Improved Ice Particle Optical Property Simulations in the Ultraviolet to Far-infrared Regime

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Abstract

To derive the bulk radiative properties of ice clouds, aircraft contrails and snow grains, which are fundamental to atmospheric radiative transfer calculations in downstream applications, it is necessary to accurately simulate the scattering of light by individual ice particles. An ice particle optical property database reported in 2013 (hereafter, TAMUice2013) is updated (hereafter, TAMUice2016) to incorporate recent advances in computation of the optical properties of nonspherical particles. Specifically, we employ the invariant imbedding T-matrix (II-TM) method to compute the optical properties of particles with small to moderate size parameters. Both versions use the Improved Geometric Optics Method (IGOM) to compute the optical properties of large ice crystals, but TAMUice2016 improves the treatment of inhomogeneous waves inside the scattering particles in the case where ice is absorptive such as at infrared wavelengths. To bridge the gap between the extinction efficiencies computed from the II-TM and the IGOM, TAMUice2016 includes spectrally dependent higher order terms of the edge effect in addition to the first order counterpart considered in TAMUice2013. Furthermore, the differences between TAMUice2013 and TAMUice2016 are quantified with respect to the computation of the bulk optical properties of ice clouds.

Keywords: Ice particles, optical properties, database, ultraviolet through far-infrared

1. Introduction

Ice particles and snow grains in the terrestrial atmosphere and cryosphere are critical components of the Earth's climate system. Atmospheric ice particles exist in the forms of cirrus clouds, aircraft contrails, type II polar stratospheric clouds (PSCs), and snowflakes. Ice particles can also coexist with supercooled water droplets in mixed-phase clouds. The optical properties of ice particles are fundamental to atmospheric radiative transfer simulations and remote sensing implementations [1-4]. For this reason, substantial computational efforts have been made to compute the optical properties of various ice crystals [5-12] and to develop data libraries in different spectral regimes with consideration of various particle microphysical properties, including particle size, habit and surface texture [13-17]. Furthermore, the bulk optical properties of an ensemble of ice crystals in ice clouds have been developed for practical applications [e.g., 18-20].

An ice optical property database (hereafter, TAMUice2013) has been developed [21] for 11 representative ice crystal habits, namely column, hollow column, plate, droxtal, 5-plate aggregate, 10-plate aggregate, 8-column aggregate, solid bullet rosette, hollow bullet rosette, sphere, and spheroid, over a spectral range of 0.2-100 μm . TAMUice2013 was an effort based on the concepts of two previous datasets [14, 22] with tremendous new computational effort to improve the computational consistency in terms of size-bin resolution and detailed particle morphologies. In addition, the degree of particle surface roughness is considered as an additional morphological parameter in TAMUice2013. To compute the optical properties of each single ice particle, in TAMUice2013 the discrete dipole approximation (DDA)

method [23, references cited therein] was used for small size parameters up to approximately 20 (the specific value varies with the incident wavelength) and the Improved Geometric Optics Method (IGOM, [10]) for size parameters exceeding 20. The IGOM results are semi-empirically adjusted to take into account the edge effect, a term introduced within the framework of a semi-classical scattering theory [24]. TAMUice2013 has already been extensively used in remote sensing applications and radiative forcing studies involving ice clouds (e.g. [25, 26]). Furthermore, the 8-column aggregate model with severe surface roughness is used for the Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 6 (C6) operational ice cloud property retrieval product [27]. A pronounced benefit of the MODIS C6 ice crystal habit model is that it gives rise to consistent optical thickness retrievals using either an algorithm based on visible and near-infrared bands or an algorithm based completely on thermal infrared bands [28].

With feedback from the research community and recent advances in light-scattering computational techniques, we update TAMUice2013. To improve the numerical accuracy of the updated database (hereafter, TAMUice2016), we use a new computational technique, the invariant imbedding T-matrix (II-TM) method, to improve the accuracy in computing the single-scattering properties of randomly oriented ice particles with small to moderate size parameters. In the Christiansen bands ([29, 30] and references therein), where the real part of the refractive index is close to unity and the imaginary part of the refractive index is relatively large, the IGOM accuracy for large particle sizes is significantly improved. In

particular, we substantially improve the treatment of inhomogeneous waves within absorbing ice particles when the imaginary part of the ice refractive index is not zero.

