

Generalization of the coupled dipole method to periodic structures

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We present a generalization of the coupled dipole method to the scattering of light by arbitrary periodic structures. This formulation of the coupled dipole method relies on the same direct-space discretization scheme that is widely used to study the scattering of light by finite objects. Therefore, all the knowledge acquired previously for finite systems can be transposed to the study of periodic structures.

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I. INTRODUCTION

In its original form, the coupled dipole method (CDM) was developed for the study, in free space, of the scattering of light by an object with finite dimensions.^{1,2} The method was subsequently extended to deal with objects near a substrate^{3,4} or inside a multilayer system.⁵ The principle of the method is always the same: the object is represented by a cubic array of N polarizable subunits, each with a size small enough compared to the spatial variations of the electromagnetic field for the dipole approximation to apply. If the CDM could be extended to deal with local scatterers near periodic structures, the CDM could then also be used, for example, to study light scattering by objects near surface gratings or by defects or cavities in photonic crystals. The first step toward such an extension is to develop a form of the CDM capable of describing periodic structures efficiently. In this paper, we present a generalization of the CDM to arbitrary periodic structures.

II. SELF-CONSISTENT FIELD FOR A PERIODIC STRUCTURE

We consider a plane substrate occupying the region $z \leq 0$. For a single object on the substrate, the self-consistent field at the i th subunit at location \mathbf{r}_i is given by

$$\mathbf{E}(\mathbf{r}_i, \omega) = \mathbf{E}_0(\mathbf{r}_i, \omega) + \sum_{j=1}^N [\mathbf{S}(\mathbf{r}_i, \mathbf{r}_j, \omega) + \mathbf{F}(\mathbf{r}_i, \mathbf{r}_j, \omega)] \alpha_j(\omega) \mathbf{E}(\mathbf{r}_j, \omega), \quad (1)$$

where $\mathbf{E}_0(\mathbf{r}_i, \omega)$ is the (initial) field at \mathbf{r}_i in the absence of the scattering object. Note that none of the subunits lies in the plane $z=0$. The tensors \mathbf{F} and \mathbf{S} are the field susceptibilities (linear responses) associated with the free space⁶ and the substrate.⁷ Note that the diagonal term of the free-space susceptibility $\mathbf{F}(\mathbf{r}_i, \mathbf{r}_i, \omega)$ is excluded from the sum in Eq. (1)

because this term is automatically taken into account in the dynamic polarizability $\alpha_i(\omega)$ (see, for instance, Ref. 8). The dynamic polarizability accounts for radiation reaction as well.^{2,9} The self-consistent field $\mathbf{E}(\mathbf{r}_i, \omega)$ is found by solving the symmetric linear system formed by writing Eq. (1) for $i=1, \dots, N$. The total field at position \mathbf{r} is computed as

$$\mathbf{E}(\mathbf{r}, \omega) = \mathbf{E}_0(\mathbf{r}, \omega) + \sum_{j=1}^N [\mathbf{S}(\mathbf{r}, \mathbf{r}_j, \omega) + \mathbf{F}(\mathbf{r}, \mathbf{r}_j, \omega)] \alpha_j(\omega) \mathbf{E}(\mathbf{r}_j, \omega). \quad (2)$$

This conventional form of the CDM is well adapted to deal with localized objects. If, instead of a single object, one wants to study a periodic structure created by the repetition of the object over a lattice located above the substrate, Eq. (1) becomes

$$\begin{aligned} \mathbf{E}(\mathbf{r}_i, \omega) = & \mathbf{E}_0(\mathbf{r}_i, \omega) + \sum_{j=1}^N \sum_{m,n=-\infty}^{\infty} [\mathbf{S}(\mathbf{r}_i, \bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega) \\ & + \mathbf{F}(\mathbf{r}_i, \bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega)] \alpha_j(\omega) \\ & \times \mathbf{E}(\bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega). \end{aligned} \quad (3)$$

The vectors \mathbf{u} and \mathbf{v} are the basis vectors of the lattice (Fig. 1). The index i runs over all the subunits of the structure. \mathbf{r}_i is the position of subunit i . The sum over j is restricted to the N subunits of a single object with position $\bar{\mathbf{r}}_j$ inside the object. The number of subunits is now infinite, and therefore so is the size of the linear system to be solved. One solution would be to truncate the infinite sum and solve the system for a large but finite number of objects, but this is impractical because the sums over the lattice converge very slowly. This problem can be circumvented by using a plane-wave decomposition of the incident field. In the case of plane-wave (propagating or evanescent) illumination, the field above the surface can be written as (we note by $\mathbf{k}_{0\parallel}$ the projection of vector \mathbf{k}_0 on a plane parallel to the surface)

