Plane-wave coupling formalism for *T*-matrix simulations of light scattering by nonspherical particles

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The computation of light scattering by the superposition *T*-matrix scheme has been restricted thus far to systems made of particles that are either sparsely distributed or of near-spherical shape. In this work, we extend the range of applicability of the *T*-matrix method by accounting for the coupling of scattered fields between highly nonspherical particles in close vicinity. This is achieved using an alternative formulation of the translation operator for spherical vector wave functions, based on a plane-wave expansion of the particle's scattered electromagnetic field. The accuracy and versatility of the present approach is demonstrated by simulating arbitrarily oriented and densely packed spheroids, for both dielectric and metallic particles.

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I. INTRODUCTION

The quantitative description of light scattering by wavelength-scale particle systems is of paramount importance for a wealth of disciplines. Examples span from the characterization and sensing of atmospheric particulates [1], the performance of astrophysical studies [2], investigations in biology [3] and biomedicine [4], to the optimization of light scattering for plasmonic devices [5], light-emitting diodes [6], and solar cells [7].

Strictly numerical simulation techniques, such as the finite-element method (FEM) and the finite-difference time-domain (FDTD) method, provide comfortable solutions for small or periodic systems. However, for larger disordered photonic systems, they require enormous computational resources, rendering the treatment of larger complex systems impossible. An efficient alternative to these numerical tools can be provided by the *T*-matrix method [8–10] in conjunction with the translation addition theorem of the spherical vector wave functions [11] to account for multiple scattering. However, the applicability of this approach is limited thus far to ensembles that are either sparsely distributed or that consist of particles with nearly spherical shape, whereas it breaks down for systems of nonspherical particles when the particle interdistance is low. In fact, the well-established superposition *T*-matrix scheme [12–14] for multiparticle systems requires that any particle's circumscribing sphere does not intersect adjacent particles. One attempt to overcome this limitation is to decompose a single scatterer into multiple subunits, which are then treated as a multiple scattering problem [15]. This way, the downsized subunits' circumscribing spheres exhibit less overlap.

In this contribution, we develop an alternative formalism to accurately describe the multiple scattering between close-by nonspherical particles. The basic idea is to transform the scattered field's spherical-wave expansion (SWE) into a plane-wave expansion (PWE), allowing the use of the much simpler plane-wave translation addition theorem instead of the spherical-wave translation addition theorem. In this way, the nonoverlap restriction of the particles' circumscribing spheres can be avoided, provided that for each pair of particles a separating plane can be found. This is always the case for convex particles. The concept extends our recent work for the case of an oblate particle near a planar interface [16].

We briefly summarize in Sec. II the procedure of the superposition *T*-matrix scheme and highlight its range of applicability. In Sec. III, we give a comprehensive description of the plane-wave coupling formalism for arbitrary orientation of the involved scattering particles.

Finally, in Sec. IV, we study light scattering at two exemplary configurations, both for dielectric and metallic spheroids. We compare results computed with both the conventional procedure based on the SWE translation addition theorem and our plane-wave coupling formalism to reference simulations using the FEM. The first example is given by a two-spheroid configuration and the second example shows a dense cluster of 20 nanorods, which are utilized, e.g., for light management in photovoltaics [17].

II. SCATTERING BY MULTIPLE PARTICLES: GENERAL *T*-MATRIX FORMALISM

One of the most powerful tools to study light scattering by nanoparticles is the *T*-matrix method [8]. For clarity, we briefly summarize its procedure. Comprehensive descriptions can be found, e.g., in Refs. [9,10].

We consider a single particle in a homogeneous, isotropic, linear, and nonabsorbing medium. The electric field $\mathbf{E}(\mathbf{r})$ can be expressed as a superposition of an incoming electric field $\mathbf{E}_{\mathrm{in}}(\mathbf{r})$ and a scattered electric field $\mathbf{E}_{\mathrm{sc}}(\mathbf{r})$,

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_{\mathrm{in}}(\mathbf{r}) + \mathbf{E}_{\mathrm{sc}}(\mathbf{r}). \tag{1}$$

In the *T*-matrix formalism, the incoming electric field is written as a sum of regular spherical vector wave functions (SVWFs) $\mathbf{M}_n^{(1)}(\mathbf{r})$,

$$\mathbf{E}_{\mathrm{in}}(\mathbf{r}) = \sum_n a_n \mathbf{M}_n^{(1)}(\mathbf{r}). \tag{2}$$

The scattered field is written in outgoing SVWFs $\mathbf{M}_n^{(3)}(\mathbf{r})$,

$$\mathbf{E}_{\mathrm{sc}}(\mathbf{r}) = \sum_n b_n \mathbf{M}_n^{(3)}(\mathbf{r}). \tag{3}$$

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Here, the summation index n subsumes the degree l and order m of the multipole, as well as the polarization p of the spherical wave, $n = (l,m,p)$.

The T-matrix of a scattering particle S is defined as the linear operator that maps the amplitudes of incoming wave functions a_n^S to the amplitudes of outgoing wave functions b_n^S,

$$b_n^S = \sum_{n'} T_{nn'} a_{n'}^S. \tag{4}$$

T contains the complete information about the scattering properties of a particle. Accurate and time-efficient methods for the computation of the T matrix are available for a broad variety of scattering particles (for a collection of computer codes, see, for example, the information portal described in Ref. [18]). In the following, we assume that the T matrix of each particle is precisely known.

