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Scattering of electromagnetic waves by a disordered two-dimensional array of spheres

N Stefanou† and A Modinos‡

† Solid State Section, University of Athens, Panepistimioupolis, New Buildings, GR-157 84 Athens, Greece

‡ Department of Physics, National Technical University of Athens, Zografou Campus, GR-157 73 Athens, Greece

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Abstract. We present a formalism for the calculation of the scattering of electromagnetic waves by a substitutionally disordered two-dimensional array of spherical particles. The formalism, which constitutes an extension of the coherent-potential approximation scheme to electromagnetic waves is valid for any frequency of the incident wave and for any size and/or concentration of the particles. We demonstrate the applicability of our formalism by applying it to the evaluation of the absorbance of disordered arrays of plasma spheres.

1. Introduction

In our earlier work [1, 2] we have shown how to calculate the scattering of electromagnetic (EM) waves from an ordered two-dimensional (2D) array of spherical particles on a substrate of a different dielectric function. We have applied the method to systems of practical interest, namely metallic particles with a diameter of about 100 Å or so on a dielectric substrate, and considered the effect of coverage, size of particles, polarization of light and other factors on the transmittance, reflectance and absorbance of light by such systems [2, 3]. The arrangement of the particles on the substrate is very rarely periodic in an actual experiment (see, e.g., [4], and references therein) and it is therefore important to be able to take into account the effect of disorder on the optical properties of such systems. When the disorder is weak, this can be taken into account, to some degree, by the so-called average **T**-matrix approximation (ATA) as we have done in previous work [3, 5]. It is well known, however, from the corresponding problem of electron scattering in substitutionally disordered alloys, that an improved treatment of disorder can be obtained from the so-called coherent-potential approximation (CPA) [6, 7]. In fact, Liebsch and Persson [8] and Persson and Liebsch [9] have already applied what is essentially a CPA scheme to the evaluation of EM scattering by three-dimensional (3D) and 2D arrays of metallic particles in the electrostatic approximation. Their treatment is valid in the long-wavelength limit and then only when the interparticle distance is relatively large, which means a low coverage in the case of particles on a substrate.

More recently, Wang *et al* [10] developed a multiple-scattering Green function formalism for EM waves and applied it to the calculation of the frequency band structure of a crystal (diamond structure) of touching vacuum spheres in a dielectric medium. An extension of their formalism to 3D substitutionally disordered systems under the CPA was also proposed in the same paper.

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In this article we present the first calculations of the transmittance, reflectance and absorbance of EM waves by 2D arrays of particles using the CPA scheme to solve Maxwell's equations exactly. Our formalism can be used together with a doubling-layer technique [11, 12] to treat also substitutional disorder in 3D layered structures. In section 2 we present the formalism, which allows one to calculate for any frequency of the incident radiation the scattering of EM waves by a disordered 2D array of spheres. We note that our formalism applies equally well to arrays of particles of any shape as long as the particles remain distinct from each other (i.e. do not overlap). In section 3 we demonstrate the method by applying it to specific examples.

2. Theory

2.1. The model

We assume a 2D lattice in the x–y plane. The sites $\boldsymbol{R}_n$ of the lattice are occupied by spheres (the centres of the spheres coincide with the lattice points) of type A, B, C, ... The spheres do not overlap each other and their arrangement on the lattice is arbitrary (non-periodic). The spheres A, B, C, ... are characterized by relative dielectric functions $\epsilon_A(\omega), \epsilon_B(\omega), \epsilon_C(\omega), \ldots$, respectively, and the surrounding medium by a relative dielectric function $\epsilon(\omega)$.

The CPA consists in replacing the disordered arrangement of spheres by an ordered (periodic) arrangement of identical effective scatterers occupying all the lattice sites. The effective scatterer need not be, and in fact is not, a spherical scatterer and its scattering properties are a rather complicated function of $\epsilon_A, \epsilon_B, \epsilon_C, \ldots$. The properties of the CPA scatterer and the justification of the method are exactly the same as in the case of electron scattering by a non-periodic arrangement of atomic scatterers [6, 7].

