High temperature (≤ 160 °C) X-ray and β - particle diamond detector

Article (Accepted Version)

Lioliou, G, Lefeuvre, G and Barnett, A M (2021) High temperature (≤ 160 °C) X-ray and β - particle diamond detector. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors, and Associated Equipment, 991. a165025 1-7. ISSN 0168-9002

This version is available from Sussex Research Online: <http://sro.sussex.ac.uk/id/eprint/96364/>

This document is made available in accordance with publisher policies and may differ from the published version or from the version of record. If you wish to cite this item you are advised to consult the publisher's version. Please see the URL above for details on accessing the published version.

Copyright and reuse:

Sussex Research Online is a digital repository of the research output of the University.

Copyright and all moral rights to the version of the paper presented here belong to the individual author(s) and/or other copyright owners. To the extent reasonable and practicable, the material made available in SRO has been checked for eligibility before being made available.

Copies of full text items generally can be reproduced, displayed or performed and given to third parties in any format or medium for personal research or study, educational, or not-for-profit purposes without prior permission or charge, provided that the authors, title and full bibliographic details are credited, a hyperlink and/or URL is given for the original metadata page and the content is not changed in any way.

High temperature (≤ 160 °C) X-ray and β^- particle diamond detector

G. Lioliou^{1*}, G. Lefeuvre², and A.M. Barnett¹

¹Space Research Group, School of Mathematical and Physical Sciences, University of Sussex, Falmer, Brighton, BN1 9QT, UK

²Micron Semiconductor Ltd., 1 Royal Buildings, Marlborough Road, Lancing Business Park, Lancing, BN15 8SJ, UK.

Abstract

A single crystal CVD diamond detector (110 μm thick) with Al contacts (11.34 mm^2) was characterised as an X-ray and β^- particle detector using an ^{55}Fe radioisotope X-ray source (Mn $K\alpha = 5.9$ keV, Mn $K\beta = 6.49$ keV) and a ^{63}Ni radioisotope β^- particle source (66 keV endpoint energy). The characterisation was performed across the temperature range 160 °C to -20 °C and at applied potential differences from -400 V to +400 V (3.6 $\text{V } \mu\text{m}^{-1}$ applied electric field strength). X-ray photocurrent and β^- particle current measurements along with theoretical calculations demonstrated that the detector could detect both types of radiation with similar apparent charge collection efficiency.

Keywords: diamond detector; current mode; X-ray photocurrent; β^- particle current; charge collection efficiency.

1. Introduction

Radiation detectors find use in a wide variety of applications ranging from space science [1] and synchrotron applications [2], to medicine [3] and the nuclear industry [4].

Semiconductor radiation detectors have been widely used, and are still being developed to optimise the trade-offs of the different requirements of the various applications. Among the different requirements, high temperature and intense radiation tolerance play important roles when selecting detector materials for harsh environments. Wide bandgap semiconductor radiation detectors which have been investigated for use in such environments include GaAs [5-8], SiC [9-11], GaN [12-13], AlGaAs [14-15], AlInP [16-18], InGaP [19-20], CdTe and CdZnTe [21-22], HgI₂ [23], ThBr [24], and diamond.

Diamond has many attributes which make it a good potential candidate material for extreme environment radiation detectors. Diamond's wide bandgap (5.45 eV [25]) results in low leakage currents and the ability to operate at high temperatures [26]. Also, compared to traditional Si radiation detectors, diamond has better radiation resistance [27] and higher carrier velocities [28-29]. Whilst natural diamond has inherent variability and relative scarcity, synthetic diamond can be grown by Chemical Vapor Deposition (CVD) [30].

CVD diamond detectors have already been shown to be spectroscopic to α particles. An energy resolution of 0.4 % for 5.486 MeV α particles from an ^{241}Am source was reported for a single crystal CVD diamond detector [31] [32]. CVD diamond detectors have been studied for their charge collection efficiencies, at temperatures up to 300 °C, using α particles from an ^{241}Am radioisotope source; charge collection efficiencies of 98.2 % for holes and 96.7 % for electrons were measured in that case at 250 °C [33].

CVD diamond detectors have been investigated in current mode (cf. particle or photon counting mode) under the illumination of both β^- particles and X-ray photons. However, they have not been shown to be spectroscopic to β^- particles and X-ray photons. A single crystal CVD diamond detector operated in current mode under the illumination of a 1 MBq $^{90}\text{Sr } \beta^-$

*Corresponding author. Tel.: +44 (0) 1273 872568. E-mail address: G.Lioliou@sussex.ac.uk

particle source (end point energy = 2.28 MeV for a $^{90}\text{Sr}/^{90}\text{Y}$ source [34]) has been demonstrated at temperatures up to 180 °C [35]. A diamond photodiode was characterised under the illumination of soft X-rays (0.25 keV to 7 keV) at room temperature [36]; the electron-hole pair creation energy was reported to be $13.25 \text{ eV} \pm 0.5 \text{ eV}$. The effect of different amorphous carbon electrodes on CVD diamond detectors has been studied [37]; whilst the devices showed low leakage currents (bulk resistivity of at least $2.8 \times 10^{14} \Omega \text{ cm}$ at applied electric field strengths of $\pm 0.3 \text{ V } \mu\text{m}^{-1}$), a reported drawback was the instability of the photocurrent generated from illumination of a Mo target X-ray tube, at high applied potential differences [37]. In other work, a single crystal CVD diamond detector was characterised under the illumination of 10.6 keV and 12.5 keV X-rays; a linear response as a function of photon flux was measured [38]. Single crystal CVD diamond detectors have also been investigated when illuminated using a Mo target X-ray tube; an approximately linear relationship between the measured current and the X-ray dose rate was reported [39].

