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# Geant4 X-ray fluorescence with updated libraries

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## Abstract

We present the results concerning the development in Geant4 of a new data driven library, called here the ANSTO HF library. This X-ray fluorescence library is based on an approach of particular interest for PIXE simulation applications; however, it can be used in any Geant4 applications where X-ray fluorescence needs to be described. The X-ray fluorescence transition probabilities were calculated within the Hartree-Fock (HF) approach, which is recognised to better reproduce PIXE experimental values compared with the Hartree-Slater approach, adopted in the current default Geant4 EADL data library. These HF X-ray fluorescence transition probabilities were integrated into a new Geant4 library and will be released within Geant4 in the near future.

In this paper, we compare the fluorescence X-ray spectra generated by the ANSTO HF library and by the currently available library (EADL-1991 [1]) within Geant4, for targets irradiated with protons and  $\alpha$  particles with energies up to 10 MeV, a range of interest for PIXE applications. The comparisons were performed for a large set of sample materials spanning a broad range of target atomic numbers. These two approaches were compared to existing experimental measurements performed at the ANSTO heavy ion microprobe beamline using 2 MeV and 3 MeV proton and 10 MeV He<sup>2+</sup> ion beams. This work represents a useful upgrade to the Geant4 atomic de-excitation package.

## Keywords

Geant4, PIXE, ionisation cross sections, X-ray fluorescence.

## Introduction

36 Particle Induced X-ray Emission (PIXE) is a well-established non-destructive analytical  
37 technique to determine the elemental composition of a sample, with very broad applications  
38 ranging from archaeometry to zoology.

39 PIXE describes the phenomenon of charged particles, such as protons,  $\alpha$  and heavier ions,  
40 interacting with matter. This includes the ionisation of atoms within the target by removing  
41 one or more electrons from the K, L or M shells, followed by atomic de-excitation. This shell  
42 vacancy is then filled by an electron from an outer shell with the subsequent emission of a  
43 characteristic X-ray. These fluorescence X-rays are then detected and used to determine the  
44 elemental composition of the irradiated target. The elements in a sample are identified by  
45 the energy of the emitted X-rays while their concentration is determined by the intensity of  
46 each specific X-ray line [2].

47 Geant4 [3] is a general-purpose Monte Carlo toolkit, describing particle interactions in matter  
48 in the energy range from a few eV to TeV. In the context of PIXE, Geant4 allows one to model  
49 the composition of both homogeneous and heterogeneous materials, gaseous samples for  
50 environmental applications, planetary samples for geological and space science studies. Like  
51 the PIXAN and GUPIX [4] packages, Geant4 models PIXE taking into account the absorption of  
52 emitted X-rays and the slowing down of the incident particle as it passes through a target.

53 The Geant4 Atomic Relaxation package includes models for the generation of vacancies in  
54 atomic shells and the subsequent emission of fluorescence X-rays and Auger electrons. The  
55 development of the Geant4 Atomic Relaxation Package was firstly documented in [5] and was  
56 more recently improved [6]. Two concepts are considered in the simulation of atomic  
57 relaxation [5]: (1) the creation of a vacancy by a primary process and (2) the generation of the  
58 relaxation cascade. The first one is handled by any Geant4 Electromagnetic Physics Package  
59 (including Standard, Livermore, Penelope, and Geant4-DNA models) which manages the  
60 primary interactions, e.g., the photoelectric effect, Compton scattering and ionisation; the  
61 second one is handled by the Atomic Relaxation package, which is used by all the primary  
62 processes generating a vacancy. The secondaries generated by the Atomic Relaxation are  
63 then passed to Geant4 tracking and managed as any other particle in the simulation.

64 Different ionisation cross sections for protons and  $\alpha$  particles can be currently used in Geant4  
65 to generate the vacancy in a shell, as described in [7]: (1) K and L shell ionisation cross sections  
66 based on empirical and semi-empirical compilations [8]–[10]; (2) The so-called Form Factor  
67 set, based on an empirical polynomial approximation to the ionisation cross sections of K, L  
68 and a selection of M shells calculated from the ECPSSR theory for incident protons and  $\alpha$   
69 particles [11], [12]. (3) The third set, named ECPSSR Analytical, is based on the ECPSSR theory  
70 [13] for the description of K and L shells ionisation for incident protons and  $\alpha$  particles [6]; (4)  
71 the fourth set, called ANSTO ECPSSR [14], based on the theoretical work of Brandt and Lapicki  
72 and documented in [15]–[18].

73 In addition, the Atomic Relaxation Geant4 component [5], [6], handling the emission of X-rays  
74 and Auger/Coster-Kronig electrons, is based on the Evaluated Data Library EADL [1], which  
75 provides the atomic electron binding energies and the radiative (fluorescence) and non-  
76 radiative (Auger and Coster–Kronig electrons) transition probabilities. Such tabulated data

77 are used to calculate the X-ray fluorescence lines [6] for Geant4 applications, where the  
78 modelling of atomic de-excitation is necessary. The radiative transition probabilities reported  
79 in the EADL were calculated according to Hartree-Slater (HS) methods [19], however following  
80 the work of Campbell et al. [20], [21] Cohen et al. [15] recommended the Hartree-Fock  
81 approach [22] rather than the Hartree-Slater model [19]. Validation studies of the G4-PIXE  
82 Package such as Guatelli et al. 2007 [5] and Pia et al. 2011 [23] showed that the EADL can  
83 reproduce the X-ray lines deriving from the K and L shell transitions within few percentage  
84 accuracy when compared to a selected experimental dataset by Deslattes et al. 2003 [24].  
85 However, such agreement may be significantly improved by using data libraries based on  
86 Hartree-Fock method [15].

87 This project is motivated by the ongoing effort to improve the physics model as well as  
88 providing the user with a broader range of dataset options in Geant4 [6]. Despite the  
89 availability of different ionisation cross sections and atomic relaxation data sets, there is the  
90 need to provide a range of options as well as a unique, accurate, self-consistent and robust  
91 recommended approach to the PIXE user community, within Geant4. This improvement also  
92 benefits applications of Geant4 in other domains, such as environmental physics, geology,  
93 archaeology, and space science.

## 94 **Methods**

95 The ECPSSR theory was developed by Brandt and Lapicki for both K and L subshell ionisation  
96 by light ions ( $Z_1/Z_2 < 0.3$ , where  $Z_1$  and  $Z_2$  refer to the charges of the projectile and the target  
97 atom, respectively [25]. Cohen and Harrigan published ECPSSR K and L subshell ionisation  
98 cross sections for both protons and  $\alpha$  particles bombardment for ion energies in the general  
99 PIXE range from 0.2 to 10 MeV and for a wide variety of target atoms, from carbon to curium  
100 [16], [17]. Their later publication included corrections to this dataset for the relativistic nature  
101 of the ions. This correction was not large for the MeV ions used in PIXE with energies between  
102 1 MeV to 3 MeV.

