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Order-*N* effective response of two-dimensional metallic structures

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Abstract

The discretization of Maxwell's equations using finite difference time domain (FDTD) methods produces real space–time equations that scale linearly with the system size, *N*. We present such an order-*N* scheme adapted to frequency-dependent dielectric functions from which we extract the effective response of metallic structures. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Yee [1] first proposed that Maxwell's equations could be discretized using a method that allowed computations to scale linearly with the system size, *N*. Most methods, such as the plane-wave expansion and transfer-matrix methods, scale as N^2 or worse. To access large systems, especially in three dimensions, computation costs quickly become prohibitive, whereby order-*N* methods can be used for calculations on systems for which other methods cannot be effectively applied.

The engineering community began to use Yee's result in finite-difference time domain (FDTD) computations for electromagnetic radiation [2–4]. Soon after, Chan et al. [5] adapted the FDTD scheme to apply to photonic crystals [6,7] as applied to simple dielectric materials. More recently, frequency-dependent dielectric functions for models of nearly free electron metals, such as the real Drude model [8], have been achieved.

Meanwhile, both experimental and theoretical studies of optical properties have produced important information [9] about composite materials, such as effective medium theories, where the composite is compared to a homogeneous material via an effective dielectric function, such as the one first derived by Maxwell-Garnett [10,11] for dilute systems of spherical particles. Accurate calculations of the effective dielectric function for periodic dielectric [12] and metallic composites [13–15] have been carried out recently, emphasizing applications to absorption and energy loss.

In this paper, we apply order-*N* methods to study the band structure of 2D structures arising from surface resonances at metal–dielectric interfaces and obtain accurate values for the strengths and positions for the modes of isolated square rods.

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2. Model and method

We consider a two-dimensional system composed of long metallic square rods embedded in an otherwise homogeneous dielectric medium (Fig. 1). The metal is modelled using the real Drude model, i.e.

$$\epsilon_{\text{rods}}(\omega)=1-\frac{\omega_{\text{p}}^2}{\omega^2} \tag{1}$$

where ω_{p} is the plasma frequency.

In general, to obtain the order-N discretization, we need to Fourier transform the electric permittivity, $\boldsymbol{D}(\boldsymbol{r},\omega)=\epsilon_0\epsilon(\omega)\boldsymbol{E}(\boldsymbol{r},\omega)$, into the time domain, which gives

$$\boldsymbol{D}(\boldsymbol{r},t)=\epsilon_0\boldsymbol{E}(\boldsymbol{r},t)+\epsilon_0\int_{-\infty}^{\infty}\boldsymbol{E}(\boldsymbol{r},\tau)\chi(t-\tau)\,\mathrm{d}\tau \tag{2}$$

where $\chi(t)=\epsilon(t)-1$ is the susceptibility. Using Eq. (1), Maxwell's equations can be combined to yield

$$\ddot{\boldsymbol{E}}(\boldsymbol{r},t)=-[c^2\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E}(\boldsymbol{r},t)+\omega_{\text{p}}^2\boldsymbol{E}(\boldsymbol{r},t)] \tag{3}$$

which can then be discretized in the time coordinate by the Halijak (central difference) method [16]. Thus, Eqs. (2) and (3) become

$$\boldsymbol{E}_{n+1}(\boldsymbol{r})=(2-\omega_{\text{p}}^2T^2)\boldsymbol{E}_n(\boldsymbol{r})-\boldsymbol{E}_{n-1}(\boldsymbol{r})-T^2c^2\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E}_n(\boldsymbol{r}) \tag{4}$$

T being the size of the discrete time step. Since the spatial derivatives apply only to $\boldsymbol{E}_n(\boldsymbol{r})$, the spatial discretization is obtained by straightforward discretization of the curl operator.

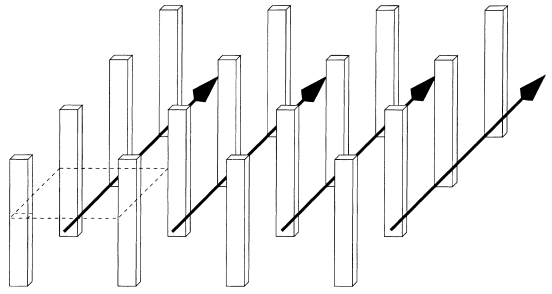

Fig. 1. Two-dimensional composite materials, consisting of square–cylindrical metal rods in a dielectric medium, showing the unit cell. Initial plane-waves are oriented perpendicular (p-polarized) to the rod orientation.

