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MONTE CARLO SIMULATION OF LIGHT SCATTERING BY INHOMOGENEOUS SPHERES

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ABSTRACT. The light scattering properties of optically inhomogeneous spherical particles are studied by means of a phase resolved 3-dimensional Monte Carlo simulation. This is equivalent to a radiative transfer process in a participating medium with spherical boundaries. For zero concentration of inhomogeneities we obtain the undisturbed scattering distribution of the coherent light with its size-dominated oscillating structure. With increasing optical depth of the scattering medium this interference structure is disturbed by multiple scattering within the particle and the corresponding loss of coherence. For large optical depth the Schoenberg result for a sphere with a Lambert surface is obtained.
Thus it is possible to quantify the influence of size and concentration of a fine dispersed phase inside a spherical droplet onto the light scattering properties of an inhomogeneous macroscopical particle. This is of importance for the application of various optical particle sizing techniques on media, that contain fine dispersed substances.

1. INTRODUCTION

Optical techniques for particle sizing are of high value for a variety of engineering applications. They are non-intrusive and can be used for on-line quality control. For the characterization of multiphase flows Information about both the local size- and velocity-distribution is required. Phase Doppler anemometry (PDA) is a well established technique in this area. It has been successfully applied to sprays of homogeneous fluids (water, mineral oil, molten metals). [l] PDA as well as all other optical particle sizing techniques relies on an accurate theoretical description of the light scattering process. Deviations from the assumptions made in the theoretical model and being part of the genealogical reality lead to incorrect sizing results and thus strongly limit the application range of these techniques. The characterization of spray drying processes, which are of high relevance in food technology - for instance spray drying of milk (lipid globules in water) or of coffee (suspension of solid powder coffee particles in water) in order to produce milk powder or instant coffee - is one example. Small emulsion or suspension (guest-) particles inside a larger host (or carrier) particle scatter and absorb the part of the incoming light, that is refracted into host. This leads to a disturbance of the scattering properties of the host particle and thus to errors in the PDA size measurement, if refracted light is used for detection. The resulting measured diameter distributions are broadened with respect to the real size distribution. [2, 3, 4] Related experimental work on the light scattering of inhomogeneous spheres has been performed by Cu et. al. [5] (angular scattering patterns) and Bronk et. al. [6] (Photon-correlation spectroscopy).

Mie theory is commonly used to calculate the scattering properties of spherical particles as a function of particle size d_{host} and index of refraction $m = n + i\,k$. Recently more sophisticated approaches were developed to overcome the limitations of Mie theory (plane wave, spherical particle shape) to take into account the Gaussian profile of laser beams and the shape of particles of less symmetry. [7, 8] The light scattering problem of spheres containing additional scatterers was addressed e.g. by Borghese [9] and Fuller [10]. Their codes yield reliable results for arbitrary positions of a single inclusion, while poor convergence and a strongly increasing requirement on computer memory and CPU time prevent an application of these approaches to optically inhomogeneous particles, that occur in spray drying processes mentioned above. Thus we can summarize that multiple scattering theories are not yet applicable for the problem at hand. Instead it seems rather appropriate to regard the scattering of light by an inhomogeneous sphere as a process of radiative transfer inside a participating (scattering and/or absorbing) medium with spherical boundaries. The source of radiation is defined by the refracted part of the initial light source to which the host particle is exposed.

Analytical solutions of the equation of radiative transfer are not available for spherical boundaries, complex distributed light sources, or even the combination of both. Therefore we developed a Monte Carlo code to gain information about the influence of size and concentration (and shape) of a second, fine dispersed phase onto the scattering properties of a large particle. Path and state of polarization of individual photons are followed throughout the host particle. The final scattering pattern results from coherent addition of the emerged photons with respect to their phases.

2. Light Scattering by Homogeneous Spheres

We start with a description of the scattering properties of a homogeneous sphere for two purposes. First, this should be the limiting case to our problem for very low concentrations $c \to 0$. Second, we assume the inhomogenities themselves to be small homogeneous spheres and use the according scattering theories in the progress of the Monte Carlo code (see below).

