Light scattering by multiple spheres: comparison between Maxwell theory and radiative-transfer-theory calculations

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We present a methodology to compare results of classical radiative transfer theory against exact solutions of Maxwell theory for a high number of spheres. We calculated light propagation in a cubic scattering region ($20\times 20\times 20$ μm^3) consisting of different concentrations of polystyrene spheres in water (diameter 2 μm) by an analytical solution of Maxwell theory and by a numerical solution of radiative transfer theory. The relative deviation of differential as well as total scattering cross sections obtained by both approaches was evaluated for each sphere concentration. For the considered case, we found that deviations due to radiative transfer theory remain small, even for concentrations up to ca. 20 vol. %. © 2009 Optical Society of America

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In modern medicine the field of biomedical optics has become an important research area. For many therapeutical and diagnostic applications of light, knowledge of its propagation in biological tissue is necessary [1]. Currently light propagation is almost always described by radiative transfer (and diffusion) theory, which is an approximation of Maxwell theory [2]. The apparent reason for this is the major drawback of Maxwell theory not to comply with the requirements of giving fast solutions in relevant simulation volumes. However, at present it is not very clear under which conditions classical radiative transfer theory without dense media extensions can be used to accurately describe light propagation in tissue. For example, Tseng and Huang found that there are significant differences in scattering cross sections calculated by both approaches, Maxwell and radiative transfer theory [3]. Recently, we showed in two dimensions that the results for cylindrical scatterers are in good agreement, referring to the angle-dependent differential scattering cross section [4]. Roux *et al.* found similar agreement for low concentrations in two dimensions [5]. Furthermore, the total scattering cross sections fit well when diffraction effects are neglected.

In this Letter we present a three-dimensional quantitative comparison between simulation results of radiative transfer theory (RTT) and Maxwell theory for light scattering by multiple spheres. For the latter case we used the Fortran codes GMM01F and GMM01S as implementations of the generalized multisphere Mie (GMM) theory [6,7]. These exact codes have been verified and were taken as reference [8]. For the solution of RTT, a Monte Carlo code was developed by our group [9]. Comparison was done by calculating the differential scattering cross sections and the relative error between the results of both theories for different sphere concentrations. The exact multisphere solution is an extension of classical Mie theory that can be deduced directly from Maxwell's equations. A collection of spheres, randomly distributed in a cubic simulation volume, is illuminated by a plane monochromatic electromagnetic wave, perpendicular to one side of the cube. Xu derived a far-field formulation of the scattered electromagnetic wave with respect to the incident wave [6]. By introduction of an amplitude scattering matrix $\mathbf{S}$, one can express the scattered field $\mathbf{E}_{\rm sca}=(E^{\parallel}_{\rm sca},E^{\perp}_{\rm sca})^T$, when regarding its parallel and perpendicular field components $E^{\parallel}$ and $E^{\perp}$, in terms of an (in z-axis propagating) incident field $\mathbf{E}_{\rm inc}=(E^{\parallel}_{\rm inc},E^{\perp}_{\rm inc})^T$ as follows [10]:

$$\mathbf{E}_{\rm sca}=\frac{e^{ik(r-z)}}{-ikr}\begin{pmatrix}S_2 & S_3\\ S_4 & S_1\end{pmatrix}\mathbf{E}_{\rm inc},$$

where the scattered field is an outgoing spherical wave with an angular intensity distribution whose information is stored inside the $\mathbf{S}$ matrix. This matrix is calculated by expanding the electromagnetic field in spherical basis functions taking into account the entire sphere distribution. The differential scattering cross section $\mathrm{d}\sigma/\mathrm{d}\Omega$ can be calculated through these matrix elements S_i $(i=1,\ldots,4)$, together with the wavenumber k.

