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1 # GaAs/$Al_{0.8}Ga_{0.2}As$ Separate Absorption and Multiplication Region X-ray
2 Spectroscopic Avalanche Photodiodes

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6

7 A GaAs/$Al_{0.8}Ga_{0.2}As$ Separate Absorption and Multiplication (SAM) X-ray avalanche photodiode (APD)
8 structure was grown by metalorganic vapour phase epitaxy. Mesa photodiodes of different diameter (200 µm and
9 400 µm) were fabricated from the structure. Two of the photodiodes (one of each diameter) were characterised
10 at 20 °C for their electrical properties and response to X-rays using an ^{55}Fe radioisotope X-ray (Mn Kα = 5.9 keV;
11 Mn Kβ = 6.49 keV) source. An energy resolution of 508 eV ± 5 eV Full Width at Half Maximum (FWHM) at
12 5.9 keV was achieved at an apparent avalanche gain, M, of 1.1. This is the best energy resolution so far reported
13 for GaAs/$Al_xGa_{1-x}As$ X-ray SAM APDs. The noise components associated with the achievable spectroscopic
14 energy resolutions are reported. Comparisons between the 200 µm and 400 µm diameter GaAs/$Al_xGa_{1-x}As$ SAM
15 X-ray APDs and recently studied GaAs p^+-i-n^+ detectors were made, showing that the inclusion of the avalanche
16 layer improves the achievable energy resolution; energy resolutions of 508 eV FWHM at 5.9 keV at M = 1.1 and
17 603 eV FWHM at 5.9 keV at M = 1.2 were achieved with the 200 µm and 400 µm diameter GaAs/$Al_xGa_{1-x}As$
18 SAM X-ray APDs respectively; this is better than was previously reported for similar devices without avalanche
19 layers: 690 eV FWHM at 5.9 keV and 730 eV FWHM at 5.9 keV for 200 µm and 400 µm diameter GaAs p^+-i-n^+
20 detectors respectively (G.Lioliou et al., J. Appl. Phys. 122, 244506 (2017)).
21

22 **KEYWORDS**
23 GaAs; AlGaAs; SAM; APD; X-ray detector; Spectroscopy
24

25 ## I. INTRODUCTION
26 Due to the limitations of narrow bandgap (typically Si) X-ray spectrometers commonly in use today [1, 2], a
27 variety of wide bandgap materials, such as GaAs [3-7], diamond [8, 9], SiC [10-12], $In_{0.5}Ga_{0.5}P$ [13, 14],
28 $Al_{0.52}In_{0.48}P$ [15-17], and $Al_xGa_{1-x}As$ [18-21], have been investigated as potential X-ray detector
29 replacements. The cooling systems and radiation shielding often required for Si X-ray spectrometers [22] place
30 substantial burdens on spacecraft mass, volume, and power consumption, limiting their suitability for certain space
31 science applications (e.g. missions to study the surface of Mercury, or the Jovian moons, where temperatures
32 and/or radiation intensities are significant). Wide bandgap materials, such as GaAs and $Al_xGa_{1-x}As$, offer an
33 alternative. Such materials can operate in a large range of thermal and radiation environments, while still
34 providing sub-keV spectral resolutions at soft X-ray energies [22]. In the case of GaAs, its wider bandgap
35 (1.43 eV for GaAs cf. 1.12 eV for Si [22]), larger X-ray absorption coefficient (837 cm^{-1} for GaAs cf. 346 cm^{-1}
36 for Si, at 5.9 keV [23]), and improved radiation hardness in comparison to Si [24], allows for superior energy
37 resolutions at high temperature [25], thinner X-ray detecting structures [26], and potentially longer instrument
38 lifetimes in intense radiation environments.

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39

40 Various prototype GaAs based X-ray spectrometers have been reported, with most work focused on GaAs p^+-i-n^+
41 mesa X-ray photodiodes operated in the non-avalanche regime. Circular mesa GaAs, 200 µm diameter, 7 µm
42 thick i layer, p^+-i-n^+ photodiodes have been characterised across the temperature range 60 °C to 0 °C, with an
43 energy resolution of 750 eV FWHM at 5.9 keV for the best performing device at 20 °C [6]. Similar GaAs
44 photodiodes (10 µm i layer) have been characterised over the temperature range 100 °C to -20 °C, with an energy
45 resolution of 690 eV FWHM at 5.9 keV reported at room temperature (20 °C) [27]. GaAs planar p^+-i-n^+
46 photodiode arrays have also been reported with very promising results; a 5 × 5 pixel, 40 µm i layer, GaAs p^+-i-n^+
47 diode array structure had a reported energy resolution of 266 eV FWHM at 5.9 keV at room temperature [3].
48 GaAs arrays of a similar structure, but with thicker epilayer layers (325 µm i layer), had a reported energy
49 resolution of 300 eV FWHM at 5.9 keV at room temperature [28].

50

51 $Al_xGa_{1-x}As$, due to the material's ability to be tailored to particular application environments by adjusting the Al
52 fraction (e.g. a reduction in Al fraction reduces the bandgap), has gained attention as an interesting material for
53 X-ray detection [21, 26, 29]. Most work has focused on $Al_xGa_{1-x}As$ p^+-i-n^+ mesa photodiodes, operated within
54 the non-avalanche regime. Circular mesa $Al_{0.8}Ga_{0.2}As$, 200 µm diameter, 1 µm i layer, p^+-i-n^+ photodiodes have
55 been characterised across the temperature range 90 °C to -30°C, with an energy resolution of 1.07 keV FWHM at
56 5.9 keV reported at 20 °C [29]. Circular mesa $Al_{0.2}Ga_{0.8}As$, 200 µm diameter, 3 µm i layer, p^+-i-n^+ photodiodes
57 have been characterised across the temperature range 20 °C to -20 °C, with an energy resolution of 1.06 keV
58 FWHM at 5.9 keV reported at 20 °C [21]. A 2 × 2 square pixel $Al_{0.2}Ga_{0.8}As$ array (each photodiode area 200 µm
59 by 200 µm, 3 µm i layer) was also recently reported with improved results; an energy resolution of 760 eV FWHM
60 at 5.9 keV at 20 °C was achieved [26].

61

62 Whilst non-avalanche GaAs and $Al_xGa_{1-x}As$ X-ray detectors show great promise, the demands they place on their
63 preamplifier electronics are more stringent than those of narrower bandgap materials like Si, because the
64 electron-hole pair creation energies of GaAs and $Al_xGa_{1-x}As$ are larger (e.g. 4.2 eV for GaAs cf. 3.6 eV for Si
65 [22]). Avalanche photodiodes (APDs) potentially reduce those demands by increasing the amount of charge
66 created from the absorption of an X-ray photon. However, the greater operating biases required can cause higher
67 leakage currents which results in more parallel white noise.

68

69 APDs are widely used in photonic detection systems, most notably within telecommunications [30-33], to improve
70 response relative to that of conventional p^+-i-n^+ photodiodes [30]. The increased response from such APDs is due
71 to the impact ionisation process, where charge carriers gain enough kinetic energy to generate electron-hole pairs
72 during collisions with atoms in the lattice [34, 35].

73

74 For photons of infrared to ultraviolet wavelength, where the photon energy is of the same order as the bandgap
75 energy of the detector material, the stochastic nature of the impact ionisation process in APDs commonly adds
76 noises (quantified by the so called the excess noise factor, N_x) to the signal [36, 37] such that

77

78 $$N_x = kM + \left(2 - \frac{1}{M}\right)(1 - k), \qquad (1)$$

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79
80 where k (= β/α) is the ratio between the hole (β) and electron (α) ionisation coefficients, which represent the
81 inverse of the mean distance between successive impact ionisation events [36]. In most compound
82 semiconductors, k can range from 0.3 to 1.0, leading to a large N_x [38]. Hence, as per Eq. 1, assuming an extension
83 of applicability of this model to the X-ray case, a poor energy resolution was originally expected [38]. For this
84 reason, it was originally thought that the use of APDs would degrade the resolution of X-ray spectrometers and
85 hence they were not extensively investigated [39]. However, recent studies have improved the understanding of
86 APDs [31, 35, 36, 38, 40]. Notably, Tan et al. [38] showed that the common model of excess noise is not directly
87 applicable at X-ray energies, since the distribution of avalanche gains tightens as the initiating photon energy, E,
88 is increased, thus leading to significantly lower additional noise for X-rays than would be expected in the case for
89 infrared, visible, or UV photons [38]. Additional design choices such as using very thin avalanche layers can also
90 be used to reduce excess noise [31].
91
92 The first X-ray APDs were complex staircase band structures [18, 41, 42]. For example, a $GaAs/Al_xGa_{1-x}As$ SAM
93 APD (320 μm × 450 μm active area) was reported to function as an X-ray detector at room temperature, utilising
94 a series of staircase multiplication regions [18]; an energy resolution of 900 eV FWHM at 13.96 keV was reported
95 at an avalanche multiplication of 4.1 at room temperature [18]. More recent work has concentrated on simpler
96 SAM APD structures [43]. For example, a thin (430 nm GaAs absorption layer, 220 nm $Al_{0.8}Ga_{0.2}As$
97 multiplication layer) $GaAs/Al_{0.8}Ga_{0.2}As$ SAM X-ray APD was reported at room temperature [44]; it had an energy
98 resolution of 1.08 keV FWHM at 5.9 keV, at an avalanche multiplication of 3.5.
99
100 In this work we report the growth, fabrication, and characterisation at room temperature (20 °C) of two new
101 circular $GaAs/Al_{0.8}Ga_{0.2}As$ SAM X-ray photodiodes of different diameter (200 μm and 400 μm). The results are
102 compared with recently reported non-avalanche GaAs p^+-i-n^+ X-ray photodiodes of the same size (200 μm and
103 400 μm diameter) and with the same absorption layer thickness (10 μm) [27]. The same measurement techniques
104 and readout electronics were used in both cases. The work shows that the addition of the avalanche layer improves
105 the energy resolution. This unambiguous comparison of the detectors both with and without an avalanche layer
106 but otherwise of the same design provides conclusive evidence that the addition of an avalanche layer can be used
107 to improve the energy resolution of a spectroscopic photon counting X-ray detector.
108

109 **II. DIODE DESIGN**

110 The $GaAs/Al_{0.8}Ga_{0.2}As$ SAM structure (see Fig. 1) was grown by metalorganic vapour phase epitaxy upon a
111 commercial GaAs n^+ substrate. The photodiode layer details are outlined in Table 1. Circular mesa structures of
112 200 μm diameter and 400 μm diameter were etched using a 1:1:1 H_3PO_4:H_2O_2:H_2O solution followed by 10 s in
113 a 1:8:80 H_2SO_4:H_2O_2:H_2O solution. Ohmic contacts consisting of 200 nm Au and 20 nm Ti were evaporated upon
114 the top p^+ side of the mesa structures and ohmic contacts consisting of 200 nm Au and 20 nm InGe were evaporated
115 upon the back side of the substrate. The top contacts covered 45% of the 200 μm diameter diode's face and 33%
116 of the 400 μm diameter diode's face.
117

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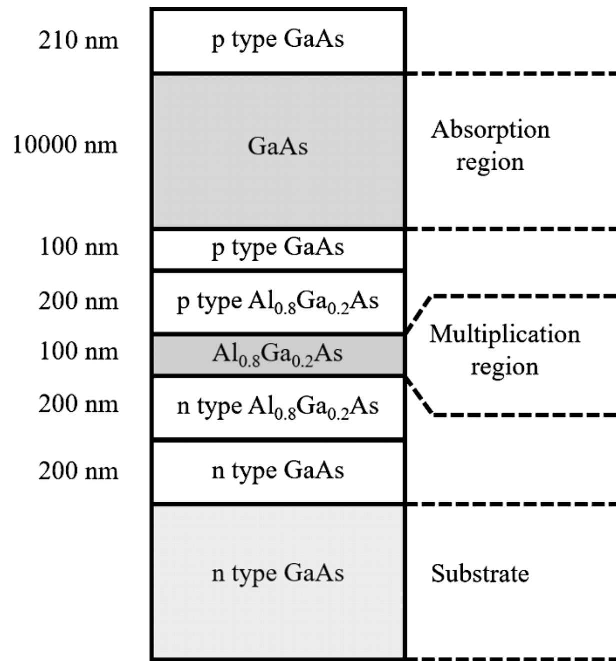

118

119 Fig. 1. Schematic of GaAs/$Al_{0.8}Ga_{0.2}As$ SAM structure.

120

121 Table 1. GaAs/$Al_{0.8}Ga_{0.2}As$ SAM layer details.

Material	Dopant	Dopant type	Thickness (nm)	Doping density (cm^{-3})
GaAs	Zn	p	10	1×10^{19}
GaAs	Zn	p	200	2×10^{18}
GaAs			10000	Undoped
GaAs	Zn	p	100	2×10^{17}
$Al_{0.8}Ga_{0.2}As$	Zn	p	200	2×10^{17}
$Al_{0.8}Ga_{0.2}As$			100	Undoped
$Al_{0.8}Ga_{0.2}As$	Si	n	200	2×10^{18}
GaAs	Si	n	200	2×10^{18}
GaAs n^+ substrate				

