

# Dyson equation for electromagnetic scattering of heterogeneous media with spatially disordered particles: properties of the effective medium

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**Abstract.** In this paper, we consider the coherent component of the electromagnetic wave field inside random media. The subject of our interest concerns a random medium, consisting of a statistical ensemble of different scattering species and artificial material structures developed on base of dielectric or metallic resonant or non-resonant particles. The starting point of our theory is the multiple scattering theory, the averaged electric field satisfies a Dyson equation with a mass operator related to the effective dielectric permittivity of the homogenized structure. Quantum multiple scattering theory has been transposed into this electromagnetic case. We give a formal solution for the mass operator by introducing the T-matrix formalism. We show that the T-matrix satisfies a Lippman-Schwinger equation. Then, we introduce the Quasi-Crystalline Coherent Potential Approximation (QC-CPA), which takes into account the correlation between the particles with a pair-distribution function. The mass operator includes geometric effects, caused by resonant behavior due to the shape and size of particles, cluster effects because of correlations between particles. Significant modifications of particle scattering properties can be observed.

## 1. Introduction

The first intent of this paper is to discuss the relation between the Dyson equation for electromagnetic scattering of heterogeneous random media and the effective dielectric constant which characterizes the coherent part of an electromagnetic wave propagating inside a random medium containing metallic or dielectric particles. In this paper we show that we can obtain a general expression of the effective dielectric constant, which is the solution of a closed system of equations. In the high frequency limit the expression includes the vectorial case generalization of the result obtained by Keller, which has been derived, using a scalar theory. We obtain an important tensorial generalization of the notion of effective permittivity. The description of electromagnetic waves propagation in random media in terms of the properties of the constituents has been studied extensively in the past decades [1-19]. In most of published works, the basic idea is to calculate several statistical moments of the electromagnetic field to understand how the wave can interact with the the random medium [7, 9, 10, 11, 14, 15].

In the second section of this paper, we are concerned by the first moment which is the average electric field. Under some assumptions, it can be shown that the average electric field propagates as if the medium where homogeneous but with a renormalized permittivity, called effective permittivity. We introduce the multiple scattering formalism, the Dyson Equation and the Mass operator. We discuss the different hypotheses under which the effective medium theory is valid. The calculation of this parameter has a long history which dates back from the work of Clausius-Mossotti and Maxwell

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Garnett [19]. Most of the studies are concerned with the quasi-static limit where retardation effects are disregarded [17-24]. In order to take into account scattering effects, quantum multiple scattering theory has been transposed into the electromagnetic case [5, 7, 9, 10, 11, 14, 15], but as a rigorous analytic solution cannot be derived, several approximation schemes have been developed [5, 7, 10, 11, 14, 15, 25-36]. In section 3, we describe the Quasicrystalline Coherent Potential Approximation (QC-CPA) which takes into account the correlation between the particles [12, 14, 25-28]. In this section we examine the different steps to obtain the system of equations verified by the effective permittivity under the (QC-CPA) approach. In section 4, we add some new approximations to the (QC-CPA) approach which give us a tractable equation for the effective permittivity. The expression obtained contains the low frequency limit of the (QC-CPA) approach. At this limit, the (QC-CPA) equations can be written as a generalized Maxwell Garnett formula and are proven to be in good agreement with the experimental results [13, 14, 37-39]. Furthermore, the formula obtained contains also the approximate formula due to Keller, which has been derived in using scalar theory, but seems to be in good agreement with the experimental data for particles larger than a wavelength [40], these results are described in section 5. In section 6, we discuss the different results of this paper and the possible extension to magnetic response of the theory of Dyson equation for multiple scattering in random media with disordered high-index dielectric scatterers. This opens up the developing field of all-dielectric nanophotonics which allows to control both magnetic and electric response of structured matter.

## 2. Dyson equation and effective permittivity

In the following, we consider harmonic waves with  $e^{-i\omega t}$  pulsation. We consider a disordered ensemble of  $N \gg I$  identical spheres of radius  $r_s$  with dielectric function  $\epsilon_s(\omega)$  within an infinite medium with dielectric function  $\epsilon_1(\omega)$ . The field produced at  $\mathbf{r}$  by a discrete source located at  $\mathbf{r}_0$  is given by the dyadic Green function  $\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0, \omega)$ , which verifies the following propagation equation:

$$\nabla \times \nabla \times \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0, \omega) - \epsilon_V(\mathbf{r}, \omega) K_{vac}^2 \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0, \omega) = \delta(\mathbf{r} - \mathbf{r}_0) \overline{\mathbf{I}} \quad (1)$$

where  $K_{vac} = \omega/c$  with  $c$  the speed of light in vacuum and

$$\epsilon_V(\mathbf{r}, \omega) = \epsilon_1(\omega) + \sum_{j=1}^N [\epsilon_s(\omega) - \epsilon_1(\omega)] \Theta_s(\mathbf{r} - \mathbf{r}_j)$$

where  $\mathbf{r}_1, \dots, \mathbf{r}_N$  are the centers of the particles and  $\Theta_s$  the spherical particle shape:

$$\Theta_s(\mathbf{r}) = \begin{cases} 1 & \text{if } \|\mathbf{r}\| < r_s \\ 0 & \text{if } \|\mathbf{r}\| > r_s \end{cases} \quad (2)$$

The equation (1) has a unique solution if we impose the radiation condition at infinity. The multiple scattering process by the particles is mathematically decomposed in introducing the Green function  $\overline{\mathbf{G}}_1^\infty$ , which describes the propagation within an homogenous medium with permittivity  $\epsilon_1(\omega)$ , which verifies the following equation:

$$\nabla \times \nabla \times \overline{\mathbf{G}}_1^\infty(\mathbf{r}, \mathbf{r}_0, \omega) - \epsilon_1(\omega) K_{vac}^2 \overline{\mathbf{G}}_1^\infty(\mathbf{r}, \mathbf{r}_0, \omega) = \delta(\mathbf{r} - \mathbf{r}_0) \overline{\mathbf{I}} \quad (3)$$

with the appropriate boundary conditions. In an infinite random medium, we have [14, 41, 42]:

$$\overline{G}_1^\infty(\mathbf{r}, \mathbf{r}_0, \omega) = \left[ \overline{I} + \frac{\nabla \nabla}{K_1^2} \right] \frac{e^{i K_1 \|\mathbf{r} - \mathbf{r}_0\|}}{4\pi \|\mathbf{r}\|} \quad (4)$$

where  $K_1^2 = \epsilon_1(\omega) K_{vac}^2$ .