103 In Geant4, atomic relaxation simulation is articulated through two stages:

- 104 1) The creation of a vacancy by a primary process e.g., photoelectric effect, Compton  
105 scattering and ionisation. The shell (or subshell) where the vacancy is created by a  
106 process is sampled based on the cross section of the given process.  
107 For the ionisation process number and shell/sub-shell of the vacancies are calculated  
108 according to the PIXE cross sections. Two ionisation cross section models were used  
109 in this work:
  - 110 a. *ECPSSR Form Factor* [11], [12]: based on a polynomial approximation of the  
111 ionisation cross sections of K, L and a selection of M shells calculated with the  
112 ECPSSR theory for incident protons and  $\alpha$  particles. This approach is the most  
113 recent available Geant4 set.
  - 114 b. *ANSTO ECPSSR* [14]: based on the ECPSSR approach of the ionisation cross  
115 sections of K, L and M sub/shells as calculated by D.D. Cohen et al. [15], [17]  
116 for incident protons and  $\alpha$  particles. This approach was implemented in Geant4  
117 and will be released in Geant4 within 2022.

- 118 2) The relaxation cascade is triggered, starting from the vacancy created by the primary  
 119 process. Fluorescence X-ray, Auger electrons or Coster-Kronig transitions are  
 120 generated through radiative and non-radiative transitions, based on the respective  
 121 transition probabilities. In the simulation, all secondary particles with energies greater  
 122 than the cut energy of 10 eV are tracked. Two transition probability libraries were used  
 123 in this work to generate the fluorescence X-rays:
- 124 a. *EADL* is the default library that is used in Geant4 to provide the transition  
 125 probability and is based on Hartree-Slater approach [19].
  - 126 b. a new fluorescence data library, called here *ANSTO HF*, from ANSTO  
 127 calculations based on Hartree-Fock approach [22]. This was written  
 128 analogously to the existing *EADL* Geant4 data library, for simplicity,  
 129 transparency and to facilitate its integration in the G4EMLOW data library.  
 130 Same binding energies of *EADL* (as in G4EMLOW7.7) were adopted in the  
 131 *ANSTO HF* library.

132 Table 1 lists the approaches used in this work.

133 *Table 1 models and approaches in this comparison*

	<b>G4-default</b>	<b>G4-ANSTO</b>
<b>Ionisation cross sections</b>	<i>ECPSSR Form Factor</i> [11], [12]	<i>ANSTO ECPSSR</i> [14]
<b>Transition probabilities</b>	<i>EADL</i> (Hartree-Slater) [19]	<i>ANSTO HF</i> (Hartree-Fock) [22]

134

135 Geant4 extended example *TestEm5* and Geant4 10.05.p01 were used. The impact of the two  
 136 fluorescence libraries, *ANSTO HF* and *EADL*, was quantified in terms of fluorescence X-ray  
 137 yields per incident particle. Nine target materials (Si, Ti, Fe, Zn, Nb, Ru, Ce, Ta, Au) were  
 138 chosen, from low to high atomic number *Z* materials.

139 When the particle's velocity matches the electron shell velocity, the chance of the K electron  
 140 shell ionisation is maximised. For example, protons and  $\alpha$  particles when incident on silicon  
 141 need a kinetic energy of  $\sim 3$  MeV and  $\sim 25$  MeV, respectively, to achieve some degree of  
 142 velocity matching [26]. This is strongly dependent on the target atomic number *Z*.  
 143 Monoenergetic pencil beams of protons (2 and 3 MeV) and  $\alpha$  particles (10 MeV) were  
 144 considered as they are representative of some common PIXE applications where  
 145 experimental data may be available for comparison with theory. These particles were incident  
 146 on 25  $\mu\text{m}$  thick targets along the direction of the incident beam.

147 The production threshold of secondary particles was ignored. The fluorescence X-rays were  
 148 counted once they were generated in the target. Note that the existing Geant4 PIXE *Empirical*  
 149 and *Analytical* cross section sets [6] were not considered in this work as they generate only K  
 150 and L vacancies.

151 Finally, the *G4-ANSTO* and *G4-default* (see Table 1) were compared to experimental  
152 measurements performed at ANSTO using the 6 MV SIRIUS Tandem Accelerator [27]. The  
153 experimental spectra were normalised to the highest line of the *G4-ANSTO* X-ray emissions.

## 154 Experimental setup

155 PIXE spectra were experimentally measured at the ANSTO heavy ion microprobe beamline  
156 [27] using 2 and 3 MeV proton and 10 MeV He<sup>2+</sup> ion beams with beam currents varying  
157 between 0.5 and 2.5 nA. For X-ray detection, a 100 mm<sup>2</sup> high purity Ge detector with a solid  
158 angle of 90 msr was used. The detector has a 25 μm thick Be window. To prevent the scattered  
159 protons from entering the detector and to reduce the low energy X-ray yield from light  
160 elements such as the underlying Si in some of the samples, a 100 μm thick Mylar absorber (or  
161 filter) was placed in front of the detector. The data were collected using the Data Acquisition  
162 System mpsys4 from Melbourne University together with a Canberra Model 2060 digital  
163 signal processor. The irradiated samples were: 1 mm silicon layer, 100 nm Au layer on silicon,  
164 50 nm TiN layer on silicon, 50 nm ZnO layer on silicon, 25 nm Ta<sub>2</sub>O<sub>3</sub> layer on graphite, 25 nm  
165 Ru oxide film on graphite, 45 nm Nb oxide film on graphite, Fe<sub>2</sub>O<sub>3</sub> layer embedded in a boron  
166 oxide pellet, and a sample of CeO<sub>2</sub> embedded in a boron oxide pellet.

## 167 Results and discussion

168 The following figures (1, 2, 4, 5, 7, 8 and 10a) show the results of *G4-ANSTO* and *G4-default*  
169 approaches, with the X axis representing energy in keV and the Y axis representing X-ray  
170 frequency per incident particle. To compare *G4-ANSTO* to the experimental spectra, the *G4-*  
171 *ANSTO* data was convoluted with a Gaussian function. The reason behind calculating the  
172 Gaussian for *G4-ANSTO* is that there were different lines adjacent to each other and,  
173 therefore, their contribution was convoluted to produce the experimentally observed peaks.

174 In these figures (1, 2, 4, 5, 7, 8 and 10a), we simply calculated the Gaussian of *G4-ANSTO* and  
175 normalised it to the *G4-ANSTO* highest line to compare with the experimental data. Doing the  
176 same for the *G4-default* approach and plotting all the data and spectra on the same plot  
177 would deceive the readers and represent the Gaussian of both *G4-ANSTO* and *G4-default* to  
178 be nearly identical, as seen in figure (1).

