COHERENT LIGHT SCATTERING

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Coherent light scattering of heterogeneous randomly rough films and effective medium in the theory of electromagnetic wave multiple scattering

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Abstract. **We have developed a general formalism based on Green's functions to calculate the coherent electromagnetic field scattered by a random medium with rough boundaries. The approximate expression derived makes it possible to determine the effective permittivity, which is generalised for a layer of an inhomogeneous random medium with different types of particles and bounded with randomly rough interfaces. This effective permittivity describes the coherent propagation of an electromagnetic wave in a random medium with randomly rough boundaries. We have obtained an expression, which contains the Maxwell–Garnett formula at the low-frequency limit, and the Keller formula; the latter has been proved to be in good agreement with experiments for particles whose dimensions are larger than a wavelength.**

Keywords: *coherent light scattering, inhomogeneously random medium, randomly rough surfaces, effective permittivity, Green's function.*

1. Introduction

The propagation of electromagnetic waves in random media has been studied extensively in the past decades [1–16]. In most of works, the basic idea is to calculate the first and second statistical moments of the electromagnetic field in order to understand how the waves interact with an inhomogeneous random medium [8, 11–13, 16]. In this paper, we are interested in the first statistical moment, which represents an average electric field. Under some assumptions, we can show that this field propagates as if the medium were homogeneous but with a renormalised permittivity, the so-called effective permittivity. The calculation of this parameter has a long history which dates back to the work of Clausius–Mossotti and Maxwell–Garnett. Since then, most of the studies are concerned with a quasi-static limit where the retardation effect is neglected. In order to take into account scattering effects, the quantum multiple scattering theory has been transposed to the electromagnetic case [6, 8, 11–13, 16]. However, as a rigorous analytical answer is unreachable, several approximation schemes have been developed [6, 8, 12, 13, 16]. One of the most advanced schemes is the quasi-crystalline coherent potential approximation (QC-CPA), which takes into account correlations between particles [13, 16].

In this paper we consider the coherent component of the electromagnetic wave field scattered by random media bounded with randomly rough surfaces. To this end, rigorous numerical methods have been developed, which are computationally cumbersome or limited to 2D geometries. Most often, the radiative transfer theory is used for the volumetric scattering. This method is well suited to compute the scattered intensity but is based on phenomenological considerations. Thus, analytical theory has been developed in order to describe the coupling between a random medium and rough boundaries. Furutsu [17, 18] formulated the rough surface scattering problem on the basis of Dyson and Bethe–Salpeter equations, in which the random medium and rough boundaries are treated on the same footing. Unfortunately, this approach is formal, and the relationship between the radiative transfer theory and the classical rough surface scattering theories is not straightforward. Mudaliar [19–21] used integral equations where the rough boundaries were considered under a perturbative development. He showed that the scattering intensity is described by a generalised transport equation. His approach is more numerically tractable than Furutsu's, but the expressions derived are determined by the choice of the perturbative development to describe the scattering by the rough surfaces. In this paper, we show that we can obtain the general expression, whatever the choice of the scattering theory used at the boundaries, by introducing the scattering operators of randomly rough surfaces (see [22] for the expressions of scattering operators for the small-amplitude perturbation theory). Furthermore, in separating the surface and the volume scattering contributions with the help of Green's functions, we can use analytical theories of waves scattered by an infinite random medium. We consider the coherent field in the random medium and study the effect of the rough boundaries of the inhomogeneous layer on the expression of the effective permittivity. The subject of our interest concerns a random medium consisting of statistical ensembles of different scattering species and artificial material structures developed on the basis of dielectric or metallic resonant particles or nanoparticles. The aim of this paper is to derive new propagation equations for a light wave and new formulas for the effective permittivity which characterises the coherent part of an electromagnetic wave propagating inside a random medium with randomly rough interfaces. The starting point of our theory is the multiple scattering theory. We present a formal solution for the scattering operator by introducing the T-operator formalism. We show that the T-operator satisfies the Lippman–Schwinger equation. Then we introduce the QC-CPA, which takes into account the correlation between the particles with the help of the pair distribution function. It is important to define a new accurate formulation of the effective permittivity and to introduce the roughness of a slab into the expression of the scattering field for the design of heterogeneous metamaterials [23]

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and the mathematical analysis of wave scattering in these media. This can provide a physical insight in the fields of wave physics of heterogeneous metamaterials and non-local dispersion. The current interest in these topics is linked to the growing attention due to ability of such structures to control the propagation of electromagnetic waves at scales larger and lesser than the wavelength and due to the elaboration of the standardised mathematical basis for optimisation of parameters of such structures. This paper presents some new perspectives: new formula of the effective permittivity, which contains the Maxwell–Garnett formula at the low-frequency limit, and the Keller formula, the contribution of the rough boundaries of the slab, new expressions of the coherent scattering field taking into account scattering from randomly rough boundaries and a random medium with different types of particles. The operator, which describes the scattering by the randomly rough boundaries of the slab, can be approximated by using the usual scattering theories by rough surface like the small-perturbation, the Kirchhoff or other more sophisticated theories (small-slope approximation, small-perturbation method at higher orders).

The paper is organised as follows. In Section 2, we introduce the multiple scattering formalism for a random layer with randomly rough surfaces and consider the Lippman–Schwinger equation. In Section 3, we derive a system of equations verified by the effective permittivity under the QC-CPA approach. Then, in Section 4, we introduce the formulation of the effective permittivity for Rayleigh scatterers, and in Section 5, we present some numerical examples for dielectric or metallic nanoparticles embedded in a dielectric medium.

2. Lippman–Schwinger equations and scattered field

In the following we consider harmonic waves with the frequency ω. The structure (Fig. 1) in question consists of a first semi-infinite medium (medium 0) with a permittivity ε_0, a random layer (medium 1) with a permittivity $\varepsilon_1(\omega)$ bounded by rough interfaces and another semi-infinite medium (medium 2) with a permittivity $\varepsilon_2(\omega)$. The layer has discrete randomly distributed scatterers. To calculate the scattered field we introduce Green's functions associated with the different media. With a source inside medium 1, Green's functions satisfy the propagation equations:

$$\nabla\times\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{01}(\boldsymbol{r},\boldsymbol{r}_0)-\varepsilon_0(\omega)K_{\mathrm{vac}}^2\boldsymbol{G}_{\mathrm{SV}}^{01}(\boldsymbol{r},\boldsymbol{r}_0)=0, \tag{1}$$

$$\nabla\times\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)-\varepsilon_{\mathrm{V}}(\boldsymbol{r},\omega)K_{\mathrm{vac}}^2\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)=\delta(\boldsymbol{r}-\boldsymbol{r}_0)\boldsymbol{I}, \tag{2}$$

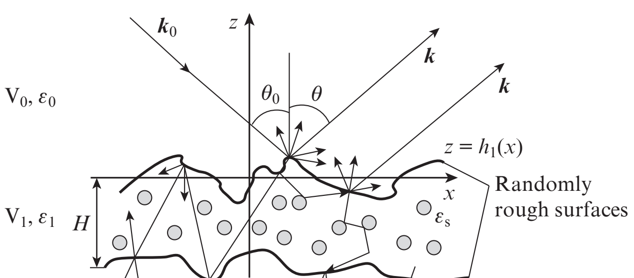

Figure 1. Definition of the random volume with rough boundaries.

$$\nabla\times\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{21}(\boldsymbol{r},\boldsymbol{r}_0)-\varepsilon_2(\omega)K_{\mathrm{vac}}^2\boldsymbol{G}_{\mathrm{SV}}^{21}(\boldsymbol{r},\boldsymbol{r}_0)=0. \tag{3}$$