122

123 **III. DETECTOR ELECTRICAL CHARACTERISATION**

124 **A. Capacitance as a function of applied reverse bias**

125 For each GaAs/$Al_{0.8}Ga_{0.2}As$ SAM photodiode (200 µm and 400 µm diameter), capacitance as a function of reverse
126 bias, V_R, was measured using an HP 4275A LCR Meter (50 mV rms signal magnitude; 1 MHz frequency). A
127 Keithley 6487 voltage source/picoammeter was used to bias the detectors. Each device was placed within a
128 custom-made, light-tight, electromagnetically screened test fixture and, for temperature control, inserted in a TAS
129 Micro MT environmental chamber. An appropriately positioned thermocouple was used to ensure thermal
130 equilibrium (20 °C) was reached between the environmental chamber and the devices. The test fixture and
131 environmental chamber were purged with dry N_2 (<5% relative humidity) in order to remove any humidity related
132 effects [6]. The environmental chamber was set to 20 °C and left for 1 hour before measuring to ensure thermal

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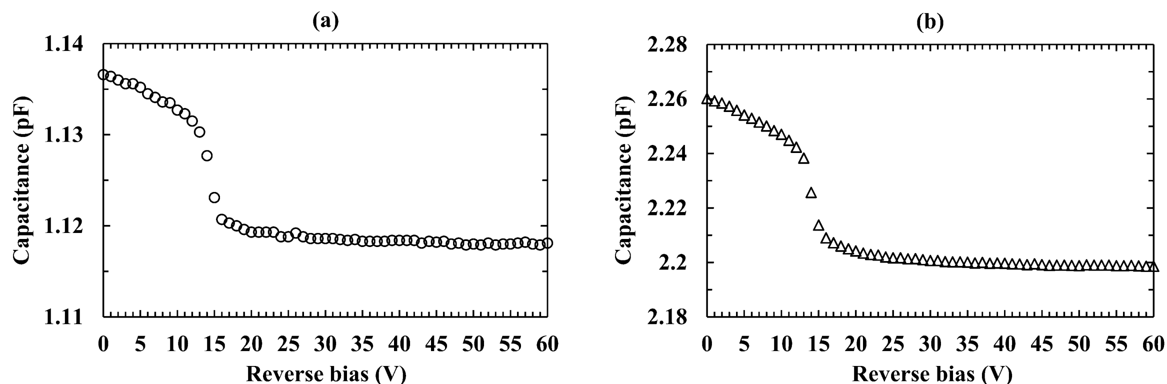
133 equilibrium. The measured capacitances as functions of reverse bias for (a) the 200 µm diameter device and (b)
134 the 400 µm diameter device is shown in Fig. 2.
135



136
137 Fig. 2. Capacitance for (a) the 200 µm diameter device and (b) the 400 µm diameter device, as a function of applied reverse
138 bias, at 20 °C. The empty package capacitance, in each case, has not been subtracted.
139

140 The measured capacitance of both packaged devices decreased with increasing reverse bias, from 1.14 pF and
141 2.26 pF at $V_R = 0$ V for the 200 µm and 400 µm diameter device respectively, to 1.12 pF and 2.20 pF at $V_R = 60$ V
142 for the 200 µm and 400 µm diameter device respectively. The uncertainty associated with each individual
143 capacitance measurement was ≈ ± 0.03 pF. However, because a set of measurements were taken without
144 modifying the conditions (e.g. no variations in electrical connections and temperature), fittings on the
145 experimental data provide a more appropriate uncertainty for relative changes [45]. Exponential fittings on the
146 measured capacitance for each device were performed as a function of reverse bias. An uncertainty of ± 0.4 fF
147 was estimated. The drop in capacitance across both the 200 µm and 400 µm diameter devices, within the range
148 $13 \text{ V} \leq V_R \leq 16$ V, indicated that the punch-through voltage (the voltage at which the multiplication region rapidly
149 depletes) was ≈ 14 V [44].
150

151 The measured capacitance, C_M, included both the diode capacitance, C_D, and the package capacitance, C_P, since
152 the devices were packaged. C_P was removed by assuming a constant capacitance density as a function of device
153 area. The capacitance density of the 200 µm diameter device and the 400 µm diameter device at each applied
154 reverse bias were compared, and the empty package capacitance calculated. A mean average empty package
155 capacitance (0.76 pF) was calculated for C_P and was subsequently subtracted from C_M for each device. Fig. 3
156 presents the capacitance densities for the 200 µm and 400 µm diameter devices.
157

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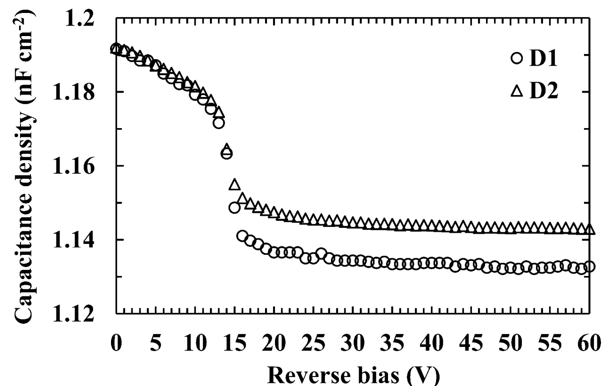

158
159 Fig. 3. Capacitance density for the 400 μm diameter device (triangles) and the 200 μm diameter device (circles) as a function
160 of applied reverse bias, at 20 °C, taking into account the empty package capacitance (0.76 pF).
161
162 The capacitances of the devices were defined primarily by the depletion layer capacitance, C_{DL} of each device
163 [34]. Therefore, the depletion width, W, could be calculated using the equation
164

165 $$W = \frac{\varepsilon_0 \varepsilon A}{C_{DL}}, \quad (2)$$

166
167 where A is the device area, ε is the relative permittivity of the material, and ε_0 is the permittivity of free space [34].
168 For the reported devices, multiple materials (GaAs and $Al_{0.8}Ga_{0.2}As$) influence C_{DL}, with their associated
169 contributions difficult to detangle. As such, in order to calculate W, the devices were approximated to be simple
170 GaAs structures (ε = 13.16 [46]). Since ε of $Al_{0.8}Ga_{0.2}As$ (= 10.628 [47]) is smaller in value, the presently reported
171 W should be taken as an upper limit. The Debye length of GaAs (0.06 μm) was also taken into account when
172 calculating the depletion width uncertainty [48]. Fig. 4 (a) shows the depletion width as a function of applied
173 reverse bias.
174
175 The depletion width increased as a function of applied reverse bias for both devices, increasing from 9.8 μm
176 ± 0.1 μm at V_R = 0 V for the 200 μm and 400 μm diameter device, to 10.3 μm ± 0.1 μm and 10.2 μm ± 0.1 μm at
177 V_R = 60 V for the 200 μm and 400 μm diameter device, respectively. As was the case in Fig. 2, the increase in
178 depletion width across both the 200 μm and 400 μm diameter devices, between 13 V and 16 V, indicated the
179 punch-through voltage. Linear least squares fitting was applied to both devices, and indicated that the devices
180 were fully depleted at $V_R \geq 50$ V.
181
182 The GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD structure quantum detection efficiency was calculated using the Beer-Lambert
183 law, assuming that the active region was solely confined to the GaAs absorption layer and that it was fully depleted
184 and active. The results can be seen in Fig. 4 (b). For photons of 5.9 keV energy, the quantum detection efficiencies
185 of the devices structure presented here were 0.56 in areas not covered by the top contact, and 0.46 in areas covered
186 by the top contact. The weighted quantum efficiency assuming uniform illumination of the devices was 0.52 and
187 0.53 for the 200 μm and 400 μm diameter detectors, respectively.
188

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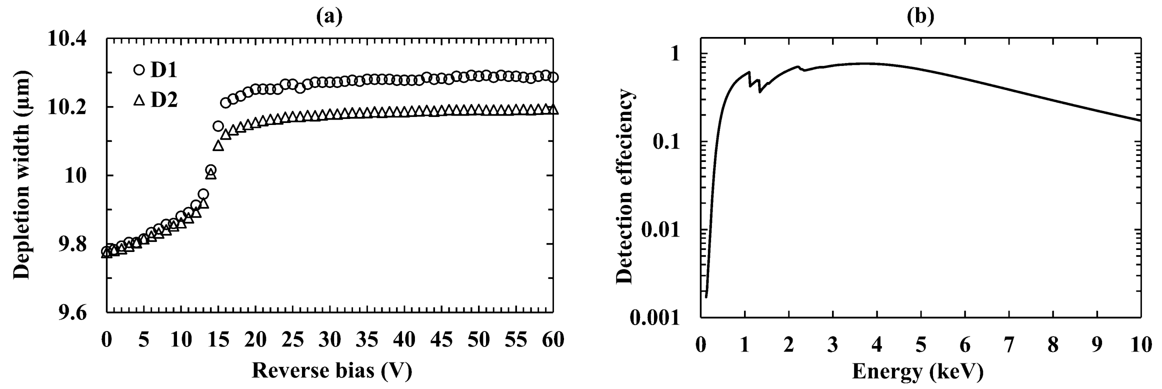

190 Fig. 4. (a) Calculated depletion width for the 200 µm diameter device (circles) and the 400 µm diameter device (triangles) as
191 a function of applied reverse bias, at 20 °C. (b) Calculated detection efficiency for the GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD structure
192 as a function of energy. The Al K, Ga L, and As L X-ray absorption edges are responsible for the detection efficiency
193 discontinuities.

194

195 The general nonuniform distributions equation [34] was used to calculate the carrier concentration of the space
196 charge region, N, where,

197

198 $$\frac{\mathrm{d}\left(1/{C_{DL}}^2\right)}{\mathrm{d}V_R} = \frac{2}{\mathrm{q}\varepsilon_0 \varepsilon N}, \quad (3)$$

199

200 where q is the elementary charge and the other symbols have previously been defined. For both devices, at a
201 calculated depletion width ≈ 10 µm, the carrier concentration reached a minimum of ≈ 7 × 10^{14} cm^{-3}. Fig. 5
202 presents the carrier concentration for the GaAs/$Al_{0.8}Ga_{0.2}As$ detectors as a function of calculated depletion width.
203 Variation in the apparent carrier concentration between the 200 µm and 400 µm devices was within the uncertainty
204 of the measurements.

205

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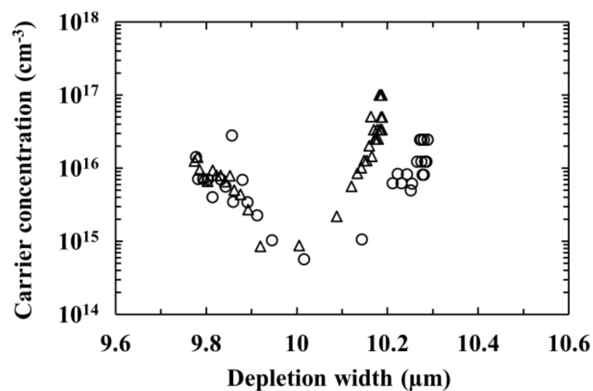

207 Fig. 5. Carrier concentration for the 200 µm (circles) and 400 µm (triangles) diameter devices as a function of calculated
208 depletion width.