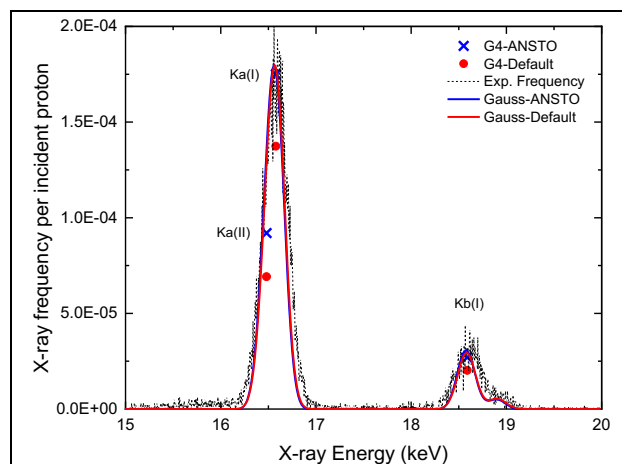


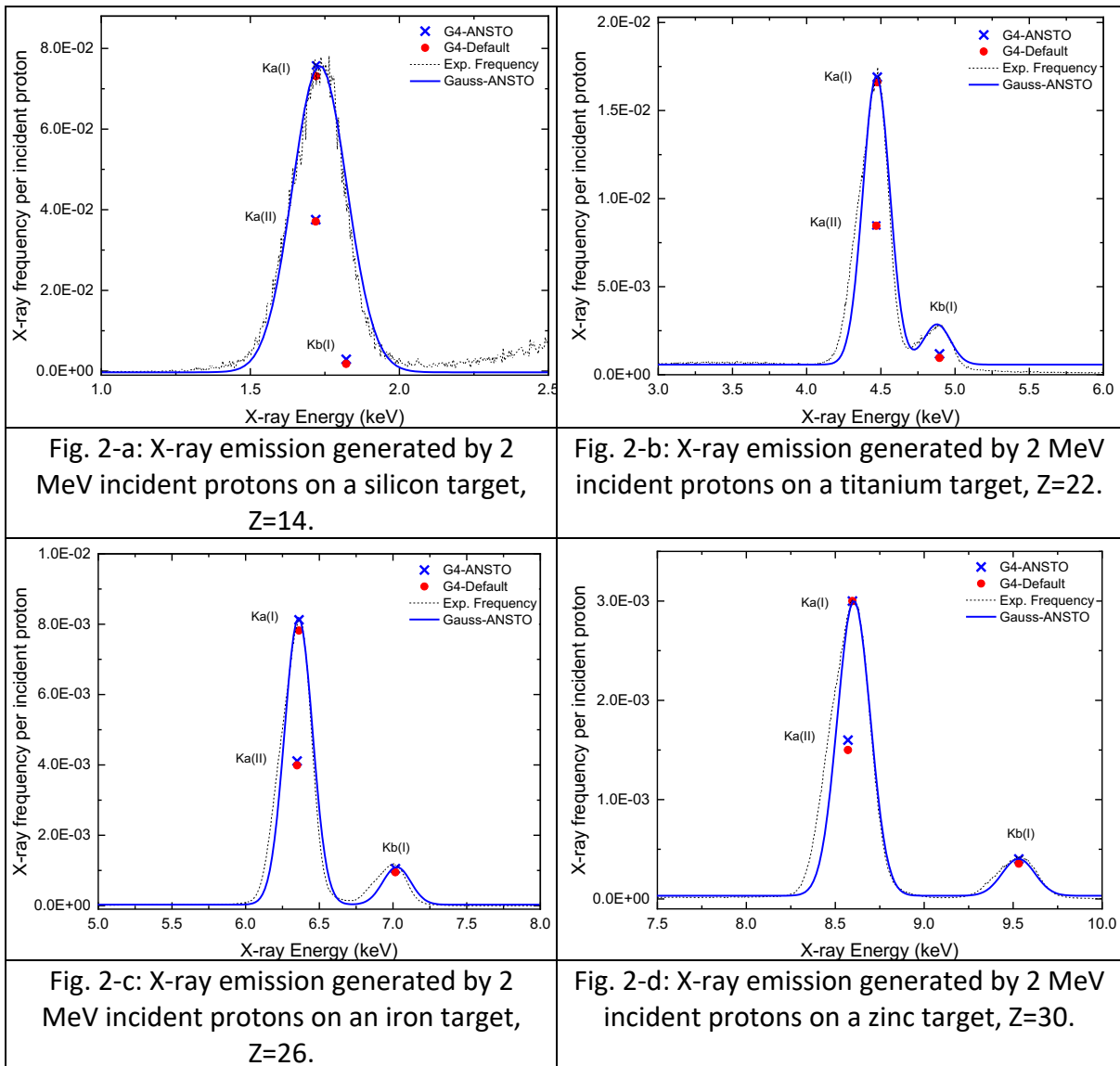
Fig. 1: X-ray emission generated by 2 MeV incident protons on a niobium target, Z=41.

179

180 Nonetheless, because the data libraries can be employed in a variety of applications ranging  
181 from space to environmental and healthcare applications, it is critical to demonstrate the  
182 absolute differences in X-ray emission yields produced with the two different data libraries.

183 **Comparison of 2 MeV incident protons results**

184 Figures 2, 3, and 4 compare the X-ray fluorescence yields obtained with *G4-default* and *G4-*  
185 *ANSTO* approaches, and experimental measurements. Figure 2 compares the X-ray emission  
186 frequencies (for the K shell) per incident particle generated by a 2 MeV proton on Si, Ti, Fe,  
187 Zn, Nb, and Ru samples. The Gaussian of *G4-ANSTO* results was calculated and compared with  
188 the experimental spectrum. The experimental spectra and the Gaussian of *G4-ANSTO* were  
189 normalised to the highest line of the *G4-ANSTO* X-ray emissions, in this case  $K\alpha(I)$ . Figure 2  
190 indicates that *G4-ANSTO* approach has higher X-ray emission rates (for  $K\alpha(I)$ ,  $K\alpha(II)$ , and  $K\beta(I)$   
191 X-ray lines) than the *G4-default* approach in all samples.



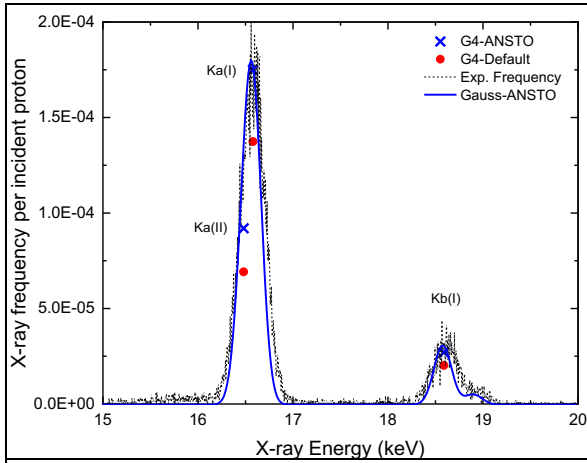


Fig. 2-e: X-ray emission generated by 2 MeV incident protons on a niobium target,  $Z=41$ .

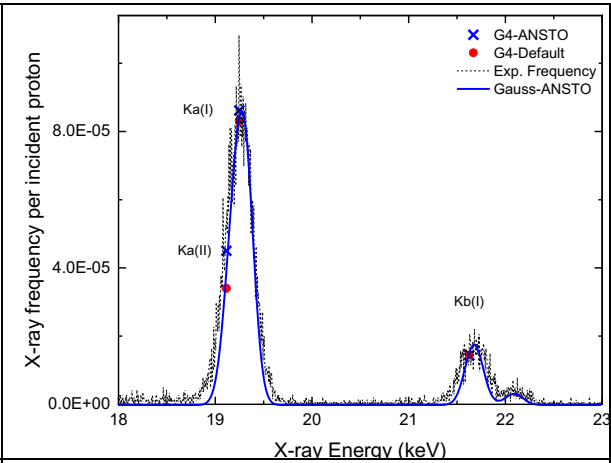


Fig. 2-f: X-ray emission generated by 2 MeV incident protons on a ruthenium target,  $Z=44$ .