Here, a single crystal CVD diamond detector with an area of $4.4 \text{ mm} \times 4.4 \text{ mm}$ ($= 19.36 \text{ mm}^2$), thickness of $110 \mu\text{m} \pm 1 \mu\text{m}$, and Al Ohmic contacts (3.8 mm diameter, corresponding to an area of 11.34 mm^2 , thus covering 59 % of the top and bottom faces of the detector) was investigated for the first time for its performance as radiation detector in current mode under the illumination of X-ray photons and β^- particles, in turn, at temperatures up to 160 °C. The detector was illuminated with 5.9 keV and 6.49 keV X-rays from an ^{55}Fe radioisotope X-ray source and β^- particles from a ^{63}Ni radioisotope β^- particle source. The detector's characteristics were investigated as a function of applied potential difference (from -400 V to +400 V, corresponding to an applied electric field strength of $3.6 \text{ V } \mu\text{m}^{-1}$) and across the temperature range 160 °C to -20 °C. The X-ray photocurrent measurements were accompanied by calculations of the X-ray photocurrent. The Monte Carlo electron (β^- particle) simulation package CASINO [40-41] was used to predict the current from illumination with the ^{63}Ni radioisotope β^- particle source. The apparent charge collection efficiency, *CCE*, (i.e. the ratio of the measured current and the predicted generated current from the radiation absorption [42]) was calculated for both types of radiation.

2. Diamond detector structure

The detector was fabricated at Micron Semiconductor Ltd, UK, using single crystal CVD diamond from Element Six Ltd, UK. During detector processing, a standard acid-clean treatment was used, followed by a proprietary cleaning step to remove any remaining surface impurities. Circular Al contacts were sputtered onto the detector's front and rear faces, to form the front and rear contacts. Since the Al contacts did not cover the whole area of the CVD diamond detector, the active volume of the detector was considered to be approximated to the cylinder of diamond material between the Al contacts. The detector was mounted and wire bonded in a custom package with SMA connections to the front and rear contacts.

3. Results and discussion

3.1. Dark current measurements of diamond detector

The dark current of the detector was measured as a function of applied potential difference and across the temperature range 160 °C to -20 °C. The detector was installed inside a light-tight die-cast Al enclosure; the enclosure was then placed inside a TAS Micro MT climatic cabinet for temperature control. The temperature was initially set to 160 °C and decreased to -20 °C, in 20 °C steps. The detector was left for 30 minutes at each temperature, prior to

initiation of the current measurements, to ensure thermal equilibrium. The environment inside the climatic cabinet was maintained dry ($< 5\%$ relative humidity) with dry N_2 flowed continually into the climatic cabinet, to mitigate any humidity related effects.

The dark current as a function of applied potential difference was measured using a Keithley 6487 Picoammeter/Voltage Source. The maximum applied potential difference was limited to ± 400 V ($3.6\text{ V }\mu\text{m}^{-1}$ applied electric field strength). The potential difference was applied at the front Al contact, and the current was measured through the rear Al contact. The uncertainty associated with the current measurements was 0.3% of the measured value plus 400 fA [43]. The same procedure, applied potential difference (0 V to 400 V), and temperature range as for the dark current measurements of the package detector was followed for an identical empty package, to separate the leakage current of the diamond detector from other parasitic leakage currents (arising from the package and the cables of the measurement set up). The leakage current of the empty package as a function of temperature, at the maximum investigated applied potential difference (400 V), can be seen in **Figure 1**. It reduced from $620\text{ pA} \pm 50\text{ pA}$ at $160\text{ }^\circ\text{C}$ to $0.6\text{ pA} \pm 0.4\text{ pA}$ at $80\text{ }^\circ\text{C}$ at 400 V applied potential difference, whereas it was below the noise floor of the Keithley 6487 Picoammeter/Voltage Source ($\pm 400\text{ fA}$) at temperatures $\leq 60\text{ }^\circ\text{C}$ at all investigated applied potential differences.

Fig. 1. Leakage current of the empty package as a function of temperature at 400 V applied potential difference.

The leakage current of the CVD diamond detector (with the leakage current of the empty package subtracted) as a function of applied potential difference within the investigated temperature range is presented in **Figure 2 (a)**. Additionally, the Arrhenius plot, the logarithm of the current, $\ln(I)$, as a function of T^{-1} , at $+400\text{ V}$ applied potential difference, was calculated and is presented in **Figure 2 (b)**. The leakage current of the package detector at applied potential differences between 0 V and -400 V was the same, within uncertainties, with the leakage current of the empty package, thus the leakage current of the diamond detector at applied potential differences between 0 V and -400 V was below the noise floor of the Keithley 6487 Picoammeter/Voltage Source ($\pm 400\text{ fA}$), within the investigated temperature range. The detector did not exhibit a symmetrical (with respect to potential difference) leakage current as a function of applied potential difference. This indicated that the front and rear Al/diamond junctions had different electrical behaviour.

Fig. 2. (a) Leakage current of the CVD diamond detector as a function of applied potential difference within the temperature range 160 °C to -20 °C; 160 °C (filled circles), 140 °C (open diamonds), 120 °C (filled squares), 100 °C (stars), 80 °C (open circles), 60 °C (dashes), 40 °C (open squares), 20 °C (filled triangles), 0 °C (crosses), and -20 °C (filled diamonds); (b) The measured $\ln(I)$ at +400 V applied potential difference, as a function of T^{-1} (Arrhenius plot). The error bars of the leakage current are omitted for clarity, whereas the error bars of the measured $\ln(I)$ were smaller than the symbol size for some data points .

The maximum measured leakage current of the diamond detector under positive applied potential difference was $70 \text{ pA} \pm 50 \text{ pA}$ ($0.7 \text{ nA cm}^{-2} \pm 0.4 \text{ nA cm}^{-2}$ dark current density), at 160 °C and +400 V applied potential difference. This value is lower compared to previous reports on other single crystal CVD diamond detectors at high temperatures. As an example, a $\sim 6 \text{ nA cm}^{-2}$ leakage current density was reported for a single crystal CVD diamond detector when operating at 150 °C and 120 °C, under the application of $1 \text{ V } \mu\text{m}^{-1}$ electric field [35]. When no external potential difference was applied, the current measured with the present system was $0.8 \text{ pA} \pm 0.4 \text{ pA}$ (rms deviance) (equivalent to $7 \text{ pA cm}^{-2} \pm 4 \text{ pA cm}^{-2}$ dark current density) across the investigated temperature range 160 °C to -20 °C. The leakage current density of the presently reported CVD diamond detector was found to remain $\leq 7 \text{ pA cm}^{-2} \pm 4 \text{ pA cm}^{-2}$ at 20 °C, across the investigated potential difference range, -400 V to +400 V. For comparison purposes, 4H-SiC Schottky diodes of the highest quality can have a leakage current density of $\sim 0.2 \text{ pA cm}^{-2}$ at the same temperature and applied electric field strength [44].

