# Macroscopic theory of the coupled dipole approximation method

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The coupled dipole approximation method was formulated by Purcell and Pennypacker to treat scattering by irregular bodies, and it has gained considerable currency in the recent past. Since the heuristic formulation of the method has a semi-microscopic flavour, here it has been given a firm macroscopic foundation by connecting it with volume integral equations. This communication also serves as a mini-review of the recent developments of this numerical technique.

#### 1. Introduction

Although scattering computations involving right circular cylinders and spheres can be carried out analytically and with considerable ease, analytical techniques for scattering by arbitrarily-shaped bodies are not available. Invariably, numerical approaches have to be resorted to in order to obtain the scattering characteristics of irregular bodies, homogeneous as well as inhomogeneous [1,2].

Any dielectric body can be thought of as consisting of elementary electric dipoles: this thought forms the basis of post-maxwellian developments in electromagnetic theory, wherein a transition from the microscopic to the macroscopic (continuum) fields is made [3]. Although the reverse transition (from the macroscopic to the microscopic) is not quite correct, it is widely utilized in constructing effective medium theories for composites [4].

That, however, is not the theme of this communication. Instead, attention is focussed on a semi-microscopic method for computing scattering from irregular objects. This numerical technique is variously known as the *coupled dipole approximation method* [5] and the *Purcell-Pennypacker method* [6] after its originators [7]. Originally developed for understanding scattering by interstellar dust particles, this method has gained considerable currency in the recent past. As it stands, the method was formulated [7] on a purely heuristic basis. The object here is to provide it with a firm macroscopic basis by connecting it with volume integral equation methods. In addition, the sequel will also serve as a mini-review of recent developments of this technique.

#### 2. Preliminary

For the sake of simplicity, we will consider a homogeneous dielectric scatterer of arbitrary shape embedded in free space in the sequel. Let the scatterer be modelled by a collection of spherical subunits; without loss of generality, let the subunits, each of radius *a* and dielectric constant  $\epsilon_r \epsilon_0$ , be identical. Then, since the *n*th subunit is electrically small, it can be modelled as a point electric dipole; hence,

$$\boldsymbol{p}_n = \alpha \boldsymbol{E}_{\mathrm{exc},n} \,, \tag{1}$$

where  $\alpha = 4\pi a^3 \epsilon_0(\epsilon_r - 1)/(\epsilon_r + 2)$  is the Clausius-Mosotti polarizability [8] of the spherical subunit,  $E_{\text{exc},n}$  is the electric field *exciting* the *n*th subunit, and  $p_n$  is the *equivalent* electric dipole moment.

The field radiated by an electric dipole in free space can be computed in terms of the free space dyadic Green's function [9]. The field exciting the *n*th subunit is composed of the field *actually incident* on the dielectric scatterer plus the fields *scattered* by all the other spherical subunits. Thus, the *exciting* field on the *n*th subunit has to be computed by solving the dyadic equation

$$\boldsymbol{E}_{\text{exc},n} = \boldsymbol{E}_{\text{inc}}(\boldsymbol{r}_n) + \sum_{m \neq n} \left( \omega^2 \mu_0 \boldsymbol{\mathfrak{G}}_{nm} \right) \cdot \boldsymbol{\alpha} \boldsymbol{E}_{\text{exc},m} , \qquad (2)$$

in which  $\mathbf{r}_n$  is the location of the center of the *n*th spherical subunit,  $E_{inc}$  is the incident field,  $E_{exc,n}$  is the exciting field for the *n*th subunit, the interaction dyadic  $\mathfrak{G}_{nm} = \mathfrak{G}_0(\mathbf{r}_n, \mathbf{r}_m)$ ,

$$\mathfrak{G}_{0}(\mathbf{r},\mathbf{r}') = (\Im + \mathcal{V}/k_{0}^{2}) [\exp(ik_{0}|\mathbf{r}-\mathbf{r}'|)/4\pi|\mathbf{r}-\mathbf{r}'|], \qquad (3)$$

is the free space Green's dyadic,  $k_0 = \omega \sqrt{\epsilon_0 \mu_0}$  is the free space wavenumber, and  $\Im$  is the identity dyadic. It is to be noted that (2) can be written in a matrix notation. Once it has been solved using matrix algebra, the scattered field at any location r outside the dielectric body can be easily computed as

$$\boldsymbol{E}_{\mathrm{sca}}(\boldsymbol{r}) = \omega^2 \mu_0 \alpha \sum_m \mathfrak{G}_0(\boldsymbol{r}, \boldsymbol{r}_m) \cdot \boldsymbol{E}_{\mathrm{exc.}m} \,. \tag{4}$$

## 3. Analysis

In order to provide a macroscopic theory for the coupled dipole approximation method, we turn our attention to the electric field integral equation [2, 10-13].