Once the scattering properties of the effective scatterer have been established, the scattering of an incident wave by the ordered array of these scatterers proceeds in the manner described earlier [1, 2].

2.2. The CPA scatterer

The electric field component $\boldsymbol{E}(r, t) = \mathrm{Re}[\tilde{\boldsymbol{E}}(r)\exp(-\mathrm{i}\omega t)]$ of the EM waves incident on and scattered by a sphere centred on the origin of coordinates is expanded in spherical waves as follows [1, 2]:

$$\tilde{\boldsymbol{E}}(r) = \sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left(\frac{\mathrm{i}}{k}a_{lmE}\nabla\times z_l(kr)\boldsymbol{X}_{lm}(\hat{\boldsymbol{r}}) + a_{lmH}z_l(kr)\boldsymbol{X}_{lm}(\hat{\boldsymbol{r}})\right) \tag{1}$$

where $k = [\epsilon(\omega)]^{1/2}\omega/c$ is the wavenumber. $\boldsymbol{X}_{lm}(\hat{\boldsymbol{r}})$ are the usual vector spherical harmonics; $z_l(kr) = j_l(kr)$ for the incident wave and $z_l(kr) = h_l^+(kr)$ for the scattered wave, where j_l and h_l^+ denote the spherical Bessel and the outgoing spherical Hankel functions, respectively. As written, the above expression applies outside the scatterer. A similar expression applies inside the scatterer. The coefficients $a_{lm\sigma}$, where $\sigma = E$ or H, in equation (1) are constants to be determined.

The expansion coefficients $a^+_{lm\sigma}$ of the scattered wave are related to the corresponding coefficients $a^0_{lm\sigma}$ of the incident wave by

$$a^+ = \mathbf{T}^{(i)}(\omega)a^0 \tag{2}$$

where here and throughout this article matrix notation in the representation $lm\sigma$ is used. $\mathbf{T}^{(i)}$, $i = A, B, C, \ldots$ is the scattering matrix for the sphere under consideration. For a spherical scatterer we have in general

$$T^{(i)}_{lm\sigma;l'm'\sigma'} = T^{(i)}_{l\sigma}\delta_{ll'}\delta_{mm'}\delta_{\sigma\sigma'}. \tag{3}$$

Explicit expressions for $T_{l\sigma}$ have been given in [1]. We shall see, however, that the diagonal form of the T-matrix described by equation (3) does not apply to the effective CPA scatterer and for that reason, in what follows, we keep the general form of equation (2). Denoting by $\langle\mathbf{T}\rangle$ the CPA T-matrix (which is to be determined), we introduce $\Delta\mathbf{T}^{(i)}$ as follows:

$$\mathbf{T}^{(i)}(\omega) = \langle\mathbf{T}(\omega)\rangle + \Delta\mathbf{T}^{(i)}(\omega). \tag{4}$$

Let us consider a plane EM wave incident on the non-periodic 2D array of spheres described in section 2.1. If we forget, for the moment, the second term of equation (4), our array becomes in effect periodic and the scattered field can be written in the form

$$\tilde{E}_{sc}(r) = \sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left(\frac{i}{k}b^{+}_{lmE}\nabla\times\sum_{R_n}\exp(ik_{\|}\cdot R_n)h^{+}_{l}(kr_n)X_{lm}(\hat{r}_n)\right.$$
$$\left. + b^{+}_{lmH}\sum_{R_n}\exp(ik_{\|}\cdot R_n)h^{+}_{l}(kr_n)X_{lm}(\hat{r}_n)\right) \tag{5}$$

with $r_n \equiv r - R_n\cdot k_{\|} = (k_x, k_y)$ is the projection of the wavevector of the incident wave on the plane of the spheres. The procedure for calculating the coefficients $b^{+}_{lm\sigma}$ has been described in [1, 2].

