Monte Carlo simulation of coherently scattered photons based on the inverse-sampling technique

Wazir Muhammad, Ying Liang, Gregory R. Hart, Bradley J. Nartowt and Jun Deng*

Department of Therapeutic Radiology, School of Medicine, Yale University, 15 York Street, New Haven, CT 06510-3221, USA. *Correspondence e-mail: jun.deng@yale.edu

Received 18 October 2019

Accepted 25 October 2019

Edited by A. Altomare, Institute of Crystallography - CNR, Bari, Italy

Keywords: Rayleigh scattering; Monte Carlo simulation; atomic form factors; anomalous scattering factors; total cross sections; differential cross sections.

The acceptance–rejection technique has been widely used in several Monte Carlo simulation packages for Rayleigh scattering of photons. However, the models implemented in these packages might fail to reproduce the corresponding experimental and theoretical results. The discrepancy is attributed to the fact that all current simulations implement an elastic scattering model for the angular distribution of photons without considering anomalous scattering effects. In this study, a novel Rayleigh scattering model using anomalous scattering factors based on the inverse-sampling technique is presented. Its performance was evaluated against other simulation algorithms in terms of simulation accuracy and computational efficiency. The computational efficiency was tested with a general-purpose Monte Carlo package named *Particle Transport in Media (PTM)*. The evaluation showed that a Monte Carlo model using both atomic form factors and anomalous scattering factors for the angular distribution of photons (instead of the atomic form factors alone) produced Rayleigh scattering results in closer agreement with experimental data. The comparison and evaluation confirmed that the inverse-sampling technique using atomic form factors and anomalous scattering factors exhibited improved computational efficiency and performed the best in reproducing experimental measurements and related scattering matrix calculations. Furthermore, using this model to sample coherent scattering can provide scientific insight for complex systems.

1. Introduction

Elastic (coherent) scattering of photons is produced by multiple mechanisms (*i.e.* Rayleigh, nuclear Thomson and Delbrück scattering). It is the dominant mode of interaction in the low-photon-energy regime (from approximately 1 keV to a few megaelectronvolts) (Carter & Cashwell, 1975). Amongst these mechanisms, Rayleigh scattering with bound atomic electrons gives a major contribution to the cross section. In the stated energy regime, the study of Rayleigh scattering is an important and effective probe in various experimental fields such as material analysis, structural chemistry and biology (Bradley *et al.*, 1999; Cianci *et al.*, 2005; Muhammad, 2012; Muhammad & Lee, 2013a), and medical physics and radiation protection (Roy *et al.*, 1993, 1999; Kissel *et al.*, 1995; Amako *et al.*, 2005; Muhammad, 2012; Muhammad & Lee, 2013a,b,c). It is mainly used to investigate the inner structure of atoms and macromolecules and to determine photon mass attenuation coefficients (Batič *et al.*, 2012). Its theoretical predictions are especially accurate due to the availability of synchrotron sources (Roy *et al.*, 1993; Kissel *et al.*, 1995; Muhammad & Lee, 2013b).

A number of general-purpose Monte Carlo (MC)-based radiation transport packages implementing Rayleigh scattering are available (*e.g.* MCNP, GEANT4, PENELOPE,

EGSnrc etc.) (Muhammad, 2012; Muhammad & Lee, 2013a). Most of these codes adopt the acceptance–rejection method to simulate Rayleigh scattering of photons based on atomic form factors (AFFs) while ignoring anomalous scattering factors (ASFs). ASFs are approximated to zero as they are considered angle-independent. However, a comprehensive study by Muhammad & Lee (2013a) showed that the angular distribution of coherently scattered photons is influenced by ASFs. Consequently, in some of these packages, ASFs are included in the total atomic cross sections. However, the total cross section of an interaction only gives the probability of that interaction (*i.e.* the number of photons that undergo the interaction). This article is about the implementation of the available AFFs and ASFs in the scattering distribution while setting aside the issue of how these factors are calculated.

A very recent study reporting an update to *GEANT4*'s coherent scattering model for photons omits ASFs, despite stating their essentiality to the accurate calculations of Rayleigh scattering amplitudes (Turturica *et al.*, 2018). However, it is presumed that that work is more focused on photon energies >1 MeV, where the contribution of Rayleigh scattering is outweighed by all the other elastic scattering processes. The other reason may be the relevance of ASFs in specific calculations such as photon beam attenuation, response of detectors, dose calculations *etc.* For these specific parameters, the ASF contributions might not be significant, which is why elastic scattering of photons has been largely ignored for the last two decades. Another issue is the unavailability of ASFs for compounds. These are the main reasons that general-purpose codes such as *GEANT4* and *MCNP* simplify this important interaction of photons. However, the emergence of high-energy and high-flux laser Compton scattering γ sources brings this matter into focus (Surzhykov *et al.*, 2018). Therefore, implementing a more realistic model for the Rayleigh scattering of photons is required.

