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An extension to the Rayleigh–Gans formula: model of partially absorbing particles

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The Rayleigh–Gans (R–G) approximation is widely employed in various optical models to simulate the optical response of transparent media. However, the R–G concept can only succeed if the refractive index of scatterers approaches that of surrounding medium. In addition, the sizes of scattering domains are assumed to be small enough. Because of these reasons, the validity of R–G solution is fairly limited. In this paper, a semi-analytical extension to the R–G theory is introduced, resulting in an approximate formula for efficiency factor for scattering Q_{sca}. It is proven numerically that this formula works much better than that for traditional R–G model. The computations have been made on absorbing particles with sizes comparable to or smaller than the wavelength of an incident radiation. The conventional R–G theory is either inapplicable or at least inappropriate for such particles.

Keywords: scattering, absorption, spherical particle, Rayleigh–Gans approximation.

1. Introduction

The Rayleigh–Gans (R–G) approximation is commonly used in diverse fields of science to simulate the optical response of various ensembles of optically soft particles [1–6]. The particles are considered to be essentially smaller than the wavelength of an incident radiation and their refractive index approaches that of surrounding medium. Most frequently it deals with scattering domains suspended in a host with similar optical properties [7] so the differences between dielectric properties of any inclusion and the host are satisfactory small. Such model was successfully employed, *e.g.*, in characterization of NaF particles during deliquescence experiments [8]. The Rayleigh–Gans concept can successfully simulate an optical behavior of both the glass-ceramics during formation of crystalline phases [9, 10] and phytoplankton dispersed in ocean water [11]. But, the R–G is also often applied to other types of particles some of which have sizes (or refractive indices) far behind the theoretical limits. For instance, the ice particles in noctilucent clouds are extremely small thus satisfying one of two specific requirements of R–G approximation. However, the refractive indices of ices violate the condition $|m-1| \ll 1$ ($m = n + in'$ is the complex refractive index with n being the real part and n' the imaginary part).

It is well recognized that the chemistry of natural particles is scarcely simple. Rather than being homogeneous media, the particles are built of various species including absorbing materials. Unfortunately, the R–G formalism is inapplicable to such particles, so its main strength (*i.e.*, the analytical formulation for spheres and cylinders [12]) is then definitely cancelled. This is one of reasons for the continuously growing interest in semi-analytical extensions to the R–G approximation for partially absorbing particles. In this paper we present a simple concept of linear absorption that is incorporated into solution scheme and enables us to find an analytical expression for efficiency factor for scattering $\tilde{Q}_{\text{sca}}$. Here we have assumed the absorbing particles with $|m| <\approx 2$ and sizes smaller than the wavelength of an incident radiation. The analytical form of $\tilde{Q}_{\text{sca}}$ is advantageous in solving the distinct inverse problems [13, 14], and, it also permits CPU non-intensive computations for the large ensembles of scattering particles.

2. Theoretical concept of the Rayleigh–Gans approximation

The Rayleigh–Gans approximation is a well-known solution to the light scattering by small particles and it has been intensively discussed across scientific literature (see, *e.g.*, [7]). In spite of this fact, we will shortly introduce some basic formulae. The R–G theory requires:

– the complex refractive index $m = n + in'$ of a particle relative to that of surrounding medium to be close to unity, *i.e.*,

$$|m-1| \ll 1 \tag{1}$$

– and the phase shift to be small enough, so

$$kd\,|m-1| \ll 1 \tag{2}$$

Here d is a characteristic linear dimension of a particle and $k = 2\pi/\lambda$ is the wavenumber (λ is the wavelength of an incident radiation).

