

Asymmetry coefficient for large optically soft spherical particles

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Abstract. The basic mechanisms describing radiation scattered by large optically soft spheres are presented in this paper. The contributions of different components of geometrical optics to formation of the integral indicatrix have been estimated. The asymptotic equations for the asymmetry coefficient have been obtained. © 2002 Society of Photo-Optical Instrumentation Engineers. [DOI: 10.1117/1.1481901]

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1 Introduction

In the last two decades, the most promising and effective experimental method used for analysis of biological fluids has been flow cytometry. The uniqueness of flow cytometry is that the measurements are performed on separate particles and with high speed (up to 300 000 particles/min). This ensures a high statistical accuracy and allows high sensitivity in revealing small populations. Thus the development of the parametric solution of the inverse light-scattering problem that provides the determination of particle parameters at the comparable rate (~ 2 ms) is a vital problem. Especially broad applications of this express method can be envisioned in further developments of cytometry systems for analyzing the elements of blood and studying the disperse structure of water ecosystems. The problem of using flow cytometry for sorting particles is discussed in detail in Ref. 1.

Among various techniques, the flying light-scattering indicatrix method (FLSI)² deserves special attention since the optical system of the scanning flow cytometer used allows one to measure light scattered by a particle using a fixed detector. Thus, during the passage ("flight") of the particle through the working area of the optical device the angular distribution of the scattered radiation, or the indicatrix (phase function), is obtained.

The angular dependency of the light-scattering intensity (indicatrix) contains significant information about particle parameters (size parameter $\rho = ka = m_0 \pi d / \lambda$ and relative refractive index $m = m_s / m_0 = n + i\chi$, where d and a are the particle diameter and radius, respectively, m_0 is the refractive index of the surrounding medium, m_s is the refractive index of the scatterer, λ is the wavelength of the incident light in vacuum, k is a wave number). Thus the indicatrix can be used to solve the inverse problem of scattering. The Mie computation is widely used for the determination of size, shape, orientation, refractive index, and aggregation of scattering biological particles. Unfortunately, this is a lengthy and complicated method, especially for spheres having two or more layers.

In order to analyze suspensions of biological particles such as erythrocytes, leucocytes, ghosts of erythrocytes, biological

cells, microorganisms, and others by an optical technique, approximation methods are widely used to describe the light-scattering pattern. Among the well-known approximating solutions for the analysis of an indicatrix, the Rayleigh–Gans–Debye approximation (RGD), anomalous diffraction (AD), geometrical optics (GO), Fraunhofer diffraction (FD), eikonal approximation, and the integral wave equation methods are most frequently used. These approximations usually assume that the relative refractive index m of particles is close to 1; thus, the particles are called optically soft.^{3–11}

One of the important characteristics of light-scattering indicatrix is the amount of light flux in the scattering angle range $\theta = [\theta_1, \theta_2]$ (θ is scattering angle).

It is well known that under condition $\rho \rightarrow \infty$ (large particles) fundamental separation of diffracted, refracted, and reflected light results from the Mie series in Ref. 6.

In Ref. 12 geometrical optics solutions for light flux, scattered from large spherical particles with the refractive index more than 1.12 were analyzed. In particular, it has been shown that for the coefficient of asymmetry of light-scattering indicatrix for such particles it is sufficient to take into account only the Fraunhofer diffraction component and three components of geometrical optics (twice refracted, reflected and twice reflected parts). References 6 and 8 also show a validity of asymptotic presentation of a scattered energy by the above mentioned components for water drops. However, the question arises: how does the light flux scattered into back hemisphere depend on m and ρ for the case of $m < 1.12$? The purpose of this work is to solve this problem.