Using this Green function, we decompose the Green function  $\overline{G}(\mathbf{r}, \mathbf{r}_0, \omega)$  under the following form [5, 7, 9, 14, 15]:

$$\overline{G} = \overline{G}_1^\infty + \overline{G}_1^\infty \cdot \overline{V} \cdot \overline{G} \quad (5)$$

where the operator notation is used:

$$[\overline{A} \cdot \overline{B}](\mathbf{r}, \mathbf{r}_0) = \int d^3 r_1 \overline{A}(\mathbf{r}, \mathbf{r}_1) \cdot \overline{B}(\mathbf{r}_1, \mathbf{r}_0) \quad (6)$$

The potential  $\overline{V}$  which describes the interaction between the wave and the N particles, is given by:

$$\overline{V} = \sum_{i=1}^N \overline{v}_{r_i} \quad (7)$$

$$\overline{v}_{r_i}(\mathbf{r}, \mathbf{r}_0, \omega) = (2\pi)^2 \delta(\mathbf{r} - \mathbf{r}_0) \overline{v}_{r_i}(\mathbf{r}, \omega) \quad (8)$$

$$\overline{v}_{r_i}(\mathbf{r}, \omega) = [K_s^2 - K_1^2] \Theta_d(\mathbf{r} - \mathbf{r}_i) \overline{I} \quad (9)$$

with  $K_s^2 = \epsilon_s(\omega) K_{vac}^2$ . It is useful to introduce the T matrix defined by [5, 6, 7, 19, 12, 14, 15]:

$$\overline{G} = \overline{G}_1^\infty + \overline{G}_1^\infty \cdot \overline{T} \cdot \overline{G}_1^\infty \quad (10)$$

By iterating equation (5) and comparing it with the definition (10), we show that the T matrix verifies the following equation:

$$\overline{T} = \overline{V} + \overline{V} \cdot \overline{G}_1^\infty \cdot \overline{T} \quad (11)$$

If we introduce the T matrix for each scatterer by:

$$\overline{t}_{r_i} = \overline{v}_{r_i} + \overline{v}_{r_i} \cdot \overline{G}_1^\infty \cdot \overline{t}_{r_i} \quad (12)$$

we can decompose the T matrix for the system in a series of multiple scattering processes for the particles [6, 11, 13, 16]:

$$\overline{T} = \sum_{i=1}^N \overline{t}_{r_i} + \sum_{i=1}^N \sum_{j=1, j \neq i}^N \overline{t}_{r_j} \cdot \overline{G}_1^\infty \cdot \overline{t}_{r_i} + \dots \quad (13)$$

This T matrix is useful to calculate the average field  $\langle \overline{G} \rangle$  since we have:

$$\langle \overline{G} \rangle = \overline{G}_1^\infty + \overline{G}_1^\infty \cdot \langle \overline{T} \rangle \cdot \overline{G}_1^\infty \quad (14)$$

The equivalent of the potential operator  $\overline{V}$  for the average Green function  $\langle \overline{G} \rangle$  is the mass operator  $\overline{\Sigma}$  defined by the following equation:

$$\langle \overline{G} \rangle = \overline{G}_1^\infty + \overline{G}_1^\infty \cdot \overline{\Sigma} \cdot \langle \overline{G} \rangle \quad (15)$$

As in equation (11), we have the following relationship between the average T matrix and the mass operator:

$$\langle \bar{\mathbf{T}} \rangle = \bar{\Sigma} + \bar{\Sigma} \cdot \bar{\mathbf{G}}_1^\infty \cdot \langle \bar{\mathbf{T}} \rangle \quad (16)$$

or:

$$\bar{\Sigma} = \langle \bar{\mathbf{T}} \rangle \cdot \left[ \bar{\mathbf{I}} + \bar{\mathbf{G}}_1^\infty \cdot \langle \bar{\mathbf{T}} \rangle \right]^{-1} \quad (17)$$

The mass operator corresponds to all irreducible diagrams in the Feynman representation [5, 7, 9, 11]. The equation (15) written in differential form is:

$$\begin{aligned} \nabla \times \nabla \times \langle \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0, \omega) \rangle - \epsilon_1(\omega) K_{vac}^2 \langle \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0, \omega) \rangle \\ - \int d^3 \mathbf{r}_1 \bar{\Sigma}(\mathbf{r}, \mathbf{r}_1, \omega) \cdot \langle \bar{\mathbf{G}}(\mathbf{r}_1, \mathbf{r}_0, \omega) \rangle = \delta(\mathbf{r} - \mathbf{r}_0) \bar{\mathbf{I}}. \end{aligned} \quad (18)$$

For a statistical homogeneous medium we have:

$$\bar{\Sigma}(\mathbf{r}, \mathbf{r}_1, \omega) = \bar{\Sigma}(\mathbf{r} - \mathbf{r}_1, \omega) \quad (19)$$

$$\langle \bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}_0, \omega) \rangle = \langle \bar{\mathbf{G}}(\mathbf{r} - \mathbf{r}_0, \omega) \rangle \quad (20)$$

Thus, we can use a Fourier transform:

$$\bar{\Sigma}(\mathbf{k}, \omega) = \int d^3 \mathbf{r} \exp(-i \mathbf{k} \cdot \mathbf{r}) \bar{\Sigma}(\mathbf{r}, \omega) \quad (21)$$

$$\bar{\mathbf{G}}(\mathbf{k}, \omega) = \int d^3 \mathbf{r} \exp(-i \mathbf{k} \cdot \mathbf{r}) \bar{\mathbf{G}}(\mathbf{r}, \omega) \quad (22)$$

and equation (18) becomes:

$$\left[ \|\mathbf{k}\|^2 (\bar{\mathbf{I}} - \hat{\mathbf{k}}\hat{\mathbf{k}}) - \epsilon_1(\omega) K_{vac}^2 \bar{\mathbf{I}} - \bar{\Sigma}(\mathbf{k}, \omega) \right] \cdot \langle \bar{\mathbf{G}}(\mathbf{k}, \omega) \rangle = \bar{\mathbf{I}} \quad (23)$$

For a statistical isotropic medium, we have:

$$\bar{\Sigma}(\mathbf{k}, \omega) = \Sigma_\perp(\|\mathbf{k}\|, \omega) (\bar{\mathbf{I}} - \hat{\mathbf{k}}\hat{\mathbf{k}}) + \Sigma_\parallel(\|\mathbf{k}\|, \omega) \hat{\mathbf{k}}\hat{\mathbf{k}} \quad (24)$$

with  $\hat{\mathbf{k}} = \mathbf{k}/\|\mathbf{k}\|$  and then:

$$\langle \bar{\mathbf{G}}(\mathbf{k}, \omega) \rangle = \left[ \|\mathbf{k}\|^2 (\bar{\mathbf{I}} - \hat{\mathbf{k}}\hat{\mathbf{k}}) - \epsilon_1(\omega) K_{vac}^2 \bar{\mathbf{I}} - \bar{\Sigma}(\mathbf{k}, \omega) \right]^{-1} \quad (25)$$

$$= \frac{\bar{\mathbf{I}} - \hat{\mathbf{k}}\hat{\mathbf{k}}}{\|\mathbf{k}\|^2 - (\epsilon_1(\omega) K_{vac}^2 + \Sigma_\perp(\|\mathbf{k}\|, \omega))} \quad (26)$$

$$- \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{\epsilon_1(\omega) K_{vac}^2 + \Sigma_\parallel(\|\mathbf{k}\|, \omega)}$$

