Optics Letters

Additivity of integral optical cross sections for a fixed tenuous multi-particle group

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Received 6 November 2018; accepted 12 December 2018; posted 17 December 2018 (Doc. ID 351120); published 14 January 2019

We use the volume integral equation formulation of frequency-domain electromagnetic scattering to settle the issue of additivity of the extinction, scattering, and absorption cross sections of a fixed tenuous group of particles. We show that all the integral optical cross sections of the group can be obtained by summing up the corresponding individual-particle cross sections, provided that the single-scattering approximation applies.

https://doi.org/10.1364/OL.44.000419

Ever since the publication of the monumental text by van de Hulst [1], additivity of the integral optical cross sections of particles forming a small group has typically been taken for granted. However, the arguments in Ref. [1] as well as in Refs. [2–4] are fragmentary as well as qualitative. Importantly, they invoke statistical randomness of particle positions in the group as a necessary condition of additivity. More recently, it was shown that randomness is not required for additivity of the individual extinction cross sections [5,6], yet it was still invoked in Refs. [5–7] to demonstrate the additivity of the scattering cross sections. Most recently [8], this issue was analyzed on the basis of the superposition T-matrix formulation of acoustic scattering by a multi-particle cluster [9], and it was concluded that randomness was not needed for the individual scattering cross sections to be additive.

Given these somewhat diverging accounts and the great practical importance of this subject, the objective of this Letter is to settle the issue of computing the optical cross sections for an N-particle group in the framework of frequency-domain electromagnetic scattering theory. Our analysis is explicitly based on the single-scattering approximation (SSA) of the volume integral equation (VIE) formulation. We discuss under what specific conditions the integral optical cross sections of the N particles are additive and how our conclusions compare with the results of the previous studies.

Throughout the Letter, we assume (and suppress) the monochromatic $\exp(-\mathrm{i}\omega t)$ time dependence of all fields, where $\mathrm{i} = \sqrt{-1}$, ω is the angular frequency, and t is time. Consider a *fixed* N-particle group embedded in an unbounded host medium that is assumed to be homogeneous, linear, isotropic, non-magnetic, and perfectly nonabsorbing (Fig. 1). The particles are made of nonmagnetic isotropic materials and, in general, can have edges, corners, and intersecting internal interfaces [10]. The total field everywhere in the three-dimensional space $\mathbb{R}^3$ is represented as the sum of the impressed incident ("inc") field and the scattered ("sca") field [11]:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\mathrm{inc}}(\mathbf{r}) + \mathbf{E}^{\mathrm{sca}}(\mathbf{r}), \qquad \mathbf{r} \in \mathbb{R}^3, \tag{1}$$

where the position vector $\mathbf{r}$ connects the common origin O and the observation point.

The total field satisfies the following VIE [10]:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{\mathrm{inc}}(\mathbf{r}) + \omega^2\mu_0 \int_{\mathbb{R}^3} \mathrm{d}^3\mathbf{r}' \overleftrightarrow{G}(\mathbf{r},\mathbf{r}') \cdot \mathbf{P}(\mathbf{r}') \qquad \mathbf{r} \in \mathbb{R}^3, \tag{2}$$

where $\mathbf{P}(\mathbf{r}) \triangleq [\varepsilon(\mathbf{r}) - \varepsilon_1]\mathbf{E}(\mathbf{r})$ is the polarization density; $\varepsilon(\mathbf{r})$ is the electric permittivity everywhere in space and ε_1 is its (constant and real) value in the host medium; μ_0 is the magnetic permeability of a vacuum; and $\overleftrightarrow{G}(\mathbf{r},\mathbf{r}')$ is the free-space dyadic Green's function. It is clear that $\mathbf{P}(\mathbf{r})$ vanishes unless $\mathbf{r} \in \cup_i V_i$, where V_i is the volume occupied by particle i (Fig. 1). In writing Eq. (2), we assume an exclusion volume (or principal value) to rigorously handle the strong singularity of $\overleftrightarrow{G}(\mathbf{r},\mathbf{r}')$ at $\mathbf{r} = \mathbf{r}'$ [10]. This is not essential for the following, since we will discuss only the influence of one particle on another.