2 Computational Scheme

This work deals with the analysis of the integral indicatrix $F(\theta_0)$, indicatrix $I(\theta)$ and coefficient of asymmetry η of the optically soft homogeneous spheres that are as follows:

$$F(\theta_0) = \frac{2\pi \int_0^{\theta_0} I(\theta) \sin \theta d\theta}{K_{SCA}},$$

$$I(\theta) = \frac{|f(\theta)|^2}{\pi a^2}, \quad (1)$$

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$$\eta = \frac{F(\pi/2)}{F(\pi) - F(\pi/2)},$$

where θ is the scattering angle, $f(\theta)$ is the scattering amplitude, a is the radius of sphere, K_{SCA} is the scattering efficiency factor.

For the analysis of these characteristics we used RGD, FD, and GO approximations. For comparison with exact solution we used Mie computation.^{5,6} For calculation of the value of flux scattered into the backward hemisphere and the asymmetry coefficient η of the light-scattering indicatrix we used the algorithm proposed in Refs. 4 and 13.

3 RGD Approximation

In the case of RGD approximation the form factor is⁶

$$G(u) = \frac{3}{u^3}(\sin u - u \cos u) = \sqrt{\frac{9\pi}{2u^3}} J_{3/2}(u), \quad (2)$$

where $u = 2\rho \sin(\theta/2)$. So the indicatrix of unpolarized light is as follows:

$$I(\theta) = \frac{2\rho^4}{9\pi} |m-1|^2 I_0 G^2(u) (1 + \cos^2 \theta). \quad (3)$$

The integral characteristic I_{θ_0} (energy scattered into solid angle with cone $2\theta_0$) is as follows:

$$I_{\theta_0} = \frac{4}{9} \rho^4 |m-1|^2 I_0 \int_0^{\theta_0} G^2[2\rho \sin(\theta/2)] \times (1 + \cos^2 \theta) \sin \theta d\theta. \quad (4)$$

After integration one can obtain

$$I_{\theta_0} = I_0 |m-1|^2 \left\{ A + 2\rho^2 + (4b^2 - 2A) \frac{\sin(4\rho b)}{4\rho b} + \left[\frac{\cos(4\rho b) - 1}{(4\rho b)^2} \right] (12b^2 - 2A) + \left(\frac{1}{2\rho^2} - 2 \right) S_1(4\rho b) \right\}, \quad (5)$$

where $A = 2 + b^2 - 1/(2b^2)$, $b = \sin(\theta_0/2)$, $S_1(x) = \int_0^x (1 - \cos y)/y dy = \ln(x) + \gamma + Ci(x)$, $Ci(x)$ is cosine integral, $\gamma = 0.57721$.

Thus, the integral indicatrix $F(2\rho b)$ for spheres in the region of validity of RGD approximation is the ratio of Eq. (5) to light-scattering efficiency factor K_{SCA} . Under conditions $\rho \rightarrow \infty$, $u_0 = 2\rho \sin(\theta_0/2) = \text{const}$ it is easy to obtain that $S_1(2u_0) = \text{const}$, $b \rightarrow 0$, $A \rightarrow 1/(2b^2)$, $K_{SCA} \rightarrow 2\rho |m-1|^2$, and thereby

$$F(\rho\theta_0) = 1 - \frac{1}{(\rho\theta_0)^2} + \frac{\sin(2\rho\theta_0)}{(\rho\theta_0)^3} + \left[\frac{\cos(2\rho\theta_0) - 1}{2(\rho\theta_0)^4} \right]. \quad (6)$$

Equation (6) is valid with the error less than 10% for $\rho \geq 5$ and the error decreases with increasing of size parameter.

In case of $\theta_0 = \pi/2$ we have $b = 1/\sqrt{2}$ and $A = 3/2$. In case of $\theta = \pi$ the same values are determined as $b = 1$ and $A = 5/2$. Thus, from Eq. (5) for large optically soft spheres ($\rho \gg 1$) it follows that $F(\pi) = 1$ and

$$F(\pi) - F(\pi/2) = \frac{1 - \ln 2}{2\rho^2}.$$

It means that the light scattering into the backward hemisphere does not depend on the relative size of a particle and is proportional to $|m-1|^2$.

