Ionisation models for nano-scale simulation

Journal of Physics: Conference Series **331** (2011) 032030 doi:10.1088/1742-6596/331/3/032030

Ionisation models for nano-scale simulation

H Seo[1], M G Pia[2], C H Kim[1] and P Saracco[2]

[1] Department of Nuclear Engineering, Hanyang University, Seoul 133-791, Korea
[2] INFN Sezione di Genova, Genova 16146, Italy

E-mail: Maria.Grazia.Pia@cern.ch

Abstract. Two theory-driven models of electron ionisation cross sections, the Binary-Encounter-Bethe and the Deutsch-Märk one, have been implemented. The resulting values have been extensively validated over a large set of experimental data. The validation process also concerned the EEDL (Evaluated Electron Data Library), which is currently exploited by various Monte Carlo codes, including Geant4, to model electron interactions. The software design and physical features of the new models are reported, along with their validation results and the validation of EEDL.

1. Introduction

The capability of simulating electron interactions over a wide range from the nano-scale to the macroscopic one is needed, in the context of HEP, to deal with requirements emerging by new detector concepts and operation environments: examples are the ongoing R&D (research and development) for nanotechnology-based tracking detectors and the study of radiation effects on semiconductor devices. Models for simulating electron interactions in any materials are available in all major Monte Carlo codes, but they are intrinsically limited to the macroscopic scale; whereas, in the lower energy end, track structure codes provide simulation capabilities limited to a single, or a small number of target materials.

This paper presents new developments to endow a large scale Monte Carlo system for the first time with the capability of simulating electron ionisation down to the scale of a few tens of electronvolts for any target elements. For this purpose, models of electron impact ionization cross sections suitable to extend Geant4 [1] [2] capabilities in the low energy range have been implemented and compared with a large set of experimental measurements. The validation process, which involves experimental data pertinent to more than 50 elements, also addresses the ionization cross sections collected in the Evaluated Electron Data Library (EEDL) [3], which are used in Geant4 low energy electromagnetic package. To the best of the authors' knowledge, this is the first time that EEDL is subject to extensive experimental benchmarks below 1 keV.

Due to the length limitations and copyright constraints imposed by the conference proceedings, this paper summarizes the main features and results of the research topic; extensive details and the full set of results will be included in a forthcoming publication in a scholarly journal.

2. Electron ionization in Geant4

The Geant4 toolkit provides various implementations of electron ionization based on a condensed-discrete particle transport scheme. Two of them, respectively based on EEDL and

on the analytical models originally developed for the Penelope [4] Monte Carlo system, are included in the low energy electromagnetic package [5, 6]; another implementation is available in the standard [7] electromagnetic package. In addition, a specialized ionization model for interactions with thin layers of material, the photoabsorption-ionization (PAI) model [8], is implemented in Geant4.

The EEDL data library tabulates electron ionization cross sections in the energy range between 10 eV and 100 GeV; nevertheless, due intrinsic limitations of its accuracy, the use of Geant4 low energy models based on this library is recommended [9] for incident electron energies above 250 eV. The lower energy limit of Penelope's applicability is generically indicated by its authors as "a few hundred electronvolts" [10].

The validation of Geant4 ionisation simulation based on the EEDL data library and on Penelope-like models is documented in [11].

Ionization models applicable down to the electronvolt scale, which operate in a discrete particle transport scheme, are available in Geant4 for electron interactions in water [12]. They have not yet been validated due to lack of pertinent experimental data.

3. Cross section models

The developments described in this paper concern the calculation of cross sections for the ionization of an atom by electron impact. The first development cycle was focused on single ionization, that is the emission of one electron from a neutral atom. The models adopted in the software implementation can deal with multiple ionized atoms and molecules as well; further extensions of the software to account for these capabilities will be the object of following development cycles.

Two ionization cross section models, which specifically address the low energy range, have been implemented: the Binary-Encounter-Bethe (BEB) model [13] and the Deutsch-Märk (DM) [14] model. BEB is a theoretical model (in that it does not depend on phenomenological constant), while DM relies on a fit of some parameters to experimental data. Their behavior has been extensively investigated in relation to experimental data and to the content of the Evaluated Electron Data Library (EEDL) [3].

3.1. The Binary-Encounter-Bethe model

The Binary-Encounter-Dipole (BED) model was first proposed by Kim and Rudd [13] to calculate electron impact ionisation cross sections. It combines the Mott cross section [15] modified by the binary-encounter theory [16] for low incident energies with the Bethe theory [17] for high energies. This model does not contain any empirical or adjustable parameters.

The Binary-Encounter-Bethe (BEB) model was elaborated as a simplification of the BED model in cases where some components of the formulation of the BED cross section would be difficult to calculate or to measure experimentally. The BEB model involves three atomic parameters for each subshell of the target atom: the electron binding energy, the average kinetic energy and the electron occupation number of the subshell.