Mie theory provides a formal solution to the scattering problem of spheres for an incident plane wave. We obtain the distribution of the scattered light (scattering phase function) $p(\theta)$ together with cross sections for scattering Qs and absorption Q_a as well as the asymmetry parameter g. Particle size d_{host}, complex index of refraction m, wavelength λ, and state of polarization are the input data. Details on the theory according to Mie and its numerical realization can be found in standard textbooks on light scattering. [11, 12]

Describing the light scattering of host particles with large diameters compared with the light wavelength λ, we are allowed to apply geometrical optics (GO). By separating the fundamental physical effects of scattering - diffraction as a consequence of the incomplete wavefront, refraction. and reflection at the host particle surface - we gain more insight into the scattering process. This is important for the further discussion, as only the refracted part of the incoming light will be the source of the radiative transfer process inside the host particle and will thus be exposed to disturbance by the dispersed particles. The reflected and diffracted part of the incoming light will not be influenced by the dispersed particles.

Fig. 1 shows the scattered intensity of a 50 µm water droplet in air according to GO and Mie theory. The course of GO-contributions is smooth. Either reflection or first order refraction is dominant in most directions. Nevertheless the interference between these scattering contributions is responsible for the oscillations clearly visible in the Mie solution: refracted and reflected photons (or rays, when speaking in terms of GO) follow different optical paths for a certain scattering angle. The path difference between these contributions varies with scattering angle and leads to the depicted oscillating structure. it is only due to this interference that we can extract Information about a particle's size from its scattering diagram, as the distribution of refracted and reflected light alone would be independent of the particle diameter.

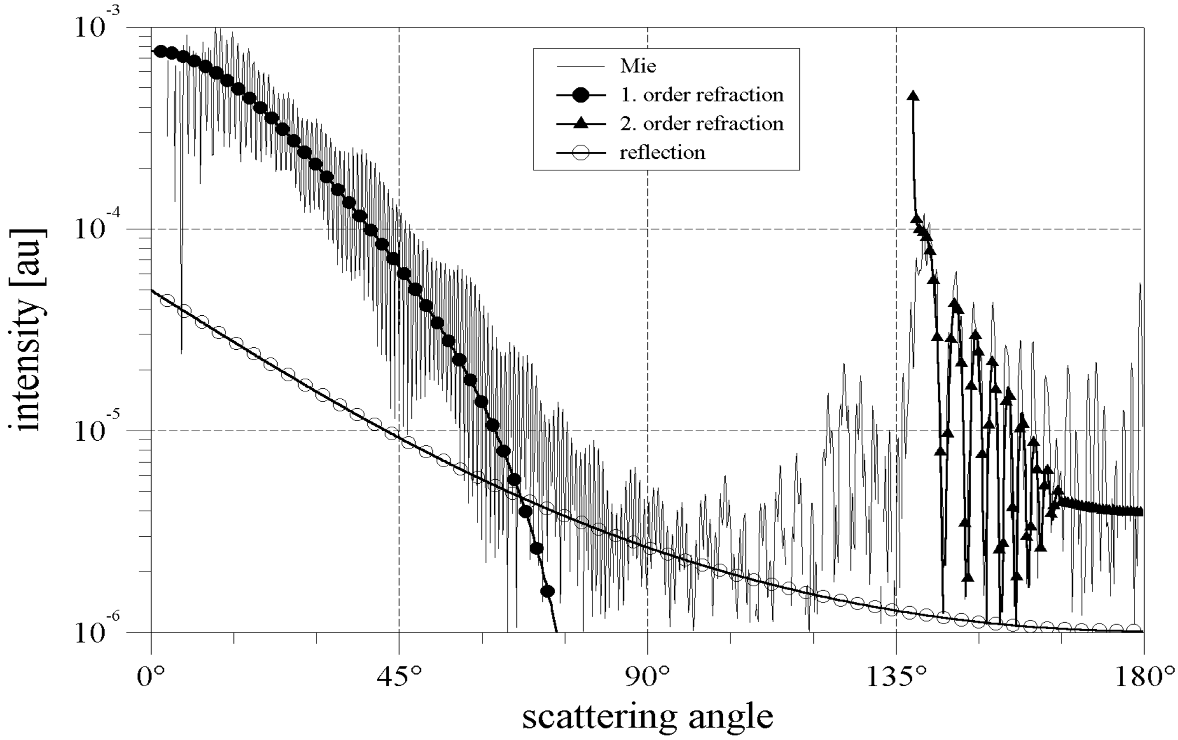

Figure 1: *Scattered intensity of a homogeneous* 50 µm *water droplet* in air (*m = 1.33 + i 0*) *for linear polarized light perpendicular to the scattering plane. The result of Mie theory (solid* line without *symbols) and the separate contributions according to geometrical optics (reflection, first and second order refraction) are depicted.*

The rainbow angle θ_{RB} is a minimum angle of deflection for second order refracted rays. According to simple GO, θ_{RB} is a function of the index of refraction m only. For all scattering angles above θ_{RB} two rays with different optical paths are contributing. Interference occurs, which is again visible as oscillations of the scattered intensity versus scattering angle.