To calculate the photonic band structure, we introduce an initial superposition of plane-waves of momentum k, perpendicular to the rod orientation. Propagation through the material is restricted to the resonance frequencies, which survive as peaks in the spectral density after sufficient time has elapsed to suppress the non-propagating frequencies. Since the frequencies are thereby generated as a function of k, the order-N method is well suited to investigate properties of systems that exhibit flat photonic band structures, as in the case of metals.

Within effective medium theory, one assumes a transverse effective dielectric function which would satisfy the (k,ω) relationship of a homogeneous material [14,15], i.e.

$$\epsilon_{\text{eff}}(\omega)=\frac{c^2k^2}{\omega^2}. \tag{5}$$

Since the dielectric function of metals is negative below the plasma frequency, surface resonances arise, and the effective response of the composite can be written as [17,18]

$$\epsilon_{\text{eff}}(\omega)=\epsilon_0\left[1-f\sum_v\frac{B_v}{\omega^2/\omega_{\text{p}}^2-m_v}\right] \tag{6}$$

where f is the filling fraction of metal to dielectric material and the strengths add up to unity, $\Sigma_v\,B_v=1$.

Similarly, the effective inverse response may also be modelled by

$$\epsilon_{\text{eff}}^{-1}(\omega)=\epsilon_0^{-1}\left[1+f\sum_v\frac{C_v}{\omega^2/\omega_{\text{p}}^2-n_v}\right] \tag{7}$$

again, with the strengths summing to unity.

For the two-dimensional metallic–dielectric composite described, s-polarized electromagnetic waves, i.e. where the electric field component is parallel to the axes of the rods, are found to be equivalent to having a homogeneous medium according to Eqs. (6) and (7) with a single non-zero mode with $B_0=C_0=1$, $m_0=0$ and $n_0=f$ [14,15]. With p-polarized waves, however, where the electric field component is perpendicular to the axes of the rods, all depolarization factors are

found to satisfy [14]

$$n_v = 1-(D-1)m_v \tag{8}$$

for dimensionality D, which is 2 in the system described above.

For a single inclusion embedded in a dielectric material, one finds that the spectral representations of Eqs. (6) and (7) coincide, which means that for $n_v < 1/D$, there exists another mode $n'_v > 1/D$ satisfying [15]

$$n'_v = 1-(D-1)n_v \tag{9}$$

thereby forming associated pairs of modes.

Since the n_v are the poles of $\epsilon_{\text{eff}}^{-1}$, they correspond to the modes obtained by setting $k=0$ in Eq. (5), thereby exploiting this advantage of the order-N method. Next, we calculate the $k=\pi/d$ results, d being the length of the unit cell, from which we can calculate the strength of the mode. Since these sets of points are well removed from the light cone, we can consider all other modes to have negligible contribution, and so we have

$$\frac{c^2k^2}{\omega_v^2(kd)} = 1-\frac{B_v}{\omega_v(kd)^2-m_v} \tag{10}$$

at $kd=0$, π, leading to

$$B_v = \omega_v(0)^2 - m_v$$

$$B_v = [\omega_v(\pi)^2 - m_v]\left[1+\frac{c^2\pi^2}{d^2\omega_v(\pi)^2}\right] \tag{11}$$

which we solve to obtain B_v and m_v. Thus, for order-N, we do not need to calculate the entire band structure to obtain the parameters for the effective response. Other schemes, such as the transfer matrix method, cannot achieve comparable results since, even at equal energy resolutions, the band flatness prevents much of the data from being extracted in addition to the disadvantage of N^2 scaling. Further, order-N can be applied to 3D structures that are not attainable with the transfer matrix.

3. Results

Taking $\hbar\omega_p = 15.8$ eV, the plasma frequency for aluminium, applied to square rods of 6 nm diameter and 36 nm between rod axes, we obtain a filling fraction of 0.028, which is sufficient to isolate the rods, and compute the band structure for p-polarized waves as described. Fig. 2 shows the most prominent photonic bands (solid lines) for the metallic rod system in Fig. 1 using 10×10 grid for the rods in a 60×60 mesh for the periodic cell (requiring 9391 s of CPU time on a DEC alpha), deviating from the homogeneous dielectric solution (dashed lines) where $\omega=\sqrt{\epsilon_0}ck$. The presence of sharp corners and flat interfaces between metal and dielectric introduces, in the limit of isolated rods, a host of resonances which manifest as very flat bands corresponding to the modes of Eqs. (6) and (7).

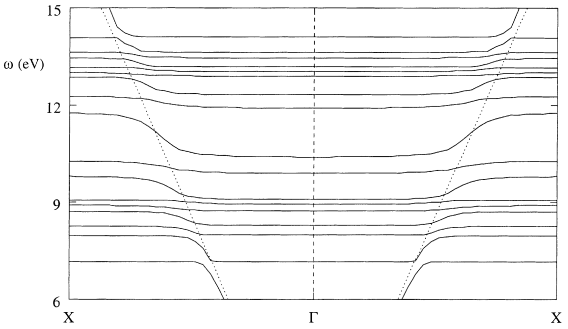

Fig. 2. Photonic band structure for p-polarization for a 10×10 grid of aluminium embedded in a 60×60 periodic unit cell.