Increasing the particle number to at least two results in a system where multiple scattering has to be taken into account. Such systems have been well studied in the framework of the superposition T-matrix scheme [19–23]. Multiple scattering denotes that each particle's scattered field $\mathbf{E}_{\mathrm{sc}}^{S'}(\mathbf{r})$ contributes to the incoming field $\mathbf{E}_{\mathrm{in}}^{S}(\mathbf{r})$ of particle S. Since the incoming field at a particle S is no longer known, (4) is not sufficient to describe the entire system. A second equation is needed to determine the incoming field for each particle,

$$a_n^S = a_n^{S,\mathrm{in}} + \sum_{S \neq S'} \sum_{n'} W_{nn'}^{SS'} b_{n'}^{S'}. \tag{5}$$

Here, $a_n^{S,\mathrm{in}}$ denotes the amplitudes of the initial incoming spherical waves at particle S, which are generated by the initial field excitation, e.g., a plane wave or a dipole field. The coupling matrix $\mathrm{W}^{SS'}$ describes how the scattered field of particle S' contributes to the incoming field of particle S. It corresponds to the transpose of the translation operator A,

$$W_{nn'}^{SS'} = A_{n'n}(\mathbf{r}_S - \mathbf{r}_{S'}). \tag{6}$$

For the translation addition theorem of SVWFs [see (A4)], the translation operator can be expressed either in a closed-form expression involving the Wigner-3j symbols [10] or be constructed from an iterative scheme, which can be found in Ref. [9].

Inserting (5) into (4) results in a self-consistent set of equations to account for light scattering by a system of multiple particles. Written in a matrix-vector notation, we obtain

$$\mathbf{b} = (1 - \mathbf{TW})^{-1}\mathbf{Ta}^{\mathrm{in}}. \tag{7}$$

Any particle system built by spheres can be described by Eq. (7). However, for different particle shapes, we have to restrict ourselves to configurations where the distance between particles is large enough to ensure that a particle's circumscribing sphere does not overlap with any other particle. The scattered field's SWE is only valid outside the particle's smallest circumscribing sphere (see Fig. 1). Inside the circumscribing sphere, the field expansion may not converge towards its true value [24]. This restriction can be slightly relaxed towards a sphere, circumscribing the singularities of the scattered field expansion [25]. The question arises of how a correct field representation in the dashed white region can be achieved.

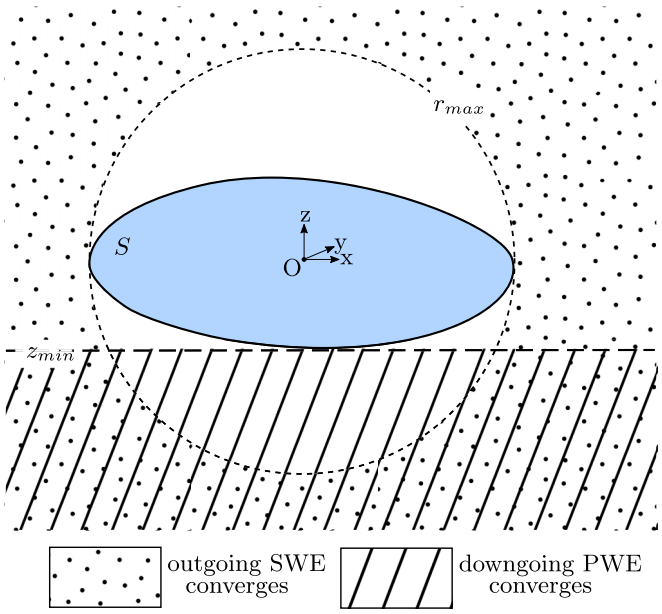

FIG. 1. Oblate scattering particle S in a homogeneous medium. The SWE of the particle's scattered field $\mathbf{E}_{\mathrm{sc}}^{S}(\mathbf{r})$ is only valid outside the particle's smallest circumscribing sphere ($r > r_{\mathrm{max}}$). A correct PWE can be obtained everywhere below the particle ($z < z_{\mathrm{min}}$).

III. NEAR-FIELD COUPLING OF NONSPHERICAL PARTICLES VIA PLANE WAVES

Utilizing an example of light scattering at a particle near a finite cylinder, Boström *et al.* [26] suggested the idea of transforming between spherical-, cylindrical-, and plane-wave representations whenever one of them is not suitable for the configuration considered. Following this idea, we propose to make use of a plane-wave representation of the scattered fields to overcome the separation restriction of the superposition T-matrix formalism.

The benefit of transforming the outgoing SWE into a PWE has been recently shown for nonspherical particles close to a layer interface [16]. In short, the intermediate transformation of the SWE into a truncated PWE acts as a regularization of the divergent part of the SWE in the near-field region. In fact, the domain of validity for a downgoing PWE is limited by a plane that is tangential to the particle from below and oriented such that its normal coincides with the z direction (see dashed region in Fig. 1), thereby allowing a correct representation of the scattered field nearby the particle, where the SWE would diverge. This holds, even if the PWE is constructed starting from a divergent SWE.

For simplicity, we consider a system of two nonspherical particles at a close distance (see Fig. 2). Each particle intersects with the other's circumscribing sphere. Therefore, the conventional T-matrix formalism based on the translation addition theorem is, in general, not suitable to model such configurations. Note that for the depicted configuration, the lower particle does not intersect the upper particle's bounding plane, such that it is entirely located in the domain where the PWE of the scattered field from the upper particle is valid.