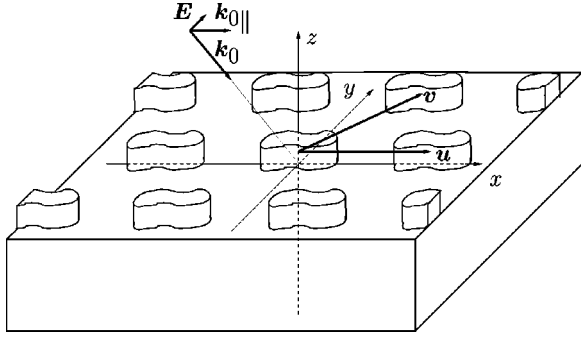


FIG. 1. Example of a periodic structure created by the repetition of an object over a lattice parallel to a substrate.

$$\mathbf{E}_0(\bar{\mathbf{r}}_i + m\mathbf{u} + n\mathbf{v}, \omega) = \mathbf{E}_0(\bar{\mathbf{r}}_i, \omega) \exp[i\mathbf{k}_{0\parallel} \cdot (m\mathbf{u} + n\mathbf{v})], \quad (4)$$

where \mathbf{k}_0 is the wave vector in free space. Because of the periodicity of the system and the translational invariance of the field susceptibilities, the self-consistent field satisfies the same relation as the incident field [Eq. (4)], and at any subunit Eq. (3) can be written as

$$\begin{aligned} \mathbf{E}(\mathbf{r}_i, \omega) = & \mathbf{E}_0(\mathbf{r}_i, \omega) + \sum_{j=1}^N \left(\sum_{m,n=-\infty}^{\infty} [\mathbf{S}(\mathbf{r}_i, \bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega) \right. \\ & + \mathbf{F}(\mathbf{r}_i, \bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega)] \\ & \left. \times \exp[i\mathbf{k}_{0\parallel} \cdot (m\mathbf{u} + n\mathbf{v})] \right) \alpha_j(\omega) \mathbf{E}(\bar{\mathbf{r}}_j, \omega). \end{aligned} \quad (5)$$

The self-consistent field on the right-hand side of Eq. (5) is independent of (m, n) and can be taken out of the infinite sum. Hence, the sum over subunits in Eq. (5) only involves $j=1, \dots, N$, that is, the number of subunits in a unit cell, which we choose to be the cell for which $m=n=0$. Moreover, because of the translational symmetry of the self-consistent field, we only need to find \mathbf{E} in one cell. Once the self-consistent field is found in the central cell, the field in any other cell is obtained by multiplying by the appropriate phase factor. Thus we only have to solve a linear system of the same size as the one describing a single object. The major issue in solving Eq. (5) is to compute efficiently the infinite, slowly convergent sums without performing a truncation of the sums. This is possible owing to the translational invariance of the field susceptibilities in a plane parallel to the surface. The dependence on $(\bar{\mathbf{r}}_i, \bar{\mathbf{r}}_j, \omega)$ can be written as $(\boldsymbol{\rho}_{ij}, z_i, z_j, \omega)$ with $\boldsymbol{\rho}_{ij} = (\bar{\mathbf{r}}_i - \bar{\mathbf{r}}_j)_{\parallel}$. Hence, the infinite sums of Eq. (5) become

$$\begin{aligned} K = & \sum_{m,n=-\infty}^{\infty} [\mathbf{S}(\bar{\mathbf{r}}_i, \bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega) + \mathbf{F}(\bar{\mathbf{r}}_i, \bar{\mathbf{r}}_j + m\mathbf{u} + n\mathbf{v}, \omega)] \\ & \times \exp[i\mathbf{k}_{0\parallel} \cdot (m\mathbf{u} + n\mathbf{v})] \\ = & \int d\mathbf{r}_{\parallel} \sum_{m,n=-\infty}^{\infty} \delta(\mathbf{r}_{\parallel} - m\mathbf{u} - n\mathbf{v}) \exp(i\mathbf{k}_{0\parallel} \cdot \mathbf{r}_{\parallel}) \\ & \times [\mathbf{S}(\boldsymbol{\rho}_{ij} - \mathbf{r}_{\parallel}, z_i, z_j, \omega) + \mathbf{F}(\boldsymbol{\rho}_{ij} - \mathbf{r}_{\parallel}, z_i, z_j, \omega)]. \end{aligned} \quad (6)$$

