Medical Physics Letter

On the Monte Carlo simulation of electron transport in the sub-1 keV energy range

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(Received 28 February 2011; revised 26 May 2011; accepted for publication 13 June 2011; published 21 July 2011)

Purpose: The validity of "classic" Monte Carlo (MC) simulations of electron and positron transport at sub-1 keV energies is investigated in the context of quantum theory.
Methods: Quantum theory dictates that uncertainties on the position and energy-momentum four-vectors of radiation quanta obey Heisenberg's uncertainty relation; however, these uncertainties are neglected in "classical" MC simulations of radiation transport in which position and momentum are known precisely. Using the quantum uncertainty relation and electron mean free path, the magnitudes of uncertainties on electron position and momentum are calculated for different kinetic energies; a validity bound on the classical simulation of electron transport is derived.
Results: In order to satisfy the Heisenberg uncertainty principle, uncertainties of 5% must be assigned to position and momentum for 1 keV electrons in water; at 100 eV, these uncertainties are 17 to 20% and are even larger at lower energies. In gaseous media such as air, these uncertainties are much smaller (less than 1% for electrons with energy 20 eV or greater).
Conclusions: The classical Monte Carlo transport treatment is questionable for sub-1 keV electrons in condensed water as uncertainties on position and momentum must be large (relative to electron momentum and mean free path) to satisfy the quantum uncertainty principle. Simulations which do not account for these uncertainties are not faithful representations of the physical processes, calling into question the results of MC track structure codes simulating sub-1 keV electron transport. Further, the large difference in the scale at which quantum effects are important in gaseous and condensed media suggests that track structure measurements in gases are not necessarily representative of track structure in condensed materials on a micrometer or a nanometer scale. [DOI: 10.1118/1.3608904]

Key words: Monte Carlo, radiation transport, low-energy electron, trajectory simulation

Monte Carlo (MC) simulations of radiation transport at sub-1 keV energies are of increasing importance as researchers strive to understand radiation-induced damage on short length scales, e.g., interactions of radiation with cellular components, particularly DNA. Studies at these low energies and short length scales often lie in the realm of "microdosimetry" or even "nanodosimetry" and find broad application across medical physics from radiation therapy to imaging, radiation protection, radiobiology, and radiochemistry. Simulations at sub-1 keV energies have traditionally been performed with specific-purpose packages (see Nikjoo *et al.*[1] and references therein); more recently, some general purpose codes have extended their range of applicability to electron volt energies. In particular, the PENELOPE package allows the simulation of electron/positron and photon transport down to 50 eV (Refs. 2 and 3) and there is an active group developing a GEANT4-based application for the simulation of radiation interactions with biological systems at the nanometer level, "GEANT4-DNA", which currently simulates electrons to 10 eV.[4] The purpose of this article is to explore the validity of the trajectory MC simulation of electron and positron transport for sub-1 keV kinetic energies in the context of quantum theory.

In MC radiation transport codes, radiation quanta (electrons, positrons, etc.) are considered to be pointlike objects with knowledge of position and energy-momentum four-vectors limited only by the computer representation of floating point numbers. Particle trajectories consist of a sequence of free-flight segments and interaction sites which are known to within the precision permitted by the computer. In reality, radiation quanta obey the laws of quantum physics, and thus the transport treatment must satisfy Heisenberg's uncertainty principle,

$$\Delta x \, \Delta p \geq \hbar \,, \tag{1}$$

which is fundamental to quantum theory.[5] Thus, for rigorous simulations of radiation transport, Δx, Δp satisfying Eq. (1) must be assigned to particle position and momentum, respectively. As long as Δx, Δp satisfy the uncertainty relation (1) and simultaneously

$$\Delta x \ll s\,, \quad \Delta p \ll p\,, \tag{2}$$

where s is the relevant length scale and p is the particle momentum, then the classic MC simulation of particle transport (in which Δx, Δp are neglected) is adequate. However, if Δx, Δp become comparable with s and p in order to satisfy the uncertainty principle (1), then the classical picture breaks down and classical MC simulations no longer provide an accurate description of particle transport.