Incident plane-wave is described by electric vector $\mathbf{E}^i$

$$\begin{pmatrix} E^i_{\|} \\ E^i_{\perp} \end{pmatrix} = \begin{pmatrix} E^i_{0\|} \\ E^i_{0\perp} \end{pmatrix} \exp\left[i(\mathbf{k}\cdot\mathbf{r} - \omega t) \right] \tag{3}$$

where E^i_0 denotes the amplitude of an incident field, $\mathbf{k}$ is the wave-vector, $\mathbf{r}$ is the position vector, ω is the angular frequency, and t is the time. In the far-field approximation, the incident E^i and scattered E^s electric fields are interrelated through the scattering matrix **S**

$$\begin{pmatrix} E_{||}^s \\ E_{\perp}^s \end{pmatrix} = \frac{\exp\left[ik(r-z)\right]}{-ikz} \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \begin{pmatrix} E_{||}^i \\ E_{\perp}^i \end{pmatrix} \tag{4}$$

where the elements of the amplitude scattering matrix for a homogenous sphere are as follows

$$\begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} = -ik^3\alpha \begin{pmatrix} \cos(\vartheta) & 0 \\ 0 & 1 \end{pmatrix} R(\vartheta, \varphi) \tag{5}$$

The scattering angle ϑ is the angle contained by directions of incident and scattered beams. Assuming V is the volume of a homogenous scatterer, the isotropic polarizability α can be expressed as

$$\alpha = \frac{m-1}{2\pi} V \tag{6}$$

The term $R(\vartheta, \varphi)$ corresponds to the form-factor given by the equation

$$R(\vartheta, \varphi) = \frac{1}{V}\int_V \exp(i\delta)\mathrm{d}V \tag{7}$$

with δ being the phase lag between any two rays scattered toward direction (ϑ, φ). For a homogenous sphere, the phase difference δ is

$$\delta = 2k\xi\sin\left(\frac{\vartheta}{2}\right) \tag{8}$$

where ξ is the distance from the origin of a local coordinate system to a plane of constant phase. Combining the previous formulae, the form-factor will read [7]

$$R(\vartheta) = \frac{1}{V}\int_{-a}^{a} \exp\left[i2k\xi\sin\left(\frac{\vartheta}{2}\right)\right] A(\xi)\mathrm{d}\xi \tag{9}$$

where $A(\xi) = \pi(a^2 - \xi^2)$ is the area of a slice cut of a spherical particle with radius a [12]. Integrating Eq. (9) one can obtain

$$R(\vartheta) = \frac{3}{u^3}\left[\sin(u) - u\cos(u)\right] \tag{10}$$

where $u = 2ka\sin(\vartheta/2)$.

The Rayleigh theory dictates that the intensity of scattered light is inversely proportional to the fourth power of λ, and, the angular pattern behaves like $(1 + \cos^2(\vartheta))$. In the Rayleigh–Gans regime the intensity of a scattered beam is determined as the product of the intensity for Rayleigh scattering and the factor $|R(\vartheta)|^2$, *i.e.*,

$$I = \frac{1 + \cos^2(\vartheta)}{2} \frac{k^4 V^2}{r^2} \left(\frac{m-1}{2\pi}\right)^2 |R(\vartheta)|^2 I_0 \tag{11}$$

where an incident light is considered to be unpolarized.

The optical behavior of disperse media is commonly characterized by the so-called scattering cross-section, C_{sca}. This parameter is determined as follows

$$C_{\text{sca}} = \frac{\pi}{k^2} \int_0^{\pi} \left(|S_1|^2 + |S_2|^2\right) \sin(\vartheta) \mathrm{d}\vartheta \tag{12}$$

For small and optically soft particles, Eq. (12) transforms to

$$C_{\text{sca}} = \frac{4}{9} \pi a^6 k^4 (m-1)^2 \int_0^{\pi} |R(\vartheta)|^2 \left[1 + \cos^2(\vartheta)\right] \sin(\vartheta) \mathrm{d}\vartheta \tag{13}$$

If the absorption cross-section C_{abs} is added to the C_{sca}, the intensity decay of a light beam can be computed for any optical material or for any optical medium. The main intention of this paper is to extend R–G approximation to make it applicable to both non-absorbing and absorbing particles with sizes smaller than or comparable to the wavelength of an incident radiation.