Here, the superscripts indicate the receiver and source locations in media 0, 1 and 2, respectively; the subscript SV shows that Green's function takes into account the interactions between the random volume and the rough interfaces; $K_{\mathrm{vac}} = \omega/c$; and c is the speed of light in vacuum. Inside the medium with the permittivity $\varepsilon_1(\omega)$, we consider a set of N scatterers which are assumed spherical with a radius r_{s} and a permittivity $\varepsilon_{\mathrm{s}}(\omega)$. In the following we consider two types of scatterers with different radii r and permittivities ε denoted by a and b, respectively. The formulas can be generalised to N types of particles. The permittivity ε_{V} for the layer is given by

$$\varepsilon_{\mathrm{V}}(\boldsymbol{r},\omega)=\varepsilon_1(\omega)+\sum_{a=1}^{N_a}[\varepsilon_a(\omega)-\varepsilon_1(\omega)]\Theta_a(\boldsymbol{r}-\boldsymbol{r}_a) + \sum_{b=1}^{N_b}[\varepsilon_b(\omega)-\varepsilon_1(\omega)]\Theta_b(\boldsymbol{r}-\boldsymbol{r}_b), \tag{4}$$

where $\boldsymbol{r}_1, ..., \boldsymbol{r}_N$ are the positions of the centres of the particles, $N_a + N_b = N$ is the total number of particles, and $\Theta_{a,b}$ describes the shape of the particles. Note that the results can be generalised to non-spherical particles. For $\Theta_{a,b}$ we have:

$$\Theta_{a,b}(\boldsymbol{r})=\begin{cases}1 \text{ at } \|\boldsymbol{r}\| < r_{a,b},\\ 0 \text{ at } \|\boldsymbol{r}\| > r_{a,b},\end{cases} \tag{5}$$

where $r_{a,b}$ are the radii of the particles. We impose boundary conditions on the two rough surfaces. For the upper rough surface, we obtain:

$$\hat{\boldsymbol{n}}_{\mathrm{s}1}\cdot\varepsilon_1(\omega)\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)=\hat{\boldsymbol{n}}_{\mathrm{s}1}\cdot\varepsilon_0(\omega)\boldsymbol{G}_{\mathrm{SV}}^{01}(\boldsymbol{r},\boldsymbol{r}_0), \tag{6}$$

$$\hat{\boldsymbol{n}}_{\mathrm{s}1}\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)=\hat{\boldsymbol{n}}_{\mathrm{s}1}\times\boldsymbol{G}_{\mathrm{SV}}^{01}(\boldsymbol{r},\boldsymbol{r}_0), \tag{7}$$

$$\hat{\boldsymbol{n}}_{\mathrm{s}1}\cdot[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)]=\hat{\boldsymbol{n}}_{\mathrm{s}1}\cdot[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{01}(\boldsymbol{r},\boldsymbol{r}_0)], \tag{8}$$

$$\hat{\boldsymbol{n}}_{\mathrm{s}1}\times[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)]=\hat{\boldsymbol{n}}_{\mathrm{s}1}\times[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{01}(\boldsymbol{r},\boldsymbol{r}_0)], \tag{9}$$

and for the lower rough surface, we derive:

$$\hat{\boldsymbol{n}}_{\mathrm{s}2}\cdot\varepsilon_1(\omega)\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)=\hat{\boldsymbol{n}}_{\mathrm{s}2}\cdot\varepsilon_2(\omega)\boldsymbol{G}_{\mathrm{SV}}^{21}(\boldsymbol{r},\boldsymbol{r}_0), \tag{10}$$

$$\hat{\boldsymbol{n}}_{\mathrm{s}2}\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)=\hat{\boldsymbol{n}}_{\mathrm{s}2}\times\boldsymbol{G}_{\mathrm{SV}}^{21}(\boldsymbol{r},\boldsymbol{r}_0), \tag{11}$$

$$\hat{\boldsymbol{n}}_{\mathrm{s}2}\cdot[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)]=\hat{\boldsymbol{n}}_{\mathrm{s}2}\cdot[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{21}(\boldsymbol{r},\boldsymbol{r}_0)], \tag{12}$$

$$\hat{\boldsymbol{n}}_{\mathrm{s}2}\times[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{11}(\boldsymbol{r},\boldsymbol{r}_0)]=\hat{\boldsymbol{n}}_{\mathrm{s}2}\times[\nabla\times\boldsymbol{G}_{\mathrm{SV}}^{21}(\boldsymbol{r},\boldsymbol{r}_0)], \tag{13}$$

where $\hat{\boldsymbol{n}}_{\mathrm{s}1}$ and $\hat{\boldsymbol{n}}_{\mathrm{s}2}$ are the exterior normals to the two rough surfaces. The solutions of the propagation equations are unique if we impose the radiation condition at infinity for media 0 and 2. In order to separate the contribution from the rough surfaces and the random medium, we introduce the dyadic Green's functions $\boldsymbol{G}_{\mathrm{S}}^{11}$, $\boldsymbol{G}_{\mathrm{S}}^{01}$, $\boldsymbol{G}_{\mathrm{S}}^{21}$, which describe scattering by a layer with rough boundaries but without a random medium taken into account. These functions satisfy similar propagation equations and boundary conditions as Green's functions $\boldsymbol{G}_{\mathrm{SV}}$, where the permittivity $\varepsilon_{\mathrm{V}}(\omega)$ due to the random medium is replaced by the effective permittivity $\varepsilon_{\mathrm{e}}(\omega)$ in the equations, with this assumption the layer is considered homogeneous. In the following sections, we will determine this effective permittivity. Let us now find the potential function, which describes the interaction between the wave and the particles and then present the expression for Green's functions. The system of differential equations with boundary conditions and radiative condition at infinity can be transformed into integral equations. For the source in medium 1, the system of integral equations related to the potential function has the form:

$$\boldsymbol{G}_{\mathrm{SV}}^{01} = \boldsymbol{G}_{\mathrm{S}}^{01} + \boldsymbol{G}_{\mathrm{S}}^{01}\cdot\boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{SV}}^{11}, \tag{14}$$

$$\boldsymbol{G}_{\mathrm{SV}}^{11} = \boldsymbol{G}_{\mathrm{S}}^{11} + \boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{SV}}^{11}, \tag{15}$$

$$\boldsymbol{G}_{\mathrm{SV}}^{21} = \boldsymbol{G}_{\mathrm{S}}^{21} + \boldsymbol{G}_{\mathrm{S}}^{21}\cdot\boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{SV}}^{11}. \tag{16}$$

Here use is made of the following operator notation:

$$[\boldsymbol{A}\cdot\boldsymbol{B}](\boldsymbol{r}_1,\boldsymbol{r}_0) = \int_{V_1} \mathrm{d}^3\boldsymbol{r}_1 \boldsymbol{A}(\boldsymbol{r},\boldsymbol{r}_1)\cdot\boldsymbol{B}(\boldsymbol{r}_1,\boldsymbol{r}_0). \tag{17}$$

and the potential function can be written as:

$$\boldsymbol{V}^{11}(\boldsymbol{r},\boldsymbol{r}_0,\omega) = \delta(\boldsymbol{r}-\boldsymbol{r}_0)\boldsymbol{V}^1(\boldsymbol{r}), \tag{18}$$

$$\boldsymbol{V}^1(\boldsymbol{r},\omega) = K_{\mathrm{vac}}^2[\varepsilon_{\mathrm{V}}(\boldsymbol{r},\omega) - \varepsilon_{\mathrm{e}}(\omega)]\boldsymbol{I}. \tag{19}$$

A direct demonstration of these equations involves integral theorems, but it is easier to use the uniqueness of the solution and verify *a posteriori* that the integral equations satisfy the propagation equations and the boundary conditions.

Our aim in this paper is to find a system of equations, with which we can calculate the effective permittivity. In solving equation (15) by iteration, we obtain that:

$$\boldsymbol{G}_{\mathrm{SV}}^{11} = \boldsymbol{G}_{\mathrm{S}}^{11} + \boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11} + \boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11} + \dots . \tag{20}$$

We can rewrite the Lippmann–Schwinger equation (15) by introducing the transition operator (T-operator) $\boldsymbol{T}_{\mathrm{SV}}^{11}$, which is defined by:

$$\boldsymbol{G}_{\mathrm{SV}}^{11} = \boldsymbol{G}_{\mathrm{S}}^{11} + \boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{T}_{\mathrm{SV}}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11}. \tag{21}$$

Using definition (21) and equation (15) we can express the operator $\boldsymbol{T}_{\mathrm{SV}}^{11}$ in terms of $\boldsymbol{V}^{11}$:

$$\boldsymbol{T}_{\mathrm{SV}}^{11} = \boldsymbol{V}^{11} + \boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{T}_{\mathrm{SV}}^{11}, \tag{22}$$

$$\boldsymbol{T}_{\mathrm{SV}}^{11} = \boldsymbol{V}^{11} + \boldsymbol{T}_{\mathrm{SV}}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{V}^{11}. \tag{23}$$

The transition operator $\boldsymbol{T}_{\mathrm{SV}}^{11}$ contains all the scattering processes occurring in a random medium. If we know this operator, then we can calculate all the electromagnetic fields in different media by using the Lippman–Schwinger equations.