209

210 **B. Current as a function of applied reverse bias**

211 The leakage current of the 200 µm and 400 µm diameter devices was measured using a Keithley 6487 voltage
212 source/picoammeter as a function of applied reverse bias. The environmental conditions were the same as for the

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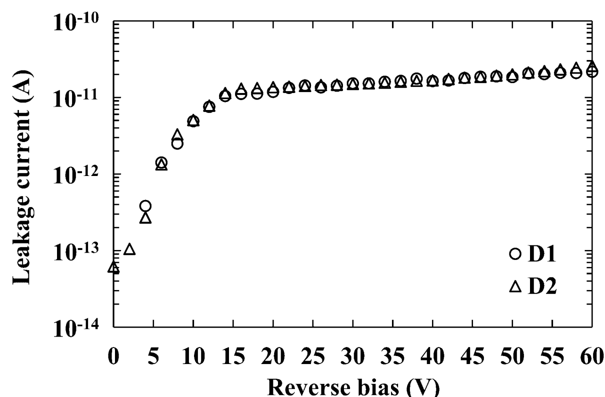
213 capacitance measurements. Fig. 6 presents the measured leakage current, I_R, of the packaged devices as a function
214 of applied reverse bias.
215



216
217 Fig. 6. Leakage current for the 200 µm diameter device (circles) and the 400 µm diameter device (triangles) as a function of
218 applied reverse bias.
219
220 For both packaged devices, the leakage current increased with increasing reverse bias. At the maximum applied
221 reverse bias (60 V), the leakage currents were measured to be 21.6 pA ± 0.8 pA for the 200 µm device and 25.7 pA
222 ± 0.5 pA for the 400 µm device. The uncertainties associated with the current measurements were dominated by
223 the uncertainty associated with a single measurement from the Keithley 6487 voltage source/picoammeter.
224
225 In order to determine the corresponding leakage current density, J_R, of the devices, the leakage current associated
226 with the package (and measurement system) was determined by measuring an empty package of identical type.
227 It was found that the empty package's leakage current was smaller than the uncertainty (± 0.4 pA) of the
228 instrument, it was therefore considered negligible. The apparent leakage current density, as shown in Fig. 7, of
229 the 400 µm diameter device was improved (lower) relative to the 200 µm diameter device (20.4 nA cm^{-2}
230 ± 0.4 nA cm^{-2} for the 400 µm diameter device cf. 68.8 nA cm^{-2} ± 2.7 nA cm^{-2} for the 200 µm diameter device, at
231 the maximum applied reverse bias (60 V). This suggested that the leakage current did not scale with junction
232 area. The presence of a non-negligible surface leakage current, possibly due to the devices being unpassivated
233 [49, 50], cannot be excluded entirely, but the measured leakage currents do not scale with circumference either.
234 A similar trend was recently reported for GaAs p^+-i-n^+ mesa X-ray photodiodes of the same size (200 µm and 400
235 µm diameter) and with the same absorption layer thickness (10 µm) [27]. For the present devices, the difference
236 in leakage current density with junction area was possibly attributable to damage caused by wirebonding to the
237 thin metal contacts, resulting in a dominating additional leakage current component.
238

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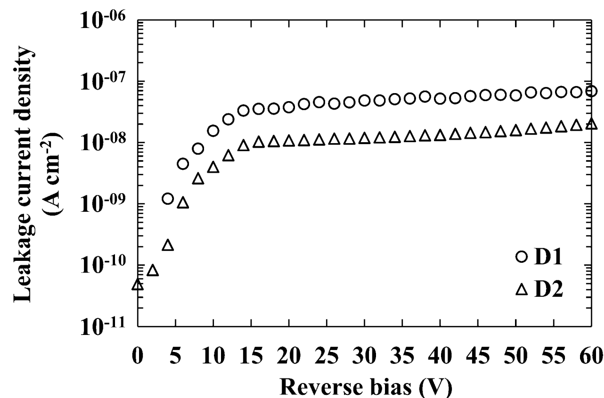

239
240 Fig. 7. Apparent leakage current density for the 200 μm (circles) and 400 μm (triangles) diameter devices as a function of
241 applied reverse bias, at 20 °C.
242
243 The leakage current densities of the presently reported devices were greater than those measured for recently
244 reported GaAs p$^+$-i-n$^+$ mesa X-ray photodiodes [27]. At an internal electric field strength, E_f, of 50 kV cm^{-1}
245 (corresponding to 51 V applied reverse bias for the GaAs/$Al_{0.8}Ga_{0.2}As$ devices, assuming E_f was uniform and
246 across only the depleted region), leakage current densities of 61.8 nA cm^{-2} ± 1.5 nA cm^{-2} and 16.4 nA cm^{-2}
247 ± 0.4 nA cm^{-2} were measured for the 200 μm diameter and 400 μm diameter devices respectively. At the same
248 E_f, leakage current densities of 15 nA cm^{-2} and 6 nA cm^{-2} were measured for the 200 μm and 400 μm GaAs
249 devices respectively, at 20 °C [27].
250

251 IV. PHOTON COUNTING X-RAY SPECTROSCOPY

252 A. Experimental measurements and determination of the spectrometers' energy resolutions

253 In order to characterise the X-ray detection performance of the GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD devices, each
254 detector was connected, in turn, to a custom-made low-noise charge-sensitive feedback-resistorless preamplifier,
255 similar in design to ref. [51]. The preamplifier, in each case, was connected to a shaping amplifier (Ortec 572A)
256 and a multi-channel analyser (Ortec Easy-MCA 8k). An ^{55}Fe X-ray (Mn Kα = 5.9 keV; Mn Kβ = 6.49 keV)
257 source (≈ 131 MBq) was positioned ≈ 4 mm above each GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD in turn. The resulting
258 spectrometers: S_{200} (using the 200 μm diameter detector) and S_{400} (using the 400 μm diameter detector) were
259 installed within a TAS Micro MT environmental chamber for temperature control. The environmental chamber
260 temperature was set to 20 °C, and allowed to stabilise for 1 hour before measurements were taken. A
261 thermocouple was positioned close to the spectrometer such that temperature equilibrium between the
262 environmental chamber and the spectrometer could be monitored. The environmental chamber was purged
263 continually throughout the measurements with dry N_2 (< 5% relative humidity) as to reduce any humidity related
264 effects [52].
265
266 Spectra were accumulated for each spectrometer as a function of detector applied reverse bias. The applied reverse
267 bias was initially set to 0 V, then increased in 1 V steps up to 50 V. The reverse bias was increased further in
268 steps of 5 V, up to 60 V. After each voltage change, the system was allowed to stabilise for 5 minutes before
269 taking a measurement. Since the two detectors had different active areas, the live time limits of each spectrum
270 were set differently: spectra accumulated with S_{200} had a live time limit of 100 s; spectra accumulated with S_{400}

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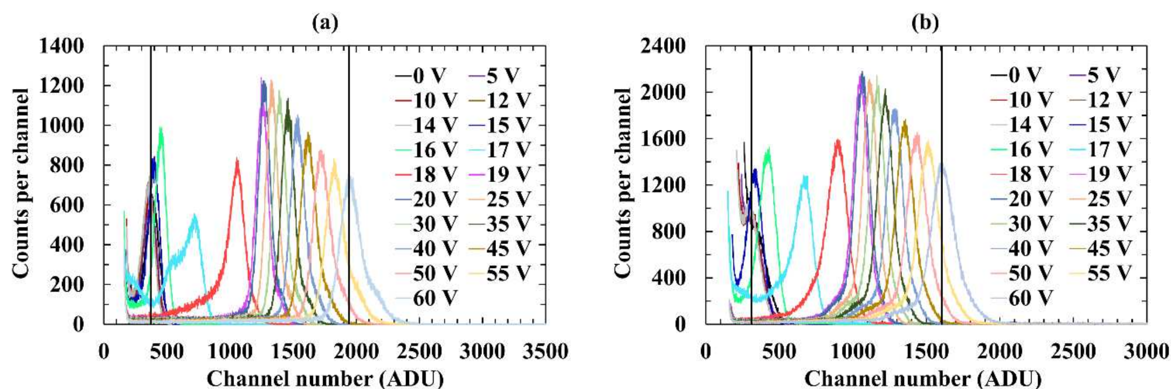
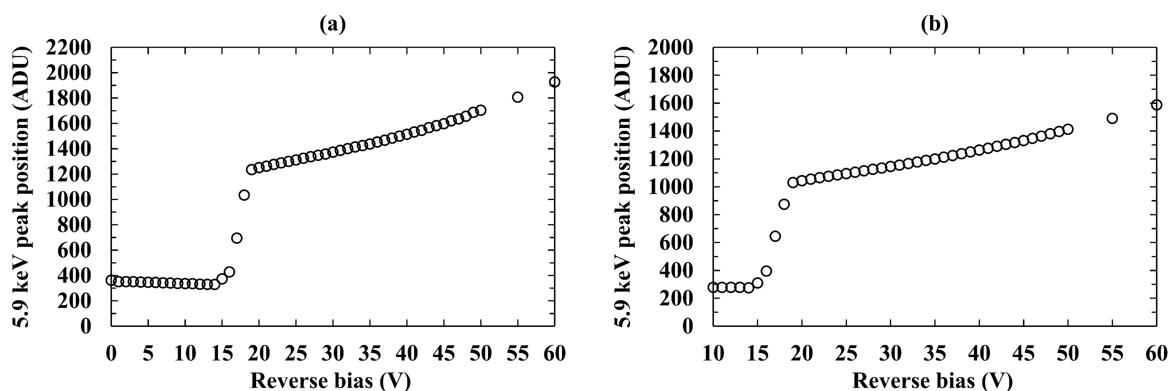
271 had a live time limit of 25 s. A shaping time of 0.5 µs was used; this was the best available shaping time for each
272 system. The accumulated ^{55}Fe spectra for the 200 µm and 400 µm diameter $GaAs/Al_{0.8}Ga_{0.2}As$ SAM APD
273 spectrometers can be seen in Fig. 8.
274



275
276 Fig. 8. Accumulated ^{55}Fe spectra using the spectrometers (a) S_{200} and (b) S_{400}. The same shaping time (0.5 µs) and temperature
277 (20 °C) were used for all spectra. The vertical black lines indicate the positions of the combined 5.9 keV (Mn Kα) and 6.49 keV
278 (Mn Kβ) X-ray photopeaks from the ^{55}Fe X-ray source accumulated with the detectors reverse biased at 0 V and 60 V.
279

280 For each accumulated spectrum from the ^{55}Fe X-ray (Mn Kα = 5.9 keV; Mn Kβ = 6.49 keV) source, Gaussian
281 fitting was applied. The relative emission ratio [53] and the relative detection efficiency of the $GaAs/Al_{0.8}Ga_{0.2}As$
282 SAM APDs at these energies were taken into account in fitting the Mn Kα and Kβ peaks. The Mn Kα and
283 Kβ peaks were not individually resolved by the spectrometer; as such, the peak detected is the combination of the
284 Mn Kα and Kβ lines. The form of spectroscopic response was consistent with a SAM APD; this was further
285 exemplified by plotting the change in 5.9 keV peak centroid position (corrected for changes in zero energy noise
286 peak position and plotted in terms of the MCA's analogue to digital units, ADU, scale) as a function of applied
287 detector reverse bias, as shown in Fig. 9.
288



289
290 Fig. 9. Difference, in ADU, between the 0 keV position and the position of the centroid of the fitted 5.9 keV peak as a function
291 of applied detector reverse bias, at a shaping time of 0.5 µs, and a temperature of 20 °C for the spectrometers (a) S_{200} and (b)
292 S_{400}.
293

294 The sharp increase in 5.9 keV peak position between 14 V and 19 V for both the 200 µm and 400 µm diameter
295 devices, as shown in Fig. 9, was attributed to an improved charge collection efficiency from reaching the

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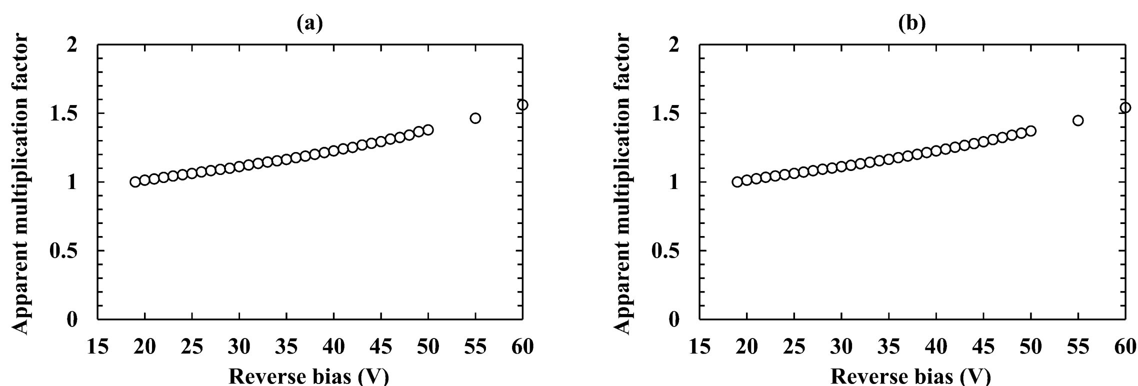
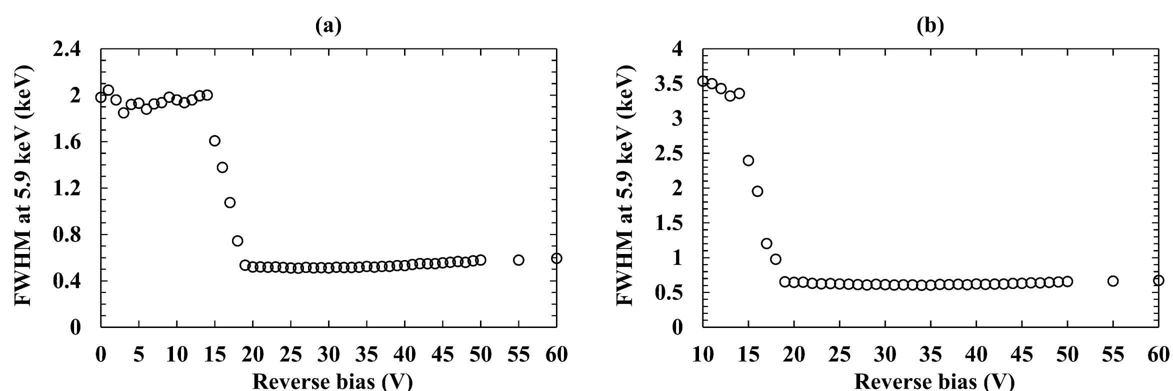
296 punch-through voltage (see Fig. 2). At applied detector reverse biases less than the punch-through voltage, charge
297 carriers cannot readily travel through the $Al_{0.8}Ga_{0.2}As$ junction (see Table 1) [18], limiting the charge collection
298 efficiency. The increasing 5.9 keV peak position as a function of applied detector bias beyond 19 V was a result
299 of increases in avalanche multiplication. The apparent multiplication factor, M, was calculated for both
300 spectrometers by calculating the ratio between the fitted 5.9 keV peak position at each bias and the fitted 5.9 keV
301 peak position at unity gain (M = 1) and complete charge collection, assumed to be achieved at V_R = 19 V applied
302 to the detector. The apparent multiplication factor as a function of applied detector reverse bias can be seen in
303 Fig. 10.
304