The dependence of the current, I , on the temperature, T , at a given applied potential difference is a function of the activation energy, E_A ,

$$I = AT^2 \exp\left(-\frac{E_A}{kT}\right), \quad (1)$$

where A is a constant and k is the Boltzmann constant [45]. Thus, a plot of the logarithm of the current, $\ln(I)$, as a function of T^{-1} , would be expected to yield a straight line whose gradient can be used to calculate the activation energy, E_A . The measured $\ln(I)$ at +400 V applied potential difference, as a function of T^{-1} (Arrhenius plot) can be seen in **Figure 2 (b)**. The line of best fit was calculated using linear least squares fitting in the temperature range 160 °C to 20 °C; the measured $\ln(I)$ for $< 20 \text{ °C}$ was within the uncertainty of the measuring system, and thus excluded from the analysis. The activation energy was found to be $0.38 \text{ eV} \pm 0.02 \text{ eV}$. This energy level for the activation energy may be associated with defects of dopant-hydrogen complex type; such defects can have an energy level of 0.3 eV for nitrogen dopants and 0.4 eV for boron dopants [46]. Those reported values (which had no uncertainties stated) are close to this calculated here.

3.2. ^{55}Fe radioisotope X-ray source illumination

3.2.1. X-ray quantum detection efficiency

X-ray photocurrent measurements were performed with an ^{55}Fe radioisotope X-ray source giving Mn K α (5.9 keV) and Mn K β (6.49 keV) emission lines [47]. The X-ray quantum detection efficiency, QE , of the detector was calculated for photons of energy up to 30 keV, accounting for the attenuation of X-rays at the front Al contact (300 nm thick with an area of 11.34 mm²), considering absorption in the thickness of the detector (110 μm), and assuming complete charge collection. The X-ray attenuation coefficients of Al and the X-ray absorption coefficients of diamond were extracted from Hubbell [48]. The quantum detection efficiency of the detector as a function of X-ray photon energy can be seen in **Figure 3**. The X-ray quantum efficiency of the detector was calculated to be 0.35 at 5.9 keV and 0.27 at 6.49 keV.

Fig. 3. Calculated X-ray quantum detection efficiency, QE , of the detector assuming complete charge collection.

3.2.2. ^{55}Fe radioisotope X-ray source photocurrent measurement

The total current (leakage current and photocurrent) with the ^{55}Fe radioisotope X-ray source placed 8.2 mm above the CVD diamond detector was measured; the same procedure as for the dark current measurements was followed (see **3.1. Dark current measurements of diamond detector**). The ^{55}Fe radioisotope X-ray source had an activity of 159 MBq. The disc source had a diameter of 6 mm and was encapsulated: the window of the capsule was a 0.25 mm thick Be layer. The ^{55}Fe X-ray photocurrent, with the dark (leakage) current subtracted from the total current, can be seen in **Figure 4**.

Fig. 4. ^{55}Fe X-ray photocurrent measured with the detector illuminated with the ^{55}Fe radioisotope X-ray source as a function of applied potential difference within the temperature

range 160 °C to -20 °C; 160 °C (filled circles), 140 °C (open diamonds), 120 °C (filled squares), 100 °C (stars), 80 °C (open circles), 60 °C (dashes), 40 °C (open squares), 20 °C (filled triangles), 0 °C (crosses), and -20 °C (filled diamonds).

The photocurrent at each temperature was found to be a function of the applied potential difference; it was found to increase with increased applied potential difference magnitude. As an example, the photocurrent measured at 20 °C increased from 3.1 pA ± 0.6 pA at 0 V, to 29.8 pA ± 0.6 pA at +400 V and to 41.3 pA ± 0.7 pA at -400 V. The increase of the photocurrent with increased applied potential difference may be attributable to a reduction in charge trapping with increased electric fields leading to improved charge transport and increased CCE, if the CCE was not 100 % at the lower temperatures [49]. Also, improved collection of electron-hole pairs generated outside of the cylinder defined by the contacts from the absorption of X-rays, as a consequence of improved carrier transport properties, could explain the photocurrent increase with increased electric field.

The photocurrent, at a given applied potential difference, was found to vary among different temperatures (see **Figure 4**). This apparent variation could be partially attributed to the increased effective diffusion lengths of minority carriers with increasing temperature (due to changes in recombination and trapping rates with temperature) [50] and/or to the reduced electron-hole pair creation energy with increasing temperature [51] [52]. However, the variation of the X-ray photocurrent was more prominent at high temperatures and high (in magnitude) applied potential differences. The rms error (root mean square of the deviations from the mean) of the photocurrent within the investigated temperature range was calculated at all applied potential differences; the data point at -400 V applied potential difference and 60 °C was considered an outlier resulting from total current instabilities and was thus excluded from the rms error calculations. The rms error increased from 10 % at no applied potential difference to 68 % at +400 V applied potential difference. Since the measured leakage current was subtracted from the total current to result in the photocurrent, potential leakage current instabilities at high temperatures and/or high (in magnitude) applied potential differences may have resulted in increased uncertainties in the photocurrent calculation, making the observed variation of the photocurrent with temperature unreal.

