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Technical Notes

AlInP X-ray photodiodes without incomplete charge collection noise

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ABSTRACT

Previously, an $Al_{0.52}In_{0.48}P$ p^+-i-n^+ spectroscopic photon counting X-ray photodiode with 2 μm thick i layer (200 μm diameter) was shown to suffer from energy-dependent incomplete charge collection noise (Lioliou et al., 2019). Subsequent measurements on a larger (400 μm diameter) $Al_{0.52}In_{0.48}P$ p^+-i-n^+ photodiode (reported here) revealed the presence of even greater incomplete charge collection noise. Given these findings, an expectation would have been that thicker $Al_{0.52}In_{0.48}P$ structures (which would be required for efficient absorption of all but the softest X-rays) would have a greater incomplete charge collection noise contribution, thus suggesting that thick $Al_{0.52}In_{0.48}P$ photodiodes may be of limited practicality as high performance detectors for photon counting X-ray spectroscopy. However, two new $Al_{0.52}In_{0.48}P$ p^+-i-n^+ photodiodes (with 6 μm i layers) were fabricated from material grown by the same technique (metalorganic vapour phase epitaxy) in the same reactor, and are now shown here to exhibit no signs of detectable incomplete charge collection noise under the illumination of X-ray photons of energy 4.95 keV to 21.17 keV. As such, now that greater experience has been built with $Al_{0.52}In_{0.48}P$, concerns about incomplete charge collection noise in X-ray detectors made from the material appear to have been unwarranted; the path towards thick $Al_{0.52}In_{0.48}P$ X-ray detectors is now clear.

$Al_{0.52}In_{0.48}P$ has been proposed recently as a detector material for X-ray photon counting spectroscopy at high (>20 °C) temperatures [1–5]. X-ray photodiodes made from the material have been shown to have low leakage current densities even at thermal extremes (e.g. <0.6 nA/cm^2 at an applied electric field of 75 kV/cm at 100 °C [3]). This key attribute of $Al_{0.52}In_{0.48}P$, which arises in part from its wide bandgap (2.31 eV at room temperature [6]), is necessary for the development of high temperature tolerant X-ray spectrometers which are required for future use in space and terrestrial applications. Although 4H–SiC X-ray detectors have been shown to have excellent high temperature performance [7,8], the larger X-ray linear absorption coefficient of $Al_{0.52}In_{0.48}P$ (1324 cm^{-1} at 5.9 keV; 12 cm^{-1} at 59.5 keV [9]) compared with that of 4H–SiC (355 cm^{-1} and 0.3 cm^{-1}, at the same energies [9]) provides the opportunity for $Al_{0.52}In_{0.48}P$ X-ray detectors to be ~4 and ~40 times thinner than SiC detectors whilst maintaining the same detection efficiency [10]. Furthermore, the lower average electron–hole pair creation energy of $Al_{0.52}In_{0.48}P$ (5.34 eV at 20 °C [11] cf. 7.8 eV for 4H–SiC [12]) suggests that better Fano-limited energy resolutions [10,13] may be achievable in future, even given the likelihood that $Al_{0.52}In_{0.48}P$ has a slightly larger (worse) Fano factor [14] than 4H–SiC (0.10 [8]) since it is a ternary material.

However, shortly after its debut as a material for X-ray detection [1,2], $Al_{0.52}In_{0.48}P$ was reported to exhibit incomplete charge collection noise; a 200 μm diameter circular mesa $Al_{0.52}In_{0.48}P$ X-ray photodiode was found to give rise to 36 e$^-$ rms incomplete charge collection noise at 21.17 keV [4]. Incomplete charge collection noise [15–17] arises due to crystal imperfections (impurity atoms, vacancies, dislocations) which can act as trapping or recombination centres, resulting in the loss of generated carriers from the radiation detected. Any such incomplete charge collection degrades the energy resolution of a spectrometer employing a detector which suffers from such effects. The noise contribution from incomplete charge collection is expected to result in a non-symmetric photopeak since it is a non-Gaussian noise source, but when it is relatively small compared with other noise sources contributing to the achieved energy resolution of the spectrometer, it is expedient to approximate its contribution to be Gaussian [18]. Thus, this gives rise to the semi-empirical formula for the energy resolution (full width at half maximum, *FWHM*) of a photon counting X-ray spectrometer that suffers from incomplete charge collection noise,

