

Experimental Estimation of Electron-Hole Pair Creation Energy in β -Ga₂O₃

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ABSTRACT

The applicability of using Electron Beam Induced Current (EBIC) measurements on Schottky barriers to obtain the mean electron-hole pair creation energy in β -Ga₂O₃ is reported. It is shown that, when combined with Monte Carlo simulation, this approach yields for Si, GaN, and 4H-SiC a data set consistent with empirical expressions proposed earlier in the literature for many different semiconductors. The method is then applied to β -Ga₂O₃, where complications related to hole trapping in the material give rise to a strong gain in EBIC and have to be carefully treated and taken into account. When this is done, the mean electron-hole pair energy formation is found to be 15.6 eV, in reasonable agreement with the values predicted by empirical expressions.

The electron-hole pair creation energy E_i is an important parameter determining the sensitivity of charged particles and x-ray detectors. As a rule, it is obtained by measurements of collected current induced in a Schottky diode by charged particles or soft x-rays whose intensity is carefully measured with a well-calibrated detector with known E_i . For such measurements it is important to keep the collection efficiency close to unity to avoid incomplete charge collection. This is usually achieved by applying a high reverse bias voltage to a detector to enhance carrier transport to the electrodes. When the collection efficiency approaches 100%, the collected current should be independent of applied bias.

In semiconductors with a small diffusion length such as GaN and β -Ga₂O₃, the collection probability is lower than 1, even at high enough biases. Additionally, in many structures the presence of current gain has been observed, even at rather small bias voltages [1-5]. Therefore, the increase of collected current as a function of applied bias cannot be considered as a universal method to measure E_i . Measurements of E_i using e-beam excitation in a scanning electron microscope are more convenient, because in such measurements, the beam energy can be adjusted and beam current can be easily measured using, for example, a Faraday cup. Therefore, it is not necessary to use a reference structure. This method has been already used in [6] for GaAs in the geometry with the e-beam parallel to the p-n junction.

However, in this geometry the collected current can be reduced due to surface recombination of carriers. The geometry with the Schottky barrier or p-n junction perpendicular to e-beam seems to give more reliable results. The advantage of such geometry is that the real collection probability can be obtained by fitting the dependence of collected current on beam energy with a calculated one [7,8] and therefore there is no need to assume it to be equal to 1.

In the present paper, the approaches to measurements of the mean electron-hole pair creation energy in the Scanning Electron Microscope (SEM) are discussed. The results

obtained were compared with the empirical dependences of E_i on bandgap. This approach is used to estimate the mean electron-hole pair creation energy for β -Ga₂O₃, an emerging wide-bandgap material showing great promise in high-power electronics and UV optoelectronics [2, 3]

First, let us discuss the basis of EBIC measurements of the mean electron-hole pair creation energy. The collected current I_c in the electron beam induced current (EBIC) method for a Schottky barrier or p-n junction can be calculated as [7-9]

$$I_c = \int_0^{\infty} \psi(z) h(z) dz, \quad (1)$$

where $\psi(z)$ is the collection probability and $h(z)$ is the depth dependent excess carrier generation rate (generation function). As shown by Donolato [9], the collection probability $\psi(z)$, which is the current induced by a unit charge at a depth z , can be obtained as a solution of the homogeneous diffusion equation with corresponding boundary conditions. For thick semiconductor structures with a Schottky barrier, the equation for $\psi(z)$ can be written as

$$\partial^2 \psi(z) / \partial z^2 - \psi(z) / L^2 = 0 \quad (2)$$

with the boundary conditions $\psi(W) = 1$ at $z=0$ and $\psi(z) \rightarrow 0$ at $z \rightarrow \infty$, where $L = (D\tau)^{0.5}$ is the excess carrier diffusion length, D is the ambipolar diffusivity equal to the minority carrier diffusivity at low excitation level and τ is the excess carrier lifetime. W is the depletion region width. For L independent of z , $\psi(z) = \exp[-(z-W)/L]$ for $z > W$ and $\psi(z) = 1$ for $z \leq W$. For p-n junctions the collection probability can be calculated using a similar diffusion equation with corresponding boundary conditions.