To circumvent the limitation of the conventional approach based on the translation addition theorem, we thus introduce

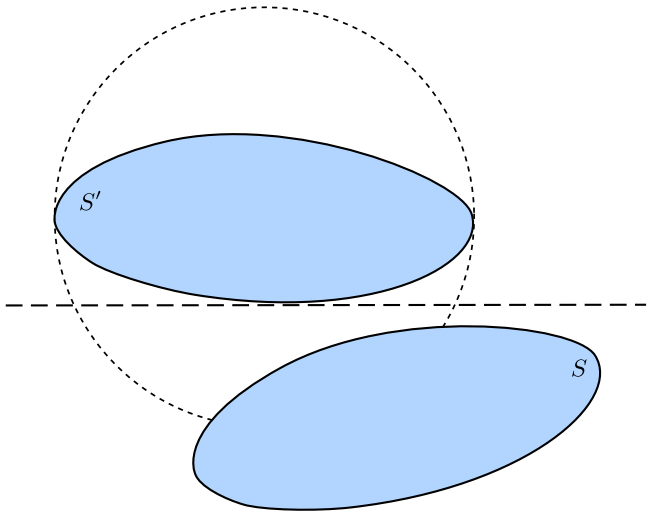

FIG. 2. Two oblate scattering particles at a close distance: particle S intersects the circumscribing sphere of particle S', but it is entirely below the bounding plane.

a formalism to couple $\mathbf{E}^{S'}_{\mathrm{sc}}$ to $\mathbf{E}^{S}_{\mathrm{in}}$ in terms of a PWE, including three main steps:

(i) a transformation of the (outgoing) SWE of $\mathbf{E}^{S'}_{\mathrm{sc}}$ into a PWE,

(ii) a translation of the PWE of $\mathbf{E}^{S'}_{\mathrm{sc}}$ to the center of particle S,

(iii) a retransformation of the PWE of $\mathbf{E}^{S'}_{\mathrm{sc}}$ into a (regular) SWE of $\mathbf{E}^{S}_{\mathrm{in}}$.

As pointed out in the previous section, the incoming field of a single particle is not explicitly known in a multiparticle system, preventing direct field transformations. Instead, we aim for a PWE formulation of the translation operator A.

We start with an outgoing SVWF $\mathbf{M}^{(3)}_n(\mathbf{r}-\mathbf{r}_{S'})$ with its center at position $\mathbf{r}_{S'}$. Expanding it in terms of a downgoing plane wave (A6) and translating it to a position $\mathbf{r}_S$ results in

$$\mathbf{M}^{(3)}_n(\mathbf{r}-\mathbf{r}_{S'}) = \frac{1}{2\pi}\int_{\mathbb{R}^2} d^2\mathbf{k}_\parallel \frac{1}{k_z k}\sum_{j=1}^{2} B_{nj}\left(\frac{-k_z}{k}\right) \times e^{im\alpha}e^{\mathbf{k}\cdot(\mathbf{r}_S-\mathbf{r}_{S'})}\mathbf{E}^-_j(\kappa,\alpha;\mathbf{r}-\mathbf{r}_S). \quad (8)$$

In our notation, $\mathbf{E}^-$ refers to a plane wave propagating in a negative z direction [see (A1)] and B denotes the transformation operator between spherical waves and plane waves [see (A8)].

Utilizing (A7), we retransform the plane wave into a regular spherical wave,

$$\mathbf{M}^{(3)}_n(\mathbf{r}-\mathbf{r}_{S'}) = \frac{2}{\pi}\int_{\mathbb{R}^2} d^2\mathbf{k}_\parallel \frac{1}{k_z k} \times \sum_{j=1}^{2} B_{nj}\left(\frac{-k_z}{k}\right)e^{im\alpha}e^{\mathbf{k}\cdot(\mathbf{r}_S-\mathbf{r}_{S'})} \times \sum_{n'} B^\dagger_{n'j}\left(\frac{-k_z}{k}\right)e^{-im'\alpha}\mathbf{M}^{(1)}_{n'}(\mathbf{r}-\mathbf{r}_S). \quad (9)$$

By comparing (9) with the translation addition theorem for SVWFs (A4), we obtain a formulation of the translation operator $A_{n'n}(\mathbf{r}_S-\mathbf{r}_{S'})$, based on a plane-wave expansion.

Writing out $\int d^2\mathbf{k}_\parallel = \int d\kappa\kappa \int d\alpha$ and utilizing $\mathbf{k}\cdot(\mathbf{r}_S-\mathbf{r}_{S'}) = \kappa\rho_{SS'}\cos(\alpha-\varphi_{SS'}) + k_z z_{SS'}$ with $(\rho_{SS'},\varphi_{SS'},z_{SS'})$ being the cylindrical coordinates of $(\mathbf{r}_S-\mathbf{r}_{S'})$, we obtain

$$W^{SS'}_{nn'} = A_{n'n}(\mathbf{r}_S-\mathbf{r}_{S'}) = \frac{2}{\pi}\sum_{j=1}^{2}\int d\kappa \frac{\kappa}{k_z k}B_{nj}\left(\frac{-k_z}{k}\right)B^\dagger_{n'j}\left(\frac{-k_z}{k}\right) \times e^{i(-k_z z_{SS'})}\int d\alpha e^{i[\kappa\rho_{SS'}\cos(\alpha-\varphi_{SS'})]}e^{i\alpha(m-m')}. \quad (10)$$

To get rid of the double integral, one can compare the second integral in (10) with the integral formulation of the Bessel function J reported in Ref. [27],

$$J_a(\varrho) = \frac{i^{-a}}{2\pi}\int_{-\pi}^{\pi} e^{i\varrho\cos\phi + ia\phi}d\phi. \quad (11)$$

Finally, we end up with

$$W^{SS'}_{nn'} = 4i^{m-m'}\sum_{j=1}^{2}\int d\kappa \frac{\kappa}{k_z k}B_{nj}\left(\frac{-k_z}{k}\right)B^\dagger_{n'j}\left(\frac{-k_z}{k}\right) \times e^{i(-k_z z_{SS'})}e^{i\varphi_{SS'}(m-m')}J_{m-m'}(\kappa\rho_{SS'}). \quad (12)$$

Note that a transformation-translation-transformation scheme to utilize the simple plane-wave addition theorem for SVWF translations has previously been suggested in Ref. [26]. However, here we use this method in order to regularize divergent near-field SWE in the context of multiple scattering by means of a truncation of the involved plane vector wave-function (PVWF) wave numbers.

