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Track-Structure Simulations for Charged Particles

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Abstract

Monte-Carlo track-structure simulations provide a detailed and accurate picture of radiation transport of charged particles through condensed matter of biological interest. Liquid water serves as surrogate for soft tissue and is used in most Monte-Carlo track-structure codes. Basic theories of radiation transport and track-structure simulations are discussed and differences to condensed history codes highlighted. Interaction cross sections for electrons, protons, alpha particles, light and heavy ions are required input data for track-structure simulations. Different calculation methods, including the plane-wave Born approximation, the dielectric theory, and semi-empirical approaches are presented using liquid water as a target. Low-energy electron transport and light ion transport are discussed as areas of special interest.

Keywords

National Council on Radiation Protection and Measurements; Monte-Carlo simulations; track structure; charged particles; interaction cross sections

Introduction

Charged particle track-structure simulations are a useful tool for the interpretation and understanding of early physical and chemical stages of radiation actions on matter. These Monte-Carlo based simulations provide detailed information on properties of the interactions including spatial distributions of energy deposition, interaction types (ionization, excitation, elastic scattering, charge change, etc.), and radical species produced. This information is used in radiation biology to explore and estimate the effects of radiation quantity and quality on the biological response and to provide detailed information on the initial patterns of radiation damage (Dingfelder 2006; Friedland et al. 2010, 2011).

Monte-Carlo track-structure simulations follow the primary, as well as all (produced) secondary particles, event-by-event, from starting or ejection energies to total stopping. This requires reliable interaction probabilities (cross sections) for all considered scattering events, including ionization, excitation, and charge-changing events of the incident charged particles (electrons, protons, alpha particles, light and heavy ions) with the atoms and molecules of the material under consideration. Liquid water is of special interest since it serves as a substitute for soft tissue.

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This paper will review and summarize different methods of radiation transport models and focus on Monte-Carlo track-structure simulations. It will discuss the calculation of charged particle interaction cross sections within commonly used approximations like the plane-wave Born approximation (PWBA) and discuss the Monte-Carlo track-structure code PARTRAC (Particle Tracks) as an example. Current and future developments as well as challenges in the field of charged particle track-structure simulations like the detailed description of light ion transport will be addressed.

Radiation Transport

Charged particles interact with the surrounding matter, transferring and depositing their energy thru electrostatic force interactions with the orbital electrons, causing excitations, ionizations, and the production of secondary electrons, as well as the excitation or charge changing events of the projectile particle. They also interact with the nuclei of surround atoms, causing projectile and target fragmentations [spallation (i.e., the breakup of the nucleus into lighter pieces)] and the production of secondary particles such as neutrons, protons, mesons, and nuclear recoil atoms. This alters the quality of the radiation field (i.e., the kinetic energy and composition). This work will focus on the interaction of charged particles with the atomic electron system and primarily on ionization and excitation. An introduction to nuclear transport codes and shielding is given elsewhere (Kim et al. 2012).

Radiation transport simulations mainly describe the energy deposition in the target material averaged over a track length of interest. Depending on the length scale, more or less details of the radiation interaction need to be considered. The condensed history approach averages over multiple interactions by determining the average energy loss of the primary radiation over a certain track length travelled and depositing it along this track length. No secondary electron production is considered. Sometimes, fast electrons from hard collisions, also called delta rays, are considered and simulated accordingly. These codes are often referred as general purpose codes. They only require the energy loss per track length, or stopping power, as input. These data can be calculated from simple theoretical models and are tabulated and published for all atomic elements. Composite materials are modeled by using weighted values of stopping powers based on their atomic composition, and by density scaling. Popular codes are PENELOPE (Salvat et al. 2008), the EGS5 code (Hirayama et al. 2005), or the MCNP code system (MCNP 2003). Condensed history codes typically follow electrons down to about 1 keV and can describe spatial resolutions down to ~ 0.1 mm. In contrast, *track-structure* simulations follow the primary particle, as well as all secondary particles produced from starting or ejection energy down to total stopping in an event-by-event manner. Track-structure simulations record the type, location, and energy deposition for each interaction considered; they also follow all created secondary particles in the same way. Track-structure codes are normally specialized to a group of problems and *homemade*. They require a large amount of input data and are not easy to operate. In contrast to general purpose codes that rely on the energy loss per track length (or stopping power), track-structure codes use the total mean free path to determine the location of the next interaction, and the relative total cross sections or inverse mean free paths (IMFP) for all considered interactions (elastic scattering, excitation levels, ionization shells, etc.) to determine the type of interactions. Once the type of interaction is determined the corresponding differential cross sections (e.g., differential in energy transfer, momentum transfer, or scattering angle, depending on the interaction type) are used to determine details of the interaction, including the creation of secondary particles. This is achieved by converting cross section data into normalized cumulative probability distributions and by random sampling. A nice introduction to Monte-Carlo simulation techniques and sampling methods is given by Salvat et al. (2009). The focus of this work is on charged particle track-structure codes used to assess radiation damage to material of biological interest, and specifically using the