The generation function can be calculated by the Monte Carlo simulation. It should be stressed that in such simulations, not only the backscattered energy but also the energy loss in the metal contact can be taken into account. To simplify further calculations, the generation

function is usually approximated by the Gaussian function. For example, for common semiconductor materials it can be described as

$$h(z) = \frac{1.76}{R} U \exp[-7.5(z/R - 0.3)^2], \quad (3)$$

for the case of Si [10]. (U in Eq. (3) is the total generation rate of excess carriers, $R(\text{nm}) = 17.1 \times E_b(\text{keV})^{1.75}$ is the electron range, and E_b is the beam energy).

For GaN, the generation function can be described as [11]

$$h(z) = \frac{3.207}{R} U \exp[-A(\frac{z}{R} - 0.11)^2], \quad (4)$$

where $R(\text{nm}) = 13.2 \times E_b(\text{keV})^{1.75}$ and $A = \begin{cases} 42.8, & z < 0.11R \\ 16.5, & z \geq 0.11R \end{cases}$.

For SiC, $h(z)$ was obtained in [12] as

$$h(z) = \frac{1.87}{R} U \exp[-7.94(z/R - 0.28)^2] \quad (5)$$

where $R(\text{nm}) = 18.25 \times E_b(\text{keV})^{1.75}$.

For Ga_2O_3 [13]

$$h(z) = \frac{1.603}{R} \exp[-A(\frac{z}{R} - 0.22)^2], \quad (6)$$

where $R(\text{nm}) = 7.34 \cdot E_b(\text{keV})^{1.75}$ and $A = \begin{cases} 12.86, & z < 0.22 \cdot R \\ 3.97, & z \geq 0.22 \cdot R \end{cases}$.

Fitting of the measured dependence of collected current on beam energy with calculated ones allows not only to obtain the diffusion length [7,8] and in some structures W [14,15], but also the total generation rate U, which is equal to

$$U = \frac{E_b I_b \eta}{E_i} \quad (7)$$

where E_b and I_b are the beam energy and current, respectively, and η is the portion of beam energy deposited inside the semiconductor. Thus, it is seen that if η is known, E_i can be obtained from the EBIC measurements.

Values of η can be obtained by the Monte Carlo simulation and they are equal to 0.93, 0.905, 0.767 and 0.781 for Si, 4H-SiC, GaN and Ga_2O_3 , respectively. Of course, for the Schottky barriers, the energy loss in metals should be taken into account, especially for the case of thick metal and/or low beam energy, however, the losses can also be obtained by the Monte Carlo simulation of two-layer structures. Thus, if U is obtained from fitting the $I_c(E_b)$ dependences, E_i values can be easily calculated.

To check the method discussed, EBIC measurements were carried out on standard Al-p-Si and Au-n-Si Schottky barriers. The obtained E_i values varied in the range of 3.63-3.65 eV that well correlates with the literature value of 3.66 eV [16]. For GaN, the difference between the calculated E_i value of 9.59 eV from the EBIC measurements and the reported value of 8.9 eV [17] is about 7%. For 4H-Si, a comparison is less informative because the previously reported values vary in the range from 5.05 [18] to 8.6 eV [19]. Our approach gives for 4H-SiC the value $E_i = 8.23$ eV.

For the prediction of E_i values in new materials, it can be useful to have the empirical relation between the bandgap and the mean electron-hole pair ionization energy. A few expressions for such relation have been proposed [20-23]. In Figure 1 these dependences are shown together with the experimental data from [17, 20] and the data obtained in the present work. It is seen that the experimental data fit well to the expressions from [20-22], with the best agreement of the experimental data to the expression [20]

$$E_i = 2.8E_g + 0.6eV \quad (8)$$

As follows from Figure 1, the E_i data obtained by the method used in the present letter correlates rather well with the experimental data obtained for different materials and the

empirical dependences. In particular, for wide-bandgap materials Eq. (8) above seems to correctly predict the pair formation energy. This confirms the applicability of the approach for measurements of the mean electron-hole pair creation energy. For the sake of convenience it is generally agreed to present the EBIC signal in the form of EBIC current I_c normalized by the product of $E_b \times I_b$, $I_c / (E_b \times I_b)$. Since at zero bias the collected current for materials with low mobilities of one or both types of carriers (as is the case for Ga_2O_3 with low hole mobility) can be seriously affected by trapping at interface states, the normalized EBIC current is usually measured at high applied voltages to mitigate this trapping. This is done below for Ga_2O_3 Schottky diodes.

