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COMMENT

Comment on "Plasmonic nanoparticle monomers and dimers: from nanoantennas to chiral metamaterials" by Chigrin et al. in Appl Phys B (2011) 105:81–97

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Abstract Chigrin et al. (Appl Phys B 105:81, 2011) have recently presented a general direct solution of the integral equation for the electric field inside the scatterer, which does not require inversion of integral operator or, equivalently, solution of large system of linear equations. This comment points out that the rigorous applicability of the considered derivation is limited to ellipsoids much smaller than the wavelength.

In Section 2.1 of reference [1], a general solution of the integral equation for the electric field inside the scatterer is provided. The core result is given by Eqs. (9), (14), and (15). If true, this would provide a breakthrough in simulation of light scattering, because any light-scattering problem would be solvable by directly computing two integrals on a dense-enough discretization of the scatterer. In comparison with current state-of-the-art volume discretization methods, e.g., the discrete dipole approximation [2], this would avoid solution of a large system of linear equations, which is a major computational bottleneck of this method.

Unfortunately, the provided derivation of the general solution has two serious flaws:

(1) Equation (10) is correct and agrees with previous publications [3, 4]. However, the newly proposed Eq. (9), which defines $\overleftrightarrow{\Xi}(\mathbf{r}')$, implicitly assumes that the internal field is *pointwise* proportional to the incident field: $\mathbf{E}(\mathbf{r}') = \overleftrightarrow{\Xi}(\mathbf{r}')\mathbf{E}^i(\mathbf{r}')$. This assumption can be partly justified for a particle much smaller than the wavelength, illuminated by field $\mathbf{E}^i$ that is constant inside the particle (e.g., a plane wave). However, it is then correct only for this narrow class of $\mathbf{E}^i$, which implies that Eq. (9) defines operator $\overleftrightarrow{\mathbf{T}}$ only for such specific $\mathbf{E}^i$ (and not for arbitrary one). Consider as a counterexample $\mathbf{E}^i$ equal to the field of the point dipole located near the particle. In other words, Eq. (9) only defines the action of $\overleftrightarrow{\mathbf{T}}$ on a constant vector, and as such cannot be used to write out $\overleftrightarrow{\mathbf{T}}\overleftrightarrow{\mathbf{t}}\mathbf{E}^i$ in Eq. (11). The only special case where Eq. (9) can be applied to $\overleftrightarrow{\mathbf{T}}\overleftrightarrow{\mathbf{t}}\mathbf{E}^i$ is when $\overleftrightarrow{\mathbf{t}}\mathbf{E}^i$ is also constant (inside the particle), which happens *only* for an ellipsoid much smaller than the wavelength. In the latter case not only $\mathbf{E}^i$ but also $\mathbf{E}$ is constant, which implies constant $\overleftrightarrow{\mathbf{t}}\mathbf{E} = \mathbf{E} - \mathbf{E}^i$, and since any constant $\mathbf{E}^i$ is a total field for another properly chosen incident field, $\overleftrightarrow{\mathbf{t}}\mathbf{E}^i$ is also constant.

Moreover, pointwise proportionality is always wrong in the general case of particle size comparable to the wavelength, as can be seen from Eq. (10) describing non-pointwise dependence and from the following example. Consider a wavelength-sized sphere illuminated by a plane wave. Rotating incident direction (i.e., $\mathbf{E}^i$) should change $\mathbf{E}$ accordingly; hence, $\overleftrightarrow{\Xi}(\mathbf{r}')$ should be spherically symmetric, and obtained $\mathbf{E}$ should have a very special structure. The latter is definitely not observed in real simulations [5]. To make Eq. (9) correct, one of the following modifications is required (preserving the linearity of the problem):

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- Replace $\overleftrightarrow{\Xi}(\mathbf{r}')\mathbf{E}^i(\mathbf{r}')$ by $\int_V \mathrm{d}^3r''\overleftrightarrow{\Xi}(\mathbf{r}',\mathbf{r}'')\mathbf{E}^i(\mathbf{r}'')$, which directly follows from Eq. (10).
- Replace $\overleftrightarrow{\Xi}(\mathbf{r}')\mathbf{E}^i(\mathbf{r}')$ by $\overleftrightarrow{\Xi}(\mathbf{r},\mathbf{r}')\mathbf{E}^i(\mathbf{r}')$, which would suffice since $\overleftrightarrow{\Xi}(\mathbf{r},\mathbf{r}')$ is further combined with $g(\mathbf{r},\mathbf{r}')$ under the integral.

However, any of these modifications would make further derivations invalid.

(2) Eq. (12) can be rewritten as $\int_V \mathrm{d}^3r'\mathbf{H}(\mathbf{r},\mathbf{r}')\mathbf{E}_0 = \mathbf{0}$, which is correct for any $\mathbf{E}_0$ and $\mathbf{r}$. The authors of [1] stated that this implies that $\mathbf{H}(\mathbf{r},\mathbf{r}') = \mathbf{0}$ for any $\mathbf{r}$ and $\mathbf{r}'$. However, this would be correct only if $\mathbf{E}_0$ is an arbitrary function of $\mathbf{r}'$. Instead, since $\mathbf{E}_0$ is just an arbitrary constant vector, the only correct implication is that not the integrand, but the whole integral is zero, i.e., $\int_V \mathrm{d}^3r'\mathbf{H}(\mathbf{r},\mathbf{r}') = \mathbf{0}$. The latter is a weaker statement that cannot be used for direct solution of the integral equation for the electric field.

Probably the second issue can be corrected by more elaborate use of (incorrect) Eq. (9), but this surely would not make the whole derivation valid. Moreover, it should be noted that the idea of general solution is not original to [1], but was proposed by Bozhevolnyi et al. [3, 4]. The details of derivation slightly differ between these papers, but each of them has serious flaws as well. In [4], the error is similar to the second issue above. The derivation of Eq. (28) heavily depends on Eq. (27), stating that $E_i^{(A)}(\mathbf{R}) = E_{0i}^{(A)}e^{\mathbf{ikR}}$. Therefore, the text immediately after Eq. (28): "$E_i^{(A)}(\mathbf{R}')$ is an arbitrary function of variable $\mathbf{R}'$" is incorrect, since Eq. (28) is valid only for $E_i^{(A)}(\mathbf{R}')$ described by Eq. (27), but not for arbitrary one. Then, the integrand does not have to be zero, but only integrate (with exponents) to a zero tensor. In [3], Eq. (23) is erroneous—$E_n^{(A)}(\vec{R}',\Omega)$ cannot be factored out, since in the second term this field is originally inside the second integral, depending on another positional variable $\vec{R}''$. While the order of integration in the double integral can be interchanged, this would also change the order of g and D, breaking all further implications.

Coming back to [1], the erroneous derivation affects only part of this paper. It is used to derive solution in the Rayleigh regime and corrections based on retardation (Sections 2.2 and 2.3). Although corresponding formulae are, at least, not proved, they may provide a certain approximation to the exact light-scattering quantities. Unfortunately, the accuracy of these approximations is unclear, because the authors have not studied it in detail. The only presented result of using these formulae is shown in Fig. 3 without comparison with other (rigorous) methods. In particular, the presented approach is valid for very small ellipsoids (as discussed above). Moreover, it is expected to be approximately correct for dielectric particles with rounded shapes, inside which the electric field is approximately constant. However, the accuracy of such approach for a particular system is hard to estimate a priori. The rest of paper [1], including abstract and conclusions, is unaffected by the error uncovered by this comment.

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