192

193 The ratio of *G4-ANSTO* to *G4-default* calculated X-ray yields of Nb and Ru derived primarily  
 194 from vacancies in the K and L shells produced by incident 2 MeV protons is shown in figure 3.

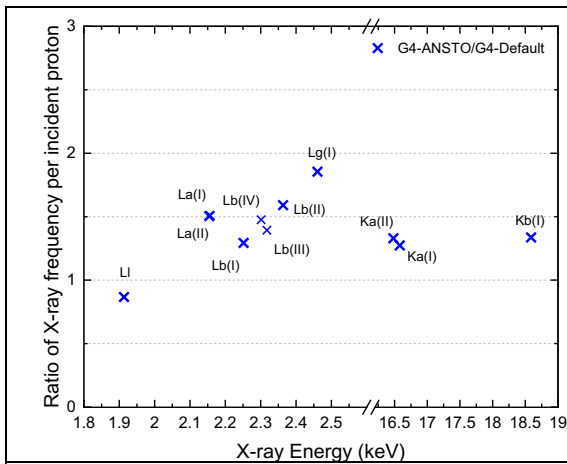


Fig. 3-a: Ratio of X-ray emission generated by 2 MeV incident protons on a niobium target,  $Z=41$ .

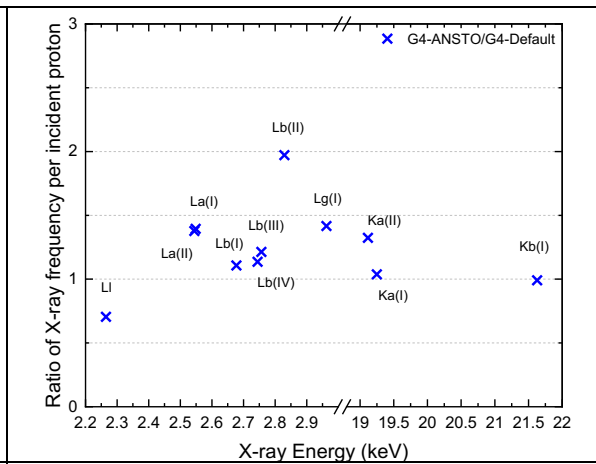


Fig. 3-b: Ratio of X-ray emission generated by 2 MeV incident protons on a ruthenium target,  $Z=44$ .

195

196 Figure 4 compares calculated X-ray yields to experimental spectra of Ce and Ta, which are  
 197 driven primarily by vacancies in the L shell created by incident 2 MeV protons.



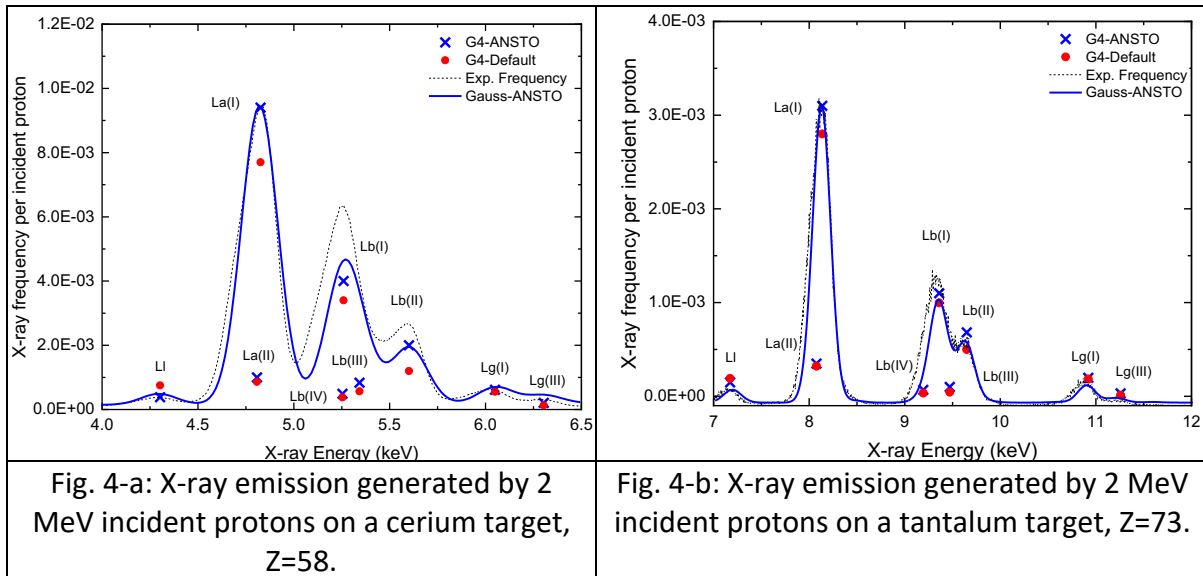


Fig. 4-a: X-ray emission generated by 2 MeV incident protons on a cerium target, Z=58.

Fig. 4-b: X-ray emission generated by 2 MeV incident protons on a tantalum target, Z=73.

198

199 The L shell X-ray emissions for Nb (Z=41) and Ru (Z=44) samples were not obtained  
 200 experimentally. Therefore, only the results of Geant4 simulation using the *G4-ANSTO* and *G4-*  
 201 *default* approaches are shown in figure 3. The ratio of the lines generated by *G4-ANSTO*  
 202 approach to the *G4-default* approach was displayed in figure 3. Most of Geant4 lines were  
 203 found to be substantially higher in the *G4-ANSTO* approach for both samples (Nb and Ru). It  
 204 can be noticed that most lines show no more ~50% difference between *G4-ANSTO* and *G4-*  
 205 *default* approaches, while it is around ~100% difference for Ly(I) and Lβ(II) for Nb and Ru,  
 206 respectively. Except the L $\alpha$  line, the electron transition from M(I) subshell to L(III) subshell, in  
 207 *G4-ANSTO* results is about ~25% lower than in *G4-default* results.

208 Figure 4 depicts the findings, which indicate a reasonable agreement between Geant4-  
 209 calculated emission X-ray spectra and the experimental measurements. We found that the  
 210 *G4-ANSTO* approach produces higher X-ray emission results than the *G4-default* one. The  
 211 difference was particularly noticeable for the L $\alpha$ (I) line. In addition, the lines (Lβ (I), (II), (III),  
 212 (IV)) calculated by means of both *G4-ANSTO* and *G4-default* approaches were lower than the  
 213 experimental spectrum, which was attributable to the fact that these four lines were adjacent  
 214 to each other and, therefore, their contribution was convoluted to produce the higher peaks  
 215 observable in the experimental data. However, the Gaussian spectra of *G4-ANSTO* approach  
 216 showed better results when compared to the experimental spectrum.

217 As shown in figure 4, the L $\alpha$  line of cerium produced by the *G4-default* approach was higher  
 218 than that generated by the *G4-ANSTO* one. The X-ray fluorescence databases of *G4-default*  
 219 and *G4-ANSTO* approaches were compared to understand the reason for these variations in  
 220 L $\alpha$  X-ray yield and it was found that only the L $\alpha$  X-ray line probability of transition was higher  
 221 in *G4-default* when compared to *G4-ANSTO*.