A detailed summary and comparison of the MC models implemented in these codes is given by Muhammad & Lee (2013a). That study also highlighted discrepancies among the MC models, as well as the available state-of-art theoretical models, and an updated MC code for scattering was proposed by the authors. This updated MC code was capable of simulating the scattering amplitudes with high accuracy compared with the available well known MC packages. However, the validity of their code has not yet been documented in a general-purpose MC package. It is also expected that the computational efficiency of conventional acceptance–rejection techniques is decreased by the complexity of a given physical model.

Coherent scattering includes Rayleigh, nuclear Thomson and Delbrück scattering. This paper is focused on modeling the Rayleigh scattering of photons because of its major contribution to elastic scattering. The focus is to identify an optimal model for the Rayleigh scattering of photons which is fitted to the reference data of the United States National Institute of Standards and Technologies (NIST) (Berger, Coursey *et al.*, 1999; Berger, Hubbell *et al.*, 1999) and of the

International Commission on Radiation Units and Measurements (ICRU) (Berger *et al.*, 1993). The model should be easy to implement in our new general-purpose MC package (named *Particle Transport in Media* or *PTM*; Liang *et al.*, 2019) without losing computational efficiency. The use of *PTM* for similar studies is reported by Surzhykov *et al.* (2018). A detailed comparison of the model with available experimental results is made in this paper to evaluate its accuracy. The computational efficiency of the traditional acceptance–rejection technique is compared with that of the inverse-transform sampling method.

2. Materials and methods

2.1. Physical model of Rayleigh scattering

A photon undergoing Rayleigh scattering changes its direction with negligible energy transfer to the bound electron in the target atom (Kane *et al.*, 1986; Roy *et al.*, 1993). The widely used approach for estimating the Rayleigh scattering cross section is the form-factor approximation (Kissel *et al.*, 1995). Based on this approximation, a differential cross section (DCS) of Rayleigh scattering can be expressed mathematically as follows (Roy *et al.*, 1999; Chadwick *et al.*, 2011; Salvat *et al.*, 2011):

$$\frac{d\sigma_{\text{coh}}(Z, E, \theta)}{d\theta} = \frac{r_0^2}{2} (1 + \cos^2 \theta) 2\pi \sin \theta F^2(Z, E, \theta). \quad (1)$$

Here, $r_0 = 2.82 \times 10^{-13}$ cm, Z and E are the classical electron radius, the atomic number of the scattering atom and the energy of the incident photon, respectively. $F(Z, E, \theta)$ is the complex-valued atomic form factor (CFF) which can be written as (Bergstrom *et al.*, 1997; Roy *et al.*, 1999; Chantler, 2000; Chadwick *et al.*, 2011; Salvat *et al.*, 2011; Muhammad & Lee, 2013a)

$$F^2(Z, E, \theta) = [f_0(Z, \theta) + f'(Z, E)]^2 + f''^2(Z, E). \quad (2)$$

Here, $f_0(Z, \theta)$ is the AFF (Hubbell *et al.*, 1975; Hubbell & Øverbø, 1979; Schaupp *et al.*, 1983; Kane *et al.*, 1986) while $f'(Z, E)$ and $f''(Z, E)$ are the real and imaginary parts of the complex-valued scattering amplitude of the ASF, respectively. The ASF has a more prominent role around the characteristic absorption edges of an atom (Pratt *et al.*, 1993; Kissel *et al.*, 1995; Chantler, 2000; Tartari *et al.*, 2002; Cianci *et al.*, 2005; Muhammad & Lee, 2013a).

2.2. Data availability

Tabulated values of $f_0(Z, \theta)$ (AFFs) as a function of momentum transfer, q ($\text{\AA}^{-1}$) = $\lambda^{-1} \sin(\theta/2)$ [where λ ($\text{\AA}$) is the wavelength of the incident photon], for all neutral atoms listed in the Periodic Table are available in the literature (Cromer & Liberman, 1970; Hubbell & Øverbø, 1979; Henke *et al.*, 1981, 1993; Cromer, 1983; Schaupp *et al.*, 1983; Kane *et al.*, 1986; Cullen *et al.*, 1997; Batič *et al.*, 2012; Cullen, 2018). Cullen (1989, 2018) computed ASF values using a relativistic dispersion relation (Pratt *et al.*, 1993). Tabulated values of ASFs [$f'(Z, E)$ and $f''(Z, E)$] as a function of incident photon

energy for all listed elements are also available in the literature (Cromer & Liberman, 1970; Henke *et al.*, 1981, 1993; Cromer, 1983; Cullen, 2018).

