ELSEVIER

Physica A 241 (1997) 72–76

PHYSICA A

Momentum-dependent electromagnetic T-matrix and dynamic effective properties of random media

Yves-Patrick Pellegrini *, Pascal Thibaudeau, D. Brian Stout

Commissariat à l'Energie Atomique, CESTA, BP 2, 33114 Le Barp, France

Abstract

We discuss the problem of the dynamic dielectric and magnetic response on the whole frequency range of a composite material. We derive exact one-body expressions of the momentum-dependent mean constitutive kernels, for an assembly of dielectric and magnetic spheres of finite size, on the basis of their T-matrix. The effective homogeneous-like constitutive constants are defined, and used in a coherent potential approximation. In a second step, we solve the dispersion relation in order to extract observable quantities.

PACS: 41.20.-q; 42.25.-p

Keywords: Disordered media; Dynamic dielectric and magnetic response; Effective response kernel; Momentum-dependent T-matrix; Coherent potential approximation; Spatial dispersion; Eddy currents; Induced diamagnetism

In spite of several attempts [1–3], it turns out that, at the present time, no clear cut expressions of the one-body order effective dynamic dielectric and magnetic constants of a composite random medium containing spheres with constitutive parameters (ε_s, μ_s), embedded in a background homogeneous medium (hereafter referred to as "bare") defined by parameters (ε_m, μ_m), are available in the literature. Though we benefit, thanks to Mie, from the knowledge of the exact solution of the one-sphere problem, this situation is partly due to considerable confusion about what "effective" dynamic constitutive parameters of such a medium ought to be, owing to spatial dispersion effects. Our purpose here is to discuss this uncomfortable situation, and propose a new perspective, which we subsequently develop with the help of the coherent potential approximation (CPA).

It is well known that statistical disorder induces spatial dispersion in the response kernels [4]. If $\mathbf{D}(\mathbf{r}) = \varepsilon_0\varepsilon(\mathbf{r})\mathbf{E}(\mathbf{r})$ and $\mathbf{B}(\mathbf{r}) = \mu_0\mu(\mathbf{r})\mathbf{H}(\mathbf{r})$ in a realization of the

* Corresponding author. Fax: 193351715427; e-mail: ypp@geocub.greco-prog.fr.

PII S0378-4371(97)00061-7

medium, then the statistical averages of these fields (denoted by $\langle\cdot\rangle$) are linked in an infinite medium by convolution products involving dyadic translation-invariant response kernels. In the Fourier representation we have, *by definition of the following kernels* (we work in the time-harmonic regime, and we omit hereafter the ω-dependence): $\langle\mathbf{D}\rangle(\mathbf{k}) = \varepsilon_0\bar{\bar{\varepsilon}}_e(\mathbf{k})\langle\mathbf{E}\rangle(\mathbf{k})$ and $\langle\mathbf{B}\rangle(\mathbf{k}) = \mu_0\bar{\bar{\mu}}_e(\mathbf{k})\langle\mathbf{H}\rangle(\mathbf{k})$. The divergence-free character of $\mathbf{B}$ implies that we can write $\bar{\bar{\mu}}_{e,ij}(\mathbf{k}) = \mu_e(k)\mathsf{P}_{\perp,ij}(\mathbf{k})$, with $\mathsf{P}_\perp(\mathbf{k})$ the transverse projector. By contrast, the dyadic permittivity kernel writes (for isotropic disorder) $\bar{\bar{\varepsilon}}_{e,ij}(\mathbf{k}) = \varepsilon_\perp(k)\mathsf{P}_{\perp,ij}(\mathbf{k}) + \varepsilon_\parallel(k)\mathsf{P}_{\parallel,ij}(\mathbf{k})$, where $\mathsf{P}_\parallel(\mathbf{k})$ is the longitudinal projector [4]. The spatial dispersion (i.e. the momentum dependence of the kernels) is what makes the difference between an averaged inhomogeneous medium and a homogeneous one. Its origin lies in the presence of the inhomogeneity length scale a (here, the sphere radius).

