ПРОБЛЕМЫ АСТРОНОМИИ

**КИНЕМАТИКА
И ФИЗИКА
НЕБЕСНЫХ
ТЕЛ том 26 № 3 2010**

UDC 523 + 523.4

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The amplitude of the coherent backscattering intensity peak for discrete random media: effect of packing density

The amplitude of the coherent backscattering intensity peak is computed for a medium composed of densely packed, randomly positioned particles. The cyclical component of the Stokes reflection matrix at exactly the backscattering direction is expressed in terms of the ladder component, and the ladder component is rigorously computed by numerically solving the vector radiative transfer equation. The effect of packing density is accounted for by multiplying the single-scattering Mueller matrix by the static structure factor computed in the Percus — Yevick approximation. It is shown that increasing packing density can substantially reduce the amplitude of the copolarized coherent backscattering peak, especially for smaller particles, and can make it significantly lower than 2. The effect of packing density on the amplitude of the cross-polarized peak is significantly weaker.

АМПЛІТУДА ЗВОРОТНОГО КОГЕРЕНТНОГО ПІКУ ІНТЕНСИВНОСТІ ДЛЯ ВИПАДКОВИХ ДИСКРЕТНИХ СЕРЕДОВИЩ: ЕФЕКТ ЩІЛЬНОЇ УПАКОВКИ, Міщенко М. І. — Розраховано амплітуду зворотного когерентного піку інтенсивності для середовища, що складається із щільно упакованих випадково розташованих частинок. Циклічний компонент матриці відбиття у поданні Стокса у напрямку точно назад виражено через драбинний компонент. Останній розраховується шляхом числового розв'язку векторного рівняння переносу. Ефект щільної упаковки враховано шляхом множення матриці Мюллера однократного розсіяння на статичний структурний фактор, розрахований у наближенні Перкуса — Євіка. Показано, що *збільшення щільності упаковки може значно послабити паралельно поляризований когерентний пік зворотного розсіяння та зменшити його амплітуду до значень, значно менших від 2. Вплив щільної упаковки на амплітуду поперечно поляризованого піку виявився набагато слабкішим.*

АМПЛИТУДА ОБРАТНОГО КОГЕРЕНТНОГО ПИКА В ИНТЕНСИВНОСТИ ДЛЯ СЛУЧАЙНЫХ ДИСКРЕТНЫХ СРЕД: ЭФФЕКТ ПЛОТНОЙ УПАКОВКИ, Мищенко М. И. — Рассчитывается амплитуда обратного когерентного пика в интенсивности для среды, состоящей из плотно упакованных случайно расположенных частиц. Циклический компонент матрицы отражения в представлении Стокса в направлении точно назад выражен через лестничный компонент. Последний рассчитывается путем численного решения векторного уравнения переноса. Эффект плотной упаковки учитывается путем умножения матрицы Мюллера однократного рассеяния на статический структурный фактор, рассчитанный в приближении Перкуса—Йевика. Показано, что увеличение плотности упаковки может значительно ослабить параллельно поляризованный когерентный пик обратного рассеяния и уменьшить его амплитуду до значений, значительно меньших 2. Влияние плотной упаковки на амплитуду поперечно поляризованного пика оказывается намного более слабым.

INTRODUCTION

Coherent backscattering of light by discrete random media has been intensively investigated during the last two decades both experimentally and theoretically [2, 13, 16, 22, 26]. Moreover, it has been shown that this phenomenon can be observed not only in laboratory conditions but also in nature in the form of the photometric and polarization opposition effects [1, 10, 14, 17—20]. The primary theoretical tool for computing the backscattering intensity peak has been the diffusion approximation [2]. However, although the diffusion approximation rather accurately predicts the angular profile of the backscattering peak, it cannot be used to compute the amplitude of the peak, i.e., the ratio of the intensity at the center of the peak to the incoherent background intensity. An additional complexifying factor is that accurate computations of the amplitude of the backscattering peak must explicitly take into account the vector nature of light since polarization effects have been shown to be extremely important in coherent backscattering [8, 15, 16].