This paper is organized as follows. In Section 2, we briefly summarize the II-TM for determining the single-scattering properties of ice particles. We focus on summarizing the intrinsic strengths of the II-TM, regarding enhancing the accuracy of the single-scattering simulations involving ice crystals. We also demonstrate an improved treatment of inhomogeneous waves in the case of absorbing particles for computing the extinction efficiency. The formalism of the new IGOM implementation is included in Appendix A. Furthermore, we also present a rigorous proof of the asymptotic extinction efficiency in the context of an electric-field volume integral equation in Appendix B. In Section 3, we compare the single-scattering properties of ice particles in TAMUice2013 and TAMUice2016. The summary and conclusions of this study are given in Section 4.

2. Computational Improvements

TAMUice2016 uses the basic TAMUice2013 methodology based on a synergistic combination of different computational methods. However, TAMUice2013 used the DDA method (specifically, the ADDA code developed in [23]) for small to moderate size parameters. TAMUice2016 uses the II-TM for that size parameter regime instead, and incorporates some new improvements of the IGOM in the infrared spectral region where ice crystals are significantly absorptive.

The basic concept of the II-TM is described by Johnson [31], and further theoretical development and a numerically stable implementation of this method are reported by Bi and Yang [11] in the case of general nonspherical particles without axial symmetry. It is worth noting that averaging the optical properties with the T-matrix technique under the random orientation condition can be performed analytically, as proved by Mishchenko [32]. This computational advantage is not provided by numerical methods such as DDA or the finite-difference time domain (FDTD) method [9, 33, 34]. Furthermore, in DDA or FDTD, the number of particle orientations required to appropriately represent the random orientation condition increases significantly with the size parameter and is also strongly sensitive to the imaginary part of the refractive index and particle's shape [35]. In particular, for highly absorptive particles, the near-backscattering phase function should be flat for randomly oriented particles within the geometric optics framework; however, if the number of orientations is insufficient, the near-backscattering phase function exhibits artifact structures or incorrectly enhanced backscattering. For TAMUice2016, in the case of absorbing convex ice particles, the size parameter region where the contribution is negligible from higher order refracted waves is identified based on a simple formula reported by Bi and Yang [36], so the size parameter domain is divided into two subdomains for using the II-TM and IGOM, respectively. This improvements enhances the accuracy of the optical properties for small and moderate size parameters, and ensures a smooth transition from the II-TM regime to the IGOM large particle regime.

The ray-tracing treatment of inhomogeneous waves in absorbing ice particles [37-39] is applied rigorously but in a simplified way as in the previous version of IGOM. The effect of an inhomogeneous wave is only taken into account in the determination of the directions of refracted rays in the scattering particle, although in an absorbing medium the electric field has a nonzero component along the ray propagation direction. The new numerical implementation of the IGOM is presented in Appendix A.

Furthermore, for an ice spheroid we employ a new formula developed by Bi and Yang [36] to compute the extinction efficiency. Although the formula is obtained semi-empirically by modifying the asymptotic formulation for a sphere based on the complex angular momentum theory, the formula for a spheroid is quite accurate in comparison with the results computed from the II-TM. For other ice crystal habits, TAMUice2016 continues to use the methodology in TAMUice2013, but includes higher order terms to bridge the extinction efficiencies computed from the II-TM and the IGOM.

3. TAMUice2013/2016 Comparison

In this section, we focus on analysis of the single-scattering properties reported in TAMUice2016 using the II-TM, and compare the results with the TAMUice2013 counterparts.

Fig. 1 shows a comparison between TAMUice2013 and TAMUice2016 for the asymmetry factor values of hexagonal ice columns. Specifically, the left panel shows the results

computed from the ADDA+IGOM. The blue line depicts a dividing line in TAMUice2013, below and above which the ADDA and IGOM are employed, respectively. The middle panel shows improved computation based on a combination of the II-TM and IGOM in TAMUice2016. The blue line depicts a dividing line for the applications of the II-TM (below the dividing line) and IGOM (above the dividing line). The general patterns of TAMUice2013 and TAMUice2016 are similar, but some differences can be noticed. The right panel shows the differences defined in terms of $(\text{TAMUice2013}-\text{TAMUice2016})/\text{TAMUice2016} \times 100\%$. The relative differences between the two databases can be as large as $\pm 15\%$, primarily in the resonance region when the radii of the circumscribed spheres of nonspherical ice crystals are comparable to the incident wavelength.

Fig. 1 Comparison of the asymmetry factor: (left panel) TAMUice2013 based on an ADDA+IGOM combination; (middle panel) TAMUice2016 based on an II-TM+IGOM

combination; (right panel) relative differences defined in terms of $(\text{TAMUice2013-TAMUice2016})/\text{TAMUice2016} \times 100\%$.