We denote by $\mathsf{G}_e = \langle\mathsf{G}\rangle$, the statistical average over the configurations of the electric Green function in the medium, related to a current source $i\omega\mu_0\mathbf{J}_s$. It completely determines $\langle\mathbf{E}\rangle$, and writes $\mathsf{G}_e = (\mathsf{G}_0^{-1} - \Sigma)^{-1}$, in terms of the so-called "self-energy" operator Σ and of the Green function of the bare medium G_0. Explicitly, $\Sigma_{ij}(\mathbf{k}) = \Sigma_\perp(k)\mathsf{P}_{\perp,ij}(\mathbf{k}) + \Sigma_\parallel(k)\mathsf{P}_{\parallel,ij}(\mathbf{k})$. Then

$$G_{e,ij}(\mathbf{k}) = \frac{1}{k^2/\mu_m - (\omega/c)^2\varepsilon_m - \Sigma_\perp(k)}P_{\perp,ij}(\mathbf{k}) - \frac{1}{(\omega/c)^2\varepsilon_m + \Sigma_\parallel(k)}P_{\parallel,ij}(\mathbf{k})\,. \tag{1}$$

The expression of G_0 is obtained by letting $\Sigma_{\perp,\parallel} \equiv 0$. The modes $k(\omega)$ which the fields can sustain are the solutions of the longitudinal and transverse dispersion equations, respectively, obtained as the roots of the denominators in the longitudinal and transverse parts of (1) – i.e., as the poles of the mean Greens function. Averaging and combining Maxwell's equations, one easily arrives at the identities

$$\begin{aligned} &k^2/\mu_m - (\omega/c)^2\varepsilon_m - \Sigma_\perp(k) = k^2/\mu_e(k) - (\omega/c)^2\varepsilon_\perp(k)\,,\\ &\varepsilon_m + (c/\omega)^2\Sigma_\parallel(k) = \varepsilon_\parallel(k)\,. \end{aligned} \tag{2}$$

They provide the transverse and longitudinal dispersion relations, respectively, when set to 0. Thus, the three kernels Σ, $\bar{\bar{\mu}}_e$ and $\bar{\bar{\varepsilon}}_e$ are not mutually independent. In particular, the self-energy does not alone determine both the average dielectric and magnetic properties of the medium. Its role is mainly to lock the fields (*and subsequently the response kernels*) at some particular values of k (which we hereafter call "observables"), via the dispersion equations, when one goes back to the direct space. We note that the longitudinal part of $\langle H\rangle(\mathbf{k})$, which does not exist in a homogeneous medium, cannot be determined by the Green function of the electric field and the average constitutive equations. We exclude it from the scope of this paper.

Let us compute the effective response kernels at one-body order. We define an electric (resp. magnetic) potential operator U_i^E (resp. U_i^M) for the ith scatterer. This permits us to expand in the direct space the operator $\varepsilon(\mathbf{r}|\mathbf{r}') \equiv \varepsilon(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}')$ (resp. $\delta(\mathbf{r} - \mathbf{r}')\nabla' \times \mu^{-1}(\mathbf{r}')\nabla'\times$) as a sum of one-body operators. Setting $\mathsf{U}_i = \mathsf{U}_i^E + \mathsf{U}_i^M$, we write down the familiar multiple-scattering series: $\mathsf{G} = \mathsf{G}_0 + \mathsf{G}_0 \sum_i \mathsf{U}_i \mathsf{G} = \mathsf{G}_0 + \mathsf{G}_0 \sum_i \mathsf{T}_i \mathsf{G}_0 + \mathsf{G}_0 \sum_{j \neq i} \mathsf{T}_i \mathsf{G}_0 \mathsf{T}_j \mathsf{G}_0 + \cdots$, wherein the individual T-matrices are $\mathsf{T}_i = \mathsf{U}_i(1 - \mathsf{G}_0 \mathsf{U}_i)^{-1}$ [5].

The average permittivity kernel is given, according to its above definition, by $\bar{\bar{\varepsilon}}_e = \langle \varepsilon \mathsf{G} \rangle \langle \mathsf{G} \rangle^{-1}$. Therefore, expanding this relation at one-body order yields, in the thermodynamic limit and at constant density of scatterers $n = N/V$ (N is the number of scatterers, V, the volume of the system. Each average $\langle \cdot \rangle$ is proportional to $1/V$)

$$\bar{\bar{\varepsilon}}_e(\mathbf{k}) = \varepsilon_m + N(c/\omega)^2 \left(\langle \mathsf{U}_i^E \rangle + \langle \mathsf{U}_i^E \mathsf{G}_0 \mathsf{T}_i \rangle \right)(\mathbf{k}) + O(n^2) . \tag{3}$$

In a similar way, we obtain, by using averaged Maxwell equations:

$$\frac{1}{\mu_e(k)} = \frac{1}{\mu_m} - \frac{N}{2k^2} \mathrm{Trace} \left\{ \left(\langle \mathsf{U}_i^M \rangle + \langle \mathsf{U}_i^M \mathsf{G}_0 \mathsf{T}_i \rangle \right)(\mathbf{k}) \right\} + O(n^2) . \tag{4}$$