3. Coherent potential approximation and effective medium theory

3.1. Definition of ensemble averages

To calculate the coherent field scattering in a random layer, we first define the averaging procedure. The symbols $\langle\dots\rangle_{\mathrm{S}}$ and $\langle\dots\rangle_{\mathrm{V}}$ denote respectively the ensemble average over the surface and volume disorder. We suppose that the rough surfaces and the random medium properties are statistically independent. The ensemble average over a random medium is defined by:

$$\langle f\rangle_{\mathrm{V}} = \int_{V_1} \mathrm{d}^3\boldsymbol{r}_1 \dots \mathrm{d}^3\boldsymbol{r}_N f(\boldsymbol{r}_1,\dots,\boldsymbol{r}_N)p(\boldsymbol{r}_1,\dots,\boldsymbol{r}_N), \tag{24}$$

where $\boldsymbol{r}_1,\dots,\boldsymbol{r}_N$ are the positions of the particles and $p(\boldsymbol{r}_1,\dots,\boldsymbol{r}_N)$ is the probability density function of finding the N particles at positions $\boldsymbol{r}_1,\dots,\boldsymbol{r}_N$ in the layer. We will use the expansion of this density function over conditional probabilities:

$$p(\boldsymbol{r}_1,\dots,\boldsymbol{r}_N) = p(\boldsymbol{r}_i)p(\boldsymbol{r}_1,\dots,\hat{\boldsymbol{r}}_i,\dots,\boldsymbol{r}_N|\boldsymbol{r}_i), \tag{25}$$

$$p(\boldsymbol{r}_1,\dots,\boldsymbol{r}_N) = p(\boldsymbol{r}_i)p(\boldsymbol{r}_j|\boldsymbol{r}_i)\,p(\boldsymbol{r}_1,\dots,\hat{\boldsymbol{r}}_i,\dots,\hat{\boldsymbol{r}}_j,\dots,\boldsymbol{r}_N|\boldsymbol{r}_i,\boldsymbol{r}_j), \tag{26}$$

where the cap ˆ indicates the terms that are not taken into account. The function $p(\boldsymbol{r}_i)$ defines the density probability to find a particle at point $\boldsymbol{r}_i$. The quantity $p(\boldsymbol{r}_j|\boldsymbol{r}_i)$ is the conditional probability to find a particle at point $\boldsymbol{r}_j$ at a given particle at point $\boldsymbol{r}_i$. If the particles are uniformly distributed inside the random medium V1, then the single particle density function is $p(\boldsymbol{r}_j) = 1/V_1$, where V_1 is the value of the volume V1. We also define the pair distribution function as $g(\|\boldsymbol{r}_j-\boldsymbol{r}_i\|) = p(\boldsymbol{r}_j|\boldsymbol{r}_i)/p(\boldsymbol{r}_j)$. This function only depends on the distance between two particles if we suppose that the distributions of the particles are statistically homogeneous and isotropic. The normalisation factor V_1 is chosen so that the particles are far away from each others, and their positions are considered uncorrelated. By using these conditional probability functions, we can define the conditional averages:

$$\langle f\rangle_{\mathrm{V}\,\boldsymbol{r}_i} = \int_{V_1} \mathrm{d}^3\boldsymbol{r}_1 \dots \widehat{d^3\boldsymbol{r}_i} \dots \mathrm{d}^3\boldsymbol{r}_N f(\boldsymbol{r}_1,\dots,\boldsymbol{r}_i,\dots,\boldsymbol{r}_N)$$
$$\times p(\boldsymbol{r}_1,\dots,\hat{\boldsymbol{r}}_i,\dots,\boldsymbol{r}_N|\boldsymbol{r}_i), \tag{27}$$

$$\langle f\rangle_{\mathrm{V}\,\boldsymbol{r}_i,\boldsymbol{r}_j} = \int_{V_1} \mathrm{d}^3\boldsymbol{r}_1 \dots \widehat{d^3\boldsymbol{r}_i} \dots \widehat{d^3\boldsymbol{r}_j} \dots \mathrm{d}^3\boldsymbol{r}_N$$
$$\times f(\boldsymbol{r}_1,\dots,\boldsymbol{r}_i,\dots,\boldsymbol{r}_j,\dots,\boldsymbol{r}_N)p(\boldsymbol{r}_1,\dots,\hat{\boldsymbol{r}}_i,\dots,\hat{\boldsymbol{r}}_j,\dots,\boldsymbol{r}_N|\boldsymbol{r}_i,\boldsymbol{r}_j). \tag{28}$$

3.2. Coherent potential approximation

The previously defined T-operator is useful to calculate the average field over the volume disorder $\langle\boldsymbol{G}_{\mathrm{SV}}^{11}\rangle_{\mathrm{V}}$. We obtain the Dyson equation in which the potential operator is replaced by the mass operator containing all irreducible diagrams in the Feynman representation. The mass operator can be expressed by $\langle\boldsymbol{T}_{\mathrm{SV}}^{11}\rangle_{\mathrm{V}}$. The averaged electric field satisfies the Dyson equation with the mass operator related to the effective dielectric permittivity of a homogenised structure. To determine $\varepsilon_{\mathrm{e}}(\omega)$ we use the coherent potential approximation (CPA), which is defined by:

$$\langle\boldsymbol{G}_{\mathrm{SV}}^{11}\rangle_{\mathrm{V}} = \boldsymbol{G}_{\mathrm{S}}^{11}. \tag{29}$$

With (21) taken into account, we can demonstrate that (29) is equivalent to the equality

$$\langle\boldsymbol{T}_{\mathrm{SV}}^{11}\rangle_{\mathrm{V}} = 0, \tag{30}$$

where

$$\boldsymbol{T}_{\mathrm{SV}}^{11} = \boldsymbol{V}^{11} + \boldsymbol{V}^{11}\cdot\boldsymbol{G}_{\mathrm{S}}^{11}\cdot\boldsymbol{T}_{\mathrm{SV}}^{11}.$$

Equations (30) and (22) give a finite closed system of equations where the unknown is the quantity $\varepsilon_{\mathrm{e}}(\omega)$.

3.3. Expression of the scattering operator $t_{r_i}^{11}$

We express (22) in terms of the scattering operator $t_{r_i}^{11}$ for a particle with a permittivity ε_{si} located at point r_i inside an infinite homogeneous medium. This scattering operator is given by:

$$t_{r_i}^{11} = [I - v_{r_i}^{11}\cdot G_1^{\infty}]^{-1}\cdot v_{r_i}^{11} \tag{31}$$

or the following different expressions:

$$t_{r_i}^{11} = v_{r_i}^{11} + v_{r_i}^{11}\cdot G_1^{\infty}\cdot t_{r_i}^{11}, \tag{32}$$

$$t_{r_i}^{11} = v_{r_i}^{11} + v_{r_i}^{11}\cdot G_1^{\infty}\cdot v_{r_i}^{11} + v_{r_i}^{11}\cdot G_1^{\infty}\cdot v_{r_i}^{11}\cdot G_1^{\infty}\cdot v_{r_i}^{11} + \ldots, \tag{33}$$

$$t_{r_i}^{11}(r, r_0) = v_{r_i}^{1}(r)\,\delta(r - r_0) + \int d^3r_1\, v_{r_i}^{1}(r)\cdot G_1^{\infty}(r, r_1)\cdot t_{r_i}^{11}(r_1, r_0). \tag{34}$$

Here, G_1^{∞} is Green's function in an unbounded homogeneous medium characterised by the effective permittivity ε_e. The scattering potential $v_{r_i}^{11}$ is given by

$$v_{r_i}^{11}(r, r_0) = (2\pi)^2\,\delta(r - r_0)\, v_{r_i}^{1}(r), \tag{35}$$

$$v_{r_i}^{1}(r) = K_{\text{vac}}^2(\varepsilon_{si} - \varepsilon_1)\,\Theta_s(r - r_i)\,I. \tag{36}$$

3.4. Introduction of the transition operator $\tilde{T}_{\text{SV}}^{11}$

To rewrite equation (22) trough the operators $t_{r_i}^{11}$, we will use the formula

$$V^{11}(r, r_0) = (2\pi)^2\,\delta(r - r_0)\, K_{\text{vac}}^2(\varepsilon_V(r) - \varepsilon_e)\, I \tag{37}$$

and define the following operators:

$$\tilde{T}_{\text{SV}}^{11} = T_{\text{SV}}^{11} + Q_{\text{SV}}^{11} \tag{38}$$

for

$$Q_{\text{SV}}^{11} = W^{11} + W^{11}\cdot G_S^{11}\cdot T_{\text{SV}}^{11}, \tag{39}$$

where

$$W^{11}(r, r_0) = (2\pi)^2\,\delta(r - r_0)\, K_{\text{vac}}^2(\varepsilon_e - \varepsilon_1)\, I, \tag{40}$$

$$\tilde{V}^{11} = V^{11} + W^{11}. \tag{41}$$

With (35)–(37), (40) and (41) taken into account, we obtain

$$\tilde{V}^{11} = \sum_{a=1}^{N_a} v_{r_a}^{11} + \sum_{b=1}^{N_b} v_{r_b}^{11}. \tag{42}$$

From (22) and (41), using definitions (38) and (39), we obtain the expressions:

$$\tilde{T}_{\text{SV}}^{11} = \tilde{V}^{11} + \tilde{V}^{11}\cdot G_S^{11}\cdot T_{\text{SV}}^{11} \tag{43}$$

$$= \tilde{V}^{11} + \tilde{V}^{11}\cdot G_S^{11}\cdot(\tilde{T}_{\text{SV}}^{11} - Q_{\text{SV}}^{11}). \tag{44}$$

With (42) and (44) taken into account, we represent $\tilde{T}_{\text{SV}}^{11}$ in the form:

$$\tilde{T}_{\text{SV}}^{11} = \sum_{a=1}^{N_a} C_{\text{SV},r_a}^{11} + \sum_{b=1}^{N_b} C_{\text{SV},r_b}^{11}, \tag{45}$$

where

$$C_{\text{SV},r_a}^{11} = v_{r_a}^{11} + v_{r_a}^{11}\cdot G_S^{11}\cdot\left(\sum_{a=1}^{N_a} C_{\text{SV},r_a}^{11} + \sum_{b=1}^{N_b} C_{\text{SV},r_b}^{11} - Q_{\text{SV}}^{11}\right), \tag{46}$$

and finally obtain a similar expression for C_{SV,r_b}^{11}.