In the following, we introduce two effective permittivity functions  $\epsilon_e^\perp$  and  $\epsilon_e^\parallel$  defined by:

$$\epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2 = \epsilon_1(\omega) K_{vac}^2 + \Sigma_\perp(\|\mathbf{k}\|, \omega) \quad (27)$$

$$\epsilon_e^\parallel(\|\mathbf{k}\|, \omega) K_{vac}^2 = \epsilon_1(\omega) K_{vac}^2 + \Sigma_\parallel(\|\mathbf{k}\|, \omega) \quad (28)$$

and (26) is written:

$$\begin{aligned}
 & \langle \overline{\mathbf{G}}(\mathbf{k}, \omega) \rangle \\
 &= \left[ \overline{\mathbf{I}} - \frac{\mathbf{k}\mathbf{k}}{\epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2} \right] \frac{1}{\|\mathbf{k}\|^2 - \epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2} \\
 &+ \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{\epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2} - \frac{\hat{\mathbf{k}}\hat{\mathbf{k}}}{\epsilon_e^\parallel(\|\mathbf{k}\|, \omega) K_{vac}^2}.
 \end{aligned} \tag{29}$$

The Green function can be expressed in the space domain by the following formulation:

$$\begin{aligned}
 & \langle \overline{\mathbf{G}}(\mathbf{r}, \omega) \rangle \\
 &= \int \frac{d^3\mathbf{k}}{(2\pi)^3} \left[ \overline{\mathbf{I}} + \frac{\nabla\nabla}{\epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2} \right] \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\|\mathbf{k}\|^2 - \epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2} \\
 &+ \int \frac{d^3\mathbf{k}}{(2\pi)^3} \left[ \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\epsilon_e^\perp(\|\mathbf{k}\|, \omega) K_{vac}^2} - \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\epsilon_e^\parallel(\|\mathbf{k}\|, \omega) K_{vac}^2} \right] \frac{\mathbf{k}\mathbf{k}}{\|\mathbf{k}\|^2}.
 \end{aligned} \tag{30}$$

After integration on the solid angle in equation (30) given by the expressions (A1) and (A2) in the appendix, we obtain the expression:

$$\begin{aligned}
 & \langle \overline{\mathbf{G}}(\mathbf{r}, \omega) \rangle \\
 &= \frac{1}{i\|\mathbf{r}\|} \int_{-\infty}^{+\infty} \frac{dK}{(2\pi)^2} \left[ \overline{\mathbf{I}} + \frac{\nabla\nabla}{\epsilon_e^\perp(K, \omega) K_{vac}^2} \right] \frac{K e^{iK\|\mathbf{r}\|}}{K^2 - \epsilon_e^\perp(K, \omega) K_{vac}^2} \\
 &+ \frac{1}{i\|\mathbf{r}\|} \nabla\nabla \int_{-\infty}^{+\infty} \frac{dK}{(2\pi)^2} \left[ \frac{e^{iK\|\mathbf{r}\|}}{\epsilon_e^\perp(K, \omega) K_{vac}^2} - \frac{e^{iK\|\mathbf{r}\|}}{\epsilon_e^\parallel(K, \omega) K_{vac}^2} \right] \frac{1}{K}
 \end{aligned} \tag{31}$$

where we have supposed that  $\epsilon_e^\perp(\|\mathbf{k}\|, \omega) = \epsilon_e^\perp(-\|\mathbf{k}\|, \omega)$  and  $\epsilon_e^\parallel(\|\mathbf{k}\|, \omega) = \epsilon_e^\parallel(-\|\mathbf{k}\|, \omega)$ .

Using the residue theorem, we easily evaluate these integrals. However, we disregard the longitudinal excitation, which are solutions of  $\epsilon_e^\parallel(K, \omega) = 0$  and  $\epsilon_e^\perp(K, \omega) = 0$  since we only consider the propagation of the transversal electromagnetic field.

Furthermore, we see that the contribution of the pole  $K = 0$  in the second term of equation (31) is null. In fact, the dyadic  $\nabla\nabla$  operates on a constant as we have  $e^{iK\|\mathbf{r}\|} = 1$  for this pole. Hence, we obtain the following expression for the Green function:

$$\langle \overline{\mathbf{G}}(\mathbf{r}, \omega) \rangle = \sum_{i=1}^n \left[ \overline{\mathbf{I}} + \frac{\nabla\nabla}{K_{e_i}^2} \right] \frac{e^{iK_{e_i}\|\mathbf{r}\|}}{4\pi\|\mathbf{r}\|} \tag{32}$$

where  $K_{e_i}$  are the roots of  $K_{e_i}^2 = \epsilon_e^\perp(K_{e_i}, \omega) K_{vac}^2$ , the roots verify the equation  $\text{Im}(K_{e_i}) > 0$  imposed by the radiation condition at infinity. Sheng has called the roots  $K_{e_i}$  the quasi-modes of the random medium [9, 31]. If we only consider the root  $K_e = K_{e_j}$  which has the smallest imaginary part ( $\text{Im}(K_{e_j}) = \min_i[\text{Im}(K_{e_i})]$ ) and then the smallest exponential factor in equation (32), we define the effective permittivity by  $\epsilon_e(\omega) = \epsilon_e^\perp(K_e, \omega)$ . The average Green function is then equal to the Green function for an infinite homogenous medium with permittivity  $\epsilon_e(\omega)$ :

$$\langle \overline{\mathbf{G}}(\mathbf{r}, \omega) \rangle = \overline{\mathbf{G}}_e^\infty(\mathbf{r}, \omega) \tag{33}$$

where

$$\overline{\mathbf{G}}_e^\infty(\mathbf{r}, \omega) = \left[ \overline{\mathbf{I}} + \frac{\nabla \nabla}{K_e^2} \right] \frac{e^{i K_e \|\mathbf{r}\|}}{4\pi \|\mathbf{r}\|} \quad (34)$$

Thus, the effective medium approach is valid if we omit the longitudinal excitation in the medium and if the propagative mode with the smallest imaginary part is the main contribution in the development (32).