So far, we have introduced a formalism to couple the scattered electric field of one particle to another by transforming the outgoing SWE into a PWE at a plane that is parallel to the xy plane ($z=0$) of our laboratory coordinate system (L). In a more general case, a plane separating two adjacent particles will not be parallel to the xy plane, but arbitrarily aligned in space. Then, one can perform the plane-wave coupling formalism in a rotated coordinate system (R), in which the separation plane is parallel to the xy plane.

Let $\mathbf{D}$ be a matrix notation of the rotation addition theorem [Eq. (A5)] with

$$D_{lmpl'm'p'}(\alpha,\beta,\gamma) = D^l_{mm'}(\alpha,\beta,\gamma)\delta_{ll'}.$$

Then we obtain the coupling matrix in the laboratory coordinate system of particles S and S' in terms of the coupling matrix in the rotated coordinate system,

$$\mathbf{W}^{SS'}(L) = \mathbf{D}^T(-\gamma,-\beta,-\alpha)\mathbf{W}^{SS'}(R)\mathbf{D}^T(\alpha,\beta,\gamma). \quad (13)$$

To determine the angles of rotation, one needs to find a plane separating the two particles. It is assured that such a plane exists if two particles, with a convex surface shape, do not touch or overlap. One way to obtain such a plane is to find the two surface points p and p' on particles S and S' that are closest to each other. Then, the separation plane is simply perpendicular to the vector $\overline{pp'}$, as illustrated in Fig. 3. Since we want the separation plane to be parallel to the xy plane in our rotated coordinate system, the angles (α,β,γ) have to transform $\overline{pp'}$ into $|\overline{pp'}|\hat{e}_z$ in the laboratory coordinate system.

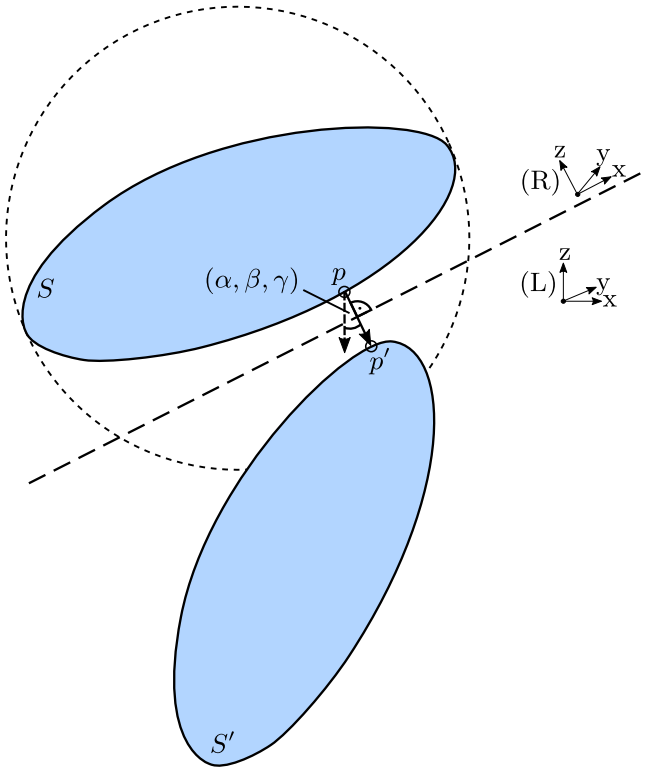

FIG. 3. A plane separating the particles S and S' is normal to the vector $\overline{pp'}$, connecting the two surface points p and p' that are closest to each other. A rotation of $\overline{pp'}$ towards the z vector $\hat{e}_z$ of the laboratory coordinate system (L) by the Euler angles (α,β,γ) ensures that the separation plane is parallel to the xy plane of the rotated coordinate system (R).

To conclude, we have introduced a formalism that couples the scattered electric field of a particle to another by making use of a plane-wave representation. An accurate near-field representation of the scattered field can thereby be achieved in a region where the SWE of the scattered field is not valid. Performing the plane-wave coupling in a rotated coordinate system allows accounting for light scattering by any pair of arbitrarily oriented, nonspherical particles.

IV. APPLICATION EXAMPLES

In this section, we evaluate the accuracy of T-matrix simulations relying on the plane-wave coupling formalism, as introduced in Sec. III. To this end, we compare them with simulations performed with the well-established FEM (available in the COMSOL MULTIPHYSICS software) used as a benchmark. The suitability of the present approach is also emphasized by introducing results obtained with the conventional superposition T-matrix scheme (see Sec. II). To demonstrate the generality of the plane-wave coupling formalism, we consider both (lossless) dielectric and (lossy) metallic nanoparticles, and scattering systems either based on two particles or on a cluster made of 20 particles. In the following, all configurations are excited by a plane wave ($\lambda = 500$ nm), which is polarized along the y direction and propagating in the negative z direction. The ambient medium is chosen to be air, $n_a = 1$.

A. Two arbitrarily oriented particles

In a first step, we study light scattering by a system consisting of two particles, which are either made of a dielectric (TiO_2) or a metallic (Ag) material. The scattering particles considered are oblate spheroids, with semimajor axes of $a = b = 200$ nm and a semiminor axis of $c = 50$ nm, corresponding to dimensionless size parameters of $ka = kb = 2.51$ and $kc = 0.63$. The refractive index of TiO_2 ($n_p = 2.5$) corresponds to the bulk value of titania in the anatase phase and at a vacuum wavelength of $\lambda = 500$ nm [28]. We note that the refractive index of nanoparticles can strongly deviate from its material's bulk value. However, the relatively large particle diameter used here justifies the use of TiO_2's bulk property. The first particle's center is placed at ($x_1 = -80$, $y_1 = 25$, $z_1 = 120$ nm), while the second particle's center is located at ($x_2 = 120$, $y_2 = -20$, $z_2 = -60$ nm). The particles' orientations are obtained by rotation of ($\alpha_1 = \frac{8}{9}\pi$, $\beta_1 = \frac{1}{3}\pi$) and ($\alpha_2 = \frac{14}{9}\pi$, $\beta_2 = \frac{5}{18}\pi$) with respect to a spheroid with its semiminor axis directed along the z axis. In this case, the minimal interparticle distance measures 18 nm. A visualization of the configuration can be found as an inset in Fig. 4(a).