222 **Comparison of 3 MeV incident protons results**

223 Figure 5 compares calculated X-ray yields to experimental spectra of Si, Ti, Fe, Zn, Nb, and Ru  
 224 derived primarily from vacancies in the K shell induced by incident 3 MeV protons.

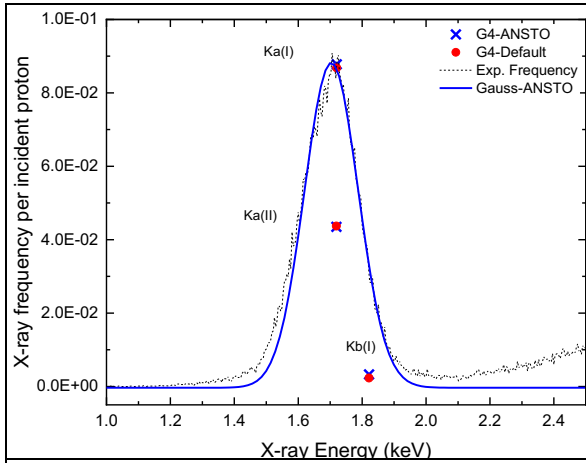


Fig. 5-a: X-ray emission generated by 3 MeV incident protons on a silicon target,  $Z=14$ .

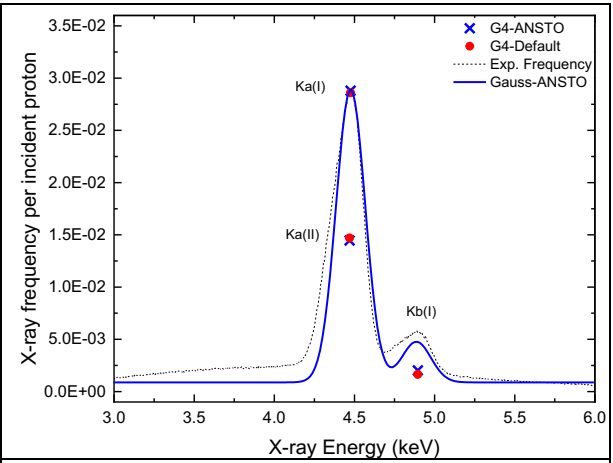


Fig. 5-b: X-ray emission generated by 3 MeV incident protons on a titanium target,  $Z=22$ .

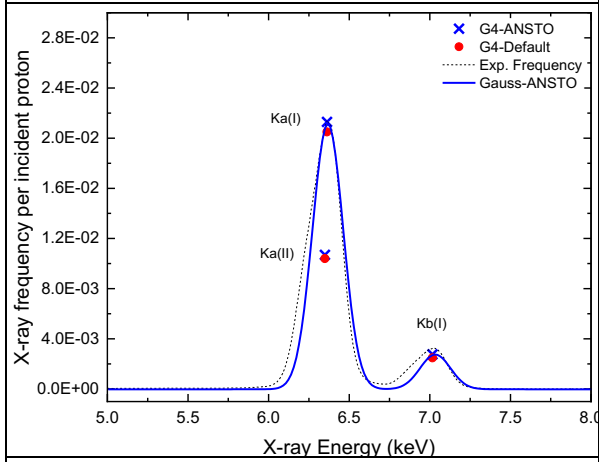


Fig. 5-c: X-ray emission generated by 3 MeV incident protons on an iron target,  $Z=26$ .

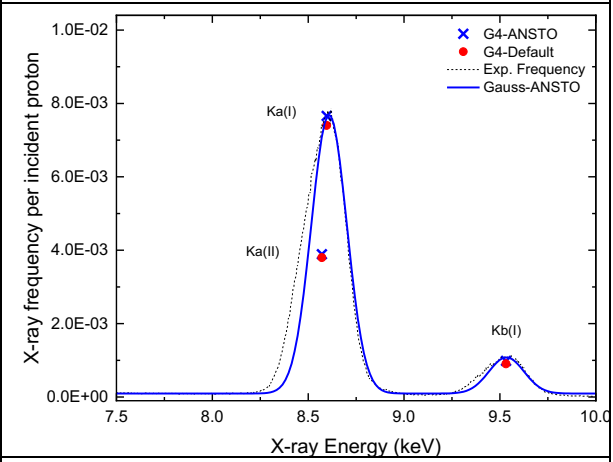


Fig. 5-d: X-ray emission generated by 3 MeV incident protons on a zinc target,  $Z=30$ .

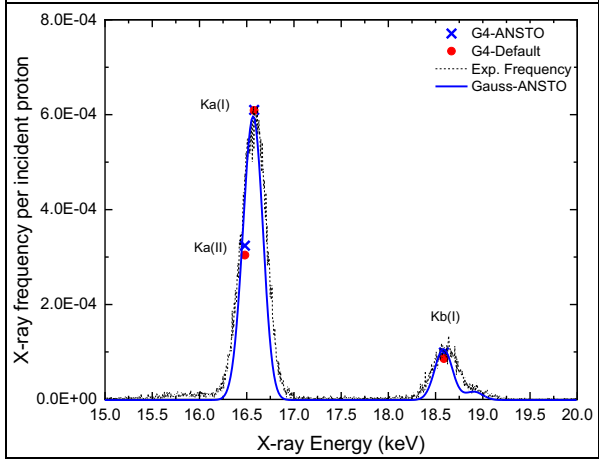


Fig. 5-e: X-ray emission generated by 3 MeV incident protons on a niobium target,  $Z=41$ .

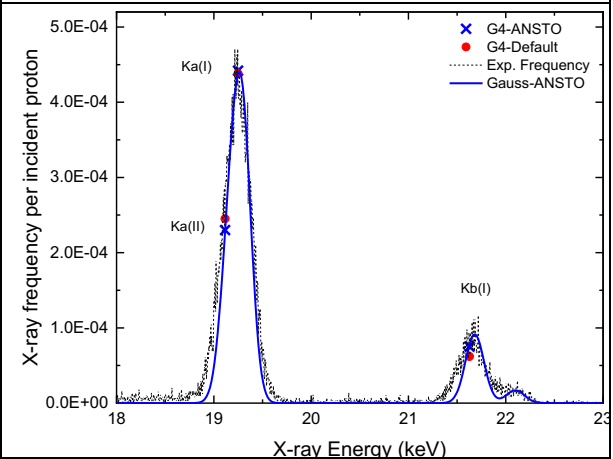


Fig. 5-f: X-ray emission generated by 3 MeV incident protons on a ruthenium target,  $Z=44$ .

226 Figure 6 depicts the measured X-ray yields of Nb and Ru, which were primarily derived from  
 227 vacancies in the L and K shells induced by incident 3 MeV protons.