Let us now replace the scatterer $\langle\mathbf{T}\rangle$ at the site $R_n = \mathbf{0}$ by the actual i scatterer. Then the scattered wave from this sphere consists of the corresponding term ($R_n = \mathbf{0}$) of (5) and, to begin with, an additional term given by (1) with $z_l(kr) = h^{+}_{l}(kr)$ and coefficients given by

$$\Delta\mathbf{T}^{(i)}(a^0 + b') \tag{6}$$

where b' represents the coefficients in the spherical-wave expansion about $R_n = \mathbf{0}$ of the wave (5) with the $R_n = \mathbf{0}$ term subtracted (see [1]). The wave (6) will be multiply scattered by the periodic array of spheres, including that at $R_n = \mathbf{0}$, producing a further contribution to the incident on the sphere at the origin wave, which is given by equation (1) with $z_l(kr) = j_l(kr)$ and with coefficients given by

$$\tilde{\mathbf{D}}^{00}\Delta\mathbf{T}^{(i)}(a^0 + b') \tag{7}$$

where $\tilde{\mathbf{D}}^{00}$ represents the contribution of all possible paths by which a wave outgoing from the site $R_n = \mathbf{0}$ produces an incident wave on the same site after scattering in all possible ways by the scatterers $\langle\mathbf{T}\rangle$ at all sites of the lattice, including $R_n = \mathbf{0}$. Because of (7) there will be a further contribution to the scattered wave from the sphere at the origin with coefficients given by

$$\Delta\mathbf{T}^{(i)}\tilde{\mathbf{D}}^{00}\Delta\mathbf{T}^{(i)}(a^0 + b'). \tag{8}$$

The process is repeated infinitely many times and this leads to the following expression for the contribution to the coefficients of the scattered wave from the sphere at $\boldsymbol{R}_n = \mathbf{0}$, due to $\Delta\mathbf{T}^{(i)}$:

$$(\mathbf{I} + \Delta\mathbf{T}^{(i)}\tilde{\mathbf{D}}^{00} + \Delta\mathbf{T}^{(i)}\tilde{\mathbf{D}}^{00}\Delta\mathbf{T}^{(i)}\tilde{\mathbf{D}}^{00} + \ldots)\Delta\mathbf{T}^{(i)}(a^0 + b') = (\mathbf{I} - \Delta\mathbf{T}^{(i)}\tilde{\mathbf{D}}^{00})^{-1}\Delta\mathbf{T}^{(i)}(a^0 + b') \tag{9}$$

where $\mathbf{I}$ is a unit matrix. In order to proceed further we need an explicit formula for $\tilde{\mathbf{D}}^{00}$. We find (see appendix) that

$$\tilde{\mathbf{D}}^{00}(\omega) = \frac{1}{S_0}\int_{SBZ} \mathrm{d}^2k_\parallel \,(\mathbf{I} - \boldsymbol{\Omega}(k_\parallel;\omega)(\mathbf{T}(\omega))]^{-1}\boldsymbol{\Omega}(k_\parallel;\omega) \tag{10}$$

where S_0 is the area of the surface Brillouin zone (SBZ) associated with the 2D periodic lattice, and

$$\boldsymbol{\Omega} \equiv \begin{bmatrix} \Omega^{(1)} & -\Omega^{(2)} \\ \Omega^{(2)} & \Omega^{(1)} \end{bmatrix} \tag{11}$$

where $\Omega^{(1)}_{l'm';lm}$ and $\Omega^{(2)}_{l'm';lm}$ are, as it turns out, the matrices given in the appendix of [2].

