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Citation: Appl. Phys. Lett. 112, 082104 (2018); doi: 10.1063/1.5011971

View online: https://doi.org/10.1063/1.5011971

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Effect of 1.5 MeV electron irradiation on β -Ga₂O₃ carrier lifetime and diffusion length

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(Received 3 November 2017; accepted 3 February 2018; published online 21 February 2018)

The influence of 1.5 MeV electron irradiation on minority transport properties of Si doped β -Ga₂O₃ vertical Schottky rectifiers was observed for fluences up to 1.43×10^{16} cm⁻². The Electron Beam-Induced Current technique was used to determine the minority hole diffusion length as a function of temperature for each irradiation dose. This revealed activation energies related to shallow donors at 40.9 meV and radiation-induced defects with energies at 18.1 and 13.6 meV. Time-resolved cathodoluminescence measurements showed an ultrafast 210 ps decay lifetime and reduction in carrier lifetime with increased irradiation. *Published by AIP Publishing*.

https://doi.org/10.1063/1.5011971

Wide bandgap semiconductors are desirable for numerous applications from solar blind UV detection to high power electronics, to which interest in the β -polymorph of Ga₂O₃ continues to develop. ^{1–3} Monoclinic β -Ga₂O₃ is the most stable polymorph, until recently available only as *n*-type, and carries an electronic bandgap (E_g) of 4.8–4.9 eV. ^{4,5} There has been significant success with various growth techniques in producing high quality crystals and thin films of β -Ga₂O₃ of large diameter. Recently, β -Ga₂O₃ has been used in Schottky rectifiers, demonstrating high reverse breakdown voltage, ⁶ and as a gate material for MOSFETs. ^{5,7}

Wide bandgap devices are often good candidates for applications implemented in harsh environments such as low earth satellite orbit, which require resistance to high fluences of radiation and extremes of temperature. The fluences of high energy protons and electrons for space borne applications depend on many factors including the somewhat unpredictable local solar weather. Defects generated by irradiation in wide bandgap semiconductors have been explored at length, and as one might expect, the types of defects created are dependent upon which type of radiation is applied. Irradiation-induced lattice defects can introduce trap states in the forbidden gap, leading to altered material properties including diffusion length, mobility, and lifetime which ultimately affect device performance. 10-14 This necessitates an exploration of the radiation hardness of β -Ga₂O₃, which is expected to be high based on its bond strength. 15

High-energy electron irradiation has been used to investigate defects in many semiconductor materials including GaN 16 and ZnO. 17 The type of defect generated from such irradiation is dependent upon the incident beam energy and displacement energy, $E_{\rm d}$, which has been found to have a dependence which is inversely proportional to the lattice constant. 15 The effects of 1.5 MeV electron irradiation on Si-doped $\beta\text{-Ga}_2\text{O}_3$ Schottky rectifiers have been observed recently, and irradiation resulted

Cathodoluminescence (CL) probes recombination behavior through electron beam excited radiative recombination. This reveals details about carrier recombination behavior and can yield information about the material composition, ¹⁸ trap activation energy, ^{19–21} defect density, ^{22,23} and plasmonic mode dispersion.^{24,25} The spatial resolution of this technique is limited by the minority carrier diffusion length and size of generation volume. 26,27 The minority carrier diffusion length can be determined by the Electron Beam Induced Current (EBIC) technique. ^{28–30} The addition of time resolution to the cathodoluminescence technique allows for the direct measurement of spectral decay, which can reveal radiative recombination lifetime, τ , 31 and the presence of stress and strain. 32 In this letter, we combine EBIC, Continuous Wave Cathodoluminescence (CWCL), and Time-Resolved Cathodoluminescence (TRCL) techniques to study the effects of 1.5 MeV electron irradiation on β-Ga₂O₃ carrier transport properties including recombination lifetime and minority carrier diffusion length.

The samples employed in this study consisted of epitaxial β -Ga₂O₃ grown on single crystal Sn-doped β -Ga₂O₃ substrates. The Sn-doped substrates were grown by the edge-defined film-fed method and showed the (001) surface orientation (Tamura Corporation, Japan). The carrier concentration in the substrates was previously determined from Hall measurements to be $3.6 \times 10^{18} \, \mathrm{cm}^{-3}.^{33}$ Epitaxial layers were Si-doped and grown by Hydride Vapor Phase Epitaxy (HVPE) courtesy of Novel Crystal Technology. As-grown layers were ~20 μ m thick, with subsequent chemical mechanical polishing reducing final thicknesses to $10 \, \mu \mathrm{m}.^1$ Majority electron concentrations in the epitaxial layers are summarized in Table I.