Therefore

$$\eta = \frac{2\rho^2}{1 - \ln 2}. \quad (7)$$

This result is confirmed by exact calculation for $\rho > 10$.⁶

4 Geometric Optics

Though in Ref. 12 it has been shown that for the flux scattered into backward hemisphere it is sufficient to take into account only the first three components of GO, the comparison with the exact calculation contradicts this statement for the optically soft particles ($m < 1.12$).

Let us numerically estimate an asymptote of η predicted by GO approximation for optically soft nonabsorbing spherical particles. For this purpose it is necessary to use the expressions for a scattering angle θ_k of the k th flux^{3,12}

$$\theta_k = (k-2)\pi + 2[\varphi - (k-1)\psi], \quad k = 2, 3, \dots \quad (8)$$

$$\theta_1 = \pi - 2\varphi,$$

where φ is the angle of incidence and ψ is the corresponding angle of refraction. The next step is to find the limits of the integration in equation

$$F_k = \frac{1}{2} \int_{\varphi_2}^{\varphi_1} (\epsilon_{k,1} + \epsilon_{k,2}) \sin \varphi d\varphi, \quad (9)$$

where incident angle values φ_1, φ_2 are selected from the condition that scattered energy hits the backward hemisphere ($\theta_k \in [\pi/2, \pi]$). Values of $\epsilon_{k,1}$ and $\epsilon_{k,2}$ are as follows:

$$\epsilon_{k,j} = (1 - R_j)^2 R_j^{k-1}, \quad j = 1, 2. \quad (10)$$

Reflectance capacity for the two kinds of polarized light can be written in the form

$$R_1 = \left| \frac{\sin(\varphi - \psi)}{\sin(\varphi + \psi)} \right|^2, \quad R_2 = \left| \frac{\tan(\varphi - \psi)}{\tan(\varphi + \psi)} \right|^2. \quad (11)$$

The range $\varphi \in 80^\circ - 90^\circ$ gives the main contribution to the value of F_k calculated with Eq. (9). For some of k th fluxes this angle range is optimal for the backward scattering, while the first and the third fluxes at these values of φ do not scatter into the backward hemisphere. So the contributions to the total scattering into the backward hemisphere of some k th fluxes are comparable to or even more than the contribution of the first and the third fluxes.

The relative values of F_k scattered in the backward hemisphere with variation of m (value of m varies from 1.001 to 1.2) are shown in Figure 1.

Note that for $m < 1.12$ the significant redistribution of the relative contribution to the total value of the energy, scattered in the backward hemisphere by different fluxes, occurs. For example, for a range $m = 1.04 - 1.08$ the fourth component of GO dominates, for $m = 1.02 - 1.04$ the fifth dominates, and so

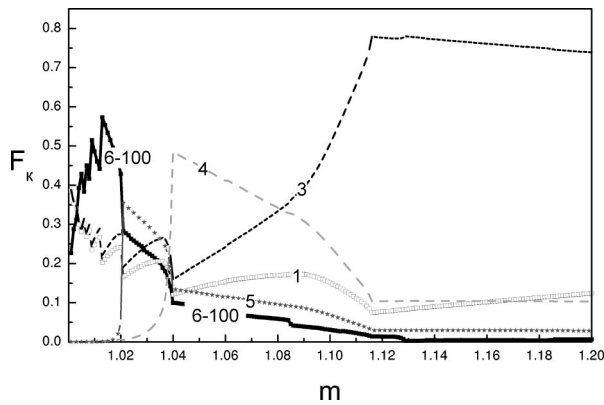


Fig. 1 Relative contribution to total value of the energy scattered in the backward hemisphere by different fluxes vs m . The numbers on the curves correspond to the n th component of GO; 6–100 denotes the sum of components from 6th to 100th.

on. This tendency is shown by a summarized flux (a sum of the components from 6th to 100th). However, with further decreasing of m there are two dominating components, the first and the third, which become equal in magnitude (in the limit $m \rightarrow 1$ the contribution of each component is 0.5). At the same time, at $m > 1.12$ the scattering into the backward hemi-

sphere practically is determined by the third component of GO. This result conforms to the abovementioned conclusion.¹⁰

Estimating scattering in the backward hemisphere, we have neglected the influence of the components of GO with $k > 100$, as their contribution is insignificant.