We obtain the CPA matrix $\langle\mathbf{T}\rangle$ in the manner originally proposed by Soven [6], by demanding that

$$\sum_i C_i(\mathbf{I} - \Delta\mathbf{T}^{(i)}\tilde{\mathbf{D}}^{00})^{-1}\Delta\mathbf{T}^{(i)} = 0 \tag{12}$$

where C_i denotes the concentration of scatterer i. Obviously

$$\sum_i C_i = 1. \tag{13}$$

Equation (12) tells us that, with $\langle\mathbf{T}\rangle$ determined in this way, the correction to the scattering due to the difference of the actual scatterers from the CPA scatterer vanishes on the average. In principle we may have a large number of terms in the sum over i, corresponding to the variety of scatterers present. In our examples (section 3) we consider the simplest case i = A, B, where B corresponds to a 'vacant' site. In the case of only two different scatterers, it can be shown that equation (12) can be written as

$$\langle\mathbf{T}\rangle = C_A\mathbf{T}^{(A)} + C_B\mathbf{T}^{(B)} - \Delta\mathbf{T}^{(A)}\tilde{\mathbf{D}}^{00}\Delta\mathbf{T}^{(B)}. \tag{14}$$

The matrix $\langle\mathbf{T}\rangle$, which enters the above equation within $\Delta\mathbf{T}^{(i)}$ and $\tilde{\mathbf{D}}^{00}$ (see equation (10)), is to be found by solving this equation numerically (see next section).

2.3. *Numerical evaluation of* $\langle\mathbf{T}\rangle$

We now turn to the numerical evaluation of $\langle\mathbf{T}\rangle$. This is achieved by solving equations (10) and (14) by an iterative procedure. Using a reasonable input for $\langle\mathbf{T}\rangle$ (the ATA matrix $C_A\mathbf{T}^A + C_B\mathbf{T}^B$) we obtain $\tilde{\mathbf{D}}^{00}$ from equation (10). Substituting $\tilde{\mathbf{D}}^{00}$ in (14) we obtain an output value for $\langle\mathbf{T}\rangle$. The whole procedure is repeated, using as input at each stage of the iteration a mixture of the input and output $\langle\mathbf{T}\rangle$ of the previous iteration, until convergence is attained. This simple mixing scheme [13] leads to convergence but not as fast as the

Chebychev iteration scheme [14] or quasi-Newton–Raphson algorithms [15]. We tried the simple mixing and the Chebychev iteration schemes. Approximately 30 iterations gave good convergence in all the cases that we examined.

The angular momentum space is truncated by introducing an angular momentum cut-off l_{max}. Therefore, all the matrices in the representation $lm\sigma$ have the dimensions $2l_{max}(l_{max}+2) \times 2l_{max}(l_{max}+2)$. We note that the CPA matrix $\langle \mathbf{T} \rangle$ is in general non-diagonal, even in the case of spherical scatterers A and B, because of the last term in equation (14). However, in this case, one can restrict $\langle \mathbf{T} \rangle$ to a block-diagonal form. Indeed, the symmetry properties of $\Omega^{(1)}$ and $\Omega^{(2)}$ are such (see appendix of [2]) that Ω has a block-diagonal form with two blocks of dimensions $l_{max}(l_{max}+2) \times l_{max}(l_{max}+2)$ each, provided that the representation basis is ordered in the following way: odd $(l+m)E$; even $(l+m)H$; odd $(l+m)H$; even $(l+m)E$. Therefore, if the input $\langle \mathbf{T} \rangle$ is block diagonal, $\tilde{\mathbf{D}}^{00}$ calculated from (10) is block diagonal and, therefore, the output $\langle \mathbf{T} \rangle$ calculated from (14) is also block diagonal. In this manner, the dimensions of the matrices involved in the calculation are reduced, and so is the CPU time.