Let the bounded region V, occupied by a dielectric medium ( $\epsilon_r \epsilon_0$ ,  $\mu_0$ ) be embedded in free space ( $\epsilon_0$ ,  $\mu_0$ ). In the absence of source currents and charges, and using a harmonic time-dependence exp( $-i\omega t$ ), the Maxwell curl equations everywhere can be written as

 $\nabla \times \boldsymbol{E} - i\omega\mu_0 \boldsymbol{H} = 0, \quad \nabla \times \boldsymbol{H} + i\omega\epsilon_0 \boldsymbol{E} = \boldsymbol{J}, \quad (5a,b)$ 

where

$$\boldsymbol{J}(\boldsymbol{r}) = -i\omega\epsilon_0(\epsilon_r - 1) \boldsymbol{E}(\boldsymbol{r}), \quad \boldsymbol{r} \in \boldsymbol{V},$$
 (6a)

$$\boldsymbol{J}(\boldsymbol{r}) = 0, \quad \boldsymbol{r} \notin \boldsymbol{V}. \tag{6b}$$

Thus, the influence of the dielectric scatterer is being treated as that due to a certain volume distribution of the electric current density in free space [10,11].

The procedure of solving (5a, b) has been detailed by Jones [10], and results in the electric field and the magnetic field integral equations that have been extensively utilized in the area of numerical electromagnetics. Let  $E_{inc}$  and  $H_{inc}$  represent the (homogeneous) solutions of (5a, b) when J=0 everywhere; in other words,  $\{E_{inc}, H_{inc}\}$  denote the field incident on the scatterer. Then, the solution of (5a, b) is obtained as [9-11]

$$\boldsymbol{E}(\boldsymbol{r}) - \boldsymbol{E}_{\text{inc}}(\boldsymbol{r}) = \mathrm{i}\omega\mu_0 \int_{\boldsymbol{v}} \mathrm{d}\boldsymbol{v}' \, \mathfrak{G}_0(\boldsymbol{r}, \boldsymbol{r}') \cdot \boldsymbol{J}(\boldsymbol{r}'), \quad (7a)$$

$$\boldsymbol{H}(\boldsymbol{r}) - \boldsymbol{H}_{\rm inc}(\boldsymbol{r}) = \boldsymbol{V} \times \int_{\boldsymbol{v}} d\boldsymbol{v}' \, \boldsymbol{\mathfrak{G}}_0(\boldsymbol{r}, \boldsymbol{r}') \cdot \boldsymbol{J}(\boldsymbol{r}') \,. \tag{7b}$$

It should be noted that (7a, b) hold for all points r, inside as well as outside V. Further, as mentioned by Jones [10], (7a, b) satisfy (i) the continuity of the tangential components of the E and the H fields across the boundary of V, and (ii) the radiation condition at infinity.

Since the scatterer is dielectric, we concentrate on (7a) now onwards [11]. When r lies inside V, then (7a) is a volume integral equation of the second kind. Noting that  $\mathfrak{G}_0(\mathbf{r}, \mathbf{r}')$  is singular at  $\mathbf{r}=\mathbf{r}'$  [14], this equation can be transformed to

$$E(\mathbf{r}) = E_{inc}(\mathbf{r}) + \mathbf{J}(\mathbf{r}) / (3i\omega\epsilon_0)$$
  
+ PV  $\left(i\omega\mu_0 \int_{\nu} d\nu' \, \mathbf{J}(\mathbf{r}') \cdot \mathfrak{G}_0(\mathbf{r}', \mathbf{r}) - \mathbf{J}(\mathbf{r}) / (3i\omega\epsilon_0)\right)$   
 $\mathbf{r} \in V,$  (8)

in which PV denotes the principal value, while the singularity at r=r' has been clearly identified. Once the solution of (8) has been obtained, it can be substituted in (7a, b) to find the field scattered by the dielectric obstacle into free space.