3.5. Definition of the scattering operator t_{S,r_i}^{11} for a particle in a random medium

If we subtract $v_{r_a}^{11}\cdot G_S^{11}\cdot C_{\text{SV},r_a}^{11}$ from the two members of equation (46), we obtain

$$(I - v_{r_a}^{11}\cdot G_S^{11})\cdot C_{\text{SV},r_a}^{11} = v_{r_a}^{11} + v_{r_a}^{11}\cdot G_S^{11}\cdot\left(\sum_{\substack{a'=1,\\ a'\neq a}}^{N_a} C_{\text{SV},r_{a'}}^{11} + \sum_{b=1}^{N_b} C_{\text{SV},r_b}^{11} - Q_{\text{SV}}^{11}\right), \tag{47}$$

which can be written as

$$C_{\text{SV},r_a}^{11} = t_{S,r_a}^{11} + t_{S,r_a}^{11}\cdot G_S^{11}\cdot\left(\sum_{\substack{a'=1,\\ a'\neq a}}^{N_a} C_{\text{SV},r_{a'}}^{11} + \sum_{b=1}^{N_b} C_{\text{SV},r_b}^{11} - Q_{\text{SV}}^{11}\right), \tag{48}$$

where

$$t_{S,r_a}^{11} = [I - v_{r_a}^{11}\cdot G_S^{11}]^{-1}\cdot v_{r_a}^{11} \tag{49}$$

$$= v_{r_a}^{11} + v_{r_a}^{11}\cdot G_S^{11}\cdot t_{S,r_a}^{11}. \tag{50}$$

The operator t_{S,r_a}^{11} is the scattering operator for a particle located at point r_a inside the layer, and this operator takes into account the interactions of the field with the rough surfaces. In writing G_S^{11} in the form

$$G_S^{11} = G_1^{\infty} + \delta G_S^{11}, \tag{51}$$

where δG_S^{11} is Green's function describing the wave interactions with the rough boundaries of the effective medium, expression (50) can be written in the form

$$[I - v_{r_i}^{11}\cdot G_1^{\infty}]\cdot t_{S,r_i}^{11} = v_{r_i}^{11} + v_{r_i}^{11}\cdot\delta G_S^{11}\cdot t_{S,r_i}^{11}. \tag{52}$$

Whence, we find

$$t_{S,r_i}^{11} = t_{r_i}^{11} + t_{r_i}^{11}\cdot\delta G_S^{11}\cdot t_{S,r_i}^{11}, \tag{53}$$

where $t_{r_i}^{11}$ is defined in (31).

3.6. Renormalisation of the particle potential

The operator $t_{r_i}^{11}$ describes the scattering of a particle in an infinite homogeneous medium (Fig. 2). If we want the operator $t_{r_i}^{11}$ to describe the scattering of a particle with the permittivity ε_s inside a medium with the permittivity ε_e, it is convenient to renormalise the permittivity of the particle by introducing the permittivity $\bar{\varepsilon} = \varepsilon_{si} - (\varepsilon_1 - \varepsilon_e)$. This yields

$$v_{r_i}^{1}(r) = K_{\text{vac}}^2(\varepsilon_{si} - \varepsilon_1)\,\Theta_s(r - r_i)\,I \tag{54}$$

$$= K_{\text{vac}}^2(\bar{\varepsilon}_s - \varepsilon_e)\,\Theta_s(r - r_i)\,I. \tag{55}$$

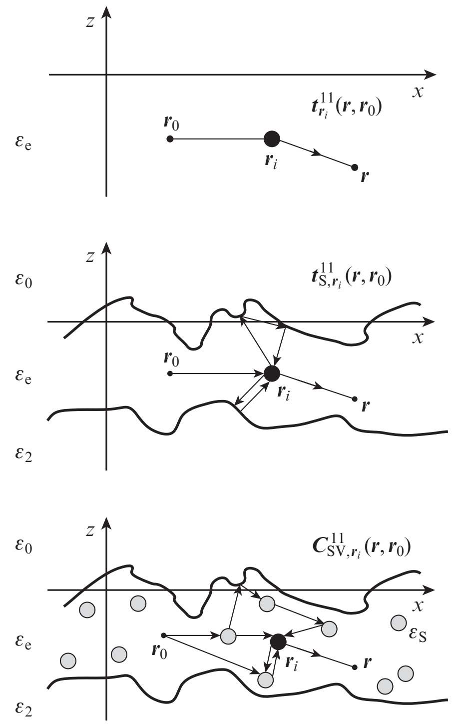

Figure 2. Representations of the operators $\boldsymbol{t}^{11}_{\boldsymbol{r}_i}$, $\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}$ and $\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i}$.

It is obvious that $\boldsymbol{t}^{11}_{\boldsymbol{r}_i}$ is the scattering operator for a particle with the permittivity $\bar{\varepsilon}_s$ inside a medium with the permittivity ε_e (see Fig. 2). In other words, the operator $\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}$ describes the scattering of a particle located at point $\boldsymbol{r}_i$ inside the layer V1 taking into account the interactions with the rough boundaries (Fig. 2). Iterating equation (53) we obtain

$$\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i} = \boldsymbol{t}^{11}_{\boldsymbol{r}_i} + \boldsymbol{t}^{11}_{\boldsymbol{r}_i}\cdot\delta\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{t}^{11}_{\boldsymbol{r}_i} + \boldsymbol{t}^{11}_{\boldsymbol{r}_i}\cdot\delta\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{t}^{11}_{\boldsymbol{r}_i}\cdot\delta\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{t}^{11}_{\boldsymbol{r}_i} + \ldots. \quad (56)$$

Note that the first term describes the scattering process of one particle and the following terms represent the interactions between a particle and rough boundaries because the operator $\delta\boldsymbol{G}^{11}_{\mathrm{S}}$ involves the interactions of the waves with the rough boundaries of the effective medium (Fig. 2). Equation (48) represents the multiple scattering process inside the layer bounded by rough surfaces:

$$\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i} = \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i} + \sum_{\substack{j=1,\\ j\neq i}}^{N} \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_j}$$
$$+ \sum_{\substack{j=1,\\ j\neq i}}^{N}\sum_{\substack{k=1,\\ k\neq j}}^{N} \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_j}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_k} + \ldots. \quad (57)$$

The quantity $\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i}$ represents the field scattered by particles located at point $\boldsymbol{r}_i$ and takes into account the interactions with other particles and rough boundaries. Due to the introduction of the effective medium $\varepsilon_e(\omega)$ in the expression for $\boldsymbol{G}^{11}_{\mathrm{S}}$, we find that in equation (48) the contributions of multiple scattering are damped by the term $\boldsymbol{Q}^{11}_{\mathrm{SV}}$.

3.7. Determination of the effective permittivity

Averaging equation (38) according to definitions (39) and (40) and under the CPA assumption (30), we have

$$(2\pi)^2\,\delta(\boldsymbol{r}-\boldsymbol{r}_0)\,\varepsilon_e K^2_{\mathrm{vac}}\boldsymbol{I}$$
$$= (2\pi)^2\,\delta(\boldsymbol{r}-\boldsymbol{r}_0)\,\varepsilon_1 K^2_{\mathrm{vac}}\boldsymbol{I} + \langle\tilde{\boldsymbol{T}}^{11}_{\mathrm{SV}}(\boldsymbol{r},\boldsymbol{r}_0)\rangle_{\mathrm{V}}. \quad (58)$$