Fig. 2 shows the effect of including higher order terms. For absorptive ice particles, the contribution from higher-order rays to the scattering phase function decreases as the size parameter increases. If the higher-order contributions were negligible, the phase function computed from convex non-spherical particles would be much closer to those of spheres. Therefore, for randomly oriented convex particles, the asymptotic phase function in the direct backscattering direction can be written in the form

$$P(180^\circ) = \frac{1}{Q_{sca}} \left| \frac{m-1}{m+1} \right|^2, \quad (1)$$

where m is the complex refractive index and Q_{sca} is the scattering efficiency. To obtain the critical value of particle size below which the higher order contributions to the scattering phase function must be considered, we compare $P(180^\circ)$ computed from the II-TM and Eq. (1), as shown in Fig. 2, for droxtal ice crystals [12]. Insignificant differences for large particle sizes (the specific values depend on the incident wavelength) indicate negligible contributions from high-order rays, because most energy associated with the first-order refracted rays is absorbed.

Figure 2. Backscattering phase function associated with droxtal ice crystals. The close agreement of the II-TM results and the dashed line for large (relative to the incident wavelength) particles indicates the region where higher-order ray contributions are negligible.

Fig. 3 illustrates the effect of the treatment of inhomogeneous waves on the extinction efficiency computed with the IGOM in the case of absorptive hexagonal ice columns, particularly in the spectral region where the real part of the refractive index is relatively small and the imaginary part of the refractive index is substantial. As shown in the left panel, the extinction efficiencies for a particle size of 10000 μm from TAMUice2013 and TAMUice2016 show large differences at wavelengths near 2.91, 11.1, 44.5 and 48 μm . In the right panel, the extinction efficiencies at the five wavelengths as functions of particle size are depicted. To illustrate the asymptotic solutions by the geometric optics approach, the edge effect contribution to the extinction efficiency is not taken into account in Fig. 3. However, it

must be clarified that the edge effect is incorporated into both TAMUice2013 and TAMUice2016.

Figure 3. Left: spectral extinction efficiencies of randomly oriented hexagonal columns with a size of 10000 μm . Right: extinction efficiencies at four different wavelengths as functions of particle size.

Bi and Yang [36] developed an analytical formula to compute the extinction efficiency of spheroids with high accuracy, which is incorporated in TAMUice2016 computation. Fig. 4 shows the extinction efficiencies computed from the II-TM and the analytical formula across the entire particle size range for all wavelengths. The blue line depicts a dividing line for the applications of the II-TM (left of the dividing line) and IGOM (right of the dividing line). For large wavelengths, the II-TM is only applied for particle sizes smaller than 1000 μm . The

general patterns in conjunction with different aspect ratios are similar, although absolute values are different with minor deviations in terms of local patterns. The II-TM essentially covers the entire resonance domain.

Figure 4. Contour plot of the extinction efficiencies of spheroids with different aspect ratio (a/c) values.

To illustrate clearly the transition of the extinction efficiencies computed with the II-TM to counterparts based on the analytical formula, Fig. 5 compares the extinction efficiencies for five selected wavelengths for six aspect ratio. The solid lines are computed with the II-TM, and the dashed lines are computed from the analytical formula. Note that there are no fitting

coefficients in the formula, and that the transition from the II-TM solution to the results based on the analytic formula is smooth in all cases.

Fig. 5. Extinction efficiencies of spheroids with six aspect ratios. For each aspect ratio, five wavelengths are selected for comparison. The solid lines are computed from the II-TM, and the dashed lines are computed from an analytical formula.

To compare the edge effect of spheroids and spheres, Fig. 6 plots the ratio of the edge effect efficiency of a spheroid to that of a sphere across the entire wavelength spectrum for a particle size of 10000 μm . It is noted that the ratio changes significantly in some spectral regions where the real part of the ice refractive index is small and the corresponding

imaginary part is large. For prolate spheroids ($a/c=0.25$), the ratio is relatively large. For oblate spheroids ($a/c=1.25$), the ratio is relatively small.

Fig.6. The ratio of spectral edge-effect efficiencies of spheroids over those of spheres.