For the “soft” particles the geometric optics asymptote of the asymmetry coefficient can be estimated as

$$\eta_{GO} = \frac{2 - \sum_{k=1,3,4,\dots} F_k}{\sum_{k=1,3,4,\dots} F_k}. \quad (12)$$

Thus the values of η_{GO} in the case of $m \leq 1.12$ are several times smaller than obtained with account of only the first four components of GO. Figure 2 shows the comparison of η calculated with Eq. (12) and the Mie theory. It is worth nothing that the exact solution gives a smaller magnitude of η than GO approximation in the specified range of ρ and m . However, it is obvious that the value of η approaches the GO asymptote with increasing of $\Delta = 2\rho|m - 1|$.

Let us trace the singularities in the behavior of η for absorptive particles with a small value of the imaginary part χ of the refractive index. In this case Eqs. (8)–(12) can be used with modifications due to absorption of the rays after their passage through the particle. In particular, it is necessary to transform Eq. (10) to

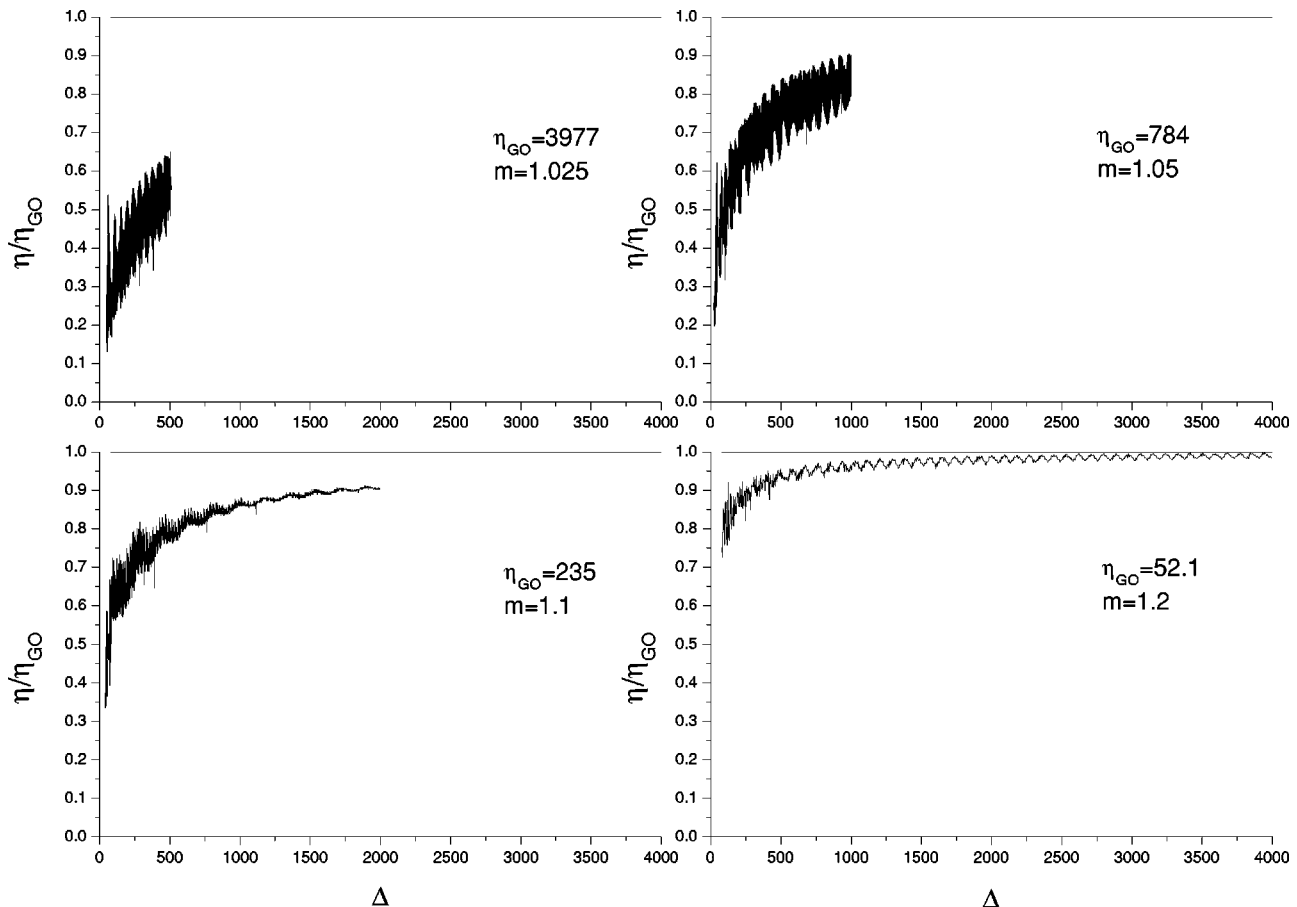


Fig. 2 The asymmetry coefficient vs the phase shift parameter Δ . Calculations have been performed with the Mie theory. Straight lines are the GO asymptotes.

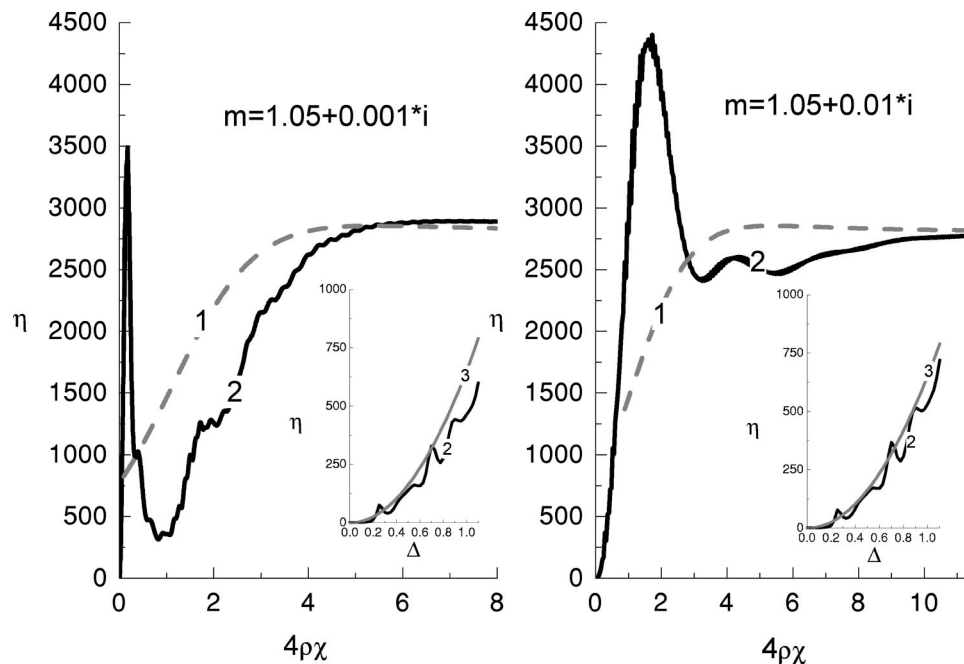


Fig. 3 The asymmetry coefficient vs the particle optical thickness. Calculations have been performed for the absorbing optically soft particles with the different relative refractive index. 1-GO, 2-Mie theory, 3-RGD Eq. (7).