On the other hand, RTT calculates the photon radiance L along a propagation path $\mathrm{d}s$ in a given medium, thus omitting the wave character of light. Contrary to Maxwell theory, the scatterers are infinitesimal small points whose spatial distribution inside the medium is given by a random number generator. True random distributions can be achieved this way, since, as opposed to extended spheres, arbitrary distances between the scatterers are possible. In steady state, the transfer equation can be written as

$$\frac{\mathrm{d}L(\mathbf{r},\mathbf{s})}{\mathrm{d}s}=-\mu_t L(\mathbf{r},\mathbf{s})+\mu_s\int_{4\pi}\mathrm{d}\Omega' p(\mathbf{s},\mathbf{s}')L(\mathbf{r},\mathbf{s}'),$$

where μ_t and μ_s denote the extinction and scattering coefficients, respectively, and Ω denotes the solid angle. The quantity $p(\mathbf{s},\mathbf{s}')$, also called normalized phase function, represents the angle-resolved scattering probability for single scattering. In our Monte Carlo simulations, the phase function equals the angular intensity distribution of a single sphere obtained from Mie theory [11].

We simulated light scattering by polystyrene microspheres with diameters of 2 μm and refractive indices of $n=1.59$ in an infinite medium with $n_m=1.33$. As a GMM scatterer model, spheres are bounded to a cubical simulation volume of $20\times20\times20\ \mu m^3$. For several concentrations, they are randomly distributed inside the volume. To achieve this, we used the Metropolis shuffling algorithm, which provides the generation of random distributions of nonoverlapping spheres [12]. The calculations were performed for ten different realizations, yielding an averaged output of the differential scattering cross sections. This procedure reduces speckle effects caused by interference resulting from Maxwell theory. Taking into account unpolarized light only, we calculated the differential scattering cross section $d\sigma/d\theta$, integrated over 360 azimuthal angles and solely depending on the scattering angle. Referring to Monte Carlo simulations, the azimuthal integration was done in the same way.

Figure 1 depicts scattering functions for sphere numbers N in the range from $N=50$ to $N=800$ and a wavelength of $\lambda=633$ nm for both GMM and Monte Carlo simulations. The most significant differences in the shape of the results are sharp peaks for the scattered intensity, obtained from Maxwell theory, at scattering angles θ close to forward ($\theta\approx0°$) and backward direction ($\theta\approx180°$), becoming steeper with an increasing number of spheres. The observed peaks in forward and backward direction are caused by two physical effects. Keeping in mind that a finite volume of spheres is irradiated by an infinite plane wave, we first get a peak in the forward direction ($\theta\lesssim2°$), caused by diffraction of the sphere cluster as a whole. In comparison to the Mie diffraction peak of an individual 2 μm sphere, also sketched in Fig. 1(a), its width is smaller. The second effect is coherent backward scattering, a result of pairwise coherent superposition along backscattering paths [2]. We remark that the dominating forward peaks vanish for a lateral infinite simulation volume as diffraction effects would disappear. These peaks do not occur at all in RTT, since the wave character of light is neglected. The area of $\theta=0°$ to $\theta\approx20°$ is shown in detail as insets of Fig. 1(a) and Fig. 1(b) to point out the shape and height of the forward-scattering peak. As a quantitative comparison of the results depicted in Fig. 1, the relative error between Monte Carlo and GMM simulations was calculated for the differential scattering cross sections $d\sigma/d\theta$. In Fig. 2(a) the angular relative error $\delta(d\sigma/d\theta)$ is shown for high sphere concentrations, calculated according to

$$\delta(d\sigma/d\theta)=\frac{(d\sigma/d\theta)_{\mathrm{GMM}}-(d\sigma/d\theta)_{\mathrm{MonteCarlo}}}{(d\sigma/d\theta)_{\mathrm{GMM}}}\times100\,\%\,.$$

Only scattering angles ranging from $\theta\approx20°$ to $\theta\approx170°$ are shown. While there was almost coincidence at certain scattering angles, we also got significant deviation up to 30% for other angles. Figure 2(b) depicts the absolute relative error