3. Optical paths of individual beams in an optically soft spherical particle

The R–G theory assumes the light beams change directions negligibly at the either interface of a spherical inclusion. This is because the difference between refractive indices of both media is considered to be small. In such a case, the geometrical model depicted in Fig. 1 becomes acceptable and the trajectory of a beam inside the sphere is given as a sum of l_1 and l_2. Here l_1 is measured from the point where the beam intersects the scatterer to the point of a scattering event. The distance l_2 is measured from the point of the scattering event to the edge of the sphere. Mathematically it is expressed as follows:

$$l_1 \approx \left| r\cos(\theta)\sin(\varphi) + \sqrt{a^2 - r^2 + r^2\cos^2(\theta)\sin^2(\varphi)} \right| \tag{14}$$

$$l_2 \approx \left| r\left[\cos(\theta)\sin(\varphi)\cos(\vartheta) + \cos(\varphi)\sin(\vartheta)\right] - \right.$$
$$\left. - \sqrt{a^2 - r^2 + \left[r\cos(\theta)\sin(\varphi)\cos(\vartheta) + r\cos(\varphi)\sin(\vartheta)\right]^2} \right| \tag{15}$$

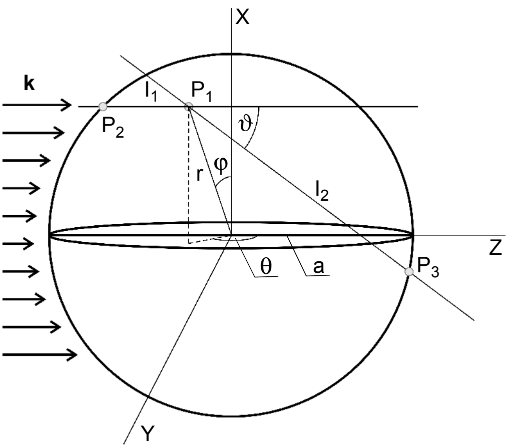

Fig. 1. The geometry of a beam trajectory inside an optically soft spherical particle. A beam that is scattered in point P_1 crosses the particle surface in P_2 and P_3. The particle radius is a.

If the real part of a particle refractive index grows, the formulae for l_1 and l_2 will change in accordance with the Snell's law of refraction. Independent of these formulae, a presence of absorbing species in the particle would cause a decay of the amplitude of a propagated wave. For a plane wave satisfying Eq. (3) the absorption term will read $\exp[-kn'(l_1 + l_2)]$. Then Eq. (7) is formally written as follows

$$R(\vartheta) = \frac{1}{V}\int_V \exp\Big[i\delta + i(n-1)k(l_1 + l_2)\Big]\exp\Big[-kn'(l_1 + l_2)\Big]\mathrm{d}V \tag{16}$$

where n' is the imaginary part of the complex refractive index of a spherical particle.

In general, the phase difference δ can be obtained in a form

$$\delta = k\Big[\mathbf{r}\cdot(\mathbf{e}_z - \mathbf{e}_r)\Big] \tag{17}$$

where $\mathbf{r}$ is the position vector of an arbitrary volume-element with respect to the center of the spherical particle, $\mathbf{e}_z$ is the unit vector parallel to the direction of propagation of the incident wave and $\mathbf{e}_r$ is the unit vector parallel to the scattered wave. The unit vectors read

$$\mathbf{e}_z = [0;\ 0;\ 1] \tag{18a}$$

$$\mathbf{e}_r = [\sin(\vartheta);\ 0;\ \cos(\vartheta)] \tag{18b}$$

After a bit of manipulations, the function $R(\vartheta)$ for an optically soft absorbing medium can be approximated by the formula

$$R(\vartheta) = \frac{1}{V}\int_0^{2\pi}\int_0^{\pi}\int_0^{a}\exp\Big[i\delta + i(n-1)k(l_1 + l_2)\Big]\exp\Big[-kn'(l_1 + l_2)\Big]r^2\sin(\varphi)\mathrm{d}r\mathrm{d}\varphi\mathrm{d}\theta \tag{19}$$