To illustrate the impact of the differences between TAMUice2013 and TAMUice2016 on practical applications, we employ the MODIS C6 ice particle habit model (for particle morphology and size distribution) [27] in conjunction with the two databases to compute the bulk optical properties. Figs. 7, 8, and 9 show the bulk extinction efficiency, single-scattering albedo, and asymmetry factor as functions of the effective particle size (r_e), respectively, at two wavelengths $\lambda = 0.65\mu m$ and $11\mu m$. From the left-upper panel in Fig. 7, it is noted that the bulk extinction efficiency based on TAMUice2016 approaches to 2 when the effective particle size is large (specifically, $r_e > 80\mu m$), whereas the TAMUice2013-based calculation

shows larger values. According to the principle of geometric optics, the asymptotic extinction efficiency of a very large particle is 2. Thus, TAMUice2016 is more accurate than TAMUice2013 from the perspective of the aforesaid physically rationale. However, the differences of the two results are small (specifically, smaller than 3%), as evident from the upper-right panel of Fig. 7. In particular, the differences are less than 1% when the effective radius is larger than $30\text{ }\mu\text{m}$. The lower two panels of Fig. 7 show the comparison of the two results at an infrared wavelength of $11\text{ }\mu\text{m}$. In this case, TAMUice2016-based results are slightly larger but the magnitudes of the differences are less than 1%. For moderate and large particle sizes ($r_e > 30\text{ }\mu\text{m}$), the differences are essentially due to the improved treatment of the inhomogeneous waves inside ice particles whereas the improvements due to the use of the II-TM in TAMUice2016 are response for the differences for small particle sizes ($r_e < 30\text{ }\mu\text{m}$).

Fig. 7. The bulk extinction efficiencies computed from using the MODIS C6 ice habit model (for particle morphology and size distribution) [27] in conjunction with the TAMUice2013 and TAMUice2016 optical property datasets.

Fig. 8 shows the bulk single-scattering albedo. For wavelength $0.65\ \mu\text{m}$, both TAMUice2013 and TAMUice2016 based simulations give single-scattering albedo values that approach essentially to unity because ice crystals are not absorptive at this wavelength. At wavelength $11\ \mu\text{m}$, the TAMUice2016 results slightly enhanced absorption (i.e., with slightly smaller single-scattering albedo), and the maximum of the differences is observed at $r_e \approx 20\ \mu\text{m}$.

Fig. 8. Same as Fig.7, except for the bulk single-scattering albedo.

Fig. 9 is similar to Figs. 7 and 8, but for the asymmetry factor. For the visible wavelength, the differences can be neglected, as evident from the upper panels of Fig. 8. For wavelength 11 μm , the magnitudes of the differences are not larger than 1.2%. However, in the infrared spectral regime, absorption dominates and the detailed phase function (thus, the asymmetry factor) is less important.

Fig. 9. Same as Figs. 7 and 8, except for the asymmetry factor.

From Figs. 7-9, it is expected that the impact of the differences between TAMUice2013 and TAMUice2016 is small for practical applications. For example, the MODIS C6 ice cloud property retrievals are based on a pair of a visible band and a near-infrared band [27]. The information content for retrieving ice cloud optical thickness is essentially from a visible band. It has been suggested that $\tau(1 - \omega g)$ is invariant [27,40], which reduces to $\tau(1 - g)$ at a

visible wavelength for which the absorption of ice is negligible. According to this invariant quantity and Fig. 9, it is expected that the update on the ice crystal optical property database has little impact on the MODIS C6 ice cloud properties. Specifically, Fig. 10 shows the ratio $\tau_{TAMUice2013} / \tau_{TAMUice2016}$ (here $\tau_{TAMUice2013}$ and $\tau_{TAMUice2016}$ indicate the optical thickness values inferred from the radiometric observations made at a visible band centered at 0.65 μm in conjunction with the use of the TAMUice2013 and TAMUice2016 datasets, respectively), which is given by $(1 - \tilde{\omega}_{TAMUice2016} g_{TAMUice2016}) / (1 - \tilde{\omega}_{TAMUice2013} g_{TAMUice2013})$, as a function of the effective particle size. It can be estimated from Fig. 10 that $|\tau_{TAMUice2013} / \tau_{TAMUice2016} - 1| \times 100\%$ is less than 0.04% and the differences can be neglected for retrieving ice cloud optics thickness from observations in a visible band.

Fig. 10. Ratio of $\tau_{TAMUice2013} / \tau_{TAMUice2016}$ estimated by assuming that $\tau(1-\omega g)$ is an invariant

quantity where τ , ω , and g are optical thickness, single-scattering albedo, and asymmetry

factor, respectively.