$$FWHM\ [\mathrm{eV}] = 2.355\sqrt{FE\omega + \left(\frac{A}{2.355}\right)^2 + aE^b}, \quad (1)$$

where the first term under the square root is the Fano noise (F is the Fano factor, E is the incident X-ray energy, ω is the electron-hole pair creation energy), A is the electronic noise, and the third term is the incomplete charge collection noise from the detector, with a and b being semi-empirical constants determined by best-fitting [19]. Assuming a Fano factor of 0.13 (i.e. equal to that of another wide bandgap semiconductor, $In_{0.5}Ga_{0.5}P$ [20], since no measurement of the Fano

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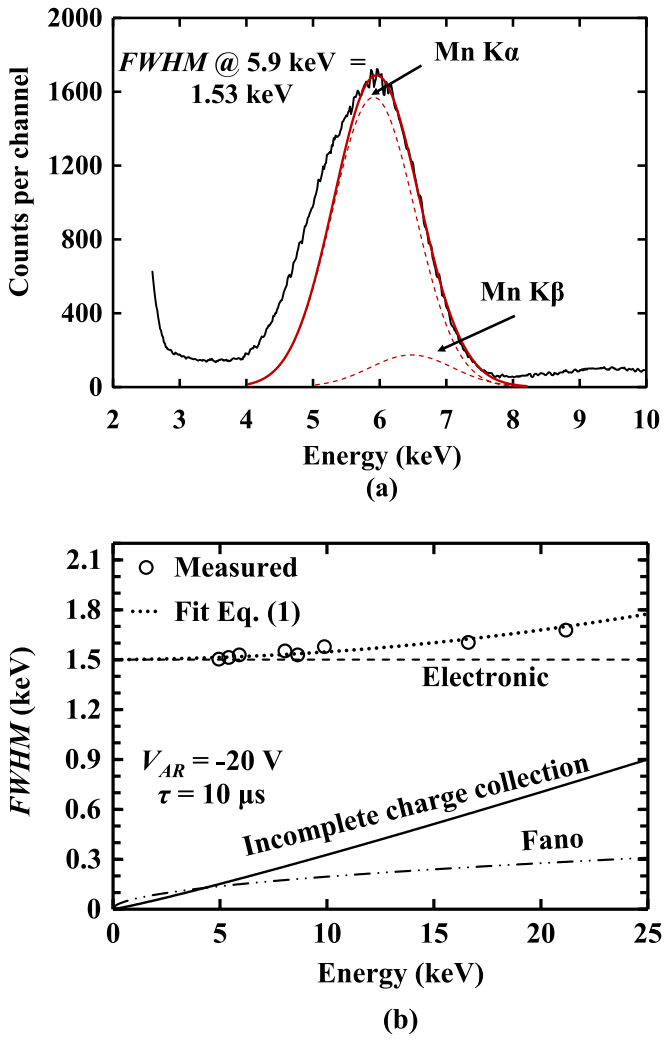

Fig. 1. (a) Mn spectrum along with the fitted Gaussians (red dashed lines), at Kα and Kβ lines, and (b) measured *FWHM* (open circles) of the primary X-ray fluorescence peak of eight different calibration foils along with the predicted *FWHM* (dotted line) from Eq. (1), the calculated Fano noise (long dash double dotted line), and the derived electronic noise (dashed line) and incomplete charge collection noise (solid line), of the 400 μm diameter $Al_{0.52}In_{0.48}P$ photodiode (2 μm i layer) based X-ray spectrometer.

factor in $Al_{0.52}In_{0.48}P$ has been reported yet), and given $Al_{0.52}In_{0.48}P$'s X-ray photon initiated electron hole pair creation energy of 5.31 eV at 30 °C [11], the expected Fano noise of $Al_{0.52}In_{0.48}P$ was calculated to increase from 138 eV at 4.95 keV to 285 eV at 21.17 keV.