We define the two-dimensional Fourier transform as $\mathcal{F}[b(\mathbf{r}_{\parallel})] = \int d\mathbf{r}_{\parallel} b(\mathbf{r}_{\parallel}) \exp(-i\mathbf{r}_{\parallel} \cdot \mathbf{h}_{\parallel})$, and its inverse as $\mathcal{F}^{-1}[B(\mathbf{h}_{\parallel})] = [1/(2\pi)^2] \int d\mathbf{h}_{\parallel} B(\mathbf{h}_{\parallel}) \exp(i\mathbf{r}_{\parallel} \cdot \mathbf{h}_{\parallel})$. Using the Parseval-Plancherel theorem, Eq. (6) becomes

$$\begin{aligned} K = & \frac{1}{(2\pi)^2} \int d\mathbf{h}_{\parallel} M \sum_{m,n=-\infty}^{\infty} \delta(\mathbf{h}_{\parallel} - m\mathbf{u}' - n\mathbf{v}' + \mathbf{k}_{0\parallel}) \\ & \times \mathcal{F}[\mathbf{S}(\boldsymbol{\rho}_{ij} - \mathbf{r}_{\parallel}, z_i, z_j, \omega) + \mathbf{F}(\boldsymbol{\rho}_{ij} - \mathbf{r}_{\parallel}, z_i, z_j, \omega)], \end{aligned} \quad (7)$$

where

$$\mathbf{u}' = 2\pi(v_y \hat{\mathbf{x}} - v_x \hat{\mathbf{y}}) / (u_x v_y - v_x u_y)$$

and

$$\mathbf{v}' = 2\pi(-u_y \hat{\mathbf{x}} + u_x \hat{\mathbf{y}}) / (u_x v_y - v_x u_y)$$

are the basis vectors of the reciprocal lattice, and $M = (2\pi)^2 / (u_x v_y - v_x u_y)$. $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are the basis vectors of the coordinate system. Using the angular spectrum representations \mathbf{W} and \mathbf{G} of tensors \mathbf{S} and \mathbf{F} , Eq. (7) becomes^{6,7}

$$\begin{aligned} K = & \frac{i}{2\pi} M \sum_{m,n=-\infty}^{\infty} \exp[i(m\mathbf{u}' + n\mathbf{v}' + \mathbf{k}_{0\parallel}) \cdot \boldsymbol{\rho}_{ij}] \\ & \times \{ \mathbf{W}(m\mathbf{u}' + n\mathbf{v}' + \mathbf{k}_{0\parallel}, \mathbf{k}_0) \exp[iw_0(z_i + z_j)] \\ & + \mathbf{G}(m\mathbf{u}' + n\mathbf{v}' + \mathbf{k}_{0\parallel}, \mathbf{k}_0) \exp[iw_0|z_i - z_j|] \}, \end{aligned} \quad (8)$$

with

$$\mathbf{G}(\mathbf{k}_{\parallel}, \mathbf{k}_0) = \begin{pmatrix} \frac{k_0^2 - k_x^2}{w_0} & -\frac{k_x k_y}{w_0} & -\gamma k_x \\ -\frac{k_x k_y}{w_0} & \frac{k_0^2 - k_y^2}{w_0} & -\gamma k_y \\ -\gamma k_x & -\gamma k_y & \frac{k_{\parallel}^2}{w_0} \end{pmatrix}, \quad (9)$$

and

$$\mathbf{W}(\mathbf{k}_{\parallel}, \mathbf{k}_0) = \begin{pmatrix} \frac{k_x^2 w_0 \Delta_p}{k_{\parallel}^2} - \frac{k_y^2 k_0^2 \Delta_s}{k_{\parallel}^2 w_0} & \frac{k_x k_y}{w_0 k_{\parallel}^2} (w_0^2 \Delta_p + k_0^2 \Delta_s) & k_x \Delta_p \\ \frac{k_x k_y}{w_0 k_{\parallel}^2} (w_0^2 \Delta_p + k_0^2 \Delta_s) & \frac{k_y^2 w_0 \Delta_p}{k_{\parallel}^2} - \frac{k_x^2 k_0^2 \Delta_s}{k_{\parallel}^2 w_0} & k_y \Delta_p \\ -k_x \Delta_p & -k_y \Delta_p & -\frac{\Delta_p k_{\parallel}^2}{w_0} \end{pmatrix}, \quad (10)$$