The evaluation of $\tilde{\mathbf{D}}^{00}$ requires a numerical integration within the SBZ (see equation (10)). Transformation of the integrand under the symmetry operations of the point group of the lattice in order to reduce the integration within the irreducible part of the SBZ is not in the present case profitable. It appears that numerical integration within the full SBZ is computationally more efficient. For this purpose, one can use a set of special points with corresponding weights, generated using the algorithm of Cunningham [16]. Also, the SBZ (square in our applications) can be subdivided into small squares, within which a nine-point integration formula [17] is very efficient. In general, using a set of about 100 points within the full SBZ, we obtained good convergence by both methods.

Once the CPA matrix $\langle \mathbf{T} \rangle$ is evaluated, the scattering of an incident wave by the ordered array of these scatterers proceeds in the manner described earlier. We note that, in applying the formulae of [2], only the term corresponding to $g = \mathbf{0}$ needs to be retained in all the cases examined.

3. Applications

We applied our method to the calculation of the absorbance of light by disordered 2D arrays of plasma spheres. Consider a square lattice with a fraction C_A of its sites occupied at random by identical plasma spheres and assume that the remaining sites are empty. Let us also assume that the optical response of the individual sphere is adequately described by the Drude dielectric function

$$\epsilon_A(\omega) = \omega_p^2/\omega(\omega + i/\tau). \tag{15}$$

Following Persson and Liebsch [9] we have taken $\hbar\omega_p = 6.93$ eV and $\hbar\tau^{-1} = 0.158$ eV which, it is assumed, are appropriate for silver particles.

Firstly, and in order to test our method, we considered a very low concentration $C_A = 0.3$ of spheres of radius $S = 50$ Å arranged on a square lattice with lattice constant $a = 100$ Å. In this case, one expects the electrostatic approximation of [9] to be adequate, and this allows a meaningful comparison between our results and their results.

One obtains a measure of the effect of disorder by comparing the absorbance of light by the disordered, partly empty lattice with lattice constant a, with that calculated for an ordered, fully occupied lattice with a lattice constant $a' = a/\sqrt{C_A}$ (= 182.57 Å) so that the coverage (number of spheres per unit area) is the same in both cases.

In the electrostatic limit, one obtains two peaks in the absorbance of an ordered square lattice of plasma spheres. The low-energy peak corresponding to a parallel mode resonance (the charge oscillates parallel to the plane of the spheres) is excited by the component of the electric field parallel to the plane. The high-energy peak corresponding to a normal mode resonance (the charge oscillates normal to the plane of the spheres) is excited by the normal component of the electric field. The resonance frequencies are given by [9]

$$\Omega_{\parallel} = \Omega(1 - \tfrac{1}{2}S^3U_0)^{1/2} \tag{16}$$

$$\Omega_{\perp} = \Omega(1 + S^3U_0)^{1/2} \tag{17}$$

where $\Omega = \omega_p/\sqrt{3}$ is the resonance frequency of a single plasma sphere and $U_0 = 9.03a'^{-3}$ for a square lattice.

ATA calculations [5] suggest that the disorder induces a shift of the parallel (normal) mode absorbance peak to lower (higher) frequencies relative to the corresponding peaks of the corresponding ordered structure. We see from table 1 that the shifts predicted by the ATA calculation are in the same direction but smaller, especially in the case of the parallel mode, than those of the CPA calculations. The ATA in the electrostatic limit replaces the partly empty (disordered) lattice by one fully occupied by spheres characterized by an average polarizability $C_A\alpha_A(\omega)$, where

$$\alpha_A(\omega) = S^3[\epsilon_A(\omega) - 1]/[\epsilon_A(\omega) + 2] \tag{18}$$

is the polarizability of the single plasma sphere. Therefore, the resonance frequencies in the ATA scheme are given by equations (16) and (17) with S^3 and a' replaced by C_AS^3 and $\sqrt{C_A}a'$, respectively. The values of $\Omega_{\parallel}$ and $\Omega_{\perp}$ evaluated in this way for the ordered structure and for the disordered structure (in the ATA scheme) are in excellent agreement with the results of the exact treatment [5] of these quantities (reported in table 1), as expected. At low coverages the electrostatic approximation is valid.