Microscopically speaking, the total elemental cross section σ_t for photons consists of four components, *i.e.* Rayleigh scattering σ_{coh} , Compton scattering σ_{cs} , photoelectric effect σ_{pe} and pair production σ_{pp} :

$$\sigma_t = \sigma_{\text{coh}} + \sigma_{\text{cs}} + \sigma_{\text{pe}} + \sigma_{\text{pp}}. \quad (3)$$

Macroscopically speaking, the total cross section of an element is $\Sigma = n_\rho \sigma_t$, where n_ρ is the number density of the element and σ_t is the microscopic cross section. For a mixture of elements or compounds, the macroscopic cross section is

$$\Sigma = N_1 \sigma_{t,1} + N_2 \sigma_{t,2} + N_3 \sigma_{t,3} \dots = \Sigma_1 + \Sigma_2 + \Sigma_3 + \dots, \quad (4)$$

or

$$\Sigma = \sum_{j=1}^i N_j \sigma_{t,j} = \sum_{j=1}^i \Sigma_j, \quad (5)$$

where Σ_j is the macroscopic cross section of the j th element or compound, and the values of N_j are dependent upon the composition of the mixture and the atomic weights and densities of the constituents. Essentially, the microscopic approach will lead to the macroscopic approach, and both approaches are the same.

To model a compound, *PTM* considers the individual atoms of a compound instead of the compound as a new material, so there is no need to include the ASFs for compounds in our data library. For example, if a collision occurs in a zone that has more than one element (*i.e.* a compound), the probability distribution function (PDF) for sampling an element i with total macroscopic cross section $\sum_i$ and a number m of elements in the zone is

$$p_i = \frac{\sum_{j=1}^m \Sigma_j}{\sum_{j=1}^m \Sigma_j}. \quad (6)$$

If a uniform random number, ξ , satisfies $P_{i-1} < \xi < P_i$, then element i is sampled. Here, $P_i = p_1 + p_2 + \dots + p_i$ is the cumulative distribution function (CDF) across sampling elements.

3. MC simulation of Rayleigh scattering

3.1. Acceptance–rejection technique

Rayleigh scattering is modeled with the form-factor approximation using equation (1). The direction of the scattered photon is sampled using the well known acceptance–rejection technique (Muhammad, 2012; Muhammad & Lee, 2013a,b,c). The PDF of the polar angle interval $d\theta$ around θ can be written as (Carter & Cashwell, 1975; Salvat *et al.*, 2011; Kling *et al.*, 2014)

$$p(\theta) d\theta = \frac{d\sigma_{\text{coh}}(Z, E, \theta) d\theta}{\sigma_{\text{coh}}(Z, E)} = \frac{r_0^2 (1 + \cos^2 \theta) F^2(Z, E, \theta) 2\pi \sin \theta d\theta}{2 \sigma_{\text{coh}}(Z, E)}. \quad (7)$$

Here, σ_{coh} is the total Rayleigh scattering cross section, while the azimuthal angle φ is uniformly distributed within the interval $(0, 2\pi)$.

Considering $\mu = \cos \theta$ and $\alpha = E/(m_0 c^2)$, where m_0 is the electron rest mass and c is the speed of light, and using half-angle $(\theta/2)$ trigonometric relations [*i.e.* $\sin^2(\theta/2) = (1 - \cos \theta)/2$], the momentum transfer $q = \sin(\theta/2)/\lambda$ is expressed as

$$q^2 = (k\alpha)^2 (1 - \mu), \quad \mu = 1 - \left[\frac{q^2}{(k\alpha)^2} \right], \quad \frac{d\mu}{(dq)^2} = \frac{-1}{(k\alpha)^2}. \quad (8)$$

Here, $k = 10^{-8} m_0 c / (2^{1/2} h) = 29.1445 \text{ \AA}^{-1}$, where h is Planck's constant (Carter & Cashwell, 1975; Salvat *et al.*, 2011; Kling *et al.*, 2014).

Using equations (8), the PDF in equation (7) can be rewritten as follows:

$$p(\mu) dq^2 = \frac{\pi r_0^2}{(k\alpha)^2 \sigma_{\text{coh}}(Z, \alpha)} (1 + \mu^2) F_0^2(Z, q) dq^2. \quad (9)$$

Now, for arbitrary q^2 and for maximum momentum transfer $\bar{q}^2$ (*i.e.* $q = 2^{1/2} k\alpha$), we have the following relations:

$$A(Z, E, q^2) = \int_0^{q^2} F^2(Z, E, q) Z^{-2} dq^2, \quad (10)$$

$$\bar{A}(Z, E, \bar{q}^2) = \int_0^{\bar{q}^2} F^2(Z, E, q) Z^{-2} dq^2, \quad (11)$$

allowing equation (9) to be rewritten as (Muhammad, 2012; Muhammad & Lee, 2013a,b,c)

$$p(q) dq^2 = \frac{2\pi r_0^2 z^2 A(Z, E, \bar{q}^2)}{(k\alpha)^2 \sigma_{\text{coh}}(Z, E)} \left(\frac{1 + \mu^2}{2} \right) \frac{F^2(Z, E, q) dq^2 z^{-2}}{A(Z, E, \bar{q}^2)} = C_0(Z, E) G(q^2) Q(q^2) dq^2. \quad (12)$$

Here,

$$Q(q^2) = \frac{F^2(Z, E, q) z^{-2}}{\bar{A}(Z, E, \bar{q}^2)}, \quad (13)$$

$$G(q^2) = \frac{1 + \mu^2}{2} \quad (14)$$

and

$$C_0(Z, E) = \frac{4\pi r_0^2 z^2 \bar{A}(Z, E, \bar{v}^2)}{\sigma_{\text{coh}}(Z, E) \bar{v}^2}. \quad (15)$$

A random number in the range 0 to 1, *i.e.*

$$\xi = \int_0^{q^2} Q(q^2) dq^2 = \frac{A(Z, E, q^2)}{\bar{A}(Z, E, \bar{q}^2)}, \quad (16)$$

is used to assign q^2 with density $Q(q^2)$ and the value of q^2 is accepted with probability $G(v^2) = (1 + \mu^2)/2 \leq 1$ (Muhammad, 2012; Muhammad & Lee, 2013a,b,c).

