Depolarization of nearly spherical particles: The Debye series approach

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Backscattering depolarization of nonspherical particles plays a critical role in active Light Detection And Ranging (LiDAR) retrievals of cloud or aerosol parameters, as well as in particle characterization techniques. However, the interpretation of backscattering light from particles is a challenging research subject. This paper addresses the depolarization of nearly spherical particles by using the Debye series approach. Specifically, the T matrix is represented as an infinite sum of terms; the terms in the expansion are correspondingly associated with diffraction and reflection ($p = 0$), and multiple transmissions ($p > 0$) from the particle to the medium as waves undergo internal reflections. We found that the enhanced depolarization for optically soft particles stems from multiple transmissions. However, this is mostly from the transmission after one internal reflection ($p = 2$), when the refractive index is larger than 1.3. Moreover, the interference among multiple transmissions was found to play an essential role in suppressing the depolarization ratio as the refractive index approaches unity. These findings have implications in interpreting the backscattering optical properties of atmospheric aerosols and hydrosols in water.

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I. INTRODUCTION

The polarized LiDAR can obtain fruitful microphysical information about atmospheric aerosols and hydrometeors. As a critical quantity, the linear depolarization ratio (LDR) is most frequently used to discriminate cloud phase or aerosol types [1–3]. Thus, extensive theoretical studies, laboratory measurements, and field campaigns have been carried out to understand the depolarization capabilities of various atmospheric particles [4]. In addition to the application of atmospheric detection and retrievals of cloud or aerosol parameters, the LDR (near backscattering, in particular) can be also used in particle characterization techniques to obtain the microphysical information of particle systems in laboratory [5]. However, there is no simple relation between the LDRs and the particle shape and refractive index. For example, the LDR peaking at aspect ratios of spheroids very close to unity was discovered by Mishchenko and Hovenier [6] and illustrates the inherent difficulty of treating the LDR as an indicator of the degree of particle nonsphericity.

Owing to the advances on the invariant imbedding T-matrix method (II-TM) [7–10], recently, depolarization capabilities of nonspherical particles have been extensively assessed in a superellipsoidal space and with a large range of refractive indices [11]. It has been shown that the enhanced LDRs are relatively common for optically soft particles (a range of refractive index from 1.05 to 1.20). Also, the large LDRs exist for nearly spherical particles in a refractive regime with the real part ranging from 1.3 to 1.7. However, the physical mechanism leading to the enhanced LDRs is unclear. As an example, Fig. 1 shows the LDRs of prolate spheroids (top panel) and oblate spheroids (bottom panel) as functions of the size parameter and refractive index. It is evident that the pronounced LDRs locate at two ranges of refractive indices, namely, (1.05, 1.2) and (1.3, 1.6).

It is not straightforward to understand the scattering mechanism leading to the phenomenon above, because the T-matrix solution contains all the effects that contribute to the scattering. This paper is indented to provide an in-depth analysis of the depolarization in the framework of Debye's series. The Debye series was first proposed by Debye for an infinite circular cylinder [12], and then has been extensively used for spherical particles [13–16], coated spherical particles [17–23], and coated cylinders [24], and further extended to spheroidal particles based on the separation of the variable method [25,26]. The recent development of Debye's approach for nonspherical particles with the extended boundary condition method (EBCM) [27,28] is a breakthrough that makes it possible to perform accurate analyzes of the scattering mechanism by nonspherical particles. The concept of Debye's series is similar to the geometric optics (GO) approach. In the GO approach, the incident beam consists of a bundle of rays. When a geometric ray is incident upon a particle, it will be partially reflected, and partially refracted into the particle, which could be absorbed into the particle or undergo an arbitrary number of internal reflections. In conjunction with each internal reflection, there will be transmitted rays from the particle to the medium, which contributes to the scattering. Different from the GO, the Debye approach is rooted rigorously in the framework of Maxwell's equations. Each term in the Debye series contains the information of the GO, but

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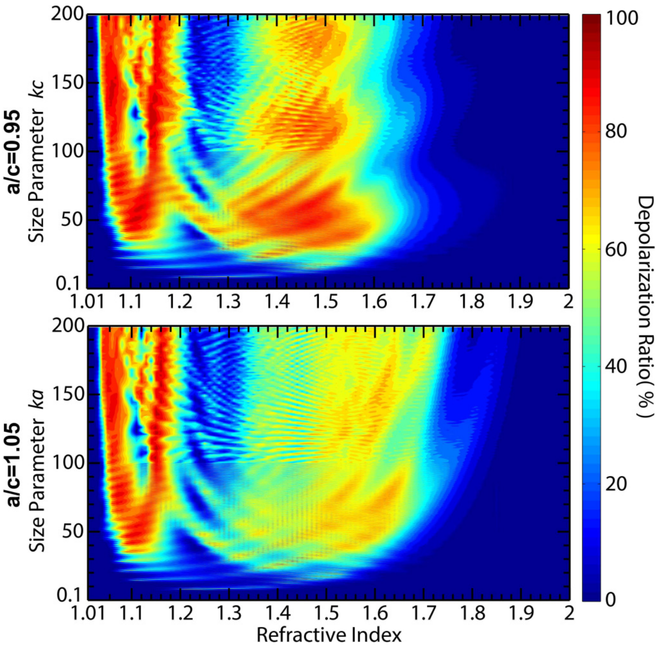