For the purpose of introducing integral optical cross sections, the incident field will often be a homogeneous plane electromagnetic wave propagating in the direction of the unit vector $\hat{\mathbf{n}}^{\mathrm{inc}}$:

$$\mathbf{E}^{\mathrm{inc}}(\mathbf{r}) = \exp(\mathrm{i}k_1\hat{\mathbf{n}}^{\mathrm{inc}} \cdot \mathbf{r})\mathbf{E}_0^{\mathrm{inc}} \qquad \mathbf{E}_0^{\mathrm{inc}} \cdot \hat{\mathbf{n}}^{\mathrm{inc}} = 0, \tag{3}$$

where $k_1 = \omega\sqrt{\varepsilon_1\mu_0}$ is the real-valued wave number in the host medium. Defining the wave impedance in the host medium as $\eta = \sqrt{\mu_0/\varepsilon_1}$, we have for the corresponding magnetic field

$$\mathbf{H}^{\mathrm{inc}}(\mathbf{r}) = \eta^{-1} \exp(\mathrm{i}k_1\hat{\mathbf{n}}^{\mathrm{inc}} \cdot \mathbf{r})\hat{\mathbf{n}}^{\mathrm{inc}} \times \mathbf{E}_0^{\mathrm{inc}}. \tag{4}$$

However, several results below are valid for an arbitrary incident field as long as it is a solution of the free-space macroscopic Maxwell equations.

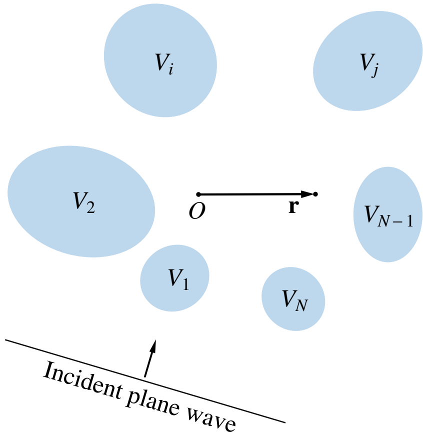

Fig. 1. Electromagnetic scattering by an arbitrary fixed N-particle group centered at the common origin O and subjected to the impressed incident field in the form of a plane electromagnetic wave.

In what follows, we always assume that N is sufficiently small and interparticle separations are sufficiently large, so that the field inside each particle is the same as it would be in the absence of all the other particles, i.e.,

$$\forall i\colon \mathbf{E}(\mathbf{r}) = \mathbf{E}^{\text{inc}}(\mathbf{r}) + \omega^2\mu_0 \int_{V_i} \mathrm{d}^3\mathbf{r}' \overleftrightarrow{G}(\mathbf{r},\mathbf{r}')\cdot\mathbf{P}(\mathbf{r}') \qquad \mathbf{r}\in V_i. \tag{5}$$

This is the very gist of the SSA [6,7]. The use of Eq. (5) as a rigorous definition serves to avoid other (often vague and/or ambiguous) notions of the SSA employed in the literature. The neglected cross influence of particles on each other is described by the integrals over $V_j (j \neq i)$ in Eq. (2).

Let us also assume that the observation point is located in the far zones of all the N particles. Then the cumulative field scattered by the N-particle group at the observation point is given by [6]

$$\mathbf{E}^{\text{sca}}(\mathbf{r}) = \sum_{i=1}^{N} g(r_i)\overleftrightarrow{A}_i(\hat{\mathbf{r}}_i, \hat{\mathbf{n}}^{\text{inc}})\cdot \mathbf{E}^{\text{inc}}(\mathbf{R}_i), \tag{6}$$

where the plane-wave incident field is given by Eq. (3). The vector notation used in this formula is explained in Fig. 2, $r_i = |\mathbf{r}_i|$, and $\hat{\mathbf{r}}_i = \mathbf{r}_i/r_i$ defines the corresponding scattering direction. Furthermore, $g(r) = \exp(\mathrm{i}k_1 r)/r$, and $\overleftrightarrow{A}_i(\hat{\mathbf{r}}_i, \hat{\mathbf{n}}^{\text{inc}})$ is the far-field scattering dyadic of particle i centered at its individual origin O_i. The SSA implies that this dyadic is not affected by the presence of all the other particles.