From tabulated values of $f_0(Z, \theta)$ against q and $f'(Z, E)$ and $f''(Z, E)$ as a function of energy, $F(Z, E, \mu)$ is calculated using equation (2) and $A(Z, E, q^2)$ is calculated for $q_i^2 = q_1^2, \dots, \bar{q}^2$

accordingly (Muhammad, 2012; Muhammad & Lee, 2013c) (*i.e.* the CFF approximation). Both $f'(Z, E)$ and $f''(Z, E)$ are approximated as zero if the anomalous effects are not considered (*i.e.* the AFF approximation) (Muhammad, 2012; Muhammad & Lee, 2013c).

3.2. Inverse-sampling technique

In this method, the CDFs (ξ) of a cosine of scattering angle less than μ are calculated numerically for all the elements present in the simulation geometry by integrating the normalized PDF in equation (9) from -1 to μ (Jia *et al.*, 2012):

$$\xi = P(\mu) = \int_{-1}^{\mu} p(\mu) d\mu. \quad (17)$$

The inverse function of equation (17), *i.e.* the scattering angle cosine μ as a function of the cumulative probability ξ , can be determined numerically (Jia *et al.*, 2012):

$$\mu = P^{-1}(\xi). \quad (18)$$

By repeating this process for different energies, two-dimensional look-up tables $(\xi, E) \rightarrow \mu$ are obtained for all the elements. The sampling of μ for a scattered photon with energy E_0 is thus reduced to the sampling of a uniform number ξ_0 within the interval $(0, 1)$. To obtain μ , a two-dimensional linear interpolation of (ξ_0, E_0) in the aforementioned look-up table (ξ, E) is used. In practice, such maps are pre-computed

for each type of material considered. The azimuthal angle φ is sampled uniformly between 0 and 2π .

4. Comparison tests

The accuracy of the simulation model was validated by comparing the computational results with the publicly available NIST and Electron Photon Interaction Cross Sections Version 2017 (EPICS2017) reference databases (Cullen *et al.*, 1997; Kawrakow *et al.*, 2010; Cullen, 2018), scattering matrix (SM) data (Batič *et al.*, 2012; Hoff, 2018), computed total cross sections (TCSs) for *GEANT4* (*G4XrayRayleighModel*) and *PENELOPE* (2008 model) (Batič *et al.*, 2012; Hoff, 2018) and available experimental results of differential cross sections (DCSs) (Pratt *et al.*, 1993; Batič *et al.*, 2012; Hoff, 2018). In the first step, the pre-calculated TCS is validated. In the second step, the sampled DCS from the simulation model is compared with the theoretical values and available experimental data (Batič *et al.*, 2012; Hoff, 2018). For the TCS, elements with both low and high Z (specifically, aluminium, $Z = 13$, copper, $Z = 29$, tin, $Z = 50$, and lead, $Z = 82$) were selected to check the validity of the model for the entire range of the Periodic Table. Further, the available experimental results and theoretical data for DCSs for copper, $Z = 29$, tin, $Z = 50$, tantalum, $Z = 73$, and lead, $Z = 82$, at a photon energy of 0.05954 MeV were compared with the simulated DCSs. The study of Ta-73 is best suited to comparing both models of AFF and CFF approximations, as the energy is in the vicinity of its K edge

Figure 1

Pre-computed look-up maps $(\xi, E) \rightarrow \mu$ for angular distributions of coherently scattered photons. (a) Oxygen (O-16) based on the AFF approximation, (b) oxygen (O-16) based on the CFF approximation, (c) lead (Pb-82) based on the AFF approximation and (d) lead (Pb-82) based on the CFF approximation.

(i.e. 67.4164 keV; Pratt *et al.*, 1993; Batič *et al.*, 2012). The goodness-of-fit test results provide a quantitative evaluation of the accuracy of the simulation model for coherent scattering.

5. Computational performance

The computational efficiency (ϵ) and accuracy are important factors of an MC model that models physical processes. The assessment of our models was performed by comparing the three different models, namely the *PTM* acceptance-rejection model (AFF approximation), the *PTM* inverse-sampling model (AFF approximation) and the *PTM* inverse-sampling model (CFF approximation). First, the data preparation time (i.e. the time required to prepare data for each element before the start of a computation) and the computation time T (for n Rayleigh scattering of photon events) for each simulation model are recorded. We checked the time for data preparation and sampling by invoking the timer at specified points within the code. Data preparation includes the time required to read data, prepare data and compute the cross sections needed for the simulation. The sampling time is the time required to sample the physical processes during the simulation. A microsecond timer is started at the main function of an example that uses one of the models mentioned above. For fair comparison, ϵ was computed as follows:

$$\epsilon = \frac{1}{T\sigma^2}. \quad (19)$$

Here, σ is standard deviation in the PDF for the angular distribution of the scattered photons. For accuracy, ϵ is checked for multiple photon histories and elements (covering the whole Periodic Table). The computational efficiency was that resulting from the use of an Intel Core i7-6700HQ CPU@2.59 GHz running the Windows 10 operating system.