Table 1. Position of absorbance peaks.

	$\hbar\Omega_{\parallel}$ (eV)	$\hbar\Omega_{\perp}$ (eV)
Ordered structure	3.81	4.36
ATA	3.64	4.61
CPA	3.33	4.70
[9]	3.20	4.80

Figure 1 shows the absorbance as a function of the photon energy of p-polarized light incident at an angle $\theta = \frac{1}{4}\pi$ ($k_{\parallel} = (k_{\parallel}, 0)$ and the electric field lies in the plane of incidence). The broken curve shows the absorbance of the ordered array of spheres. We see quite clearly in the CPA results the shift of the parallel mode resonance to lower energies and that of the normal mode resonance to higher energies. The positions of the shifted peaks are almost identical with those evaluated in the electrostatic approximation by Persson and Liebsch [9] (see table 1). The same applies to the shape of the peaks, and especially to that of the parallel mode resonance which is broadened to a larger degree because of the variance in the distribution of the interaction between neighbours (see [9]).

We must now emphasize that our method, unlike that in [9], is not restricted to low frequencies and/or small concentrations of spheres. To demonstrate this we calculated the

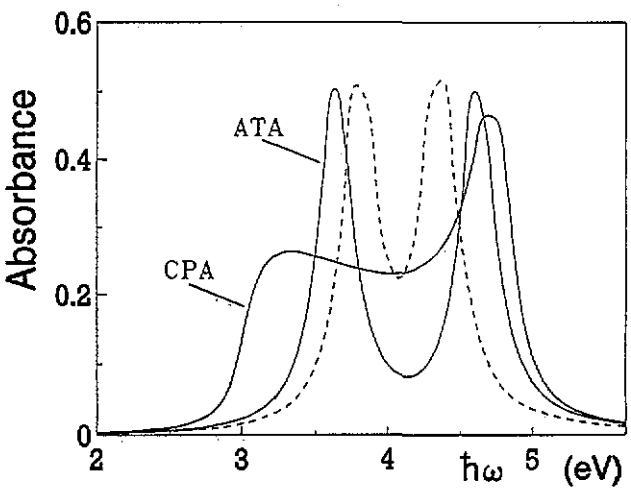

Figure 1. Absorbance of p-polarized light of angular frequency ω incident at an angle $\theta = \frac{1}{4}\pi$ on a disordered array of plasma spheres of radius $S = 50$ Å occupying randomly 25% of the sites of a square lattice of lattice constant $a = 100$ Å (——) and on an ordered square array of plasma spheres of radius $S = 50$ Å with lattice constant $a = 182.57$ Å (– – –).

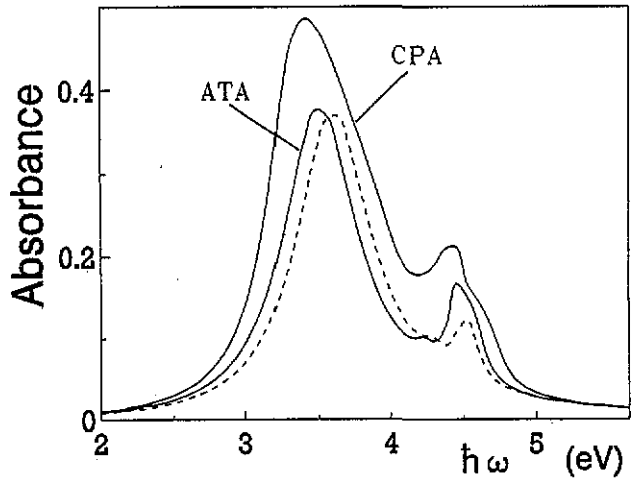

Figure 2. Absorbance of normally incident light of angular frequency ω on an array of plasma spheres of radius $S = 80$ Å occupying randomly 75% of the sites of a square lattice of lattice constant $a = 200$ Å (——). The absorbance of the corresponding ordered lattice ($a = 230.94$ Å) is also shown (– – –).