The integral optical cross sections of the entire group are computed by integrating the Poynting vector over a closed surface enclosing the group [12]. Since the host medium is nonabsorbing, the result must be independent of the choice of the integration surface. Therefore, we simplify the problem by considering a spherical surface S centered at the common origin O and such that its radius tends to infinity: $r \to \infty$ (Fig. 2). Then the scattered field is an outgoing spherical wave centered at O [6]:

$$\mathbf{E}^{\text{sca}}(\mathbf{r}) \underset{r\to\infty}{=} g(r)\mathbf{E}_1^{\text{sca}}(\hat{\mathbf{r}}), \qquad \mathbf{E}_1^{\text{sca}}(\hat{\mathbf{r}})\cdot\hat{\mathbf{r}} = 0. \tag{7}$$

Note that the dimension of $\mathbf{E}_1^{\text{sca}}$ differs from that of the electric field by a factor of length (the dimension of $\overleftrightarrow{A}$). The corresponding magnetic field is given by

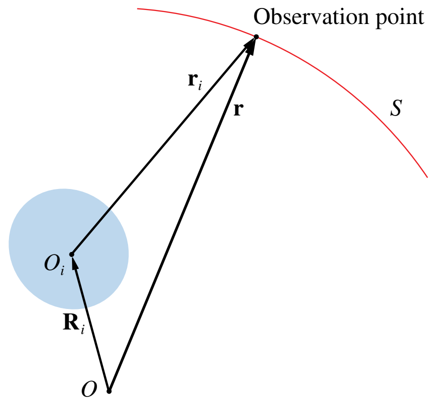

Fig. 2. Vector notation used in Eq. (6).

$$\mathbf{H}^{\text{sca}}(\mathbf{r}) = \eta^{-1} g(r)\hat{\mathbf{r}} \times \mathbf{E}_1^{\text{sca}}(\hat{\mathbf{r}}). \tag{8}$$

In the case of plane-wave illumination,

$$\mathbf{E}_1^{\text{sca}}(\hat{\mathbf{r}}) = \overleftrightarrow{A}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\text{inc}})\cdot\mathbf{E}_0^{\text{inc}}, \tag{9}$$

where

$$\overleftrightarrow{A}(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\text{inc}}) = \sum_{i=1}^{N} \exp(\mathrm{i}\Delta_i)\overleftrightarrow{A}_i(\hat{\mathbf{r}}, \hat{\mathbf{n}}^{\text{inc}}) \tag{10}$$

is the scattering dyadic of the entire N-particle object, and $\Delta_i = k_1(\hat{\mathbf{n}}^{\text{inc}} - \hat{\mathbf{r}})\cdot\mathbf{R}_i$.

The time-averaged Poynting vector $\mathbf{S}(\mathbf{r})$ can be represented as the sum of three terms [12]:

$$\mathbf{S}(\mathbf{r}) = \frac{1}{2}\,\mathrm{Re}\{\mathbf{E}(\mathbf{r})\times[\mathbf{H}(\mathbf{r})]^*\} = \mathbf{S}^{\text{inc}}(\mathbf{r}) + \mathbf{S}^{\text{sca}}(\mathbf{r}) + \mathbf{S}^{\text{ext}}(\mathbf{r}), \tag{11}$$

where "Re" stands for "the real part of"; the asterisk stands for "the complex-conjugate value of";

$$\mathbf{S}^{\text{inc}}(\mathbf{r}) \triangleq \frac{1}{2}\,\mathrm{Re}\{\mathbf{E}^{\text{inc}}(\mathbf{r})\times[\mathbf{H}^{\text{inc}}(\mathbf{r})]^*\} \tag{12}$$

and

$$\mathbf{S}^{\text{sca}}(\mathbf{r}) \triangleq \frac{1}{2}\,\mathrm{Re}\{\mathbf{E}^{\text{sca}}(\mathbf{r})\times[\mathbf{H}^{\text{sca}}(\mathbf{r})]^*\} \tag{13}$$

are the Poynting vector components associated with the incident and scattered fields, respectively; and

$$\mathbf{S}^{\text{ext}}(\mathbf{r}) \triangleq \frac{1}{2}\,\mathrm{Re}\{\mathbf{E}^{\text{inc}}(\mathbf{r})\times[\mathbf{H}^{\text{sca}}(\mathbf{r})]^* + \mathbf{E}^{\text{sca}}(\mathbf{r})\times[\mathbf{H}^{\text{inc}}(\mathbf{r})]^*\} \tag{14}$$

is the extinction term. At infinity,

$$\mathbf{S}^{\text{sca}}(\mathbf{r}) \underset{r\to\infty}{=} \frac{1}{2\eta r^2}|\mathbf{E}_1^{\text{sca}}(\hat{\mathbf{r}})|^2\hat{\mathbf{r}}. \tag{15}$$