PARTRAC code (Dingfelder et al. 2008a; Friedland et al. 2011) as an example. Some other track-structure codes used are the NOREC (New Oak Ridge Electron Transport Code) code (Semenenko et al. 2003; Dingfelder et al. 2008a) and the KURBUC (Kyushu University and Radiobiology Unit Code) code (e.g., Endo et al. 2002). A review of the most common codes is provided by Nikjoo et al. (2006), a good introduction into the theory of track-structure is provided by Paretzke (1987, 1988). Track-structure codes typically model the transport of particles down to 10 eV (or lower) and can describe spatial resolutions down to a nanometer, or even an Angstrom in length.

Charged particles of interest include electrons, positrons, protons, alpha particles, light ions like carbon, and heavy ions, especially high atomic number, high energy (HZE) particles from the galactic cosmic rays. Charged particle kinetic energies range from 1 eV (or lower) for electrons to a GeV per nucleon (or higher) for HZE particles. Two topics of special interest include particles with high relativistic energies and low-energy electrons. Track-structure simulations started out with non-relativistic electron transport modeling but now have been revised to include high relativistic energies. Condensed phase codes (like the Bethe theory) use simplified approaches for calculating the energy loss per track length (or stopping power) and are available in a consistent relativistic formulation. Low-energy electron transport is important as it describes the *track ends* [i.e., the spatial locations of solvated (totally stopped) electrons]. Low-energy electrons also have short mean-free paths and short ranges, which can lead to multiple ionizations or excitations within small volumes. This gives them an enhanced ability to produce an effect, for example, clustered DNA damage (from direct effects, indirect effects, and combinations), including simple and complex double-strand breaks.

Cross Sections

Ionization and excitation cross sections

It is most common to calculate ionization and excitation cross sections for track-structure codes within the PWBA. The PWBA is a first-order perturbation approach and considers the incoming particle as a point particle [electrons, positrons, protons, bare (i.e., fully ionized) ions)] traveling with speeds much faster than the (bound) atomic electrons. The target material is either described by the generalized oscillator strength in the microscopic atomic picture or the dielectric response function (DF) in a macroscopic condensed phase picture. Both quantities can be related to each other. Within the PWBA the nonrelativistic double differential IMFP shown by Equation 1, includes a kinematic factor (T) for the primary particle and an energy loss function [$\varepsilon(\omega,k)$] that is a target material property:

$$\frac{d^2\sum}{d\omega dk} = \frac{1}{\pi a_0 T}\frac{1}{k}\mathrm{Im}\left(\frac{-1}{\varepsilon(\omega,k)}\right). \quad (1)$$

Here, Σ is the IMFP or macroscopic cross section and can be expressed as $N\sigma$, where σ the microscopic cross section, and N the number density of the material under consideration. ω is the frequency related to the energy transfer by $E = \hbar\omega$, where $\hbar$ is the reduced Planck's constant equal to Planck's constant/2π, and k is the wave number related to the momentum transfer in the collision by $q = \hbar k$, a_0 is the Bohr radius, T the kinetic energy of the primary particle, and $\varepsilon(\omega,k)$ the DF of the target material. Im(.) indicates the imaginary part of the argument. The IMFP Σ is then obtained from eq. (1) by integrating over the momentum transfer and energy transfer. The DF is a function of the energy and momentum transfer and is a characteristic function of the target material. It does not depend on the incoming particle. The DF can be obtained from calculations or experimental data, or can be modeled using available experimental information and theoretical constraints. Further details on the

nonrelativistic PWBA theory can be found in Dingfelder et al. (1998, 2008a), and the relativistic description in Fernandez-Varea et al. (2005).