absorbance as a function of the photon energy of light incident normally on a plane of spheres of radius $S = 80$ Å occupying randomly 75% of the sites of a square lattice of lattice constant $a = 200$ Å. The results are shown in figure 2. The broken curve in this figure gives the absorbance of the corresponding (in the manner defined earlier) ordered lattice. The main feature in the absorbance curve is the appearance of a resonance peak which does not exist within the electrostatic approximation. In order to obtain this additional

peak which comes about through interparticle scattering of light, we need to maintain in the angular momentum expansions terms up to $l_{max} = 4$. We see that the disorder does not destroy the additional peak. The main effect of the disorder appears to be a slight shift of the resonance peaks accompanied by broadening of the peaks. At the same time we see an increase in the overall absorbance by about 30%.

Finally, we would like to add that the formalism that we have presented in this paper can be applied to a variety of problems (for a list of these see, e.g., [9]) and we hope to be able to deal with some of these problems in future publications.

Appendix

An outgoing wave spherical about $\boldsymbol{R}_m$, given by

$$\tilde{\boldsymbol{E}}(\boldsymbol{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left(\frac{\mathrm{i}}{k} b^{+}_{lmE}(\boldsymbol{R}_m) \boldsymbol{\nabla} \times h_l^{+}(kr_m) \boldsymbol{X}_{lm}(\hat{\boldsymbol{r}}_m) + b^{+}_{lmH}(\boldsymbol{R}_m) h_l^{+}(kr_m) \boldsymbol{X}_{lm}(\hat{\boldsymbol{r}}_m) \right) \tag{A1}$$

can be expanded into spherical waves about $\boldsymbol{R}_n$, incident on $\boldsymbol{R}_n$. We have

$$\tilde{\boldsymbol{E}}(\boldsymbol{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \left(\frac{\mathrm{i}}{k} b'_{lmE}(\boldsymbol{R}_n) \boldsymbol{\nabla} \times j_l(kr_n) \boldsymbol{X}_{lm}(\hat{\boldsymbol{r}}_n) + b'_{lmH}(\boldsymbol{R}_n) j_l(kr_n) \boldsymbol{X}_{lm}(\hat{\boldsymbol{r}}_n) \right) \tag{A2}$$

where

$$b'(\boldsymbol{R}_n) = \tilde{\Omega}(\boldsymbol{R}_{nm}) b^{+}(\boldsymbol{R}_m) \tag{A3}$$

with $\boldsymbol{R}_{nm} = \boldsymbol{R}_n - \boldsymbol{R}_m$. The matrix $\tilde{\Omega}(\boldsymbol{R}_{nm}) \equiv \tilde{\Omega}^{nm}$ can be obtained from the formalism in [1, 2]. We obtain

$$\tilde{\Omega}^{nm}_{l'm'E;lmE} = \tilde{\Omega}^{nm}_{l'm'H;lmH} = \sum_{\mu=-1}^{1} C(l1l; m-\mu\mu) G_{lm-\mu;l'm'-\mu}(\boldsymbol{R}_{nm}) C(l'1l'; m'-\mu\mu) \tag{A4}$$

$$\begin{aligned} \tilde{\Omega}^{nm}_{l'm'E;lmH} &= -\tilde{\Omega}^{nm}_{l'm'H;lmE} \\ &= \sqrt{\frac{2l'+1}{l'+1}} \sum_{\mu=-1}^{1} C(l1l; m-\mu\mu) G_{lm-\mu;l'-1m'-\mu}(\boldsymbol{R}_{nm}) C(l'-11l'; m'-\mu\mu) \end{aligned} \tag{A5}$$