In the case of plane-wave illumination,

$$\mathbf{S}^{\text{inc}}(\mathbf{r}) = I^{\text{inc}}\hat{\mathbf{n}}^{\text{inc}}, \qquad I^{\text{inc}} = \frac{1}{2\eta}|\mathbf{E}_0^{\text{inc}}|^2, \tag{16}$$

where I^{inc} is the intensity of the incident plane wave.

According to the Poynting theorem, the net time-averaged flow of electromagnetic power entering the volume bounded by the spherical surface S (i.e., being absorbed inside this volume) is given by [13]

$$W^{\mathrm{abs}}=-\oiint_S \mathrm{d}^2\mathbf{r}\mathbf{S}(\mathbf{r})\cdot\hat{\mathbf{r}}\geq 0. \tag{17}$$

According to Eq. (11), W^{abs} can be written as a combination of three terms:

$$W^{\mathrm{abs}}=W^{\mathrm{inc}}-W^{\mathrm{sca}}+W^{\mathrm{ext}}, \tag{18}$$

where

$$W^{\mathrm{inc}}=-\oiint_S \mathrm{d}^2\mathbf{r}\mathbf{S}^{\mathrm{inc}}(\mathbf{r})\cdot\hat{\mathbf{r}}, \tag{19}$$

$$W^{\mathrm{sca}}=\oiint_S \mathrm{d}^2\mathbf{r}\mathbf{S}^{\mathrm{sca}}(\mathbf{r})\cdot\hat{\mathbf{r}}, \tag{20}$$

$$W^{\mathrm{ext}}=-\oiint_S \mathrm{d}^2\mathbf{r}\mathbf{S}^{\mathrm{ext}}(\mathbf{r})\cdot\hat{\mathbf{r}}. \tag{21}$$

It is quite obvious that since the host medium is nonabsorbing, W^{inc} vanishes exactly for any free-space solution of the macroscopic Maxwell equations. The other two contributions are nonzero and, in the case of plane-wave illumination, can be used to define the scattering and extinction cross sections as follows:

$$C^{\mathrm{sca}}\triangleq W^{\mathrm{sca}}/I^{\mathrm{inc}}, \qquad C^{\mathrm{ext}}\triangleq W^{\mathrm{ext}}/I^{\mathrm{inc}}. \tag{22}$$

The absorption cross section is then given by

$$C^{\mathrm{abs}}\triangleq W^{\mathrm{abs}}/I^{\mathrm{inc}}=C^{\mathrm{ext}}-C^{\mathrm{sca}}\geq 0. \tag{23}$$

In the case of plane-wave illumination, the W^{ext} of an object is directly expressed in terms of the object's forward-scattering dyadic $\overleftrightarrow{A}(\hat{\mathbf{n}}^{\mathrm{inc}},\hat{\mathbf{n}}^{\mathrm{inc}})$ [5–7]. Since all the Δ_i vanish identically in the exact forward-scattering direction, we have from Eq. (10) $\overleftrightarrow{A}(\hat{\mathbf{n}}^{\mathrm{inc}},\hat{\mathbf{n}}^{\mathrm{inc}})=\sum_i\overleftrightarrow{A}_i(\hat{\mathbf{n}}^{\mathrm{inc}},\hat{\mathbf{n}}^{\mathrm{inc}})$. Hence, $W^{\mathrm{ext}}=\sum_i W_i^{\mathrm{ext}}$, and

$$C^{\mathrm{ext}}=\sum_{i=1}^N C_i^{\mathrm{ext}}. \tag{24}$$

In other words, the extinction cross section of the fixed N-particle object is equal to the sum of the individual-particle extinction cross sections computed as if all the other particles were absent [5,6,14]. Note that each C_i^{ext} is independent of the particle position $\mathbf{R}_i$ with respect to the common origin O.