4. Approximate analytical extension to the R–G formula for particles with |*m*| <≈ 2

The integral form of Eq. (19) is too complex to be solved analytically. However, a set of more-or-less convenient simplifications can be found depending on approximation taken into account. Usually an underintegral function can be averaged over the integration interval, thus allowing us to proceed with further analytical derivations. We have made an averaging over azimuth angle for every slice perpendicular to the line in the scattering plane that bisects the angle $(\pi - \vartheta)$ between the incident and scattered beam (see the R–G concept in [12]). After some mathematical manipulations we obtained the analytical solution to Eq. (13) except for the following integral

$$\int_{-1}^{1} \exp\left\{-2a\tilde{q}_1(1+t) - 2a\tilde{q}_2\sqrt{1-t^2}\right\}(1+t^2)\,\mathrm{d}t \tag{20}$$

that needs to be approximated. Here $\tilde{q}_1$ and $\tilde{q}_2$ are scaling constants resulting from previous azimuthal averaging. Both depend on particle microphysical properties and wavelength of an incident radiation. The optimum values of $\tilde{q}_1$ and $\tilde{q}_2$ can be retrieved by minimizing the differences between exact and approximate computations. We have found that

$$\tilde{q}_i \approx k\sqrt{n' B_i |m-1|} \tag{21}$$

with B_i $(i = 1, 2)$ being the free parameters. The extensive mathematical treatment resulted in the analytical expression for the efficiency factor for scattering $Q_{\mathrm{sca}} = \pi a^2 C_{\mathrm{sca}}$, which has now the form

$$\tilde{Q}_{\mathrm{sca}} = Q_{\mathrm{sca}}^{\mathrm{R-G}}\left(\frac{3}{8}\right)^2\left\{\frac{1-\exp\left[-4\tilde{q}a\right]}{\tilde{q}a} + \frac{1-\exp\left[-4\tilde{q}a\right]\left[8(\tilde{q}a)^2+4\tilde{q}a+1\right]}{4(\tilde{q}a)^3} + \right.$$
$$\left. + \frac{\exp\left[-4\tilde{q}a\right]\left[4\tilde{q}a+1\right]-1}{2(\tilde{q}a)^2}\right\}^2 \tag{22}$$

with $\tilde{q}$ satisfying Eq. (21) and $B \approx 0.05$. Finally,

$$Q_{\mathrm{sca}}^{\mathrm{R-G}} = |m-1|^2\left\{\frac{5}{2} + 2x^2 - \frac{\sin(4x)}{4x} - \frac{7\left[1-\cos(4x)\right]}{16x^2} + \right.$$

$$+\left(\frac{1}{2x^2}-2\right)\left[\gamma+\ln(4x)-Ci(4x)\right]\Bigg\} \qquad (23)$$

is a well-known efficiency factor for scattering in Rayleigh–Gans approximation [12]. The $\gamma = 0.577$ is Euler's constant and $x = ka$ is the so-called size parameter. Because of Eq. (21), the $\tilde{q}a$ is a product of size parameter x and a scaling factor $\sqrt{n'B|m-1|}$. If either $n' \to 0$ or $a \to 0$, the $\tilde{Q}_{\rm sca}$ approaches $Q_{\rm sca}^{\rm R-G}$ since the rest part of Eq. (22) approaches unity. If the particle radius grows to infinity, the $Q_{\rm sca}^{\rm R-G}$ will be proportional to x^2, but the factor at the right-hand side of Eq. (22) behaves like x^{-2}. Therefore, Eq. (22) should work better than Eq. (23) and it is expected to be applicable to moderate size parameters, too.