Measurements subsequent to those of Ref. [4] using a larger (400 μm diameter) photodiode fabricated from the same 2 μm i layer wafer revealed the presence of an even more significant, non-Gaussian, contribution of incomplete charge collection noise than was presented in Ref. [4]; an example X-ray fluorescence spectrum, of an Mn calibration foil, can be seen in Fig. 1(a). Gaussians were fitted to the right-hand side of the primary peaks of the accumulated spectra, i.e. excluding part of the contribution of the incomplete charge collection noise. The measured *FWHM* as a function of energy, the calculated Fano noise, and the extracted electronic noise and incomplete charge collection noise from fitting of Eq. (1) to the measured *FWHM*, can be seen in Fig. 1(b). The presence of 750 eV (60 e^- rms) at 21.17 keV incomplete charge collection noise was suggested, excluding the left-hand side broadening of the photopeaks.

Whilst incomplete charge collection noise is a relatively common feature of many wide bandgap compound semiconductor detectors (e.g. CdZnTe [21], TlBr [22], and semi-insulating 4H–SiC [23]), it can be difficult to eliminate and thus its detection can discourage adoption of detectors made from the material, particularly when other competing detector media have shown negligible incomplete charge collection noise (e.g. epitaxial 4H–SiC [23] and InGaP [20]).

Following the detection of significant incomplete charge collection noise in early $Al_{0.52}In_{0.48}P$ X-ray detectors ([4] and Fig. 1), as part of efforts to better understand the noise mechanisms, a new epitaxial $Al_{0.52}In_{0.48}P$ p^+-i-n^+ structure was grown by metalorganic vapour phase epitaxy on a n^+ GaAs substrate. The structure had a thicker (3×, i.e. 6 μm) i layer than the thickest $Al_{0.52}In_{0.48}P$ X-ray photodiodes reported previously [4]. The epitaxial p^+ (5×10^{17} cm^{-3}) and n^+ (2×10^{18} cm^{-3}) layers had thicknesses of 0.2 μm and 0.1 μm, respectively. A 0.01 μm thick p^+ GaAs (1×10^{19} cm^{-3}) cap was grown atop the $Al_{0.52}In_{0.48}P$ p^+ layer to facilitate adhesion of a top quasi-annular Ohmic contact (20 nm of Ti and 200 nm of Au). An n side planar contact, 20 nm of InGe and 200 nm of Au, was formed on reverse of the substrate. Mesa photodiodes of two different diameters (217 μm and 409 μm) were fabricated from the material by wet chemical etching: 1:1:1 $K_2Cr_2O_7$:HBr:CH_3COOH followed by 10 s in 1:8:80 H_2SO_4:H_2O_2:H_2O.

The doping density of the epitaxial i layer was determined by measurements of the devices' capacitances as functions of applied reverse bias to be 10^{15} cm^{-3} at 33 °C; at 100 V applied reverse bias, the capacitances of the devices were 0.65 pF ± 0.07 pF and 2.48 pF ± 0.09 pF, respectively. The corresponding depletion widths were 5.7 μm ± 0.9 μm and 5.3 μm ± 0.8 μm. Measurements of the devices' leakage currents as functions of reverse bias were also made at the same temperature; both devices had leakage currents <5.5 pA ± 0.4 pA at 100 V applied reverse bias (167 kV/cm corresponding electric field strength).