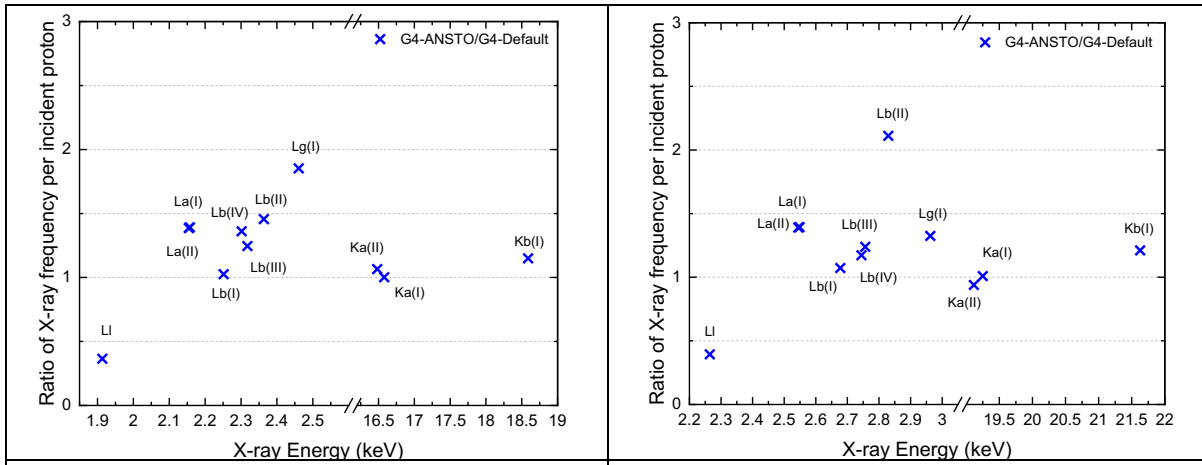


Fig. 6-a: Ratio of X-ray emission generated by 3 MeV incident protons on a niobium target, Z=41.

Fig. 6-b: Ratio of X-ray emission generated by 3 MeV incident protons on a ruthenium target, Z=44.

228

229 Figure 7 compares calculated X-ray yields to experimental spectra of Ce, Ta, and Au, which  
 230 were driven primarily by vacancies in the L shell induced by incident 3 MeV protons.

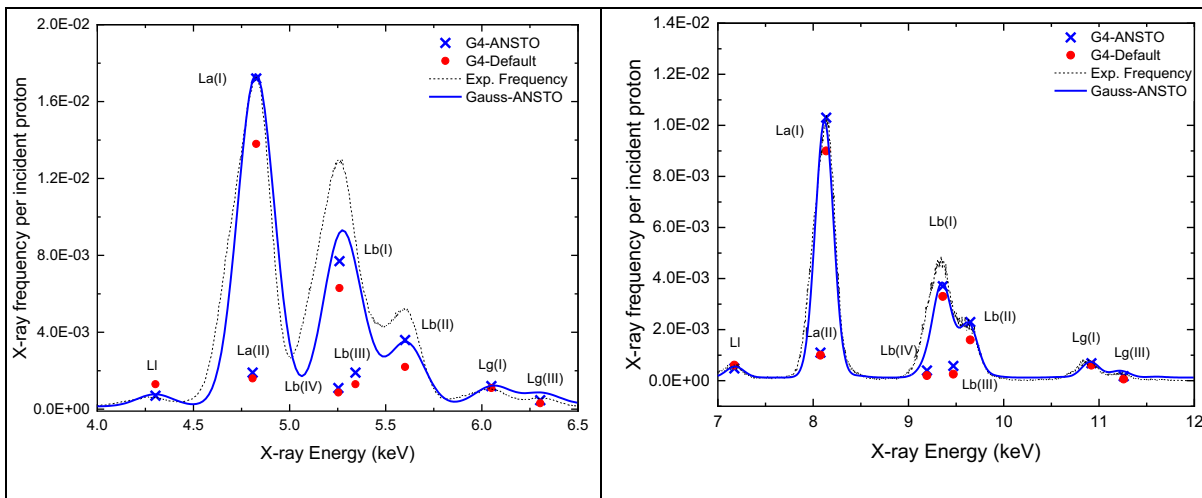
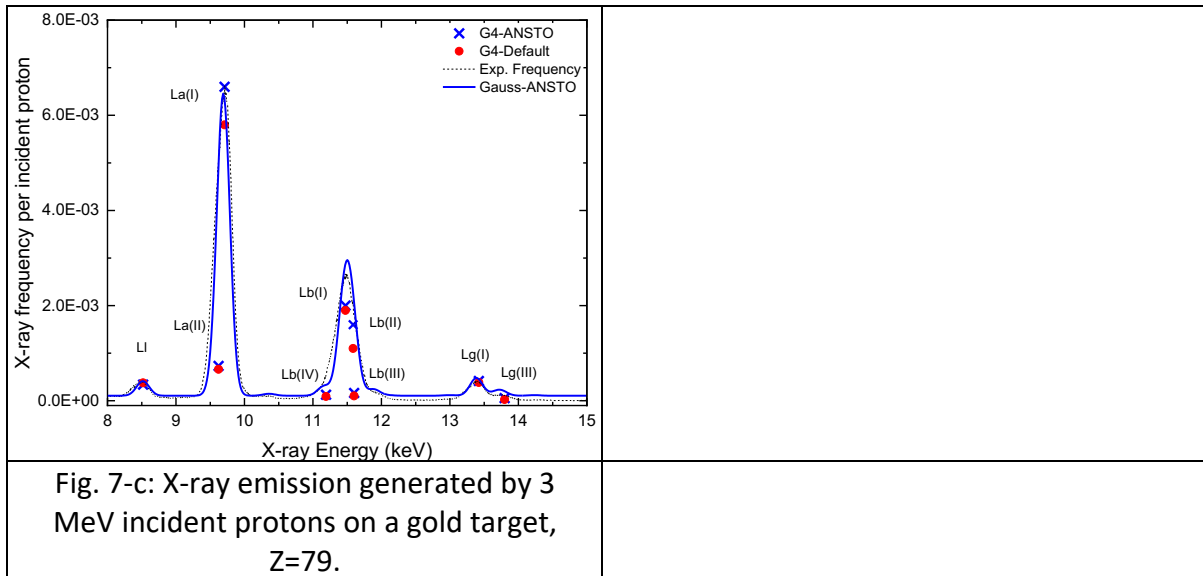


Fig. 7-a: X-ray emission generated by 3 MeV incident protons on a cerium target, Z=58.

Fig. 7-b: X-ray emission generated by 3 MeV incident protons on a tantalum target, Z=73.



231

232 Figures 5, 6, and 7 provide a comparison of the *G4-ANSTO* and *G4-default* approaches' results.  
 233 Figure 5 compares the calculated *G4-ANSTO* and *G4-default* X-ray emission frequencies (for  
 234 the K shell) per incident particle, produced by a 3 MeV proton on Si, Ti, Fe, Zn, Nb, and Ru  
 235 samples to experimental measurements. Additionally, the experimental spectrum was  
 236 compared to the Gaussian of *G4-ANSTO* results. The experimental spectra and the Gaussian  
 237 of *G4-ANSTO* were normalised to the highest line of the *G4-ANSTO* X-ray emissions, in this  
 238 case  $K\alpha(I)$ . Figure 5 shows that the *G4-ANSTO* approach provides higher X-ray emission  
 239 frequencies than the *G4-default* approach in all samples, except for  $K\alpha(II)$  X-ray line for Ru  
 240 (see figure 5-f).

241 Since the L shell experimental spectra for these samples are not available, figure 6 only shows  
 242 the results of the Geant4 simulation, the ratio of lines generated by the *G4-ANSTO* approach  
 243 to lines generated by the *G4-default* approach. It is worth noting that all lines show less than  
 244 ~50% difference between *G4-ANSTO* and *G4-default* approaches, while  $L\gamma(I)$  and  $L\beta(II)$  for Nb  
 245 and Ru, respectively, show about ~100% difference.

