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Nuclear spectroscopy with GEANT4

The superheavy challenge

Luis G. Sarmiento[a]

Department of Physics, Lund University, 22100 Lund, Sweden

Abstract. The simulation toolkit GEANT4 was originally developed at CERN for high-energy physics. Over the years it has been established as a *swiss army knife* not only in particle physics but it has seen an accelerated expansion towards nuclear physics and more recently to medical imaging and γ- and ion- therapy to mention but a handful of new applications.

The validity of GEANT4 is vast and large across many particles, ions, materials, and physical processes with typically various different models to choose from. Unfortunately, atomic nuclei with atomic number $Z > 100$ are not properly supported. This is likely due to the rather novelty of the field, its comparably small user base, and scarce evaluated experimental data.

To circumvent this situation different workarounds have been used over the years. In this work the simulation toolkit GEANT4 will be introduced with its different components and the effort to bring the software to the heavy and superheavy region will be described.

1. Introduction

During the past decade, ^{48}Ca-induced fusion-evaporation reactions on radioactive actinide targets have been the driving force behind the observations of several correlated α-decay chains, which all terminate by spontaneous fission. These decay chains are interpreted to originate from the production of atomic nuclei with proton numbers $Z = 113$–118 [1]. However, neither their mass, A, nor their atomic number, Z, have been measured directly thus far.

Monte Carlo simulations are already an established method to assess various experimental aspects in nuclear and particle physics among others disciplines. Simulation packages of the passage – and interaction – of particles through matter such as the GEANT4 simulation toolkit [2] have been used systematically to benchmark and characterize complete experimental set-ups.

GEANT4 has its origins in high-energy particle physics. The initial scope quickly widened when it became apparent that such a tool would also benefit the nuclear, accelerator, space, and medical physics community. Lack of theoretical understanding, scarce experimental data, and a relative small community have hindered the expansion of the toolkit towards simulation of atomic nuclei with atomic number $Z > 100$. Recently GEANT4 has been used to verify the coherence and consistency of decay schemes of heavy and superheavy nuclei [3–6] showing that the – arguably new – field of heavy and superheavy nuclei can benefit greatly from native support in the simulation toolkit.

[a] e-mail: Luis.Sarmiento@nuclear.lu.se

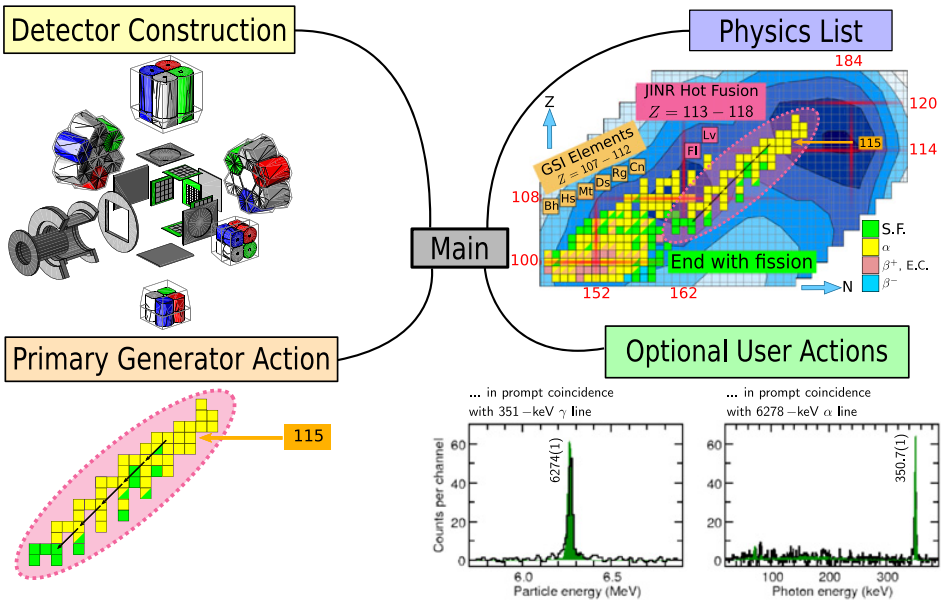

Figure 1. Schematic presentation of the different components of a Monte Carlo simulation using GEANT4. The "laboratory" (geometry, materials, positions) is defined in the *Detector Constructor*. The relevant particles and physical processes to be included in the simulation should go in the *Physics List*. In the *Primary Generator Action* the properties and definitions of the initial particles coming for the "source" to be simulated are included. On top of these three mandatory components the user has the possibility to use some *Optional User Actions*. See text for details.