PWBA calculations require a reliable DF or generalized oscillator strength of the material under consideration. Track-structure simulations use liquid water as transport medium. Liquid water is the main constituent of the human body and serves as surrogate for soft biological tissue. Also, DNA in the cell nucleus is surrounded by a water shell. Locations of ionizations and excitations in the surrounding water serve as starting point for the chemical stage of the biological damage of radiation to DNA (i.e., the formation and transport of radicals). The DF of liquid water is modeled by representing the DF by a superposition of functions representing individual excitation and ionization levels in the optical limit (i.e., for momentum transfer zero) and fitting it to available experimental data. Then, a suitable extension algorithm based on physical principles (and experimental data) is applied to model the DF or Bethe surface. Currently, there are three major models for the DF used in cross-section calculations: The PARTRAC model (Dingfelder et al. 1998, 2008a; Dingfelder and Inokuti 1999), the OREC (Oak Ridge Electron Transport Code) model (Ritchie et al. 1991; Dingfelder et al. 2008a), and the Emfietzoglou model (Emfietzoglou et al. 2005; Emfietzoglou and Nikjoo 2007). They all consider five excitation levels of water and five ionization levels and fit the optical DF to experimental data while retaining theoretical constraints such as sum rules and average quantities. The OREC and PARTRAC models are mainly based on the old optical reflectance measurements by Heller et al. (1974), carried out at a liquid water surface, while the Emfietzoglou model is mainly based on the new measurement of the Bethe ridge by Hayashi et al. (2000). All three models use slightly different extension algorithms leading to slightly different ionization and excitation cross sections, especially at low incident energies. However, at these energies, it is common to apply additional corrections to account for the nonvalidity of the PWBA and for higher-order perturbation effects.

Alternative methods to calculate ionization and excitation cross sections include the Bethe approximation, or semi-empirical models. The Bethe formula is asymptotic to the PWBA at high energies and requires only optical information (i.e, the dielectric function in the optical limit, or optical oscillator strengths) (Dingfelder et al. 1998; Fernandez-Varea et al. 2005). However, it only provides energy loss differential cross sections and no information about momentum transfer. Angular deflections need to be determined separately, normally within the binary-collision theory. Semi-empirical models are either based on theoretical asymptotic behavior or simple functional dependencies. Parameters are fitted to experimental data. Often, when experimental data are unavailable for certain materials, parameters are inter- or extrapolated within known values. Like the Bethe approximation most semi-empirical models only provide energy differential cross sections. An often used example for a semi-empirical model is the Rudd model for proton transport (e.g., Dingfelder et al. 2000; Plante and Cucinotta 2008). Semi-empirical models may also be useful if materials other than liquid water are required for track-structure simulations. Using the PWBA to create new data sets of interaction cross sections for new materials is cumbersome. Currently, there are efforts on the way to create datasets for metallic calcium, a major component of cortical and trabecular bone (Jorjishvili et al. 2009), and for thin metal foils like copper and gold (Travia and Dingfelder 2011).

The Bethe approximation and semi-empirical models are in general numerically simpler and easier to use. However, like the PWBA, their applicability is limited. The PWBA is applicable if the incoming charged particle is fast compared to the bound atomic electrons. In the case of electron transport, PWBA cross sections are justified for electron energies above 300–500 eV, and in the case of proton transport for energies above 100 keV. Below these energies, corrections need to be applied. This can be done by semi-empirical

correction factors (Dingfelder et al. 1998), using more sophisticated theories like the distorted wave Born approximation (Champion 2003; Segui et al. 2003), or higher order Born corrections (Emfietzoglou and Nikjoo 2007). In the case of electron transport, exchange effects need to be considered as well as relativistic effects for energies above 5 keV.

Elastic cross sections

Charged particles also scatter elastically with the target. In these collisions, the primary particle does not lose energy, but changes its direction. Elastic scattering events also prolong the mean free path. They are important mainly at lower energies and for electron or positron impact. A current review of the state-of-the-art calculations and cross sections for electron and positron elastic scattering was recently published by the International Commission on Radiation Units and Measurements as Report 77 (ICRU 2007). Protons and heavier particles also scatter elastically. However, due to the large mass difference, the momentum transfer is very small, resulting in very small deviation angles. Since elastic scattering for protons only becomes important for very low energies (below kiloelectron volt levels) it is in general neglected.

Ion transport

Ionization and excitation cross sections for bare heavier ions [i.e., fully ionized, including for example, alpha particles (He^{2+}), carbon ions (C^{6+}) or iron ions (HZE particles; Fe^{26+})] can be obtained within the PWBA or Bethe theories. Both theories allow the calculation of ionization and excitation cross sections from the corresponding proton impact cross sections by using scaling laws for velocity and charge. The double differential IMFP for an ion with nuclear charge Z and velocity v is given by:

$$\frac{d^2\sum_{\text{ion}}}{d\omega dk}(v)=Z^2\frac{d^2\sum_{\text{proton}}}{d\omega dk}(v). \quad (2)$$