The diffraction part is not depicted in fig. 1 for reasons of clarity. The separate diffraction solution for a sphere can be easily obtained. [12] it contributes only within a small area of forward scattering and is thus of no importance for our purposes.

Finally it should be pointed out, that GO is only an approximation to the exact scattering problem. Full agreement with Mie theory cannot be expected in all cases. Therefore any other approach which is based on GO - like our Monte Carlo algorithm described below - cannot do better than GO when compared with an exact solution.

3. Light Scattering by Diffuse Reflecting Spheres

The reflectance of a participating medium of infinite optical depth is known to be a function solely of the albedo w, which quantifies the contribution of scattering to the total extinction of the medium. Independent from *w* the angular reflectance of such a medium is diffuse and incoherent (depending on the length of coherence) due to multiple scattering inside the medium. A sphere composed from a participating medium of infinite optical depth will therefore be equivalent to a sphere whose surface follows Lambert's cosθ law. The scattering distribution for a sphere with a Lambert surface was derived by Schoenberg: [13]

$$p(\theta) = \frac{8}{3\pi}(sin\theta - \theta cos\theta) \quad (1)$$

In addition the surface of the host particle will contribute its undisturbed coherent reflectance distribution. Thus, for the case of strongly scattering particles in high concentration within a large host we have another limiting case to our Monte Carlo simulation.

4. Light Scattering by Inhomogeneous Spheres: Monte Carlo Approach

As common in Monte Carlo simulations of radiative transfer we follow single photons on their path through the chosen setup until they hit some predefined detection area. [14] The complexity of the problem is reduced by splitting the life of a photon into a series of interactions. for which simple and well known physical laws exist: plane layer reflection and refraction at the host particle surface, and single scattering at a small particle. The solutions of Maxwell's equations - Fresnel Formulas and Mie theory, respectively - though derived for waves of electromagnetic radiation rather than discrete photons-matter-interaction, can be used in a statistical manner to calculate the outcome of each interaction event.

We have to be sure to correctly trace the phase of the photons to yield the undisturbed scattering properties in the low concentration limit. This distinguishes our application from other Monte Carlo codes, where only incoherent radiation fields can be assumed in advance due to multiple scattering.

The photon tracing is performed in three dimensional geometry. This will enable us to simulate the light scattered on PDA detectors located at different positions in space. We are also able to assume host particle shapes other than spherical, as long as the surface of the host can be described by an analytical function.

Fig. 2 shows a flow diagram of the complete simulation process for a photon. Details on- the single steps are described below.

We use a standard code for the Generation of pseudo random numbers. [1,3] According to the authors this routine passes all known statistical tests as long as the number of calls is less than 10^8.

The photon starting position is chosen to yield either a plane wave or a Gaussian beam profile to account for strongly focused Gaussian beams if necessary. Until now the initial direction of photon propagation is chosen to be parallel to the z-axis of our coordinate system.

We use standard Fresnel-formulas to calculate the coefficients (used as probabilities) for refraction and reflection at the tangential surface to the point where the photon hits the host boundary. We have to take into account the angle a between the photon direction and the tangential plane normal, the state of polarization with respect to the plane of incidence, and the optical properties of the media involved. Phase jumps upon reflection are also considered. Mie theory is employed to calculate the scattering efficiency for Q_s and the parameter of asymmetry g of the scattering inhomogenities from their size dinh and index of refraction $n + i\,k$, relative to the host material. Absorption from neither the host medium nor the dispersed phase has been considered yet. This can be implemented without difficulty. As we restricted ourselves to mono-sized scattering particles, no size averaging has been implemented For the calculation of the scattering angles from random numbers we prefer to use the well known Henyey Greenstein phase function $p_{HG}(\theta,g)$ rather than the detailed Mie phase function $p_{Mie}(\theta, d_{inh}, n + i\,k)$, though the value for g is taken from Mie calculations. [16] The main advantage in using p_{HG} instead of p_{Mie}, lies in the fact that p_{HG} can be inverted analytically to