2. GEANT4 overview

Any GEANT4 simulation requires some basic "ingredients". Figure 1 cartoons the ingredients in particular for the simulation of the TASISPEC experimental set-up [7]. First, we describe the physical space, for instance a laboratory containing a detector set-up, in the *Detector Constructor*. There the geometry, materials, and positions of the different components are defined. Second, since the simulation toolkit covers a wide range of energies and particles, the relevant ones to be used for a specific simulation are included in the *Physics List*; in this example we include the ones in the realm of a typical nuclear physics experiment (α, β^+, or β^- particles, γ rays, ions, electromagnetic interactions, radioactive decay, etc.) and we leave out the high-energy ($\sim$ GeV) and very-low energy ($\sim$ meV) specific physics processes and particles. Third, in the *Primary Generator Action* the properties and definitions of the initial particles to be simulated are included, for instance a radioactive source or an ion beam, in our case a radioactive beam of $^{288}115$ atoms. Last, on top of these three mandatory components the user has the possibility to use some *Optional User Actions* to have more fine control of the the simulation. Simulation control parameters, gating procedures, and user-defined simulation outputs are examples of possible *actions*.

In particular for TASISPEC, the geometry was included in the most realistic, yet practical manner (see upper left of Fig. 1). The remaining *ingredients* of the simulation have been carefully tailored and benchmarked in such a way that it was even possible to use experimental data from calibration sources to determine the experimental thicknesses of the dead layers of the silicon detectors for more than 2000 pixels on a pixel-by-pixel basis [8], and to implement these in the final GEANT4-simulated geometry.

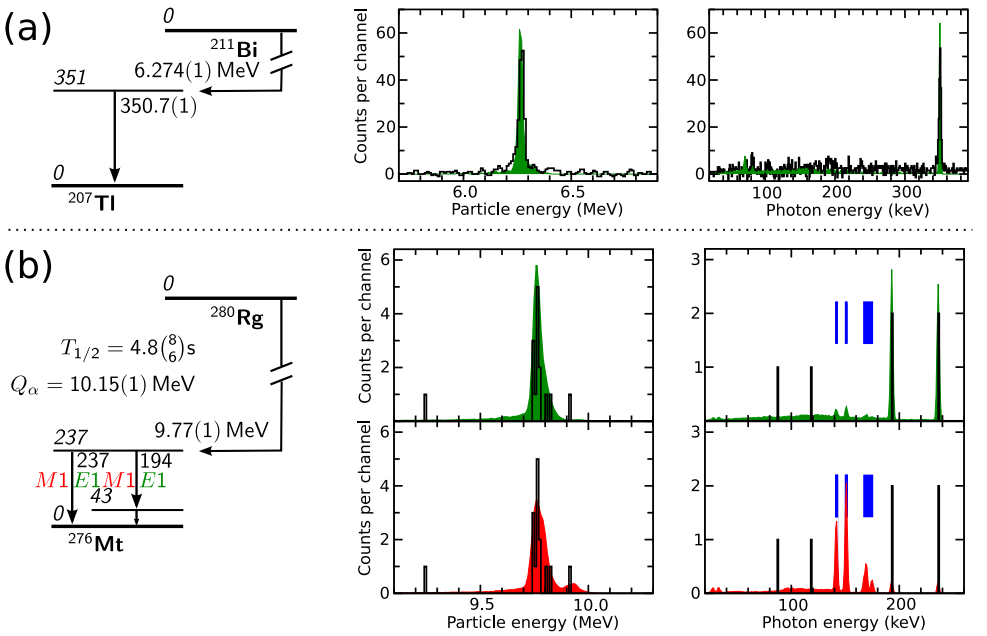

Figure 2. Experimental (black) and simulated (green and red) particle- and photon-energy spectra for the (a) ^{211}Bi $\rightarrow$ ^{207}Tl and (b) ^{280}Rg $\rightarrow$ ^{276}Mt decays. The simulations used 10^5 decays following the schemes shown on the left hand side. The level scheme shown in (a) contains only statistical uncertainties of the peak fit. Level energies and γ-ray energies are in keV. In blue, the K X-ray energies of Mt are indicated in the photon spectra. See text for details.