3.2.3. ⁵⁵Fe radioisotope X-ray source photocurrent calculation

The photocurrent at 20 °C under the illumination of the ⁵⁵Fe radioisotope X-ray source was calculated. The generated photocurrent, I_{ph} , from the absorption of the 5.9 keV and 6.49 keV X-ray photons within the CVD diamond detector (110 µm thick with an active area of 11.34 mm² assuming the active area was confined to that encompassed by the contacts) depended on the quantum detection efficiency, QE , and the total X-ray power, P , incident on the front face of the detector (accounting for the attenuation of X-rays within the Be window of the capsule of the source) at each corresponding energy, E_i , assuming complete charge collection,

$$I_{ph} = \sum_i QE_i * P_i * \frac{E_i}{E_{ehp}} * q. \quad (2)$$

In Eq. (2), q is the charge of an electron and E_{ehp} is the average energy needed for the creation of an electron-hole pair in diamond (= 13.2 eV [52]). Summing the photocurrent generated from the absorption of the 5.9 keV and 6.49 keV X-ray photons, a photocurrent of 196 pA was calculated, accounting for photocurrent generation within the cylinder of diamond material between the Al contacts. A comparison between the measured at 20 °C and the

predicted X-ray photocurrent as a function of applied potential difference can be seen in **Figure 5**.

Fig. 5. Comparison between the measured at 20 °C (filled triangles) and calculated (dashed line) ⁵⁵Fe X-ray photocurrent as a function of potential difference.

The measured photocurrent at all applied potential differences was lower than the calculated photocurrent. The discrepancy between the measured and the calculated ⁵⁵Fe X-ray photocurrent was attributed to incomplete charge collection. The measured photocurrent at 0 V was 2 % of the calculated ⁵⁵Fe X-ray photocurrent, whereas the apparent *CCE* increased from this value to 15 % and 21 % at +400 V and -400 V applied potential difference (3.6 V μm⁻¹ electric field strength), respectively.

Such *CCE* are lower than some previously reported CVD diamond detectors, e.g. a *CCE* of 98.2 % for holes and 96.7 % for electrons (at ~ 1 V μm⁻¹ and 250 °C) using α particles [33]. The incomplete charge collection of the currently presented diamond detector could be due to poorer electron and hole transport in this particular material. However, a fixed space charge that locally compensated the applied electric field could also explain the apparent limited *CCE*, similarly to a previous report on a diamond detector with a *CCE* of 30 % at 3 V μm⁻¹ (using α particles) which was created during priming [53]. Evidence of such a fixed space charge region is the non-zero *CCE* at 0 V applied potential difference. The photocurrent flowing between the electrodes (Ohmic contacts) of a semiconductor detector is directly proportional to the applied potential difference, in the absence of a built in potential (which is the case for the reported diamond detector) [54], thus a zero *CCE* was expected when no potential difference was applied to the detector. The presence of a fixed space charge region would create an internal electric field, even at zero applied potential difference, accounting for the non-zero *CCE*. Polarisation, the creation of space charge towards the contacts leading to a modified electric field distribution in the detector with a local minimum in the central region, is unlikely to be responsible for the relatively low apparent *CCE*, since polarisation is less prominent with soft X-rays and β⁻ particles cf. alpha particles [55] and hysteresis in the current as a function of voltage characteristics was not observed for the reported detector [56]. However, further measurements with variable photon flux would be required to explore this hypothesis [57]. Despite the nuances discovered, the results demonstrate that the detector can be used to detect X-rays at ⁵⁵Fe energies (5.9 keV and 6.49 keV) in current mode at temperatures up to 160 °C.

3.3. ⁶³Ni radioisotope β⁻ particle source illumination

3.3.1. Percentage of β⁻ particle energy absorbed

Initially, the percentage of β^- particle energy absorbed within the CVD diamond detector, as a function of β^- particle energies (1 keV to 66 keV), was calculated using CASINO simulations. A total of 4000 β^- particles were simulated in CASINO at each energy (1 keV to 66 keV, in 1 keV steps) to be incident on the detector's face. The simulated detector geometry included the front Al contact (300 nm thick) and the active volume of the detector (assuming that the active volume of the detector was limited to the cylinder defined by the contacts). The computed trajectories of the simulated β^- particles were used to calculate the energy deposited in the detector from each of them. The ratio of the total β^- particle energy deposited in the detector by the total energy incident on the detector's face as a function of β^- particle energy was calculated. This resulted in calculation of the percentage of β^- particle energy absorbed in the detector and it can be seen in **Figure 6**.

Fig. 6. Percentage of β^- particle energy absorbed in the detector (taking into account the front Al contact), as a function of β^- particle energy.

The percentage of β^- particle energy absorbed was found to be 0 % for β^- particle energies < 5 keV, which was attributed to the attenuation of β^- particles in the front Al contact. As the energy was increased, the percentage of β^- particle energy absorbed increased, reaching 90 % at 19 keV, 97 % at 50 keV and remaining at 97 % up to 66 keV. This suggested that the thickness of the CVD diamond detector (110 μ m) was not a limiting factor for the absorption of the β^- particles of the investigated energy range. The 300 nm thick front Al contact was the main limiting factor attenuating the percentage of β^- particle energy absorbed within the detector; without its presence, 96 % and 98 % of β^- particle energy would have been absorbed at 1 keV and 66 keV, respectively. The rear Al contact did not affect the percentage of β^- particle energy absorbed within the detector; all β^- particles, up to 66 keV, were fully absorbed before reaching the rear Al contact.

3.3.2. ^{63}Ni radioisotope β^- particle source current measurement

The total current (leakage current and photocurrent) with a ^{63}Ni radioisotope β^- particle source placed 13 mm above the CVD diamond was measured; the same procedure as for the dark current measurements was followed (see **3.1. Dark current measurements of diamond detector**). The ^{63}Ni radioisotope β^- particle source had an actual activity of 185 MBq. Its apparent activity, due to self-absorption [58], was 136 MBq. ^{63}Ni has an endpoint energy of 66 keV. An inactive Ni overlayer (1 μ m thick) covered the active side of the ^{63}Ni radioisotope β^- particle source having been electroplated on there by the manufacturer to satisfy local laboratory safety protocols; this reduced the energy of the particles emitted from the source and was accounted for during the simulations and measurements. The ^{63}Ni β^- particle current of the CVD diamond detector, with the dark (leakage) current subtracted from the total current can be seen in **Figure 7**.