Summary

This effort updates a database of the optical database of the optical properties of ice crystals from a previous version referred to as TAMUice2013 to an updated version referred to as TAMUice2016. The new version improves the accuracy of the single-scattering properties of

ice particles with small to moderate size parameters. In particular, the transition of the asymmetry factor from the electrodynamics wave domain to the geometric optics domain is much smoother. Furthermore, we have also extended the spectral region into the far-infrared and microwave region, which is reported in a separate paper [41].

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Appendix A: Treatment of Inhomogeneous Waves

This section documents the formulas used in a new version of the IGOM code to compute the optical properties. Consider a general inhomogeneous plane wave propagating in a medium with a refractive index $m = m_r + im_i$:

$$E(\mathbf{r}) = E_0 \exp(ik_0 [N_a \mathbf{e}_a + iN_b \mathbf{e}_b] \cdot \mathbf{r}), \quad (\text{A1})$$

where E_0 is the amplitude of electric field, k_0 the wave number in vacuum space, N_a and N_b the real part and the imaginary part of effective refractive indices, respectively, $\mathbf{e}_a$ the wave propagation direction, and $\mathbf{e}_b$ determines the constant-amplitude wave front. Eq. (A1) must satisfy the vector Helmholtz equation in the form

$$\nabla^2 E(\mathbf{r}) + m^2 k_0^2 E(\mathbf{r}) = 0. \quad (\text{A2})$$

Substituting Eq. (A1) into Eq. (A2), we have

$$N_a^2 - N_b^2 = m_r^2 - m_i^2, \quad (\text{A3})$$

$$N_a N_b \mathbf{e}_a \cdot \mathbf{e}_b = m_r \cdot m_i. \quad (\text{A4})$$

If the medium is non-absorptive, i.e., $m_i = 0$, there are two sets of solutions from Eq. (A3) and (A4), given by

$$N_a = m_r, N_b = m_i = 0, \quad (\text{A5})$$

$$N_a^2 - N_b^2 = m_i^2, \mathbf{e}_a \cdot \mathbf{e}_b = 0. \quad (\text{A6})$$

The wave corresponding to Eq. (A5) is the homogeneous wave. In the case of Eq. (A6), the wave is inhomogeneous in a non-absorptive medium. A straightforward example is the

evanescent wave arising from total internal reflection. For an absorbing particle ($m_i \neq 0$), consider a beam (constant phase front) emerging from the particle to air. The amplitude of the electric field within the cross section in air is nonuniform because the path length in the particle is generally variable except in certain special cases such as a parallel slab. Because $\hat{e}_a \cdot \hat{e}_b = 0$, the amplitude of the electric field in air increases or decreases along a vector perpendicular to $\hat{e}_a$. In this case, N_a and N_b are undetermined in Eq. (A6) and can only be defined through boundary conditions.

For an inhomogeneous wave, a complex wave vector can be defined to be

$$\hat{k} = k_0 [N_a \hat{e}_a + iN_b \hat{e}_b] \quad (\text{A7})$$

It can either be defined as

$$\hat{k} = mk_0 \hat{k} \quad (\text{A8})$$

with $\hat{k}$ a unit complex vector (i.e., $\hat{k} \cdot \hat{k} = 1$). From Eq. (A7), $\hat{k} = [N_a \hat{e}_a + iN_b \hat{e}_b] / m$. The electric-field vector associated with inhomogeneous waves can have a component along the ray-tracing direction (for simplicity, a natural choice is $\hat{e}_a$). Thus, to describe the polarization state, three components are required to describe the field oscillating along three orthogonal unit vectors,

$$\hat{E}_0 = E_{0,\hat{e}_a} \hat{e}_a + E_{0,\perp} \hat{e}_\perp + E_{0,a} \hat{e}_a, \quad (\text{A9})$$

where $\hat{e}_\parallel$ and $\hat{e}_\perp$ are two unit vectors parallel and perpendicular to a plane of reference. In the following, we do not use Eq. (A9) for the description of field polarization, but introduce two complex unit vectors to specify the electric field vector:

$$\vec{E}_0 = E_{0,\alpha} \hat{e}_\alpha + E_{0,\beta} \hat{e}_\beta. \quad (\text{A10})$$