Note, that various approximations to $Q_{\rm sca}$ can be found in scientific literature. The common aim is to find a formula that is simple or even analytical, computationally non-intensive, and well applicable to various problems of the optics of disperse media. However, many of sufficiently accurate approximations are neither analytical nor convenient for rapid numerical computations. Especially, it is important in solving the inverse problems where $Q_{\rm sca}$ usually poses as the kernel of an integral equation. In fact, the analytical form of Eq. (22) is advantageous because it makes the theoretical interpretation of scattering features of small particles easily possible. Implementation of Eq. (22) is simple since it does not require any numerical integration.

One of the analytical solutions to the inverse problems is based on Mellin's transform, where the kernel of integral equation satisfies the condition $Q_{\rm sca}(\tilde{q}, a) = Q_{\rm sca}(\tilde{q}a)$. This concept can be used to determine the size distribution of small particles dispersed in a transparent medium. If the transform of Eq. (22) is found, then the computation of the particle size distribution is extremely fast and straightforward (see, *e.g.*, [15]).

5. Numerical demonstrations

The Rayleigh–Gans formalism is usually valid for optically soft particles with $n \approx 1$ and $n' \ll 1$. But, the real part of particle refractive index n usually ranges from 1.3 to 2 in the visible spectral range. For instance, n is about 1.3 for water ice [16], 1.33 for water under normal atmospheric conditions [17], and even higher for other species like space materials, silicates, sulphides, or carbides [18]. In such a case Eq. (23) becomes inaccurate. If incorporated into optical models without a detail analysis, the subsequent physical interpretations may fail. But, since the $Q_{\rm sca}^{\rm R-G}$ is frequently used as an underintegral function in solving diverse problems of the optics of polydisperse systems, the extreme behavior of $Q_{\rm sca}^{\rm R-G}$ may be partly suppressed. As discussed in [19], the R–G approximation has been applied to atmospheric aerosols. For example, the Shifrin's expansion tends to be applicable to both nonspherical particles and particles with moderate values of size parameters [15]. Nevertheless, the existence of analytical solution for $Q_{\rm sca}^{\rm R-G}$ encourages many scientists to use it also for absorbing particles. Unfortunately, obtained results then deviate significantly from those obtained by means of rigorous Mie theory.

The computations based on Eq. (22) clearly demonstrate that $\tilde{Q}_{\rm sca}$ represents one of appropriate approximations to particles with refractive indices up to 2 and particles with radii smaller than or comparable to λ.

Figures 2 and 3 document the overall behavior of $Q_{\rm sca}^{\rm R-G}$ and $\tilde{Q}_{\rm sca}$. Both are related to the reference data computed using the rigorous Mie theory. As shown here, the R−G model significantly deviates from Mie theory when size parameters x are larger than 2. The real and the imaginary parts of the complex refractive index are varied over large range. If using the traditional R−G formula (Eq. (23)), the ratio of $Q_{\rm sca}^{\rm R-G}/Q_{\rm sca}^{\rm Mie}$ can be 10 or more for $x \approx 6$. Such an error is definitely non-acceptable and it makes Eq. (23) inappropriate for any practical use, especially for particles with elevated refractive indices. The concept of Eq. (22) appears to be more convenient, since the error margin is significantly lower. The peak errors of $\tilde{Q}_{\rm sca}$ are found for large values of n and small values of n'. As n' grows, the discrepancies between $\tilde{Q}_{\rm sca}$