305
306 Fig. 10. Apparent multiplication factor of the 5.9 keV [55]Fe photopeak as a function of applied detector reverse bias for the
307 spectrometers (a) S_{200} and (b) S_{400}, at 20 °C. Unity gain was set to 19 V.
308
309 The spectra were energy calibrated by assuming a linear variation of output charge with energy and using the
310 positions of the so called zero energy noise peak and fitted 5.9 keV peak. The energy resolution (FWHM at
311 5.9 keV) was then calculated for each accumulated spectrum. Fig. 11 presents the energy resolution of each
312 spectrometer as a function of applied detector reverse bias.
313



314
315 Fig. 11. FWHM at 5.9 keV for the spectrometers (a) S_{200} and (b) S_{400} as a function of applied detector reverse bias at a shaping
316 time of 0.5 μs, and at 20 °C.
317
318 The improved charge collection efficiency, due to overcoming the punch-through voltage of the detectors, resulted
319 in an abrupt improvement in energy resolution (FWHM at 5.9 keV) of the spectroscopic systems around the punch
320 through voltage, as shown in Fig. 11. At V_R = 14 V, the FWHM at 5.9 keV was 2.00 keV ± 0.05 keV and 3.36 keV

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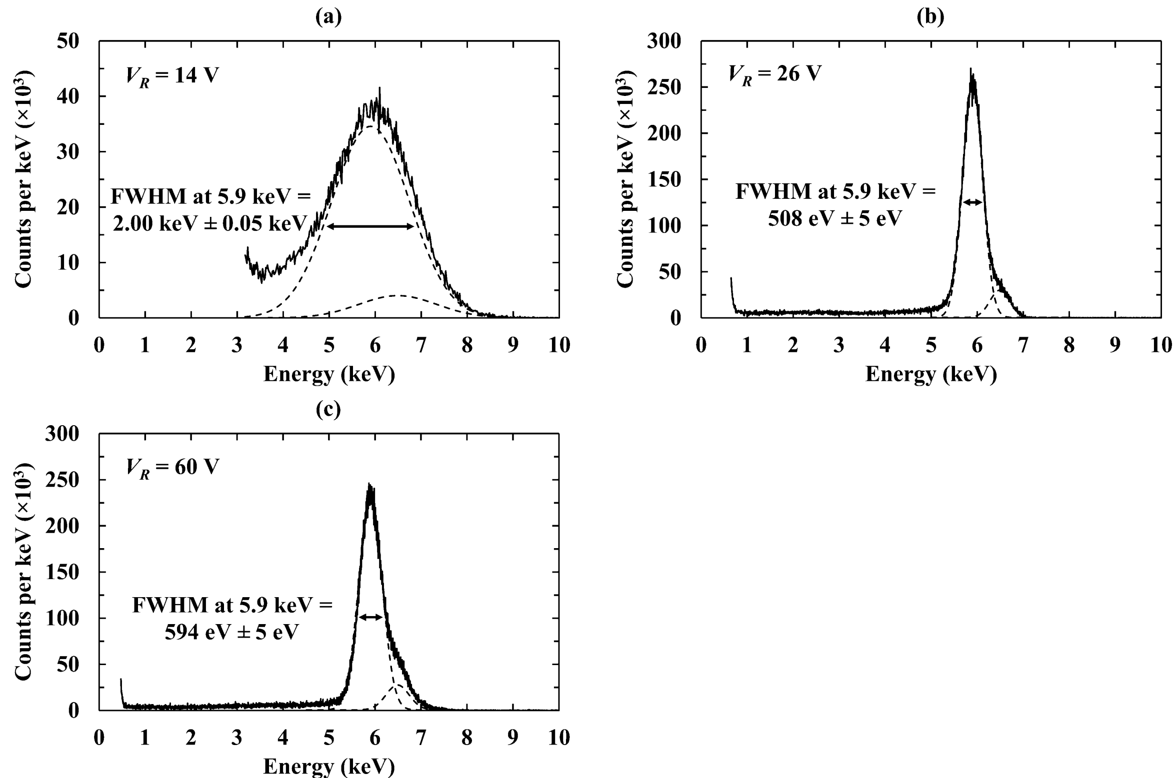
321 ± 0.05 keV for the spectrometers S_{200} and S_{400}, respectively. At V_R = 19 V, the FWHM at 5.9 keV was 534 eV
322 ± 5 eV and 653 eV ± 6 eV for S_{200} and S_{400}, respectively. The best measured energy resolution for the spectrometer
323 S_{200} was 508 eV ± 5 eV, achieved at V_R = 26 V, corresponding to an apparent multiplication factor of 1.1. The
324 best measured energy resolution for the spectrometer S_{400} was 603 eV ± 6 eV, achieved at V_R = 34 V,
325 corresponding to an apparent multiplication factor of 1.2. ^{55}Fe X-ray spectra accumulated with the spectrometers
326 S_{200} and S_{400} can be seen in Fig. 12 and Fig. 13 respectively.
327
328 Both the 200 μm diameter and 400 μm diameter GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD devices reported here, had
329 improved performance relative to the recently studied GaAs p^+-i-n^+ photodiodes [27]. At 20 ºC, energy resolutions
330 of 690 eV and 730 eV FWHM at 5.9 keV were reported for the 200 μm and 400 μm GaAs p^+-i-n^+ photodiode,
331 respectively [27]. The presently reported spectrometers also had improved performance compared to the
332 previously reported GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD spectrometers which had a FWHM at 5.9 keV = 1.08 keV at
333 an avalanche gain of M = 3.5 at room temperature [44].
334



335
336 Fig. 12. ^{55}Fe X-ray spectra accumulated with the 200 μm diameter detector based spectrometer, at 20 °C, a shaping time of
337 0.5 μs, and a reverse bias of (a) 14 V, (b) 26 V, and (c) 60 V. The fitted 5.9 keV (Mn Kα) and 6.49 keV (Mn Kβ) peaks have
338 been plotted (dashed lines). The accumulated spectra have been normalised into counts per keV in order to account for the
339 differing channel widths.
340

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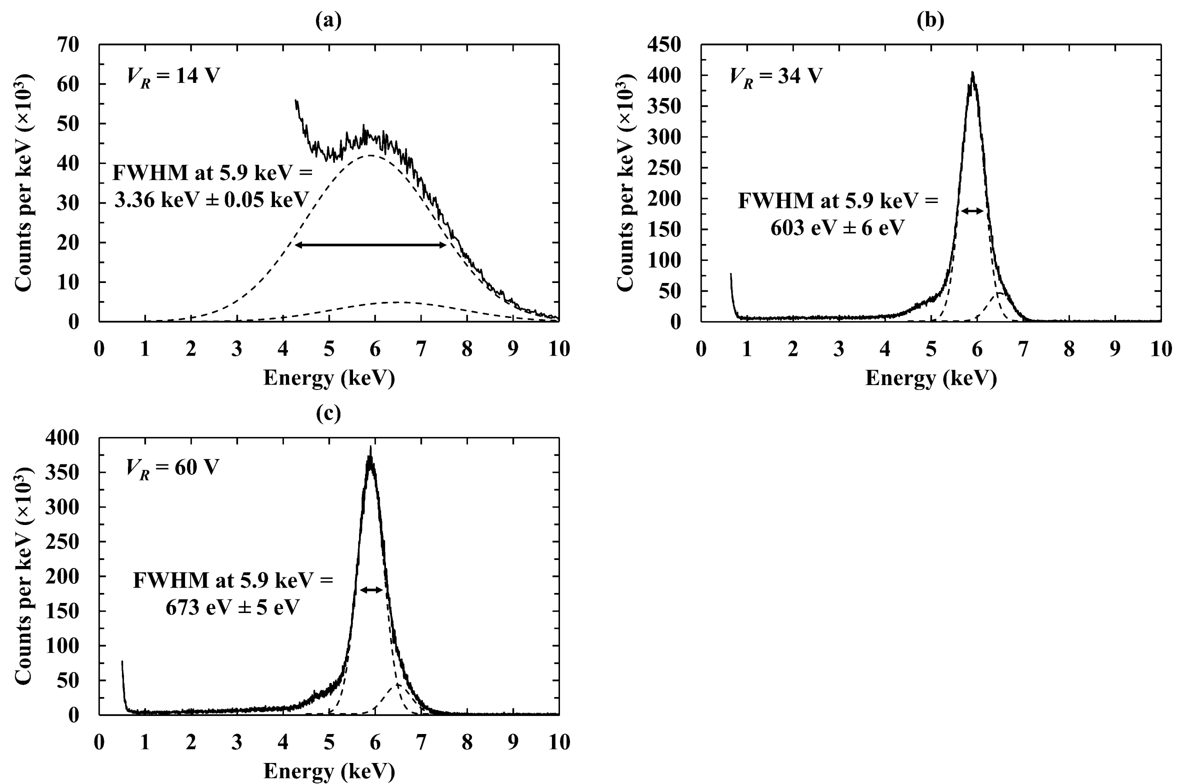

341
342 Fig. 13. ^{55}Fe X-ray spectra accumulated with the 400 μm diameter detector based spectrometer, at 20 °C, a shaping time of
343 0.5 μs, and a reverse bias of (a) 14 V, (b) 34 V, (c) and 60 V. The fitted 5.9 keV (Mn Kα) and 6.49 keV (Mn Kβ) peaks have
344 been plotted (dashed lines). The accumulated spectra have been normalised into counts per keV in order to account for the
345 differing channel widths.
346
347 **B. Origin of the secondary peak in the obtained X-ray spectra**
348 As could be seen in Fig. 13, at sufficiently high applied detector biases ($V_R \geq 17$ V), a small secondary peak
349 appears to the left (low energy) side of the fitted peaks. The secondary peak appears as a shoulder on the main
350 combined peak as they are not resolved from each other. This secondary peak arises as a consequence of the
351 GaAs layer not having a 100 % absorption efficiency for the X-ray photons (see Fig. 4b); some photons are
352 transmitted through the GaAs layer and absorbed in the $Al_{0.8}Ga_{0.2}As$ layers. This can be proven by consideration
353 of the electron-hole pair creation energies of each material.
354
355 Due to the difference in electron-hole pair creation energy of GaAs (ω_{GaAs} = 4.19 eV ± 0.03 eV at 20 °C [54]) and
356 $Al_{0.8}Ga_{0.2}As$ (ω_{AlGaAs} = 5.07 eV ± 0.08 eV at 20 °C [55]), the average number of charge carriers generated in each
357 material by the absorption of a photon of energy, E, also differs.
358
359 The ratio of the average numbers of charge carriers (N_{AlGaAs} for $Al_{0.8}Ga_{0.2}As$ and N_{GaAs} for GaAs) created by the
360 absorption of a photon of energy, E, in conjunction with the known electron-hole pair creation energy of GaAs,
361 can be used to determine ω_{AlGaAs}, where
362
363 $$\omega_{AlGaAs} = \omega_{GaAs}\left(\frac{N_{GaAs}}{N_{AlGaAs}}\right). \quad (4)$$
364