The presence of incomplete charge collection noise was then examined by investigating the X-ray spectroscopic response of the X-ray spectrometer employing the new $Al_{0.52}In_{0.48}P$ X-ray detectors. The detectors were each, in turn, connected to a custom-made low-noise feedback resistorless charge-sensitive preamplifier (similar to Ref. [24]). X-rays from a Mo target X-ray tube were collimated before impinging on nine different high-purity (≥98.7%) metal foils (V, Cr, Mn, Cu, Zn, Au, Ge, Nb, and Pd), each in turn, producing X-ray fluorescence. The fluorescence X-rays then illuminated the $Al_{0.52}In_{0.48}P$ devices through a 4 μm thick Al X-ray window which provided complete rejection of visible light. The output of the preamplifier was connected to an ORTEC 572A shaping amplifier, the output of which was connected to an ORTEC EASY-MCA 8k multi-channel analyser (MCA). X-ray fluorescence spectra of foils were accumulated; the detectors were reverse biased at 100 V, thus proving full depletion. The optimum available shaping times, 2 μs and 3 μs, were used for the 217 μm and 409 μm diameter photodiode based spectrometers, respectively.

A non-linear X-ray energy (charge output) response of an X-ray spectrometer is one typical indication of incomplete charge collection within the X-ray detector [25], but a linear response is not in itself conclusive evidence of complete charge collection. Indeed, it was the case that the charge output of the X-ray spectrometer employing the 2 μm think i layer $Al_{0.52}In_{0.48}P$ p^+-i-n^+ photodiode, which exhibited incomplete charge collection noise, had a linear relationship with incident photon energy [4]. Nevertheless, such an investigation is a common diagnostic tool for incomplete charge collection noise when it returns a positive result. As such, the X-ray energy (charge output) response of the $Al_{0.52}In_{0.48}P$ detector based spectrometers was investigated by recording the positions of the primary X-ray fluorescence peak (centroid channel number) in each of the accumulated X-ray fluorescence spectra, and plotting those positions as functions of the corresponding X-ray photopeak energy. This can be seen in Fig. 2, along with the lines of best fit as calculated using linear least squares fitting. The error bars associated with the fitting (±3 channels and ±2 channels for the 217 μm and 409 μm diameter photodiode based spectrometer, respectively) were smaller than the analytical uncertainty associated

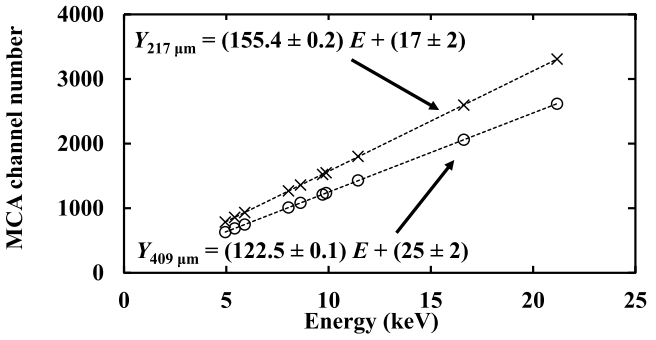

Fig. 2. The X-ray energy (charge output) response of the 217 μm (× symbols) and 409 μm (circles) diameter photodiode based X-ray spectrometer over the X-ray photon energy range 4.49 keV–21.17 keV. The lines of best fit, for the positions of the primary X-ray fluorescence peak (in terms of centroid MCA channel number) of the obtained X-ray fluorescence spectra, *Y*, as functions of their corresponding X-ray photon energy, *E*, were calculated using linear least squares fitting.