FIG. 1. LDRs for randomly oriented spheroids (aspect ratio is 0.95 and 1.05). The horizontal and vertical axes of a spheroid are denoted as a and c, respectively. $k = 2\pi/\lambda$, where λ is the wavelength of the incident light.

also includes semiclassical effects that cannot be interpreted in the GO [29–31]. According to the conventional notations for a sphere, the zeroth-order term ($p = 0$) corresponds to diffraction and reflection by particle surfaces (note that the diffraction and external reflection are essentially bundled together, but can be separated from each other approximately based on the GO and physical-geometric optics approaches), and the pth term indicates the contribution to scattering from transmitted waves after making $p - 1$ internal reflections.

In this paper, we explore the powerfulness of the Debye-series-based T-matrix method and, in particular, interpret the depolarization of incident polarized electromagnetic waves by nearly nonspherical particles, assumed to be spheroids with an aspect ratio close to unity. To analyze depolarization, an explicit formalism is developed in this paper by expanding the T matrix in terms of Debye's series. The EBCM is used to compute reflection and transmission matrices in Debye's series (see details in Sec. II). Thus, the phase matrices associated with each order of Debye's series for either oriented particles or randomly oriented particles could be accurately obtained. Specifically, the contributions from waves associated with diffraction, external reflection, and transmission (with and without interference) can be cleary identified. This paper is organized as follows. In Sec. II, we briefly summarize the theoretical formalism that represents the T matrix as a series. Representative results with discussions are given in Sec. III. Section IV is the summary and conclusion of this paper.

II. THEORETICAL FORMALISM

Here, we outline a formalism that is developed to assess the underlying mechanism of backscattering. In the framework of the T matrix, the incident field and the scattered field are expanded in terms of vector spherical wave functions [32]:

$$\mathbf{E}^{\mathrm{inc}}(\mathbf{r}) = \sum_{l=1}^{\infty} a_l \mathrm{Rg}\mathbf{M}_l(k\mathbf{r}) + b_l \mathrm{Rg}\mathbf{N}_l(k\mathbf{r}), \tag{1}$$

$$\mathbf{E}^{\mathrm{sca}}(\mathbf{r}) = \sum_{l=1}^{\infty} p_l \mathbf{M}_l(k\mathbf{r}) + q_l \mathbf{N}_l(k\mathbf{r}), \tag{2}$$

where k is the wave vector in the medium, $\mathrm{Rg}\mathbf{M}_l$ and $\mathrm{Rg}\mathbf{N}_l$ are the regular vector spherical functions, and $\mathbf{M}_l$ and $\mathbf{N}_l$ are the irregular vector spherical functions. For simplicity, two subindices of vector spherical functions are combined as one index via $l = n(n+1) + m$, where n is the total angular momentum and m is the projected angular momentum. The T matrix $\mathbf{T}$ is defined as a transition matrix that transfers the coefficients of the incident field to those of the scattered field. Explicitly, we have the following equation:

$$\begin{bmatrix} p_1 \\ q_1 \\ \vdots \\ p_{l_{\max}} \\ q_{l_{\max}} \end{bmatrix} = \begin{bmatrix} \mathrm{T}_{11}^{11} & \mathrm{T}_{11}^{12} & \cdots & \mathrm{T}_{1l_{\max}}^{11} & \mathrm{T}_{1l_{\max}}^{12} \\ \mathrm{T}_{11}^{21} & \mathrm{T}_{11}^{22} & \cdots & \mathrm{T}_{1l_{\max}}^{21} & \mathrm{T}_{1l_{\max}}^{22} \\ \vdots & \vdots & \cdots & \vdots & \vdots \\ \mathrm{T}_{l_{\max}1}^{11} & \mathrm{T}_{l_{\max}1}^{12} & \cdots & \mathrm{T}_{l_{\max}l_{\max}}^{11} & \mathrm{T}_{l_{\max}l_{\max}}^{12} \\ \mathrm{T}_{l_{\max}1}^{21} & \mathrm{T}_{l_{\max}1}^{22} & \cdots & \mathrm{T}_{l_{\max}l_{\max}}^{21} & \mathrm{T}_{l_{\max}l_{\max}}^{22} \end{bmatrix} \begin{bmatrix} a_1 \\ b_1 \\ \vdots \\ a_{l_{\max}} \\ b_{l_{\max}} \end{bmatrix}, \tag{3}$$