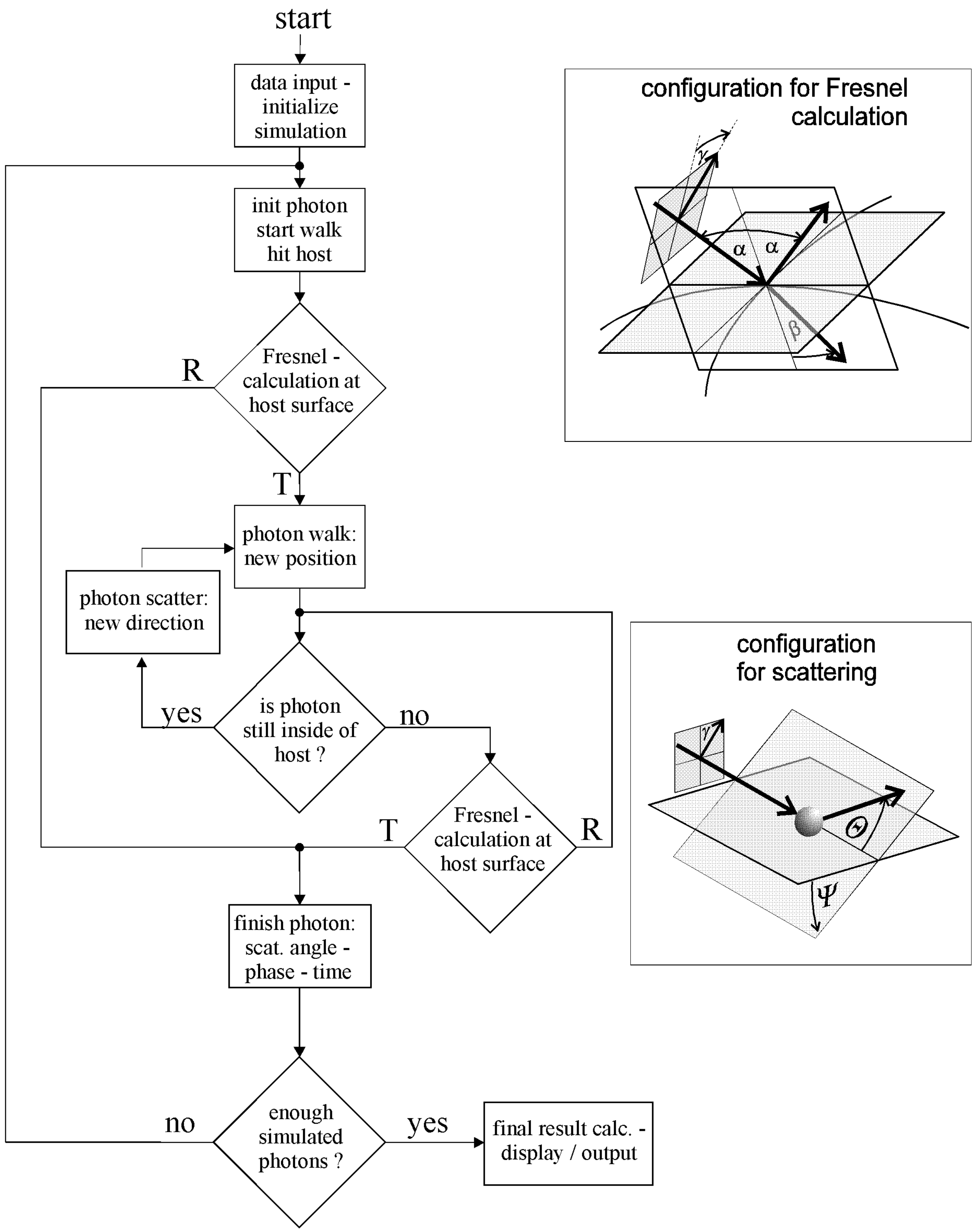

Figure 2: *Flow diagram of the Monte Carlo algorithm. Photons which are refracted into the host are repeatedly moved forward (change of photon position) and scattered (change in photon direction*). On *each crossing of the host boundary Fresnel equations* are employed. *The geometries for scattering (lower right),* reflection, and refraction (*upper right) are also depicted.*

yield a scattering angle θ from a random number $R \in [0;1]$:

$$\cos\theta = \frac{1}{2g}\left(1 - g^2 + -\left(\frac{1-g^2}{1+g-2gR}\right)^2\right) \tag{2}$$