We turn now to the measurements of E_i in $\beta\text{-Ga}_2\text{O}_3$. In the course of our previous and current work, we have looked at more than 20 epitaxial films of $\beta\text{-Ga}_2\text{O}_3$ deposited by Halide Vapor Phase Epitaxy (HVPE) on bulk $\beta\text{-Ga}_2\text{O}_3$ substrates prepared by Edge-defined Film-fed Growth (EFG). The samples were acquired from Tamura/Novel Crystals Ltd company in Japan, the main commercial manufacturer of high-quality $\beta\text{-Ga}_2\text{O}_3$ bulk and epitaxial material widely used to fabricate high-power devices based on $\beta\text{-Ga}_2\text{O}_3$ [24, 25]. All these films were doped to n-type by Si and had net donor concentration between $10^{15} - 10^{17} \text{ cm}^{-3}$. The orientation of the films was (001), the thickness was $\sim 10 \mu\text{m}$. They were prepared on $\beta\text{-Ga}_2\text{O}_3$ substrates heavily doped with Sn to donor concentration of $3 \times 10^{18} \text{ cm}^{-3}$ cut from bulk crystals prepared by EFG. The Schottky diodes were made by e-beam evaporation of 20 nm of Ni. The back Ohmic contacts to the substrate side were made by e-beam evaporation of Ti/Au (20nm/80 nm) [4, 5]. Some of the samples were additionally subjected to irradiation with fast reactor neutrons [5], with 10 MeV or 20 MeV protons and 20 MeV a-particles [13, 26-28], and some subjected to treatment in dense Ar plasmas [29]. EBIC measurements were performed as described in Ref. [4, 5, 13]. The samples were also characterized by current-voltage (I-V) measurements in the dark and under monochromatic light illumination

(wavelength range 259-940 nm), capacitance-voltage (C-V) profiling in the dark and under illumination (CV or LCV profiling [27, 30]), and by Deep Level Transient Spectroscopy with electrical (DLTS) or optical (ODLTS) injection. Experimental details can be found in our earlier papers [4, 5, 13, 26-29].

All EBIC measurements results fell into two unequal groups. For group I, the normalized EBIC signal $I_c/(E_b \times I_b)$ was independent of the applied bias for beam energies such that they produced electron-hole pairs well within the space charge region (SCR). This is the kind of behavior observed for all previously studied semiconductor materials and predicted by Eq. (1-7). It is simply the consequence of complete charge collection inside the SCR. Hence, once the energy deposited by the electron beam inside the SCR is calculated taking into account the energy losses due to the absorption in the metal contact and the backscattering of the incident electrons, the electron-hole energy formation E_i can be calculated using the measured value of the normalized EBIC signal and Eq. (7). Such dependence is presented in Fig. 2 for the beam energy 4 keV for one of the studied β -Ga₂O₃ samples that had net donor concentration $1.3 \times 10^{16} \text{ cm}^{-3}$. As shown in Ref. [5], under these conditions, the charge carriers are generated at the depth lower than 50 nm, taking into account the energy absorption in the top Ni layer. The SCR width at 0 V was close to 0.3 μm . Experimental results are shown for this sample by solid blue squares. The data follows the behavior predicted by Eq.(1-7). The initial slight increase of the signal with applied bias is most likely due to the charge collection loss near the interface with the Schottky diode metal because of the hole trapping by the surface states. For higher biases, this effect is suppressed and the normalized EBIC signal versus bias plateau observed can be used to extract the e-h formation energy from Eq. (7) as $E_i=15.6 \text{ eV}$, close to the one predicted by the empirical expression (8) describing the trends for many other semiconductors (corresponding value for Ga₂O₃ derived from the data in Fig. 2 is shown in Fig. 1).