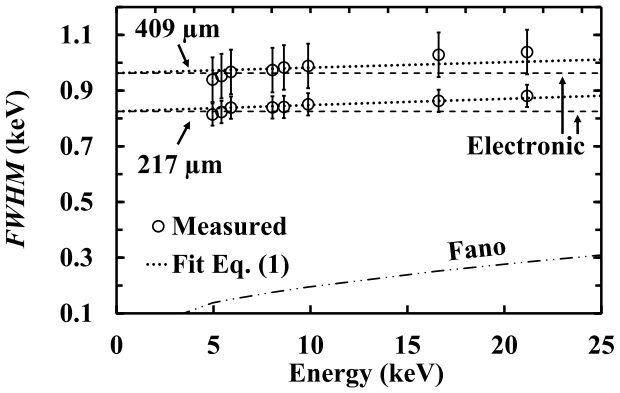

Fig. 3. Measured *FWHM* (open circles) of the primary X-ray fluorescence peak of eight different metal foils with the 217 μm diameter and 409 μm diameter photodiode based X-ray spectrometer. The calculated Fano noise (long dash double dotted line), the derived electronic noise (dashed line), and the predicted *FWHM* (round dotted line) from Eq. (1), are also shown. The Au photopeaks were excluded from this figure due to the difficulty of deconvolving them with sufficient accuracy.

with the determination of the centroid of each peak. Thus, the X-ray spectrometers' energy responses were linear over the investigated X-ray photon energy range within the associated uncertainties; the presence of incomplete charge collection noise was not detected via this technique.

Given that incomplete charge collection noise is a photon energy dependent phenomenon, the different noise contributions to the energy resolutions of the $Al_{0.52}In_{0.48}P$ photodiode X-ray spectrometers were then investigated as functions of energy. The MCA's charge scale for each of the accumulated X-ray fluorescence spectra was energy calibrated using the corresponding relationship presented in Fig. 2. Gaussians were fitted to the primary X-ray photopeak in each spectrum taking into account the appropriate emission ratios [26] and the relative quantum detection efficiencies of the detectors at the corresponding X-ray energies when more than one X-ray line was encompassed in a single photopeak. The *FWHM* of the primary X-ray fluorescence peak for each foil as detected using the 217 μm and 409 μm diameter photodiode based X-ray spectrometers was measured, and all are presented in Fig. 3. The achieved energy resolutions (*FWHM*) of the 217 μm and 409 μm diameter photodiode based X-ray spectrometers over the energy range 4.49 keV–21.17 keV ranged from 0.81 keV ± 0.04 keV to 0.88 keV ± 0.04 keV and from 0.94 keV ± 0.08 keV to 1.04 keV ± 0.08 keV, respectively. The stated uncertainties (shown as the error bars in Fig. 3) reflected the uncertainties in the Gaussians fitted to the photopeaks, and the uncertainties in the measurements.

Initially, as a working hypothesis, the incomplete charge collection noise in the detectors was considered to be negligible. The calculated Fano noise can be seen in Fig. 3. The electronic noise of the 217 μm and 409 μm $Al_{0.52}In_{0.48}P$ photodiode based X-ray spectrometers was then extracted from fitting of Eq. (1) to the measured *FWHM*; the results were found to be 0.83 keV and 0.96 keV, respectively. The predicted *FWHM*, Eq. (1), of the spectrometers as a function of X-ray photon energy can be seen in Fig. 3. The predicted *FWHM* of both X-ray spectrometers were in good agreement (within uncertainties) with the measured *FWHM*, across the energy range from 4.95 keV to 21.17 keV, therefore, the results suggested the absence of detectable incomplete charge collection noise when the photodiodes were operated at 100 V reverse bias.

The smallest detectable amount of incomplete charge collection noise being present at each spectrometer, given the predicted *FWHM* as a function of energy and the uncertainties associated with the measured *FWHM* for each spectrometer, was calculated. This lower limit of detectable incomplete charge collection noise was found to be 26 e^- rms and 40 e^- rms for the 6 μm i layer 217 μm diameter and 409 μm diameter photodiode based X-ray spectrometers, respectively, at the highest investigated X-ray energy, 21.17 keV, at which the incomplete charge collection noise was expected to have its highest contribution. Hence, it was concluded that the 6 μm thick i layer detectors exhibited <26 e^- rms (217 μm diameter) and <40 e^- rms (409 μm diameter) incomplete charge collection noise, whilst the previously reported 2 μm thick $Al_{0.52}In_{0.48}P$ detectors exhibited 36 e^- rms (200 μm diameter) [4] and >60 e^- rms (400 μm diameter) of incomplete charge collection noise.