The transition between the "high energy" regime in which the classical picture of pointlike particles is acceptable and the "low energy" regime in which the classical picture breaks down and quantum physics must be considered may be quantified as follows. Suppose that the maximum uncertainties in position and momentum for which the classical picture holds are

$$\Delta x = \varepsilon s\,, \quad \Delta p = \varepsilon p\,, \tag{3}$$

respectively, for a small (dimensionless) quantity ε. Inserting these expressions into the uncertainty relation (1) and using the low energy, nonrelativistic approximation for the kinetic energy, $E \simeq p^2/2m$, where m is the particle mass, one finds that ε must satisfy

$$\varepsilon \geq \varepsilon_c \equiv \sqrt{\frac{\hbar}{s\sqrt{2mE}}}. \tag{4}$$

The critical value ε_c represents a lower bound on the fractional uncertainties on the position (relative to length scale s) and on the momentum of a massive radiation quantum of kinetic energy E for an application characterized by length scale s. In order for the classical MC transport treatment to be valid, the uncertainties Δx, Δp must be greater than this lower bound while simultaneously satisfying Eq. (2). This is generally the case for radiotherapy treatment planning calculations (for which $s \sim 1$ mm and energy cutoffs $E \geq 1$ keV; $\varepsilon_c < 8 \times 10^{-5}$); however, it is not always true for lower energy simulations (e.g., for microdosimetry), as follows.

Low energy simulations are typically concerned with particle track structure.[1] In this context, the relevant length scale for a particle of kinetic energy E is set by the macroscopic cross section $\Sigma(E)$ (number of interactions per unit length) or, equivalently, the mean free path $s = 1/\Sigma(E)$. The critical value ε_c is then

$$\varepsilon_c = \sqrt{\frac{\hbar\,\Sigma(E)}{\sqrt{2mE}}}. \tag{5}$$

The parameter ε_c will generally be small for heavy and/or neutral particles; hence classical MC simulations will generally be valid, and thus simulation of, e.g., neutrons to thermal energies is acceptable. However, electrons and positrons are light and have large cross sections, leading to larger values of ε_c at lower energies and implying a lower energy bound on the validity of classical MC simulations.

Figure 1 presents ε_c as a function of kinetic energy for electrons in water (unit density liquid, vapor, and solid–ice) and in air. For electron kinetic energies of 10 keV and above, ε_c is generally 0.01 or less; hence 1% uncertainties on electron momentum and position (relative to the length scale s) satisfy the uncertainty principle. For 1 keV electrons in liquid water ε_c is 5% and increases to 17%–20% at 100 eV. Below 100 eV, ε_c is even larger. Hence, for most applications of interest for radiotherapy treatment planning ($E \geq 1$ keV), ε_c is sufficiently small that classical MC simulations are adequate. However, for electrons in water with kinetic energies of 1 keV or less, ε_c becomes significant and the uncertainties Δx, Δp no longer satisfy Eq. (2).