Unfortunately, the samples belonging to group I are in the minority among the Ga_2O_3 samples we have measured. Only two samples showed such a behavior. In both cases, the E_i values were very close to 15.6 eV. For the majority of the samples, the bias dependence of the normalized EBIC signal for low beam energies was quite strong, as shown for one of such group II samples by olive squares in Fig. 2. These results were obtained for a sample with net donor density $5 \times 10^{15} \text{ cm}^{-3}$ under excitation with the same low beam energy of 4 keV creating charge carriers at a depth 50 nm from the interface, while the SCR width at 0V for the sample was $\sim 0.5 \mu\text{m}$. Obviously, for this second group of samples, the EBIC data indicates the presence of some multiplication mechanism resulting in the External Quantum Efficiency (EQE) much higher than unity. The existence of this internal gain mechanism has been observed for as-grown HVPE films [4], films subjected to fast reactor neutron irradiation [5], HVPE epilayers subjected to proton irradiation [26-28] or treatment in Ar plasmas [29]. In all cases, we observed a similarly high gain in the photocurrent as in EBIC for Schottky diodes illuminated with above-band-gap light (for some as-grown samples this has been demonstrated in Ref. [4], for neutron irradiated samples it was shown in Ref. [5], for proton irradiated samples and Ar plasma treated samples we have also shown recently this to be the case, the actual results will be published separately).

The existence of gain mechanisms has been widely invoked to explain the apparently high EQE of photocurrent in Ga_2O_3 and variously attributed to impact ionization [1-3], the operation of polaronic Self-Trapped Hole (STH) states [31] or to trapping by deep hole traps [3-5]. We can definitely rule out in our case the contribution of the impact ionization simply because the electric field strength was very far from the expected threshold of impact ionization near 5-8 MV/cm [32]. The reasons we believe the STH impact cannot be a major factor in the observed phenomena at room temperature is that EBIC results obtained for β - Ga_2O_3 Schottky diodes cannot be explained without assuming that, at room temperature and

above, holes in this material are mobile [4]. As for the participation of deep hole traps, we have shown in Ref. [4, 5] that the amount of increase in EQE definitely correlates with the increased concentration of deep hole traps with optical ionization threshold near 2.3 eV and 3.1 eV, often attributed to Ga vacancy acceptors, V_{Ga} , or V_{Ga} complexes with interstitial Ga, V_{Gai} [33, 34]. Recently we have shown that such correlation also holds for samples irradiated with 20 MeV protons or subjected to Ar plasma treatment (the results are to be published separately). In all these cases, the holes trapped by the deep acceptors change the space charge density in the part of the SCR, thus increasing the electric field strength and decreasing the effective Schottky barrier height and enhancing the electron current flow through the diode [3, 5]. The photocurrent and EBIC current then consists in Ga_2O_3 Schottky diodes of the “normal” part common for all semiconductor materials and the “gain” part $J_{dark}[\exp(\Delta V_{bi}/k_B T)-1]$ [3], where J_{dark} is the dark current, k_B is the Boltzmann constant, T is temperature, and ΔV_{bi} is the change of the Schottky barrier height due to trapping of holes on deep acceptors). In Ref. [5] we show that the change of the Schottky barrier height is closely related to the change in the density of deep traps N_{deep} as $\Delta V_{bi}=qN_{deep}w_0^2/(2\epsilon\epsilon_0)$, where q is the electronic charge, w_0 is the thickness of the layer where the deep hole traps are recharged by light or electron beam, ϵ_0 is the dielectric constant, and ϵ is the relative permittivity. The distinguishing feature of the group I samples that separates them from the group II samples with the high current gain is the much lower density of the deep acceptor hole traps with optical threshold 3.1 eV related to V_{Ga} and of traps with optical threshold 2.3 eV related to V_{Gai} . For the two samples in Fig. 2, LCV spectra measurements [27-30] give the concentration of the 2.3 eV V_{Ga}^i hole traps as $2\times 10^{14} \text{ cm}^{-3}$ for the sample from group I and $7\times 10^{14} \text{ cm}^{-3}$ for the sample from group II in which we also detected $2.5\times 10^{14} \text{ cm}^{-3}$ of the V_{Ga} acceptors.