Vertical Schottky rectifiers were fabricated by electron beam evaporation of top Schottky and bottom Ohmic contacts. The Ti/Au (20/80 nm) Ohmic contact was deposited over the entire bottom area, and the Ni/Au Schottky contacts were

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in a carrier removal rate—defined as loss in carrier density (cm⁻³) per fluence (cm⁻²)—of ~4.9 cm⁻¹ and a significant reduction in reverse-bias current density for higher voltages.¹

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TABLE I. Room temperature material parameters for Si-doped β -Ga₂O₃ in response to 1.5 MeV electron irradiation.

Fluence	n (cm ⁻³)	L (nm)	L ₀ (nm)	$\Delta E_a (meV)$	τ (ps)
Non-irradiated					
$1.79 \times 10^{15} \text{cm}^{-2}$					
$1.43 \times 10^{16} \mathrm{cm}^{-2}$	3.32×10^{16}	243 ± 6	186 ± 19	13.6 ± 2.4	138 ± 15

patterned on the top face using lift-off photolithography. The diameter of the circular Schottky contacts was $210\,\mu\text{m}$. A schematic cross-section can be found in Fig. 1(a). Current-Voltage characteristics were recorded before and after irradiation with 1.5 MeV electrons. Irradiation was performed at Korea Atomic Energy Research Institute for fluences of 1.79×10^{15} and $1.43 \times 10^{16}\,\text{cm}^{-2}$ at a current of 1 mA.

The minority carrier diffusion length, L, was determined using the EBIC technique on Schottky barriers (contacts) in the planar configuration. 19 EBIC is minority carrier sensitive due to the charge collection through the Schottky contact due to the electric field in the space charge region. Although both types (electrons and holes) of excited non-equilibrium carriers are generated via electron beam diffuse, only minority carriers are collected by the Schottky diode and the resultant current recorded. Therefore, only minority carriers contribute to the EBIC signal. The EBIC was recorded during line-scans of 10s duration performed with a Philips XL-30 Scanning Electron Microscope (SEM). Using a beam energy of 20 keV, the EBIC was measured for each sample, without bias, as a function of the distance from the Schottky contact, x, as depicted in Fig. 1(a). To prevent the possible effects of SEM beam electron injection, each EBIC line-scan was performed on a previously unexposed region. 19-21 EBIC was recorded for sample temperatures 295, 330, 370, and 395 K by using a temperature-controlled stage monitored by an integrated platinum resistance thermometer with 0.5°K accuracy.

CL measurements were conducted at room temperature on an Attolight Allalin 4027 Chronos SEM. The accelerating voltage used for all CL measurements was 10 keV. Dispersion was accomplished using a single grating with 150 grooves/mm blazed at 500 nm. CWCL was recorded using a Newton 920 CCD from Andor, which is sensitive in the 180–1100 nm range. TRCL was collected using an Optronis Streak camera for a 2 ns range using an SEM beam pulse of ~8 ps duration at 80 MHz.

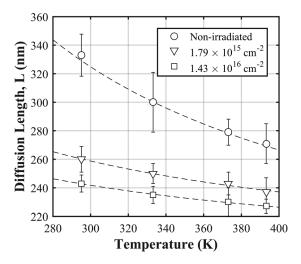


FIG. 2. Dependence of the minority carrier (hole) diffusion length on temperature for irradiated and non-irradiated samples with exponential fits (dotted lines) revealing activation energy, $\Delta E_a.$

EBIC line-scans were used to determine L according to 34,35

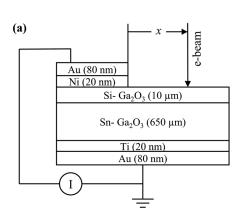
$$I(x) = I_0 x^{\alpha} \exp\left(-\frac{x}{L}\right). \tag{1}$$

Here, I_0 is a scaling constant, x is the beam-to-junction distance, L is the minority (hole) carrier diffusion length, and α is a constant related to surface recombination velocity. The zero-bias depletion region width in Au/β - Ga_2O_3 Schottky diodes has been estimated previously to be $d=14\,\mathrm{nm}$ and does not significantly interfere with these measurements since $x\gg d$. To ensure accuracy, the determination of the diffusion length is measured at x>2L. 28,37 To achieve linearity, the value of α was estimated as -0.5, indicating a low influence of surface recombination velocity. EBIC data are displayed in Fig. 1(b) for the non-irradiated and most irradiated samples along with a linear fit generated by Eq. (1). The values of L are presented in Fig. 2 for all temperatures and doses tested.