Reflection of polarized light by a discrete random medium can be fully described by a 4 4 Stokes reflection matrix. In [8], Saxon’s reciprocity principle [21] was used to derive a rigorous relationship between the cyclical and ladder components of the reflection matrix at exactly the backscattering direction. Two factors make this relationship very useful.

First, in the derivation of this relationship the vector nature of light has been fully taken into account. Second, the ladder component of the reflection matrix can be rigorously computed by solving the vector radiative transfer equation (VRTE) with one of the well established numerical techniques [7, 16]. Both spherical and nonspherical scattering particles can be treated [16]. Therefore, this relationship can be used to compute the cyclical component of the reflection matrix in the center of the backscattering peak and, thus, the amplitude of the peak. This approach has been used in [5, 6, 9, 12, 16] to extensively study the properties of the coherent backscattering effect for different representations of polarization.

Numerical solutions of the VRTE used in [5, 6, 9, 12, 16] imply sparsely distributed, "independently scattering" particles and hence are not necessarily applicable to densely packed media, for example owing to correlations among particle positions. Therefore, it is the aim of this paper to extend the approach developed in [8] to media with nonzero packing density and to examine the effect of packing density on the amplitude of the coherent backscattering peak. Unlike the rigorous and sophisticated approach pursued in [23—25], we will use a rather simple and inherently approximate approach based on the so-called structure factor formalism.

THEORY AND COMPUTATIONS

Let the discrete scattering medium be a homogeneous semi-infinite slab composed of sparsely and randomly distributed particles. The slab is illuminated by a quasi-monochromatic parallel beam of light of infinite lateral extent incident in the direction of the unit vector $\hat{\mathbf{n}}_0 = \{ \ _0 \quad /2, \ \ _0\}$, where $\ _0$ is the corresponding zenith angle measured from the positive direction of the z axis and $\ _0$ is the corresponding azimuth angle measured from the positive direction of the x axis in the clock-wise sense when looking in the positive direction of the z axis (Fig. 1). In what follows, we will assume for simplicity that $\ _0 = 0.$ The Stokes column vector has four Stokes parameters as its components: $\mathbf{I} \quad [I\ Q\ U\ V]^T$ where T stands for "transposed". Let $\mathbf{R}_b$ be the 4 4 Stokes reflection matrix for exactly the backscattering direction $\hat{\mathbf{n}}_b = \{ \ = \ - \ _0, \ \ _0 = \ \}$. This matrix yields the specific Stokes column vector of the backscattered light as follows:

$$\tilde{\mathbf{I}}_b = \frac{1}{\ } \ {}_0 \mathbf{R}_b \mathbf{I}_0, \tag{1}$$

where $\ _0 \quad \cos \ _0$ and $\mathbf{I}_0$ is the Stokes column vector of the incident beam [16].

Under the simplifying assumption of a macroscopically isotropic and mirror-symmetric particulate medium, the backscattering matrix $\mathbf{R}_b$ has the following block-diagonal structure [7, 16]:

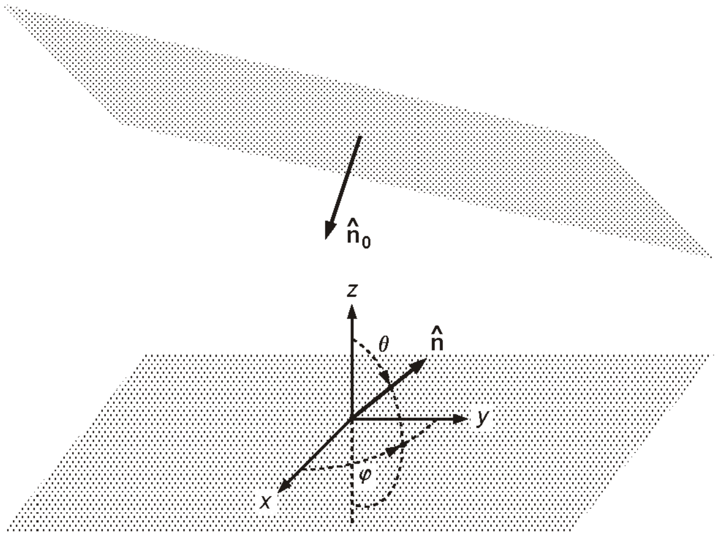


Fig. 1. Right-handed laboratory coordinate system with origin at the upper boundary of a semi-infinite particulate slab. The direction of light propagation is specified by a unit vector $\hat{\mathbf{n}}$ = { , }. The slab is illuminated by a quasi-monochromatic parallel beam of light incident from above in the direction of the unit vector $\hat{\mathbf{n}}_0$

$$\mathbf{R}_b = \begin{matrix} R_{b11} & R_{b12} & 0 & 0 \\ R_{b12} & R_{b22} & 0 & 0 \\ 0 & 0 & R_{b33} & R_{b34} \\ 0 & 0 & R_{b34} & R_{b44} \end{matrix} . \tag{2}$$

In accordance with the microphysical theory of coherent backscattering by sparse discrete random media [13, 16], the backscattering Stokes matrix $\mathbf{R}_b$ can be decomposed as follows:

$$\mathbf{R}_b \quad \mathbf{R}_b^1 \quad \mathbf{R}_b^M \quad \mathbf{R}_b^C, \tag{3}$$

where $\mathbf{R}_b^1$ is the contribution of the first-order scattering, $\mathbf{R}_b^M$ is the diffuse component consisting of all the ladder terms of scattering orders ($n \quad 2$), and $\mathbf{R}_b^C$ is the cumulative contribution of all the cyclical terms. The matrices $\mathbf{R}_b^1$ and $\mathbf{R}_b^M$ can be found by numerically solving the VRTE [16]. Then the matrix $\mathbf{R}_b^C$ can be determined from the following exact relation derived in [8, 16]:

$$\mathbf{R}_b^C = \begin{matrix} R_{b11}^C & R_{b12}^M & 0 & 0 \\ R_{b12}^M & R_{b22}^C & 0 & 0 \\ 0 & 0 & R_{b33}^C & R_{b34}^M \\ 0 & 0 & R_{b34}^M & R_{b44}^C \end{matrix} , \tag{4}$$

where

$$R_{b11}^{C} \quad 0.5(R_{b11}^{M} \quad R_{b22}^{M} \quad R_{b33}^{M} \quad R_{b44}^{M}), \tag{5}$$

$$R_{b22}^{C} \quad 0.5(R_{b11}^{M} \quad R_{b22}^{M} \quad R_{b33}^{M} \quad R_{b44}^{M}), \tag{6}$$

$$R_{b33}^{C} \quad 0.5(\quad R_{b11}^{M} \quad R_{b22}^{M} \quad R_{b33}^{M} \quad R_{b44}^{M}), \tag{7}$$

$$R_{b44}^{C} \quad 0.5(R_{b11}^{M} \quad R_{b22}^{M} \quad R_{b33}^{M} \quad R_{b44}^{M}). \tag{8}$$

Once all the components of the backscattering Stokes matrix are computed, they can be used to calculate the amplitude of the coherent backscattering peak for different states of polarization of the incident and scattered beams [8, 16]. Specifically, if the incident light is fully linearly polarized in the vertical direction, then the amplitudes of the copolarized and cross-polarized peaks are expressed in terms of the backscattering Stokes matrix as

$$_{vv} = \frac{R_{b11}^{1} \quad R_{b22}^{1} \quad 2R_{b11}^{M} \quad 4R_{b12}^{M} \quad 2R_{b22}^{M}}{R_{b11}^{1} \quad R_{b22}^{1} \quad R_{b11}^{M} \quad 2R_{b12}^{M} \quad R_{b22}^{M}}, \tag{9}$$