Eq. (8) provides the *macroscopic* basis of the coupled dipole approximation method. Let the volume V be broken up into mutually disjoint volumes  $V_n$  (n=1, 2, ..., N), and let  $r_n$  denote the center of  $V_n$ . It is then assumed that the field is constant *inside*  $V_n$ , i.e.,  $E(r) = E(r_n) = E_n$  for  $r \in V_n$ . Then (8) simplifies to

$$E_n = [3/(\epsilon_r + 2)] E_{inc}(r_n) + \sum_{m \neq n} (\omega^2 \mu_0 \alpha_m) \mathfrak{G}_{nm} \cdot E_m, \qquad (9)$$

where

$$\alpha_n = 3V_n \epsilon_0(\epsilon_r - 1) / (\epsilon_r + 2) , \qquad (10)$$

it being assumed that all volumes  $V_n$  have sufficiently isotropic shapes.

In order to convert (9) into (2), the correct iden-

tification of  $E_n$  is necessary. This  $E_n$  is the field existing inside the dielectric volume  $V_n$ . Since  $V_n$  is electrically small, there is no harm in thinking of it as being spherical. This allows  $\alpha_n$  to formally have the status of the Clausius-Mosotti polarizability of a dielectric sphere. Next, for example, in section (9.5) of van Bladel [15], it is shown that if  $E_{exc}$  excites a small dielectric sphere, the field inside the sphere is given by  $[3/(\epsilon_r+2)] E_{exc}$ . Therefore, the field exciting the spherical  $V_n$  can be obtained from (9) as

$$\boldsymbol{E}_{\mathrm{exc},n} = \boldsymbol{E}_{\mathrm{inc}}(\boldsymbol{r}_n) + \sum_{m \neq n} \left( \omega^2 \mu_0 \boldsymbol{\mathfrak{G}}_{nm} \right) \cdot \boldsymbol{\alpha}_m \boldsymbol{E}_{\mathrm{exc},m} \,, \quad (11)$$

which is the core of the Purcell-Pennypacker formalism as well as of its variants. In particular, let all  $V_n$  be identical in volume so that each can be thought of in terms of the equivoluminal of radius *a*. It is easy to see then that  $\alpha_n$  of (11) is the  $\alpha$  of (2).

### 4. Discussion

In the coupled dipole approximation method, there is no need that the scatterer be homogeneous. Indeed, the only restriction is that each spherical subunit be so. This is also borne out by Jones' treatment of (7a, b).

Further, the spherical subunits may have magnetic properties and chiral properties as well, which results in a coupling of electric and magnetic fields [16]. The subunits may even be anisotropic [17], and even nonspherical [5,18]. Even the scattering object need not be compact: it may simply be an agglomerate of distinct particles [19,20].

The straightforward solution of (2) requires the inversion of a matrix [5,17], which puts considerable strain on computing facilities and introduces errors. To some extent, errors arising from matrix inversion algorithms can be minimized by using the conjugate gradient method [21], and the storage requirements can be reduced by exploiting any geometric symmetries the scatterer and the incident field may have [22,23]. The matrix itself may be bypassed by resorting to iterative solution techniques [7,20], or by taking recourse to an order-of-scattering approximation [24-26]; convergence of the an-

swer has to be carefully checked, however.

Inevitably, the coupled dipole approximation method has its limitations, besides the matrix storage and inversion problems. These become apparent when the subunits are either (i) strong scatterers by themselves, in which case the off-diagonal interaction terms (represented by  $\mathfrak{G}_{nm}$  in (2)) may overshadow the diagonal terms; or (ii) strong absorbers in which case the internal fields  $E_n$  may be highly concentrated in some regions.

Ill-conditioning from either of these factors is intensified if the subunits lie in close proximity of each other. Thus, as of now, a treatable scatter may not have electrically large dimensions, nor can its properties contrast sharply from that of free space (i.e., the ambient medium). The method, however, may be ideally suitable for tenuous scatterers such as fractal clusters [20,27], carbonaceous smoke [28,29], snow crystals [30], cirrus clouds [31], pharmacological suspensions [32], macromolecules [33] comet tails [34], particulate surfaces [35] and interstellar particulate matter [7,21].