Similarly to the ^{55}Fe X-ray photocurrent (**Figure 5**), the ^{63}Ni β^- particle current was found to be a function of the applied potential difference; it was found to increase with increased applied potential difference magnitude at a given temperature. In addition, the measured ^{63}Ni β^- particle current was found to vary among different temperatures. This variation was more prominent at high temperatures and high (in magnitude) applied potential differences; the rms error of the β^- particle current at a given applied potential difference within the investigated temperature range increased from 11 % at -50 V applied potential difference to 67 % at -400 V applied potential difference.

Fig. 7. ^{63}Ni β^- particle current of the CVD diamond detector as a function of applied potential difference within the temperature range 160 °C to -20 °C; 160 °C (filled circles), 140 °C (open diamonds), 120 °C (filled squares), 100 °C (stars), 80 °C (open circles), 60 °C (dashes), 40 °C (open squares), 20 °C (filled triangles), 0 °C (crosses), and -20 °C (filled diamonds).

The ^{63}Ni β^- particle current was measured to increase from $5.4 \text{ pA} \pm 0.6 \text{ pA}$ (rms deviance) at 0 V, to $148 \text{ pA} \pm 1 \text{ pA}$ at +400 V and $209 \text{ pA} \pm 1 \text{ pA}$ at -400 V, at 20 °C. The increase of the β^- particle current with increased applied potential difference may be attributed to the improved charge transport with increased electric field, resulting in less charge being trapped. Similarly to the ^{55}Fe X-ray photocurrent, potential leakage current instabilities at high temperatures and/or at high (in magnitude) applied potential differences, could have resulted in increased uncertainties in the β^- particle current calculation at such applied potential differences, making the observed variation of the β^- particle current with temperature (**Figure 7**) unreal.

3.3.3. ^{63}Ni radioisotope β^- particle current calculation

The ^{63}Ni β^- particle spectrum to be incident on the detector was calculated by simulating the spectrum emitted by the ^{63}Ni radioisotope β^- particle source (adjusted for self-absorption) and identifying the β^- particle energy losses between the surface of the ^{63}Ni radioisotope β^- particle source and the front face of the CVD diamond detector (i.e. within the inactive Ni overlayer and air). Different numbers of β^- particles were simulated (in CASINO) for each energy to account for their different emission probability, P_i , from the ^{63}Ni radioisotope β^- particle source including consideration of self-absorption of the source [59]. The trajectories of all β^- particles within the inactive Ni overlayer (thickness of 1 μm and density of 8.908 g cm^{-3}) and the air layer (thickness of 13 mm and density of 0.0012 g cm^{-3}) were computed and were used to calculate the total remaining energy of each β^- particle when it reached the detector, thus taking into account lost energy in the Ni overlayer and air layer. The summation of the distribution of β^- particle energy remaining for each β^- particle simulated resulted in the spectrum predicted to be incident on the CVD diamond detector and it is presented in **Figure 8**.

Fig. 8. Simulated ^{63}Ni β^- particle spectra as emitted from the source (solid line) and as incident on the front face of the detector (dotted line).

Consideration was then given to calculating the spectrum of β^- particles to be usefully absorbed by the detector in order to calculate the current generated due to illumination with the ^{63}Ni radioisotope β^- particle source.

The energy deposited in the CVD diamond detector from the computed trajectories of each β^- particle simulated to be incident on the detector was used in conjunction with the calculations of the percentage of β^- particle energy absorbed in the detector (**Figure 6**) to determine the usefully absorbed energy for each β^- particle emitted from the ^{63}Ni . Given that the electron-hole pair creation energy, E_{ehp} , in single crystal CVD diamond has been reported to be 13.2 eV [52], the current at 20 $^{\circ}\text{C}$ under the illumination of the ^{63}Ni radioisotope β^- particle source was calculated. The β^- particle current generated,

$$I_{\beta^-} = \sum_{i=0}^{66} F_i * \frac{A}{2} * \frac{A_{det}}{A_s} * P_i * \frac{E_i}{E_{ehp}} * q, \quad (3)$$

depended on the fraction, F_i , of the useful β^- particles (defined below and calculated using CASINO simulations) and the number of β^- particles potentially arriving at the front face of the diamond detector, assuming complete charge collection. The number of useful β^- particles was calculated using half of the apparent activity, $A/2$, of the radioactive source (i.e.

that emitted towards the detector), the ratio of the detector's area over the source's area, A_{del}/A_s , and the emission probability, P_i , of each β^- particle [59].

The fraction, F_i , of the useful β^- particles is the ratio of the number of β^- particles absorbed in the detector to the number of emitted β^- particles from the ^{63}Ni , as a function of β^- particle energy 1 keV to 66 keV. This fraction, F_i , accounted for losses in the protective Ni overlayer of the radioactive source, the air between the source and the front of the detector (13 mm), and the percentage of β^- particle energy absorbed in the detector (i.e. losses in the Al contact layer as well as absorption within the diamond itself).

Using Eq. (3), the β^- particle current was calculated to be 911 pA at 20 °C; a comparison between the calculated generated current and the measured ^{63}Ni β^- particle current at 20 °C, as a function of applied potential difference can be seen in **Figure 9**.

Fig. 9. Comparison between the measured at 20 °C (filled triangles) and calculated (dashed line) ^{63}Ni β^- particle generated current as a function of applied potential difference.

The measured β^- particle current at all applied potential differences was lower than the calculated current, I_{β^-} . The discrepancy between the measured and the calculated ^{63}Ni β^- particle current was attributed to incomplete charge collection, i.e. $CCE < 100\%$. The apparent charge collection efficiency at 20 °C was found to increase from 0.6 % at 0 V to 16 % and 23 %, at +400 V and -400 V applied potential difference, respectively. Similarly to the X-ray photocurrent measurements, the non-zero CCE at 0 V applied potential and the relatively low apparent CCE even at the highest investigated applied electric field strengths measured with β^- particles, were partially attributed to the presence of a fixed space charge region reducing the electric field strength, the origin of which is currently unknown. This hypothesis is even stronger for β^- particles than for X-rays; the most probable β^- particles arriving at the detector had an energy ranging from 10 keV to 15 keV (**Figure 8**) which have a penetration depth (intensity of radiation dropped to 1/e of its original value), according to CASINO simulations, of $< 1.1\ \mu\text{m}$. Hence, a high charge density generated close to the front Al contact could result in a strong space charge region [60]. Nevertheless, the results demonstrate that the CVD diamond detector can be used for β^- particle detection at ^{63}Ni energies in current mode at temperatures up to 160 °C

4. Conclusions

A prototype single crystal CVD diamond detector has been characterised for its X-ray and β^- particle detection performance as a function of applied potential difference (from -400 V to +400 V, corresponding to an applied electric field strength of $\pm 3.6\ \text{V}\ \mu\text{m}^{-1}$) within the temperature range 160 °C to -20 °C.