### 3.The coherent-potential and quasi-crystalline approximations

Previously, we have shown how the mass operator is related to the effective permittivity. To calculate the mass operator, we can use equations (13) and (17). However, we can improve this system of equations, thus we rewrite the Green function development (5) by replacing the Green function  $\overline{\mathbf{G}}_1^\infty$  with  $\overline{\mathbf{G}}_e^\infty$  :

$$\overline{\mathbf{G}} = \overline{\mathbf{G}}_e^\infty + \overline{\mathbf{G}}_e^\infty \cdot \overline{\mathbf{V}}_e \cdot \overline{\mathbf{G}} \quad (35)$$

where we have introduced a new potential  $\overline{\mathbf{V}}_e$  :

$$\overline{\mathbf{V}}_e = \sum_{i=1}^N \tilde{\mathbf{v}}_{e,r_i} \quad (36)$$

$$\tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \mathbf{r}_0, \omega) = (2\pi)^2 \delta(\mathbf{r} - \mathbf{r}_0) \tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega) \quad (37)$$

$$\begin{aligned} \tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega) &= [K_s^2 - K_e^2] \Theta_s(\mathbf{r} - \mathbf{r}_i) \overline{\mathbf{I}} \\ &+ [K_1^2 - K_e^2] \overline{\mathbf{I}}. \end{aligned} \quad (38)$$

As in the previous section, we introduce a T matrix:

$$\overline{\mathbf{G}} = \overline{\mathbf{G}}_e^\infty + \overline{\mathbf{G}}_e^\infty \cdot \overline{\mathbf{T}}_e \cdot \overline{\mathbf{G}}_e^\infty \quad (39)$$

which can be decomposed as following:

$$\overline{\mathbf{T}}_e = \sum_{i=1}^N \tilde{\mathbf{t}}_{e,r_i} + \sum_{i=1}^N \sum_{j=1, j \neq i}^N \tilde{\mathbf{t}}_{e,r_j} \cdot \overline{\mathbf{G}}_e^\infty \cdot \tilde{\mathbf{t}}_{e,r_i} + \dots \quad (40)$$

where we have defined a renormalized T matrix for the particles:

$$\tilde{\mathbf{t}}_{e,r_i} = \tilde{\mathbf{v}}_{e,r_i} + \tilde{\mathbf{v}}_{e,r_i} \cdot \overline{\mathbf{G}}_e^\infty \cdot \tilde{\mathbf{t}}_{e,r_i} \quad (41)$$

We impose the following condition on the average field:

$$\langle \overline{\mathbf{G}}(\mathbf{r}, \omega) \rangle = \overline{\mathbf{G}}_e(\mathbf{r}, \omega) \quad (42)$$

or,

$$\langle \overline{\mathbf{T}}_e \rangle = \overline{\mathbf{0}} \quad (43)$$

considering equation (39). The condition (42) is the Coherent-Potential Approximation (CPA) [9, 11, 14, 25]. The expressions (43) and (40) form a closed system of equations for the unknown permittivity  $\varepsilon_e(\omega)$ . At the first order in density of particles, this system of equations gives:

$$\sum_{i=1}^N \langle \tilde{\mathbf{t}}_{e,r_i} \rangle = \mathbf{0} \quad (44)$$

In Fourier-space, the T matrix for one scatterer verifies the property:

$$\tilde{\mathbf{t}}_{e,r_i}(\mathbf{k}|\mathbf{k}_0) = e^{-i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_i} \tilde{\mathbf{t}}_{e,o}(\mathbf{k}|\mathbf{k}_0) \quad (45)$$

where  $\tilde{\mathbf{t}}_{e,o}$  is the T matrix for a particle located at the origin of coordinates. The average of the exponential term, introduced in equation (44) with the equation (45), gives for a statistical homogeneous medium:

$$\langle \sum_{i=1}^N e^{-i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_i} \rangle = N \int d^3\mathbf{r} \frac{1}{\mathcal{V}} e^{-i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}} \quad (46)$$

$$= n (2\pi)^2 \delta(\mathbf{k} - \mathbf{k}_0) \quad (47)$$

where we have defined the density of scatterers  $n = N / \mathcal{V}$  with  $\mathcal{V}$  the volume of the random medium. The condition (44) becomes:

$$\tilde{\mathbf{t}}_{e,o}(\mathbf{k}_0|\mathbf{k}_0) = \mathbf{0} \quad (48)$$

This CPA condition has been used in several works [9, 31, 43]. It is worth mentioning that operator  $\tilde{\mathbf{t}}_{e,r_i}(\mathbf{k}|\mathbf{k}_0)$  is not the T matrix describing the scattering by a particle of permittivity  $\varepsilon_s(\omega)$  surrounded by a medium of permittivity  $\varepsilon_e(\omega)$ . To describe this electromagnetic interaction, the operator (38) should have the following form:

$$\tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega) = [K_s^2(\omega) - K_e^2(\omega)] \Theta_s(\mathbf{r} - \mathbf{r}_i) \bar{\mathbf{I}} \quad (49)$$

However, we see that  $\tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega)$  is different from the operator in equation (38), and especially we have  $\tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega) = [K_1^2 - K_e^2] \bar{\mathbf{I}}$  for  $\|\mathbf{r} - \mathbf{r}_i\| > r_s$ , which does not correspond to definition (49), where  $\tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega) = \mathbf{0}$  when  $\mathbf{r}$  is outside the particle.

Thus,  $\tilde{\mathbf{t}}_{e,r_i}$  is a non-local operator and cannot be obtained from the classical Mie theory [3, 25]. To overcome this difficulty, in some works [9, 43]  $\tilde{\mathbf{t}}_{e,r_i}$  is replaced by the scattering operator of a "structural unit". Nevertheless, this approach does not seem to have any theoretical justification. Hence, we prefer to use the more rigorous approach introduced in the scattering theory by disordered liquid metal [28] and adapted in the electromagnetic scattering case by Tsang *et al.* [11, 14].

In this approach, the non-local term  $[K_1^2 - K_e^2] \bar{\mathbf{I}}$  is correctly taken into account by averaging equations (39) where we use the correct potential  $\tilde{\mathbf{v}}_{e,r_i}(\mathbf{r}, \omega)$ , defined by (38). A system of hierarchical equations is obtained where correlation functions between two or more particles are successively introduced.

The chain of equations is closed by using the Quasi-Crystalline Approximation (QCA), which disregards the fluctuation of the effective field, acting on a particle located at  $\mathbf{r}_j$ , due to a deviation of a particle located at  $\mathbf{r}_i$  from its average position [26].