where $l_{\max}$ is the truncation number. Based on Debye's concept, the T matrix can be expanded in terms of a series:

$$\begin{aligned} \mathbf{T} &= -\frac{1}{2}\left[\mathbf{1} - \tilde{\mathbf{R}}_{11} - \tilde{\mathbf{T}}_{21}\left(\sum_{n=0}^{\infty}(\tilde{\mathbf{R}}_{22})^n\right)\tilde{\mathbf{T}}_{21}\right] \\ &= -\frac{1}{2}\left[\mathbf{1} - \tilde{\mathbf{R}}_{11} - \tilde{\mathbf{T}}_{12}\frac{1}{\mathbf{1} - \tilde{\mathbf{R}}_{22}}\tilde{\mathbf{T}}_{21}\right], \end{aligned} \tag{4}$$

where $\tilde{\mathbf{R}}_{11}$ is defined as a matrix that transforms the coefficient of the incident vector spherical wave field to those of the reflected field, $\tilde{\mathbf{T}}_{21}$ is defined as a matrix that transforms the coefficient of the incident vector spherical wave field to those of the refracted field, $\tilde{\mathbf{R}}_{22}$ is defined as a matrix that transforms the coefficient of the internal outgoing vector spherical wave field to those of the reflection field within the particle, and $\tilde{\mathbf{T}}_{12}$ is defined as a matrix that transforms the coefficient of the internal outgoing vector spherical wave field to those of the transmitted field exiting the particle. $\tilde{\mathbf{R}}_{11}$, $\tilde{\mathbf{T}}_{21}$, $\tilde{\mathbf{R}}_{22}$, and $\tilde{\mathbf{T}}_{12}$ are computed from the EBCM [27]. Note that the subindices (11, 12, 21, 22) in Eq. (4) are not intended to indicate the elements of matrices but symbolize the particle (as 2) and the medium (as 1). The first equality of Eq. (4) is obtained from physical concept involved in Debye's series. It can be directly validated for homogeneous spherical particles, which have analytical solutions. However, for nonspherical particles, it is not straightforward to prove Eq. (4), because analytical solutions do not exist. Instead, Eq. (4) has been numerically validated through the second equality to avoid truncation errors in summation; the left and right sides are computed from the II-TM and the EBCM, respectively. The agreement of phase matrices computed from both sides of Eq. (4) also indirectly verified the equation (shown in Sec. III).

To separate the contributions from diffraction, reflection, and higher-order transmissions, the following quantities are defined:

$$\mathbf{T}_0 = -\frac{1}{2}[\mathbf{1} - \tilde{\mathbf{R}}_{11}], \tag{5}$$

$$\mathbf{T}_1 = \frac{1}{2}\tilde{\mathbf{T}}_{12}\tilde{\mathbf{T}}_{21}, \tag{6}$$

$$\mathbf{T}_2 = \frac{1}{2}\tilde{\mathbf{T}}_{12}\tilde{\mathbf{R}}_{22}\tilde{\mathbf{T}}_{21}, \tag{7}$$

$$\mathbf{T}_N = \frac{1}{2}\tilde{\mathbf{T}}_{12}(\tilde{\mathbf{R}}_{22})^{N-1}\tilde{\mathbf{T}}_{21}, \quad N \geqslant 1, \tag{8}$$

$$\mathbf{T}_{p<=N} = \sum_{k=0}^{N}\mathbf{T}_k = -\frac{1}{2}\left[\mathbf{1} - \tilde{\mathbf{R}}_{11} - \tilde{\mathbf{T}}_{12}\left(\sum_{n=0}^{N-1}(\tilde{\mathbf{R}}_{22})^n\right)\tilde{\mathbf{T}}_{21}\right]. \tag{9}$$

The physical implications of all the above terms are clear. $\mathbf{T}_0$ contains all the information associated with diffraction and reflection. In particular, the edge effect associated with a tunneling process can be rigorously obtained, which has been studied in Bi *et al.* [33] and Lin *et al.* [34]. $\mathbf{T}_1$ contains all the information associated with the contribution to the scattered field from the waves that transmitted to the particle without internal reflection. $\mathbf{T}_N$ represents the contribution from transmitted waves after $N-1$ internal reflections. For simplicity, $\mathbf{T}_{p<=N}$ is defined to sum all the contributions up to the Nth order. For a polarized plane-wave incident field, the phase matrix can be derived from the T matrix. The phase matrix of randomly oriented particles (with mirror symmetry) that determines the change in the Stokes vector $[I_{\text{inc}}, Q_{\text{inc}}, U_{\text{inc}}, V_{\text{inc}}]^T$ of the incident wave to that of scattered waves $[I_{\text{sca}}, Q_{\text{sca}}, U_{\text{sca}}, V_{\text{sca}}]^T$ is given by [35]