In summary, the thickest (6 μm i layer) $Al_{0.52}In_{0.48}P$ X-ray photodiodes so far reported have been produced. Unlike previous thinner (2 μm i layer) $Al_{0.52}In_{0.48}P$ detectors, the thicker devices do not suffer from incomplete charge collection noise at detectable levels within the energy range investigated. Better energy resolutions were also achieved with the new detectors compared with the original devices; this was attributed to the absence of detectable incomplete charge collection noise and a reduction in electronic noise due to the lower capacitance of the new devices. These results are important for the development of new X-ray detectors because they demonstrate that incomplete charge collection noise is not an inherent feature of $Al_{0.52}In_{0.48}P$; as such, the material is a highly promising candidate for future X-ray spectrometers. It may be possible in future to make additional measurements of the charge collection efficiency (potentially with a precision better than 0.1%) by obtaining X-ray spectra as a function of detector internal electric field and application of the Hecht equation [27]. This will be explored and reported as part of future work on these emerging $Al_{0.52}In_{0.48}P$ X-ray detectors.

Declaration of competing interest

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

CRediT authorship contribution statement

S. Zhao: Methodology, Formal analysis, Investigation, Writing - original draft, Visualization. **G. Lioliou:** Methodology, Formal analysis, Writing - review & editing, Visualization. **S. Butera:** Formal analysis, Writing - review & editing. **A.B. Krysa:** Resources, Writing - review & editing. **A.M. Barnett:** Conceptualization, Methodology, Writing - review & editing, Supervision, Project administration, Funding acquisition.

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Data underlying this work are subject to commercial confidentiality. The Authors regret that they cannot grant public requests for further access to any data produced during the study, however the key findings are fully included within the article.

References

[1] A. Auckloo, J.S. Cheong, X. Meng, C.H. Tan, J.S. Ng, A. Krysa, R.C. Tozer, J.P.R. David, $Al_{0.52}In_{0.48}P$ avalanche photodiodes for soft X-ray spectroscopy, J. Instrum. 11 (2016) P03021.

[2] S. Butera, G. Lioliou, A.B. Krysa, A.M. Barnett, Characterisation of $Al_{0.52}In_{0.48}P$ mesa $p-i-n$ photodiodes for X-ray photon counting spectroscopy, J. Appl. Phys. 120 (2016) 024502.

[3] S. Butera, T. Gohil, G. Lioliou, A.B. Krysa, A.M. Barnett, Temperature study of $Al_{0.52}In_{0.48}P$ detector photon counting X-ray spectrometer, J. Appl. Phys. 120 (2016) 174503.

[4] G. Lioliou, S. Butera, A.B. Krysa, A.M. Barnett, X-ray spectroscopy with an AlInP photodiode, Nucl. Instrum. Methods Phys. Res. A 943 (2019) 162467.

[5] S. Zhao, S. Butera, G. Lioliou, A.B. Krysa, A.M. Barnett, AlInP photodiode x-ray detectors, J. Phys. D 52 (2019) 1–9.

[6] J.S. Cheong, J.S.L. Ong, J.S. Ng, A.B. Krysa, J.P.R. David, $Al_{0.52}In_{0.48}P$ SAM-APD as a blue-green detector, IEEE J. Sel. Top. Quantum Electron. 20 (2014) 3801305.

[7] G. Bertuccio, R. Casiraghi, A. Cetronio, C. Lanzieri, F. Nava, Silicon carbide for high resolution X-ray detectors operating up to 100 °C, Nucl. Instrum. Methods Phys. Res. A 522 (2004) 413–419.

[8] G. Bertuccio, S. Caccia, D. Puglisi, D. Macera, Advances in silicon carbide X-ray detectors, Nucl. Instrum. Methods Phys. Res. A 652 (2011) 193.