This is consistent with the familiar result $\Sigma = N \langle \mathsf{T}_i \rangle + O(n^2)$ and relations, Eq. (2), since by definition, $\mathsf{T}_i = \mathsf{U}_i + \mathsf{U}_i \mathsf{G}_0 \mathsf{T}_i = (\mathsf{U}_i^E + \mathsf{U}_i^E \mathsf{G}_0 \mathsf{T}_i) + (\mathsf{U}_i^M + \mathsf{U}_i^M \mathsf{G}_0 \mathsf{T})$. The explicit computation of these two $\mathbf{k}$-dependent response kernels via that of T_i, in terms of a Mie dyadic series expansion over the vector spherical harmonics basis, constitutes our main result [6].

We can now address the question of effective parameters in their usual sense, that is, as k-independent constants describing the average medium (both in its longitudinal and transverse response) as if it were homogeneous. Strictly speaking, such quantities do not exist (i.e. are not observable) because of the dispersion relations. On the other hand, they are required for the conventional CPA treatment of the one-body problem, since all our T-matrix calculations have been performed starting from a homogeneous bare medium. The main problem we face is that, in opposition to the homogeneous case, *different* permittivities appear in the longitudinal and transverse parts of the mean Greens function at *any* (real) finite k. Statistical isotropy however implies that $\varepsilon_{\parallel}(k = 0) = \varepsilon_{\perp}(k = 0)$. This common value thus emerges as the *only* possible choice for the effective permittivity constant ε_{eff}. The spatial dispersion obscures what the correct choice for μ_{eff} would be. Indeed, the formulation of the problem in an infinite medium is invariant under the transformations $1/\mu_e(k) \to 1/\mu'_e(k) = 1/\mu_e(k) + (\omega/c)^2 \delta\varepsilon_{\perp}(k)/k^2$, $\varepsilon_{\perp}(k) \to \varepsilon'_{\perp}(k) = \varepsilon_{\perp}(k) - \delta\varepsilon_{\perp}(k)$, where $\delta\varepsilon_{\perp}(k)$ is any part of $\varepsilon_{\perp}(k)$ proportional to k^2. These transformations are accompanied by related new definitions for the fields $\langle \mathbf{D} \rangle$ and $\langle \mathbf{H} \rangle$, which correspond to a re-shuffling of the currents [2,7]. Performing such transformations, and taking μ_{eff} as the limit $k \to 0$ of the result for the obtained $\mu_e(k)$, are clearly non-commuting operations. We therefore imposed as a guide the physical constraints that μ_{eff}: (1) be finite when either ε_s or $\mu_s \to 0$; (2) remain finite when $\omega \to \infty$; and (3) obey the Kramers–Kronig relations, with positive imaginary parts. These led us to a final form of μ_{eff} matching that of ε_{eff}, with ε_m, ε_s replaced by μ_m, μ_s (and vice versa). Direct coupling terms between the dielectric and magnetic response are absent in our both ε_{eff} and μ_{eff}, these quantities being coupled only through common spherical functions $h_1^{(1)}(ak_s)$ and $j_1(ak_m)$.