6. Results

6.1. Look-up maps

To illustrate the inverse-sampling method, the computed look-up maps $(\xi, E) \rightarrow \mu$ for coherent scattering of photons using the AFF [i.e. $f'(Z, E) = f''(Z, E) = 0$] and CFF (i.e. AFF + ASF) approximations are plotted in Fig. 1 for oxygen (O-16) and lead (Pb-82). A visible difference is observed for the higher atomic number element (i.e. Pb-82) on the scale of this graph. These maps are further used to sample coherent scattering based on inverse sampling for a specified energy of photons.

6.2. Total and differential cross sections

Comparison of TCSs calculated through simulation models based on the AFF and CFF approximations with the corre-

Figure 2

Comparisons of the *PTM*-calculated total cross section as a function of energy calculated based on the AFF and CFF approximations with the corresponding programs *EPDL2018*, *PENELOPE-2008* (Batič *et al.*, 2012), *G4XrayRayleighModel* (Batič *et al.*, 2012) and *SM* for (a) aluminium ($Z = 13$), (b) copper ($Z = 29$), (c) tin ($Z = 50$) and (d) lead ($Z = 82$).

sponding results from *EPDL* version 2018 (*EPDL2018*; Cullen, 2018), *PENELOPE-2008* (Batič *et al.*, 2012; Hoff, 2018) and *G4XrayRayleighModel* (Batič *et al.*, 2012; Hoff, 2018) for aluminium (Al-13), copper (Cu-29), tin (Sn-50) and lead (Pb-82) is shown in Fig. 2. The simulation model based on the AFF approximation reproduces the results from *G4XrayRayleighModel*. However, it is unable to retrieve the underlying atomic structure below 0.2 MeV of photon energy. The results of the model based on the CFF approximation are best fitted to the *EPDL2018*, *SM* and *PENELOPE-2008* data.

Sampled angular distributions of photons scattered coherently by Pb-82 with energies equivalent to its elemental absorption edges (*i.e.* M_5 edge = 0.002484 MeV, L_{III} edge = 0.0130352 MeV, L_{II} edge = 0.0152 MeV and K edge = 0.0880045 MeV) used in the AFF and CFF approximations are shown in Fig. 3. A significant difference in the distribution is observed. An increase in the forward scattering for all edge energies of Pb-82 is evident in Fig. 3, and it is more prominent for the energy equivalent to the K -edge energy. The results shown in Fig. 3 confirm that the angular distribution of coherent scattering is ASF-dependent.

Fig. 4 shows a comparison of sampled DCSs (as a function of scattering angle θ) for 59.54 keV photons calculated through *PTM* (CFF approximation), the *SM* data, the theoretical (CFF approximation) data and the results for simula-

tion through *PENELOPE* for the listed elements, all alongside the corresponding experimental results. The *PTM* results are closer to the corresponding *SM* and theoretical (CFF approximation) data for light elements. As the atomic number of the scattering element increases the deviation increases. The same observation can be made for the corresponding experimental data, except for Pb-82, where a higher difference between the experimental data, the corresponding *SM* data and the theoretical CFF approximation data is observed. However, for all elements, *PTM* (CFF approximation) showed better results than *PENELOPE-2008*. Overall, the *PTM* inverse-sampling model (CFF approximations) provides the most accurate representation of the Rayleigh scattering of photons.

6.3. Computational performance

The mean data-preparation times per element for the *PTM* acceptance–rejection model (AFF approximation), *PTM* inverse-sampling model (AFF approximation) and *PTM* inverse-sampling model (CFF approximation) are 1.7831, 2.336 and 2.6949 s, respectively. These results show that the data-preparation time is increased for the *PTM* inverse-sampling models (AFF and CFF approximations) compared with the *PTM* acceptance–rejection model. However, the most

Figure 3

Angular distributions of the coherently scattered photons through Pb-82 with listed energies equivalent to the absorption edges of Pb-82 (*i.e.* M_5 edge = 0.002484 MeV, L_{III} edge = 0.0130352 MeV, L_{II} edge = 0.0152 MeV and K edge = 0.0880045 MeV) based on the AFF and CFF approximations by simulating 1 million photons.

Table 1
Summary of computational times and efficiency of different MC models for 1 million Rayleigh scattering events.