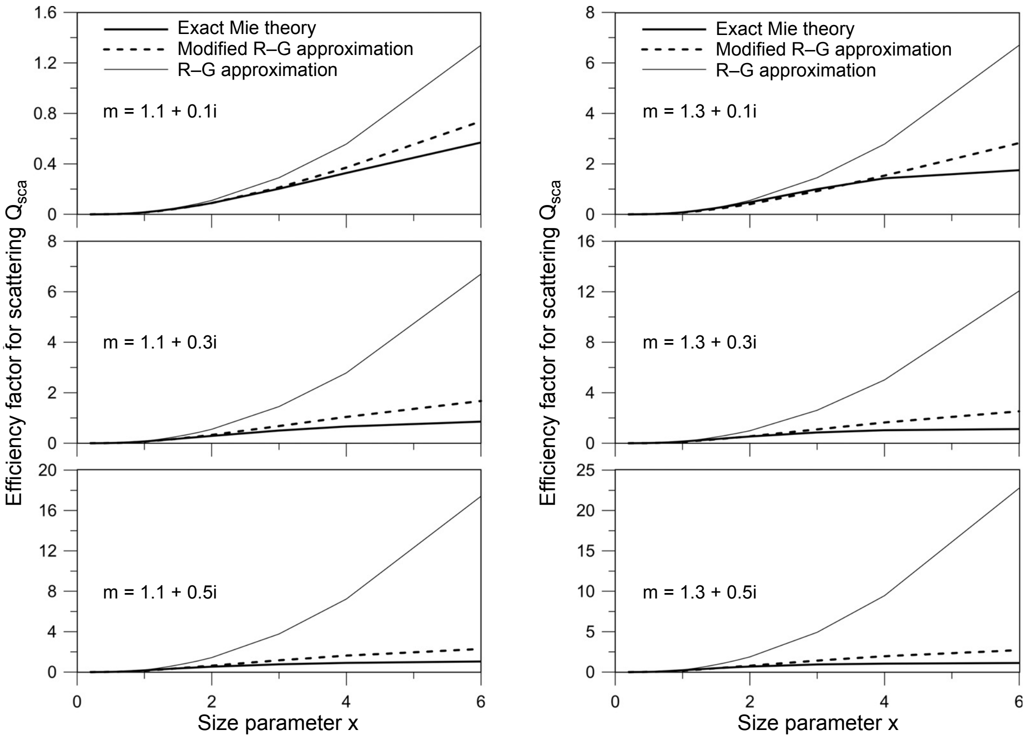

Fig. 2. The efficiency factor for scattering computed using Rayleigh–Gans approximation, Mie theory and Eq. (22). The real parts of particle refractive indices are considered to be 1.1 (left pane) and 1.3 (right pane). The imaginary part n' varies from 0.1 to 0.5.