where $\gamma = \text{sgn}(z_i - z_j)$, $\mathbf{k}_{\parallel} = m\mathbf{u}' + n\mathbf{v}' + \mathbf{k}_{0\parallel} = k_x\hat{\mathbf{x}} + k_y\hat{\mathbf{y}}$, and w_0 is the component along z of the wave vector \mathbf{k}_0 , i.e., $w_0 = (k_0^2 - k_x^2 - k_y^2)^{1/2}$. Δ_p and Δ_s are the Fresnel reflection coefficients for the substrate. Sums involving different susceptibility tensors (free space or surface) will have a different behavior, due to the different arguments of the exponential terms ($z_i + z_j$ and $|z_i - z_j|$). They will be computed separately.

For the surface term, the convergence of the sum is ensured by the exponential term. As m and n increase, the magnitude of \mathbf{k}_{\parallel} increases and the nature of the plane wave changes from propagating to evanescent. Because $z_i + z_j$ never vanishes, and because the subunits are never exactly on the surface, this exponential term is always present and ensures the rapid convergence of the sum. Hence, the periodic field susceptibility associated to the surface is computed in the reciprocal space.

For the free-space part, the argument of the exponential term is $|z_i - z_j|$ and the rapid convergence of the sums is not as trivial. We use the method introduced to derive the Green function of a two-dimensional (2D) square grating.¹⁰ We consider two cases. The first case pertains to the interaction between elements from different ‘‘layers’’ of the lattice, and corresponds to the case $z_i \neq z_j$. This case is similar to the surface case where the rapid convergence of the sum is ensured by the exponential term. Accordingly, this sum is also computed in the reciprocal space.

In the second case $z_i = z_j$ and the exponential term disappears making the convergence of the sum in the reciprocal space slow. Therefore, another strategy is needed to compute this term efficiently. We cast the free-space part of the infinite sum in two different forms. Let $\mathbf{a}(\mathbf{r}_{\parallel}, z_i - z_j)$ be the sum expressed in direct space [the \mathbf{F} terms in Eq. (6)], and let $\mathbf{A}(\mathbf{k}_{\parallel}, z_i - z_j)$ be the sum in reciprocal space [the \mathbf{G} terms in Eq. (8)]. Note that these two sums represented the same quantity (sum of free-space terms) expressed in two forms, one in direct space and the other in reciprocal space. By combining these two forms we can improve the convergence of the sum in the case $z_i = z_j$. When $z_i = z_j$, we write the sum as

$$\mathbf{a}(\mathbf{r}_{\parallel}, 0) = \mathbf{A}(\mathbf{k}_{\parallel}, h) + [\mathbf{a}(\mathbf{r}_{\parallel}, 0) - \mathbf{a}(\mathbf{r}_{\parallel}, h)], \quad (11)$$

where h is an offset parameter. We emphasize that $\mathbf{A}(\mathbf{k}_{\parallel}, h)$ and $\mathbf{a}(\mathbf{r}_{\parallel}, h)$ represent the same sum expressed in reciprocal and direct space, respectively. The auxiliary sum in the reciprocal space [$\mathbf{A}(\mathbf{k}_{\parallel}, h)$] can be computed efficiently owing to the presence of an exponentially decreasing term. The difference of direct-space sums $\mathbf{a}(\mathbf{r}_{\parallel}, 0) - \mathbf{a}(\mathbf{r}_{\parallel}, h)$ goes as $1/r_{\parallel}^2$ and can also be computed efficiently. With Eq. (11) we can ensure a rapid convergence of the sums in a discretization plane despite the absence of an exponentially decreasing term in the original sum.