The detector had low but asymmetric (with respect to the polarity of applied potential difference) leakage current; it remained ≤ 400 fA (4 pA cm^{-2} current density) and $\leq 70 \text{ pA} \pm 50 \text{ pA}$ ($0.7 \text{ nA cm}^{-2} \pm 0.4 \text{ nA cm}^{-2}$ current density) under negative and positive applied potential difference, respectively, at temperatures up to 160°C .

The X-ray detection performance of the CVD diamond detector was characterised with an ^{55}Fe radioisotope X-ray source across the same temperature range. The measured X-ray photocurrent increased with increasing (in magnitude) applied potential difference at a given temperature. Reduced charge trapping with increased electric field, due to improved charge transport, and/or improved collection of charge generated outside the cylinder defined by the contacts could potentially explain this increase. The X-ray photocurrent was predicted to be 196 pA if the detector had complete charge collection, assuming that the active volume of the detector was limited to the cylinder defined by the contacts; the apparent *CCE* at 20°C was 15% and 21% at $+400 \text{ V}$ and -400 V applied potential difference, respectively.

The β^- particle detection performance of the CVD diamond detector was characterised with a ^{63}Ni radioisotope β^- particle source across the same temperature range as was used for the other measurements. CASINO Monte Carlo simulations predicted that the percentage of β^- particle energy absorbed within the detector was $> 90\%$ for β^- particle energies $> 19 \text{ keV}$, reaching a value of 97% at 50 keV . The measured β^- particle current was found to increase with increasing (in magnitude) applied potential difference at all investigated temperatures. The β^- particle current at 20°C was predicted from the simulations to be 911 pA in a detector with complete charge collection, assuming that the active volume of the detector was limited to the cylinder defined by the contacts. The apparent *CCE* were determined to be 16% and 23% at $+400 \text{ V}$ and -400 V applied potential difference, respectively.

The results demonstrate that CVD diamond devices can be used for soft X-ray (5.9 keV and 6.49 keV) and soft β^- particle ($< 66 \text{ keV}$) detection (with similar *CCE* for both radiation types) in high temperature environments (up to 160°C). The limited apparent *CCE* was attributed, at least in part, to the presence of a space charge region.

DATA AVAILABILITY

Whilst all data from the study and the findings are contained within the paper, further requests for information may be addressed to the authors.

ACKNOWLEDGEMENTS

A.M.B. acknowledges funding from the Leverhulme Trust, United Kingdom, in the form of a 2016 Philip Leverhulme Prize.

REFERENCES

- [1] M. Bavdaz, A. Peacock, A. Owens, Future space applications of compound semiconductor X-ray detectors. *Nucl. Instrum. Methods Phys. Res. A* **458** (2001) 123-131.
- [2] A. De Sio, E. Pace, G. Cinque, A. Marcelli, J. Achard, A. Tallaire, Diamond Detectors for Synchrotron Radiation X-ray Applications. *Spectrochim. Acta B* **62** (2007) 558-561.
- [3] J. Seco, B. Clasié, M. Partridge, Review on the Characteristics of Radiation Detectors for Dosimetry and Imaging. *Phys. Med. Biol.* **59** (2014) R303-R347.
- [4] P. Bergonzo, A. Brambilla, D. Tromson, C. Mer, B. Guizard, R.D. Marshall, F. Foulon, CVD Diamond for Nuclear Detection Applications. *Nucl. Instrum. Methods Phys. Res. A* **476** (2002) 694-700.