Figure 4 shows the comparison between the conventional superposition T-matrix scheme, for which the translation operator A [see (A4)] has been computed by making use of the Wigner-3j symbols, and the T-matrix scheme relying on the plane-wave coupling formalism (Sec. III). For the computation of all T matrices, a FORTRAN code based on the null-field method with discrete sources (NFM-DS) [9] has been used. For reference, we compare our results to FEM simulations. In Fig. 4(a), the differential scattering cross section (DSCS) in the yz plane is shown. In this example, the SWE has been performed up to a maximal multipole order of $l_{\max} = 15$. A substantial deviation of the blue-dotted line from the FEM solution (black dots) indicates that the exact scattering behavior of the spheroid ensemble is not correctly reproduced by the conventional T-matrix formalism. Such a mismatch is to be expected since one particle's circumscribing sphere intersects the second particle. For the T-matrix simulation utilizing the PVWF coupling (orange line), we obtain a very good agreement with the FEM simulation. For the PWE, the integral over all in-plane wave numbers κ [compare (12)] has been considered up to the truncation value $\kappa_{\text{trunc}} = 3k$. For applicability reasons, the infinite integral has to be truncated at some finite value. As stated in our previous work [16], one has to ensure that for a fixed maximal multipole order $l_{\max}$, only values of the in-plane wave vector $\mathbf{k}_\parallel$ are considered, for which the angular power spectrum has converged against its true value. Very recently, a phenomenological formula for the estimation of κ_{trunc} has been proposed [29].

For a quantification of the accuracy of our simulation results, the relative deviation of both T-matrix formalisms from FEM-based solutions can be found in Fig. 4(c). The relative deviation refers to the L^2 norm of the differential scattering cross sections and is shown for maximal multipole orders $l_{\max} = 1$ up to $l_{\max} = 20$, while the truncation of κ is kept constant at $\kappa_{\text{trunc}} = 3k$. For low values of the maximal multipole order, a convergence of the angular power spectrum is not achieved for the fixed value of $\kappa_{\text{trunc}} = 3k$. By increasing the maximal multipole order above $l_{\max} = 7$, the relative