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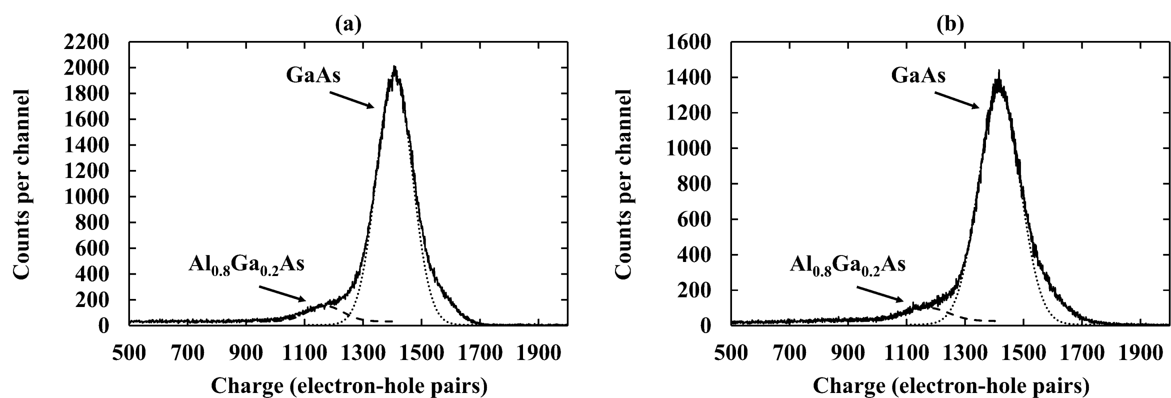
365 The primary and secondary peak of the spectra obtained with S_{400}, with the detector biased at 34 V and 60 V
366 respectively, were fitted with Gaussians for the ^{55}Fe 5.9 keV (Mn Kα) and 6.49 keV (Mn Kβ) peak emissions in
367 the accepted ratio [53], see Fig. 14; the relative detection efficiencies of the Mn Kα and Mn Kβ photons were also
368 taken into account [49]. The accepted value of ω_{GaAs} was then used in conjunction with Eq. 4 to calculate ω_{AlGaAs}.
369 With the detector biased at 34 V and 60 V, ω_{AlGaAs} was found to be 5.04 eV ± 0.08 eV and 5.06 eV ± 0.08 eV
370 respectively, which are in agreement with the accepted value (ω_{AlGaAs} = 5.07 eV ± 0.08 eV at 20 °C [55]). Thus,
371 it was demonstrated that the secondary, left shoulder, peak arises as a consequence of X-ray absorption in the
372 $Al_{0.8}Ga_{0.2}As$ layers by computation of the electron-hole pair creation energy of $Al_{0.8}Ga_{0.2}As$ from the spectra.
373
374 The secondary peak, clearly visible as a shoulder in Fig. 13 and Fig. 14 (400 µm diameter detector), was less
375 visible in Fig. 12 (200 µm diameter detector). This was attributed to the size difference between the two
376 investigated detectors, where the front face of the 400 µm diameter detector received four times more photons
377 than the 200 µm diameter detector.
378



379
380 Fig. 14. ^{55}Fe X-ray spectra accumulated with S_{400} at a reverse bias of (a) 34 V, and (b) 60 V. Charge calibration was achieved
381 using the positions of the zero energy noise peak of the preamplifier and the GaAs peak, together with the accepted ω_{GaAs}
382 value. The dashed and dotted lines are the fitted 5.9 keV (Mn Kα) peaks for the $Al_{0.8}Ga_{0.2}As$ and GaAs materials respectively;
383 the 6.49 keV Mn Kβ peaks were considered and included in the analysis but are not shown in the figures for clarity
384

385 **C. Noise analysis**

386 Four sources of noise influence the energy resolution of a charge-sensitive preamplifier coupled to a
387 semiconductor detector operating in avalanche mode, these are: the Fano noise, N_F [56]; incomplete charge
388 collection noise, R [22]; the electronic noise, A [57]; and the excess noise factor, N_x [44].
389
390 The Fano noise, assuming that X-ray photons incident on the reported detectors are absorbed only within the GaAs
391 absorption region, can be calculated using the equation
392

393 $$N_F = 2.355\omega_{GaAs}\sqrt{FE/\omega_{GaAs}}, \quad (5)$$

394
395 where F (0.12 [58]) is the Fano factor of GaAs and the other symbols have previously been defined. N_F was
396 calculated to be 13 e^- rms at 5.9 keV (128 eV FWHM at 5.9 keV) for the $GaAs/Al_{0.8}Ga_{0.2}As$ SAM APD. Since

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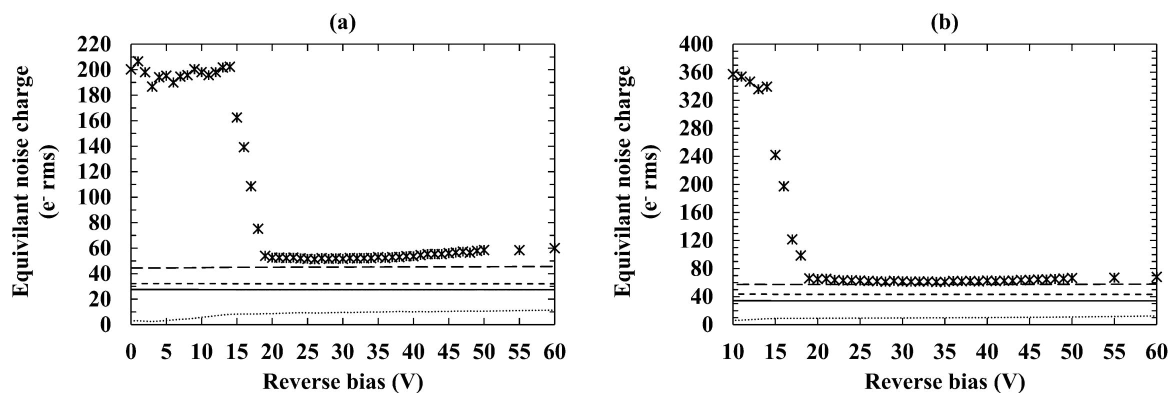
397 the calculated Fano noise was less than the reported spectrometers measured energy resolution, other noise
398 contributions were clearly affecting the spectroscopic system.
399
400 The electronic noise from the detector and preamplifier of a photodiode X-ray spectrometer consists of dielectric
401 noise, N_D, series white noise (including the induced gate drain current noise), N_{SW}, parallel white noise, N_{PW}, and
402 1/f series noise, $N_{1/f}$ [19]. Dielectric noise is induced by lossy dielectrics close to the input of the spectrometer
403 (e.g. the packaging of the preamplifier input Junction Field-Effect Transistor (JFET) and detector) [59]. Series
404 white noise is caused by capacitances at the input of the spectrometer (e.g. the input JFET (JFET capacitance
405 = 2 pF [60]) and the detector (see Fig. 2)) [27]. Parallel white noise is induced by leakage currents from the input
406 JFET (JFET leakage current = 1 pA [60]) and the detector (see Fig. 6) [27]. For a review of the various noise
407 components, the reader is directed to refs [59, 61, 62].
408
409 The dielectric noise, series white noise, parallel white noise, and 1/f series noise contributions were calculated for
410 each spectrometer (S_{200} and S_{400}) via the standard methods for semiconductor X-ray devices connected to
411 charge-sensitive preamplifiers [59]. The results can be seen in Fig. 15. It should be noted that, in the case of the
412 dielectric noise contributions, only a lower bound value could be calculated directly; whilst dielectric noise from
413 the JFET, feedback capacitor, and GaAs/$Al_{0.8}Ga_{0.2}As$ SAM APD device could be estimated [19, 25], additional
414 lossy dielectrics close to the preamplifier input could have also contributed to the noise. Similarly, due to the
415 prototype nature of the preamplifier, the presence of unknown capacitances may have also added to the series
416 white noise contribution. The dielectric noise and series white noise were thus considered in two parts: known
417 noise contributions and stray noise contributions. Subtracting the expected Fano noise and the electronic noise
418 contributions (1/f noise, parallel white noise, known series white noise (including induced gate drain current
419 noise), and known dielectric noise) from the measured FWHM in quadrature, yields a combination of stray series
420 white noise, stray dielectric noise, incomplete charge collection noise, excess noise, and possibly stray parallel
421 white noise contributions (so called the remaining noise contribution). Changes in detector capacitance were
422 included in the known series white noise and known dielectric noise calculations, whilst the JFET was considered
423 to contribute a constant capacitance (2 pF [60]) to both calculations. A constant JFET leakage current (1 pA [60])
424 was included in the known parallel white noise calculation.
425



426
427 Fig. 15. Calculated noise contributions of the spectrometers (a) S_{200} and (b) S_{400} as a function of applied detector reverse bias
428 at a shaping time of 0.5 µs, and at 20 °C: total noise (stars); sum of the calculated noise contributions (long dashed line); known
429 series white noise (short dashed line); known dielectric noise (solid line); calculated parallel white noise (dotted line).

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430
431 As per Fig. 15, the total sum of the calculated noise contributions (Fano noise, 1/f noise, known series white noise,
432 known dielectric noise, and parallel white noise, added in quadrature) did not account for the measured total noise
433 (FWHM at 5.9 keV) of the 200 µm and 400 µm diameter detector based spectroscopic systems. At an applied
434 detector reverse bias of 10 V, the total noise of each system was 198 e^- rms ± 5 e^- rms, and 357 e^- rms ± 5 e^- rms
435 for S_{200} and S_{400}, respectively. At the same applied bias (10 V), the total sum of the calculated noise contributions
436 was 44.7 e^- rms ± 0.4 e^- rms and 57.4 e^- rms ± 0.2 e^- rms, respectively. This discrepancy was attributed in part to
437 incomplete charge collection noise, where charge carriers cannot readily travel through the $Al_{0.8}Ga_{0.2}As$ junction
438 before the punch-through voltage [18] (see Section IV.A). Stray dielectric noise, stray series white noise, and any
439 stray parallel white noise contributions, arising from the spectroscopic systems, would have also contributed to
440 the measured total noise.
441
442 As the applied detector reverse bias was further increased (14 V < V_R < 19 V), the measured total noise (FWHM
443 at 5.9 keV) of each spectrometer improved (reduced). At 19 V applied detector reverse bias, the total noise was
444 54.0 e^- rms ± 0.5 e^- rms and 66.0 e^- rms ± 0.6 e^- rms for the spectrometers S_{200} and S_{400}, respectively. The total
445 sum of the calculated noise contributions, at the same applied detector reverse bias (19 V), was 45.0 e^- rms
446 ± 0.4 e^- rms and 57.3 e^- rms ± 0.2 e^- rms for the spectrometers S_{200} and S_{400}, respectively. This, in part, indicated
447 that incomplete charge collection noise reduced as the punch-through voltage (≈ 14 V) was exceeded.
448
449 At the optimal applied detector reverse bias for each spectrometer (26 V and 34 V for the spectrometers S_{200} and
450 S_{400} respectively), the measured apparent noise was 51.4 e^- rms ± 0.5 e^- rms and 61.0 e^- rms ± 0.6 e^- rms for the
451 spectrometers S_{200} and S_{400}, respectively. The total sum of the calculated noise contributions, at the same optimal
452 applied detector reverse bias (26 V and 34 V for S_{200} and S_{400} respectively), was 45.1 e^- rms ± 0.4 e^- rms and
453 57.4 e^- rms ± 0.2 e^- rms for the spectrometers S_{200} and S_{400}, respectively. The apparent decrease in remaining
454 noise contribution between 19 V and the optimal applied detector reverse bias of each spectrometer indicated a
455 net benefit due to avalanche multiplication.
456
457 At the maximum applied detector reverse bias (60 V) an apparent noise of 60.1 e^- rms ± 0.5 e^- rms and 68.0 e^- rms
458 ± 0.5 e^- rms was measured for S_{200} and S_{400}, respectively. The total sum of the calculated noise contributions at
459 the same applied detector reverse bias (60 V), was 45.6 e^- rms ± 0.4 e^- rms and 57.9 e^- rms ± 0.2 e^- rms,
460 respectively. The increased discrepancy between the measured apparent noise (FWHM at 5.9 keV) and the total
461 sum of the calculated noise contributions may have arisen from a larger than expected parallel white noise at high
462 biases or from increasing excess noise due to avalanche multiplication.
463

464 D. Improvements in energy resolution due to avalanche multiplication

465 In order to determine whether avalanche multiplication affected the energy resolution of the reported
466 spectrometers, the measured energy resolution (FWHM at 5.9 keV) was compared to the expected non-avalanche
467 energy resolution of each spectrometer.
468