Nevertheless, the concentration of the deep hole traps in the group I sample is not totally negligible and the current “gain” contribution to EBIC is still present, albeit its magnitude is small for very low beam energies with very small thickness of the w_o region where the traps can be recharged by the electron beam (the upper limit of w_o is 50 nm for $E_b=4$ keV). The impact of these hole traps, can, however, become much more pronounced for higher beam energies recharging the hole traps in the thicker w_o region. In Fig. 3 we compare the results of EBIC signal measurements as a function of bias for the group I sample from Fig. 2 when the beam energy is increased to 25 keV. In that case, the electrons range will be close to 2.4 μm [13], so that up to ~ 25 V applied bias, a considerable part of the electron-hole pairs will be created outside the space charge region and collected due to the holes diffusion to the SCR boundary with the diffusion length close to 0.6 μm . Thus, for biases lower than 25 V, the collection efficiency will be much lower than that observed in the case of $E_b=4$ keV [13], but the contribution of the energy losses in the metal will be much lower than in the $E_b=4$ keV case. Hence, without the current “gain” term, the normalized EBIC signal should look as shown by the open diamonds in Fig. 3. It can be seen, however, that the actual measurements go much higher than the calculated curve. This is because of the enhanced contribution of the “gain” term in EBIC current. Indeed, the upper limit of the w_o value in the ΔV_{bi} expression above is increased by about 50 times compared to the 4 keV case (2.4 μm versus 0.05 μm), which, for the same concentration of the deep acceptors, will produce an enormous increase in gain (these are only crude estimates not taking into account the diffusion of holes towards the Schottky diode). Thus, even for group I samples, the estimated E_i value contains a small contribution from the “gain” term.

Taking into account that any losses of collected current inside the depletion region lead to the increase of calculated E_i value, the obtained value should be considered as the upper limit of E_i . Also, the presence of even a small amount of deep hole traps could produce some

small error in the E_i estimate. This is because the presence of the gain due to the hole trapping on deep acceptors will not be cancelled out, even if the E_i is determined from comparison with the signal from the well calibrated detectors, because in Schottky diodes, there still will be a contribution coming from the hole trapping and effective Schottky barrier decrease while in photoconductors this hole trapping will give rise to additional enhancement of photocurrent because of the increased lifetime of electrons.

The reason why the concentration of deep traps can strongly vary from sample to sample needs better understanding. The increase in the level of donor doping from $\sim 10^{16} \text{ cm}^{-3}$ to 10^{17} cm^{-3} seems to somewhat increase the concentration of deep acceptors [5], but the data has been collected for very few samples with larger donor density (most of the samples we studied had net donor concentration closer to $(1-3) \times 10^{16} \text{ cm}^{-3}$ often used for high-power rectifiers work). Since the deep acceptors responsible seem to be related to Ga vacancies or their complexes, one would expect that changing the VI/III flows ratio in HVPE should have a more pronounced effect than varying the donor density in a rather narrow range, but no systematic studies of that sort can be performed on commercially available samples. The E_i measurements performed on the samples with the lowest possible deep acceptors concentrations is at the moment the best we can do in order to estimate the E_i value in β - Ga_2O_3 . The E_i value obtained above is about 10% larger than the 14.2 eV predicted by the expression (8). The expressions proposed in References [21, 22] give values of 13.3 and 13.79 eV, respectively. There is some uncertainty in the value of the bandgap of β - Ga_2O_3 that is differently quoted as ranging from 4.7 eV to 4.9 eV.

Nevertheless, our estimated value differs from the smallest value of 13.3 eV by 15% only. Taking into account that the state-of-the-art β - Ga_2O_3 Schottky diodes due to reasons discussed above do not allow to obtain the E_i value more precisely, the value obtained in the present work from the EBIC measurements or those calculated using the empirical

expressions from [20-22] can be used for the mean electron-hole pair creation energy in β - Ga_2O_3 for the prediction of charged particle detector parameters and other cases where the knowledge of E_i value is necessary.

To conclude, the electron-hole pair formation in Ga_2O_3 indeed can be reasonably accurately determined by fitting the normalized EBIC signal collection efficiency, but these measurements have to be performed at low beam energies when the excitation depth is much lower than the space charge region width. In addition, it is important to check the independence of the collected current on bias and that the density of deep hole traps is low to avoid photoconductive gain effects.

ACKNOWLEDGMENTS

The work at NUST MISiS was supported in part by Grant № K2-2020-040 under the Program to increase Competitiveness of NUST MISiS among the World Leading Scientific and Educational centers (Program funded by the Russian Ministry of Science and Education). The work at IMT RAS was supported in part by the State Task No 075-00920-20-00. The work at UF was sponsored by Department of the Defense, Defense Threat Reduction Agency, part of Interaction of Ionizing Radiation with Matter University Research Alliance (IIRM-URA) under award HDTRA1-20-2-0002 (Jacob Calkins) and also by NSF DMR 1856662 (James Edgar).