Complex wave vector $\hat{k}$ naturally helps to define basis vectors, $\hat{e}_\alpha$ and $\hat{e}_\beta$, which can be defined by

$$\hat{e}_\alpha = \frac{\hat{k} \times \hat{e}_b}{\sqrt{(\hat{k} \times \hat{e}_b) \cdot (\hat{k} \times \hat{e}_b)}}, \quad \hat{e}_\beta = \hat{k} \times \hat{e}_\alpha, \quad (\text{A11})$$

with $\hat{e}_\alpha \cdot \hat{e}_\alpha = \hat{e}_\beta \cdot \hat{e}_\beta = 1$ and $\hat{e}_\alpha \cdot \hat{e}_\beta = 0$. The intensity I of the field is now given by

$$I = 0.5 \left(|E_{0,\alpha}|^2 \bar{e}_\alpha \cdot \bar{e}_\alpha + |E_{0,\beta}|^2 \bar{e}_\beta \cdot \bar{e}_\beta \right). \quad (\text{A12})$$

Note that $\hat{e}_\alpha \cdot \hat{e}_\alpha$ and $\hat{e}_\beta \cdot \hat{e}_\beta$ are non-unity in general. For a homogenous wave, $\hat{e}_\alpha$ and $\hat{e}_\beta$ are arbitrary mutual orthogonal real vectors perpendicular to the wave vector.

To obtain Snell's law and Fresnel's formula, the incident wave, reflected wave and refracted/transmitted wave are assumed to have the following formalism:

$$\vec{E}_0^i = (E_{0,\alpha}^i \hat{e}_{\alpha,i} + E_{0,\beta}^i \hat{e}_{\beta,i}) \exp(ik_0 [N_{a,i} \hat{e}_{\alpha,i} + iN_{b,i} \hat{e}_{\beta,i}] \cdot \vec{r}) \quad (\text{A13})$$

$$\vec{E}_0^r = (E_{0,\alpha}^r \bar{e}_{\alpha,r} + E_{0,\beta}^r \bar{e}_{\beta,r}) \exp(ik_0 [N_{a,r} \bar{e}_{\alpha,r} + iN_{b,r} \bar{e}_{\beta,r}] \cdot \vec{r}) \quad (\text{A14})$$

$$\vec{E}_0^t = (E_{0,\alpha}^t \hat{e}_{\alpha,t} + E_{0,\beta}^t \hat{e}_{\beta,t}) \exp(ik_0 [N_{a,t} \hat{e}_{\alpha,t} + iN_{b,t} \hat{e}_{\beta,t}] \cdot \vec{r}). \quad (\text{A15})$$

Consider the phase-matching condition,

$$\hat{n} \times \vec{k}_i = \hat{n} \times \vec{k}_r = \hat{n} \times \vec{k}_t, \quad (\text{A16})$$

and

$$\vec{k}_i \cdot \vec{k}_i = \vec{k}_r \cdot \vec{k}_r, \hat{n} \cdot \vec{k}_i = -\hat{n} \cdot \vec{k}_r. \quad (\text{A17})$$

We have the following Snell's law:

$$N_{a,i} = N_{a,r}, N_{b,i} = N_{b,r}, \quad (\text{A18})$$

$$N_{a,i} \vec{e}_{a,i} \cdot \vec{t} = N_{a,t} \vec{e}_{a,t} \cdot \vec{t} = N_{b,t} \vec{e}_{b,t} \cdot \vec{t}, \quad (\text{A19})$$

where

$$\begin{aligned} & N_{a,t}^4 - [m_r^2 - m_i^2 + N_{a,i}^2 (\vec{e}_{a,i} \cdot \vec{t}_a) + N_{b,i}^2 (\vec{e}_{b,i} \cdot \vec{t}_b)] N_{a,t}^2 \\ & + N_{a,i}^2 (\vec{e}_{a,i} \cdot \vec{t}_a) [m_r^2 - m_i^2 + N_{b,i}^2 (\vec{e}_{b,i} \cdot \vec{t}_b)] - [m_r m_i - N_{a,i} N_{b,i} (\vec{e}_{a,i} \cdot \vec{t}_a) (\vec{e}_{b,i} \cdot \vec{t}_b) (\vec{t}_a \cdot \vec{t}_b)] = 0 \end{aligned} \quad (\text{A20})$$

Note that the plane of incidence previously defined in the case of a homogenous wave cannot be easily specified, because $\vec{e}_a$, $\vec{e}_b$ and the normal direction of the local flat surface are no longer coplanar. Thus, the traditional TM and TE modes do not exist. Following Chang et al. [38] to define Fresnel's formula, we have the PE mode

$$r_\beta = \frac{K_i - K_t}{K_i + K_t}, t_\beta = \frac{2K_i}{K_i + K_t}, \quad (\text{A21})$$

and the PM mode

$$r_\alpha = \frac{m_t^2 K_i - m_i^2 K_t}{m_t^2 K_i + m_i^2 K_t}, t_\alpha = \frac{2m_i m_t K_i}{m_t^2 K_i + m_i^2 K_t}, \quad (\text{A22})$$

where $K_i = \hat{n} \cdot \hat{k}_i, K_t = \hat{n} \cdot \hat{k}_t$ Eq. (A1)-(A22) provide a self consistent formalism for ray tracing in an absorbing or non-absorbing medium in the new implementation of the IGOM.