246 Figure 7 indicates that the Gaussian of *G4-ANSTO* emission X-ray spectra and the  
 247 experimental measurements agreed reasonably well. The experimental spectrum and the  
 248 Gaussian of *G4-ANSTO* were normalised to the highest line of the *G4-ANSTO* approach. We  
 249 found that the *G4-ANSTO* approach produces higher X-ray emission results than the *G4-*  
 250 *default* results. The differentiation was especially evident in the  $L\alpha(I)$  line.

251 Figures 6 and 7 show that only the  $L\alpha(I)$  X-ray yield is about ~60% higher in *G4-default* approach  
 252 than in *G4-ANSTO* one since the probability of transition for the  $L\alpha(I)$  X-ray line was higher in *G4-*  
 253 *default* approach.

254 **Comparison of 10 MeV incident  $\alpha$  particles results**

255 Figure 8 compares calculated X-ray yields to experimental spectra of Ti, Zn, Nb, and Ru derived  
 256 primarily from vacancies in the K shell induced by incident 10 MeV  $\alpha$  particles.

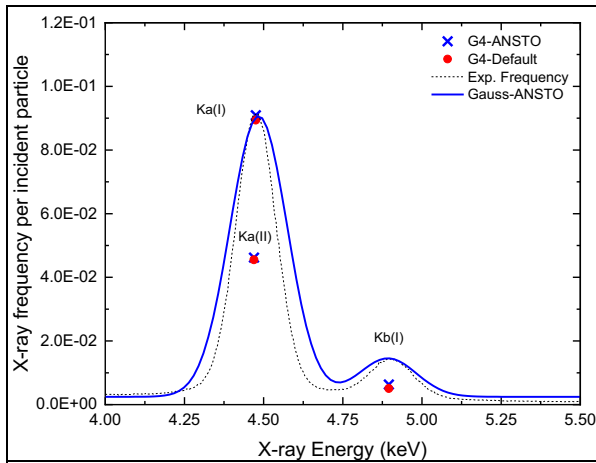


Fig. 8-a: X-ray emission generated by 10 MeV incident  $\alpha$  particles on a titanium target,  $Z=22$ .

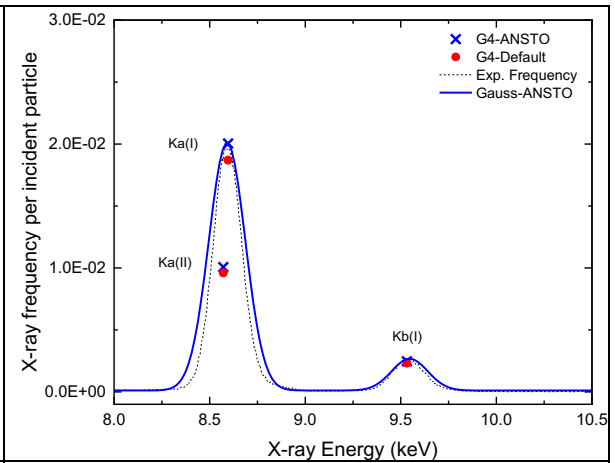


Fig. 8-b: X-ray emission generated by 10 MeV incident  $\alpha$  particles on a zinc target,  $Z=30$ .

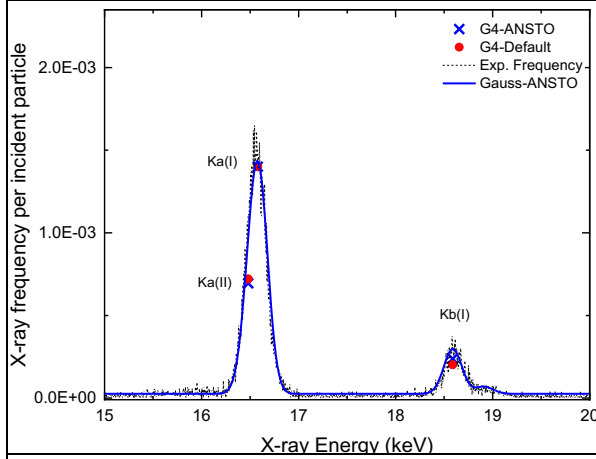


Fig. 8-c: X-ray emission generated by 10 MeV incident  $\alpha$  particles on a niobium target,  $Z=41$ .

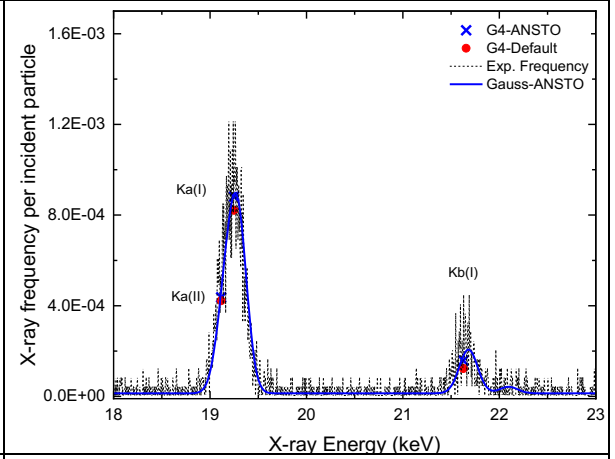


Fig. 8-d: X-ray emission generated by 10 MeV incident  $\alpha$  particles on a ruthenium target,  $Z=44$ .

257

258 Figure 9 depicts the calculated X-ray yields of Nb and Ru, which result primarily from vacancies  
 259 in the L and K shells induced by incident 10 MeV  $\alpha$  particles.

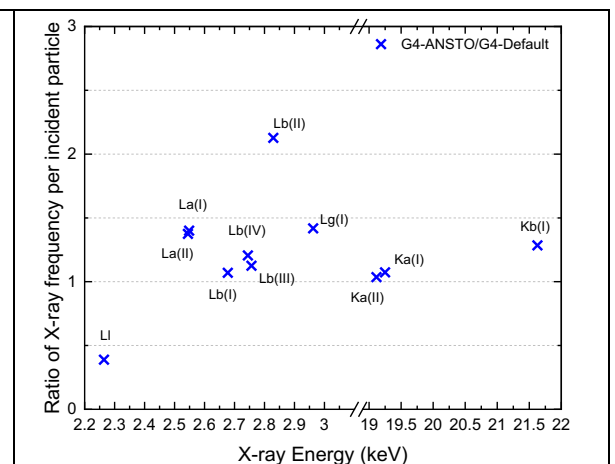
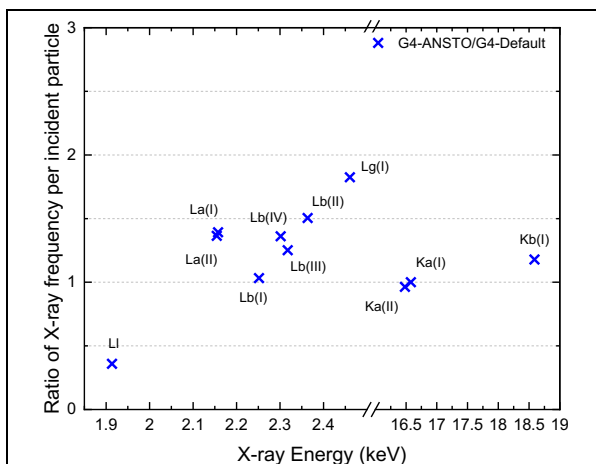


Fig. 9-a Ratio of X-ray emission generated by 10 MeV incident  $\alpha$  particles on a niobium target, Z=41.