where

$$G_{lm;l''m''}(\boldsymbol{R}_{nm}) = \sum_{l'=0}^{\infty} \sum_{m'=-l'}^{l'} 4\pi (\mathrm{i})^{l-l'-l''} (-1)^{m'+m''} B_{lm}(l'm'; l''m'') h_{l'}^{+}(kR_{nm}) Y_{l'-m'}(\hat{\boldsymbol{R}}_{nm}) \tag{A6}$$

$$B_{lm}(l'm'; l''m'') \equiv \int Y_{lm}(\hat{\boldsymbol{r}}) Y_{l'm'}(\hat{\boldsymbol{r}}) Y_{l''-m''}(\hat{\boldsymbol{r}}) \, \mathrm{d}\hat{\boldsymbol{r}} \tag{A7}$$

and $C(l_1 l_2 l; m_1 m - m_1)$ are Clebsch–Gordan coefficients [18].

We now introduce the propagator $\tilde{\mathbf{D}}^{nm}$ which gives the coefficients (in an expansion such as (A2)) of the wave incident on the sphere at $\boldsymbol{R}_n$, due to an outgoing wave from the sphere at $\boldsymbol{R}_m$. An outgoing wave (A1) from the mth sphere can reach the nth sphere directly and in that way contributes to the incident wave on that sphere the term (A2) or can reach it after scattering any number of times by any number of scatterers (including those at m and n) of the periodic array of spheres. Let $\tilde{\mathbf{D}}^{nm}$ be the sum of the contribution to the coefficients of the incident wave on $\boldsymbol{R}_n$ from all possible scattering paths originating from an outgoing wave from the mth sphere. One can easily prove by iteration the following equation:

$$\tilde{\mathbf{D}}^{nm} = \tilde{\Omega}^{nm} + \sum_{p \neq n} \tilde{\Omega}^{np} \langle \mathbf{T} \rangle \tilde{\mathbf{D}}^{pm} \tag{A8}$$

where $\langle \mathbf{T} \rangle$ is the matrix (to be determined within the CPA formalism) which describes the scattering by a single effective scatterer, as we have already explained. $\tilde{\Omega}^{nm}$ is given by equations (A4) and (A5) for $n \neq m$ and, by definition, equals zero for $n = m$.

We solve equation (A8) through a Fourier transformation as follows. Because of the 2D periodicity of the system, $\tilde{\mathbf{D}}^{nm}$ is given by a Fourier integral over the SBZ:

$$\tilde{\mathbf{D}}^{nm}(\omega) = \frac{1}{S_0} \int_{SBZ} \mathrm{d}^2 k_\parallel \exp(\mathrm{i} k_\parallel \cdot \boldsymbol{R}_{nm}) \mathbf{D}(k_\parallel; \omega) \tag{A9}$$

with

$$\mathbf{D}(k_\parallel; \omega) = \sum_{R_{nm}} \tilde{\mathbf{D}}^{nm}(\omega) \exp(-\mathrm{i} k_\parallel \cdot \boldsymbol{R}_{nm}). \tag{A10}$$

If we multiply (A8) by $\exp(-\mathrm{i} k_\parallel \cdot \boldsymbol{R}_{nm})$, take the sum over $\boldsymbol{R}_{nm}$ and use (A10), we finally obtain

$$\mathbf{D}(k_\parallel; \omega) = [\mathbf{I} - \Omega(k_\parallel; \omega) \langle \mathbf{T}(\omega) \rangle]^{-1} \Omega(k_\parallel; \omega) \tag{A11}$$

where

$$\Omega(k_\parallel; \omega) = \sum_{R_{nm} (\neq \mathbf{0})} \exp(-\mathrm{i} k_\parallel \cdot \boldsymbol{R}_{nm}) \tilde{\Omega}^{nm}(\omega). \tag{A12}$$

Substitution of (A11) into (A9) gives, when $n = m = 0$, equation (10).

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