In dealing with compact scatterers, considerable errors stems from the piecewise constant assumption of the internal fields. This situation is reminiscent of similar problems in the method of moments [36]. If, however, the sole aim is to obtain the scattered field in the far zone, then this error may have only a limited effect.

A greater problem is demonstrated by the non-satisfaction [6,21] of the optical theorem [15] when the incident field is a plane wave and  $\epsilon_r$  is purely real. Even an isolated spherical subunit must attenuate the incident plane wave so that the equivalent dipole moment cannot be in phase with the incident plane wave. But a real refractive index implies a purely real Clausius-Mosotti polarizability  $\alpha$ . Hence, the  $\alpha$  to be used in (2) and (4) must have an imaginary component.

A complex polarizability can be provided in at least two ways. Instead of obtaining  $\alpha$  from the Clausius-Mosotti formula [8], it can be obtained from the lowest order term in the Mie solution itself [37]. Or, a radiative correction may be done by replacing the Clausius-Mosotti  $\alpha$  by  $\alpha/[1-(1/6\pi\epsilon_0)ik_0^3\alpha]$  as has been done by Draine [21] <sup>#1</sup>.

<sup>&</sup>lt;sup>#1</sup> Draine [21] has used gaussian units, as also have some others.

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There has been yet another development to make the coupled dipole method satisfy the optical theorem. Since the spherical subunits are usually arranged uniformly on a cartesian lattice, in truth each subunit is a cube. This has been utilized as a correction [6] to the diagonal terms in the matrix involved in (9)  $^{#2}$ .

In the introductory section, it was mentioned that the coupled-dipole approximation method is a *semi*microscopic method. In order to justify that appellation, consider (2) rewritten as

$$\boldsymbol{p}_n/\boldsymbol{\alpha} = \boldsymbol{E}_{\text{inc}}(\boldsymbol{r}_n) + \sum_{m \neq n} \left( \omega^2 \mu_0 \boldsymbol{\mathfrak{G}}_{nm} \right) \cdot \boldsymbol{p}_m \,. \tag{12}$$

Thus, (12) has a *microscopic* basis since it represents a collection of discrete dipoles in free space immersed in the incident field  $E_{inc}$ . In fact, (12) is used in section 2.4 of Born and Wolf's book [38] and elsewhere [e.g., 39–41] to determine the equivalent, and therefore *macroscopic*, properties of (dielectric) matter.

On the other hand, Purcell and Pennypacker [7] used the Clausius-Mosotti polarizability [8] involving the dielectric constant in (2). The dielectric constant is a macroscopic entity. Hence, it would no matter how large N (the number of spherical units) and how small a (the subunit dimension) be, the subunits remain pieces of a continuum and can never become molecules in (2). Were (12) to be used with even an estimated molecular polarizability instead of the Clausius-Mosotti polarizability, the Purcell-Pennypacker formulation would indeed be microscopic. As it is, the use of the macroscopic dielectric constant in  $\alpha$  suggests that the formulation is only a semi-microscopic approach to scattering.

In summary, it is to be noted that a firm macroscopic basis has been provided for the coupled dipole approximation method, by connecting (2) with the electric field integral equation. Thus, this method has been connected with the various other numerical techniques commonly used for scattering problems.

Finally, at the suggestion of an anonymous reviewer and in the context of composite and inhomogeneous materials, attention is drawn to the proceedings of two conferences – Electronic, Transport and Optical Properties of Inhomogeneous Media. It must be that the enormous proliferation of scientific literature in recent years is responsible for the absence of these proceedings from the shelves of the Penn State Libraries.

This work is affectionately dedicated to Craig Frederic Bohren, Distinguished Professor of Meteorology at the Pennsylvania State University, on the occasion of his 50th birthday.