It scales from the corresponding proton IMFP at the same velocity with Z^2. This approximation is valid as long as the ions remain fully ionized. This is the case if the velocity of the ion remains high. However, these ions are atomic systems and can pick up electrons from the target material when slowing down. The atomic orbital electrons then shield part of the positive nuclear charge, so that the incoming particle only sees an effective (screened) Coulomb potential. This scenario is referred to as partially ionized or partially dressed. In this case, the total nuclear charge Z is replaced by an effective or average charge Z_{eff}, which depends on the ion's kinetic energy, and scaling is applied only to the (single) energy differential mean free paths instead of the double differential ones. The effective or average charge Z_{eff} is commonly determined using semi-empirical formulas like the Barkas formula (Barkas 1963), or calculated using atomic wave functions as the CasP (convolution approximation for swift particles) code (e.g., Schiewitz and Grande 2001, 2009 and references therein) does.

It should be noted that the PWBA as described above refers to single electron processes only. This includes single ionization or single excitation of the target and a point like description of the projectile using an effective or average charge. However, the ion is like the target material an atomic system, which can also be excited or ionized. In addition, a moving atomic system can capture electrons from the surrounding or lose electrons by stripping. Both processes change the charge state of the ion. While multi-ionization by electron impact is unlikely and therefore neglected, it is much more probable for heavier ions and should be considered. In summary, in (heavy) ion: target collisions, multi-event

processes should be considered, especially at lower energies when the ion is already partially dressed. Events may include single or multi ionizations and excitations of the target and the projectile, as well as single or multiple electron capture and loss processes of the projectile. Furthermore, all processes are charge state dependent: ionization probabilities for alpha particles (He^{2+}), He^{+}, and neutral He^{0} differ, as well as the electron capture probability from neutral He^{0} and He^{+}. Currently, the track-structure code PARTRAC simulates protons and alpha-particle transport from relativistic energies down to 1 keV. In the case of proton transport, single ionization and excitation of the protons and neutral hydrogen is considered together with the one electron loss of the proton and the one electron capture by the neutral hydrogen atom. In the case of alpha particle transport, single ionization and excitation by He^{2+}, He^{+}, and He^{0} is considered. Charge-changing processes include one electron loss from He^{2+} and He^{+}, two electron loss from He^{2+}, one electron capture by He^{0} and He^{+}, and two electron capture by He^{0}. In both cases, all cross sections below 1 MeV are based on semi-empirical models and experimental data, mainly for water vapor as target. Details can be found in the literature (Dingfelder et al. 2000, 2005; Friedland et al. 2011). Furthermore, experimental and theoretical work is in progress to better understand the interactions of partially dressed ions. Carbon is of special interest due to the operation of carbon cancer irradiation facilities in Europe and Japan. Liamsuwan et al. (2011) for example report on a project to calculate single electron capture and loss cross sections for various charge states of carbon with water. Ledakir et al. (2009) study single-electron loss cross sections of DNA and RNA bases impacted by energetic multicharged ions, including carbon.

Discussion

Monte-Carlo track-structure codes describe well the initial pattern of energy depositions in material of biological interest within the limitations of the validity of their transport models and input data [i.e., for point particles (electrons, and bare, fully ionizied ions)] at moderate and high energies. Semi-empirical models based on scarce experimental data and theoretical information are used for the transport of low-energy electrons, protons, and alpha particles, and for the transport of dressed ions at moderate energies. All these approaches are based on a one-active-electron model, and multi-electron processes are neglected. This is valid for electron impact, where double and multi-ionization events are rare and can be neglected. However, in the case of ion impact, multi-electron events consisting of single or multiple ionizations, excitations, and electron capture and loss, and combinations of these, may need to be considered. Modeling the transport of dressed ions will benefit from more theoretical calculations and experimental data in this area, especially for biologically relevant materials like liquid water. Only limited data for light ions (alpha particle, lithium, carbon) exist for some gas phase targets.

Relativistic theories describe total cross sections and stopping powers well. However, details on differential cross sections at relativistic energies may not be as accurate as desired. This applies especially to (heavy) ion induced secondary electron emission spectra, where simple semi-empirical models are used. Again, only limited experimental data exist to benchmark ion induced secondary electron emission spectra at higher (relativistic) energies.

Recent experiments on electron emission from thin layers of amorphous solid water after proton impact (Toburen et al. 2010) allows testing the low-energy electron transport model of liquid water as implemented in track-structure codes by simulating the experiment (Dingfelder et al. 2008b). The PARTRAC track-structure model reproduces secondary electron emission yields for electron energies above 50 eV, but clearly overestimates the yields at lower energies. Work is in progress to improve the electron transport model at low energies.

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