$$\begin{bmatrix} I_{\text{sca}}(\theta) \\ Q_{\text{sca}}(\theta) \\ U_{\text{sca}}(\theta) \\ V_{\text{sca}}(\theta) \end{bmatrix} \propto \begin{bmatrix} P_{11}(\theta) & P_{12}(\theta) & 0 & 0 \\ P_{21}(\theta) & P_{22}(\theta) & 0 & 0 \\ 0 & 0 & P_{33}(\theta) & P_{34}(\theta) \\ 0 & 0 & P_{43}(\theta) & P_{44}(\theta) \end{bmatrix} \begin{bmatrix} I_{\text{inc}} \\ Q_{\text{inc}} \\ U_{\text{inc}} \\ V_{\text{inc}} \end{bmatrix}, \tag{10}$$

where θ is the scattering angle ranging from 0 to 180 deg, $P_{12} = P_{21}$, and $P_{43} = -P_{34}$. For a linearly polarized incident wave with the Stokes vector $[1\,1\,0\,0]^T$, the Stokes vector of the scattered wave is $[P_{11} + P_{12} P_{21} + P_{22}\,0\,0]^T$. Given the phase matrix, the LDR (defined as the intensity of the electric field perpendicular to the scattering plane $|E_\perp|^2$ compared to that parallel to the scattering plane $|E_\parallel|^2$) can be straightforwardly computed by

$$\text{LDR} = \frac{|E_\perp|^2}{|E_\parallel|^2} = \frac{I_{\text{sca}} - Q_{\text{sca}}}{I_{\text{sca}} + Q_{\text{sca}}} = \frac{P_{11} - P_{22}}{P_{11} + 2P_{12} + P_{22}} = \frac{1 - P_{22}/P_{11}}{1 + 2P_{12}/P_{11} + P_{22}/P_{11}}. \tag{11}$$

At direct backscattering, the LDR can be shown to be less than or equal to 100%; this is because P_{12} is zero, and $P_{22}/P_{11} \geqslant 0$ according to the reciprocity theorem [6]. However, for the side scattering, the LDR could be much larger than 100% (examples will be shown in Sec. III).

The concept and technical details of the T-matrix formulation, the II-TM, and the EBCM are not iterated here because they were thoroughly described in previous studies [7–10,27,32,36,37]. The present paper focuses on the optical properties of spheroids with aspect ratios close to unity, although other nearly spherical particles could also have enhanced backscattering depolarization.

Now we consider two computational schemes.

(1) Compute the phase matrix from $\mathbf{T}_{p<=N}$, and understand the convergence of the T matrix and contributions of each order of waves to the scattered field. Note that the interferences between the different orders of waves are taken into account.

(2) Compute the phase matrix from $\mathbf{T}_N$, and then sum all the phase matrices with weighting functions. The comparison of this phase matrix from the summations and the phase matrix computed from the T matrix can be used to assess the interference effect among the different orders of waves. To compute the final phase matrix, the weights, namely, the "scattering cross section" for each term, should be computed. The scattering cross sections can be employed to understand the relative contribution of each order of scattered waves. The pth order scattering cross section of a randomly oriented particle can be computed from the following formula:

$$C_{\text{sca}}^p = -\frac{2\pi}{k^2}\sum_{l=1}^{l_{\max}}\sum_{l'=1}^{l_{\max}}\left[\left|\mathrm{T}_{p,ll'}^{11}\right|^2 + \left|\mathrm{T}_{p,ll'}^{12}\right|^2 + \left|\mathrm{T}_{p,ll'}^{21}\right|^2 + \left|\mathrm{T}_{p,ll'}^{22}\right|^2\right], \tag{12}$$

where $\mathrm{T}_{p,ll'}^{11}$, $\mathrm{T}_{p,ll'}^{12}$, $\mathrm{T}_{p,ll'}^{21}$, and $\mathrm{T}_{p,ll'}^{22}$ are the elements of $\mathbf{T}_p$ [see Eq. (3) for the explicit definition of $\mathbf{T}$]. The phase matrix elements neglecting the interference are computed by

$$P_{ij} = \sum_{p=0}^{\infty} P_{ij}^p C_{\text{sca}}^p \quad i, j = 1, 4, \tag{13}$$

where ${P^p}_{ij}$ is the phase matrix element computed from the pth-order Debye series.

III. RESULTS

A. Numerical validation

Figure 2 shows a comparison of the phase matrix computed from the II-TM and Debye series. The II-TM directly computes the T matrix by employing an invariant embedding procedure, whereas the Debye series approach first computes the reflection and transmission matrices $\tilde{\mathbf{R}}_{11}$, $\tilde{\mathbf{R}}_{22}$, $\tilde{\mathbf{T}}_{21}$, and $\tilde{\mathbf{T}}_{22}$, and then computes the T matrix through Eq. (4). Once the T matrix is obtained, the two methods share the same algorithm to compute the phase matrix of randomly oriented particles. An excellent agreement between the two scattering phase matrices validates Debye's approach, as well as its numerical implementation. Note that the second formula with the inverse of $\mathbf{1} - \tilde{\mathbf{R}}_{22}$ in Eq. (4) is employed in the computation to avoid possible truncation errors in Fig. 2.