To improve further on the convergence of the sums (both the sums in direct space and reciprocal space for the field susceptibility of free space and the surface), we use Shanks’ transformation.¹¹ The principle is the following: consider the sum $S = \sum_{i=1}^{\infty} a_i$. Let us define the partial sum $S_n = \sum_{i=1}^n a_i$ and a new sequence $e_{s+1}(S_n)$ such that

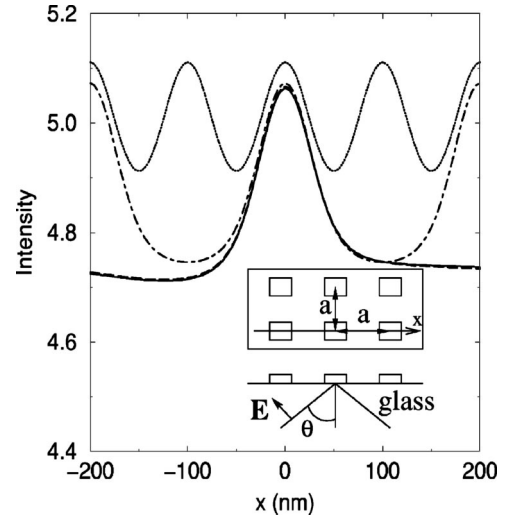


FIG. 2. Intensity of the electric field above a dielectric substrate in the direction of the x axis with a 2D grating of parallelepipeds. The inset shows the geometry used. The solid line is for an isolated parallelepiped. The other curves are obtained for the 2D grating with $a = 100$ nm (dotted line), $a = 200$ nm (dot dashed line), $a = 1000$ nm (dashed line).

$$e_{s+1}(S_n) = e_{s-1}(S_{n+1}) + \frac{1}{e_s(S_{n+1}) - e_s(S_n)} \quad (12)$$

for $s = 1, \dots, n-1$ with

$$e_0(S_n) = S_n, \quad \text{and} \quad e_1(S_n) = \frac{1}{e_0(S_{n+1}) - e_0(S_n)}. \quad (13)$$

The new sequence formed by the even-order terms $e_{2p}(S_n)$ converges toward S faster than the original sum $S = \sum_{i=1}^{\infty} a_i$.

Because we are dealing with double sums (over m and n), one way to evaluate the double sums would be to apply successively Shanks’ accelerator to the inner (n) and outer (m) sums (as suggested in Ref. 12). The problem with this approach is that in our case, the convergence of the inner sum (over n) can be very slow for high values of m (outer sum). A better solution consists in defining one element l of the Shanks series as the sum over $m = -l, l$ for $n = -l, \dots, l$ and $n = -l, l$ for $m = -l+1, \dots, l-1$. This strategy gets rid of the inner/outer sum problem and results in a faster convergence and an easier implementation of the Shanks’ algorithm.

Note that there is another way of computing efficiently the free-space term. As we did earlier, when we introduced a parameter h , it is possible to split the infinite sum (\mathbf{F}) terms in Eq. (6) in two parts; one in the direct space and one in the reciprocal space, where these two sums converge quickly owing to a damping function.^{13,14} The convergence is the best when $h = \sqrt{\pi/(u_x v_y - v_x u_y)}$. Poppe *et al.* introduced this method to study the optical response of an atomic monolayer; the period of the structure was therefore very small compared to the wavelength.

Once the periodic susceptibility tensors are known, we solve the linear system of Eq. (5) to find the self-consistent

TABLE I. Computation time in seconds for the coefficients of the linear system [Eq. (6)] used to solve Eq. (5). N is the number of subunits. CDM is the time for the classical CDM for one parallelepiped. CDM₁ is the time for the periodic CDM when the free-space contribution is computed with Eq. (8), and CDM₂ is the time for the periodic CDM when the free-space contribution is computed with Ref. 13. The infinite sums of the series are stopped when the relative error is less than 10^{-3} (10^{-6}).

	N	32	256	500	1372
	CDM	2	18	39	137
$a =$	CDM ₁	0.3 (2)	4 (17)	10 (34)	43 (116)
100 nm	CDM ₂	0.2 (0.4)	2.7 (5.5)	7 (13)	29 (54)
$a =$	CDM ₁	0.7 (2)	12 (30)	30 (75)	119 (300)
200 nm	CDM ₂	0.4 (1)	7 (16)	18 (40)	72 (158)
$a =$	CDM ₁	5.7 (16)	96 (281)	246 (684)	949 (4020)
1 μm	CDM ₂	5.6 (16)	96 (276)	233 (674)	900 (2460)

field at each site. Once the field at all subunits is known, the scattered field at any position \mathbf{r} , above, below, or inside the periodic structures is readily computed through Eq. (5) with the exchange $\mathbf{r} \leftrightarrow \mathbf{r}_i$. Notice that the new linear system is no longer symmetric. This is due to the fact that the elements of the system depend on the incident plane wave via the exponential term in Eq. (5).