A numerical integration of the phase function is therefore redundant. The overall effects of this approximation on the result of radiative transfer simulations have been shown to be negligibly small in plane parallel geometry. [17]

If c is the volume concentration of scatterers inside the host, the number density n/V is found to be

$$\frac{n}{V} = \frac{6c}{\pi d_{inh}^{3}} \tag{3}$$

The scattering coefficient E_s, which is equivalent to the reciprocal of the mean free distance for photons $\bar{l}$ is then given by

$$E_s = \frac{l}{\bar{l}} = \frac{n}{V} Q_s^{Mie} \frac{\pi d_{inh}^{2}}{4} = \frac{3}{2} c Q_s^{Mie} \frac{1}{d_{inh}} \tag{4}$$

The path length $\bar{l}$ of photons between succeeding scattering processes is chosen randomly on a logarithmic scale with an average value of $\bar{l}$.

Finally we assign an optical depth τ to the host particle by choosing an average photon pathlength of $\pi d_{host}/2$:

$$\tau = E_s \pi \frac{d_{host}}{2} = \frac{3}{2} c Q_s^{Mie} \frac{d_{host}}{d_{inh}} \tag{5}$$

The simulation process of a photon is finished when it leaves the host particle after a walk process and the Fresnel calculation decides for a refraction. Only for the limiting case of vanishing concentration of scattering particles do we have to terminate photons which are captured inside the host due to total reflection. Otherwise, they would walk around somewhere close the host surface forever. There is no limitation concerning high orders of refraction or long photon path due to multiple scattering and reflection at the host surface.

The final direction and phase of the photons are evaluated. The contribution of each photon to the scattering pattern on an imaginary detector placed in the far field of the host is calculated. We consider both coherent and incoherent addition of photons. As mentioned before the coherent addition is necessary to obtain the undisturbed scattering pattern in the homogeneous case. Incoherent addition will be important when multiple scattering takes places. Here the photon phases will be randomly distributed. Thus result of a coherent addition will vanish.

5. Results and Discussion

We performed Monte Carlo light scattering simulations for a 50 µm water ($m = 1.33 + i\,0$) sphere, a light wavelength of $\lambda = 633$ nm, and a plane incident wave in a 2d-geometry. The properties of the dispersed phase were chosen to be close to those of lipid in milk. [18] For all following simulations we used $m = 1.46 + i\,0$, a fixed globule size of $d_{inh} = 0.5$ µm, $g = 0.80$, and $Q_s = 1.0$.

Fig. 3 shows the Monte Carlo result for the case of a homogeneous sphere. All important features of the Mie result can be observed. First and second order rainbows, as well as additional oscillations due to interference with reflected photons can be seen. The frequency and phase of the oscillations is in perfect agreement with the Mie result in those angular areas, where reflected and first order refracted photons contribute. Due to minor deviations of the reflected contribution in forward direction as well as the missing diffraction part the amount of the oscillations differs slightly from the Mie result.