This approximation describes the correlation between the particles, only with a two-point correlation function  $g(\mathbf{r}_i, \mathbf{r}_j) = g(|\mathbf{r}_i - \mathbf{r}_j|)$ . Under the QC-CPA scheme, we obtain the following expression for the mass operator [11, 14, 25, 28]:

$$\bar{\Sigma}(\mathbf{k}_0, \omega) = n \bar{\mathbf{C}}_{e,o}(\mathbf{k}_0 | \mathbf{k}_0) \quad (50)$$

$$\begin{aligned} \bar{\mathbf{C}}_{e,o}(\mathbf{k} | \mathbf{k}_0) &= \bar{\mathbf{t}}_{e,o}(\mathbf{k} | \mathbf{k}_0) \\ &+ n \int \frac{d^3 \mathbf{k}_1}{(2\pi)^3} h(\mathbf{k} - \mathbf{k}_1) \bar{\mathbf{t}}_{e,o}(\mathbf{k} | \mathbf{k}_1) \cdot \bar{\mathbf{G}}_e^\infty(\mathbf{k}_1) \cdot \bar{\mathbf{C}}_{e,o}(\mathbf{k}_1 | \mathbf{k}_0) \end{aligned} \quad (51)$$

where

$$\bar{\mathbf{t}}_{e,o} = \bar{\mathbf{v}}_{e,o} + \bar{\mathbf{v}}_{e,o} \cdot \bar{\mathbf{G}}_e^\infty \cdot \bar{\mathbf{t}}_{e,o} \quad (52)$$

$$\bar{\mathbf{v}}_{e,o}(\mathbf{r}, \mathbf{r}_0) = (2\pi) \delta(\mathbf{r} - \mathbf{r}_0) \bar{\mathbf{v}}_{e,o}(\mathbf{r}) \quad (53)$$

$$\bar{\mathbf{v}}_{e,o}(\mathbf{r}) = [K_s^2 - K_1^2] \Theta_s(\mathbf{r}) \bar{\mathbf{I}} \quad (54)$$

and

$$h(\mathbf{r}) = g(\mathbf{r}) - 1 \quad (55)$$

$$h(\mathbf{k} - \mathbf{k}_1) = \int d^3 \mathbf{r} \exp(-i(\mathbf{k} - \mathbf{k}_1) \cdot \mathbf{r}) h(\mathbf{r}) \quad (56)$$

$$\bar{\mathbf{G}}_e^\infty(\mathbf{k}) = \int d^3 \mathbf{r} \exp(-i\mathbf{k} \cdot \mathbf{r}) \bar{\mathbf{G}}_e^\infty(\mathbf{r}) \quad (57)$$

If we rewrite the potential (54) under the following form:

$$\bar{\mathbf{v}}_{e,o}(\mathbf{r}) = [\tilde{K}_s^2 - K_e^2] \Theta_s(\mathbf{r}) \bar{\mathbf{I}} \quad (58)$$

where we have defined a new wave number  $\tilde{K}_s^2 = K_s^2 - K_1^2 + K_e^2$ , we see that the operator  $\bar{\mathbf{t}}_{e,o}$  is the T matrix for a scatterer of permittivity  $\tilde{\epsilon}_s = \epsilon_s - \epsilon_1 + \epsilon_e$  in a medium of permittivity  $\epsilon_e$ . As described in the previous section, the effective propagation constant  $K_e$  is the root, which has the smallest imaginary part, of the equation:

$$K_e^2 = K_1^2 + \Sigma_\perp(K_e, \omega) \quad (59)$$

where the mass operator is decomposed under the form (24). Once the effective wave number  $K_e$  is obtained, the effective permittivity is given by:

$$\epsilon_e(\omega) = K_e^2 / K_{vac}^2 \quad (60)$$

#### 4. Some further approximations

Solving numerically the previous system of equations (50-60) is a difficult task. However, for the low frequency limit of this system of equation, an analytical solution can be obtained and has shown to be in good agreement with the experimental results [11, 14]. We have also to mention that the numerical resolution of the quasicrystalline approximation without the coherent potential approximation has been

developed [13, 38]. To reduce the numerical difficulties for this system of equations (50-60), we add two new approximations to the QC-CPA scheme:

→ *A far-field approximation*, for an incident plane wave:

$$\mathbf{E}^i(\mathbf{r}) = \mathbf{E}^i(\mathbf{k}_0) e^{i\mathbf{k}_0 \cdot \mathbf{r}} \quad (61)$$

transverse to the propagation direction  $\hat{\mathbf{k}}_0$ :

$$\mathbf{E}^i(\mathbf{k}_0) \cdot \hat{\mathbf{k}}_0 = 0 \quad (62)$$

where  $\mathbf{k}_0 = K_e \hat{\mathbf{k}}_0$  and  $\hat{\mathbf{k}}_0 \cdot \hat{\mathbf{k}}_0 = 1$ , the scattered far-field, by a particle within a medium of permittivity  $\varepsilon_e(\omega)$ , is described by an operator  $\bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0)$ . We obtain the following equation:

$$\mathbf{E}^s(\mathbf{r}) = \frac{e^{iK_e \|\mathbf{r}\|}}{\|\mathbf{r}\|} \bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) \cdot \mathbf{E}^i(\hat{\mathbf{k}}_0) \quad (63)$$

which verifies transversality conditions:

$$\bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) \cdot \hat{\mathbf{k}}_0 = 0 \quad (64)$$

$$\hat{\mathbf{k}} \cdot \bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) = 0 \quad (65)$$

Moreover, the scattered field in the general case is expressed with the operator  $\bar{\mathbf{t}}_{e,o}$  by:

$$\mathbf{E}^s(\mathbf{r}) = \int d^3\mathbf{r}_1 d^3\mathbf{r}_2 \bar{\mathbf{G}}_e^\infty(\mathbf{r}, \mathbf{r}_1) \cdot \bar{\mathbf{t}}_{e,o}(\mathbf{r}_1|\mathbf{r}_2) \cdot \mathbf{E}^i(\mathbf{r}_2) \quad (66)$$

Using the properties of the Green function in equation (66), we obtained the scattered far-field in function of the operator  $\bar{\mathbf{t}}_{e,o}(\mathbf{k}|\mathbf{k}_0)$ , and comparing the result with equation (63), we can write:

$$4\pi \bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) = (\bar{\mathbf{I}} - \hat{\mathbf{k}}\hat{\mathbf{k}}) \cdot \bar{\mathbf{t}}_{e,o}(K_e \hat{\mathbf{k}}|K_e \hat{\mathbf{k}}_0) \cdot (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0) \quad (67)$$

Our far-field approximation disregards the longitudinal component and the off-shell contribution in the operator  $\bar{\mathbf{t}}_{e,o}$ , and we write:

$$\bar{\mathbf{t}}(K_e \hat{\mathbf{k}}|K_e \hat{\mathbf{k}}_0) \simeq 4\pi \bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) \quad (68)$$

$$= 4\pi (\bar{\mathbf{I}} - \hat{\mathbf{k}}\hat{\mathbf{k}}) \cdot \bar{\mathbf{f}}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) \cdot (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0) \quad (69)$$

where the last equality comes from the properties (64-65).