	<i>PTM</i> acceptance and rejection			<i>PTM</i> inverse sampling					
				AFF approximation			CFF approximation		
	Time (ms)	SD	Efficiency (ms ⁻¹)	Time (ms)	SD	Efficiency (ms ⁻¹)	Time (ms)	SD	Efficiency (ms ⁻¹)
Mean	670.8	1.6281	6.199×10^{-4}	453.7	1.59377	9.414×10^{-4}	454.5	1.65251	8.637×10^{-4}
Minimum	646.0	1.3230	2.914×10^{-4}	434.0	1.30962	4.761×10^{-4}	430.0	1.38587	4.319×10^{-4}
Maximum	692.0	2.3049	8.631×10^{-4}	490.0	2.19994	1.207×10^{-4}	481.0	2.20557	1.082×10^{-4}

important thing is the computational efficiency ϵ in the MC simulation. Table 1 shows a summary of computation times T and computational efficiency ϵ for the acceptance–rejection and inverse-sampling (both AFF and CFF approximations) models for 1 million Rayleigh events. The results show that T is reduced for the *PTM* inverse-sampling models (AFF and CFF approximations) compared with the *PTM* acceptance–rejection model. Overall, the efficiency per event is improved by factors of 1.52 and 1.4 for the *PTM* inverse-sampling models with the AFF and CFF approximations, respectively.

7. Discussion

MC models based on the AFF approximation use a single set of AFF values as a function of momentum transfer for each element. Therefore, a single pre-calculated data library is

easily provided and implemented in MC models based on the acceptance–rejection technique. On the other hand, the MC model based on the CFF approximation uses a set of CFF values as a function of both momentum transfer and energy. Thus, live calculation of the data for each run is needed to handle the sampling for photons encountered with various energy values. To implement the inverse-sampling technique, the pre-calculated look-up maps $(\xi, E) \rightarrow \mu$, as shown in Fig. 1, were calculated. The visible difference on the scale of these figures shows the variation in the angular distribution around the elemental absorption edges of the selected elements.

The plots of TCS in Fig. 2 show that simulations based on the CFF approximation are the best model to reproduce the corresponding *EPDL* and *SM* results. In contrast, the MC model based on the AFF approximation overestimates the TCSs at lower energies (<0.2 MeV of photon energy) with

Figure 4
Comparisons of *PTM*-sampled (based on the CFF approximation) DCSs with literature-collected experimental, *SM*, theoretical (*i.e.* the CFF approximation) and *PENELOPE-2008* data as a function of scattering angle (θ) for 59.54 keV photons after coherent scattering by neutral atoms of (a) copper ($Z = 29$), (b) tin ($Z = 50$), (c) tantalum ($Z = 73$) and (d) lead ($Z = 82$).

respect to the *EPDL2017* and *SM* results. Furthermore, it is unable to predict or retrieve the underlying atomic structure. Lastly, it is approximately valid only above the *K* edge. This model is not useful where coherent scattering is of primary interest. For higher energies on the scale of Fig. 2, the TCSs agree with the *EPDL2017* and *SM* results for the MC model based on both AFF and CFF approximations. The angular distributions of coherently scattered photons (presented in Fig. 3) show that they are dependent on the ASFs, owing to the increase in forward scattering around the elemental absorption edges. These results show that the MC model based on the CFF approximation for Rayleigh scattering of photons can calculate coherent scattering amplitudes that are more accurate than MC-based radiation transport packages ignoring anomalous scattering factors (AFF approximation).

Fig. 4 shows that the DCS decreases as a function of scattering angle (θ). The results for the angular distribution of photons by the simulation model based on the CFF approximation are in good agreement with experimental elastic scattering measurements for photons with the same energy and scattering element. The results are also best fitted to the corresponding experimental results and theoretical data. It is noted here that we are simulating Rayleigh scattering only. However, a measured DCS for coherent scattering is a combination of several physical processes (e.g. Rayleigh, nuclear Thomson and Delbrück scattering, and nuclear resonance) whose contribution depends on *Z* and the energy of the incident photon. The contribution of these processes increases with *Z*, which is evident from Fig. 4. On the other hand, the simulation model presented here only evaluates Rayleigh scattering DCSs, while the *SM* calculations account for both Rayleigh and Thomson scattering amplitudes (Batič *et al.*, 2012). Currently, almost all major MC packages ignore the contribution of nuclear Thomson, Delbrück and nuclear resonance amplitudes, considering their contribution to the coherent scattering amplitude to be small. Ideally, each of these processes should be included to give the most accurate simulation of the coherent scattering amplitudes. Another important observation from Fig. 4(c) is the maximum deviation of the *PENELOPE-2008*-calculated DCSs from the corresponding experimental and *SM* data. This confirms that the ASFs are ignored while sampling coherently scattered photons. However, Fig. 1 confirms that the ASFs are used by *PENELOPE-2008* while using TCSs.