Now we turn to the contributions of waves (of various orders) to the phase matrix of randomly oriented spheroids. As expected, the contribution from the diffraction and external reflection to the phase function is featureless, except for a

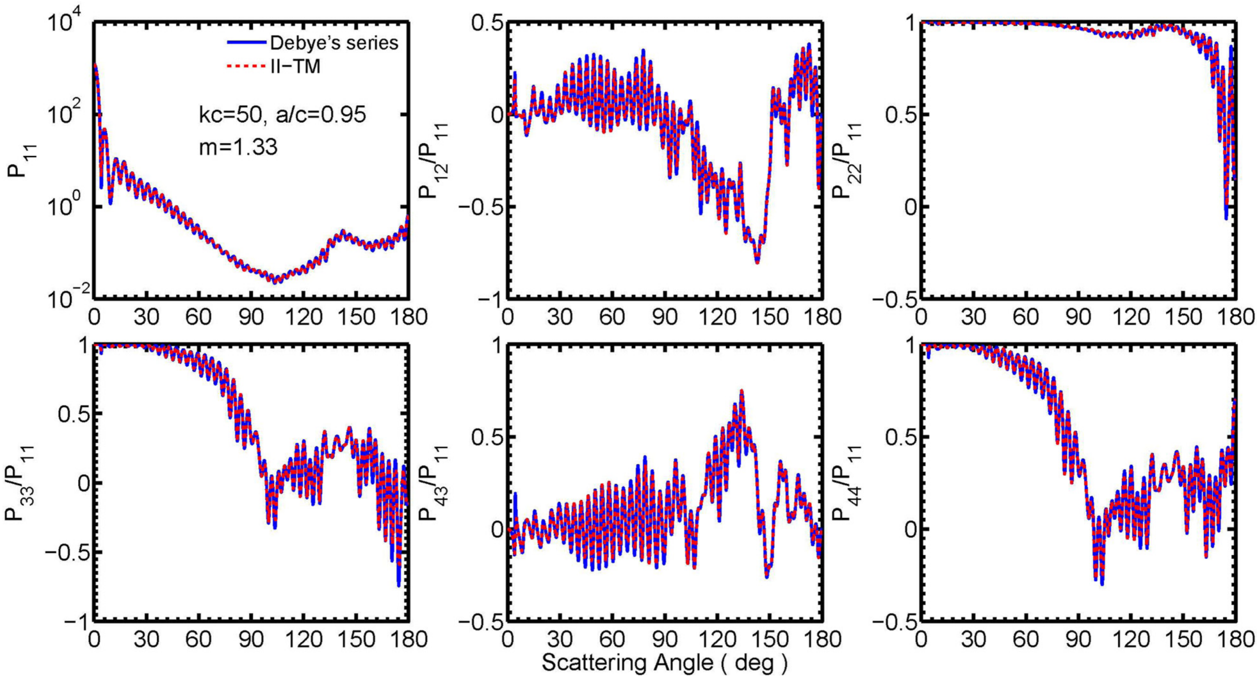

FIG. 2. Comparison of the phase matrix of randomly oriented spheroids (the aspect ratio $a/c = 0.95$) computed from the invariant imbedding T-matrix and Debye's series. The refractive index is 1.33, and the size parameter is $kc = 50$.

strong diffraction peak. A comparison of the diffraction plus reflections computed from the zeroth-order Debye series and physical optics approximation has been given in Bi and Yang [38], showing an excellent agreement. From this paper, it is evident from Fig. 3 that the superposition of the first-order transmission and diffraction and reflection yields close results with the II-TM for scattering angles less than 85 deg. Addition of the transmission after one internal refection dominates the scattering contributions in the scattering angle from 130 to 180 deg. The transmission after two internal reflections has critical contributions to the phase function at scattering angles between 85 and 130 deg. It is evident from Fig. 3 that the summation of wave contributions with $p \leqslant 3$ reasonably explains the phase function computed from the II-TM, although the accuracy can be improved by including higher-order terms. However, from the P_{22}/P_{11} comparison, the maximum of summation terms should be sufficiently large (up to a few tens) to guarantee the convergence of P_{22}/P_{11} between 60 and 120 deg.