→ *A forward scattering approximation*: for scatterers whose dimensions are large compared to a wavelength, the magnitude of the scattered field is predominantly located in the forward direction

(i.e.  $|f(\hat{\mathbf{k}}_0|\hat{\mathbf{k}}_0)| \gg |f(-\hat{\mathbf{k}}_0|\hat{\mathbf{k}}_0)|$ ). With our forward approximation we only conserve the contribution of the amplitude of diffusion  $f(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0)$  in the direction of the incident wave  $\hat{\mathbf{k}}_0$ . Using the hypothesis (68), we can write:

$$\bar{t}_{e,o}(K_e \hat{\mathbf{k}}|K_e \hat{\mathbf{k}}_0) = 4\pi \bar{f}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) \quad (70)$$

$$\simeq 4\pi \bar{f}(\hat{\mathbf{k}}_0|\hat{\mathbf{k}}_0) \quad (71)$$

$$= 4\pi (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0) f(K_e, \omega) \quad (72)$$

where:

$$f(K_e, \omega) = \frac{i}{K_e} S_1(0) = \frac{i}{K_e} S_2(0) \quad (73)$$

with  $S_1(0) = S_2(0)$  given by the Mie theory [3, 4, 44]. It is worth mentioning that the approximation (71) is also valid for small scatterers (Rayleigh scatterers).

In this case, the scattering amplitude  $f(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0)$  does not depend on the directions of the incident  $\hat{\mathbf{k}}_0$  and scattered  $\hat{\mathbf{k}}$  wave vectors, since we have:

$$\bar{t}_{e,o}(\mathbf{k}|\mathbf{k}_0) = t_{e,o}(\omega) \bar{\mathbf{I}} \quad (74)$$

From equation (67), we show that:

$$\bar{f}(\hat{\mathbf{k}}|\hat{\mathbf{k}}_0) = \bar{f}(\hat{\mathbf{k}}_0|\hat{\mathbf{k}}_0) \quad (75)$$

and we also obtain the coefficient  $f(K_e, \omega)$ :

$$4\pi f(K_e, \omega) = t_{e,o}(\omega) \quad (76)$$

Furthermore, we see from equation (51), that for the order zero in density, we have:

$$\bar{\mathbf{C}}_{e,o}(\mathbf{k}|\mathbf{k}_0) = \bar{t}_{e,o}(\mathbf{k}|\mathbf{k}_0) \quad (77)$$

and the forward approximation (71) can be applied to the operator  $\bar{\mathbf{C}}_{e,o}(\mathbf{k}|\mathbf{k}_0)$  in the limit of low density. We will suppose that the forward approximation is valid for the operator whatever the order in density and we write:

$$\bar{\mathbf{C}}_{e,o}(\mathbf{k}|\mathbf{k}_0) \simeq \bar{\mathbf{C}}_{e,o}(\mathbf{k}_0|\mathbf{k}_0) \quad (78)$$

$$\simeq (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0) C_{e,o}^\perp(\|\mathbf{k}_0\|, \omega) \quad (79)$$

With this hypothesis, only the path of type 1 in figure 1 is considered. This approximation also implies that the operator  $\bar{\mathbf{C}}_{e,o}(\mathbf{k}|\mathbf{k}_0)$  is transverse to the propagation direction  $\hat{\mathbf{k}}_0$ .

From the previous hypothesis and the QC-CPA equations (51), we obtain an expression of  $\bar{\mathbf{C}}_{e,o}(\mathbf{k}|\mathbf{k}_0)$

$$\begin{aligned} \bar{\mathbf{C}}_{e,o}(\mathbf{k}_0|\mathbf{k}_0) &= 4\pi f(K_e, \omega) \bar{\mathbf{I}}_\perp(\hat{\mathbf{k}}_0) \\ &+ 4\pi n f(K_e, \omega) \bar{\mathbf{m}}(\mathbf{k}_0) \cdot \bar{\mathbf{C}}_{e,o}(\mathbf{k}_0|\mathbf{k}_0) \end{aligned} \quad (80)$$

where we have introduced the notation:

$$\bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) = (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0) \quad (81)$$

$$\bar{\mathbf{m}}(\mathbf{k}_0) = \int \frac{d^3 \mathbf{k}_1}{(2\pi)^3} h(\mathbf{k}_0 - \mathbf{k}_1) \cdot \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) \cdot \bar{\mathbf{G}}_e^{\infty}(\mathbf{k}_1) \cdot \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) \quad (82)$$

Then, we have:

$$\bar{\mathbf{C}}_{e,o}(\mathbf{k}_0|\mathbf{k}_0) = \left[ \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) - 4\pi n f(K_e, \omega) \bar{\mathbf{m}}(\mathbf{k}_0) \right]^{-1} \cdot \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) 4\pi f(K_e, \omega). \quad (83)$$

Using the classical properties of the Fourier transform, we obtain:

$$\bar{\mathbf{m}}(\mathbf{k}_0) = \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) \cdot \int d^3 \mathbf{r} e^{-i\mathbf{k}_0 \cdot \mathbf{r}} h(\mathbf{r}) \bar{\mathbf{G}}_e^{\infty}(\mathbf{r}) \cdot \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) \quad (84)$$

where we have used the translation invariance of the Green function:  $\bar{\mathbf{G}}_e^{\infty}(\mathbf{r} - \mathbf{r}_0) = \bar{\mathbf{G}}_e^{\infty}(\mathbf{r}, \mathbf{r}_0)$ . The Dyadic Green function has a singularity which can be extracted in introducing the principal value of the Green function [13, 14, 41, 45]:

$$\bar{\mathbf{G}}_e^{\infty}(\mathbf{r}) = P.V. \bar{\mathbf{G}}_e(\mathbf{r}) - \frac{1}{3K_e^2} \delta(\mathbf{r}) \bar{\mathbf{I}} \quad (85)$$

where the principal value is defined by:

$$\begin{aligned} & P.V. \int d^3 \mathbf{r}_0 \bar{\mathbf{G}}_e^{\infty}(\mathbf{r} - \mathbf{r}_0) \cdot \bar{\phi}(\mathbf{r}_0) \\ &= \lim_{a \rightarrow 0} \int_{S_a(\mathbf{r})} d^3 \mathbf{r}_0 \bar{\mathbf{G}}_e^{\infty}(\mathbf{r} - \mathbf{r}_0) \cdot \bar{\phi}(\mathbf{r}_0), \end{aligned} \quad (86)$$