Still considering the case of plane-wave illumination, let us rewrite Eq. (20) as

$$W^{\mathrm{sca}}=\sum_{i=1}^N W_{ii}^{\mathrm{sca}}+\sum_{i=1}^N\sum_{j(\neq i)=1}^N W_{ij}^{\mathrm{sca}}, \tag{25}$$

where

$$W_{ij}^{\mathrm{sca}}\underset{r\to\infty}{=}\frac{1}{2\eta r^2}\oiint_S \mathrm{d}^2\mathbf{r}\hat{\mathbf{r}}\exp[\mathrm{i}(\Delta_i-\Delta_j)]$$
$$\times\{[\overleftrightarrow{A}_i(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}_0^{\mathrm{inc}}]\cdot[\overleftrightarrow{A}_j(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}_0^{\mathrm{inc}}]^*\}. \tag{26}$$

Recalling that

$$C_i^{\mathrm{sca}}=\frac{1}{I^{\mathrm{inc}}}\frac{1}{2\eta}\int_{4\pi}\mathrm{d}\hat{\mathbf{r}}|\overleftrightarrow{A}_i(\hat{\mathbf{r}},\hat{\mathbf{n}}^{\mathrm{inc}})\cdot\mathbf{E}_0^{\mathrm{inc}}|^2 \tag{27}$$

is the $\mathbf{R}_i$-independent scattering cross section of particle i in the absence of all the other particles [5–7], we have $W_{ii}^{\mathrm{sca}}=C_i^{\mathrm{sca}}I^{\mathrm{inc}}$. The computation of the double sum in Eq. (25) is not as straightforward. It can be shown, however, that this double sum vanishes exactly. Indeed, let us rewrite Eq. (20) in the volume-integral form:

$$W^{\mathrm{sca}}=\frac{\omega}{2}\int_{\mathbb{R}^3}\mathrm{d}^3\mathbf{r}\,\mathrm{Im}[\mathbf{E}^{\mathrm{sca}}(\mathbf{r})\cdot\mathbf{P}^*(\mathbf{r})], \tag{28}$$

where "Im" stands for "the imaginary part of." This representation is a direct consequence of the divergence theorem and the Maxwell curl equations [15]. Obviously,

$$W^{\mathrm{sca}}=\sum_{i=1}^N W_i^{\mathrm{sca}}, \tag{29}$$

where

$$W_i^{\mathrm{sca}}=\frac{\omega}{2}\int_{V_i}\mathrm{d}^3\mathbf{r}\,\mathrm{Im}[\mathbf{E}^{\mathrm{sca}}(\mathbf{r})\cdot\mathbf{P}^*(\mathbf{r})]. \tag{30}$$

In the framework of the SSA defined by Eq. (5), $\mathbf{E}^{\mathrm{sca}}(\mathbf{r})$ and $\mathbf{P}(\mathbf{r})$ inside V_i are the same for an isolated particle and for the whole system. Hence, each W_i^{sca} is an individual-particle contribution computed in the absence of all the other particles. Therefore, Eq. (29) expressly states the general additivity of these contributions for any type of incident field.

In the case of plane-wave illumination, Eqs. (25) and (29) must give identical results. We can therefore conclude that $W_i^{\mathrm{sca}}=W_{ii}^{\mathrm{sca}}$ for all i and that the double sum on the right-hand side of Eq. (25) is exactly equal to zero. We thus finally have

$$C^{\mathrm{sca}}=\sum_{i=1}^N C_i^{\mathrm{sca}}. \tag{31}$$

This outcome is consistent with the conclusion reached in Ref. [8] for the case of acoustic scattering. Of course, the same additivity property applies to the total absorption cross section:

$$C^{\mathrm{abs}}=\sum_{i=1}^N C_i^{\mathrm{abs}}. \tag{32}$$

Again, the individual-particle scattering and absorption cross sections are independent of particles' positions and of the presence of all the other particles.