L depends on temperature according to

$$L(T) = L_0 \exp\left(\frac{\Delta E_a}{2kT}\right). \tag{2}$$

Here, L_0 is the asymptotic diffusion length, ΔE_a is the thermal activation energy, k is the Boltzmann constant, and T is



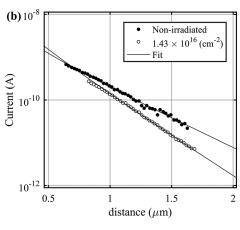


FIG. 1. (a) Schematic side view of the EBIC setup and sample showing the electron beam, distance, x, for the planar (Schottky) configuration, and current probe. (b) Room temperature EBIC data collected from irradiated and non-irradiated samples with the corresponding fit.

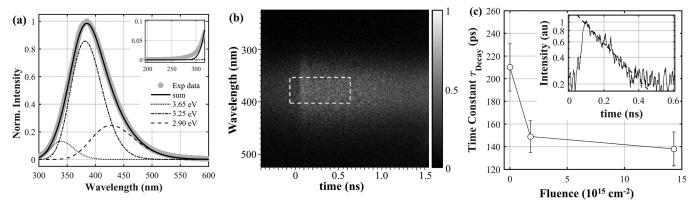


FIG. 3. (a) CWCL emission peak at 380 nm with a Gaussian spectral decomposition showing emission from bands at 3.65, 3.25, and 2.95 eV and the absence of band-to-band emission (inset). (b) TRCL output from a streak camera centered at 380 nm. The dashed white rectangle indicates the region where τ was determined. (c) Cathodoluminescence decay time constant (open circles) for non-irradiated and irradiated samples, with the inset showing a non-irradiated sample normalized decay spectrum at 380 nm with an exponential fit (dotted line).

the temperature. ΔE_a determined by Eq. (2) is related to a charge trap in the forbidden gap and is a parameter for determining the temperature dependence in L.38 The room temperature value of L was initially 335 nm for the nonirradiated sample and tended to decrease for increasing temperature for all samples. The reduction of L with temperature can be caused by increased scattering or recombination; the temperature dependent measurement of lifetime could assist in discerning which dominates. However, present hardware limitations prevent measuring lifetime for higher temperatures and will be addressed in future studies. The values of ΔE_a , listed in Table I, were 40.9 meV for the non-irradiated sample and reduced to 18.1 and ultimately 13.6 meV in response to higher irradiation doses. The introduction of these trap states results in a reduction in L likely due to an increased recombination rate.

The CWCL emission spectrum is presented in Fig. 3(a) showing no band-to-band emission at 255 nm [4.9 eV, cf. Fig. 3(a), inset] and a broad emission centered at \sim 380 nm $(\sim 3.26 \,\mathrm{eV})$ with a Full-Width at Half Maximum (FWHM) bandwidth of 80 nm. A Gaussian decomposition of the spectrum revealed that the emission can be represented by a sum of Gaussian peaks centered at 3.65 eV, 3.25 eV, and 2.9 eV. The band at 3.65 eV is consistent with the charge transition level for one of the three inequivalent oxygen vacancy (V_O) sites.³⁹ The emission at 3.25 eV is attributed to the recombination of free electrons with self-trapped holes, 40 while that of 2.9 eV is attributed to donor-acceptor pairs. 41 The emission spectrum showed no changes after irradiation. High resolution SEM imaging and polychromatic CWCL did not reveal any changes in the surface morphology, which indicates that no significant changes in the β -Ga₂O₃ structure was induced by 1.5 MeV electron irradiation.

TRCL measurements of lifetime, τ , shown in Fig. 3(b) were carried out with a 20 nm bandwidth centered at 380 nm. The transient CL intensity obeys a single exponential decay of the form

$$C(t) = C_0 \exp\left(-\frac{t}{\tau}\right) + A. \tag{3}$$

Here, C_0 is the initial integrated CL intensity, A is the extended decay, t is the delay time after excitation pulse, and

 τ is the decay time constant. The extended decay, A, is meant to describe the background luminescence persisting longer than the excitation repetition period of 12.5 ns (80 MHz). Consistent with the short term UV decay behavior in the study by Harwig and Kellendonk⁴² and Binet and Gourier,⁴³ we observed a persistent CL signal (t > 2 ns). At the wavelengths shown, in the time scale observed, and using a very short excitation pulse of ~8 ps, we claim that A is effectively a constant and is employed as such in the determination of τ from Eq. (3).