Taking into account the definitions of the conditional averages, from (47) we obtain

$$\langle\tilde{\boldsymbol{T}}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} = \sum_{a=1}^{N_a}\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_a}\rangle_{\mathrm{V}} + \sum_{b=1}^{N_b}\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_b}\rangle_{\mathrm{V}}$$
$$= \sum_{a=1}^{N_a}\int_{V_1}\mathrm{d}^3\boldsymbol{r}_a p(\boldsymbol{r}_a)\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_a}\rangle_{\mathrm{V}\boldsymbol{r}_a} + \sum_{b=1}^{N_b}\int_{V_1}\mathrm{d}^3\boldsymbol{r}_b p(\boldsymbol{r}_b)\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_b}\rangle_{\mathrm{V}\boldsymbol{r}_b}$$
$$= n_a\int_{V_1}\mathrm{d}^3\boldsymbol{r}_a\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_a}\rangle_{\mathrm{V}\boldsymbol{r}_a} + n_b\int_{V_1}\mathrm{d}^3\boldsymbol{r}_b\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_b}\rangle_{\mathrm{V}\boldsymbol{r}_b}, \quad (59)$$

where $n_i = N_i/V_1$ defines the density of the particles of type a or b. In (59), we used the propriety $\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i}\rangle_{\mathrm{V}\boldsymbol{r}_i} = \langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j}\rangle_{\mathrm{V}\boldsymbol{r}_j}$ for $i \neq j$, and the assumption of a random medium, which is statistically homogeneous. We average equation (48) following the definition of the conditional average $\langle\ldots\rangle_{\mathrm{V}\boldsymbol{r}_i}$, and using (29) we obtain ($i = a$ or b)

$$\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i}\rangle_{\mathrm{V}\boldsymbol{r}_i} = \langle\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}\rangle_{\mathrm{V}\boldsymbol{r}_i}$$
$$+ \sum_{\substack{j=1,\\ j\neq i}}^{N}\int_{V_1}\mathrm{d}^3\boldsymbol{r}_j p(\boldsymbol{r}_j|\boldsymbol{r}_i)\,\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\langle\boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j}\rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j}$$
$$- \langle\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\boldsymbol{Q}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}\boldsymbol{r}_i}. \quad (60)$$

Note that $\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}$ is the scattering operator for a particle located at point $\boldsymbol{r}_i$, which is independent of the variable $\boldsymbol{r}_j$ for $j \neq i$. It follows that the process of averaging $\langle\ldots\rangle_{\mathrm{V}\boldsymbol{r}_i}$ does not affect the expression for $\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}$. Moreover, the averaging of equation (38) leads to the fact that

$$\langle\boldsymbol{Q}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}\boldsymbol{r}_i} = \boldsymbol{W}^{11} + \boldsymbol{W}^{11}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\langle\boldsymbol{T}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}\boldsymbol{r}_i}. \quad (61)$$

This equation can be simplified by using the CPA approximation, $\langle\boldsymbol{T}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} = 0$ and can be written as

$$\int_{V_1}\mathrm{d}^3\boldsymbol{r}_i p(\boldsymbol{r}_i)\langle\boldsymbol{T}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}\boldsymbol{r}_i} = 0. \quad (62)$$

Identity (62) is valid for any volume V_1. Thus, we have $\langle\boldsymbol{T}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}\boldsymbol{r}_i} = 0$. Accordingly, from equation (61) we obtain

$$\langle\boldsymbol{Q}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}\boldsymbol{r}_i} = \boldsymbol{W}^{11}. \quad (63)$$

Taking into account definitions (38), (39) and the coherent potential approximation, we can write the expressions:

$$\langle\boldsymbol{Q}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} = \boldsymbol{W}^{11} + \boldsymbol{W}^{11}\cdot\boldsymbol{G}^{11}_{\mathrm{S}}\cdot\langle\boldsymbol{T}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} \quad (64)$$
$$= \boldsymbol{W}^{11}, \quad (65)$$

and

$$\langle\tilde{\boldsymbol{T}}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} = \langle\boldsymbol{T}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} + \langle\boldsymbol{Q}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}} \quad (66)$$
$$= \langle\boldsymbol{Q}^{11}_{\mathrm{SV}}\rangle_{\mathrm{V}}. \quad (67)$$

Taking into account expressions (63), (65), (67) and (59), we obtain for $i \in [1, N_i]$

$$\langle \boldsymbol{Q}^{11}_{\mathrm{SV}} \rangle_{\mathrm{V}\boldsymbol{r}_i} = \langle \tilde{\boldsymbol{T}}^{11}_{\mathrm{SV}} \rangle_{\mathrm{V}} \tag{68}$$

$$= n_i \int_{V_1} \mathrm{d}^3\boldsymbol{r}_j \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_j}. \tag{69}$$

Then, equation (60) is written in the form

$$\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i} \rangle_{\mathrm{V}\boldsymbol{r}_i} = \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i} + n_i \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i} \cdot \boldsymbol{G}^{11}_{\mathrm{S}} \times \int_{V_1} \mathrm{d}^3\boldsymbol{r}_j [g(\|\boldsymbol{r}_j - \boldsymbol{r}_i\|) \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j} - \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_j}], \tag{70}$$

where we used the approximation $n_i \simeq (N_i - 1)/V_1$, which is valid for a large number of particles ($N \gg 1$).

Following the same procedure, we can average equation (48) with $\langle \ldots \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j}$, obtain an equation for $\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j}$, which depends on the function $\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_k} \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j,\boldsymbol{r}_k}$, and iterate this procedure. We generate a system of equations for the unknown functions $\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i} \rangle_{\mathrm{V}\boldsymbol{r}_i}$, $\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j}$, $\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_k} \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j,\boldsymbol{r}_k}$, ... We can close this infinite system by using the QCA, which requires the fulfilment of the condition

$$\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_i,\boldsymbol{r}_j} = \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_j}. \tag{71}$$

This approximation is strictly met when the particles have fixed positions, as in a crystal. The quasi-crystalline approximation consists in neglecting the fluctuation of the electromagnetic field interacting with a particle located at point $\boldsymbol{r}_j$, due to a deviation of a particle located at point $\boldsymbol{r}_i$ from its average position. With this approximation and equation (58) taken into account, the effective permittivity $\varepsilon_{\mathrm{e}}(\omega)$ satisfies the system of equations

$$(2\pi)^2 \delta(\boldsymbol{r} - \boldsymbol{r}_0) \varepsilon_{\mathrm{e}} K^2_{\mathrm{vac}} \boldsymbol{I} = (2\pi)^2 \delta(\boldsymbol{r} - \boldsymbol{r}_0) \varepsilon_1 K^2_{\mathrm{vac}} \boldsymbol{I} + n_a \int_{V_1} \mathrm{d}^3\boldsymbol{r}_a \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_a}(\boldsymbol{r},\boldsymbol{r}_0) \rangle_{\mathrm{V}\boldsymbol{r}_a} + n_b \int_{V_1} \mathrm{d}^3\boldsymbol{r}_b \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_b}(\boldsymbol{r},\boldsymbol{r}_0) \rangle_{\mathrm{V}\boldsymbol{r}_b}, \tag{72}$$

$$\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i} \rangle_{\mathrm{V}\boldsymbol{r}_i} = \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i} + n_i \boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i} \cdot \boldsymbol{G}^{11}_{\mathrm{S}} \times \int_{V_1} \mathrm{d}^3\boldsymbol{r}_j [g(\|\boldsymbol{r}_j - \boldsymbol{r}_i\|) - 1] \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_j}. \tag{73}$$

The function $g(\|\boldsymbol{r}_j - \boldsymbol{r}_i\|)$ is the pair distribution function defined by

$$g(\|\boldsymbol{r}_j - \boldsymbol{r}_i\|) = p(\boldsymbol{r}_j|\boldsymbol{r}_i)/p(\boldsymbol{r}_j). \tag{74}$$

This is one of the main results of this paper. Equations (72) and (73) can be simplified if we assume that, in the expression $\boldsymbol{G}^{11}_{\mathrm{S}} = \boldsymbol{G}^{\infty}_1 + \delta\boldsymbol{G}^{11}_{\mathrm{S}}$, the contribution $\delta\boldsymbol{G}^{11}_{\mathrm{S}}$ of the rough surface scattering can be neglected when the condition $K''_{\mathrm{e}}H \gg 1$ with $K''_{\mathrm{e}} = \mathrm{Im}\, K_{\mathrm{e}}$ is satisfied. We define the extinction length as $l_{\mathrm{e}} = 1/(2K''_{\mathrm{e}})$. This condition means that the layer thickness must be greater than the extinction length. In this condition we replace in (73) Green's function $\boldsymbol{G}^{11}_{\mathrm{S}}$ by $\boldsymbol{G}^{\infty}_1$ and the operator $\boldsymbol{t}^{11}_{\mathrm{S},\boldsymbol{r}_i}$ by $\boldsymbol{t}^{11}_{\boldsymbol{r}_i}$. Then, we obtain

$$\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i} \rangle_{\mathrm{V}\boldsymbol{r}_i} = \boldsymbol{t}^{11}_{\boldsymbol{r}_i} + n_i \boldsymbol{t}^{11}_{\boldsymbol{r}_i} \cdot \boldsymbol{G}^{\infty}_1 \times \int_{V_1} \mathrm{d}^3\boldsymbol{r}_j [g(\|\boldsymbol{r}_j - \boldsymbol{r}_i\|) - 1] \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_j} \rangle_{\mathrm{V}\boldsymbol{r}_j}. \tag{75}$$

In this procedure we neglect the surface effects in calculating the effective permittivity. To express (75) in Fourier space, we use the definition

$$f(\boldsymbol{k}|\boldsymbol{k}_0) = \iint \frac{\mathrm{d}^2\boldsymbol{r}}{(2\pi)^3} \frac{\mathrm{d}^2\boldsymbol{r}_0}{(2\pi)^3} \exp(-\mathrm{i}\boldsymbol{k}\boldsymbol{r} + \mathrm{i}\boldsymbol{k}_0\boldsymbol{r}_0) f(\boldsymbol{r},\boldsymbol{r}_0). \tag{76}$$