with  $\bar{\phi}(\mathbf{r}_0)$  a test function and  $S_a(\mathbf{r})$  a spherical volume of radius  $a$  centered at  $\mathbf{r}$ . This principal value can be easily calculated, and we obtain [11, 14]:

$$\begin{aligned} P.V. G_e(\mathbf{r}) = \frac{e^{iK_e \|\mathbf{r}\|}}{4\pi \|\mathbf{r}\|} & \left[ \left( 1 - \frac{1}{iK_e \|\mathbf{r}\|} - \frac{1}{K_e^2 \|\mathbf{r}\|^2} \right) \bar{\mathbf{I}} \right. \\ & \left. - \left( 1 - \frac{3}{iK_e \|\mathbf{r}\|} - \frac{3}{K_e^2 \|\mathbf{r}\|^2} \right) \hat{\mathbf{r}} \hat{\mathbf{r}} \right]. \end{aligned} \quad (87)$$

Using polar coordinate in the integral (84):

$$\int d^3 \mathbf{r} = \int_0^{+\infty} r^2 dr \int_{4\pi} d^2 \hat{\mathbf{r}} \quad (88)$$

and the integral on solid angles given in Appendix, we obtain the following result:

$$\bar{\mathbf{m}}(\mathbf{k}_0) = \left[ -\frac{h(0)}{3K_e^2} + m(K_e) \right] \bar{\mathbf{I}}_{\perp}(\hat{\mathbf{k}}_0) \quad (89)$$

with

$$m(K_e) = \int_0^{+\infty} r dr p(K_e r) [g(r) - 1] e^{iK_e r} \quad (90)$$

$$p(x) = \frac{\sin x}{x} - \left( \frac{\sin x}{x^3} - \frac{\cos x}{x^2} \right), \tag{91}$$

$$- \left( \frac{1}{ix} + \frac{1}{x^2} \right) \left( \frac{\sin x}{x} - 3 \left[ \frac{\sin x}{x^3} - \frac{\cos x}{x^2} \right] \right)$$

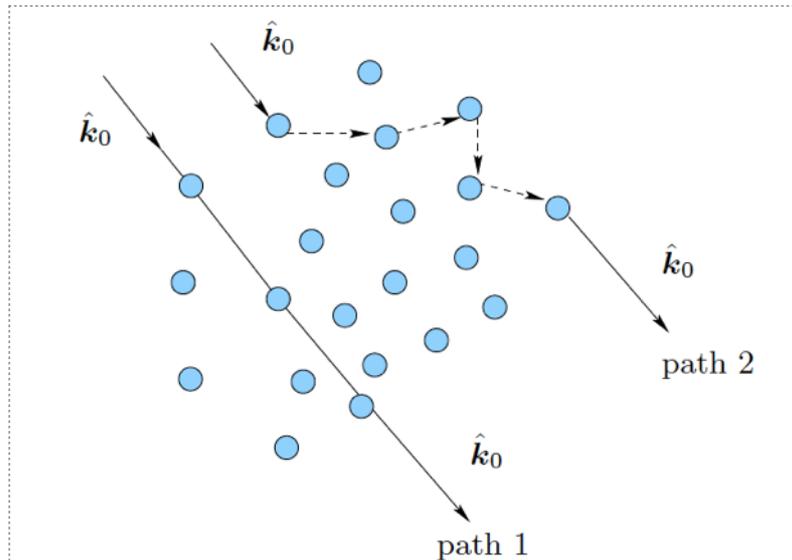
where we have assumed  $\|\mathbf{k}_0\| = K_e$  because the mass operator (50) is evaluated with this value in the equation (59) to obtain the effective permittivity. As the particles cannot overlap, we have  $g(0) = 0$  and then  $h(0) = -1$ . With equations (59, 24, 50, 83, 89), we derive an expression for the effective wave number  $K_e$  :

$$K_e^2 = K_1^2 + \frac{4\pi n f(K_e, \omega)}{1 - 4\pi n f(K_e, \omega) \left( \frac{1}{3K_e^2} + m(K_e) \right)} \tag{92}$$

where the scalar  $m(K_e)$  is defined by equations (90,91).

The scalar  $f(K_e, \omega)$  is the forward scattering amplitude:  $\bar{f}(\hat{\mathbf{k}}_0|\hat{\mathbf{k}}_0) = (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0\hat{\mathbf{k}}_0)f(K_e, \omega)$  for a particle of permittivity  $\tilde{\epsilon}_s = \epsilon_s - \epsilon_1 + \epsilon_e$  within a medium of permittivity  $\epsilon_e$ . The relationship between the effective wave number  $K_e$  and the effective permittivity  $\epsilon_e$  is given by:

$$\epsilon_e(\omega) = K_e^2 / K_{vac}^2 \tag{93}$$



**Figure 1.** Two different paths which contribute to the mass operator  $\bar{\Sigma}(\|\mathbf{k}_0\|, \omega) = n \bar{C}_{e,o}(\mathbf{k}_0|\mathbf{k}_0)$  in the (QC-CPA) approach. Only the path of kind 1 is taken into account in our forward scattering approximation (79).

### 5.High frequency and low frequency limits

In section 3, we have derived a general system of equation to obtain the effective permittivity. With some approximations we obtain a system which can be solved more easily. We have derived [46] a

general formulation for an effective permittivity for a layer of an inhomogeneous random medium with different types of particles and bounded with randomly rough surfaces. We obtained an analytical expression for Rayleigh-type scatterers for the low frequency limit. This formula is important because it takes into account a broader model of nanoscale particles if we introduce the regularization parameter [46] of the principal value of the Green function. This parameter is linked to resonances of the considered scatterer. We are now going to show that the relation (92) that we have obtained contains also the Keller formula [40] in the high frequency limit. This formula has been shown to be in good agreement with experimental results for particles larger than a wavelength. The Keller formula can be obtained in considering the QC-CPA approach in the scalar case [11, 14]. The equations are formerly identical to equation (50-57) where the dyadic Green function

$$\overline{\mathbf{G}}_e^\infty(\mathbf{r}, \mathbf{r}_0, \omega) = \left[ \overline{\mathbf{I}} + \frac{\nabla \nabla}{K_e^2} \right] \frac{e^{i K_e \|\mathbf{r} - \mathbf{r}_0\|}}{4\pi \|\mathbf{r}\|} \quad (94)$$

has to be replaced by the scalar Green function:

$$G_e^\infty(\mathbf{r}, \mathbf{r}_0, \omega) = \frac{e^{i K_e \|\mathbf{r} - \mathbf{r}_0\|}}{4\pi \|\mathbf{r}\|} \quad (95)$$

Using equation (A1), the first iteration of the scalar version of equation (51) gives:

$$\begin{aligned} K_e^2 &= K_1^2 + 4\pi n f(K_e, \omega) \\ &+ (4\pi)^2 n^2 f^2(K_e, \omega) \int_0^{+\infty} dr \frac{\sin K_e r}{K_e} [g(r) - 1] e^{i K_e r} + \dots \end{aligned} \quad (96)$$

As was shown by Waterman et al [27], this development is valid if the following condition is verified:

$$(4\pi)^2 n |f(K_e, \omega)|^2 / K_e \ll 1 \quad (97)$$

In the geometric limit, the scattering cross section  $\sigma_s$  for a single particle is in good approximation given by  $\sigma_s \simeq 2 \pi r_s^2$ .