Fig. 9-a Ratio of X-ray emission generated by 10 MeV incident  $\alpha$  particles on a ruthenium target, Z=44.

260

261 Figure 10 compares calculated X-ray yields to experimental Ta spectra derived primarily from  
 262 vacancies in the L shell induced by 10 MeV incident  $\alpha$  particles.

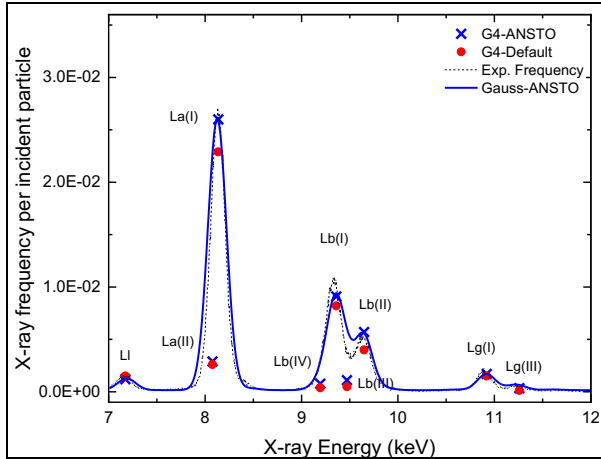


Fig. 10-a: X-ray emission generated by 10 MeV incident  $\alpha$  particles on a tantalum target, Z=73.

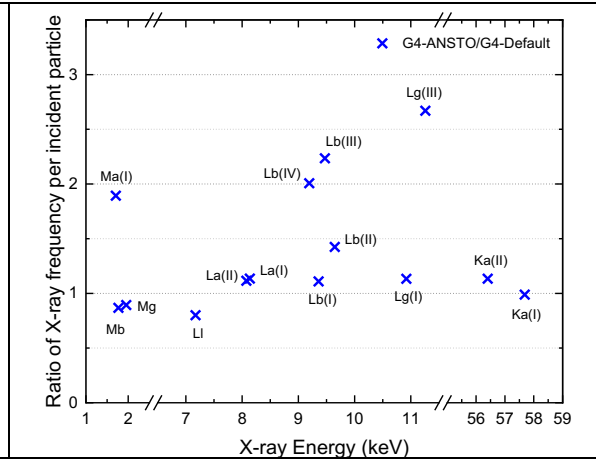


Fig. 10-b: X-ray emission generated by 10 MeV incident  $\alpha$  particles on a tantalum target, Z=73.

263

264 Figures 8, 9, and 10 compare the results of the *G4-ANSTO* and *G4-default* approaches. Figure  
 265 8 compares the X-ray emission frequencies (for the K shell), produced by 10 MeV  $\alpha$  particles  
 266 on Ti, Zn, Nb, and Ru samples, calculated using the *G4-ANSTO* and *G4-default* approaches to  
 267 experimental measurements. The Gaussian of *G4-ANSTO* results were compared to the  
 268 experimental spectrum. The experimental spectra and the Gaussian of *G4-ANSTO* were  
 269 normalised to the highest line of *G4-ANSTO* approach, in this case  $K\alpha(I)$ . Figure 8 shows that  
 270 the *G4-ANSTO* approach indicates about  $\sim 5\%$  higher X-ray emission frequencies than the *G4-*  
 271 *default* approach in all samples. Figure 9 depicts the L and K shells X-ray emissions using the  
 272 *G4-ANSTO* and *G4-default* approaches; the *G4-ANSTO* results were significantly higher than  
 273 the *G4-default* results, a maximum of 50% difference for most lines, while  $L\gamma(I)$  and  $L\beta(II)$  for  
 274 Nb and Ru, respectively, showed about  $\sim 100\%$  difference. Besides the  $L\gamma(I)$  line, which was  $\sim 60\%$   
 275 lower using *G4-ANSTO* approach than *G4-default* approach.

276 Figure 10 depicts the calculated Ta X-ray yields resulting primarily from vacancies in the L and  
 277 M shells induced by 10 MeV incident  $\alpha$  particles. In Figure 10-a, the Gaussian of *G4-ANSTO* X-  
 278 ray emission results showed a reasonably good agreement with the ANSTO experimental  
 279 data. Figure 10-b, on the other hand, depicts the M and L shells X-ray emissions for both the  
 280 *G4-ANSTO* and *G4-default* approaches under consideration. Except for the  $L\gamma(I)$  X-ray line, all *G4-*  
 281 *ANSTO* X-ray lines created from vacancies in L shells were higher (maximum of 100% besides  
 282 of  $L\beta(III)$  and  $L\gamma(III)$  are about 150% higher) than the *G4-default* ones, due to the higher  
 283 probability of transition in the *G4-default* approach, with the sole exception of the  $M\beta$  and  
 284  $M\gamma$  X-ray lines.

## 285 **Conclusions**

286 The *G4-ANSTO* approach, which includes the *ANSTO ECPSSR* ionisation cross sections [14]  
287 and *ANSTO HF* fluorescence yield [22] libraries, was implemented in Geant4 and compared to  
288 *G4-default* approach. The *G4-default* approach includes the *ECPSSR Form Factor* ionisation  
289 cross sections [11], [12] and *EADL* [19] fluorescence yield libraries. Geant4-calculated X-ray  
290 emission spectra using the *G4-ANSTO*, and the *G4-default* approaches were compared in  
291 terms of fluorescence X-ray yields per incident particle. Nine target materials (Si, Ti, Fe, Zn,  
292 Nb, Ru, Ce, Ta, Au) were under study, from low to high atomic number *Z* materials.  
293 Monoenergetic beams of protons (2 and 3 MeV) and  $\alpha$  particles (10 MeV) were used.

294 For all samples, most lines were observed to be significantly higher when using the *G4-ANSTO*  
295 approach with the exception of the L<sub>I</sub> line, corresponding to the electron transition from the  
296 M(I) subshell to the L(III) subshell. The study showed that the Geant4-calculated X-ray  
297 emission spectra and the ANSTO experimental measurements were in a reasonably good  
298 agreement. However, we are only analysing a few peaks with PIXE, but in reality, *G4-ANSTO*  
299 has significant differences with *G4-default* for other lines, which may be important in other  
300 application domains.

301 The novel *G4-ANSTO* X-ray fluorescence data libraries can be coupled with the *G4-ANSTO*  
302 cross sections [14] or with the Geant4 default cross sections, handling the generation of  
303 atomic vacancies (e.g., the photoelectric effect, Compton scattering and ionisation).

304 The *G4-ANSTO* approach will be included in the public release of Geant4 in the near future  
305 and will be usable by means of user interface commands on top of any electromagnetic  
306 physics configurations. The Geant4 PIXE user community will be able to use the *G4-ANSTO*  
307 libraries and models, which provide a unique, self-consistent, and stable recommended  
308 approach.

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