Note that by analogy with Eq. (28), we can re-write Eqs. (17) and (21) as

$$W^{\mathrm{abs}}=-\frac{\omega}{2}\int_{\mathbb{R}^3}\mathrm{d}^3\mathbf{r}\,\mathrm{Im}[\mathbf{E}(\mathbf{r})\cdot\mathbf{P}^*(\mathbf{r})], \tag{33}$$

$$W^{\mathrm{ext}}=-\frac{\omega}{2}\int_{\mathbb{R}^3}\mathrm{d}^3\mathbf{r}\,\mathrm{Im}[\mathbf{E}^{\mathrm{inc}}(\mathbf{r})\cdot\mathbf{P}^*(\mathbf{r})]. \tag{34}$$

Then the additivity of the individual-particle contributions W_i^{ext} to W^{ext} for any incident field immediately follows from Eq. (34), since in the framework of the SSA, all functions under the integral are the same for an isolated particle and for the whole N-particle group. We thus have for any incident field

$$W^{\mathrm{ext}}=\sum_{i=1}^N W_i^{\mathrm{ext}}, \tag{35}$$

where each W_i^{ext} is computed as if all the other particles were absent. In the case of plane-wave illumination, the ratio $W_i^{\mathrm{ext}}/I^{\mathrm{inc}}$ must give the position-independent extinction cross section of particle i, and we thus recover Eq. (24). Proving the general additivity of W_i^{abs},

$$W^{\mathrm{abs}} = \sum_{i=1}^{N} W_i^{\mathrm{abs}}, \tag{36}$$

and recovering Eq. (32) in the case of plane-wave illumination are completely analogous using Eq. (33).

In summary, all our results are based on only two fundamental premises. First, the host medium has been assumed to be perfectly nonabsorbing, since otherwise it would be impossible to give meaningful definitions of the scattering and absorption cross sections [13]. Second, we have relied explicitly on the SSA of the VIE formulation according to which the internal field inside each particle is the same as it would be in the absence of all the other particles [Eq. (5)]. These two assumptions have been sufficient to prove the general additivity of W_i^{ext}, W_i^{sca}, and W_i^{abs} for a fixed multi-particle group and additivity of C_i^{ext}, C_i^{sca}, and C_i^{abc} in the case of plane-wave illumination. Importantly, both types of additivity exactly reproduce energy conservation in the case of a group of nonabsorbing particles. Thus, fundamentally, the assumption of random particle positions is unnecessary (cf. Refs. [1–7]). The same conclusion was reached in Ref. [8] for the case of acoustic scattering.

Legitimate as it may be mathematically, the *ad hoc* assumption (5) may not be easy to interpret in terms of acceptable cluster morphologies and compositions. Of course, among the likely qualitative criteria of applicability of the SSA are the following: (i) the number N of particles in the group is sufficiently small, and (ii) interparticle distances are sufficiently large. However, for practical purposes, it is highly desirable to perform a thorough quantitative analysis of this issue, e.g., by using the numerically exact superposition T-matrix method [16,17]. Doing that can still be problematic, although initial results have already been published [18,19]. Apart from choosing a proper functional norm to evaluate the internal fields, such systematic analysis should address the influence of the magnitude of $[\varepsilon(\mathbf{r}) - \varepsilon_1]$, which is evident from Eq. (5) but is not explicit in requirements (i) and (ii) above.

Requirement (i) is key regardless of which additional assumptions are made. Indeed, even if particles are widely separated, as they are in a typical liquid-water cloud in the terrestrial atmosphere, taking the limit $N \to \infty$ invariably invalidates the SSA and breaks the additivity of the individual-particle optical cross sections [7].

Although randomness of particle positions is not needed for additivity of the integral optical cross sections of a tenuous cluster of particles, it is an essential requirement in modeling "differential" optical observables [6,7], as it serves to extinguish the notorious speckle that is typical of fixed scattering objects [20,21] but is rarely observed in practice. This can also be achieved by averaging over a range of frequencies [22]. Moreover, both types of averaging can facilitate achieving practical validity of the SSA in the first place.

Funding. Goddard Space Flight Center (GSFC) (N/A); Russian Science Foundation (RSF) (18-12-00052); National Aeronautics and Space Administration (NASA) Radiation Sciences Program.

Acknowledgment. We thank Alexander Moskalensky for a discussion of the volume-integral representation of the scattering cross section.

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