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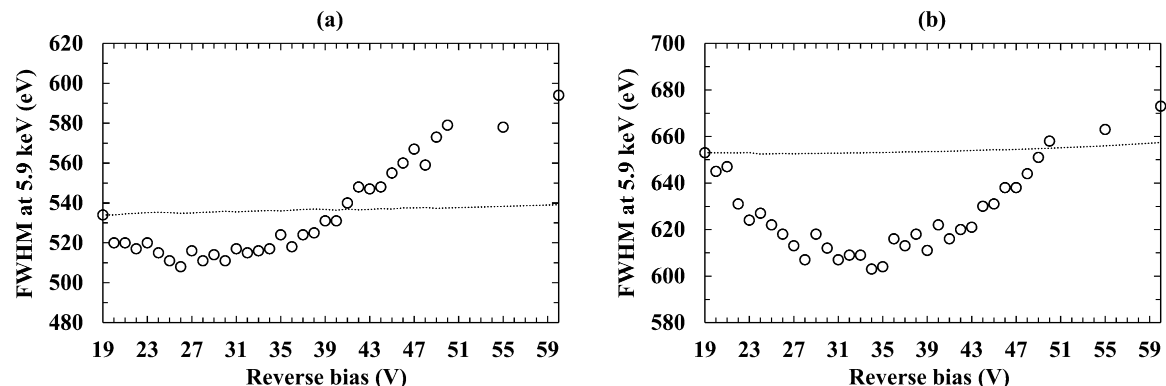
469 The expected non-avalanche energy resolution was calculated by assuming incomplete charge collection noise
470 became negligible at $V_R \geq 19$ V, avalanche multiplication was not present at $V_R \leq 19$ V, and any stray noises
471 contributing to the remaining noise contribution (see Section IV.C) were independent of applied reverse bias.
472 Given these assumptions, the remaining noise contribution at $V_R = 19$ V for both spectrometers represents the
473 non-avalanche mode remaining noise contribution across the applied reverse bias range (19 V $\leq V_R \leq$ 60 V). The
474 remaining noise contribution at $V_R = 19$ V was calculated to be 29.8 e^- rms ± 1.1 e^- rms and 32.8 e^- rms ± 1.6 e^- rms
475 for S_{200} and S_{400}, respectively.
476
477 Adding in quadrature the remaining noise contribution at $V_R = 19$ V to the calculated known noise contributions
478 at each investigated applied detector reverse bias yields the expected non-avalanche energy resolution as a
479 function of applied reverse bias within the range 19 V $\leq V_R \leq$ 60 V. The expected non-avalanche energy resolution,
480 and measured avalanche energy resolution, at each applied reverse bias for the spectrometers S_{200} and S_{400}, can be
481 seen in Fig. 16.
482
483 At an applied detector reverse bias of 26 V (M = 1.1), an expected non-avalanche energy resolution of 535 eV
484 ± 7 eV FWHM at 5.9 keV was calculated for the spectrometer S_{200}. Given the same spectrometer and the same
485 applied detector reverse bias (26 V), an energy resolution of 508 eV ± 5 eV was measured. As for the spectrometer
486 S_{400}, at an applied detector reverse bias of 34 V (M = 1.2), an expected non-avalanche energy resolution of 653 eV
487 ± 8 eV FWHM at 5.9 keV was calculated. At the same applied detector reverse bias (34 V), an energy resolution
488 of 603 eV ± 6 eV was measured. As such, it can be concluded that a noticeable benefit from small avalanche
489 multiplication gains (M ≤ 1.4) was measured. This conclusion is further supported by recent results from non-
490 avalanche GaAs p^+-i-n^+ photodiodes. Those photodiodes were of identical design to the devices reported here
491 except that they did not have the AlGaAs avalanche layer [27]. At 20 ºC, energy resolutions of 690 eV and 730 eV
492 FWHM at 5.9 keV were reported for the 200 µm and 400 µm GaAs p^+-i-n^+ photodiode, respectively [27]. Thus
493 it has been demonstrated that the addition of an avalanche layer can lead to improvement of the energy resolution
494 in X-ray photodiodes.
495



496
497 Fig. 16. Expected non-avalanche FWHM at 5.9 keV, assuming no incomplete charge collection noise at $V_R \geq 19$ V (dotted
498 line), as a function of applied detector reverse bias to the spectrometers (a) S_{200} and (b) S_{400}. The measured FWHM at 5.9 keV
499 (circles) has been included.
500
501 **V. CONCLUSION**

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502 A 200 µm diameter and a 400 µm diameter, custom-made, circular GaAs/$Al_{0.8}Ga_{0.2}As$ separate absorption and
503 multiplication (SAM) X-ray photodiode have each been characterised at room temperature (20 °C), and
504 investigated for their response to ^{55}Fe X-rays (Mn Kα = 5.9 keV; Mn Kβ = 6.49 keV) source. Each device
505 consisted of a 10 µm thick GaAs absorption layer and a 0.1 µm thick $Al_{0.8}Ga_{0.2}As$ multiplication layer.
506
507 Capacitance measurements indicated a punch-through voltage of ≈ 14 V for both devices. A capacitance of
508 1.12 pF and 2.20 pF (each ± 0.4 fF) was measured for the 200 µm and 400 µm diameter devices respectively at
509 the maximum applied reverse bias (60 V). The capacitance measurements indicated that both devices were fully
510 depleted at $V_R \geq 50$ V, reporting a depletion width consistent with growth specifications (see Table 1).
511
512 The best measured energy resolution achieved at 20 °C for the spectrometer S_{200} was 508 eV ± 5 eV FWHM at
513 5.9 keV, at an applied detector reverse bias of 26 V, corresponding to an apparent multiplication factor of 1.1.
514 The best measured energy resolution at 20 °C for the spectrometer S_{400} was 603 eV ± 6 eV FWHM at 5.9 keV, at
515 an applied detector reverse bias of 34 V, corresponding to an apparent multiplication factor of 1.2. Further
516 increasing the applied detector reverse bias increased (worsened) the energy resolution (e.g. 594 eV ± 5 eV
517 FWHM at 5.9 keV and 673 eV ± 5 eV FWHM at 5.9 keV for the spectrometers S_{200} and S_{400} respectively, at an
518 applied detector reverse bias of 60 V). This indicated that any benefits from further increasing avalanche gain
519 were exceeded by increases in excess noise and/or parallel white noise.
520
521 In order to determine whether avalanche multiplication affected the energy resolution of the spectrometers, the
522 measured energy resolution (FWHM at 5.9 keV) was compared to the expected non-avalanche energy resolution
523 of each spectrometer (see Section IV.D). The results indicated a noticeable benefit from small avalanche
524 multiplication gains ($M \leq 1.4$). At an applied detector reverse bias of 26 V ($M = 1.1$), an expected non-avalanche
525 energy resolution of 535 eV FWHM at 5.9 keV was calculated for the spectrometer S_{200}. Given the same
526 spectrometer and the same applied detector reverse bias (26 V), an energy resolution of 508 eV FWHM at 5.9 keV
527 was measured. Similarly, at an applied detector reverse bias of 34 V ($M = 1.2$), an expected non-avalanche energy
528 resolution of 653 eV FWHM at 5.9 keV was calculated for the spectrometer S_{400}. Given the same spectrometer
529 and the same applied detector reverse bias (34 V), an energy resolution of 603 eV FWHM at 5.9 keV was
530 measured. The results were supported further (and set in context) by a recent study using non-avalanche GaAs
531 p^+-i-n^+ photodiodes which did not have an AlGaAs avalanche layer [27]. At 20 ºC, energy resolutions of 690 eV
532 and 730 eV FWHM at 5.9 keV were reported for the 200 µm and 400 µm GaAs p^+-i-n^+ photodiodes, respectively
533 [27]. The results indicated that introducing a separate AlGaAs multiplication layer can be beneficial to GaAs
534 photodiodes.
535
536 The energy resolution (FWHM at 5.9 keV) reported here is the best so far reported for GaAs/$Al_xGa_{1-x}As$ SAM
537 APD X-ray spectrometers at room temperature. Energy resolutions of 1.08 keV FWHM at 5.9 keV [44] and
538 900 eV FWHM at 13.96 keV have been reported previously [18]. The measured energy resolution was also better
539 than recently investigated non-avalanche $Al_xGa_{1-x}As$ detector based X-ray spectrometers, where an energy
540 resolution of 760 eV FWHM at 5.9 keV was reported at room temperature [26]. However, the energy resolutions
541 reported here are modest when compared to the best reported results for the best non-avalanche GaAs based X-ray

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spectrometers (266 eV [3] and 300 eV [28] FWHM at 5.9 keV at room temperature) and the best Si based X-ray spectrometers (141 eV FWHM at 5.9 keV by [63] and 134 eV FWHM at 5.9 keV [64]), when those detectors are coupled to ultra-low-noise electronics better than those used for the investigations reported in the present article.

Despite the achieved energy resolutions of the presently reported GaAs/$Al_xGa_{1-x}As$ SAM APDs being not yet as good when compared to other more developed materials, X-ray spectrometers with modest energy resolutions can still provide important scientific contributions. For example, D-CIXS aboard SMART-1 [65] had an energy resolution of 420 eV FWHM at 4.5 keV [66] and measured, for the first time, Ti Kα (4.51 keV) X-ray fluorescence on the lunar surface [66]. An X-ray spectrometer with similar or slightly improved energy resolution, that is also radiation hard and temperature tolerant, could therefore find utility in future space science missions to harsh environments.

In future, the temperature dependence of the devices' electrical characteristics and X-ray detection performance will be studied. New SAM APDs will be fabricated in array format and characterised. Devices with thicker absorption regions will be grown to improve the quantum efficiency (thereby also reducing/eliminating the observed secondary photopeak as discussed in Section IV.B), and staircase avalanche regions implemented. A thicker absorption region would also reduce the detector capacitance, reducing the series white noise, and potentially improving the energy resolution of the spectrometer.

AUTHORS' DATA STATEMENT

The data that supports the findings of this study are available within the article [and its supplementary material].

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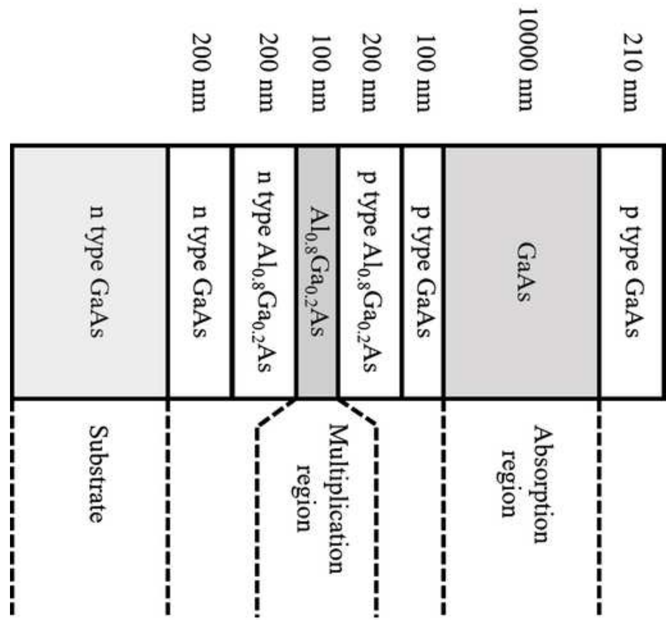

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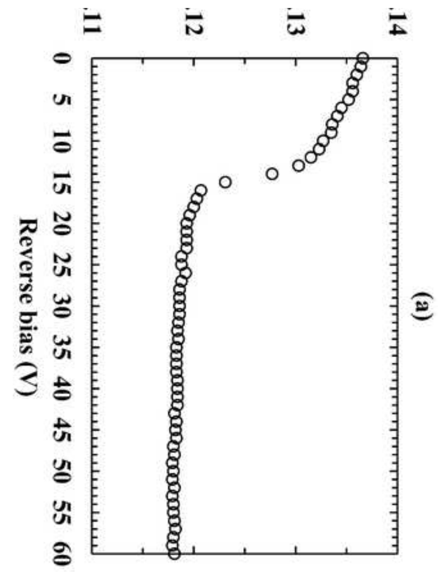

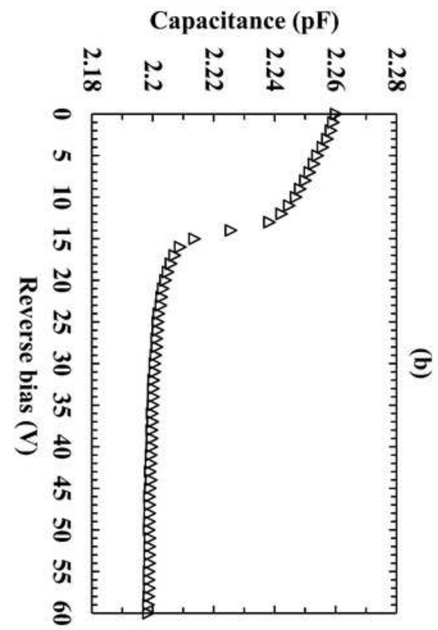

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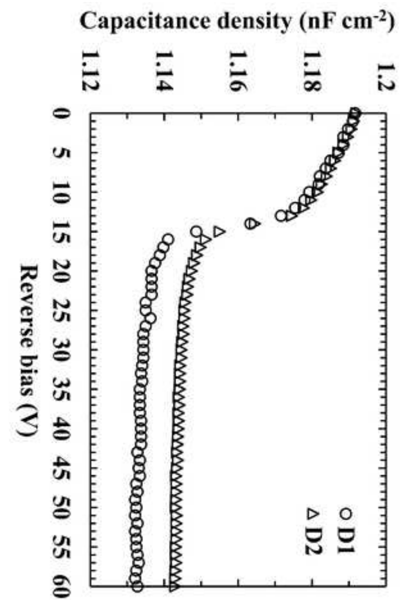

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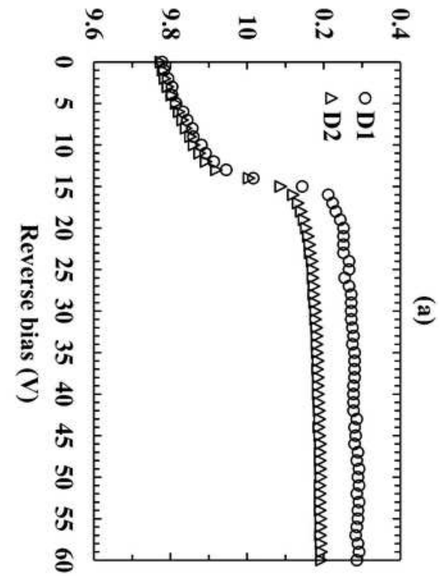