$$_{hv} = \frac{R_{b11}^{1} \quad R_{b22}^{1} \quad R_{b11}^{M} \quad R_{b22}^{M} \quad R_{b33}^{M} \quad R_{b44}^{M}}{R_{b11}^{1} \quad R_{b22}^{1} \quad R_{b11}^{M} \quad R_{b22}^{M}}, \tag{10}$$

respectively. In the case of fully circularly polarized incident beam, we have for the amplitudes of the helicity-preserving and opposite-helicity peaks:

$$_{hp} \quad \frac{R_{b11}^{1} \quad R_{b44}^{1} \quad 2R_{b11}^{M} \quad 2R_{b44}^{M}}{R_{b11}^{1} \quad R_{b44}^{1} \quad R_{b11}^{M} \quad R_{b44}^{M}}, \tag{11}$$

$$_{oh} \quad \frac{R_{b11}^{1} \quad R_{b44}^{1} \quad R_{b11}^{M} \quad R_{b22}^{M} \quad R_{b33}^{M} \quad R_{b44}^{M}}{R_{b11}^{1} \quad R_{b44}^{1} \quad R_{b11}^{M} \quad R_{b44}^{M}}. \tag{12}$$

For spherical particles $R_{b22}^{1} \quad R_{b11}^{1}$ and $R_{b44}^{1} \quad R_{b11}^{1}$, which implies that

$$_{vv} \quad 2 \quad \frac{2R_{b11}^{1}}{2R_{b11}^{1} \quad R_{b11}^{M} \quad 2R_{b12}^{M} \quad R_{b22}^{M}}, \tag{13}$$

$$_{hv} \quad 1 \quad \frac{R_{b33}^{M} \quad R_{b44}^{M}}{R_{b11}^{M} \quad R_{b22}^{M}}, \tag{14}$$

$$_{hp} \quad 2, \tag{15}$$

$$_{oh} \quad 1 \quad \frac{R_{b22}^{M} \quad R_{b33}^{M}}{2R_{b11}^{1} \quad R_{b11}^{M} \quad R_{b44}^{M}}. \tag{16}$$

To solve numerically the VRTE, one needs to know single-scattering characteristics of particles comprising the medium. The single scattering of polarized light by a particle is described by the 4 4 Stokes scattering matrix **F** [16]. For spherical particles, the elements of the scattering matrix depend on the particle refractive index and size parameter x (defined as $x = 2 \quad r / \quad$, where r is particle radius and λ is the wavelength in the surrounding medium) as well as on the scattering angle Θ (i.e., the angle between the incidence and scattering directions). The (1, 1) element of the scattering matrix is called the differential scattering cross section and is often denoted as $dC_{sca}/d\Omega$. The scattering cross section C_{sca} is obtained by integrating the differential scattering cross section over all scattering directions:

$$C_{sca} = \int_{4\pi} d\Omega \, \frac{dC_{sca}}{d\Omega}. \tag{17}$$

The quantity

$$p(\Theta) = \frac{4\pi}{C_{sca}} \frac{dC_{sca}}{d\Omega} \tag{18}$$

is called the phase function and satisfies the normalization condition

$$\frac{1}{4\pi} \int_{4\pi} d\Omega \, p(\Theta) = 1. \tag{19}$$

Finally, the asymmetry parameter (or the mean cosine of the scattering angle) is defined as

$$\langle \cos\Theta \rangle = \frac{1}{4\pi} \int_{4\pi} d\Omega \, p(\Theta) \cos\Theta. \tag{20}$$

The standard techniques for computing the single-scattering matrix (e.g., the conventional Lorenz — Mie theory for spherical scatterers and the T-matrix method for nonspherical particles [16]) for discrete random media imply that particles are widely separated and their positions are independent of each other [13, 16]. However, in densely packed media both conditions are violated. In particular, spatial correlations between particles may become important. As has been suggested in [29], in the first approximation the corresponding modification of the single-scattering matrix can be accounted for via multiplying **F** by the so-called static structure factor S which depends on the filling factor f (i.e., the fraction of the scattering volume occupied by the particles) and the product

$$u = 4x \sin(\Theta/2). \tag{21}$$

The computation of the static structure factor using the Percus—Yevick approximation for hard, impenetrable, monodisperse spheres is described, e, g., in [11]. Figure 2 shows the structure factor computed for several values of the filling factor. It is seen that increasing packing density results in an angular redistribution of the scattered intensity which is especially pronounced at $u < 10$.