The BEB cross section for the ionization of subshell i is given by:

$$\sigma_{BEB,i} = \frac{S}{t + (u+1)/n}\left[\frac{\log(t)}{2}\left(1 - \frac{1}{t^2}\right) + 1 - \frac{1}{t} - \frac{\log(t)}{t+1}\right] \tag{1}$$

where:

$$t = \frac{T}{B}, \qquad u = \frac{U}{B}, \qquad S = 4\pi a_0^2 N \left(\frac{R}{B}\right)^2 \tag{2}$$

In the above equations T is the incident electron energy, B and N are respectively the binding energy and the occupation number of the subshell, U is the kinetic energy, t and u are normalized

incident and kinetic energies, n is the principal quantum number, a_0 is the Bohr radius and R is the Rydberg constant. $\sigma_{BEB,i}$ is proportional to N, therefore clearly electron-electron correlations are neglected (i.e. a binary encounter); this sets implicitly a lower limit on the applicability of the model. The sum over all subshells i gives the total (counting) cross section.

3.2. The Deutsch-Märk model

The Deutsch-Märk (DM) model has its origin in a classical binary encounter approximation derived by Thomson [18] and the improved form of Gryzinski [19].

The DM model calculates the atomic cross section σ_{DM} for single ionization as the sum over all partial ionization cross sections corresponding to the removal of a single electron from a given atomic subshell, characterized by quantum numbers n and l as:

$$\sigma_{DM} = \sum_{n,l} g_{nl} \pi r_{nl}^2 \xi_{nl} b_{nl}^{(q)}(u) \left[\frac{ln(c_{nl}u)}{u} \right] \tag{3}$$

where: r_{nl} is the radius of maximum radial density of the atomic subshell with quantum numbers n and l, ξ_{nl} is the electron occupation number in that subshell, g_{nl} are weighting factors, which were determined by the original creators of the model from a fit to experimental data. The quantity u represents the reduced energy E/E_{nl}, where E is the energy of the incident electron and E_{nl} is the ionization energy of the subshell identified by n and l quantum numbers. The sum extends over all the subshells of the target atom.

3.3. Electron ionization in the EEDL data library

The Evaluated Electron Data Library (EEDL) include tabulations of ionization cross sections resulting from theoretical calculations. For close collisions, the present data base uses Seltzer's modification of the Möller binary collision cross section, which takes into consideration the binding of the atomic electron in a given subshell. For distant collisions, Seltzer's modification [20] of the Weizs´acker-Williams method was used. To take into account the density effect, the relativistic cross section of Scofield [21] was used.

4. Software development

The software adopts a policy-based class design, which has also been exploited in recent developments [22, 23] for photon interactions. The policy relevant to this context is associated with a CrossSection function, whose arguments characterize the involved incident particle and target.

The software implementation is based on the most recent documented analytical formulations and associated parameters of the BEB and DM models, which are documented in the literature. The values of the atomic parameters used by the two cross section models were taken from the same sources documented by the original authors, whenever possible; otherwise, in the cases where the original values could not be retrieved, values tabulated in the Evaluated Atomic Data Library (EADL) [24] or available from the NIST web site were used.

The implemented models allow the calculation of ionization cross sections for any element.

5. Verification

Verification tests were performed to check whether the cross section values calculated by the software were consistent with those calculated by the original authors of the models, which are documented in the literature. In most cases the software implementation reproduces the original values consistently; in a few cases some discrepancies were observed, which could be tracked to different values of model parameters in the software implementation and in the original

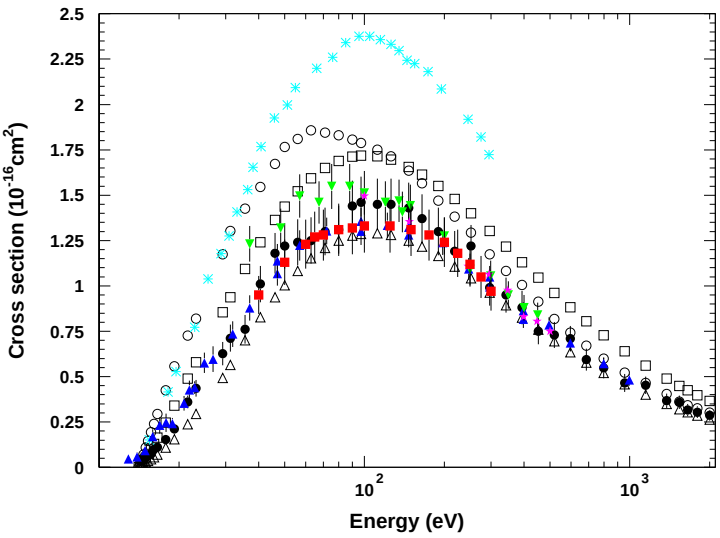

Figure 1. Cross section, Z=8: EEDL (empty circles), BEB model (empty squares), DM model (empty triangles) and experimental data from [25] (black circles), [26] (turquoise asterisks), [27] (blue triangles), [28] (green upside-down triangles), [29] (pink stars) and [30] (red squares).

calculations. A thorough analysis was performed to estimate whether the values resulting from the code implementation were statistically compatible with the original ones, using alternative compilations of atomic parameters.