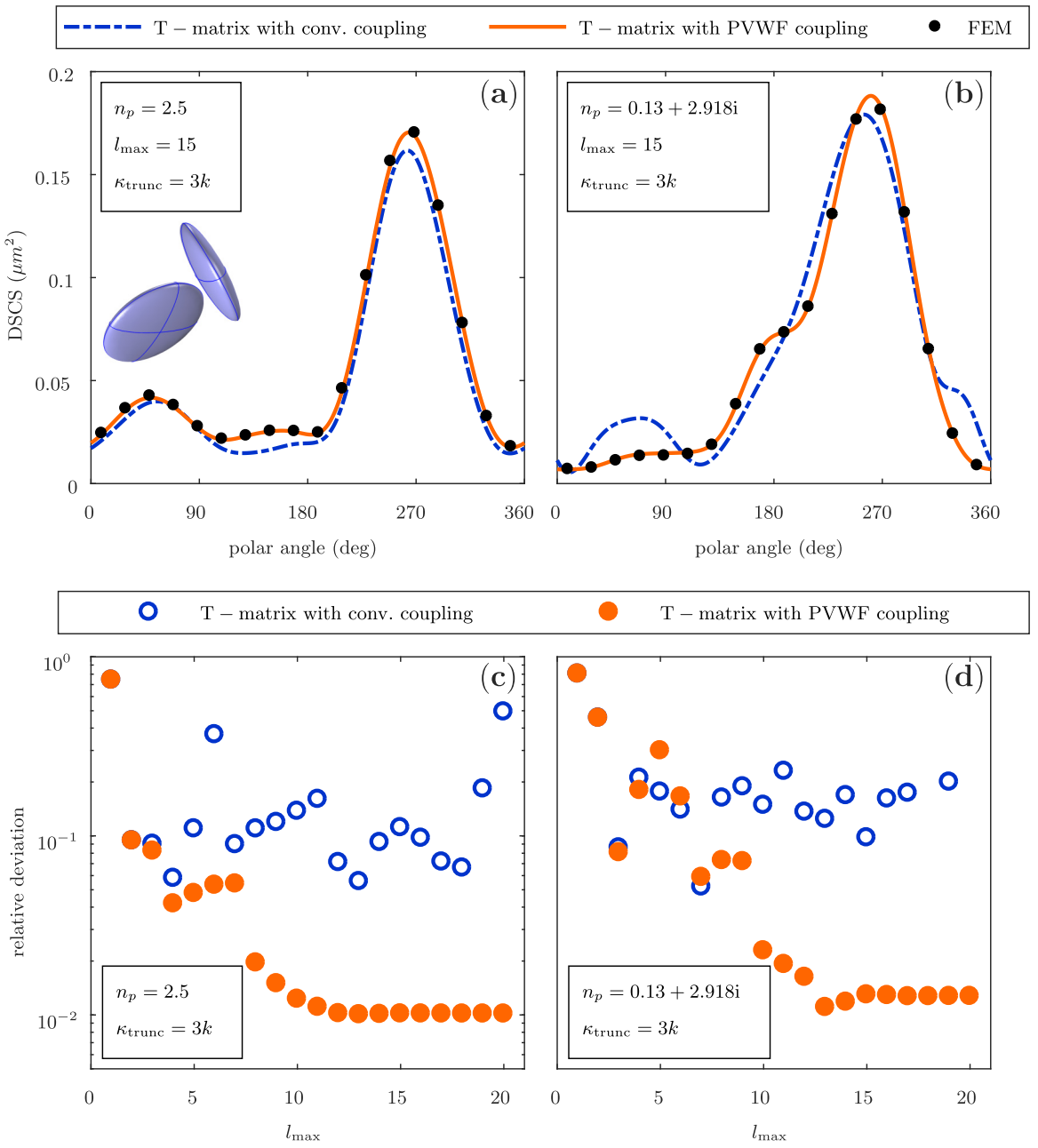

FIG. 4. Light scattering at two oblate spheroids with semiaxes ($a = b = 200, c = 50$ nm). The particles are excited by a plane wave ($\lambda = 500$ nm), polarized in the y direction and propagating in the negative z direction. The ambient medium is air ($n_a = 1$). (a) DSCS of two TiO_2 particles ($n_p = 2.5$). For the SWE, multipole orders up to $l_{\max} = 15$ are considered. The PWE is truncated at $\kappa_{\text{trunc}} = 3k$. (b) DSCS of two oblate Ag particles ($n_p = 0.13 + 2.918i$). The relative deviation of the DSCS for the conventional T-matrix formalism and the PVWF coupling procedure with respect to the FEM simulations is shown for (c) TiO_2 and (d) Ag. The maximal multipole order is varied from $l_{\max} = 1$ up to $l_{\max} = 20$, while the PWE truncation is kept constant at $\kappa_{\text{trunc}} = 3k$.

deviation of the PVWF coupling formalism (orange dots) converges towards a minimal relative deviation of 1%.

For the conventional superposition T-matrix formalism (blue circles), no convergence of the relative deviation can be obtained. Moreover, the relative deviation fluctuates around 10% and strongly increases for large multipole orders ($l_{\max} \geqslant 19$). Such divergent behavior in the near-field coupling has to be expected, as it reflects the divergence of the SWE in the near field with growing multipole order.

In a second example, we consider the two spheroids illustrated in Fig. 4(a) to be made of silver with a refractive index of $n_p = 0.13 + 2.918i$ at $\lambda = 500$ nm [30]. Figure 4(b) shows the DSCS for the silver spheroids at a maximal multipole order of $l_{\max} = 15$ and $\kappa_{\text{trunc}} = 3k$. Again, the coupling via plane waves enables a good agreement with the FEM simulations, unlike the conventional superposition T-matrix formalism results, which strongly differ from the FEM reference. As shown in Fig. 4(d), relative deviations, comparable to the TiO_2 case, are obtained