As follows from Eqs. (13) and (16), the copolarized and opposite-helicity amplitudes depend explicitly on the (1, 1) element of the single-scattering component of the Stokes backscattering matrix $\mathbf{R}^1_{b11}$. If this component is neglected, which is the case in the framework of the diffusion

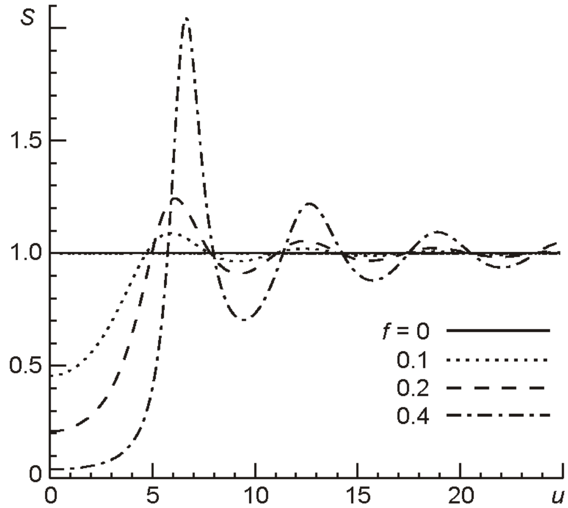


Fig. 2. Static structure factor S for different values of the filling factor f

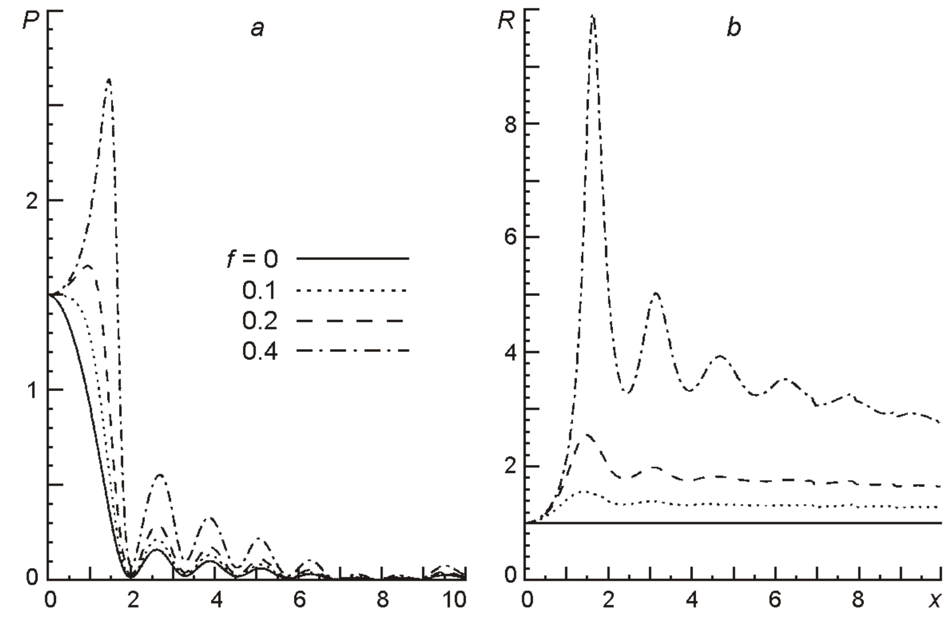
approximation, the copolarized amplitude becomes equal to 2. Therefore, it is the deviation of R^{1}_{b11} from zero that makes the copolarized amplitude smaller than 2. The single-scattering component of the Stokes backscattering matrix is in turn proportional to the backscattering phase function $p(180°)$. Specifically, in the case of a semi-infinite medium [7, 16],

$$R^{1}_{b11} \qquad \frac{p(180\ \ )}{8}, \tag{22}$$

where is the single-scattering albedo (for nonabsorbing media considered below, = 1).