As a result of the verification process, the software implementation was acknowledged to render the original cross section values with adequate precision. Further details of the verification process will be available in a dedicated paper after this conference.

6. Validation

The validation process involved the comparison with experimental data. It concerned the two new model implementations, as well as the ionization cross sections tabulated in EEDL already used by Geant4.

A survey in the literature identified more than one hundred sets of experimental data concerning electron ionization cross sections in the low energy range below 1 keV, which are pertinent to more than 50 target elements. The quality of the experimental data is highly variable over the different samples; measurements in patent disagreement are documented in the literature.

Some examples of comparisons with experimental data are shown in Figures 1 and 2.

The validation process exploited rigorous statistical analysis methods to estimate quantitatively the compatibility between the new simulation models, EEDL data and experimental data. It involved two stages: first goodness-of-fit tests [32, 33] to evaluate the hypothesis of compatibility with experimental data, then categorical analysis exploiting contingency tables to determine whether the various modelling options differ significantly in accuracy. Contingency tables were analyzed with the χ^2 test and with Fisher's exact test.

The comparisons between the results of the implementation and experimental data were performed over selected energy ranges to verify any dependence of the modelling accuracy on the application energy.

Further tests were performed to investigate whether the validation results could be biased by characteristics of the experimental data. The experimental references included different types of

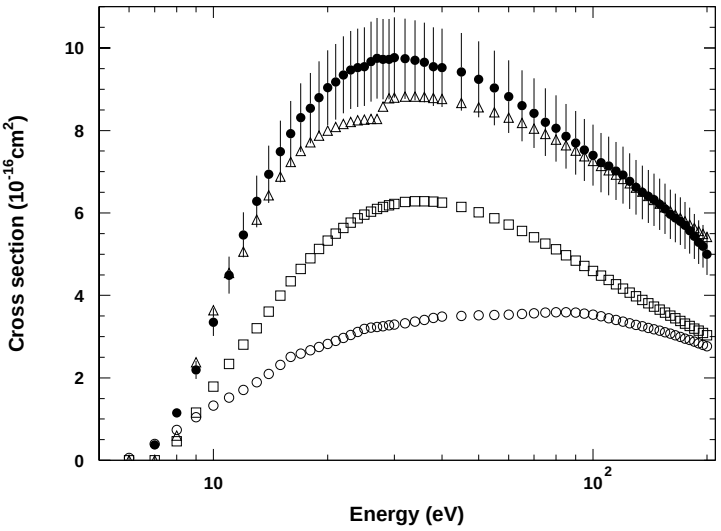

Figure 2. Cross section, Z=50: EEDL (empty circles), BEB model (empty squares), DM model (empty triangles) and experimental data from [31] (black circles).

data: cross sections for single or total ionization, absolute cross section measurements or relative to other data sources. Tests were performed on each data category separately; their outcome was evaluated with statistical methods to ascertain any dependence of the software accuracy on different types of experimental conditions.

According to the results of the tests, the DM model exhibits the best accuracy with respect to experimental data over the whole energy range and all the target elements subject to test. Its predictions are found to be compatible at 95% confidence level with experimental measurements for a fraction of tested target elements varying between 78% and 93%, depending on the energy range of the interacting electron. The BEB model is comparable in accuracy to the DM model for electron energies below 100 eV at 95% confidence level.

EEDL cross sections are compatible at 95% confidence level with experimental data above 250 eV, but they are not enough accurate at lower energies; they are statistically equivalent in accuracy to the DM ones in the energy range between 250 eV and 1 keV.

The conclusions of the statistical data analysis comparing the accuracy of the various cross section options hold whatever type of data is considered: single or total ionization, absolute or relative measurements.

The results of the validation process will be documented in detail in a dedicated paper.

7. Conclusions

Two models for the calculation of electron impact ionization, the Binary-Encounter-Bethe and the Deutsch-Märk model, have been implemented; they are specialized for application in the low energy domain below 1 keV. The software is intended for use with Geant4; it extends Geant4 simulation capabilities in an energy range not yet covered by other general purpose Monte Carlo codes.