After the simulated experimental set-up has been benchmarked against calibration sources it is desirable to test its performance under "in-beam" conditions and compare it to the experimental results in the most direct possible way. By using the *Optional User Actions* it is possible to accommodate the output of the simulation into experimental-like events that can be analyzed in the exact same fashion as the experimental data. The deposition of ^{211}Bi ions into TASISpec was investigated; the deposition profile, event rate, and energy distribution of the ions were set to match the experimental conditions after the TASCA separator [9, 10] during the 2012 experiment. The results of the comparison between the simulation and the experiment can be found in the lower-right part of Fig. 1 and they are featured later in Fig. 2.

Once the performance of the simulation has been benchmarked under different conditions the next step is to use the – at this stage – well-established simulation in a self-consistent way to shed light in the interpretation of newly available experimental data in a process dubbed Geant4-aided nuclear spectroscopy.

For the Geant4-aided nuclear spectroscopy we iterate over user-defined nuclear decay schemes which are *fed* into the simulation in a self-consistent procedure. The definition of new decay schemes (or override of the existing ones) is readily possible using User Interface Commands provided by the toolkit itself. The commands allow the user to manipulate the simulation physics input in two ways: (*i*) the radioactive decay (decay type, branching ratios, etc.), and (*ii*) the subsequent nuclear de-excitation (γ-ray transitions, emission of conversion electrons, etc.). The interplay between those two inputs provides valuable information in the analysis of the complex, and oftentimes scarce, experimental data.

3. GEANT4 and the decay of heavy and superheavy atomic nuclei

Ideally, to simulate the implantation and decay of heavy and superheavy atomic nuclei, for example element 115, one should apply the same "recipe" as for the in-beam simulation of ^{211}Bi decays. This means we have already included the nuclear physics processes we are interested in; in particular electromagnetic interactions, radioactive decays, and atomic relaxation. As for the *Primary Generator Action*, i.e. an initial nucleus of element 115, it should be a matter of creating the corresponding particle and its electromagnetic decay properties of the full chain, and include them in the simulation using the existing tools for it. Unfortunately, until rather recently the GEANT4 source code had a hard-coded upper limit in the atomic number of $Z = 100$.

While – now – it is supported to use $Z > 100$, i.e. element 115, as a *particle* – in the toolkit sense of it – and its decay properties (radioactive decay and photon de-excitation) via user-interface commands, elements with $Z > 100$ are most certainly far from being fully supported. There are over 100 GEANT4 classes in the source code that are silently forced to $Z \leq 100$ or even lower atomic numbers, for instance the electronic stopping power for materials which is valid only for $Z \leq 91$ [11].

Of paramount importance in the investigation of heavy and superheavy atomic nuclei is the possibility to investigate their atomic relaxation processes as highly converted γ transitions are responsible for summing effects [6] in the α-decay energies and perhaps more importantly in the emission of characteristic X rays [3]. However, that is one of the aspects which is more difficult to extend in the toolkit due to the complicated many physical processes and databases intertwined for the radiative (fluorescence photons), and non-radiative (Auger and Coster-Kronig electrons) atomic relaxation.

As a workaround it is possible to hard-code the atomic relaxation process as a whole as part of the *Primary Generator Action* [5] for a stationary nucleus, or it is possible to *disguise* heavy nuclei as lighter ones by modifying the atomic properties such as electron binding energies, occupancy, X-ray and Auger energies, etc., of the heaviest odd number element to be taken as the heaviest odd element available in GEANT4, i.e. to convert $Z = 115$ to $Z = 99$ and $Z = 113$ to $Z = 97$, in their atomic properties and study their decay on a step-by-step basis as this method allows most dynamic aspects of the ions to be simulated [12]. Other approaches have ignored the atomic relaxation but allowed hard-coded decay paths covering only γ rays and conversion electrons using the electron binding energies of the heaviest available atom in GEANT4, Fm ($Z = 100$) [13].

The electron binding energies for elements up to $Z = 130$ are readily available in the literature [14]. Fluorescence and Auger energies and their corresponding transitions probabilities can be obtained using a linear extrapolation of the data files for low energy electromagnetic processes for elements $Z = 95$ to 100, which are available in GEANT4 [15]. Proper stopping power tables, bulk density values, and many other quantities associated with a *properly defined* element – or material – are still to be provided.