Figure 3, *a* shows the backscattering phase function $P = p(180°)$ vs. size parameter x computed for monodisperse spherical latex particles in water (relative refractive index 1.195) for several values of the filling factor ranging from 0 to 0.4. In addition, Fig. 3, *b* shows the ratio R of $p(180°)$ for densely packed particles ($f > 0$) to that for widely separated particles ($f = 0$). It is seen that the effect of particle correlations can be strong and is especially pronounced for particles with size parameters smaller than about 2, and that increasing packing density always increases $p(180°)$. Note, however, that the phase function for Rayleigh particles (x 1) remains intact with increasing packing density. Interestingly, $p(180°)$ is an oscillating rather than a monotonically decreasing function of size parameter, and, thus, one should not expect a monotonic dependence of the copolarized amplitude $_{vv}$ on x [see Eq. (13)]. For comparison, Fig. 3, *c* shows the asymmetry parameter vs. size parameter for the same values of the filling factor. One sees that for filling factors not exceeding 0.4 and size parameters larger than about 1.5, cos is a monotonically increasing function of size parameter. Therefore, we have to conclude that there is no direct correlation between the asymmetry parameter on one hand and the backscattering phase function (and, thus, the copolarized amplitude) on the other hand.

Figures 4, *a—c* show the copolarized, $_{vv}$, cross-polarized, $_{hv}$, and opposite-helicity, $_{oh}$, amplitudes vs. size parameter computed for a semi-infi-



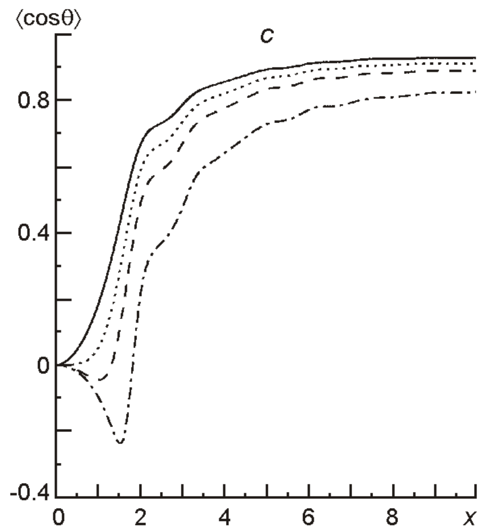


Fig. 3. Backscattering phase function $P = p(180°)$, the ratio R of the backscattering phase function for densely packed particles ($f > 0$) to that of sparsely distributed particles ($f = 0$), and the asymmetry parameter cos vs. size parameter for monodisperse spherical particles with refractive index 1.195 and different values of the filling factor f

nite scattering medium which is composed of monodisperse spherical particles with the index of refraction 1.195 and is illuminated perpendicularly to its boundary. For both sparsely and densely packed particles, the VRTE was rigorously solved using the technique described in [3, 16]. For densely packed particles, the single-scattering characteristics were modified by means of the static structure factor.

It is clearly seen from Figs 4, *a*—*c* that increasing packing density affects all the amplitudes. The effect is especially strong for small size parameters and is much more noticeable for $_{vv}$ and, to a lesser degree, for $_{oh}$ because these amplitudes depend on the single-scattering component R^1_{b11} explicitly [Eqs. (13) and (16)].