As the cross-section is connected to the scattering amplitude by the relation  $\sigma_s = \frac{8\pi}{3} |f(K_e, \omega)|^2$  and as the maximum density is  $n = 1/v_s$ , we see that for particles larger than a wavelength the condition (97) is satisfied:

$$(4\pi)^2 n |f(K_e, \omega)|^2 / K_e \simeq 1 / K_e r_s \ll 1 \quad (98)$$

The equation (96) has been derived by Keller [40] and this expression is in good agreement with experiments for large particles compared to the wavelength. If we now use a Taylor development in equation (92), we obtain:

$$\begin{aligned} K_e^2 &= K_1^2 + (4\pi)^2 n f(K_e, \omega) \\ &+ (4\pi)^2 n^2 f^2(K_e, \omega) \left[ \frac{1}{3 K_e^2} + m(K_e) \right] + \dots \end{aligned} \quad (99)$$

This development is valid if the condition (97) is satisfied. In the geometrical limits, we can approximate the function  $p(K_e r)$  in definition (90). For  $K_e r \gg 1$  we have from equation (91):

$$p(x) \simeq \frac{\sin x}{x}, \quad x \gg 1 \quad (100)$$

The relation (99) becomes:

$$K_e^2 = K_1^2 + 4\pi n f(K_e, \omega) + (4\pi)^2 n^2 f^2(K_e, \omega) \left[ \frac{1}{3K_e^2} + \int_0^{+\infty} dr \frac{\sin K_e r}{K_e} [g(r) - 1] e^{iK_e r} \right] \quad (101)$$

We see that equation (101) differs from the equation (96), only by the factor  $1/3K_e^2$ , due to the singularity of the vectorial Green function and this factor cannot be derived from the scalar theory developed by Keller. We also notice that to solve numerically our new equation (92), we can utilize the same procedure that is used for the original Keller formula (96) with the Muller theory. Therefore we have derived a numerical tractable approximation to the (QC-CPA) scheme.

## 6. Discussion

The intent of this paper has been to establish a new formula for the effective dielectric constant which characterizes the coherent part of an electromagnetic wave propagating in a random medium containing disordered particles. The particles can be metallic, dielectric, spherical or non-spherical. We have discussed the relation between the Dyson equation and the effective permittivity of the medium. With this formulation, we have introduced the mass operator. The mass operator or self-energy operator includes geometric effects, caused by resonant behavior due to the shape and size of particles, cluster effects because of correlations between particles. The problem is solved if we obtain an expression of the mass operator. For that, the relation between the mass operator and the scattering operator is expressed. We give a formal solution for the scattering operator by introducing the T-operator formalism. We show that the T-operator, or the scattering operator satisfies a Lippman-Schwinger equation. This operator includes all the multiple scattering. Then we have introduced the Quasi-Crystalline Coherent Potential Approximation (QC-CPA), which takes into account the correlation between the particles with a pair-distribution function. This function describes the correlation between two distinct particles. To simplify the numerical calculation of the effective permittivity under the (QC-CPA) approach, we have added far-field and forward scattering approximations to (QC-CPA) scheme. In the low frequency limit, our equation is identical with the usual result obtained under the (QC-CPA) scheme and we have generalized the formulation for Rayleigh scatterers by introducing near plasmon resonances. For particles whose dimensions are of the order of wavelength or in the high frequency limit the expression includes the generalization, in the vectorial case, of the result obtained by Keller. We obtained an important tensorial generalization of the notion of effective permittivity. This technique of Dyson equation in wave multiple scattering by spatially disordered discrete medium leads to a dielectric permittivity tensor. This expression of the permittivity developed in this paper has been extended to random media with different types of particles and bounded by rough surfaces [46]. This extended formulation of the effective permittivity is then used to calculate the coherent fields and incoherent intensities scattered from a three-dimensional disordered medium with discrete scatterers and randomly rough interfaces.

The authors [47] show the physical nature of diamagnetic property in the effective magnetic permeability at coherent electromagnetic wave multiple scattering by statistical ensemble of independent perfectly reflected non-magnetic small spherical particles. This observation is made with the aid of the technique of Dyson equation developed in this paper for a random medium with disordered particles.

## Appendix

$$\int_{4\pi} d^2 \hat{\mathbf{r}} e^{-i \mathbf{k}_0 \cdot \hat{\mathbf{r}} \|\mathbf{r}\|} = 4\pi \frac{\sin \|\mathbf{k}_0\| \|\mathbf{r}\|}{\|\mathbf{k}_0\| \|\mathbf{r}\|} \quad (A1)$$

$$\begin{aligned}
 \int_{4\pi} d^2\hat{r} e^{-i\mathbf{k}_0 \cdot \hat{r} \|\mathbf{r}\|} \hat{r} \hat{r} = 4\pi & \left[ \frac{\sin \|\mathbf{k}_0\| \|\mathbf{r}\|}{\|\mathbf{k}_0\|^3 \|\mathbf{r}\|^3} \right. \\
 & \left. - \frac{\cos \|\mathbf{k}_0\| \|\mathbf{r}\|}{\|\mathbf{k}_0\|^2 \|\mathbf{r}\|^2} \right] (\bar{\mathbf{I}} - \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0) + 4\pi \left[ \frac{\sin \|\mathbf{k}_0\| \|\mathbf{r}\|}{\|\mathbf{k}_0\| \|\mathbf{r}\|} \right. \\
 & \left. + 2 \frac{\cos \|\mathbf{k}_0\| \|\mathbf{r}\|}{\|\mathbf{k}_0\|^2 \|\mathbf{r}\|^2} - 2 \frac{\sin \|\mathbf{k}_0\| \|\mathbf{r}\|}{\|\mathbf{k}_0\|^3 \|\mathbf{r}\|^3} \right] \hat{\mathbf{k}}_0 \hat{\mathbf{k}}_0, \tag{A2}
 \end{aligned}$$

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