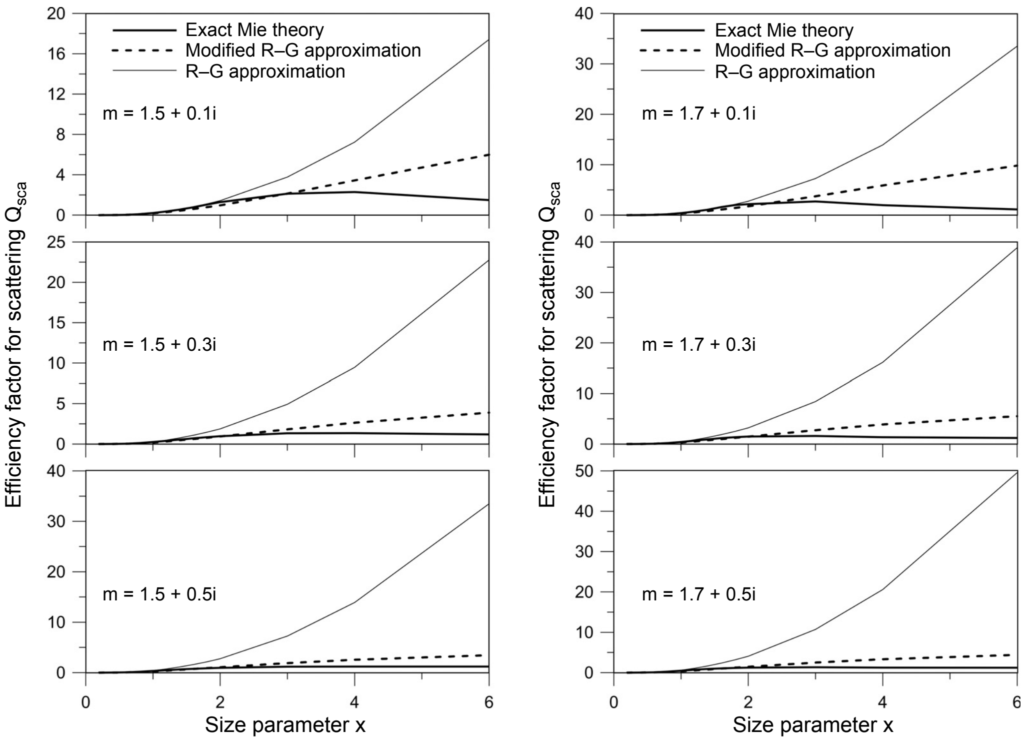

Fig. 3. The same as in Fig. 2 but for real parts of particle refractive indices 1.5 (left pane) and 1.7 (right pane).

and Q_{sca}^{Mie} decrease essentially. Equation (22) shows enhanced accuracy in comparison with Eq. (23). Assuming the size parameters are below 4, the $\tilde{Q}_{sca}$ can be used in solving the forward and/or inverse problems for optically transparent media composed of polydisperse system of scattering/absorbing particles.

6. Concluding remarks

The Rayleigh–Gans approximation is regularly used in simulating the optical behavior of small particles distributed in media with similar optical properties. Thanks to the analytical form, the R–G computations are extremely fast thus making the R–G approach very attractive in various fields of science. However, the validity of R–G model is strictly limited to the particles with $|m-1| \ll 1$ and $kd|m-1| \ll 1$, where d and m are characteristic size and refractive index of the particle, respectively. The parameter $k = 2\pi/\lambda$ is the wavenumber that is inversely proportional to the wavelength of an incident radiation λ. If R–G concept is applied to large absorbing particles, the numerical results become inaccurate and this may lead to an incorrect interpretation of measured data. This has also important consequences, *e.g.*, in evaluation of an optical response of interstellar dust particles. Although these particles are usually smaller than 1 μm, many of them can be composed of highly absorbing materials. Another typical misuse of R–G approach is an evaluation of glass transparency in the course of lithium disilicate glass crystallization. This process can be related to both nucleation of new crystalline phases and the subsequent growth of respective nuclei. Even if the optical behavior of these particles is perfectly consistent with R–G theory at early phases of crystallization, the R–G approach fails to describe this system in later phases of crystallization when the initially transparent glass transforms into semitransparent or opaque glass-ceramics material. To obtain a reasonable result, the R–G model has to be replaced by more accurate Mie theory. Such a modeling might become cumbersome (at least under certain circumstances) because of more complex numerical implementation and CPU consumption.

In this paper we have derived an approximate formula for $\tilde{Q}_{\mathrm{sca}}$ that improves the conventional Rayleigh–Gans formula for $Q_{\mathrm{sca}}^{\mathrm{R-G}}$. Both functions are interrelated through the factor that depends on particle refractive index $m = n + in'$ and particle radius a. This factor approaches unity if either a or n' are as small as zero. The numerical experiments have proven that the improved model simulates the efficiency factor for scattering much better than R–G theory (Eq. (23)). Since the new model is applicable to real values of n and n', Eq. (22) is a good candidate for routine usage in processing the optical data, and, in solving the inverse problems where $\tilde{Q}_{\mathrm{sca}}$ represents a kernel of the integral equation. Typically it deals with multispectral transparency data, where the particle size parameter is below 4–6. In contrast to $Q_{\mathrm{sca}}^{\mathrm{R-G}}$, the $\tilde{Q}_{\mathrm{sca}}$ can provide better fit to optical properties of a polydisperse system composed of scattering/absorbing particles. For such modeling purposes it is usually necessary to repeat the entire set of calculations. Therefore the new analytical formula can save the computational time significantly, keeping the error margin below that of R–G.

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