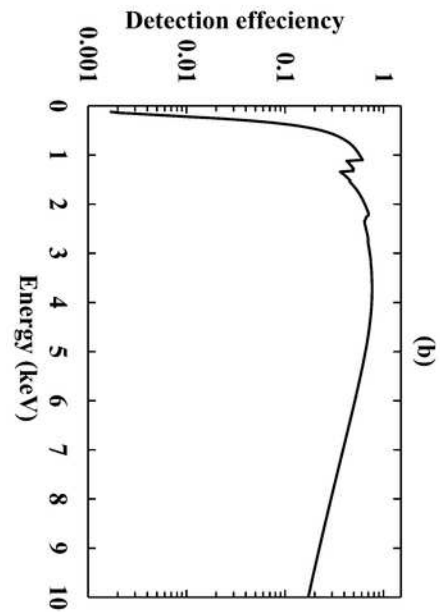

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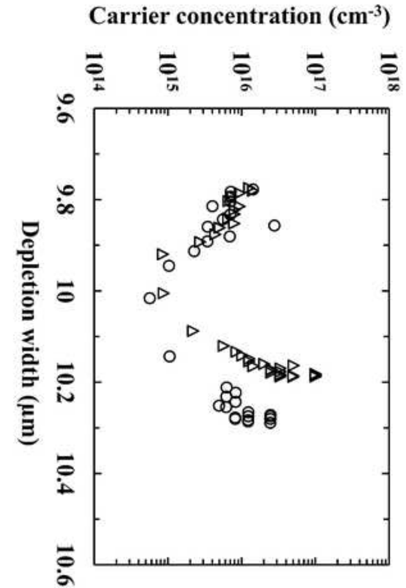

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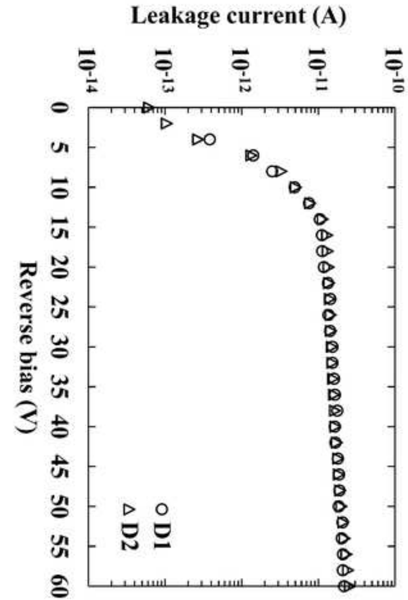

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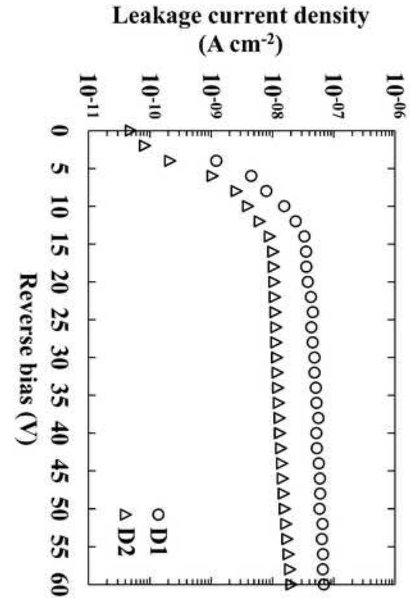

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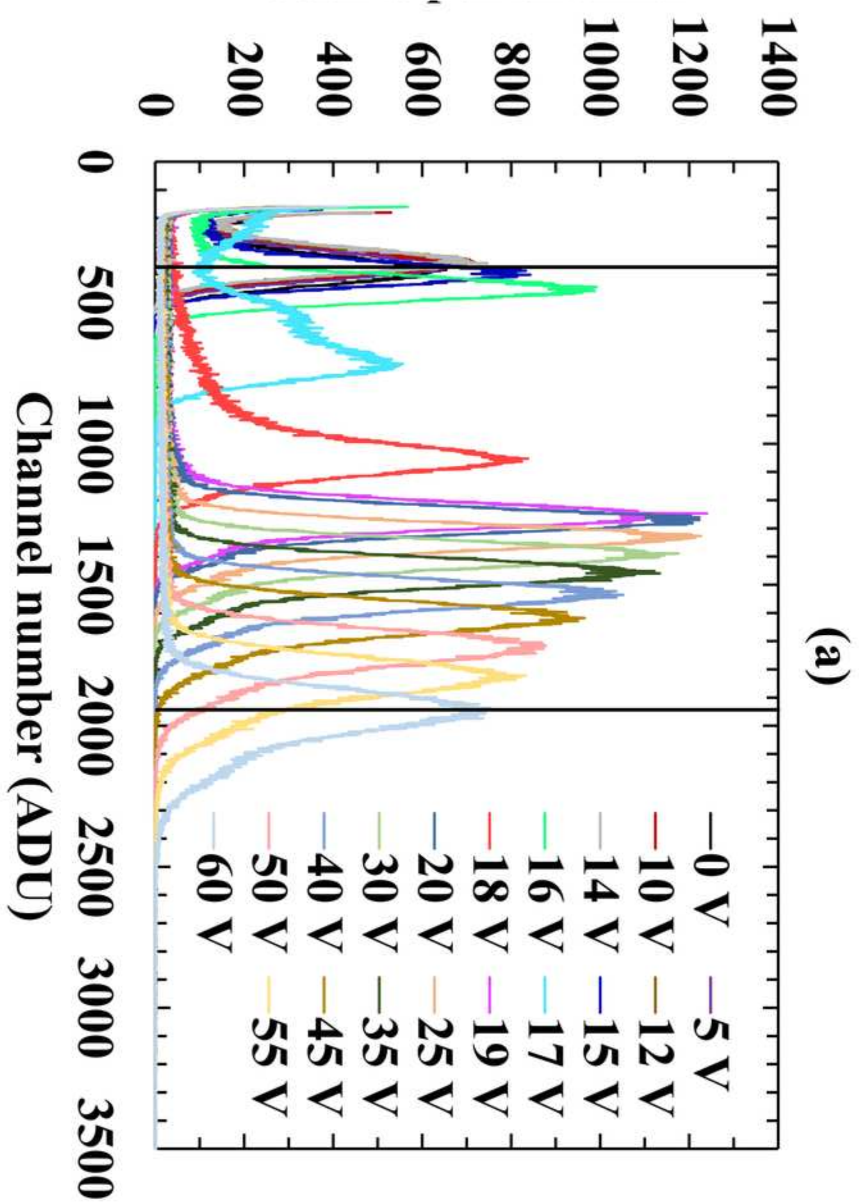

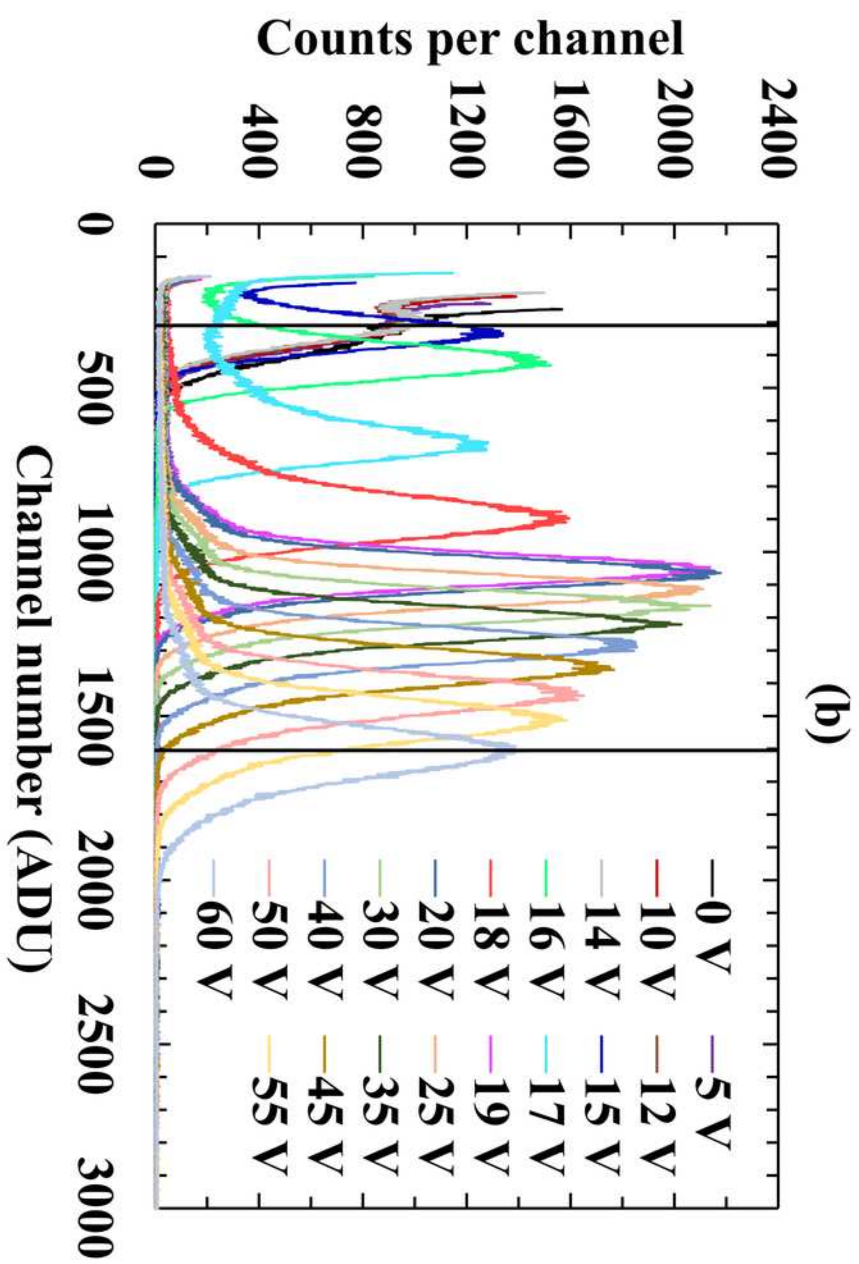

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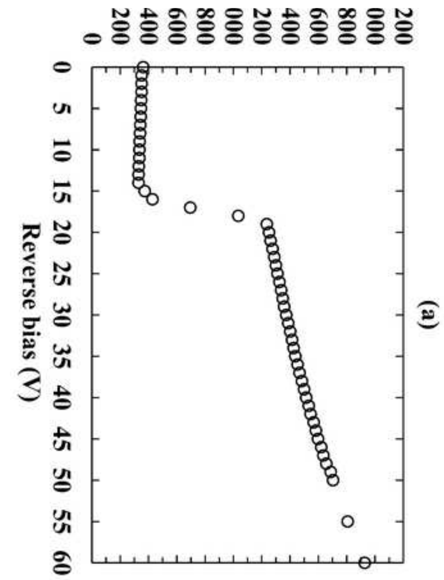

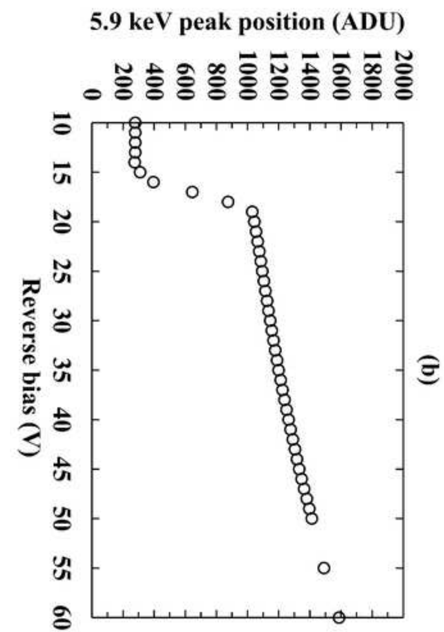

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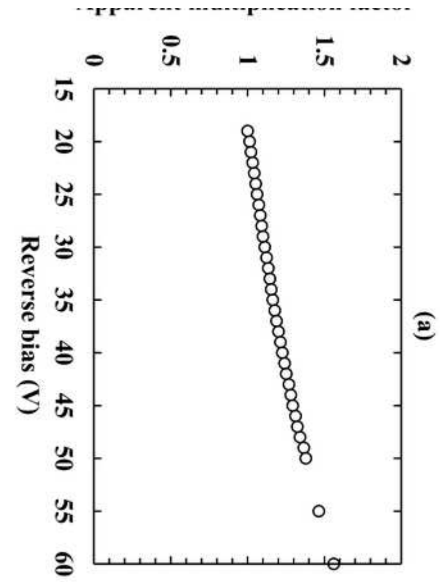