In Fourier space, the translation invariance of the operator $\boldsymbol{t}^{11}_{\boldsymbol{r}_i}$ can be expressed as

$$\boldsymbol{t}^{11}_{\boldsymbol{r}_i}(\boldsymbol{k}|\boldsymbol{k}_0) = \exp[-\mathrm{i}(\boldsymbol{k} - \boldsymbol{k}_0)\boldsymbol{r}_i]\, \boldsymbol{t}^{11}_{\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0), \tag{77}$$

where $\boldsymbol{t}^{11}_{\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0) = \boldsymbol{t}^{11}_{\boldsymbol{r}_i=0}(\boldsymbol{k}|\boldsymbol{k}_0)$ is the scattering operator for a particle located at the coordinate origin. Using (77) and equation (75), we show that $\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i}(\boldsymbol{k}|\boldsymbol{k}_0) \rangle_{\mathrm{V}\boldsymbol{r}_i}$ has a similar property:

$$\langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i}(\boldsymbol{k}|\boldsymbol{k}_0) \rangle_{\mathrm{V}\boldsymbol{r}_i} = \exp[-\mathrm{i}(\boldsymbol{k} - \boldsymbol{k}_0)\boldsymbol{r}_i]\, \boldsymbol{C}^{11}_{i,\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0). \tag{78}$$

Here, $\boldsymbol{C}^{11}_{i,\mathrm{o}} = \langle \boldsymbol{C}^{11}_{\mathrm{SV},\boldsymbol{r}_i=0}(\boldsymbol{k}|\boldsymbol{k}_0) \rangle_{\mathrm{V}\boldsymbol{r}_i=0}$. Substituting (77) and (78) into equations (72) and (75), we obtain

$$\varepsilon_{\mathrm{e}} K^2_{\mathrm{vac}} \boldsymbol{I} = \varepsilon_1 K^2_{\mathrm{vac}} \boldsymbol{I} + \boldsymbol{C}^{11}_{\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0), \tag{79}$$

$$\boldsymbol{C}^{11}_{\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0) = n_a \boldsymbol{t}^{11}_{a,\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0) + n_b \boldsymbol{t}^{11}_{b,\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_0) + \int \frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^3} h(\boldsymbol{k} - \boldsymbol{k}_1)[n_a \boldsymbol{t}^{11}_{a,\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_1) + n_b \boldsymbol{t}^{11}_{b,\mathrm{o}}(\boldsymbol{k}|\boldsymbol{k}_1)] \cdot \boldsymbol{G}^{\infty}_1(\boldsymbol{k}_1) \cdot \boldsymbol{C}^{11}_{\mathrm{o}}(\boldsymbol{k}_1|\boldsymbol{k}_0), \tag{80}$$

where ($i = a$ or b)

$$\boldsymbol{t}^{11}_{i,\mathrm{o}} = \boldsymbol{v}^{11}_{i,\mathrm{o}} + \boldsymbol{v}^{11}_{i,\mathrm{o}} \cdot \boldsymbol{G}^{\infty}_1 \cdot \boldsymbol{t}^{11}_{i,\mathrm{o}}, \tag{81}$$

$$\boldsymbol{v}^{11}_{i,\mathrm{o}}(\boldsymbol{r},\boldsymbol{r}_0) = \delta(\boldsymbol{r} - \boldsymbol{r}_0) \boldsymbol{v}^{11}_{i,\mathrm{o}}(\boldsymbol{r}), \tag{82}$$

$$\boldsymbol{v}^{11}_{i,\mathrm{o}}(\boldsymbol{r}) = K^2_{\mathrm{vac}}(\bar{\varepsilon}_{\mathrm{s},i} - \varepsilon_{\mathrm{e}}) \Theta_{\mathrm{s},i}(\boldsymbol{r}) \boldsymbol{I}, \tag{83}$$

$$h(\boldsymbol{k} - \boldsymbol{k}_1) = \int \mathrm{d}^3\boldsymbol{r} \exp[-\mathrm{i}(\boldsymbol{k} - \boldsymbol{k}_1)\boldsymbol{r}][g(\|\boldsymbol{r}\|) - 1], \tag{84}$$

$$\boldsymbol{G}^{\infty}_1(\boldsymbol{k}) = \int \mathrm{d}^3\boldsymbol{r} \exp[-\mathrm{i}\boldsymbol{k}\boldsymbol{r}]\, \boldsymbol{G}^{\infty}_1(\boldsymbol{r}). \tag{85}$$

4. Effective medium theory for Rayleigh scatterers

4.1. Rayleigh scatterers

In this section we calculate the effective permittivity according to equations (79) and (80) in the case of Rayleigh scatterers. We consider spheres whose radii are small compared to the incident wavelength ($r_{\mathrm{d}} \ll \lambda$). We define ε_{d} as the permittivity of the Rayleigh scatterers, which behave as dipoles polarised by the incident field. To characterise a dipole located at the coordinate origin in a medium with the permittivity ε_{e}, we introduce its polarisability α^1_{pol}, which is related to the dipolar moment $\boldsymbol{p}_{\mathrm{dip}}$ and the electrical field $\boldsymbol{E}^{1i}(\boldsymbol{r})$ interacting with the dipole by the relation

$$\boldsymbol{p}_{\mathrm{dip}} = \varepsilon_{\mathrm{vac}} \alpha^1_{\mathrm{pol}} \boldsymbol{E}^{1i}(0). \tag{86}$$

To calculate α^1_{pol} we consider the polarisability α^1_{B}, which does not take into account the medium surrounding the sphere:

$$\boldsymbol{p}_{\rm dip} = \varepsilon_{\rm vac}\alpha^1_{\rm B}\boldsymbol{E}^1_{\rm local}(0). \qquad (87)$$

Here, $\boldsymbol{E}^1_{\rm local}$ is the local field, which acts on the dipole. Then, the following relation

$$\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}=0) \approx \Lambda^{-1}_{\rm Lor}\boldsymbol{E}^{1i}(0) \qquad (88)$$

is valid, where

$$\Lambda_{\rm Lor} = \frac{\varepsilon_{\rm d} + 2\varepsilon_1}{3\varepsilon_1} \qquad (89)$$

is the Lorentz depolarisation factor.

The polarisability of a Rayleigh scatterer in a medium is given by the relation

$$\alpha^1_{\rm pol} = \frac{3\varepsilon_1\alpha^1_{\rm B}}{\varepsilon_{\rm d} + 2\varepsilon_1}. \qquad (90)$$

We express $\alpha^1_{\rm B}$ as a function of the permittivity of the medium and the scatterers. In the case of particles with a radius smaller than the wavelength, the polarisability $\boldsymbol{p}_{\rm dip}$ can be written in terms of the polarisation vector $\boldsymbol{P}(\boldsymbol{r})$:

$$\boldsymbol{p}_{\rm dip} = \int {\rm d}^3\boldsymbol{r}\,\boldsymbol{P}(\boldsymbol{r}), \qquad (91)$$

For a sphere with a radius $r_{\rm d}$ with the permittivity $\varepsilon_{\rm d}$ in the medium with the permittivity ε_1, the polarisation vector is given by

$$\boldsymbol{P}(\boldsymbol{r}) = \varepsilon_{\rm vac}(\varepsilon_{\rm r} - \varepsilon_1)\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}), \qquad (92)$$

where $\varepsilon_{\rm r}$ is the relative permittivity of the scatterer or the medium surrounding the scatterer. This permittivity is defined by the relation:

$$\varepsilon_{\rm r} = \varepsilon_1 + (\varepsilon_{\rm d} - \varepsilon_1)\Theta_{\rm d}(\boldsymbol{r}). \qquad (93)$$

The polarisation vector $\boldsymbol{P}(\boldsymbol{r})$ is given by

$$\boldsymbol{P}(\boldsymbol{r}) = \varepsilon_{\rm vac}(\varepsilon_{\rm d} - \varepsilon_1)\Theta_{\rm d}(\boldsymbol{r})\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}). \qquad (94)$$

Using (93), the polarisability of the sphere is written in the form

$$\boldsymbol{p}_{\rm dip} = \varepsilon_{\rm vac}(\varepsilon_{\rm d} - \varepsilon_1)\int {\rm d}^3\boldsymbol{r}\,\Theta_{\rm d}(\boldsymbol{r})\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}). \qquad (95)$$

Since the scatterer is small compared to the wavelength, we assume that the field $\boldsymbol{E}^1_{\rm local}$ interacting with the sphere is uniform, i.e., $\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}) \approx \boldsymbol{E}^1_{\rm local}(0)$, and therefore

$$\boldsymbol{p}_{\rm dip} = \varepsilon_{\rm vac}(\varepsilon_{\rm d} - \varepsilon_1)\,V_{\rm d}\,\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}=0), \qquad (96)$$

where $V_{\rm d} = 4\pi r^3_{\rm d}/3$ is the scatterer volume. Comparing this expression with definition (87) for $\alpha^1_{\rm B}$, we obtain the relation:

$$\alpha^1_{\rm B} = (\varepsilon_{\rm d} - \varepsilon_1)V_{\rm d}. \qquad (97)$$