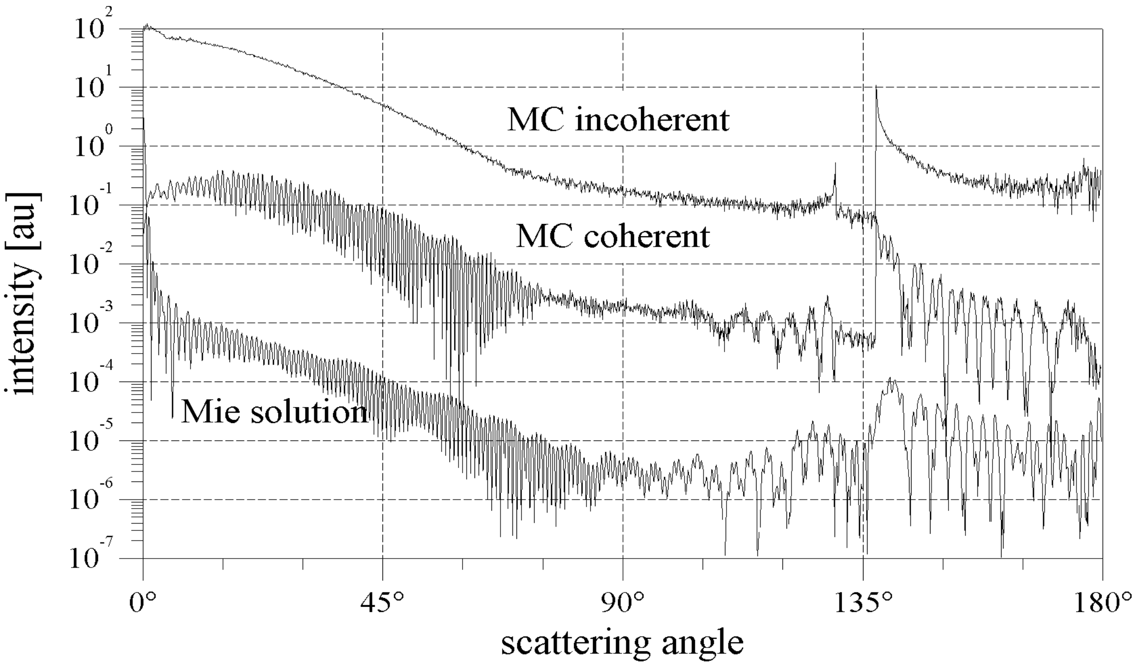

Figure 3: *Monte Carlo and Mie results (from top: coherent sum, incoherent sum, Mie) for a homogeneous 50 µm water sphere.* $2 \cdot 10^6$ *photons haue been followed, the angular resolution is 2.18 mrad (1440 classes).*

In fig. 4 the scattering distributions of inhomogeneous spheres are depicted. The left figure shows the combined contribution of all photons according to a coherent addition in a restricted angular area. The typical size-dependent oscillations due to interference between first order refracted and reflected photons can be seen. The bottom curve is the Mie result. Each curve is shifted in vertical direction for reasons of clarity. The volume concentration c is given as a label to each curve. *c* was increased from *c* = 0 % (homogeneous sphere) to *c* = 5.3 %.

The right figure concentrates on the scattering pattern in backward direction. Actually only the area of the first order rainbow is depicted. We compare the contribution of the second order refracted photons (and their interference with each other) with the exact solution according to the laws of geometrical optics. This leads to the Young, solution for the rainbow. Again, c was increased from the homogeneous case up to *c* = 2.7

With increasing concentration a gradual transition from the scattering pattern of the homogeneous sphere takes place in both areas. The loss of structure in the scattering pattern is clearly visible at the rainbow angle and above. This is obvious as second and higher order photons have to take a long path through the inside of the host and are therefore strongly subjected to scattering by the dispersed phase.

This is different in the area where first order refraction is dominant (left figure). Surprisingly we find an increase of the modulation depth for higher concentration of scatterers c. This is due to the fact that the dominance of the first order refracted photons is reduced by multiply scattered photons and their corresponding loss of coherence whit respect to undisturbed refracted photons. Thus reflection gains importance. Actually the reflected part of the scattering pattern is the only one to remain visible for very large concentrations c. In this case all phase information is lost. Only those photons which do not enter the host contribute to the coherent scattering pattern.