#### References

- [1] R. Mittra (Ed.), Numerical and asymptotic techniques in electromagnetics (Springer-Verlag, New York, 1975).
- [2] R.F. Harrington, Field computation by moment methods (MacMillan, New York, 1968).
- [3] J.Z. Buchwald, From Maxwell to microphysics (Univ. Chicago Press, Chicago, 1985).
- [4] L. Ward, The optical constants of bulk materials and films (Adam Hilger, Bristol, 1989).
- [5] S.B. Singham and G.C. Salzman, J. Chem. Phys. 84 (1986) 2658.
- [6] G.H. Goedecke and S.G. O'Brien, Appl. Optics 27 (1988) 2431.
- [7] E.M. Purcell and C.R. Pennypacker, Astrophys. J. 186 (1973) 705.
- [8] C. Kittel, Introduction to solid state physics (Wiley Eastern, New Delhi, 1974).
- [9] H.C. Chen, Theory of electromagnetic waves (McGraw-Hill, New York, 1983).
- [10] D.S. Jones, J. Inst. Math. Applics. 23 (1979) 421.
- [11] C.T. Tai, Electromagnetics 5 (1985) 79.
- [12] K.M. Chen, Interaction of electromagnetic fields with biological bodies, in: Research topics in electromagnetic wave theory, ed. J.A. Kong (Wiley, New York, 1980).
- [13] D.S. Jones, Methods in electromagnetic wave propagation (Oxford Univ. Press, Oxford, 1979).
- [14] W. Weiglhofer, Amer. J. Phys. 57 (1989) 455.
- [15] J. van Bladel, Electromagnetic fields (Hemisphere, Washington, D.C., 1985).
- [16] S.B. Singham, Chem. Phys. Lett. 130 (1986) 139.
- [17] V.V. Varadan, A. Lakhtakia and V.K. Varadan, IEEE Trans. Antennas Propagat. 37 (1989) 800.
- [18] V.K. Varadan, A. Lakhtakia and V.V. Varadan, J. Wave-Mater. Interact. 2 (1987) 153.
- [19] A. Lakhtakia, R. Messier, V.V. Varadan and V.K. Varadan, J. Phys. A: Math. Gen. 20 (1987) 1615.
- [20] J. Frey, J.J. Pinvidic, R. Botet and R. Jullien, J. Phys. France 49 (1988) 1969.
- [21] B.T. Draine, Astrophys. J 333 (1988) 848.

<sup>&</sup>lt;sup>#2</sup> Goedecke and O'Brien [6] have actually worked with (9). This should be contrasted with (2), which represents an exciting field formalism.

- [22] P.J. Flatau, G.L. Stephens and B.T. Draine, J. Opt. Soc. Am. A 7 (1990) 693.
- [23] Y.L. Yung, Appl. Optics 17 (1978) 3707.
- [24] P. Chiapetta, J. Phys. A: Math. Gen. 13 (1980) 2101.
- [25] S.B. Singham and C.F. Bohren, Optics Lett. 12 (1988) 10.
- [26] S.B. Singham and C.F. Bohren, J. Opt. Soc. Am. A 5 (1988) 1967.
- [27] J. Nelson, J. Mod. Optics 36 (1989) 1031.
- [28] I. Colbeck, E.J. Hardman and R.M. Harrison, J. Aerosol Sci. 20 (1989) 765.
- [29] H.Y. Chen, M.F. Iskander and J.E. Penner, J. Mod. Opt. 37 (1990) 171.
- [30] S.G. O'Brien and G.H. Goedecke, Appl. Optics 27 (1988) 2439.
- [31] P.J. Flatau, G.L. Stephens and B.T. Draine, Radiative properties of visible and subvisible cirrus: scattering on hexagonal ice crystals, in: first ISSCP Regional Experiment

Science Team Meeting, NASA Conf. Publ. (NASA, Langley, Virginia, to be published), cited in ref. [22].

- [32] H.B. Bohidar, Colloid Polym. Sci. 267 (1989) 159.
- [33] S. Zietz, A. Belmont and C. Nicolini, Cell Biophys. 5 (1983) 163.
- [34] G.A. Shah, Bull. Astr. Soc. India 17 (1989) 114.
- [35] M. Wolff and A. Dollfus, Appl. Optics 29 (1990) 1496.
- [36] H. Massoudi, C.H. Durney and M.F. Iskander, IEEE Trans. Microwave Theory Tech. 34 (1986) 653.
- [37] W.T. Doyle, Phys. Rev. B 39 91989) 9852.
- [38] M. Born and E. Wolf, Principles of optics (Pergamon, Oxford, 1980).
- [39] F.E. Fedorov, Theory of gyrotropy (in russian) (Nauka i Technika, Minsk, 1976).
- [40] J.J. Sein, Optics Comm. 2 (1970) 170.
- [41] D. Bedeaux and P. Mazur, Physica 67 (1973) 23.