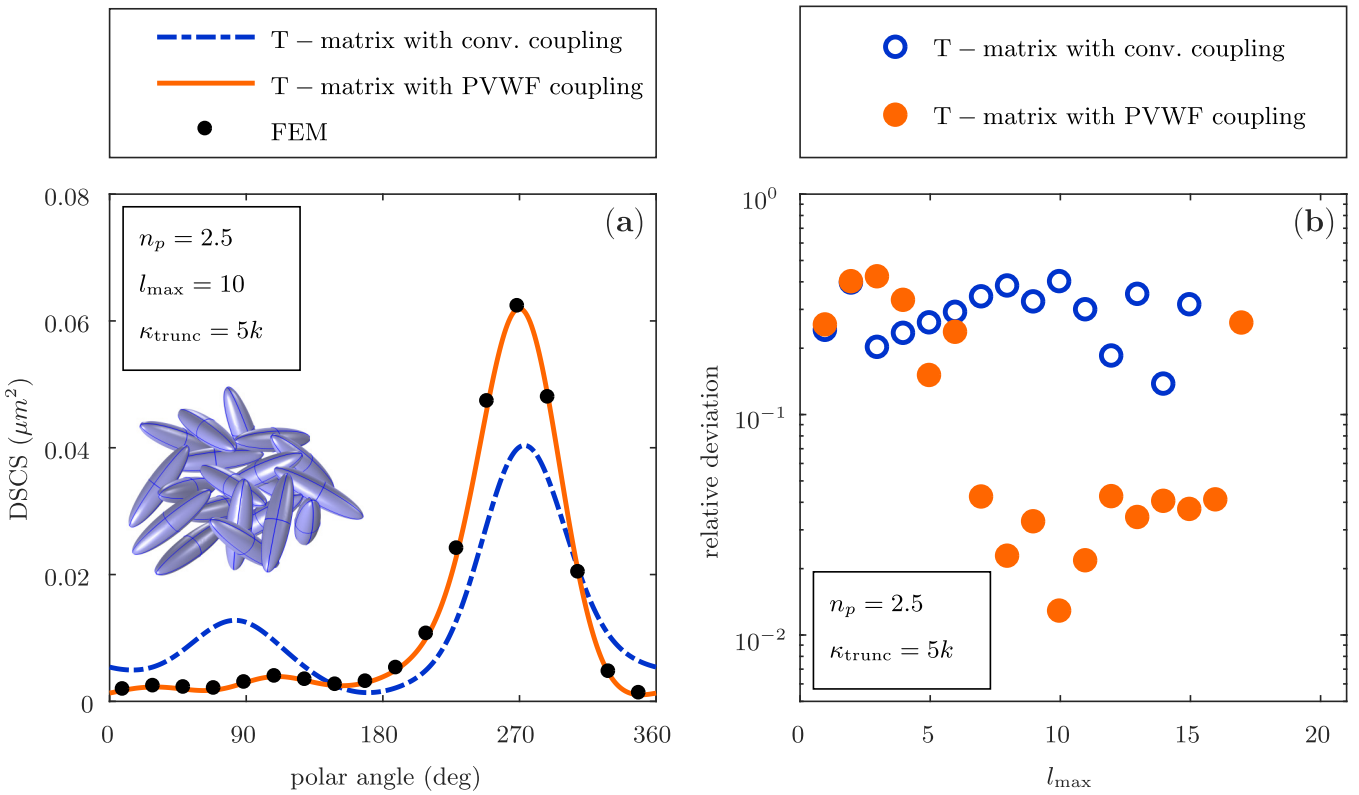

FIG. 5. Light scattering by 20 prolate spheroids ($a = b = 30, c = 120$ nm). TiO_2 particles ($n_p = 2.5$) are excited by a plane wave ($\lambda = 500$ nm), polarized in the y direction and propagating in the negative z direction. The ambient medium is air ($n_a = 1$). (a) Differential scattering cross section of the spheroid cluster. For the SWE, multipole orders up to $l_{\max} = 10$ are considered. The PWE is truncated at $\kappa_{\text{trunc}} = 5k$. (b) The relative deviation of the DSCS for the conventional T-matrix formalism and the PVWF coupling procedure with respect to the FEM simulations. The maximal multipole order is varied from $l_{\max} = 1$ up to $l_{\max} = 20$, while the PWE truncation is kept constant at $\kappa_{\text{trunc}} = 5k$.

for the metallic nanoparticles. Thus, above a multipole order of $l_{\max} = 10$, the relative deviation starts converging towards 1.3% for the plane-wave coupling, while it varies between 10% and 20% for the conventional formalism based on the spherical-wave translation addition theorem.

B. Cluster of spheroids

In this example, we extend the validation of our approach to a more complex scattering system. The latter consists of a cluster made of 20 prolate TiO_2 spheroids ($n_a = 2.5$) with semiminor axes of $a = b = 30$ nm and a semimajor axis of $c = 120$ nm. The cluster is formed by arbitrarily oriented particles [for visualization, see Fig. 5(a)]. Such scattering clusters find applications in dye-sensitized solar cells, where the TiO_2 rods are exploited as a scattering layer for improving light harvesting [17,31]. Smaller, very dense clusters consisting of a few TiO_2 particles can also be found in white paint, which can be used as light-trapping back reflectors in photovoltaics [32].

The cluster considered herein functions as an extreme challenge for the plane-wave coupling formalism. The prolate shape of the scattering particles and an aspect ratio of 4 allows for very low distances between particle centers, in comparison to the particle diameters. In some cases, the high packing factor leads to a minimal distance between adjacent particles below 1 nm and to overlapping of the circumscribing sphere of one particle with multiple neighboring particles.

Figure 5(a) compares the calculated DSCS of the spheroid cluster for the conventional superposition T-matrix scheme in conjunction with the translation addition theorem (blue-dotted line), the PVWF coupling formalism (orange line), and FEM simulations (black dots). The spherical-wave expansion has been taken into account up to a multipole order of $l_{\max} = 10$, while the plane-wave expansion has been truncated at $\kappa_{\text{trunc}} = 5k$. A good agreement between the T-matrix simulations relying on the PVWF coupling formalism and the FEM can be observed, while the conventional T-matrix scheme's results do not match the FEM simulation. For $\kappa_{\text{trunc}} = 5k$ and large multipole orders ($l_{\max} > 16$), the relative deviation [see Fig. 5(b)] between both T-matrix and FEM simulations show divergent behavior. In this configuration, very low distances below 1 nm lead to large values for the spherical Hankel function of the first kind $h_l^{(1)}$ [see definition of the outgoing SVWFs (A2)]. This can lead to an ill-conditioning of the linear system (5), when too large multipole orders $l_{\max}$ are considered. Such divergence has been reported for decreasing distances between a spheroid and an interface [25]. Doicu *et al.* state that for each fixed distance, a domain of the maximal multipole order $l_{\max}$ exists for which small deviations in the computed scattering response are obtained.

Such a plateau can be observed for the relative deviation between the PVWF coupling formalism and FEM. For maximal multipole orders of $l_{\max} = 7, \ldots, 16$, the relative deviation

does not exceed a value of 4%, with a minimal deviation of 1.3% at $l_{\max} = 10$. In comparison, the conventional superposition T-matrix approach using the translation addition theorem for SVWFs shows a minimal deviation of 13.7% at $l_{\max} = 14$ and typically exceeds 25%.

V. DISCUSSION AND CONCLUSIONS

We have shown that the T-matrix approach can be suitable to evaluate light scattering by dense systems of highly nonspherical particles, even if the circumscribing spheres intersect adjacent particles. To account for multiple scattering of neighboring particles, the SVWF translation operator can be expressed in a plane-wave expansion. In practice, one has to ensure that for a given maximal multipole order of the SWE, the in-plane wave number of the PWE is truncated in a regime where the angular spectrum converges [16]. For low values of the maximal multipole order $l_{\max}$, the accuracy is limited by the multipole truncation error, whereas for large $l_{\max}$, the poor condition number of the linear system becomes prohibitive; compare [16,25].

In this contribution, we have applied the plane-wave coupling formalism to the case of spheroids. In general, our approach works for any nonspherical particle with a convex surface shape (or arbitrary particles, as long as the convex hulls do not overlap).

Regarding the computation time, the presented formalism cannot compete with the conventional superposition T-matrix scheme using the SVWF translation addition theorem, but exceeds it by a factor of 10 in terms of accuracy for the shown examples. The additional effort of the plane-wave coupling can be reduced to a minimum by utilizing the conventional scheme for coupling between particles that are not within a very low distance. This way, the additional effort brought by the plane-wave coupling scheme scales only linearly with the number of involved particles, and thereby becomes negligible with growing particle numbers.

We conclude that the range of applicability of the T-matrix approach is much larger than typically expected. It has the potential to solve light-scattering problems in large disordered systems, where strictly numerical approaches such as the FEM or the FDTD method struggle in terms of hardware requirements.

