficient sink for MPD [16]. Thus, the number of point defects available to form stable damage decreases and the maximum of their distribution shifts deeper to the bulk. This leads to the apparent shift of BDP position deeper to the crystal bulk and eventually to its saturation since the number

of point defects needed to promote its further growth [32] decreases. However, this mechanism could not be the case of $\beta$-$Ga_2O_3$ since practically no shift of the BDP position is observed in this material. Instead, the following scenario can be proposed to explain the BDP behavior in this polymorph. It is initially created in the region where a lot of ion-beam-induced displacements of target atoms take place, which suggests efficient interaction between MPDs and the formation of various immobile defect complexes. The damage accumulation rate in $\beta$-$Ga_2O_3$ is very high. In such a narrow dose range, the position of stable damage cannot change much, since it simply cannot collect enough defects to move anywhere.

In contrast to both $\beta$-$Ga_2O_3$ and GaN, ion-beam-generated MPDs in $\alpha$-$Ga_2O_3$ can probably form complexes with implanted impurities that may explain coincidence of the BDP position with the depth distribution of the implanted ions.

Reasons underlying saturation of the radiation damage level below amorphization in gallium compounds are still unknown. Possibly implantation-induced phase transitions can play a role in gallium oxides. For example, as was shown in [33] an implantation-induced phase transition from $\beta$ to $\kappa$-$Ga_2O_3$ can take place during ion irradiation which dramatically change both the damage accumulation kinetics as well as channeling conditions during the RBS/C analysis. Thus, more detailed studies are required to better understand the physical nature of the discussed effects.

## 5 Conclusions

The kinetics of damage accumulation in three gallium compounds: $\alpha$- and $\beta$-$Ga_2O_3$ and GaN under 40 keV P ion irradiation at RT has been experimentally studied and compared. Our results indicate that the radiation resistance of $\alpha$- and $\beta$-$Ga_2O_3$ polymorphs is dramatically different and lower as compared to that of GaN. At the same time, the damage accumulation rate both in the crystal bulk and at the surface of $\beta$-$Ga_2O_3$ is about an order of magnitude higher than those of $\alpha$-$Ga_2O_3$ and GaN indicating that $\beta$-$Ga_2O_3$ is more susceptible to ion irradiation damage. Moreover, in $\beta$-$Ga_2O_3$ the BDP is situated at the depth corresponding well to the position of the maximum of primary defect generation, while that in $\alpha$-$Ga_2O_3$ is found near the maximum of implanted atom distribution. The BDP maximum position formed in $\beta$-$Ga_2O_3$ and $\alpha$-$Ga_2O_3$ only slightly changes with the ion dose. This finding points out the difference in the mechanisms of radiation damage formation in the two gallium oxide polymorphs. Moreover, diffusion lengths of simple point defects generated by stopping ions in the $Ga_2O_3$ polymorphs have to be different. The BDP value saturates below full amorphization in all these semiconductors; however, the saturation level of both $Ga_2O_3$ polymorphs is approximately twice larger than that of GaN (0.9 vs. 0.45).

## Acknowledgements

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Kinetics of ion irradiation damage accumulation in $\alpha$- and $\beta$-$Ga_2O_3$ and GaN is studied

$\alpha$-$Ga_2O_3$ is more susceptible to radiation damage than GaN

$\alpha$-$Ga_2O_3$ is considerably higher radiation resistant than the stable $\beta$-$Ga_2O_3$

Mechanisms of radiation damage formation in the $\alpha$- and $\beta$-$Ga_2O_3$ are different

**Declaration of interests**

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: