

ON THE IMPORTANCE OF LOCAL-FIELD CORRECTIONS FOR POLARIZABLE PARTICLES ON A FINITE LATTICE: APPLICATION TO THE DISCRETE DIPOLE APPROXIMATION

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Received 2003 November 27; accepted 2004 February 16

ABSTRACT

We investigate the influence of local-field effects on the electromagnetic response of a collection of dipoles. We derive the local-field corrected static polarizability for a collection of dipoles in the case of a scatterer with uniform depolarization. We then use this correction within the discrete dipole approximation to study the scattering of an electromagnetic wave by a spherical particle. The local-field correction leads to a new formulation of the discrete dipole approximation that is exact in the long-wavelength limit and more accurate at finite frequencies. We also discuss the feasibility of a generalization of the local-field correction to arbitrary scatterers.

Subject headings: dust, extinction — scattering

1. INTRODUCTION

The scattering of electromagnetic waves by irregular objects can be addressed from many viewpoints. For simple or special configurations, analytical solutions can be found. However, in the majority of cases the scattering problem is not amenable to an analytic solution and numerical methods must be used. Many computational approaches exist, relying on different strategies (Mishchenko et al. 2000; Kahnert 2003). Among these methods one finds volume integral methods, of which the discrete dipole approximation (DDA) is a discretized version. The DDA was introduced by Purcell & Pennypacker (1973) to study the scattering of light by interstellar dust grains with arbitrary shapes. Dust grains can alter the electromagnetic signature of stars and galaxies, some wavelengths being attenuated or, conversely, strengthened by the scattering process (Draine 2003). An accurate description of the scattering of light by arbitrary dust grains is therefore an essential part of the astrophysics of the interstellar medium. The theoretical foundation of the DDA stems from a simple observation. When an object interacts with an electromagnetic field, it develops a polarization. If one considers a small enough volume inside the object, the induced polarization will be uniform within this volume and hence that small region can be represented by an electric dipole. Accordingly, in the DDA the scatterer is discretized over a cubic lattice and its electromagnetic properties are described by those of a collection of coupled electric dipoles. Therefore, the central quantity in the DDA is the electric polarizability associated with the dipoles (polarizable regions forming the scatterer).

The original formulation of the DDA (Purcell & Pennypacker 1973) used the Clausius-Mossotti (CM) polarizability. However, the CM polarizability is only exact in the long-wavelength (static) regime, and for an isolated dipole in free-space (or a dipole in an infinite lattice). Consequently, problems such as the violation of the optical theorem arise when the DDA is

used at finite frequencies, a critical issue for the calculation of absorption cross sections or optical forces and torques (Draine & Weingartner 1996; Chaumet & Nieto-Vesperinas 2000; Chaumet et al. 2002). Subsequent formulations of the DDA improved on the CM polarizability by accounting for retardation and propagation effects (Draine & Flatau 1994 and references therein). For instance, Draine (1988) introduced a radiation-reaction correction to the CM polarizability, thereby ensuring that the optical theorem is satisfied (i.e., the total electromagnetic energy is conserved). Later, Draine & Goodman (1993) introduced the lattice dispersion relation (LDR) correction to derive a polarizability such that the lattice would reproduce the propagation properties of a continuum. Other forms of the polarizability were also proposed in order to improve the performance of the DDA at finite frequencies (Dungey & Bohren 1991; Lakhtakia 1992). A point worth emphasizing is that although several prescriptions exist for the polarizability, *they all reduce to the Clausius-Mossotti expression in the long-wavelength (static) limit*. In other words, it has been widely accepted that the CM polarizability was the correct starting point and that any improvement of the DDA in describing electromagnetic scattering has to come from an improvement of how dynamic (i.e., finite frequency) effects are accounted for.

However, in his study of light scattering by spherical particles Draine (1988) pointed out some discrepancies between the DDA and the exact Mie calculation in the long-wavelength limit. These discrepancies are most noticeable for large values of $|n - 1|$ (n being the complex refractive index of the sphere), and are not due to a mere convergence issue since increasing the number of dipoles does not solve the problem. In the long-wavelength regime radiative corrections are irrelevant; therefore, the problem pointed out by Draine suggests that the conventional form of the DDA overlooks some fundamental issues that exist in the long-wavelength regime.

While this long-wavelength anomaly may hinder high-precision scattering calculations at any wavelength (the

finer the discretization, the closer to the long-wavelength limit), it will have particularly dramatic consequences in the infrared (IR) part of the electromagnetic spectrum ($1 \mu\text{m} < \text{wavelength} < 300 \mu\text{m}$). Indeed, consider the scattering of electromagnetic radiation by interstellar dust particles. In the IR region, two effects will contribute to the dramatic increase of the long-wavelength anomaly. First, for a given size of the scatterer as the wavelength gets larger one moves deeper into the long-wavelength regime. The second effect results from the large refractive index (and the usually concomitant large absorption) that most materials exhibit as the wavelength increases, which demands a fine discretization in the DDA and hence ensures again that the long-wavelength regime will be attained. These two effects can drastically reduce the accuracy of the DDA, putting the method in a delicate situation. A coarse discretization will fail at describing accurately the scattering properties of the particle. But on the other hand, a fine discretization will hit the long-wavelength anomaly. The only way to overcome this problem is by understanding the physics of the scattering of electromagnetic waves by a collection of dipoles in the long-wavelength regime.

In this paper, we address the problem of the static polarizability for a dipole in a finite lattice, by deriving an expression of the polarizability that takes into account the particular environment of each dipole. Because the correction is made on the static polarizability, the usual radiative corrections, such as the LDR, still apply. We illustrate the relevance of local-field corrections by computing the scattering properties of a spherical particle and comparing them to the exact Mie results. We also discuss the possibility of extending this approach to arbitrary scatterers.

2. DERIVATION OF A STATIC POLARIZABILITY THAT ACCOUNTS FOR LOCAL-FIELD EFFECTS

The idea behind our derivation is quite simple. When a static electric field is applied to a collection of small polarizable particles, each particle will develop an induced dipole moment that will depend on the applied field but also on the field resulting from all other induced dipoles. This local-field effect will be responsible for all particle not having an identical polarization, which in other terms means that not all particles will have the same effective polarizability. The simplest way to see this is to consider a slab of matter discretized over a cubic lattice with infinite extension along directions x and y , and a finite thickness, larger than a few layers, along z . From symmetry considerations, a dipole near the center of the slab should have equal polarizabilities along directions x , y , and z . On the other hand, for a dipole at the surface of the slab one should expect that the response (polarizability) to an applied field within the xy plane will differ from the response to a field applied along z . We now express this idea in a more formal way. For the sake of clarity we reproduce here the derivation of the static polarizability as described in Rahmani et al. (2002). However, we will make an essential distinction between two classes of scattering objects that was not made in Rahmani et al. (2002). The first class, with uniform depolarization (response of the material due to polarization charges when an electric field is applied), is discussed in the next section. The second class, with nonuniform depolarization, will be discussed in § 3.2.

2.1. Class A: Objects with Uniform Depolarization

From a mathematical viewpoint, the first class of objects comprises shapes for which the depolarization tensor is

uniform over the volume of the object. The depolarization tensor \mathbf{L} can be viewed as a geometrical factor whose value is determined by the shape of the object (Yaghjian 1980). From a physical viewpoint, these are objects that respond to a uniform static electric field by exhibiting a uniform electric polarization (not necessarily parallel to the initial field). This class includes objects such as slabs, infinite cylinders, spheres, and spheroids. Consider a class A homogeneous scatterer with permittivity ϵ (assumed to be scalar for simplicity), whose electromagnetic properties are approximated by a set of N electric dipoles with electric polarizability α_i ($i = 1, N$), arranged on a cubic lattice with spacing d . The self-consistent local field at subunit i is

$$\mathbf{E}_i^{\text{loc}}(\omega) = \mathbf{E}_i^0(\omega) + \sum_{j \neq i} \mathbf{F}_{ij}(\omega) \alpha_j(\omega) \mathbf{E}_j^{\text{loc}}(\omega). \quad (1)$$

$\mathbf{E}_i^0(\omega)$ is the incident field at subunit i , $\mathbf{F}(\omega)$ is the free-space field susceptibility (Green tensor), and ω is the angular frequency of the electromagnetic wave. The sum over j runs over all the subunits forming the scatterer. Note that the term $j = i$ is not included in the sum; this term is automatically dealt with by accounting for the finite volume of the dipoles.

We make the electrostatic approximation ($\omega = 0$; the angular frequency will be omitted in the equations henceforth) and consider a uniform applied field \mathbf{E}^0 . In the case where the macroscopic field \mathbf{E}^m is uniform over the lattice, it can be related to the applied field through the (uniform) depolarization tensor \mathbf{L} such that

$$\left(1 + \frac{\epsilon - 1}{4\pi} \mathbf{L}\right) \mathbf{E}^m = \mathbf{E}^0, \quad (2)$$

where $\mathbf{1}$ is the identity tensor. Once the macroscopic field is introduced in equation (1) we obtain

$$\mathbf{E}_i^{\text{loc}} = \left(1 + \frac{\epsilon - 1}{4\pi} \mathbf{L}\right) \mathbf{E}^m + \sum_{j \neq i} \mathbf{F}_{ij} \alpha_j \mathbf{E}_j^{\text{loc}}. \quad (3)$$

By definition, the local-field tensor $\mathbf{\Lambda}_i$ satisfies

$$\mathbf{E}_i^{\text{loc}} = \mathbf{\Lambda}_i \mathbf{E}^m. \quad (4)$$

Using the fact that the (uniform) polarization can be written as

$$\mathbf{P} = \frac{\alpha_i}{d^3} \mathbf{E}_i^{\text{loc}} = \frac{\epsilon - 1}{4\pi} \mathbf{E}^m = \frac{\epsilon - 1}{4\pi} \mathbf{\Lambda}_i^{-1} \mathbf{E}_i^{\text{loc}}, \quad (5)$$

we can express the polarizability in terms of the local-field tensor:

$$\alpha_i = \frac{\epsilon - 1}{4\pi} \mathbf{\Lambda}_i^{-1} d^3. \quad (6)$$

The local-field tensor is derived by using equations (4)–(6) to express the local field in terms of the macroscopic field in equation (3), leading to

$$\mathbf{\Lambda}_i = \mathbf{1} + \frac{\epsilon - 1}{4\pi} \mathbf{L} + \sum_{j \neq i} \mathbf{F}_{ij} \frac{\epsilon - 1}{4\pi} d^3. \quad (7)$$

The local-field corrected static polarizability (LFCSP) of equation (6) reduces to the CM expression only for a single,

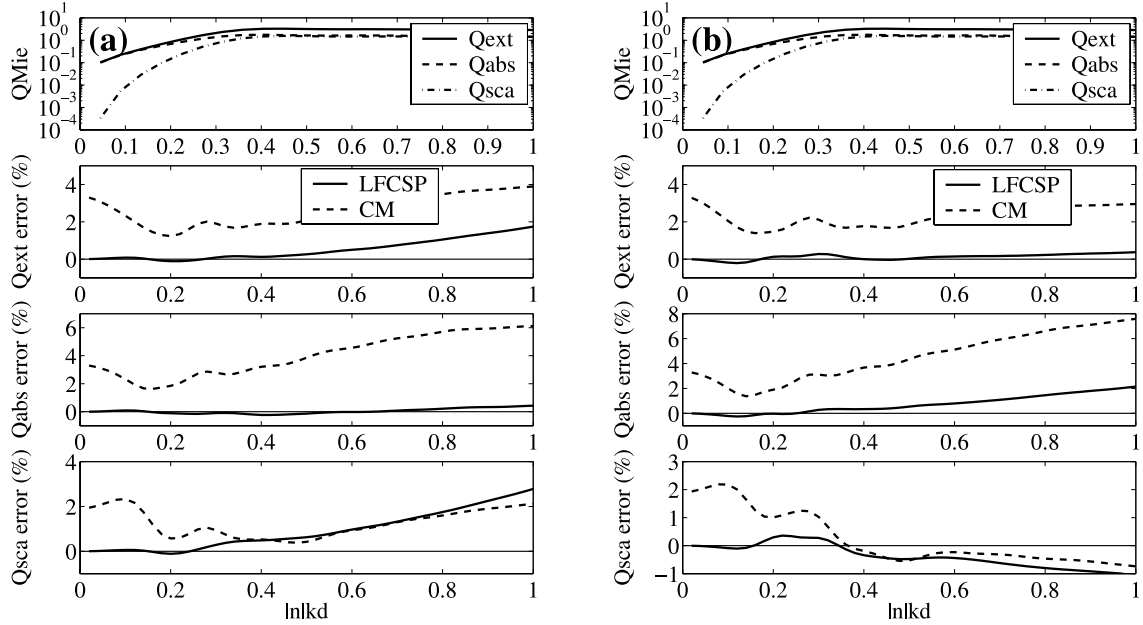


FIG. 1.—Scattering properties of a pseudosphere with $N = 17904$ and $\epsilon = n^2 = 5 + 5i$. The incident wave vector is along direction $[1\ 1\ 1]$ of the lattice. The electric field is polarized along direction $[2\ \bar{1}\ 1]$ of the lattice. From top to bottom the figure shows extinction, absorption, and scattering efficiencies (Q_{ext} , Q_{abs} , and Q_{sca} ; top panel) as given by Mie as a function of the normalized lattice spacing $|n|kd$, where k is the free-space wave vector of the incident light; and the relative error in the DDA computed values of the extinction, absorption, and scattering efficiencies (lower three panels). The static polarizability (dashed lines) is defined according to the Clausius-Mossotti relation. The static polarizability (solid lines) is defined according to eq. (6). A dynamic correction is applied to both forms of the static polarizability. (a) Lattice dispersion relation (LDR); (b) radiation reaction.

isolated dipole in free space, or a dipole in an infinite lattice (for any practical purpose, a dipole more than a few lattice sites away from any interface can in general be considered as immersed in an infinite lattice).

We emphasize that our correction affects the static polarizability, i.e., the LFCSP prescription is used as a replacement of the CM expression. Consequently, previously derived corrections that account for radiation-reaction (Draine 1988) or propagation effects (Draine & Goodman 1993) can be applied to the LFCSP.

Rahmani et al. (2002) illustrated the relevance of the LFCSP by computing the field inside and outside a slab. Note that the slab was actually treated as a three-dimensional problem using a generalization of the DDA to periodic systems (Chaumet et al. 2003). This made it possible to use the conventional, three-dimensional expressions for the field susceptibility and the polarizability. Using the example of a slab, we showed that accounting for the influence of the local environment of the dipoles on their electromagnetic response led to a more accurate estimate of the macroscopic field inside the slab as well as a more accurate calculation of reflected and transmitted fields.

However, it is of interest to test the prescription of equations (6) and (7) on a fully three-dimensional scatterer. In the next section, we consider the case of a spherical scatterer.

2.2. Example: Light Scattering by a Homogeneous Spherical Particle

The case of a homogeneous spherical scatterer is of particular interest because an analytic solution is known in the form of a Mie series. We shall use the Mie result as our reference in the computation of the scattering properties of spheres. The Mie scattering numerical code we use is the Bohren-Huffman Mie scattering subroutine modified by Draine. Our DDA light-scattering code is derived from our DDA spontaneous

emission code (Rahmani et al. 2001; Rahmani & Bryant 2002) by solving for the electric field instead of the field susceptibility. The local field at each lattice site is found by solving a linear system using the QMR iterative solver developed by Freund & Nachtigal (1991). For free-space computations, we perform matrix-vector multiplications using the Temperton fast Fourier transform routine (FFT; Temperton 1992) as implemented in the DDSCAT code of Draine and Flatau.¹

For a homogeneous sphere with permittivity ϵ , the depolarization tensor \mathbf{L} is constant within the volume of the sphere and equal to $4\pi/3$ (note that our definition of \mathbf{L} and that of Yaghjian 1980 differ by a factor 4π). Following equation (7), the local-field tensor becomes

$$\Lambda_i = \frac{\epsilon + 2}{3} + \frac{\epsilon - 1}{4\pi} d^3 \sum_{j \neq i} \mathbf{F}_{ij}, \quad (8)$$

where the sum involves the static limit of the free space field susceptibility. It is of interest to note that d^3 times the sum is a finite lattice sum that depends only on the relative position of the dipoles on the lattice. Therefore, the sum pertaining to a homogeneous sphere with a given number N of dipoles need not be computed again if the wavelength of the incident light, the permittivity of the sphere, the lattice spacing or any other parameter is changed.

We plot in Figures 1, 2, and 3 the relative error (using Mie as a reference) for the extinction, absorption, and scattering efficiencies, as a function of $|n|kd$ where n is the complex refractive index of the sphere, k the magnitude of the wave

¹ DDSCAT is a software package developed by Draine and Flatau that applies the DDA to calculate scattering and absorption of electromagnetic waves by targets with arbitrary geometries and complex refractive index. DDSCAT is available at <http://www.astro.princeton.edu/~draine/DDSCAT.6.0.html>. Note that the routines we use come from ver. 5.10 of DDSCAT.

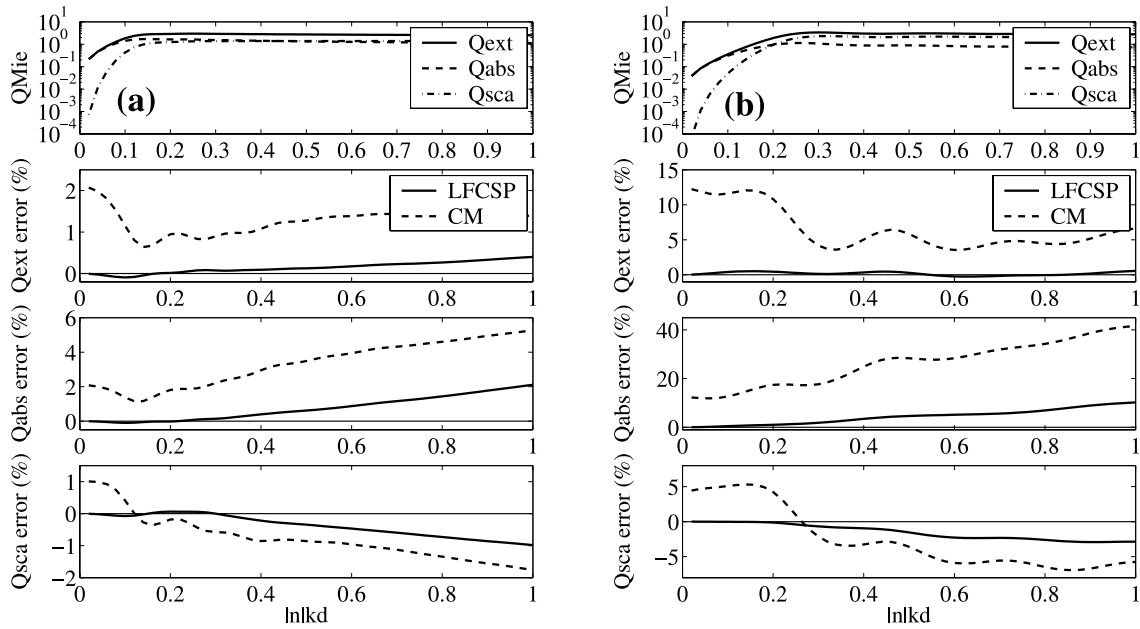


FIG. 2.—Same as Fig. 1, but for (a) silicate at $20 \mu\text{m}$ ($\epsilon = n^2 = 2.3 + 4i$) and (b) silicon carbide at $12 \mu\text{m}$ ($\epsilon = n^2 = -9 + 20i$). The radiation-reaction correction is used in both cases.

vector in vacuum of the incident light, and d the DDA lattice spacing. The scatterer is a pseudosphere composed of $N = 17,904$ dipoles. The plots in all the figures are for an incident field with a wave vector parallel to direction $[111]$ of the lattice and polarized along direction $[2\bar{1}1]$. Other combinations of propagation direction and polarization lead to similar results for the effectiveness of the LFCSP prescription. The extinction and absorption efficiencies are calculated from the respective cross sections (Draine 1988, eqs. [3.02] and [3.06]). The scattering efficiency is computed as the difference of the two other efficiencies after we check that it yields a value in agreement with what can be found by a direct computation (Draine 1988, eq. [3.07]).

A good assessment of the relevance of the LFCSP prescription can be made by considering a scatterer with $|\epsilon^2 - 1| > 1$. We consider in Figures 1a and 1b a sphere with permittivity $\epsilon = 5 + 5i$. The two figures differ by the nature of the radiative correction. On Figure 1a, the LDR prescription is used (in addition to the “static prescription”; either CM or LFCSP), whereas the radiation-reaction corrective term is used in Figure 1b. We consider two dynamic correction terms merely to illustrate the fact that the LFCSP leads to an improvement of the accuracy of the DDA irrespective of the specifics of the additional correction procedure used to account for finite frequency effects. The overall effect of the LFCSP is to bring the DDA computation closer to the Mie result. In

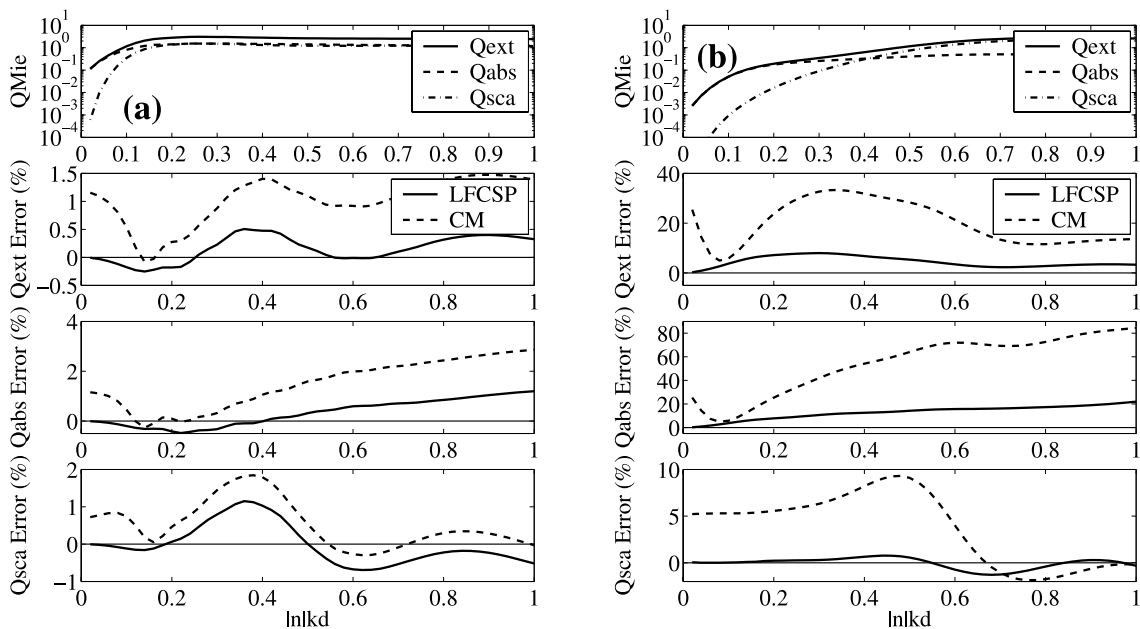


FIG. 3.—Same as Fig. 1, but for graphite at $20 \mu\text{m}$. (a) Electric field parallel to c -axis, $\epsilon = n^2 = 3 + 1.4i$; (b) electric field perpendicular to c -axis, $\epsilon = n^2 = -34 + 140i$. The radiation-reaction correction is used.

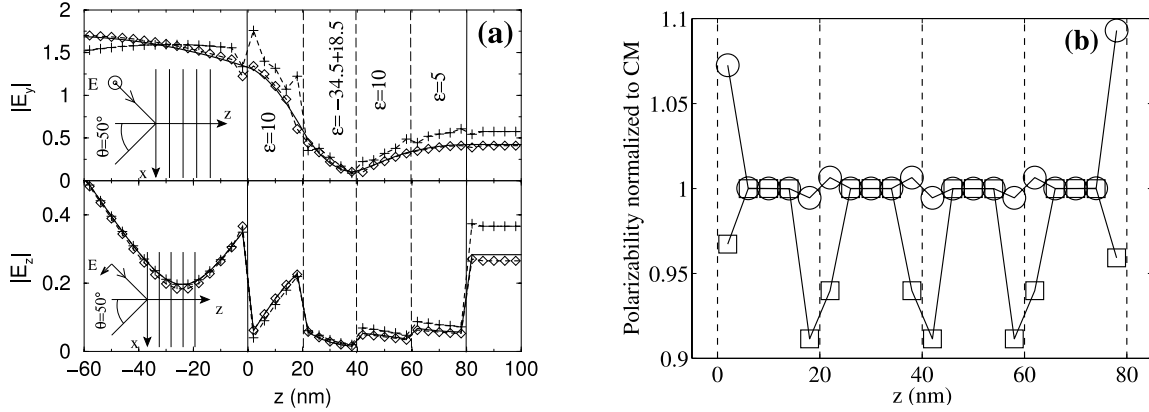


FIG. 4.—(a): Reflected, internal, and transmitted electric field for a multilayered slab. The polarization of the incident light is shown on the figures. *Solid line*: exact result; *dashed line with crosses*: DDA using CM; *dashed line with diamonds*: DDA with LFCSP. The radiation-reaction prescription is applied to both forms of the static polarizability. (b): Plot of the xx (squares) and the zz (circles) components of the LFCSP normalized to the CM result for the structure of Fig. 3a. The dashed vertical lines mark the interfaces.

particular, the tendency of the DDA to overestimate the absorption is noticeably moderated by the use of the LFCSP. Another striking feature is that the local-field corrected DDA converges toward the exact result, whereas the conventional DDA (using the CM polarizability) converges toward a result with a relative error of a few percent in the long-wavelength limit ($|n|kd \rightarrow 0$). We emphasize that this does not depend on the permittivity of the sphere; the local-field correction of equation (8) was designed precisely to yield the exact result in the static limit. This point is illustrated further in the next figures. As we mentioned previously, the DDA might have great difficulties at describing accurately scattering processes in the infrared region of the spectrum. Figures 2a and 2b pertain to a spherical particle with permittivities $\epsilon = n^2 = 2.3 + 4i$ and $\epsilon = n^2 = -9 + 20i$, respectively. These rounded-off values are close to the permittivity of silicate around $20 \mu\text{m}$ and that of silicon carbide around $12 \mu\text{m}$ (Laor & Draine 1993). Figures 3a and 3b pertain to a spherical particle with permittivities $\epsilon = n^2 = 3 + 1.4i$ and $\epsilon = n^2 = -34 + 140i$, respectively, which corresponds to graphite around $20 \mu\text{m}$ with the electric field parallel (Fig. 3a) or perpendicular (Fig. 3b) to the c -axis (Laor & Draine 1993).

One can see that, irrespective of the permittivity, the LFCSP prescription leads to a higher accuracy of the DDA. Most striking is the fact that the DDA using our new prescription *always* converges toward the exact result in the long-wavelength limit. Moreover, we see again that the LFCSP lessens the tendency of the DDA to overestimate absorption. This is most noticeable in Figure 3b, where the imaginary part of the permittivity is large.

These examples demonstrate the dramatic effect that the local-field correction has on the computation of the electromagnetic properties of a sphere with the DDA, despite the fact that the correction is significant only for the dipoles at the surface of the sphere. In the next section, we discuss the possibility of a generalization of this approach to arbitrary scatterers.

3. SCATTERERS WITH ARBITRARY SHAPE

3.1. Class A: Objects with Uniform Depolarization

For this class of objects, the LFCSP is readily found using equation (7) with the appropriate value of \mathbf{L} in place of the $4\pi/3$ value for a sphere. For objects from this class, or

sometimes a collection of objects as illustrated in Figure 4a, the use of the LFCSP prescription will make the DDA converge toward the exact result in the long-wavelength limit and should improve significantly the performances of the DDA at finite frequencies, not only for far-field computations but for internal fields as well. We emphasize again that the LFCSP will differ from the CM polarizability only near interfaces. To illustrate this point, we plot in Figure 4b the xx (squares) and zz (circles) components of the LFCSP, normalized to the CM result, for the multilayered structure of Figure 4a. Figure 4b shows that only the dipoles at the interfaces have optical responses that depart notably from the CM prescription.

3.2. Class B: Objects with Nonuniform Depolarization

This class comprises objects that would exhibit a nonuniform polarization when placed in a uniform static field. One of the simplest examples of such an object is a dielectric cube. For this class of objects, the derivation described above or in Rahmani et al. (2002) is not valid.²

While investigating the possibility of extending the LFCSP to class B objects, we found that Karam (1997) derived the general expression for the macroscopic field at position \mathbf{r} inside an arbitrary object placed in a static field \mathbf{E}^0 . The macroscopic field reads

$$\mathbf{E}^m(\mathbf{r}) = \mathbf{E}^0(\mathbf{r}) - \frac{(\epsilon - 1)}{4\pi} \int_S \frac{\hat{\mathbf{R}} ds'}{|\mathbf{r} - \mathbf{r}'|^2} \mathbf{E}^m(\mathbf{r}'), \quad (9)$$

where ϵ is the permittivity of the object, $\hat{\mathbf{R}} = (\mathbf{r} - \mathbf{r}')/|\mathbf{r} - \mathbf{r}'|$, and the integration is performed over the surface S enclosing the object. Note that when the macroscopic field is uniform, equation (9) reduces to equation (2), with the depolarization tensor given by

$$\mathbf{L} = \int_S \frac{\hat{\mathbf{R}} ds'}{|\mathbf{r} - \mathbf{r}'|^2}. \quad (10)$$

² Note that because the necessity of a uniform depolarization was not explicit, eq. (8) in Rahmani et al. (2002) is misleading as it does not include the simplifications that result from the depolarization tensor being uniform. However, it is clear that eq. (8) is not valid for a class B object since it would yield a nonsymmetric static polarizability; a physical impossibility.

Therefore, the obstacle in deriving the LFCSP for a class B objects lies in the fact that a simple term of proportionality between the incident and the macroscopic field cannot be found. However, it will be of great interest to investigate the possibility of estimating numerically the surface effect of equation (9) in order to derive a more appropriate expression of the static polarizability when the DDA is used to represent class B objects. We are currently working to extend the LFCSP to this class of objects.

4. CONCLUSION

We have considered an electric dipole on a lattice and we have shown that its electrostatic response (static polarizability) depends on its local environment (local-field effect). From this observation, we have derived a new prescription for the static polarizability to be used in the DDA in place of the Clausius-Mossotti expression. Our general derivation is valid for any value of the refractive index and always converging to the exact result in the long-wavelength limit. We

also showed that, at finite frequencies, the usual radiative corrections to the polarizability can still be used. As a result, when the DDA is used to model light scattering by a spherical scatterer, the accuracy of the computation is globally enhanced and the long-wavelength problem pointed out by Draine (1988) is resolved. Moreover, our prescription improves significantly the description of strongly absorbing material. The derivation of the static polarizability presented in this work is valid for any scatterer whose depolarization tensor, and by extension macroscopic field, is uniform. For other shapes, there is no simple factor of proportionality between the macroscopic field and the incident field, which hinders the derivation of a static polarizability that account for local-field effects. However, as we have shown, the local-field correction can lead to significant improvements in the modeling of scattering and internal properties of scatterers, particularly in the infrared region. Therefore, even an approximate solution of the local-field problem is worth investigating.

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