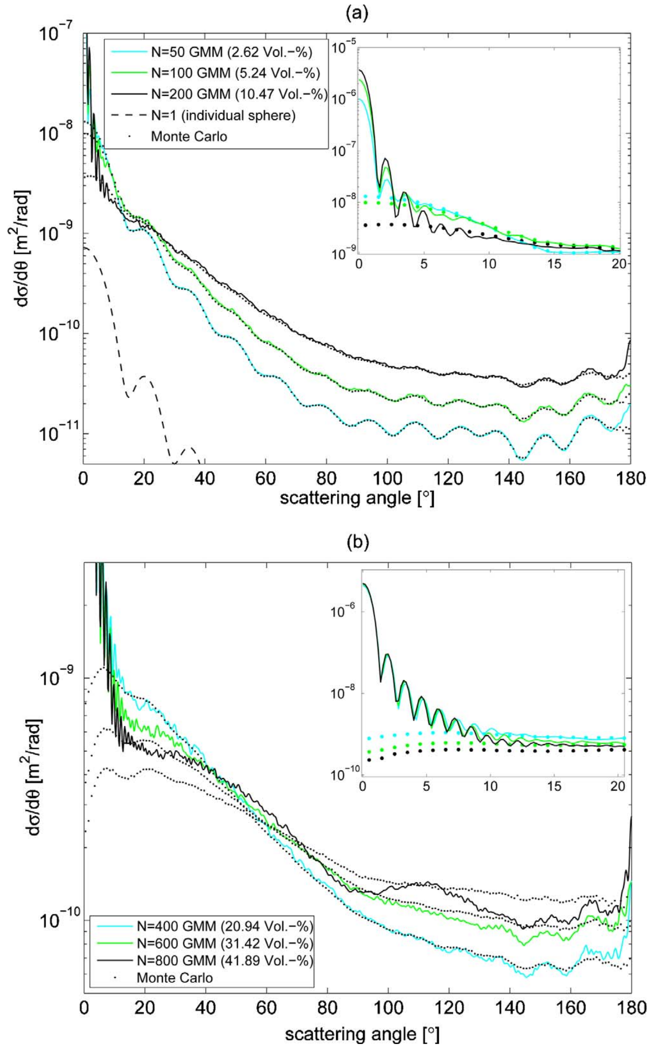

Fig. 1. (Color online) Differential scattering cross sections $d\sigma/d\theta$ due to an incident wave at $\lambda=633$ nm for different sphere concentrations. The results of GMM simulations (solid curves) and Monte Carlo simulations (dots) are compared. In (a) the sphere numbers are $N=50$, 100, and 200, in (b) $N=400$, 600, and 800.

$$\langle\delta(d\sigma/d\theta)\rangle=\frac{1}{n_{st}}\sum_{i=1}^{n_{st}}|\delta_i(d\sigma/d\theta)|,$$

averaged over a limited angle range against the simulated number of spheres. We used absolute values to quantify differences $\delta_i(d\sigma/d\theta)$ between both theories in order to avoid subtractive cancellation. The number of angle steps is $n_{st}=751$ resulting from an angular resolution of $\Delta\theta=0.2°$ and a range from $\theta=20°$ to $\theta=170°$. Within this range, the deviation of the Monte Carlo model results are below ca. 3% for a sphere number smaller than up to 400. The reason for the increasing error at higher concentrations is the field interaction of nearby spheres, referred to as dependent scattering, according to Maxwell theory. Alternatively, the radiative transfer equation de-

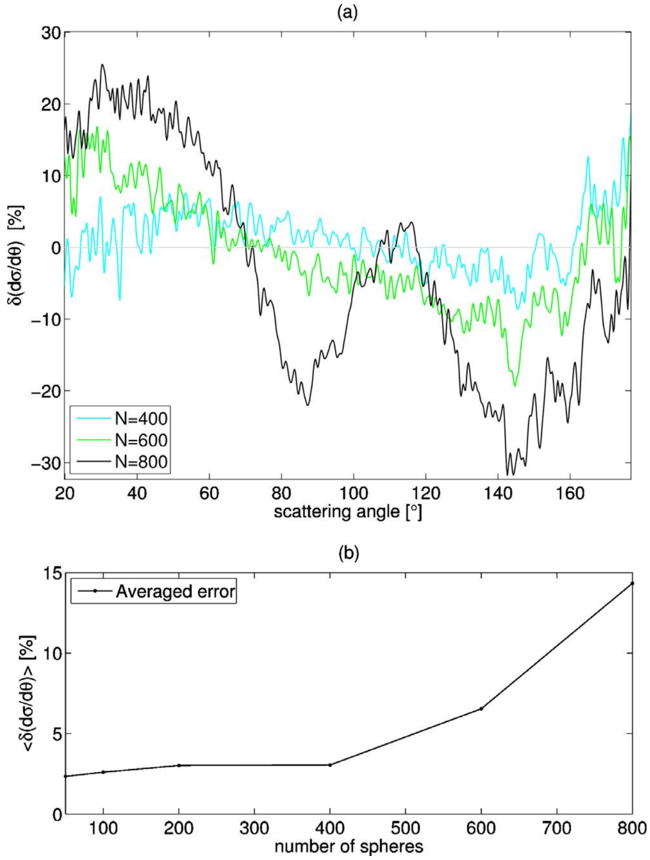

Fig. 2. (Color online) (a) Relative error $\delta(\mathrm{d}\sigma/\mathrm{d}\theta)$ of GMM versus Monte Carlo results for $N=400$, 600, and 800 plotted against the scattering angle that is cropped to a range of $\theta=20°$ to $\theta=170°$ to exclude the sharp forward- and backscattering peaks. (b) Averaged error $\langle\delta(\mathrm{d}\sigma/\mathrm{d}\theta)\rangle$, plotted against number of spheres.

scribes light propagation as sequential, independent scattering events, thus neglecting dependent interaction aspects.

From the simulation data the total scattering cross section C_{sca} was also calculated,

$$C_{\mathrm{sca}} = \int_0^{\pi} \mathrm{d}\theta \sin\theta \frac{\mathrm{d}\sigma}{\mathrm{d}\theta}.$$

We found significant differences between the calculated scattering cross sections of both theories, essentially due to diffraction, provided that both approaches are compared over the whole angular range (see Fig. 3). Again, in analogy to previous considerations, the angular range was truncated to a region from $\theta=20°$ to $\theta=170°$ to exclude differences due to coherent effects. With this correction, the cross sections calculated by either method are in good agreement, e.g. there is an averaged error of $<2\%$ for 400 spheres (corresponding to a sphere-packing density of ca. 21%) and smaller sphere numbers in contrast to an error of ca. 50% if uncorrected.

In summary, we showed our methodology to quantify the validity of classical RTT to describe multi-

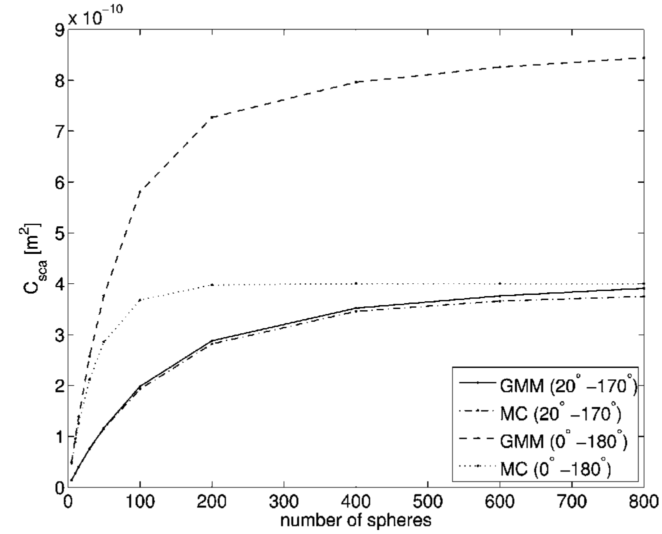

Fig. 3. Simulated total scattering cross section against number of spheres. While uncorrected cross sections (dashed and dotted curves) differ by a factor of approximately 2, the corrected ones are in good agreement. For $N \gtrsim 15$ the curves deviate from linear increase because of multiple scattering.

sphere light scattering for different volume concentrations. Since RTT-based methods are preferably applied to complex inhomogeneous media but are known to fail in case of high densities, the range of validity of classical RTT can now be systematically investigated. With the present model we have the possibility to compare Maxwell and RTT for arbitrary sizes and refractive indices of the scatterers and thus also for more realistic models of biological tissue.

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