Appendix B: A Proof of Asymptotic Extinction Efficiency

In the case of an absorbing particle, the asymptotic amplitude scattering matrix is determined by diffraction and external reflection. According to the optical theorem, the extinction efficiency can be computed from the forward scattering amplitude and should be 2, but this statement is not obvious from the electric field volume integral equations. Here, we provide an analytical proof of the asymptotic extinction efficiency and, particularly, illustrate the importance of treating the nature of inhomogeneous waves in order to obtain the correct value.

Starting from an electric-field volume equation, the amplitude scattering matrix can be obtained as follows

$$\begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} = \sum_{i=1}^{N_f} D_i \frac{(m^2 - 1) \cos \theta_t}{N_a + i\tilde{N}_b - \hat{r} \cdot \hat{e}_t} \begin{bmatrix} \tilde{S}_2 & \tilde{S}_3 \\ \tilde{S}_4 & \tilde{S}_1 \end{bmatrix}, \quad (\text{B1})$$

where N_f is the total number of facets that face the incident wave, θ_t the refraction angle, $\tilde{N}_b = N_b \cos\theta_t$, $\hat{r}$ the scattering direction, $\hat{e}_t$ the direction of the refracted wave, and D_i is the shape factor associated with the i -th local facet. In the direct forward scattering direction,

$$\begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} = \frac{k^2}{4\pi} \sum_{i=1}^{N_f} S_i \frac{(m^2 - 1) \cos\theta_t}{N_a + i\tilde{N}_b - \cos(\theta_i - \theta_t)} T_\beta \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}, \quad (\text{B2})$$

where S_i is the area of the i -th local facet, θ_i the incident angle and T_β the transmission coefficient, given in the form

$$T_\beta = \frac{2 \cos\theta_i}{\cos\theta_i + m \cos\tilde{\theta}_t} = \frac{2 \cos\theta_i}{\cos\theta_i + N_a \cos\theta_t + iN_b}, \quad (\text{B3})$$

In Eq. (B3), $\tilde{\theta}_t$ is the complex refraction angle. Eq. (B2) is simplified as

$$\begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} = \frac{k^2}{4\pi} \sum_{i=1}^N S_i \frac{2(m^2 - 1) \cos\theta_t \cos\theta_i}{[N - \cos(\theta_i - \theta_t)] [\cos\theta_i + m \cos\tilde{\theta}_t]} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \quad (\text{B4})$$

The denominator can be first expanded into multiple terms, represented as follows

$$\begin{aligned}
& \left[N - \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) \right] \left[\cos \boldsymbol{\theta}_i + m \cos \tilde{\boldsymbol{\theta}}_t \right] \\
&= \left[N - \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) \right] \left[\cos \boldsymbol{\theta}_i + N \cos \boldsymbol{\theta}_t + i \tilde{N}_b \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_t \right] \\
&= \left[N - \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) \right] \left[\cos \boldsymbol{\theta}_i / \cos \boldsymbol{\theta}_t + N + i \tilde{N}_b \tan^2 \boldsymbol{\theta}_t \right] \cos \boldsymbol{\theta}_t \\
&= \left[N \cos \boldsymbol{\theta}_i / \cos \boldsymbol{\theta}_t + N^2 + i N_a \tilde{N}_b \tan^2 \boldsymbol{\theta}_t - \tilde{N}_b^2 \tan^2 \boldsymbol{\theta}_t \right. \\
&\quad \left. - \cos^2 \boldsymbol{\theta}_i - \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_i \cos \boldsymbol{\theta}_i - N \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) - i \tilde{N}_b \tan^2 \boldsymbol{\theta}_t \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) \right] \cos \boldsymbol{\theta}_t \quad (\text{B5})
\end{aligned}$$