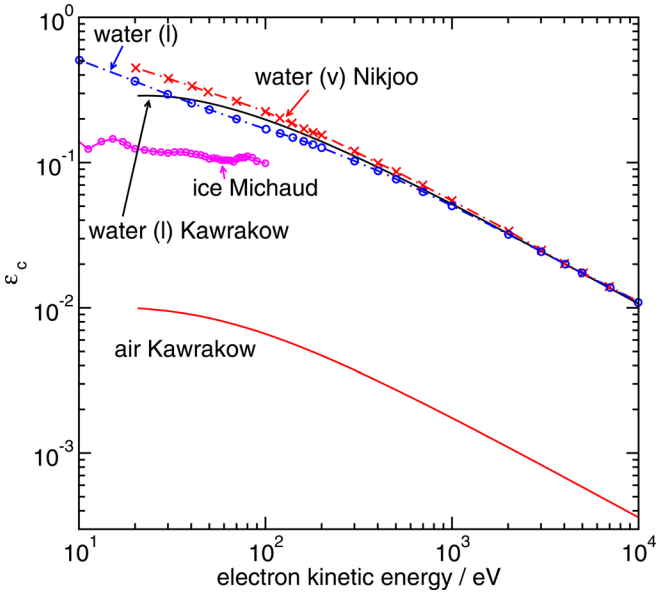

FIG. 1. The critical fractional uncertainty on position and momentum, ε_c, as a function of electron kinetic energy. Fractional uncertainties on electron position (relative to the length scale, s) and momentum must be greater than ε_c to satisfy the uncertainty principle. Values of ε_c are calculated using cross sections from different sources: the line labeled "water (l)" employs cross sections for liquid water with the elastic scattering contribution from the model of Champion *et al.* (Ref. 17) and inelastic scattering contribution from Emfietzoglou and Nikjoo (Ref. 8); the Kawrakow model (Ref. 12) provides data for liquid water and air; cross sections in ice are from experimental measurements of Michaud *et al.* (Ref. 10); finally, data for gaseous water (vapor) are from Nikjoo *et al.* (Ref. 1). The calculations with water in different phases assume unit density.

For electrons in air, $\varepsilon_c < 1\%$ due to the smaller interaction cross sections in air than in water, justifying the simulation of electrons down to electron volt energies in air in, e.g., proportional counters. In experimental micro and nano-dosimetry, energy deposition in a tissue volume of diameter d is often measured in a tissue-equivalent gas volume of diameter $d\rho_t/\rho_g$ where ρ_t and ρ_g are the tissue and gas densities, respectively. In this way, measurements on millimeter scales in gas media are used to study track structure in condensed media on a micrometer or even nanometer scale.[6,7] However, the large difference between ε_c for a gaseous medium such as air and a condensed material such as water indicates that electron track structure measurements performed in a low density medium on a millimeter scale are not necessarily representative of the track structure in a condensed medium on a micrometer scale.

The variation in the parameter ε_c in Fig. 1 derived from different water cross section data sets reflects the large uncertainty in sub-1 keV cross sections. Notable variations in electron cross section result from different model approximations.[8] Uncertainties of order 20%–40% or larger are

expected between 100 eV and 1 keV; cross section calculations below 100 eV are highly uncertain.[1,8] The line labeled "water (l)" in Fig. 1 for liquid water was calculated using inelastic cross sections published by Emfietzoglou and Nikjoo in 2005 (Ref. 8); these authors subsequently reported inelastic cross sections for a subset of electron energies (100 eV to 1 keV) based on an improved dielectric description of the Bethe surface.[9] Values of ε_c calculated with the later results for 100 eV to 1 keV electrons are nearly coincident with the circles on the line labeled "water (l)" calculated using the earlier data. Results are similar if the liquid water cross sections presented by Incerti *et al.*[4] and employed in "GEANT4-DNA" are used to calculate ε_c. The overall error on the cross sections measured in amorphic ice is ±30%–45%.[10] Although the uncertainties in cross sections affect the precise value of ε_c derived at a particular energy, it is generally observed that significant uncertainties on position and momentum are required for sub-1 keV electrons in water.