Not surprisingly, local maxima of $_{vv}$ (Fig. 4, *a*) exactly follow local minima of the backscattering phase function $p(180°)$ (Fig. 3, *a*). Since in-

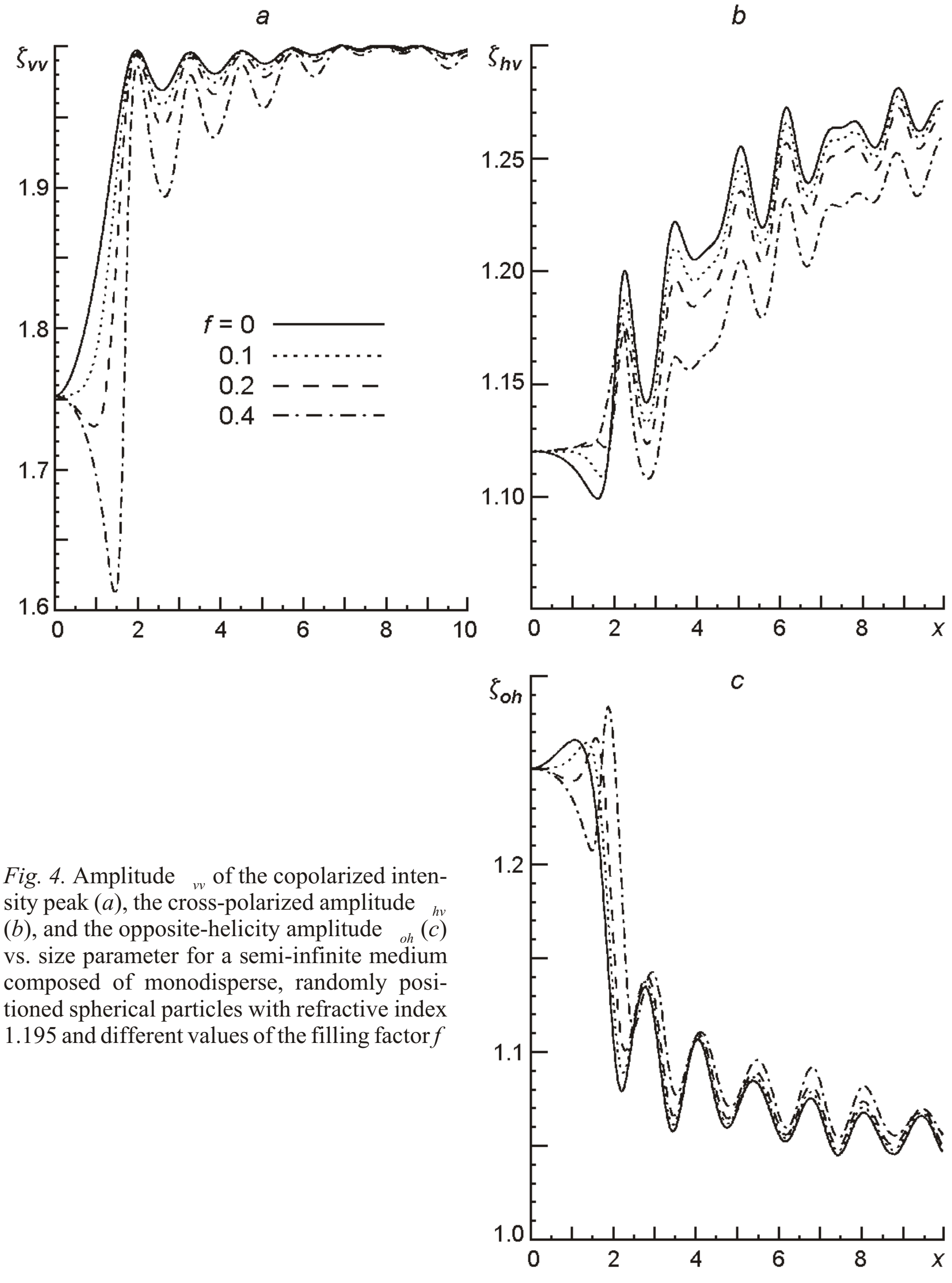


Fig. 4. Amplitude $_{vv}$ of the copolarized intensity peak (*a*), the cross-polarized amplitude $_{hv}$ (*b*), and the opposite-helicity amplitude $_{oh}$ (*c*) vs. size parameter for a semi-infinite medium composed of monodisperse, randomly positioned spherical particles with refractive index 1.195 and different values of the filling factor *f*