- [5] A. Owens, M. Bavdaz, A. Peacock, A. Poelaert, High Resolution X-Ray Spectroscopy Using GaAs Arrays. *J. Appl. Phys.* **90** (2001) 5376-5381.
- [6] A.M. Barnett, J.E. Lees, D.J. Bassford, J.S. Ng, C.H. Tan, N. Babazadeh, R.B. Gomes, The Spectral Resolution of High Temperature GaAs Photon Counting Soft X-ray Photodiodes. *Nucl. Instrum. Methods Phys. Res. A* **654** (2011) 336-339.
- [7] G. Lioliou, M.D.C. Whitaker, A.M. Barnett, High temperature GaAs X-ray detectors. *J. Appl. Phys.* **122** (2017) 244506.
- [8] G. Lioliou, C.L. Poyser, S. Butera, R.P. Champion, A.J. Kent, A.M. Barnett, 30 μm thick GaAs X-ray $\text{p}^+\text{-i-n}^+$ photodiode grown by MBE. *Nucl. Instrum. Methods Phys. Res. A* **946** (2019) 162670.
- [9] G. Bertuccio, S. Caccia, D. Puglisi, D. Macera, Advances in Silicon Carbide X-ray Detectors. *Nucl. Instrum. Methods Phys. Res. A* **652** (2011) 193-196.
- [10] G. Lioliou, H.K. Chan, T. Gohil, K.V. Vassilevski, N.G. Wright, A.B. Horsfall, A.M. Barnett, 4H-SiC Schottky Diode Arrays for X-ray Detection. *Nucl. Instrum. Methods Phys. Res. A* **840** (2016) 145-152.
- [11] G. Lioliou, N.R. Gemmell, M. Mazzillo, A. Sciuto, A.M. Barnett, 4H-SiC Schottky diodes with Ni_2Si contacts for X-ray detection. *Nucl. Instrum. Methods Phys. Res. A* **940** (2019) 328-336.
- [12] A. Owens, A. Barnes, R.A. Farley, M. Germain, P.J. Sellin, GaN Detector Development for Particle and X-ray Detection. *Nucl. Instrum. Methods Phys. Res. A* **695** (2012) 303-305.
- [13] T. Gohil, J. Whale, G. Lioliou, S. V. Novikov, C. T. Foxon, A. J. Kent, A. M. Barnett, X-ray Detection with Zinc-Blende (Cubic) GaN Schottky Diodes. *Sci. Rep.* **6** (2016) 29535.
- [14] A.M. Barnett, J.E. Lees, D.J. Bassford, J.S. Ng, A Varied Shaping Time Noise Analysis of $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$ and GaAs soft X-ray Photodiodes Coupled to a Low-Noise Charge Sensitive Preamplifier. *Nucl. Instrum. Methods Phys. Res. A* **673** (2012) 10-15.
- [15] M.D.C. Whitaker, G. Lioliou, S. Butera, A.M. Barnett, $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ 2×2 square pixel X-ray photodiode array. *Nucl. Instrum. Methods Phys. Res. A* **899** (2018) 106-114.
- [16] S. Butera, T. Gohil, G. Lioliou, A.B. Krysa, A.M. Barnett, Temperature Study of $\text{Al}_{0.52}\text{In}_{0.48}\text{P}$ Detector Photon Counting X-ray Spectrometer. *J. Appl. Phys.* **120** (2016) 024502.
- [17] A. Auckloo, J.S. Cheong, X. Meng, C.H. Tan, J.S. Ng, A. Krysa, R.C. Tozer, J.P.R. David, $\text{Al}_{0.52}\text{In}_{0.48}\text{P}$ Avalanche Photodiodes for Soft X-ray Spectroscopy. *J. Instrum.* **11** (2016) P03021.
- [18] S. Zhao, S. Butera, G. Lioliou, A.B. Krysa, A.M. Barnett, AlInP photodiode X-ray detectors. *J. Phys. D Appl. Phys.* **52** (2019) 225101.
- [19] S. Butera, G. Lioliou, A.B. Krysa, A.M. Barnett, InGaP (GaInP) Mesa p-i-n Photodiodes for X-ray Photon Counting Spectroscopy. *Sci. Rep.* **7** (2017) 10206.
- [20] G. Lioliou, A.B. Krysa, A.M. Barnett, Energy response characterization of InGaP X-ray detectors. *J. Appl. Phys.* **124** (2018) 195704.
- [21] C.T.C. Hansson, A. Owens, J. van den Biesen, X-ray, γ -ray and Neutron Detector Development for Future Space Instrumentation. *Acta Astronaut.* **93** (2014) 121-128.
- [22] A. Owens, M. Bavdaz, H. Andersson, T. Gagliardi, M. Krumrey, S. Nenonen, A. Peacock, I. Taylor, L. Trögere, The X-ray Response of CdZnTe. *Nucl. Instrum. Methods Phys. Res. A* **484** (2002) 242-250.
- [23] A. Owens, H. Andersson, M. Bavdaz, L. van den Berg, A. Peacock, A. Puig, The Hard X-ray Response of a Large Area HgI_2 Detector. *Nucl. Instrum. Methods Phys. Res. A* **487** (2002) 90-95.
- [24] V. Kozlov, H. Andersson, V. Gostilo, M. Leskelä, A. Owens, M. Shorohov, H. Sipilä, Improved Process for the TlBr Single-Crystal Detector. *Nucl. Instrum. Methods Phys. Res. A* **591** (2008) 209-212.

- [25] S.F. Kozlov, E. Belcarz, M. Hage-Ali, R. Stuck, P. Siffert, Diamond Nuclear Radiation Detectors. *Nucl. Instrum. Methods Phys. Res.* **117** (1974) 277-283.
- [26] Z. Minglong, X. Yiben, W. Linjun, S. Hujiang, Response of Chemical Vapor Deposition Diamond Detectors to X-ray. *Solid State Commun.* **130** (2004) 425-428.
- [27] H. Kagan, Diamond Radiation Detectors may be Forever!, *Nucl. Instrum. Methods Phys. Res. A* **546** (2005) 222-227.
- [28] M. Pomorski, E. Berdermann, W. de Boer, A. Furgeri, C. Sander, J. Morse, Charge Transport Properties of Single Crystal CVD-Diamond Particle Detectors. *Diam. Relat. Mater.* **13** (2007) 1066-1069.
- [29] J. Isberg, J. Hammersberg, E. Johansson, T. Wikström, D.J. Twitchen, J. A. Whitehead, S.E. Coe, G.A. Scarsbrook, High Carrier Mobility in Single-Crystal Plasma-Deposited Diamond. *Science* **297** (2002) 1670-1672.
- [30] Anon *The Element Six CVD Diamond Handbook*. (Element Six Technologies).
- [31] J.H. Kaneko, T. Tanaka, T. Imai, Y. Tanimura, M. Katagiri, T. Nishitani, H. Takeuchi, T. Sawamura, T. Iida, Radiation Detector Made of a Diamond Single Crystal Grown by a Chemical Vapor Deposition Method. *Nucl. Instrum. Methods Phys. Res. A* **505** (2003) 187-190.
- [32] Y. Sato, H. Murakami, T. Shimaoka, M. Tsubota, J.H. Kaneko, Charge-Collection Efficiency and Long-Term Stability of Single-Crystal CVD Diamond Detector under Different Carrier-Drift Conditions, *Jpn. J. Appl. Phys.* **55** (2016) 046401.
- [33] M. Tsubota, J.H. Kaneko, D. Miyazaki, T. Shimaoka, K. Ueno, T. Tadokoro, A. Chayahara, H. Watanabe, Y. Kato, S. Shikata, H. Kuwabara, High-Temperature Characteristics of Charge Collection Efficiency Using Single CVD Diamond Detectors. *Nucl. Instrum. Methods Phys. Res. A* **789** (2015) 50-56.
- [34] G.F. Knoll, *Radiation Detection and Measurement* (John Wiley & Sons, 2010).
- [35] R. Pilotti, M. Angelone, S. Loreti, G. Pagano, M. Pillon, F. Sarto, M. Marinelli, E. Milani, G. Prestopino, C. Verona, G. Verona-Rinati, High Temperature Performances of CVD Single Crystal Diamond Detectors. *1st EPS Conference on Plasma Diagnostics Frascati*; Italy (2015).
- [36] J.W. Keister, J. Smedley, Single Crystal Diamond Photodiode for Soft X-ray Radiometry. *Nucl. Instrum. Methods Phys. Res. A* **606** (2009) 774-779.
- [37] F. Schirru, A. Lohstroh, K.D.G.I. Jayawardena, S.J. Henley, P.J. Sellin, X-ray Induced Photocurrent Characteristics of CVD Diamond Detectors with Different Carbon Electrodes. *J. Instrum.* **8** (2013) C12046.
- [38] N. Tartoni, M. Angelone, M. Pillon, S. Almagia, M. Marinelli, E. Milani, G. Prestopino, C. Verona, G.V. Rinati, M.A. Roberts, X-Ray Detection by Using CVD Single Crystal Diamond Detector. *IEEE Trans. Nucl. Sci.* **56** (2009) 849-852.
- [39] A. Lohstroh, P.J. Sellin, F. Boroumand, J. Morse, High Gain Observed in X-ray Induced Currents in Synthetic Single Crystal Diamonds. *Phys. Status Solidi A* **204** (2007) 3011-3016.
- [40] P. Hovington, D. Drouin, R. Gauvin, CASINO: A new Monte Carlo Code in C Language for Electron Beam Interaction —part I: Description of the Program. *Scanning* **19** (1997) 1-14.
- [41] D. Drouin, P. Hovington, R. Gauvin, CASINO: A new Monte Carlo Code in C language for Electron Beam Interactions—part II: Tabulated Values of the Mott Cross Section. *Scanning* **19** (1997) 20-28.
- [42] A. Owens, Semiconductor materials and radiation detection. *J. Synchrotron Rad.* **13** (2006) 143-450.
- [43] Anon *Model 6487 Picoammeter/Voltage Source Reference Manual*, 6487-901-01 Rev C (Keithley Instruments, 2011).