The exact value of the lower bound on the product of position and momentum uncertainties depends on the interpretation of the uncertainties which is an active area of research in fundamental quantum theory.[11] For multiple measurements, the uncertainty principle may be written as $\sigma_x\sigma_p \geq \hbar/2$, where σ_x is the standard deviation of the position measured for a sample of particles prepared in a wave function ψ while σ_p is the standard deviation of the momentum measured for a second sample of particles also prepared in the state ψ.[11] Schürmann and Hoffmann recently showed that particles initially prepared with a projection in Δx satisfy $\sigma_p\Delta x \geq \pi\hbar$.[11] Particles localized to within a finite interval Δx have their momentum uncertain by Δp where the product satisfies Eq. (1).[5] For particle trajectory simulations, the uncertainties are not standard deviations corresponding to multiple measurements of position and momentum; hence $\Delta x\Delta p \geq \hbar$ is employed in the present work. Regardless of the exact value of the lower bound on the product of uncertainties, the uncertainties Δx, Δp must be significant for sub-1 keV electrons in water.

Electron "spread" is characterized by the de Broglie wavelength $\lambda\,(\mathrm{nm}) = \sqrt{1.50/E(\mathrm{eV})}$ (valid for electrons with kinetic energy $E \ll mc^2$) which grows from 0.04 nm for a 1 keV electron, to 0.12 nm (100 eV), to 0.17 nm (50 eV), and to 0.39 nm at 10 eV. As electron energy decreases below 1 keV, its de Broglie wavelength becomes a significant fraction of the mean free path, s; e.g., for a 100 eV electron, $\lambda/s \sim 0.24$ to 0.32 based on the cross sections of Kawrakow for liquid water[12] and Nikjoo *et al.* for water vapor,[1] respectively, both with unit density. Furthermore, for sub-1 keV energies, the de Broglie wavelength is comparable to the interatomic spacing (~2–3 Å in condensed media) and coherent scattering from multiple centers becomes appreciable:[2] the classical trajectory description ceases to be applicable.

Related issues regarding the delocalization of energy in ionizing radiation, quantum effects in radiation transport for low energy electrons, and the quantum uncertainty principle have been considered elsewhere. Kaplan and Miterev[13] demonstrated that a particle of speed v_x transferring energy ΔE has a position uncertainty $\Delta x \geq \hbar v_x/\Delta E$ due to the quantum uncertainty principle. Subsequent work in radiochemistry has considered this delocalization and that due to collective excitations of molecular species and the diffusion of free radicals produced in interactions.[14,15] In the context of MC track structure calculations, Emfietzoglou *et al.* discussed the minimum delocalization of an energy-loss event due to the uncertainty principle, noting that uncertainties are of order 3 nm along the particle's trajectory and 10 nm radially at the interaction site and concluding that results pertaining to ~nanometer-sized regions should be interpreted with care.[16] Salvat *et al.* stated that results from simulations with any MC trajectory code for energies below 1 keV should be considered "to have only a qualitative (or, at most, semiquantitative) value" due to the fact that interaction models become less accurate as electron energy decreases and when the de Broglie wavelength is comparable to the interatomic spacing.[2] In a series of papers (Ref. 15 and references therein), Liljequist has compared trajectory transport of very low energy (generally sub-20 eV) electrons with full quantum modeling for certain simple configurations and has demonstrated that the magnitude of quantum effects depends on the assumed structure of the transport medium and the approximations involved in the transport treatment.

The results of the present work suggest that the classical MC transport treatment is incorrect for sub-1 keV electrons and positrons in condensed water (often used as a biological tissue surrogate) as uncertainties on position and momentum must be large in order to satisfy the uncertainty principle. The quantum properties of electrons and positrons should not be neglected at these short length scales and low energies in water or other tissuelike media. Simulations of radiation transport which do not reflect the quantum nature of electrons and positrons are not faithful representations of the physical reality at these low energies, calling into question the results of MC track structure codes for sub-1 keV electron and positron transport. The present work underlines the need for a transport treatment consistent with quantum theory for low energy electrons in condensed media. It also suggests that measurements of electron track structure in gaseous media on millimeter length scales may not be representative of track structure in condensed media on micrometer or nanometer scales as quantum effects differ in the two cases.

ACKNOWLEDGMENTS

RMT acknowledges support from the Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grant program and the Carleton University Research Office.

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