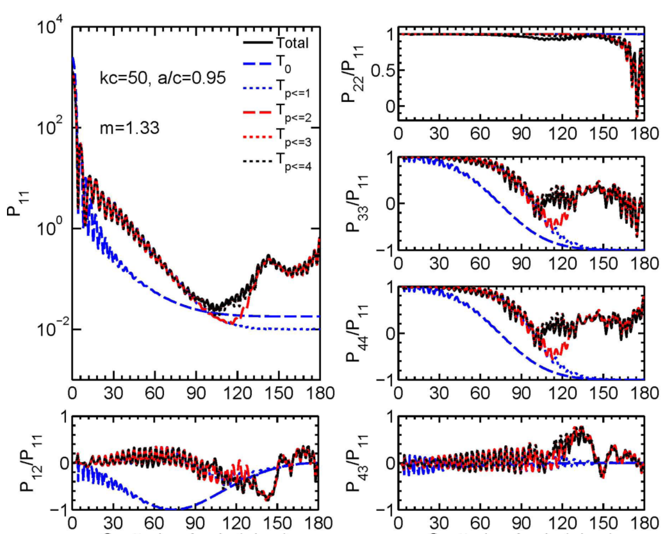

FIG. 3. The contributions of Debye's series of various order to six nonzero phase matrix elements for randomly oriented spheroids with the aspect ratio of 0.95.

B. Linear depolarization ratios

For nearly spherical particles, the LDRs are quite small when the scattering angles are less than 160 deg. Figure 4 shows the depolarization ratio at the scattering angles larger than 160 deg at six refractive indices. $T_{p<=1}$ indicates the result with transmission without internal reflections. It is evident that the LDR is zero for all the refractive indices. This is because the diffraction and reflection produce no depolarization, and the first-order transmission has no contribution to the backscattering scattering angles. For the refractive indices (1.10 and 1.20), a superposition of different orders has an obvious contribution to the depolarization. However, for the refractive indices (1.33, 1.45, 1.55, and 1.70), the contribution from the second-order transmission dominates in the backscattering depolarization. A little impact from $p > 2$ can also be seen at the refractive index of 1.33.

Figure 5 shows the comparison of the LDR as a function of particle orientation. The incident angle is defined as the angle between the direction of the incident light and the symmetric axis of spheroids. For the refractive index 1.10, high-order transmissions could significantly increase the depolarization. However, at the orientation 70°, the depolarization is almost solely from the $p = 2$ term. At the refractive index 1.20, the multiple transmission decreases the depolarization in general.

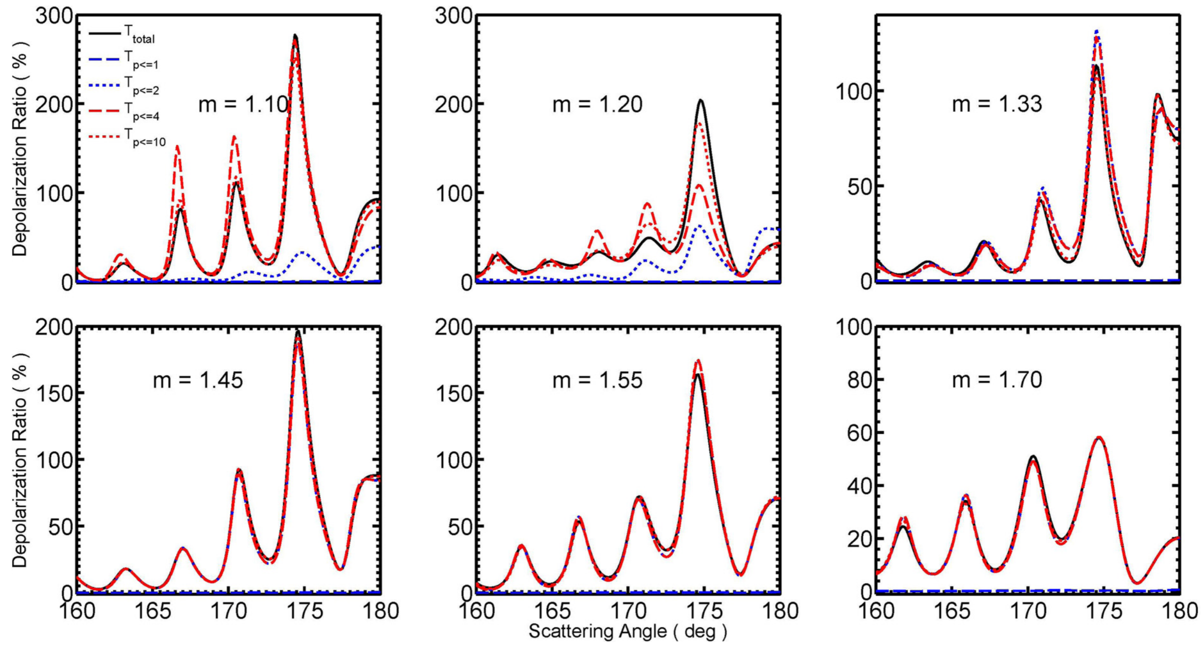

FIG. 4. Depolarization ratios at scattering angles ranging from 160 to 180 deg for six selected refractive indices. The size parameter is kc = 50. The aspect ratio a/c = 0.95.