4. Current state of affairs

As it was previously stated it is necessary to benchmark the simulation performance before GEANT4-aided nuclear spectroscopy can be performed. Figure 2a shows the experimental (black) and simulated (green) particle and photon spectra of the transfer-reaction background isotope, ^{211}Bi, and its α-decay daughter, ^{207}Tl. It is remarkable how well the simulation reproduces the experimental data. The simulation comprised 10^5 decays following the

decay scheme shown on the left hand side. The experimentally measured energy values, $E_\alpha = 6274(1)\,\mathrm{keV}$ and $E_\gamma = 350.7(1)\,\mathrm{keV}$, differ slightly from $E_\alpha = 6278.2(7)\,\mathrm{keV}$ and $E_\gamma = 351.07(5)\,\mathrm{keV}$ found in the literature [16], but for the experimental numbers also systematic uncertainties of $E_\alpha \lesssim 10\,\mathrm{keV}$ and $E_\gamma \lesssim 1\,\mathrm{keV}$ need to be considered. The experimental and simulated data was treated in an identical fashion to improve the validity and consistency of the physical assessment of the interpretation of results.

The particle energy spectrum as seen by the simulated set-up was normalized to the number of α particles detected experimentally. The α-photon coincidence efficiency of the simulation is notably very well under control, because the yield of the 351-keV line in the photon energy spectrum is nicely reproduced using a normalization factor identical to the one of the particle spectrum [12].

Once the simulation has been proven to work it is possible to use it to interpret newly available experimental data. This is demonstrated in the study of the α decay of ^{280}Rg. In this case it was necessary to *disguise* the superheavy nuclei as described in the previous section. Figure 2b shows the result of the comparison between the experimental data and the simulation for two cases, namely varying the electromagnetic character of the simulated 194-keV and 237-keV γ-ray transitions: either $E1$ (green) or $M1$ (red). Just like for the ^{211}Bi α decay, the photon spectra were normalized to the number of α particles detected experimentally. It is possible to assign the 194-keV and 237-keV transitions as $E1$ transitions in the decay scheme of Fig. 2b. If the γ-ray transitions had $M1$ character instead, the simulation would suggest the detection of K_α X rays, because $M1$ transitions with energies just above the K-binding energy are highly converted. This initial GEANT4-aided assignment was later independently confirmed [4]. The $E1$ character of those transitions implies very interesting nuclear structure physics [3, 17–19].

5. Future native extension of GEANT4 towards superheavy elements

Currently GEANT4 uses by default the Evaluated Atomic Data Library EADL [20] and optionally the compilation from Bearden [21] for the electron binding energies. Unfortunately those datasets cover only the range $Z \leq 100$. There are many different compilations of binding energies of atomic properties in the literature like the ones from Carlson [22], Sevier [23], Firestone [24], and Lide [25].

In its current – development – form the GEANT4 uses the compilation of electron binding energies which constitute the backbone to the BRICC code for evaluating theoretical conversion coefficients [26, 27]. The purpose of this choice is to be consistent with the nuclear deexcitation processes simulated in the discrete so called *photon evaporation* where the BRICC code is used.

The new dataset of electron binding energies includes not only new – and more – values but also brings slightly different electron occupancies of certain orbitals for a number of elements. In order to make the changes backwards-compatible the code need to be tested and the current development effort is being made towards the implementation of a user-interface command that switches to the new databases `fluor_SHE` and `auger_SHE`[1] as well as expanding the applicability of the related data reading routine and the associated sizes of information container arrays throughout the relevant parts across the entire GEANT4 source code.

[1] Working names.

6. Closing remarks

It is shown that it is possible to tune the "ingredients" of a GEANT4 simulation to provide a self-consistent cross check of the interpretation of the sometimes very scarce experimental data. Even though quantities such as implantation depth or recoil energy of the nuclei after the α decay are not *strictly* correct they do not necessarily compromise the validity of the spectroscopic results as long as the relevant atomic decay information is properly accounted for. The use of a high-resolution spectroscopic set-up in conjunction with comprehensive Monte Carlo simulations proves to be an effective probe to study the nuclear structure properties of heavy and superheavy man-made elements.

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