- [44] G. Bertuccio, D. Puglisi, D. Macera, R. Di Liberto, M. Lamborizio, L. Mantovani, Silicon Carbide Detectors for in vivo Dosimetry. *IEEE Trans. Nucl. Sci.* **61**, (2014) 961-966.
- [45] G. Bertuccio, D. Puglisi, A. Pullia, C. Lanzieri, X and gamma Ray Spectroscopy With Semi-Insulating 4H-Silicon Carbide. *IEEE Trans. Nucl. Sci.* **60** (2013) 1436-1441.
- [46] J.P. Goss, P.R. Briddon, R. Jones, S. Sque, Donor and Acceptor States in Diamond. *Diam. Relat. Mater.* **13** (2004) 684-690.
- [47] U. Schötzgig, Half-Life and X-ray Emission Probabilities of ^{55}Fe . *Appl. Radiat. Isot.* **53** (2000) 469-472.
- [48] J.H. Hubbell, Photon mass attenuation and energy-absorption coefficients. *Int. J. Appl. Radiat. Isot.* **33**, (1982) 1269-1290.
- [49] A. Owens, *Compound Semiconductor Radiation Detectors* (CRC Press, 2012).
- [50] K. Scharf, R.K. Mohr, Temperature Dependence of Photocurrents Produced by X and Gamma Rays in Silicon Radiation Detectors. *J. Res. Natl. Bur. Stand.* **75A** (1971) 579-589.
- [51] G. Bertuccio, D. Maiocchi, Electron-Hole Pair Generation Energy in Gallium Arsenide by x and γ Photons. *J. Appl. Phys.* **92** (2002) 1248-1255.
- [52] F. Nava, C. Canali, M. Artuso, E. Gatti, P.F. Manfredi, S.F. Kozlov, Transport Properties of Natural Diamond Used as Nuclear Particle Detector for a Wide Temperature Range. *IEEE Trans. Nucl. Sci.* **26**, (1979) 308-315.
- [53] A. Brambilla, D. Tromson, P. Bergonzo, C. Mer, F. Foulon, Thin CVD diamond detectors with high charge collection efficiency. *IEEE Trans. Nucl. Sci.* **49** (2002) 277-280.
- [54] S.M. Sze, K.K. Ng, *Physics of Semiconductor Devices* (John Wiley & Sons, 2007).
- [55] T. Behnke, P. Huntemeyer, A. Oh, J. Steuerer, A. Wagner, W. Zeuner, The Charge Collection Properties of CVD Diamond. *Nucl. Instrum. Methods Phys. Res. A* **414** (1998) 340-356.
- [56] C. Manfredotti, A. Lo Giudice, E. Vittone, F. Fizzotti, Y. Garino, E. Pace, Memory effects in CVD diamond. *Diam. Relat. Mater.* **15** (2006) 1467-1471.
- [57] F. Kassel, M. Guthoff, A. Dabrowski, W. de Boer, Severe signal loss in diamond beam loss monitors in high particle rate environments by charge trapping in radiation induced defects. *Phys. Status Solidi A* **213** (2016) 2641-2649.
- [58] T.R. Alam, M.A. Pierson, Principles of Betavoltaic Battery Design. *J. Energy* **3** (2016) 11-41.
- [59] Y.-p. Liu, X.-b. Tang, Z.-h. Xu, L. Hong, H. Wang, M. Liu, D. Chen, Influences of Planar Source Thickness on Betavoltaics with Different Semiconductors. *J. Radioanal. Nucl. Chem.* **304** (2015) 517-525.
- [60] Y. Sato, H. Murakami, T. Shimaoka, M. Tsubota, J.H. Kaneko, Charge-collection efficiency of single-crystal CVD diamond detector for low-energy charged particles with energies ranging from 100 keV to 2 MeV. *Nucl. Instrum. Methods Phys. Res. A* **834** (2016) 218-222.