Because

$$N \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) = N \cos \boldsymbol{\theta}_t \cos \boldsymbol{\theta}_i + \sin^2 \boldsymbol{\theta}_i + i N_a \tilde{N}_b \sin^2 \boldsymbol{\theta}_t \quad (\text{B6})$$

$$i \tilde{N}_b \tan^2 \boldsymbol{\theta}_t \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) = i \tilde{N}_b \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_i \cos \boldsymbol{\theta}_i + i N_a \tilde{N}_b \tan^2 \boldsymbol{\theta}_t \sin^2 \boldsymbol{\theta}_t \quad (\text{B7})$$

we then have

$$\begin{aligned}
& \left[N - \cos(\boldsymbol{\theta}_t - \boldsymbol{\theta}_i) \right] \left[\cos \boldsymbol{\theta}_i + m \cos \tilde{\boldsymbol{\theta}}_t \right] = \left[N^2 - \tilde{N}_b^2 \tan^2 \boldsymbol{\theta}_t - 1 \right] \cos \boldsymbol{\theta}_t \\
& + \cos \boldsymbol{\theta}_i \left[N \cos \boldsymbol{\theta}_i / \cos \boldsymbol{\theta}_t + i N_a \tilde{N}_b \tan^2 \boldsymbol{\theta}_t - \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_i \cos \boldsymbol{\theta}_i \right. \\
& \left. - N \cos \boldsymbol{\theta}_t \cos \boldsymbol{\theta}_i + i N_a \tilde{N}_b \sin^2 \boldsymbol{\theta}_t - i \tilde{N}_b \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_i \cos \boldsymbol{\theta}_i - i N_a \tilde{N}_b \tan^2 \boldsymbol{\theta}_t \sin^2 \boldsymbol{\theta}_t \right] \\
& = \left[N^2 - \tilde{N}_b^2 \tan^2 \boldsymbol{\theta}_t - 1 \right] \cos \boldsymbol{\theta}_t + \cos \boldsymbol{\theta}_t \left[N \cos \boldsymbol{\theta}_i / \cos \boldsymbol{\theta}_t - \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_i \cos \boldsymbol{\theta}_i \right. \\
& \left. - N \cos \boldsymbol{\theta}_t \cos \boldsymbol{\theta}_i - i \tilde{N}_b \tan \boldsymbol{\theta}_t \sin \boldsymbol{\theta}_i \cos \boldsymbol{\theta}_i \right] \quad (\text{B8})
\end{aligned}$$

The second term in the last expression of Eq. (B8) is zero, which can be proved with the following derivation,

$$\begin{aligned}
& \cos\theta_t \left[N \cos\theta_i / \cos\theta_t - \tan\theta_t \sin\theta_i \cos\theta_i \right. \\
& \quad \left. - N \cos\theta_t \cos\theta_i - i\tilde{N}_b \tan\theta_t \sin\theta_t \cos\theta_i \right] \\
& = N \cos\theta_i - N_a \sin^2\theta_t \cos\theta_i - N \cos^2\theta_t \cos\theta_i - i\tilde{N}_b \sin^2\theta_t \cos\theta_i \\
& = N \sin^2\theta_t \cos\theta_i - N_a \sin^2\theta_t \cos\theta_i - i\tilde{N}_b \sin^2\theta_t \cos\theta_i \\
& = 0
\end{aligned} \tag{B9}$$

Therefore, we obtain

$$\left[N - \cos(\theta_t - \theta_i) \right] \cos\theta_i + m \cos\tilde{\theta}_t = \left[N^2 - \tilde{N}_b^2 \tan^2\theta_t - 1 \right] \cos\theta_t \tag{B10}$$

By using the following identity,

$$m^2 = N^2 - \tilde{N}_b^2 \tan^2\theta_t \tag{B11}$$

we have

$$\begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} = \sum_{i=1}^N \frac{k^2}{2\pi} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \langle S_i \rangle = \frac{k^2}{2\pi} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \sum_{i=1}^N \langle S_i \rangle = \frac{k^2 \langle S \rangle}{2\pi} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}, \tag{B12}$$

where $\langle S \rangle$ is the projected cross section. Eq. (B11) can be obtained from Eqs. (A3) and (A4).

According to the optical theorem, the extinction cross section is obtained as follows:

$$C_{ext} = \frac{2\pi}{k^2} [S_1 + S_2] = 2 \langle S \rangle . \quad (B13)$$

Thus, the extinction efficiency is given by

$$Q_{ext} = \frac{C_{ext}}{\langle S \rangle} = 2 . \quad (B14)$$