The software has been subject to rigorous validation with respect to a large collection of experimental measurements, concerning more than 50 target elements. The validation project involved ionization cross sections included in EEDL as well; the accuracy of EEDL for electron energies below 1 keV has been quantitatively evaluated; to the best of the authors' knowledge, this is the first validation of EEDL below 1 keV. These results are relevant to the use of currently

available Geant4 models based on EEDL in experimental applications.

The new developments described in this paper open for the first time the possibility of performing microdosimetry simulations for any target elements in a general purpose Monte Carlo system.

The complete set of results is documented and discussed in depth in a dedicated paper.

Acknowledgements

The authors thank Andreas Pfeiffer for his significant help with data analysis tools throughout the project. They express their gratitude to Elisabetta Gargioni and Vladimir Grichine for advice concerning the Binary-Encounter-Bethe model, H. Deutsch and K. Becker for advice concerning the Deutsch-Märk model, and Sergio Bertolucci and Simone Giani for valuable discussions.

The support of the CERN Library has been essential to this study; the authors are especially grateful to Tullio Basaglia.

This work was partly supported by the Nuclear Research and Development Program in Korea through the Radiation Technology Development Program and the Basic Atomic Energy Research Institute (BAERI). Support also was received from the Korean Ministry of Knowledge Economy (2008-P-EP-HM-E-06-0000)/the Sunkwang Atomic Energy Safety Co., Ltd.

References

[1] Agostinelli S et al. 2003 *Nucl. Instrum. Meth. A* **506** 250
[2] Allison J et al. 2006 *IEEE Trans. Nucl. Sci.* **53** 270
[3] Perkins S Tet al. 1997 Tables and Graphs of Electron-Interaction Cross Sections from 10 eV to 100 GeV Derived from the LLNL Evaluated Electron Data Library (EEDL), UCRL-50400 Vol. 31
[4] Baro J et al. 1995 *Nucl. Instrum. Meth. B* **100** 31
[5] Chauvie S et al. 2001 *Proc. Computing in High Energy and Nuclear Physics* 337
[6] Chauvie S et al. 2004 *Conf. Rec. 2004 IEEE Nucl. Sci. Symp.* N33-165
[7] Burkhardt H et al. 2005 *Proc. 2005 Conf. on Monte Carlo Method: Versatility Unbounded in a Dynamic Computing World*
[8] Apostolakis J et al. 2000 *Nucl. Instrum. Meth. A* **453** 597
[9] Apostolakis et al. 1999 *INFN/AE-99/18*
[10] Salvat F et al. 2008 NEA-06416
[11] Lechner A et al. 2009 *IEEE Trans. Nucl. Sci.*, **56** 398
[12] Chauvie S et al. 2007 "*IEEE Trans. Nucl. Sci.* **54** 2619
[13] Kim Y K and M. E. Rudd M E 1994 *Phys. Rev. A* **50** 3954
[14] Deutsch H and Märk D T1987 *Int. J. Mass Spectrom. Ion Processes* **79** R1
[15] Mott N F1930 *Proc. Royal Soc. A* **126** 259
[16] Vriens L1966 *Phys. Rev.* **141** 88
[17] Bethe H 1930 *Ann. Phys.* **397** 325
[18] Thomson J J1912*Philos. Mag.* **23** 449
[19] Gryzinski M 1965*Phys. Rev. A* **138** 305
[20] Seltzer SM1988 *Monte Carlo Transport of Electrons and Photons* Chap. 4.
[21] Scofield J H 1978*Phys. Rev. A* **18** 963
[22] Augelli M et al. 2009 *IEEE Nucl. Sci. Symp. Conf. Rec.* 177
[23] Pia M G et al. 2010 *J. Phys.: Conf. Ser.* **219** 042019
[24] Perkins S T et al 1991 *Tables and Graphs of Atomic Subshell and Relaxation Data Derived from the LLNL Evaluated Atomic Data Library (EADL), Z=1-100* UCRL-50400 Vol. 30
[25] Thompson W R et al. 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** 1321
[26] Kieffer L J and Dumm G H 1966 *Rev. Mod. Phys.* **38** 1
[27] Brook E et al. 1978 *J. Phys. B: Atom. Molec. Phys.* **11** 3115
[28] Fite W L and Brackmann R T 1959 *Phys. Rev.* **113** 815
[29] Rothe E W et al. 1962 *Phys. Rev.* **125** 582
[30] Zipf E C 1985 *Planet. Space Sci.* **33** 1303
[31] Freund R S et al. 1990 *Phys. Rev. A* **41** 3575
[32] Cirrone G A P et al. 2004 *IEEE Trans. Nucl. Sci.* 51 2056
[33] Mascialino B et al. 2006 *IEEE Trans. Nucl. Sci.* 53 3834