From Figs. 5(c)–5(e), we can see that the impact of multiple transmissions becomes smaller and smaller as the refractive index increases.

Figure 6 shows the "scattering cross section" as a function of the refractive index. From Fig. 6(a), we can see that the scattering cross section increases as the refractive index

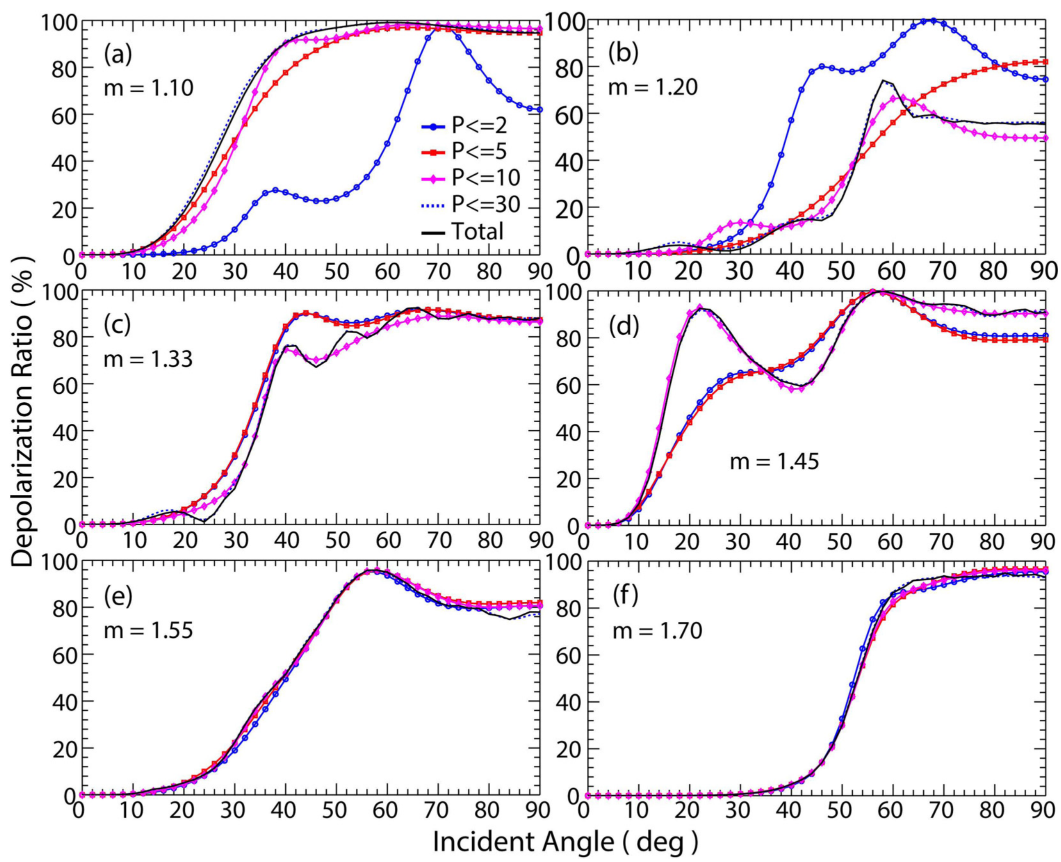

FIG. 5. Depolarization ratios of a particle with different orientations.