The CFF approximation model based on the inverse-sampling technique is successfully implemented in our general-purpose radiation transport code, *PTM*. The computational efficiency is improved for *PTM* inverse-sampling models based on the AFF and CFF approximations compared with the conventional acceptance–rejection technique. The distributions of photons scattered by Rayleigh scattering are also reproduced accurately. While MC modeling of coherent scattering of photons can play an important role in studying complex systems, the accuracy depends on how accurately and effectively it is able to reproduce the actual physical model for known problems. By implementing a CFF-based MC model for coherent scattering, general-purpose MC packages like

PTM are expected to be valuable tools to gain precise scientific insight into complex systems. Computationally, using inverse sampling is best suited for a GPU platform, since the GPU supports hardware linear interpolation with a higher efficiency. In this case, a very high efficiency is expected.

8. Conclusions

An updated MC model based on the CFF approximation for Rayleigh scattering of photons with anomalous scattering was implemented in a general-purpose radiation transport code, *PTM*. The model was tested against the *GEANT4* and *PENELOPE-2008* MC packages, as well as corresponding theoretical and experimental results. Comparisons with the experimental results and other benchmark MC codes have indicated that the updated model provides a more accurate representation of the coherent scattering of photons. Computational efficiency is also increased. By implementing the inverse-sampling technique based on the CFF approximation, the computational efficiency for $\sim 10^6$ coherent scattering events is improved by a factor of 1.4 over the conventional acceptance–rejection technique (based on the AFF approximation).

Acknowledgements

The authors would like to thank Gabriela Hoff from Pontificia Universidade Catolica do Rio Grande do Sul, Brazil, for providing the database to perform the validation.

Funding information

The following funding is acknowledged: National Institute of Biomedical Imaging and Bioengineering (award No. R01EB022589). The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health.

References

- Amako, K., Guatelli, S., Ivanchenko, V. N., Maire, M., Mascalino, B., Murakami, K., Nieminen, P., Pandola, L., Parlati, S., Pia, M. G., Piergentili, M., Sasaki, T. & Urban, L. (2005). *IEEE Trans. Nucl. Sci.* **52**, 910–918.
- Batič, M., Hoff, G., Pia, M. G. & Saracco, P. (2012). *IEEE Trans. Nucl. Sci.* **59**, 1636–1664.
- Berger, M. J., Coursey, J. S., Zucke, M. A. & Chang, J. (1999). *Stopping-Power and Range Tables for Electrons, Protons, and Helium Ions: Physical Reference Data*. National Institute of Standards and Technology, Maryland, USA. <https://www.nist.gov/pml/stopping-power-range-tables-electrons-protons-and-helium-ions>.
- Berger, M. J., Hubbell, J. H., Seltzer, S. M., Chang, J., Coursey, J. S., Sukumar, R., Zucker, D. S. & Olsen, K. (1999). *XCOM: Photon Cross Sections Database*. Version 1.2. <http://physics.nist.gov/xcom>.
- Berger, M. J., Inokuti, M., Andersen, H. H., Bichsel, H., Powers, D., Seltzer, S. M., Thwaites, D. & Watt, D. E. (1993). International Commission on Radiation Units and Measurements (Bethesda, Maryland) ICRU Report 49 (*Journal of the International Commission on Radiation Units and Measurements*, Vol. os25, Issue 2, page NP).