Table 1
Prominent pairs of depolarization factors and corresponding strengths for the effective dielectric function

$m_v \approx n_v$	B_v	$m'_v \approx n'_v$	B_v
0.206	0.023	0.796	0.018
0.255	0.025	0.745	0.020
0.275	0.025	0.726	0.021
0.305	0.025	0.695	0.021
0.321	0.077	0.679	0.044
0.332	0.026	0.665	0.045
0.393	0.182	0.608	0.095
0.432	0.235	0.567	0.272

As the mesh is refined, additional bands appear about the existing ones, all remaining in the 6–15 eV region, but are dominated by the strengths of a few prominent peaks. Because of the unit sum between depolarization factors of corresponding modes, we see that the upper region is more densely packed than the lower, allowing for the

lower modes to be broader and more prominent by having larger B_v. Thus, we must use Eq. (9) and consider pairs of modes when determining which ones are important.

Extracting the values for Eq. (6) from the band structure calculation, we find that as the grid is refined, additional modes appear which may cause a slight repositioning of the previous modes until convergence to the continuum is obtained. Most of the contribution, however, is determined by a few prominent pairs. In Table 1, we show the eight pairs for the 60×60 mesh which reproduce the modes shown in Fig. 2. The sum of the strengths of these prominent modes exceeds unity due to the finiteness of the grid. As the grid is refined, the strength sum converges to unity. Furthermore, these locations and strengths are independent of the dielectric properties of the metallic material, the results arise solely from the microgeometry of the rods.

4. Conclusions

We have investigated the properties of the effective dielectric function arising from the surface resonances at the interfaces of a periodic composite of square metallic rods embedded in a dielectric medium using an FDTD order-N method of discretizing Maxwell's equations. The depolarization factors and strengths are obtainable directly from the $k=0$ and $k=\pi/d$ calculations using this method, rather than requiring knowledge of the entire band structure, and we have been able to test the accuracy of our results through the fulfilment of sum rules.

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References

[1] K.S. Yee, IEEE Trans. Ant. Prop. 14 (1966) 302.
[2] R.J. Luebbers, F. Hunsberger, K.S. Kunz, R.B. Standler, M. Schneider, IEEE Trans. Electromagn. Compat. 32 (1990) 222.
[3] R.M. Joseph, S.C. Hagness, A. Taflove, Opt. Lett. 16 (1991) 1412.
[4] A. Taflove, Computational Electrodynamics: The Finite-Difference Time-Domain Method, Artech House, Boston, 1995.
[5] C.T. Chan, Q.L. Yu, K.M. Ho, Phys. Rev. B 51 (1995) 16 635.
[6] J.B. Pendry, J. Phys.: Condens. Matter 8 (1996) 1085.
[7] V. Kuzmiak, A.A. Maradudin, Phys. Rev. B 55 (1997) 7427.
[8] J. Arriaga, A.J. Ward, J.B. Pendry, Phys. Rev. B 59 (1999) 1874.
[9] D.J. Bergman, D. Stroud, in: H. Ehrenreich, D. Turnbull (Eds.), Solid State Physics Vol. 46, Academic Press, New York, 1992, p. 147. and references cited therein.
[10] J.C. Maxwell-Garnett, Philos. Trans. R. Soc. Lond. 203 (1904) 385.
[11] J.C. Maxwell-Garnett, Philos. Trans. R. Soc. Lond. 205 (1906) 237.
[12] S. Datta, C.T. Chan, K.M. Ho, C.M. Soukoulis, Phys. Rev. B 48 (1993) 14 936.
[13] F.J. García-Vidal, J.M. Pitarke, J.B. Pendry, Phys. Rev. Lett. 78 (1997) 4289.
[14] J.M. Pitarke, F.J. García-Vidal, J.B. Pendry, Phys. Rev. B 57 (1998) 15 261.
[15] J.M. Pitarke, F.J. García-Vidal, J.B. Pendry, Surf. Sci. 433 (1999) 605.
[16] T.T. Hartley, G.O. Beale, S.P. Chicatelli, Digital Simulation of Dynamic Systems: A Control Theory Approach, Prentice-Hall, Englewood Cliffs, NJ, 1994.
[17] D. Bergman, Phys. Rep. 43 (1978) 377.
[18] G. Milton, J. Appl. Phys. 52 (1981) 5286.