creasing packing density always increases the backscattering phase function, it always reduces the copolarized amplitude and, for small particles, can even make it lower than the Rayleigh-limit value of 1.752 [16]. On the other hand, the size-parameter dependences of the amplitudes $_{hv}$ and $_{oh}$ do not quite follow that of R^1_{b11}. Apparently, this can be explained by the implicit dependence of the ratio $_{hv}$ on the single-scattering matrix and by a complicated size parameter dependence of the diagonal elements of the ladder component $\mathbf{R}_b^M$ [see Eqs. (14) and (16)].

DISCUSSION

The standard theories of radiative transfer and coherent backscattering are based on the assumption that particles forming a discrete random medium are widely separated and totally uncorrelated [13, 16]. The approach pursued in this paper can only be considered to be a rather trivial approximate "patch" intended to take into account the effect of spatial correlations for closely spaced particles. It goes without saying that it cannot replace a rigorous theory directly based on the Maxwell equations. In fact, it will be very interesting to compare our approximate results with exact computations when the latter have become available.

As mentioned above, the diffusion approximation predicts the amplitude of the copolarized backscattering peak exactly equal to 2 [2]. However, some experimentalists have claimed that they found evidence for an amplitude smaller than two. As a result, a discussion as to the actual value of the copolarized amplitude has taken place (see, e.g., [4] and references therein). In order to partially explain the discrepancy between the prediction of the diffusion approximation and laboratory experiments, the effect of the backscattering contribution from nonself-avoiding closed light paths, in addition to the ladder and cyclical contributions, was studied [27, 28].

As follows from Eq. (13), the amplitude of the copolarized peak is never equal to 2 since, for real scattering particles, R^1_{b11} is never exactly equal to zero but rather is a positive number. Moreover, it was shown in [16] that $_{vv}$ is always smaller than 2. The degree of deviation of the copolarized amplitude from the value 2 depends primarily on the value of the backscattering phase function. For latex particles in water $p(180°)$ is usually small (except for particles with size parameters less than about 1) because the corresponding relative refractive index (1.195) is small. However, for larger refractive indices $p(180°)$ can be much larger, thereby causing copolarized amplitude values significantly smaller than 2 [9, 12].

As follows from the calculations reported in this paper, spatial correlations among scattering particles caused by nonzero packing density can be an additional important factor which can substantially increase the backscattering phase function and, thus, reduce the amplitude of the copolarized backscattering peak. The effect is especially pronounced for small particles (but not for Rayleigh scatterers) and weakens as the particle size parameter becomes much greater than 1. On the other hand, the amplitude of the helicity-preserving amplitude $_{hp}$ for spherical particles is not influenced at all by increasing packing density and is identically equal to 2 independently of the filling factor. Apparently, this can explain, at least qualitatively, why in the measurements for latex particles in water reported in [4] the amplitude of the helicity-preserving backscattering peak was always equal to 2 (within the measurement accuracy), while the amplitude of the copolarized peak was substantially smaller than 2, especially for small particles.

Finally we note that for spherical particles the equalities $R^1_{b22} = R^1_{b11}$ and $R^1_{b44} = \quad R^1_{b11}$ do not, in general, hold. Therefore, the helicity-preserving amplitude $_{hp}$ can explicitly depend on the single-scattering component of the Stokes backscattering matrix $\mathbf{R}^1_b$ [Eq. (11)] and, thus, can be appreciably smaller than 2 [5, 6, 16] and change with increasing packing density. Therefore, unlike the case with spherical particles, the calculation of the helicity-preserving amplitude for nonspherical particles should explicitly take into account the effect of packing density.

The author is grateful to P. V. Litvinov, Yu. G. Shkuratov, V. P. Tishkovets, and E. G. Yanovitskij for many fruitful discussions. Partial funding for this research was provided by the NASA Radiation Sciences Program managed by H. Maring.

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Received March 26, 2009