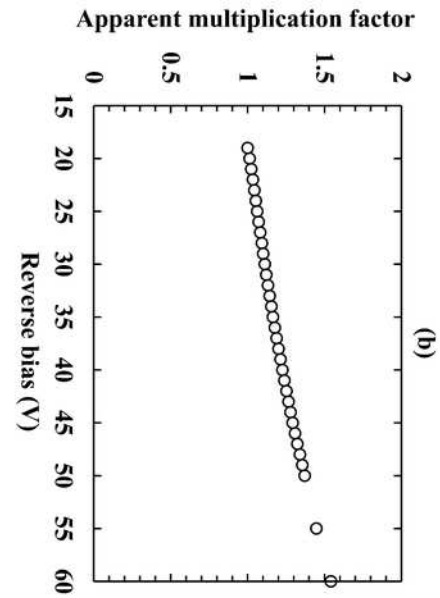

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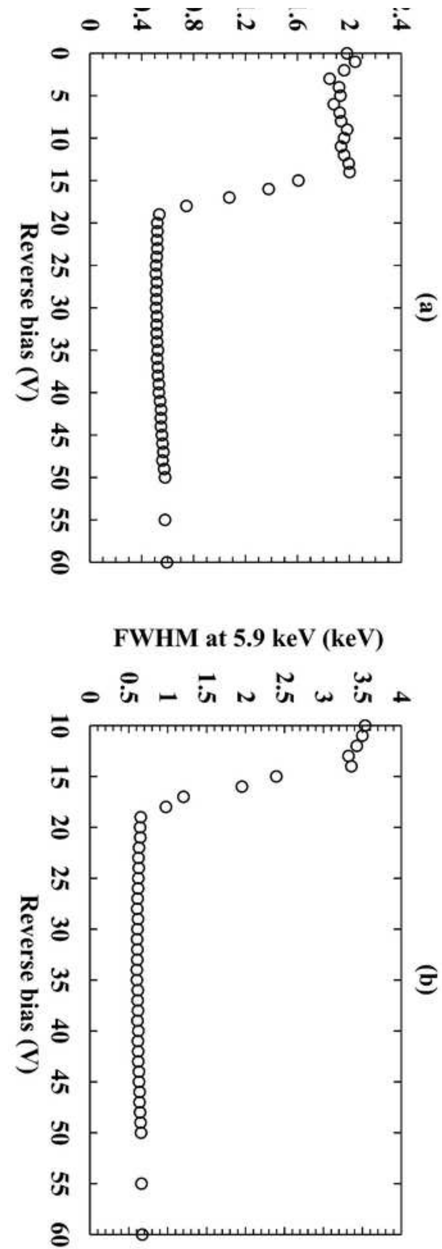

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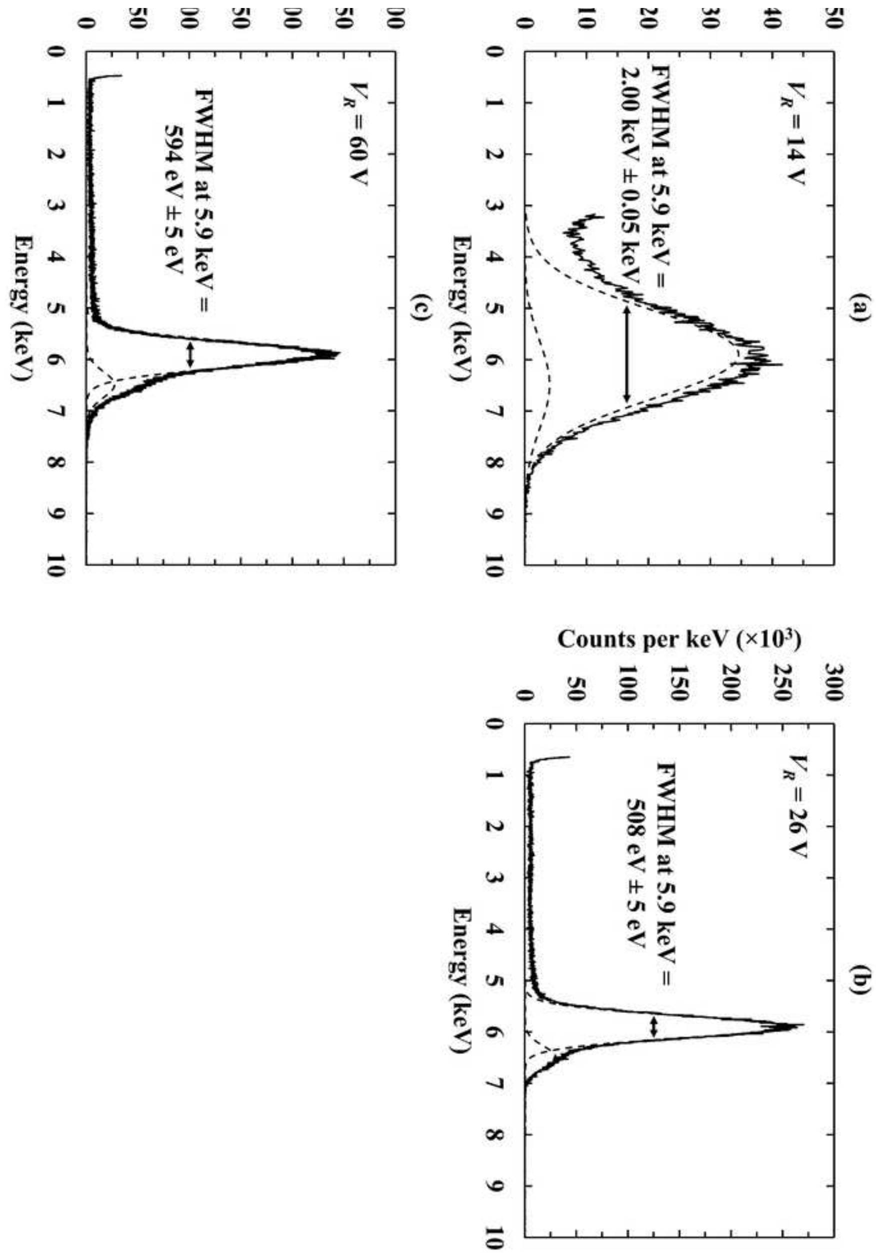

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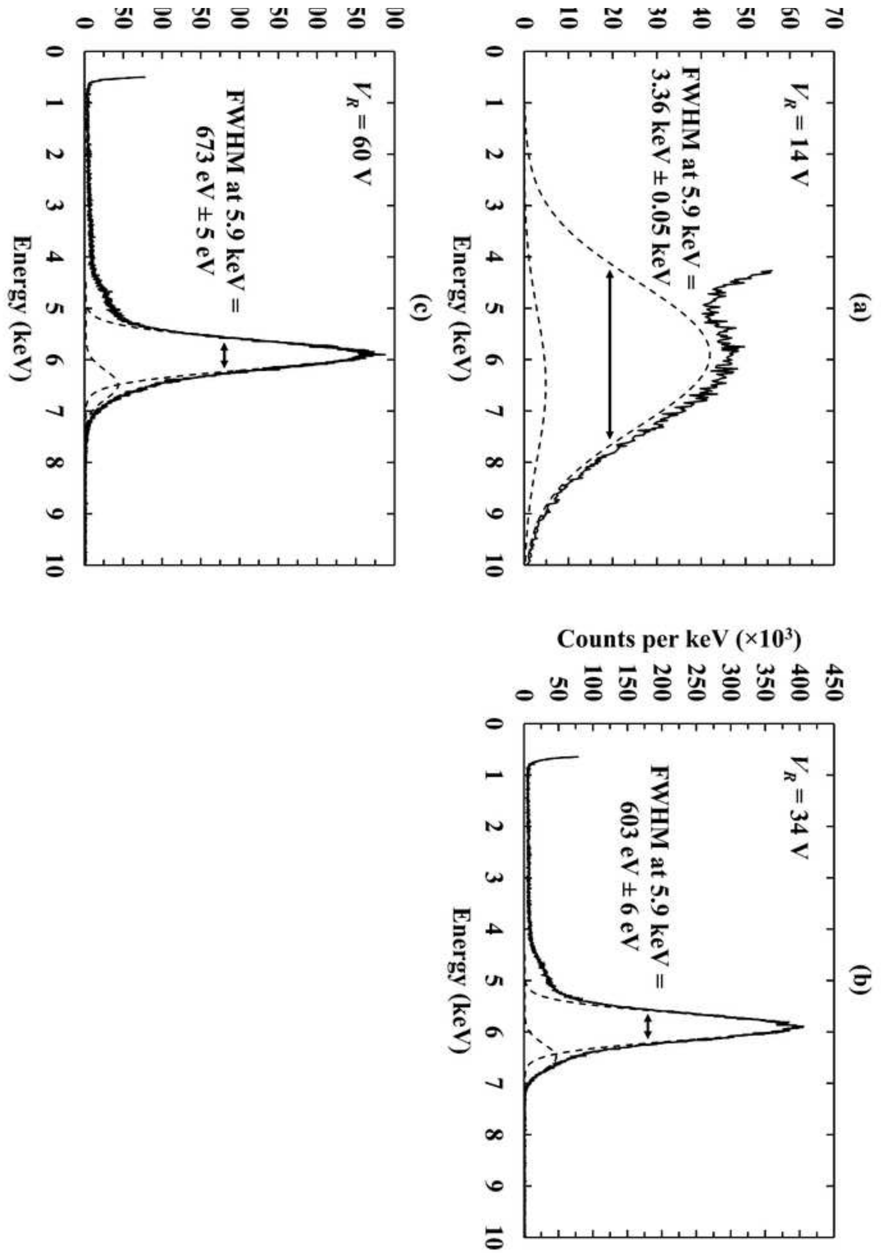

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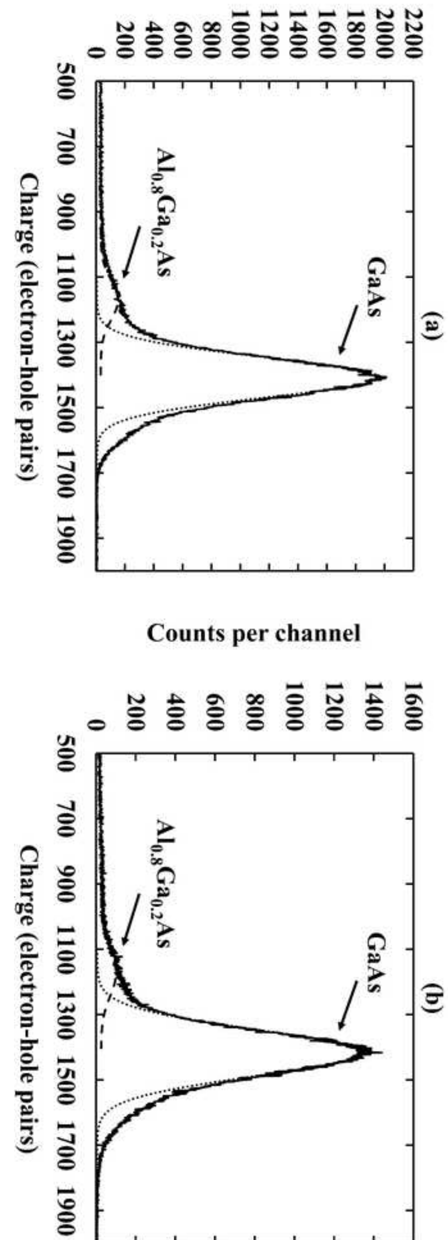

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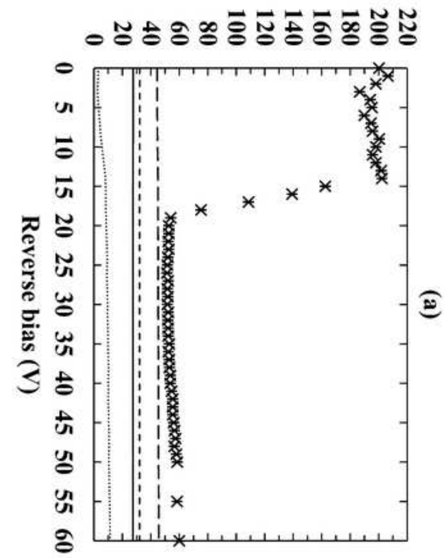

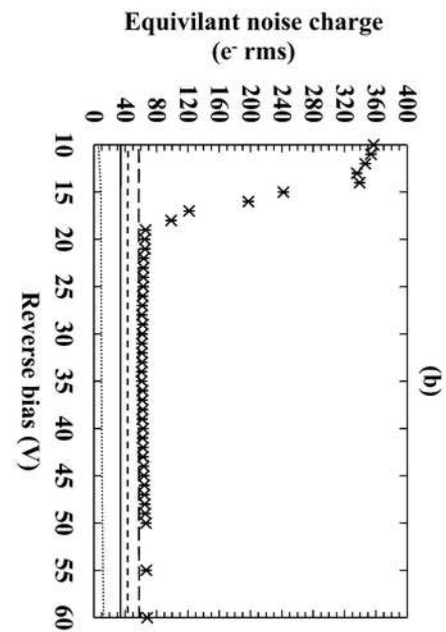

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