[9] B.L. Henke, E.M. Gullikson, J.C. Davis, X-ray interactions: Photoabsorption, scattering, transmission, and reflection at $E = 50 - 30,000$ eV, $Z = 1 - 92$, At. Data Nucl. Data Tables 54 (1993) 181–342.

[10] B.G. Lowe, R.A. Sareen, Semiconductor X-ray Detectors, Taylor & Francis Inc, Florida, 2014.

[11] S. Butera, G. Lioliou, A.B. Krysa, A.M. Barnett, Measurement of the electron–hole pair creation energy in $Al_{0.52}In_{0.48}P$ using X-ray radiation, Nucl. Instrum. Methods Phys. Res. A 879 (2018) 64–68.

[12] G. Bertuccio, R. Casiraghi, Study of silicon carbide for X-ray detection and spectroscopy, IEEE Trans. Nucl. Sci. 50 (2003) 175–185.

[13] G. Bertuccio, The silence of the Amps: Integrated circuits for very-low-noise processing of random signals from radiation detectors, IEEE Solid-State Circuits Mag. 4 (2012) 36–45.

[14] U. Fano, Ionization yield of radiations. II. The fluctuations of the number of ions, Phys. Rev. 72 (1947) 26–29.

[15] R.B. Day, G. Dearnaley, J.M. Palms, Noise, trapping and energy resolution in semiconductor gamma-ray spectrometers, IEEE Trans. Nucl. Sci. 14 (1967) 487–491.

[16] G.F. Knoll, D.S. Mcgregor, Fundamentals of semiconductor detectors for ionizing radiation, Mater. Res. Soc. Symp. Proc. 302 (1993) 3–17.

[17] G. Bertuccio, C. Canali, F. Nava, Energy resolution in GaAs X- and γ-ray detectors, Nucl. Instrum. Methods Phys. Res. A 410 (1998) 29–35.

[18] A. Owens, Compound Semiconductor Radiation Detectors, Taylor & Francis Inc, Florida, 2012.

[19] C. Erd, A. Owens, G. Brammertz, M. Bavdaz, A. Peacock, V. Lämsä, S. Nenonen, H. Andersson, N. Haack, Hard X-ray test and evaluation of a prototype 32 × 32 pixel gallium–arsenide array, Nucl. Instrum. Methods Phys. Res. A 487 (2002) 78–89.

[20] G. Lioliou, A.B. Krysa, A.M. Barnett, Energy response characterization of InGaP X-ray detectors, J. Appl. Phys. 124 (2018) 195704.

[21] A. Owens, M. Bavdaz, H. Andersson, T. Gagliardi, M. Krumrey, S. Nenonen, A. Peacock, I. Taylor, L. Tröger, The X-ray response of CdZnTe, Nucl. Instrum. Methods Phys. Res. A 484 (2002) 242–250.

[22] A.G. Kozorezov, J.K. Wigmore, A. Owens, R. den Hartog, A. Peacock, Hala A. Al-Jawhari, Resolution degradation of semiconductor detectors due to carrier trapping, Nucl. Instrum. Methods Phys. Res. A 546 (2005) 209–212.

[23] 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.

[24] G. Bertuccio, P. Rehak, D. Xi, A novel charge sensitive preamplifier without the feedback resistor, Nucl. Instrum. Methods Phys. Res. A 326 (1993) 71–76.

[25] G. Knoll, Radiation Detection and Measurement, John Wiley, New Jersey, 2010.

[26] M. Sánchez del Rio, A. Brunetti, B. Golosio, A. Somogyi, A. Simionovici, XRAYLIB Tables (X-Ray Fluorescence Cross-Section), European Synchrotron Radiation Facility and University of Sassari, 2003.

[27] G. Bertuccio, Prospect for energy resolving X-ray imaging with compound semiconductor pixel detectors, Nucl. Instrum. Methods Phys. Res. A 546 (2005) 232–241.