Data Availability Statement

Data available in article. The data that supports the findings of this study are available within the article

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Figure Captions

Figure 1. Experimental values of E_i from [17, 20] (olive symbols) and obtained in the present work (blue symbols) Lines present the empirical expressions from [20] (olive line), [21] (magenta line), [22] (wine line) and [23] (violet line)

Fig. 2. Normalized collected current as a function of applied reverse bias for two Schottky diodes on $\beta\text{-Ga}_2\text{O}_3$ measured at $E_b = 4$ keV.

Fig. 3. Normalized collected current as a function of applied reverse bias measured at beam energy 4 keV with beam current of 7 pA (blue squares) and 100 pA (olive squares) and at beam energy 25 keV and beam current 10 pA (solid wine squares); also shown is the calculated bias dependence of the normalized EBIC signal without taking into account the current gain term (open diamonds).

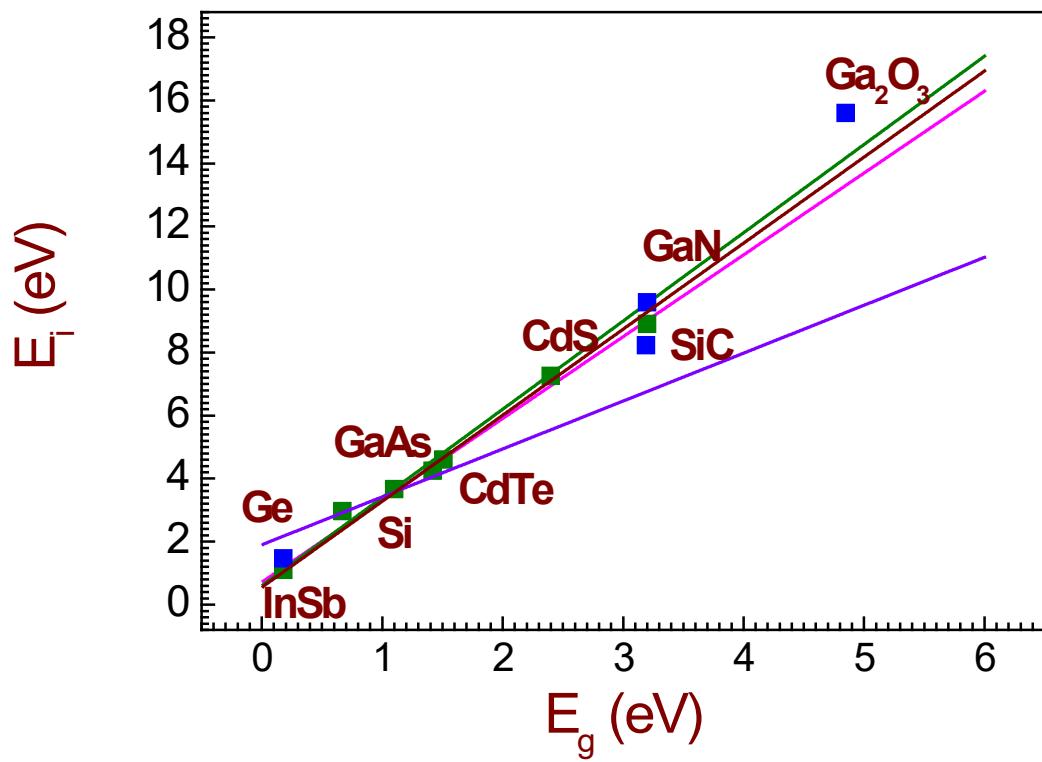


Fig 1.