Thus, the expression of the polarisability $\alpha^1_{\rm pol}$ of a Rayleigh scatterer can be defined as a function of the permittivity:

$$\alpha^1_{\rm pol} = 3\varepsilon_1 V_{\rm d}\,\frac{\varepsilon_{\rm d} - \varepsilon_1}{\varepsilon_{\rm d} + 2\varepsilon_1}. \qquad (98)$$

Note that the approximation for the polarisation vector (98) can be written as

$$\boldsymbol{P}(\boldsymbol{r}) = \varepsilon_{\rm vac}(\varepsilon_{\rm d} - \varepsilon_1)V_{\rm d}\,\delta(\boldsymbol{r})\boldsymbol{E}^1_{\rm local}(\boldsymbol{r}). \qquad (99)$$

The Rayleigh scatterer approximation can be expressed mathematically by the relation:

$$\Theta_{\rm d}(\boldsymbol{r}) = V_{\rm d}\,\delta(\boldsymbol{r}). \qquad (100)$$

4.2. Expression of the transition operator for Rayleigh scatterers

In previous sections we defined the transition operator (34)

$$\boldsymbol{t}^{11}_{\boldsymbol{r}_i}(\boldsymbol{r},\boldsymbol{r}_0) = \boldsymbol{v}^1_{\boldsymbol{r}_i}(\boldsymbol{r})\,\delta(\boldsymbol{r}-\boldsymbol{r}_0) + \int {\rm d}^3\boldsymbol{r}_1\,\boldsymbol{v}^1_{\boldsymbol{r}_i}(\boldsymbol{r})\cdot\boldsymbol{G}^\infty_1(\boldsymbol{r},\boldsymbol{r}_1)\cdot\boldsymbol{t}^{11}_{\boldsymbol{r}_i}(\boldsymbol{r}_1,\boldsymbol{r}_0)$$

and the scattering potential (36)

$$\boldsymbol{v}^1_{\boldsymbol{r}_i}(\boldsymbol{r}) = K^2_{\rm vac}(\varepsilon_{si} - \varepsilon_1)\,\Theta_{\rm s}(\boldsymbol{r}-\boldsymbol{r}_i)\boldsymbol{I}.$$

The operator $\boldsymbol{G}^\infty_1(\boldsymbol{r},\boldsymbol{r}_1)$ has a singularity at $\boldsymbol{r} = \boldsymbol{r}_1$. To calculate the integral in expression (34), we define it as

$$\boldsymbol{G}^\infty_1(\boldsymbol{r},\boldsymbol{r}_1) = {\rm P.V.}\{\boldsymbol{G}^\infty_1(\boldsymbol{r},\boldsymbol{r}_1)\} - \frac{1}{K^2_1}\,\delta(\boldsymbol{r}-\boldsymbol{r}_1)\,\frac{\boldsymbol{I}}{3}, \qquad (101)$$

where P.V. is the principal value of the generalised function, $\delta(\boldsymbol{r}-\boldsymbol{r}_1)$ is the singularity of the operator $\boldsymbol{G}^\infty_1$, and $K^2_1 = \varepsilon_1 K^2_{\rm vac}$. This singularity is the mathematical formulation of the notion of excluded volume introduced by Lorentz to calculate the depolarisation factor (89). Substituting (101) into equation (34) and using the expressions for the Rayleigh scatterers, we obtain

$$\boldsymbol{t}^{11}_{\boldsymbol{r}_i}(\boldsymbol{r},\boldsymbol{r}_0) = \boldsymbol{v}^1_{{\rm Lor}\,\boldsymbol{r}_i}(\boldsymbol{r})\,\delta(\boldsymbol{r}-\boldsymbol{r}_0) + \int {\rm d}^3\boldsymbol{r}_1\,\boldsymbol{v}^1_{{\rm Lor}\,\boldsymbol{r}_i}(\boldsymbol{r})\cdot[{\rm P.V.}\{\boldsymbol{G}^\infty_1(\boldsymbol{r},\boldsymbol{r}_1)\}]\cdot\boldsymbol{t}^{11}_{\boldsymbol{r}_i}(\boldsymbol{r}_1,\boldsymbol{r}_0), \qquad (102)$$

where

$$\boldsymbol{v}^1_{{\rm Lor}\,\boldsymbol{r}_i}(\boldsymbol{r}) = \left[1 + \frac{\boldsymbol{v}^1_{\boldsymbol{r}_i}(\boldsymbol{r})}{3K^2_1}\right]^{-1}\boldsymbol{v}^1_{\boldsymbol{r}_i}(\boldsymbol{r}) \qquad (103)$$

$$= K^2_{\rm vac}\,3\varepsilon_1\,\frac{\varepsilon_{\rm d} - \varepsilon_1}{\varepsilon_{\rm d} + 2\varepsilon_1}\,\Theta_{\rm d}(\boldsymbol{r})\boldsymbol{I} \qquad (104)$$

$$= K^2_{\rm vac}\alpha^1_{\rm pol}\,\frac{\Theta_{\rm d}(\boldsymbol{r})}{V_{\rm d}}\,\boldsymbol{I} \qquad (105)$$

and $\alpha^1_{\rm pol}$ is given by equation (98). The potential $\boldsymbol{v}^1_{{\rm Lor}\,\boldsymbol{r}_i}(\boldsymbol{r})$ is expressed as a function of the polarisability $\alpha^1_{\rm pol}$ of each dipole contained in the particles whose geometrical shape is given by the factor $\Theta_{\rm d}(\boldsymbol{r})$. Thus, the integral equation (102) describes all the scattering processes for each dipole of polarisability $\alpha^1_{\rm pol}$ contained in the particle. Using approximation (100) for the Rayleigh scatterers, the potential $\boldsymbol{v}^1_{{\rm Lor}\,\boldsymbol{r}_i}(\boldsymbol{r}))$ can be written in the form

$$\boldsymbol{v}^1_{{\rm Lor}\,\boldsymbol{r}_i}(\boldsymbol{r}) \approx K^2_{\rm vac}\alpha^1_{\rm pol}\,\delta(\boldsymbol{r})\boldsymbol{I}. \qquad (106)$$

The Dirac distribution $\delta(\boldsymbol{r})$ allows us to calculate analytically the transition operator $\boldsymbol{t}^{11}_{\boldsymbol{r}_i}$ with the help of (102):

$$\boldsymbol{t}^{11}_{\boldsymbol{r}_i}(\boldsymbol{r}_1,\boldsymbol{r}_2) = \delta(\boldsymbol{r}_1-\boldsymbol{r}_2)\,\delta(\boldsymbol{r}_1)\boldsymbol{t}^1(\omega), \qquad (107)$$

$$\boldsymbol{t}^1(\omega) = K^2_{\rm vac}\boldsymbol{\alpha}^1_{\rm ray}, \qquad (108)$$

$$\boldsymbol{\alpha}^1_{\rm ray} = \alpha^1_{\rm pol}[\boldsymbol{I} - K^2_{\rm vac}\alpha^1_{\rm pol}{\rm P.V.}\{\boldsymbol{G}^\infty_1(\boldsymbol{r}=0)\}]^{-1}. \qquad (109)$$

Here, we took into account the fact that the free-space Green's function is translation-invariant $\boldsymbol{G}_1^\infty(\boldsymbol{r},\boldsymbol{r}_0) = \boldsymbol{G}_1^\infty(\boldsymbol{r}-\boldsymbol{r}_0)$. The calculation shows that

$$\text{P.V.}\{\boldsymbol{G}_1^\infty(\boldsymbol{r}=0)\} = \lim_{\|r\|\to 0}\left[\frac{1}{6\pi\|\boldsymbol{r}\|} + \frac{\mathrm{i}K_1}{6\pi} + O(\|\boldsymbol{r}\|)\right]\boldsymbol{I}. \quad (110)$$

The first term diverges as δ^{-1}, which is due to the use of the approximation of the punctual scatterer. We regularise this term by putting

$$\text{P.V.}\{\boldsymbol{G}_1^\infty(\boldsymbol{r}=0)\} = \left[\frac{\Lambda_{\mathrm{T}}}{6\pi} + \frac{\mathrm{i}K_1}{6\pi}\right]\boldsymbol{I}, \quad (111)$$

where Λ_{T} is a parameter that we must fix with the same order of magnitude as $1/r_{\mathrm{d}}$. The introduction of the parameter Λ_{T} can be very useful, because the scatterer can present a resonance. Thus, the addition of this term provides a broader model of point scatterers. In the following we only keep the second term of expression (111) because we only consider Rayleigh scatterers. The transition operator for a punctual scatterer is defined by replacing the permittivity ε_{d} of the scatterer by the permittivity $\bar{\varepsilon}_{\mathrm{d}} \equiv \varepsilon_{\mathrm{d}} - \varepsilon_1 + \varepsilon_{\mathrm{e}}$ and the permittivity of the medium ε_1 by the effective permittivity ε_{e} (Fig. 2). The coherent potential approximation involves the notion of an effective medium in the formulation of the transition operators $\boldsymbol{t}_{(\mathrm{e})\mathrm{o}}^{11}$.