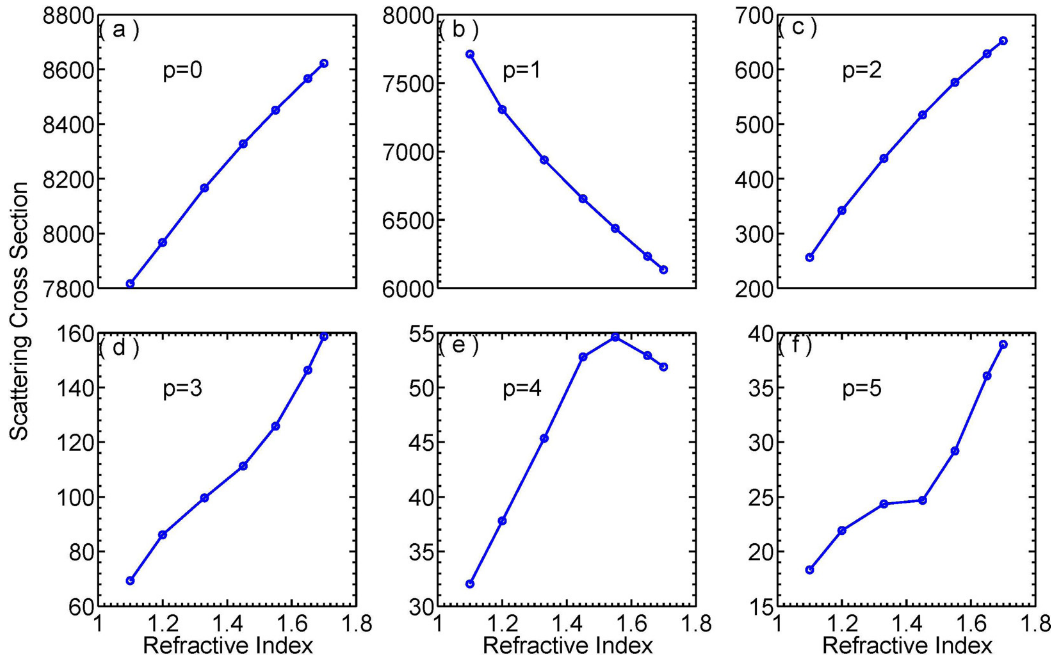

FIG. 6. Scattering cross sections for the different orders of waves.

increases. In this case, the diffraction energy is about the averaged projected area, but the energy from reflected light increases as the refractive index increases, whereas the scattering cross section associated with the transmission ($p = 1$) shown in Fig. 6(b) decreases as the refractive index increases. For higher-order terms, the scattering cross section could first increase and then decrease as the refractive index increases [see Fig. 6(e)].

Based on the scattering cross sections given in Fig. 6, the phase matrices neglecting the phase interference can be computed. Thus, the interference effect on the LDR can be investigated. Figure 7 shows the depolarization ratio as a function of the refractive index for randomly oriented spheroids. The blue curve (with high oscillations) is the rigorous solution computed from the T-matrix method. The red curve (relatively smooth) is computed from the Debye series. However, the interference among the different orders of transmissions is neglected. It is evident that the interference can be reasonably neglected for the refractive index larger than 1.5. The main reason for this is that the $p = 2$ term dominates in the backward scattering. In the 1.3 to 1.5 refractive region, the interference effect is obvious, but the results without interference can still capture the general trend of LDRs as a function of the refractive index. However, as the refractive index decreases, the difference between the two results can be huge. In particular, for the refractive index approaching 1.0, the interference effect plays an essential role in suppressing the depolarization. From this comparison, it is easy to understand how, when ignoring interference, the accuracy of the geometric-optics method becomes poor as the refractive index decreases. Because the LDRs are sensitive to the interference among the different orders of waves, modeling LDRs for optically soft particles should be done carefully when modeling the particle geometry and choosing computational methods (geometric optics could lead to large uncertainties).

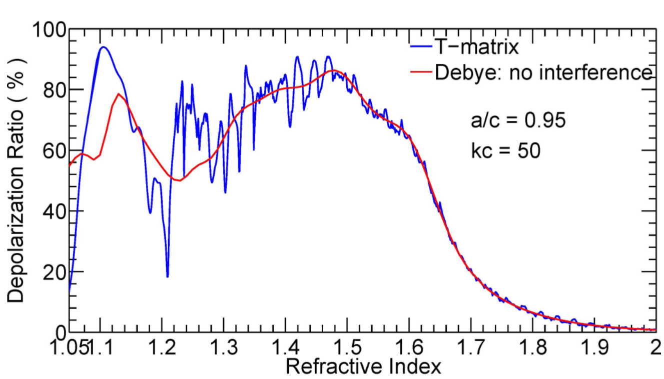

FIG. 7. Interference effect on depolarization ratio.

IV. SUMMARY AND CONCLUSIONS

In this paper, we explored the use of Debye's series to compute the phase matrix of spheroids with either random or fixed orientations. By defining the T matrix with different orders and computing the associated phase matrix, we analyzed the contribution from the different transmitted waves to the scattering. As expected, diffraction and external reflection cause no depolarization. For optically soft particles, the superposition of some transmitted waves is critical to the depolarization. But for a particle with the refractive index larger than 1.3, the depolarization of backscattered light is dominated by the $p = 2$ terms. Namely, the backscattering contributed to the transmission after one internal reflection. These findings have strong implications in LiDAR retrievals of cloud or aerosol parameters, as well as in particle characterization techniques. This paper only focuses on nearly spherical particles. The existing formalism of T-matrix Debye series [27] is applicable to convex-shape nonspherical particles other than the spheroids discussed here. However, some efforts on numerical implementation are necessary to decompose the electromagnetic scattering by large-size and large-aspect-ratio particles (such as particles with size parameter over 100 and aspect ratio over 3–5). One highly possible way to resolve this constraint is to apply the invariant imbedding procedure [7] to the Debye series, which has been proved by our progress on calculating total light scattering by arbitrarily shaped large nonspherical particles [8–10]. This will be our future research subject.

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