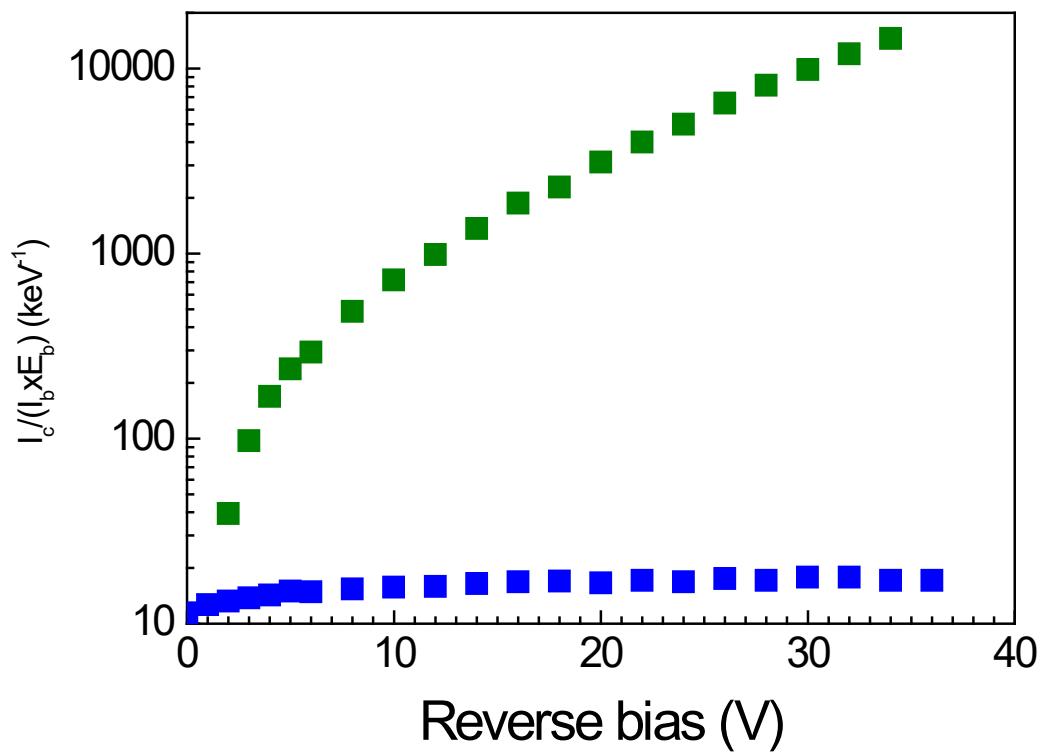


Fig 2.

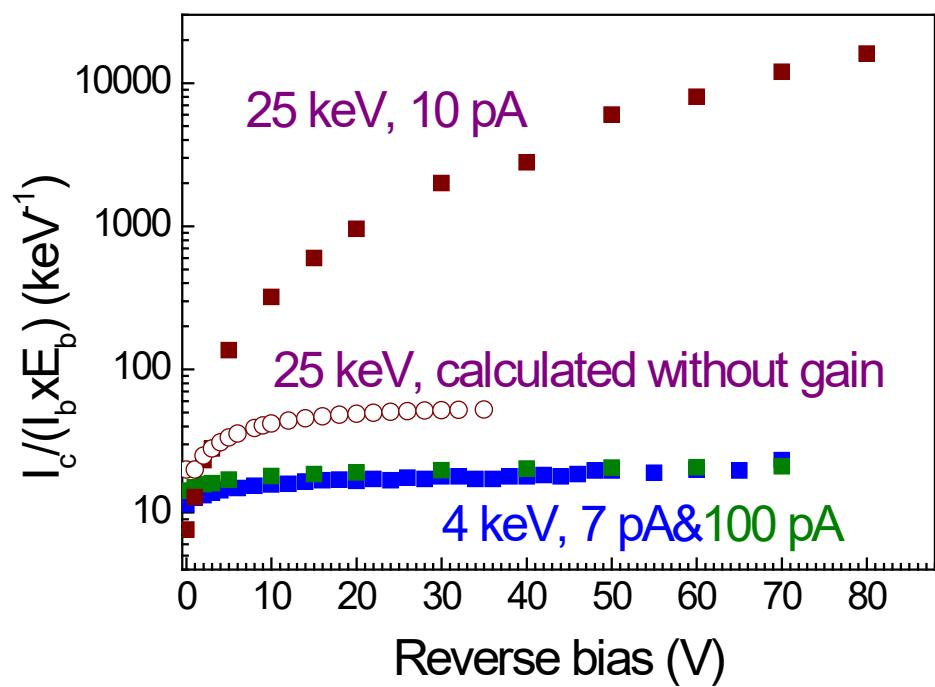


Fig 3.