The value of τ was 215 ps for the non-irradiated sample and reduced to 151 and 138 ps, indicating an increase in the recombination rate in response to irradiation. These are much faster than the lifetime of ~30 ns found in other studies. ^{43,44} In the study by Binet and Gourier, ⁴³ the Time Resolved Photoluminescence (TRPL) decay from the UV (3.2 eV) luminescence was not observed at t < 100 ns. This indicates that the ultrafast [t < 2 ns, cf. Fig. 3(b)] radiative recombination reported in this work may differ from that found at longer delay times. The lifetime we report in this paper applies to non-equilibrium holes for the n-type β -Ga₂O₃.

The effects on L and τ are reflected in the diffusivity constant, D, and mobility, μ , by way of the Einstein relation

$$L = \sqrt{D\tau} = \sqrt{\mu k T \tau / q}.$$
 (4)

Here, q is the charge of the electron. The room temperature minority carrier mobility immediately after electron beam excitation calculated from Eq. (4) using independently measured values of L and τ decreased with 1.5 MeV electron dose from 204 to 176 and ultimately $166\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$. This hole mobility is comparable to an electron mobility of $153\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ found in Ref. 45. Recently, hole mobilities have been measured at $0.2\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$, well above their computationally predicted value of $1\times10^{-6}\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ using the Einstein relation [Eq. (4)]. In contrast to the transient behavior studied by us from the TRCL decay [cf. Fig. 3(c), inset], Yamaoka *et al.* succeeded in measuring the TRPL rise time at 8 K of 24 ps corresponding to the tunneling time through the potential barrier from free to trapped exciton states. In fact, the stability of the self-trapped hole is poor above $90-120\,\mathrm{K}$, lending an increase in hole mobility.

The calculated hole mobility in this work originates from the short recombination lifetimes observed in the ultrafast dynamics of TRCL. The diffusion length of minority holes determined in this study is an average quantity involving all mobile minority carriers excited during electron beam excitation and may vary for other crystal orientations such as (010) or ($\overline{2}01$). Previously, the ultrafast majority carrier mobility was shown to be dependent upon delay time and excitation pulse magnitude in GaAs⁴⁹ and GaP.⁵⁰ At such a short timescale as in this report, the mobility is transient, only holding this value for very brief instance since the nonequilibrium carrier concentration decays very quickly, and should not be compared to the steady-state Hall mobility. Therefore, we conclude that the hole mobility determined in this study represents the transient response to \sim 8 ps pulses of electron injection.

The activation energy of 40.9 meV, found prior to irradiation, matches closely with the donor ionization energy found in previous studies. 43,45,51,52 These shallow donor levels have been attributed to Si doping.⁵³ The irradiationinduced reduction in L and ΔE_a is due to the generation of trap states between the conduction band and the shallow donor levels. The monotonic reduction in activation energy is due to the introduction of these shallow interband traps serving as more energetically favorable pathways for trapped electrons to return to the conduction band and consequently recombine with holes.⁴³ This is confirmed by the simultaneous reduction in lifetime. The trap states formed are likely due to oxygen displacement, which can result in either oxygen vacancies, pairs of oxygen and gallium vacancies, or complexes of both. After irradiation, the recombination pathway remains energetically unchanged as evidenced by the comparison of CWCL measurements. While deeper levels may be formed, experimentally we did not observe them. Post-irradiation annealing may repair irradiation-induced defects⁵⁴ and move the activation energy toward its preirradiated value⁵⁵ although it was not performed in this study.

The temperature dependence of the minority carrier diffusion length was observed by EBIC and allowed for the extraction of activation energy before and after 1.5 MeV electron irradiation. Exposure to 1.5 MeV electrons led to a marked decrease in L and lifetime, suggesting the generation of defects interfering with minority carrier transport. The carrier lifetime was measured by TRCL to determine the effects of irradiation on the ultrafast recombination rate. The initial activation energy was attributed to shallow donor levels commonly found in $\beta\text{-Ga}_2\text{O}_3$ samples, while the irradiated samples displayed reduced activation energy from irradiation-induced trap states. The induced trap states assisted recombination, as evidenced by the reduction in lifetime.

The research at the University of Central Florida was supported in part by the National Science Foundation Award ECCS No. 1624734 and the US-Israel BSF Award No. 2014020. The work at the University of Florida was partially supported by the Department of the Defense, Defense Threat Reduction Agency HDTRA1-17-1-0011 (Jacob Calkins, monitor). The content of the information does not

necessarily reflect the position or the policy of the federal government, and no official endorsement should be inferred. We appreciate the electron irradiations performed at Korea University through Professor Jihyun Kim.

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