$$\epsilon_{k,j} = \exp(-4[k-1]\rho\chi \cos \psi)(1-R_j)^2 R_j^{k-2}, \quad j=1,2 \quad (13)$$

and instead of 2 in the numerator of Eq. (12) to use expression $1 + \sigma$, where σ is the value of the total flux scattered according to the laws of the geometric optics

$$\sigma = \frac{1}{2} \sum_{j=1}^2 \int_0^{90} \left(R_j + \frac{(1-R_j)^2 \exp(-4\chi \rho \cos \psi)}{1-R_j \exp(-4\chi \rho \cos \psi)} \right) \times \sin 2\varphi d\varphi.$$

Figure 3 shows the value of η as functions of the optical thickness of the particle τ ($\tau = 4\rho\chi$) for the two values of the index of refraction with the identical real and different imaginary parts of m . The calculation with the GO mechanism was carried out for the first ten components.

It is obvious that the value of η reaches the stationary value rather fast; the stationary value corresponds to the contribution of the first component of GO. It is as follows:

$$\eta_{GO} = \frac{1}{F_1}. \quad (14)$$

The abovementioned value of η is achieved at $\tau = 4-8$. It can be accounted for by the decreasing influence of the fluxes that have passed through the particle. At the large values of τ their total contribution becomes negligible. In this case the components diffracted and reflected from the surface of the particle dominate.

Data for the RGD region are also presented in Figure 3. As seen under the condition $\Delta < 1$, the value of η can be described with Eq. (7).

In Figure 4 the dependence of the GO asymptote (η_{GO}) for the different values of m is presented (for the both nonabsorbing and absorbing particles). The asymptote for the nonab-

sorbing particles, for example, has the breaks in the points $m = 1.02, 1.04$. These are determined by involvement in the backward hemisphere of scattering of the fourth, fifth, etc., fluxes, respectively. The change in a functional dependence of the asymmetry coefficient from $\sim |m-1|^{-2}$ ($m < 1.003$) to $\sim |m-1|^{-1}$ ($m > 1.12$) through these nonmonotone zones occurs.

In the case of the nonabsorbing particles with the small value of χ and $m < 1.2$ within the whole range of the variation of m , $\eta_{GO} \sim |m-1|^{-2}$. It is easy to find the analytical solution of the abovementioned dependence for these particles.

Under condition $m \rightarrow 1$ the first component of GO determines the integral flux reflected in the backward hemisphere. It gives

$$\eta_{GO} \cong \frac{2}{|m-1|^2(1-\ln 2)}. \quad (15)$$

Equation (15) can be used with the error less than 14% for $m < 1.1$ and decreasing with decrease of m . Let us mark also a rather interesting result for the case of the nonabsorbing particles. With the decrease of m (from $m < 1.04$) the flux with one interior reflection (the third component) actually coincides with the reflected one (Figure 1). Thus, the total flux reflected from the exterior and the interior surface of the particle in the backward hemisphere is

$$F_1 + F_3 = (m-1)^2(1-\ln 2). \quad (16)$$

The RGD approximation gives the same result.

5 Conclusion

In conclusion, the asymmetry coefficient is significantly dependent on the redistribution of the energy among the k fluxes. Specifically, for the estimation of the asymmetry coef-