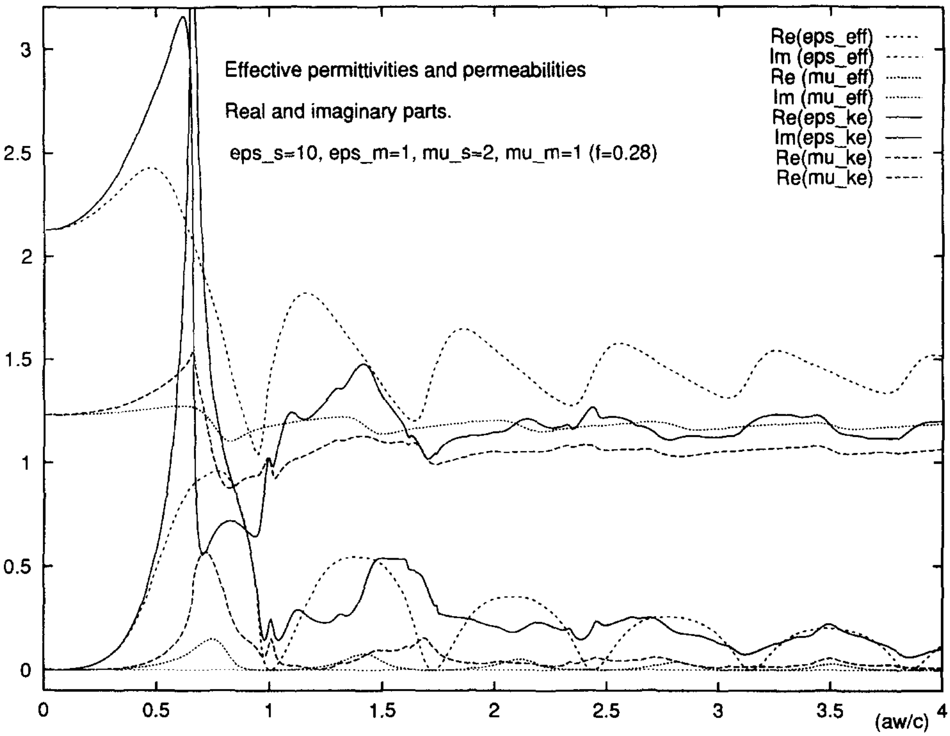

Fig. 1. CPA ($\varepsilon_{\rm eff}$, $\mu_{\rm eff}$) and final (ε_{k_e}, μ_{k_e}) effective parameters as a function of the reduced frequency ($a\omega/c$) (see text). The inner constitutive parameters are $\varepsilon_s = 10$, $\mu_s = 2$, and the outer ones are $\varepsilon_m = \mu_m = 1$. The volume concentration of spheres is $f = 0.28$. The same line style applies to the real and imaginary part of the same quantity. The imaginary parts begins at $(0, 0)$.

On the basis of these $\varepsilon_{\rm eff}$ and $\mu_{\rm eff}$, and of the $O(n)$ correction they define, we worked out a self-consistent CPA calculation (that is, a dynamic version of the Bruggeman formula [1]), in which the matrix is described as zero-radius spheres embedded with the finite-sized ones in the effective medium. We found the iterative solution of the system for the self-consistent $\varepsilon_{\rm eff}$ and $\mu_{\rm eff}$ to always converge for any concentration of spheres, at any frequency. Using the obtained values as the background effective medium values, we were then able to iteratively solve at any frequency the transverse dispersion relation (which now involves two different types of inclusions) for an observable mode k_e, up to the volume concentration $f = 0.28$ of finite-sized spheres (for example, Fig. 1). This value of k_e was then substituted into Eqs. (3) and (4) in order to obtain the (observable) effective permittivity and permeability constants $\varepsilon_{k_e} = \varepsilon_{\perp}(k_e)$ and $\mu_{k_e} = \mu_e(k_e)$ pertaining to this transverse mode (we summed 50 terms in the Mie series) [6].

In this theory, it is only in the last step that we can recover, for instance, the dielectric–magnetic coupling leading to an effective diamagnetic permeability in a composite made of metal spheres [2,3] (not shown here). However, as discussed above, an always possible re-shuffling of the currents makes different values for the couple $(\varepsilon_{k_e}, \mu_{k_e})$ equally acceptable, even though some combinations lead to a negative imaginary part in μ_{k_e} in the high-frequency regime [7]. The problem of finding a combination with an always positive imaginary part in the whole frequency range in the metallic case (which may be of better experimental relevance), and that of numerically computing k_e for any concentration, are currently under investigation.

References

[1] D. Stroud and F.P. Pan, Phys. Rev. B 27 (1983) 5098; P. Chýlek and V. Srivastava, Phys. Rev. B 30 (1984) 1008; G.D. Mahan, Phys. Rev. B 38 (1988) 9500; K. Busch and C.M. Soukoulis, Phys. Rev. Lett. 75 (1995) 3442.
[2] A.N. Lagarkov et al., J. Electr. Waves Appl. 6 (1992) 1159.
[3] D. Rousselle et al., J. Appl. Phys. 74 (1993) 475.
[4] P. Sheng, Introduction to Wave Scattering, Localization and Mesoscopic Phenomena (Academic Press, New York, 1995) and references therein; R. Balian and J.-J. Niez, J. Phys. I France 5 (1995) 7; M. Barthélémy, H. Orland and G. Zérah, Phys. Rev. E 52 (1995) 1123; Y.-P. Pellegrini, P. Thibaudeau and D.B. Stout, in: Proc. Journées Maxwell '95, Bordeaux-Lac, France, ed. J.-F. Eloy (CEA-CESTA, BP2, 33114 LE BARP, France, 1996) p. 335.
[5] L. Tsang and J.A. Kong, J. Appl. Phys. 51 (1980) 3465.
[6] Y.-P. Pellegrini, D.B. Stout and P. Thibaudeau, in preparation.
[7] P.C. Martin, Phys. Rev. 161 (1967) 143.