III. EXAMPLE: SCATTERING BY A PERIODIC STRUCTURE LYING ON A SUBSTRATE

To illustrate the method we consider the case of a dielectric substrate (the relative permittivity is 2.25) on which lies a 2D grating of parallelepipeds with the same permittivity. The structure is illuminated in TM polarization from the substrate side by total internal reflection at an angle of incidence $\theta=45^\circ$; then

$$\mathbf{k}_{0\parallel} = \left(\frac{2\pi}{\lambda} \sin \theta \sqrt{2.25}, 0 \right).$$

The wavelength in vacuum is $\lambda=632.8$ nm, and the basis vectors of the lattice $\mathbf{u}=(a,0)$, $\mathbf{v}=(0,a)$. The parallelepipeds have a square base of 40×40 nm², and a height of 20 nm (see inset in Fig. 2). In Fig. 2, we present the intensity of the electric field, normalized to the incident field, along the x axis, 60 nm above the dielectric substrate for different value of a . The curves are obtained for $N=256$, hence the size of the subunit is $5 \times 5 \times 5$ nm³ (but convergence is already achieved for $N=32$). Notice that the solid line is for an isolated parallelepiped on the substrate, i.e., the electric field is computed with the conventional CDM.⁴ When a is small, the computed curves for the electric field are notably different from the single object case. This denotes a strong coupling between parallelepipeds. Conversely, for large a ($a=1000$ nm), the curve is very similar to the curve for an isolated parallelepiped.

Table I presents the computation time for the coefficients of the linear system [Eq. (6)] used to solve Eq. (5), for different values of N , and three values of a . The factor h has an

important influence on the computation time, therefore we have chosen the optimal value of h for each case. As a reference, we use the conventional CDM to compute the field for a single parallelepiped.¹⁵

Table I shows three computation times: CDM is the time for the classical CDM for one parallelepiped. CDM₁ is the time for the periodic CDM when the free-space contribution is computed with Eq. (8), and CDM₂ is the time for the periodic CDM when the free-space contribution is computed with Ref. 13. CDM₂ is faster than CDM₁ for small periods. For $a=1$ μm , the computation times are similar. For larger periods, CDM₂ fails to converge to the reference result because the method of Ref. 13 used to compute the free-space term does not work well for large a . We note that the computation time increases with a . This is mainly due to the surface term. The convergence of the series depends on the term $\exp[iw_0(z_i+z_j)]$. In our case the moduli of the vectors of the reciprocal basis are $|\mathbf{u}'|=|\mathbf{v}'|=2\pi/a$. Hence when a decreases, the modulus of the vector basis increases, w_0 becomes imaginary for smaller values of (m,n) , and the exponential term produces a stronger damping. Obviously, when N increases, the computation time increases due to the increased number of subunits involved. But there is another effect of the surface term. As the size of the subunit becomes smaller (N increases), there are more subunits close to the substrate with a small value of z_i+z_j and a slower exponential decay. When we compare the classical CDM to the periodic CDM, we see that for a smaller than 200 nm the computation time of the periodic CDM is shorter. When the size of the period becomes larger than the wavelength used, the convergence becomes slower.

IV. CONCLUSION

In conclusion we have generalized the coupled dipole method (CDM) to periodic structures. We have discussed explicitly the case of a three-dimensional structure, periodic in two directions, placed on a substrate. However, the principle of the approach described here applies to a broad range of configurations with one-, two-, or three-dimensional structures. The main advantage of this formulation is that it relies on the same straightforward, direct-space discretization scheme that is used for a single localized object. Therefore, all the knowledge acquired previously in CDM modeling of finite systems can be transposed to the study of periodic structures.¹⁶ Optical anisotropy, for instance, can be included by taking the appropriate permittivity tensor. Also, as shown here, the symmetry of the periodic lattice can be arbitrary. Here, we have considered the case of plane-wave illumination. In the case of arbitrary illumination, each spectral component of the incident field must be treated individually. An interesting extension of the present work would be to merge the periodic CDM and the conventional CDM into a single approach to light scattering. This would be particularly useful in dealing with localized defects in periodic structures or the interaction between a near-field probe (microscope tip, fluorescing particle, . . .) and a periodic system. The periodic generalization of the coupled dipole method can also be used to draw a better physical picture of the local-field corrections that appear during the multiple scattering of light by a discrete set of scatterers.¹⁷

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