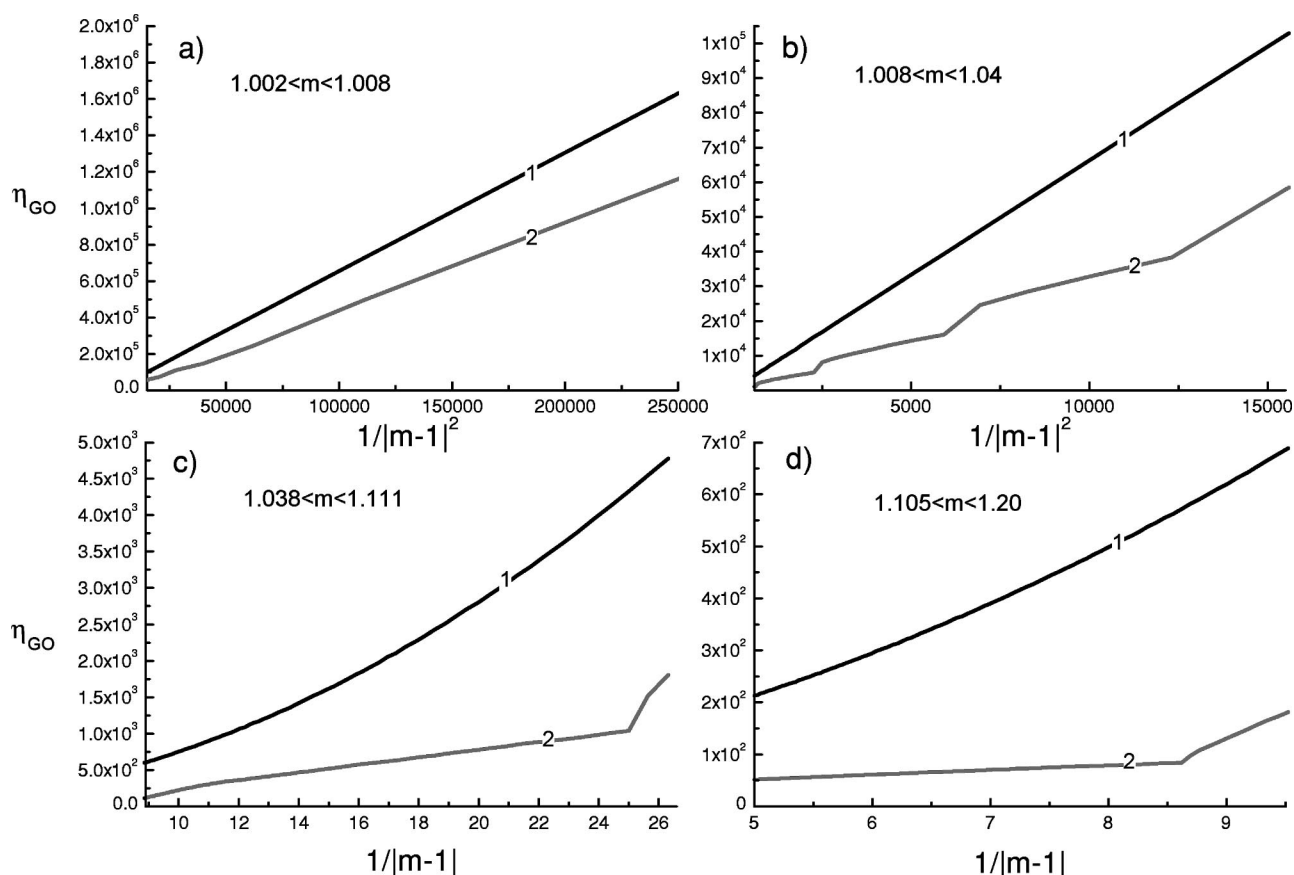


Fig. 4 η_{GO} vs $|\mu - 1|^{-2}$ (a,b) and $|m - 1|^{-1}$ (c,d) for the absorbing (1) and nonabsorbing (2) optically soft spherical particles.

cient of scattering (η) of the nonabsorbing optically soft particles ($m < 1.12$) a significant number of such fluxes must be taken into account. For such particles the approach to the GO asymptote of η occurs at very large values of Δ . At the same time for the particles with absorption, the asymptotic value of η is achieved much faster.

Obtained in the work asymptotic relations for η [Eqs. (7), (14), (15)] are perspective for the solution of the inverse light-scattering problem in the case of the optically soft homogeneous spheres.

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