- Bergstrom, P. M., Kissel, L., Pratt, R. H. & Costescu, A. (1997). *Acta Cryst.* **A53**, 7–14.
- Bradley, D. A., Roy, S. C., Kissel, L. & Pratt, R. H. (1999). *Radiat. Phys. Chem.* **56**, 175–195.
- Carter, L. L. & Cashwell, E. D. (1975). *Particle-Transport Simulation with the Monte Carlo Method*. Report TID-26607. Technical Information Center, Office of Public Affairs, US Energy Research and Development Administration, Oak Ridge, Tennessee, USA.
- Chadwick, M. B., Herman, M., Obložinský, P., Dunn, M. E., Danon, Y., Kahler, A. C., Smith, D. L., Pritychenko, B., Arbanas, G., Arcilla, R., Brewer, R., Brown, D. A., Capote, R., Carlson, A. D., Cho, Y. S., Derrien, H., Guber, K., Hale, G. M., Hoblit, S., Holloway, S., Johnson, T. D., Kawano, T., Kiedrowski, B. C., Kim, H., Kunieda, S., Larson, N. M., Leal, L., Lestone, J. P., Little, R. C., McCutchan, E. A., MacFarlane, R. E., MacInnes, M., Mattoon, C. M., McKnight, R. D., Mughabghab, S. F., Nobre, G. P. A., Palmiotti, G., Palumbo, A., Pigni, M. T., Pronyaev, V. G., Sayer, R. O., Sonzogni, A. A., Summers, N. C., Talou, P., Thompson, I. J., Trkov, A., Vogt, R. L., van der Marck, S. C., Wallner, A., White, M. C., Wiarda, D. & Young, P. G. (2011). *Nucl. Data Sheets*, **112**, 2887–2996.
- Chantler, C. T. (2000). *J. Phys. Chem. Ref. Data*, **29**, 597–1056.
- Cianci, M., Helliwell, J. R., Helliwell, M., Kaucic, V., Logar, N. Z., Mali, G. & Tusar, N. N. (2005). *Crystallogr. Rev.* **11**, 245–335.
- Cromer, D. T. (1983). *J. Appl. Cryst.* **16**, 437.
- Cromer, D. T. & Liberman, D. (1970). *J. Chem. Phys.* **53**, 1891–1898.
- Cullen, D. E. (1989). *Program SCATMAN: A Code Designed to Calculate Photon Coherent Scattering Anomalous Scattering Factors and Cross Sections*. Report UCRL-ID-103422. Lawrence Livermore National Laboratory, California, USA.
- Cullen, D. E. (2018). *A Survey of Electron Cross Section Data for Use in EPICS2017*. Document IAEA-NDS-0226. IAEA, Vienna, Austria. <https://www.nndc.bnl.gov/endf/epics/>.
- Cullen, D. E., Hubbell, J. H. & Kissel, L. (1997). *EPDL97: The Evaluated Photon Data Library, '97 Version*. Report UCRL-50400, Vol. 6, Revision 5. Lawrence Livermore National Laboratory, California, USA.
- Henke, B. L., Gullikson, E. M. & Davis, J. C. (1993). *At. Data Nucl. Data Tables*, **54**, 181–342.
- Henke, B. L., Lee, P., Tanaka, T. J., Shuimabukuro, R. L. & Fujikawa, B. K. (1981). *AIP Conf. Proc.* **75**, 340.
- Hoff, G. (2018). Personal communication.
- Hubbell, J. H. & Øverbø, I. (1979). *J. Phys. Chem. Ref. Data*, **8**, 69–106.
- Hubbell, J. H., Veigele, W. J., Briggs, E. A., Brown, R. T., Cromer, D. T. & Howerton, R. J. (1975). *J. Phys. Chem. Ref. Data*, **4**, 471–538.
- Jia, X., Yan, H., Gu, X. & Jiang, S. B. (2012). *Phys. Med. Biol.* **57**, 577–590.
- Kane, P. P., Kissel, L., Pratt, R. H. & Roy, S. C. (1986). *Phys. Rep.* **140**, 75–159.
- Kawrakow, I. M.-H., Rogers, E. D. W. O., Tessier, F., Walters, B. R. B. & Rogers, D. (2010). NRCC Report PIRS-701. Ottawa, ON: National Research Council of Canada.
- Kissel, L., Zhou, B., Roy, S. C., Sen Gupta, S. K. & Pratt, R. H. (1995). *Acta Cryst.* **A51**, 271–288.
- Kling, A., Barao, F. J., Nakagawa, M., Tavora, L. & Vaz, P. (2014). *Advanced Monte Carlo for Radiation Physics, Particle Transport Simulation and Applications*, Proceedings of the Monte Carlo 2000 Conference, Lisbon, Portugal, 23–26 October 2000, p. 69. Springer Science and Business Media.
- Liang, Y., Muhammad, W., Hart, G., Nartowt, B., Chen, Z., Yu, B., Roberts, K., Duncan, J. & Deng, J. (2019). *Sci. Rep.* Submitted.
- Muhammad, W. (2012). Thesis. Kyungpook National University, Daegu, South Korea.
- Muhammad, W. & Lee, S. H. (2013a). *Acta Cryst.* **A69**, 297–308.
- Muhammad, W. & Lee, S. H. (2013b). *J. Radiat. Res.* **54**, 190–201.
- Muhammad, W. & Lee, S. H. (2013c). *PLoS One*, **8**, e69608.
- Pratt, R. H., Bergstrom, P. M. Jr & Kissel, L. (1993). *New Relativistic S-Matrix Results for Scattering – Beyond the Usual Anomalous Factors/Beyond Impulse Approximation*. Report UCRL-JC-114583. Lawrence Livermore National Laboratory, California, USA.
- Roy, S. C., Kissel, L. & Pratt, R. H. (1999). *Radiat. Phys. Chem.* **56**, 3–26.
- Roy, S. C., Pratt, R. H. & Kissel, L. (1993). *Radiat. Phys. Chem.* **41**, 725–738.
- Salvat, F., Fernández-Varea, J. M. & Sempau, J. (2011). *PENELOPE-2011: A Code System for Monte Carlo Simulation of Electron and Photon Transport*. *PENELOPE-2011 Workshop and Training Course*, Barcelona, Spain, 4–7 July 2011. Report NEA/NSC/DOC(2011)5. Organisation for Economic Co-operation and Development Nuclear Energy Agency Data Bank, Issy-les-Moulineaux, France.
- Schaupp, D., Schumacher, M., Smend, F., Rullhusen, P. & Hubbell, J. H. (1983). *J. Phys. Chem. Ref. Data*, **12**, 467–512.
- Surzhykov, A., Yerokhin, V., Fritzsche, S. & Volotka, A. (2018). *Phys. Rev. A*, **98**, 053403.
- Tartari, A., Taibi, A., Bonifazzi, C. & Baraldi, C. (2002). *Phys. Med. Biol.* **47**, 163–175.
- Turturica, G., Iancu, V., Suliman, G. & Ur, C. (2018). *Nucl. Instrum. Methods Phys. Res. B*, **436**, 68–73.