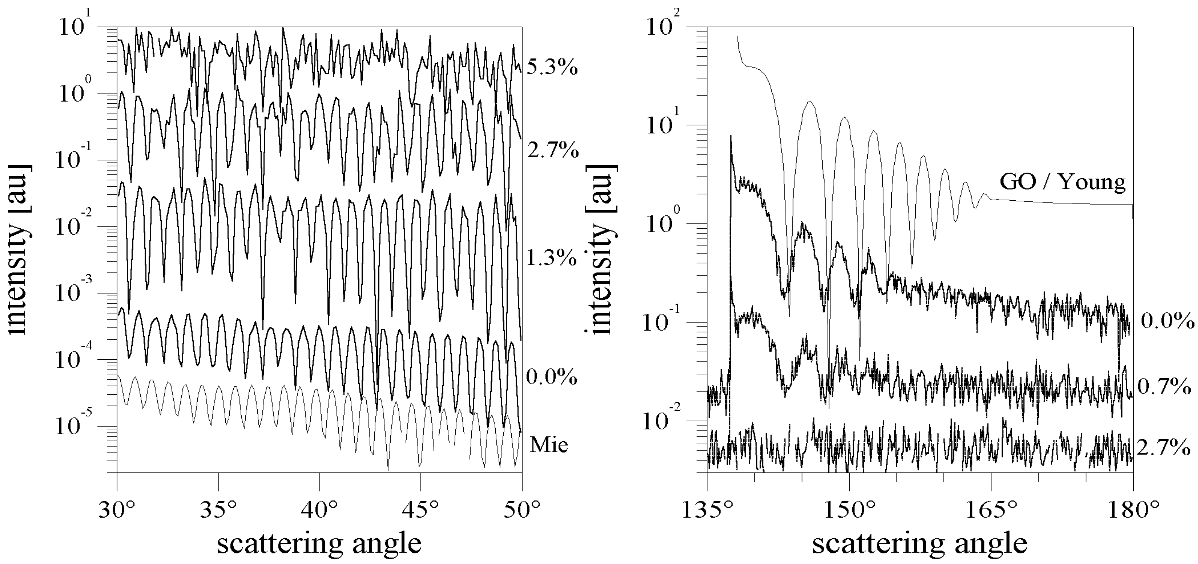

Figure 4: *Monte Carlo results for the seattered intensiv of inhomogeneous spheres (d_{host} = 50 µm, d_{inh} = 0.5 µm). The left diagram shows the result for the coherent addition of photons of all possible contributions, while the right diagram shows only the second order refracted photons. The curves are labeled with the underlying volume concentration of scatterers. The reference curves are from Mie theory and the Young rainbow theory, respectively. All curves are shifted in vertical direction. The angular resolution is 2.18 mrad (1440 classes).*

Finally the Schoenberg solution of a sphere with a Lambert surface (eqn. 1) is obtained. We chose an optical depth of 7 = 20 for which the medium can be regarded as optically dense. Fig. 5 shows the resulting scattering pattern of that sphere. The incoherent pattern is shown, as the coherence of all refracted photons is totally lost through multiple scattering,. In accordance with the model described in sec. 3 the contribution of reflected photons has to be subtracted to yield the correct result for a diffuse reflecting sphere.

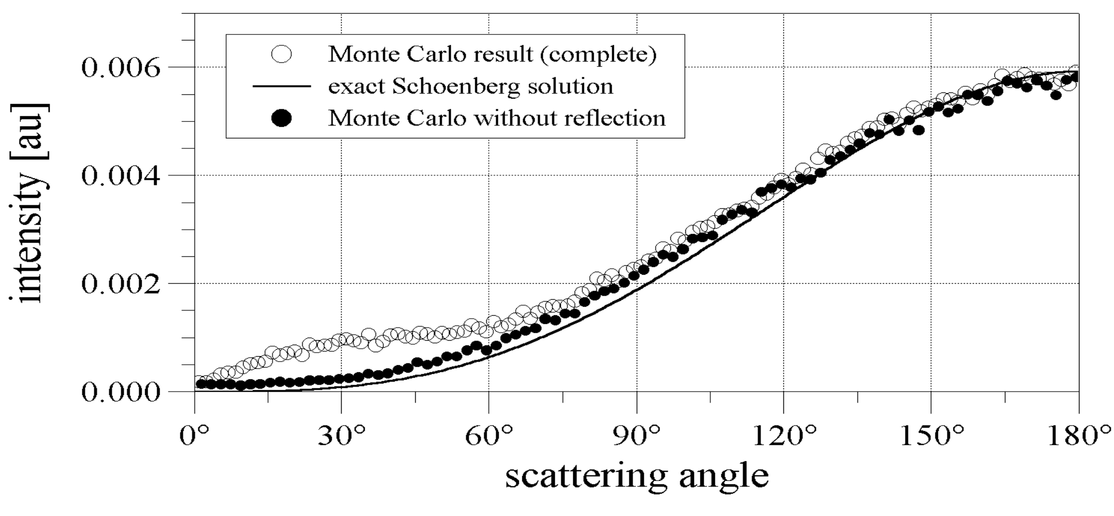

Figure 5: *Comparison between the Monte Carlo result for a 20Mm water sphere with large optical depth (τ = 62.8). The Schoenberg solution is obtained when the contribution of the reflected photons is subtracted from the total incoherent result.*

6. Conclusion

We showed that the scattering pattern of a sphere can be obtained by means of a phase resolved 3-Monte Carlo simulation. The pattern is disturbed when small particles are seeded inside the host. Due to multiple scattering the phase information smears out in those areas where mainly refracted photons contribute.

As no exact solution of the general multiple scattering problem is available at present - reference results exist only for the limiting cases - the veracity of our approach might be questionable to some degree. This is mainly due to the fact that we apply the concept of a radiative continuum to cases of quite limited spacial size and eventually small optical depth. Furthermore we do not pay attention to so called dependent scattering effects, which are known to occur at high volume fractions of scatterers in a participating medium.