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APPENDIX: WAVE FUNCTIONS AND TRANSFORMATIONS

The plane vector wave functions are defined as

$$\mathbf{E}_j^{\pm}(\kappa,\alpha,\mathbf{r}) = \exp(i\mathbf{k}^{\pm}\cdot\mathbf{r})\hat{\mathbf{e}}_j. \tag{A1}$$

Here, $(\kappa,\alpha,\pm k_z)$ define the cylindrical coordinates of the wave vector $\mathbf{k}^{\pm}$, with $k_z = \sqrt{k^2-\kappa^2}$ and the wave number $k = n_0\omega$. The plus sign corresponds to waves propagating in the positive z direction; the minus sign refers to waves propagating in the negative z direction. Index j of $\mathbf{E}_j$ denotes the polarization (1 = TE and 2 = TM) realized by the unit vectors $\hat{\mathbf{e}}_1 = \hat{\mathbf{e}}_\alpha$ and $\hat{\mathbf{e}}_2 = \hat{\mathbf{e}}_\beta$, which belong to the azimuthal and polar angle of $\mathbf{k}^{\pm}$.

Besides plane-wave functions, we make use of spherical vector wave functions, which read [9]

$$\mathbf{M}_{lm1}^{(\nu)}(\mathbf{r}) = \frac{1}{\sqrt{2l(l-1)}}\nabla\times\left[\mathbf{r}z_l^{(\nu)}(kr)P_l^{|m|}(\cos\theta)e^{im\phi}\right], \tag{A2}$$

$$\mathbf{M}_{lm2}^{(\nu)}(\mathbf{r}) = \frac{1}{k}\nabla\times\mathbf{M}_{lm1}^{(\nu)}(\mathbf{r}), \tag{A3}$$

where (r,θ,ϕ) are the spherical coordinates of the position vector $\mathbf{r}$. Index (ν) indicates whether the wave function is of a regular kind ($\nu = 1$) or if it represents an outgoing wave ($\nu = 3$). In the case of regular spherical waves, the radial wave function $z_l^{(\nu)}$ stands for the spherical Bessel function of the order of l, $z_l^{(1)} = j_l$. Outgoing spherical waves involve the spherical Hankel function of the first kind, $z_l^{(3)} = h_l^{(1)}$. $P_l^{|m|}$ denotes the normalized associated Legendre functions. The spherical-wave functions $\mathbf{M}_{lmp}^{(\nu)}$ are specified by the following indices: $l = 1,2,\ldots$ describes the angular index with respect to θ, $m = -l,\ldots,l$ describes the angular index with respect to ϕ, and p describes the spherical polarization (1 = TE,2 = TM). For a more condensed notation, the indices are subsumed into a multi-index $(lmp)\to n$.

A translation of SVWFs can be accounted for by making use of the translation addition theorem [11],

$$\mathbf{M}_n^{(3)}(\mathbf{r}+\mathbf{d}) = \sum_{n'} A_{nn'}(\mathbf{d})\mathbf{M}_{n'}^{(1)}(\mathbf{r}) \quad \text{for } r < d. \tag{A4}$$

The translation operator $A(\mathbf{d})$ can be obtained by making use of recurrence formulas given in [9,33]. Alternatively, one can use expressions involving the so-called Wigner-3j symbols found, e.g., in Refs. [10,11,34].

Transforming SVWFs from a laboratory coordinate system (L) to a rotated coordinate system (R) can be achieved, utilizing the rotation addition theorem for SVWFs [34],

$$\mathbf{M}_{lmp}^{(1,3)}(R) = \sum_{m'=-l}^{l} D_{mm'}^{l}(\alpha,\beta,\gamma)\mathbf{M}_{lm'p}^{(1,3)}(L). \tag{A5}$$

Function D refers to the so-called Wigner D functions. Recurrence formulations for D can be found, e.g., in Ref. [9] or Ref. [10]. The rotation between the two coordinate systems is defined by the Euler angles (α,β,γ) in $zy'z'$ convention.

Spherical vector wave functions can be expanded in plane vector wave functions, and vice versa [26],

$$\mathbf{M}_n^{(3)}(\mathbf{r}) = \frac{1}{2\pi}\int_{\mathbb{R}^2} d^2\mathbf{k}_\parallel \frac{1}{k_z k}\sum_{j=1}^{2} B_{nj}\left(\frac{\pm k_z}{k}\right) \times \mathbf{E}_j^{\pm}(\kappa,\alpha;\mathbf{r})e^{im\alpha} \quad \text{for } z \gtrless 0. \tag{A6}$$

The integral is performed over the in-plane components of the wave vector $\mathbf{k}_\parallel$ with its polar coordinates κ,α.

A plane-wave representation in terms of regular spherical vector wave functions reads

$$\mathbf{E}_j^{\pm}(\kappa,\alpha;\mathbf{r}) = 4\sum_n e^{-im\alpha} B_{nj}^{\dagger}\left(\frac{\pm k_z}{k}\right)\mathbf{M}_n^{(1)}(\mathbf{r}). \tag{A7}$$

The transformation operator B is given by

$$B_{nj}(x) = -\frac{1}{i^{l+1}}\frac{1}{\sqrt{2l(l+1)}}(i\delta_{j1}+\delta_{j2})\left[\delta_{pj}\tau_l^{|m|}(x) + (1-\delta_{pj})m\pi_l^{|m|}(x)\right], \tag{A8}$$

where the spherical functions π and τ are defined as

$$\pi_l^m(\cos\theta) = \frac{P_l^m(\cos\theta)}{\sin\theta}, \quad \tau_l^m(\cos\theta) = \partial_\theta P_l^m(\cos\theta).$$

In the "daggered" version of the transformation operator $B^{\dagger}$, all explicit i are set to $-i$.

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