Nevertheless it seems to be possible to quantify the influence of size, concentration, and optical properties of a dispersed phase on the scattering behavior of large host particles. An application of our method will follow in order to predict the response of optical sizing techniques to inhomogeneous particles. A gain of information about the composition of a scattering medium (scaled optical depth, albedo) from measurements of the disturbed scattering pattern of a sphere of this medium is also a possible application. Further investigations will focus on a better agreement with GO for the homogeneous sphere limit. The influence of inhomogenities will be studied in detail in a small angular intervals in order to enable comparisons with experimental results. For the measurement of disturbed scattering patterns a setup is available which consists of a electrodynamical particle trap., a laser, and a CCD line sensor.

REFERENCES

[1] Bauckhage, K., The Phase-Doppler-Difference-Method, a New Laser-Doppler Technique for Simultaneous Size and Velocity Measurements Part I: Description of the Method, Part. Part. Syst. Charact., Vol.5, pp.16-22, 1988.

[2] Köser, 0. and Wriedt, T., Iterative inversion of phase-Doppler-anemometry size distributions from sprays of optically inhomogeneous liquids, Applied Optics, Vol.35, No.15, pp.2537-2543, 1996.

[3] Wriedt, T., Mitschke, M. and Lehmann, P., Evaluation of PDA measuring results obtained from sprays of inhomogeneous liquids, Optical Techniques in Fluid, Thermal, and Combustion Flow, Editors Cha, S.S., Trolinger,J.D., Proc. SPIE 2545, pp.435-443, 1995.

[4] Manasse, U., Wriedt, T. and Bauckhage, K., Reconstruction of Real Size Distributions Hidden in Phase-Doppler Anemometry Results Obtained from Droplets of Inhomogeneous Liquids, Part. Part. Syst. Charact., Vol.11, pp.84-90, 1994.

[5] Gu, J., Ruekgauer, T.E.. Xie, J.G., Armstrong, R.L., Effect of particulate seeding on microdroplet angular scattering, Optics Letters, Vol.18, No.16, pp.1293-1295, 1993.

[6] Bronk, B.V., Smith, ivl.J., Arnold, S., Photon-correlation spectroscopy from small spherical inclusions in a micrometer-sized electrodynamically levitated droplet, Optics Letters, Vol.18, No.2, pp.93-95, 1993.

[7] Wriedt, T., Available electromagnetic-scattering programs, IEEE Antennas and Propagation Magazine Vol.36, No.6, pp.61-63, 1994. the list is also available via WWW: http.-//

imperator.cz'p-iwl.uni-bremen.de/'fg01/codes2.htmL
[8] Evers, T., Dahl, H., Wriedt, T., Extension of the program 3D MMP with a 5th order Gaussian beam, Electronic Letters, Vol.32, No.15, pp.1356-1357, 1996.
[9] Borghese, F., Denti, P., and Saija, R., Optical properties of spheres containing several spherical inclusions, Applied Optics, Vol.33, No.3, pp.484-493, 1994.
[10] Fuller, K.A., Scattering and absorption cross section of compounded spheres. 111 Spheres containing arbitrarily located spherical inhomogenities, J. Opt. Soc. Am. A, Vol.12, No.5, pp.893-904, 1995.
[11] Bohren, C.F., Huffman, D.R., Absorption and Scattering of Light by Small Particles, John Wiley k Sons, 1983.
[12] van de Hulst, H.C., Light Scattering by Small Particles, Dover Publications, 1981.
[13] Schoenberg: Handb. Astrophysik Vol.2, pp.255, 1929.
[14] Siegel, R., Howell, J.R., Thermal Radiation Heat Transfer, Hemisphere Publishing Corp., third edition, 1992.
[15] Press, W.H., Flannery, B.P., Teukolsky, S.A., Vetterling, W.T., Numerical Recipes in C: The Art of Scientific Computing, Cambridge University Press, 1992.
[16] Henyey, L.G., and Greenstein,J.L., Diffuse Radiation in the Galaxy, Astrophys. J., Vol.93, pp.70-83, 1941.
[17] Toublanc, D., Henyey-Greenstein and Mie phase functions in Monte Carlo radiative transfer computations, Applied Optics, Vol.35, No.18, pp.3270-3274, 1996.
[18] Nallasamy. M., Mie scattering functions for milk fat globules, Rev. Sci. Instrum., VOI.55, pp.814-815', 1984.