$$\boldsymbol{t}^1(\omega) = K_{\mathrm{vac}}^2\boldsymbol{\alpha}_{\mathrm{ray}}^1,$$

$$\boldsymbol{\alpha}_{\mathrm{ray}}^1 = \alpha_{\mathrm{pol}}^1[1 - K_{\mathrm{vac}}^2\alpha_{\mathrm{pol}}^1\frac{\mathrm{i}K_1}{6\pi}]^{-1}, \quad (112)$$

$$\alpha_{\mathrm{pol}}^1 = 3\varepsilon_{\mathrm{e}}V_{\mathrm{d}}\frac{\bar{\varepsilon}_{\mathrm{d}} - \varepsilon_{\mathrm{e}}}{\bar{\varepsilon}_{\mathrm{d}} + 2\varepsilon_{\mathrm{e}}}. \quad (113)$$

According to (107), the transition operator $\boldsymbol{t}_{r_i}^{11}$ has the following form in Fourier space:

$$\boldsymbol{t}_{r_i}^{11}(\boldsymbol{k}|\boldsymbol{k}_0) = \iint\frac{\mathrm{d}^2\boldsymbol{r}}{(2\pi)^3}\frac{\mathrm{d}^2\boldsymbol{r}_0}{(2\pi)^3}\exp(-\mathrm{i}\boldsymbol{k}\boldsymbol{r} + \mathrm{i}\boldsymbol{k}_0\boldsymbol{r}_0)\boldsymbol{t}_{r_i}^{11}(\boldsymbol{r},\boldsymbol{r}_0) \quad (114)$$

$$= \boldsymbol{t}^1(\omega)\,\delta(\boldsymbol{k}-\boldsymbol{k}_0)\boldsymbol{I}. \quad (115)$$

4.3. Expression of the operator $\boldsymbol{C}_{\mathrm{o}}^{11}$

Using the results obtained in the previous sections, we can write the operator $\boldsymbol{C}_{\mathrm{o}}^{11}$ (80) in the form

$$\boldsymbol{C}_{\mathrm{o}}^{11}(\boldsymbol{k}|\boldsymbol{k}_0) = n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega) + \int\frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^3}h(\boldsymbol{k}-\boldsymbol{k}_1)[n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)] \cdot\boldsymbol{G}_1^\infty(\boldsymbol{k}_1)\cdot\boldsymbol{C}_{\mathrm{o}}^{11}(\boldsymbol{k}_1|\boldsymbol{k}_0). \quad (116)$$

Hence, we obtain the expression:

$$\boldsymbol{C}_{\mathrm{o}}^{11}(\boldsymbol{k}|\boldsymbol{k}_0) = \left[\boldsymbol{I} - \int\frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^3}h(\boldsymbol{k}-\boldsymbol{k}_1)[n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)]\cdot\boldsymbol{G}_1^\infty(\boldsymbol{k}_1)\right] \cdot[n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)]. \quad (117)$$

Using the classical relationship between the convolution product of the two functions and the product of their Fourier transform, we write

$$\int\frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^3}h(\boldsymbol{k}_1-\boldsymbol{k}_0)\boldsymbol{G}_1^\infty(\boldsymbol{k}_1) = \int\mathrm{d}^3\boldsymbol{r}\exp(-\mathrm{i}\boldsymbol{k}_0\boldsymbol{r})h(\boldsymbol{r})\boldsymbol{G}_1^\infty(\boldsymbol{r}). \quad (118)$$

Substituting the expansion $\boldsymbol{G}_1^\infty(\boldsymbol{k}_1)$ into equation (118) and taking into account the fact that the range of the correlation function for Rayleigh scatterers is very small:

$$h(\boldsymbol{r}_j - \boldsymbol{r}_l) \approx \delta(\boldsymbol{r}_j - \boldsymbol{r}_l)\int\mathrm{d}^3\boldsymbol{r}\,h(\boldsymbol{r}), \quad (119)$$

we obtain:

$$\int\frac{\mathrm{d}^3\boldsymbol{k}_1}{(2\pi)^3}h(\boldsymbol{k}_1-\boldsymbol{k}_0)\boldsymbol{G}_1^\infty(\boldsymbol{k}_1) = -\frac{h(0)}{3K_{\mathrm{e}}^2} + \text{P.V.}\{\boldsymbol{G}_1^\infty(\boldsymbol{r}=0)\}\int\mathrm{d}^3\boldsymbol{r}\,h(\boldsymbol{r}). \quad (120)$$

Using approximation (111) with $\Lambda_{\mathrm{T}} = 0$ in (120), according to (117), we obtain

$$\boldsymbol{C}_{\mathrm{o}}^{11}(\boldsymbol{k}|\boldsymbol{k}_0) = [n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)]\boldsymbol{I} \times\left\{1 - \left(-\frac{h(0)}{3K_{\mathrm{e}}^2}[n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)] + \frac{\mathrm{i}K_{\mathrm{e}}}{6\pi}\left[\int\mathrm{d}^3\boldsymbol{r}\,h_a(\boldsymbol{r})n_a\boldsymbol{t}_a^1(\omega) + \int\mathrm{d}^3\boldsymbol{r}\,h_b(\boldsymbol{r})n_b\boldsymbol{t}_b^1(\omega)\right]\right)\right\}^{-1}. \quad (121)$$

By using the Percus–Yevick pair distribution, we can calculate the integral of the function $h(\boldsymbol{r})$:

$$n_i\int\mathrm{d}^3\boldsymbol{r}\,h_i(\boldsymbol{r}) = -1 + w_i, \quad (122)$$

where

$$w_i = \frac{(1-f_{\mathrm{vol}}^i)^4}{(1+2f_{\mathrm{vol}}^i)^2} \quad (123)$$

n_i is the number of the particles of type i, V_i is their volume and $f_{\mathrm{vol}}^i = n_iV_i$ is the their volume fraction.

Using the definition $h(r) = g(r) - 1$, we can deduce that $h(0) = -1$, because $g(r)$ is the correlation function of two scatterers. Indeed, $g(0) = 0$ because two different scatterers cannot be located at the same point. Taking into account the previous results, the expression for $\boldsymbol{t}_i^1(\omega)$ is given by ($i = a$ or b for the two type of scatterers):

$$\boldsymbol{t}_i^1(\omega) = 3K_{\mathrm{vac}}^2\varepsilon_{\mathrm{e}}V_i(\varepsilon_i - \varepsilon_1) \times\left\{(\varepsilon_i - \varepsilon_1)\left[1 - \frac{\mathrm{i}3K_{\mathrm{e}}K_{\mathrm{vac}}^2}{6\pi}\varepsilon_{\mathrm{e}}V_i\right] + 3\varepsilon_{\mathrm{e}}\right\}^{-1}, \quad (124)$$

where $K_{\mathrm{e}}^2 = \varepsilon_{\mathrm{e}}K_{\mathrm{vac}}^2$.

4.4. Equation for the effective permittivity

According to equations (121) and (122), equation (79) of the effective permittivity can be rewritten in the form

$$\varepsilon_{\mathrm{e}} = \varepsilon_1 + \frac{1}{K_{\mathrm{vac}}^2}[n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)] \times\left\{1 - \left(\frac{1}{3K_{\mathrm{e}}^2}[n_a\boldsymbol{t}_a^1(\omega) + n_b\boldsymbol{t}_b^1(\omega)]\right.\right.$$

$$+ \frac{\mathrm{i}K_\mathrm{e}}{6\pi}[(w_a - 1)\boldsymbol{t}_a^1(\omega) + (w_b - 1)\boldsymbol{t}_b^1(\omega)]\Bigg)\Bigg\}^{-1}. \tag{125}$$

If we put $a = b$ and $n_a + n_b = n$, we obtain the equation for identical particles. Equation (125) is a nonlinear equation relating the effective permittivity ε_e to the permittivities of the layer ε_1 and the scatterers ε_a and ε_b. This equation can be generalised to N types of Rayleigh scatterers.

5. Applications and numerical estimates of the effective permittivity for a random medium with nanoparticles

In this section, we consider different examples of media consisting of statistical ensembles of different scattering species and artificial material structures developed on the basis of dielectric or metallic nanoparticles. The incident laser wavelength is $\lambda = 800$ nm. Let us now examine the procedure to solve the nonlinear equation (125) for ε_e, which gives a new formulation of the effective permittivity for two types of particles. By writing equation (125) for one type of scatterers, we obtain the expression, which is a generalised Maxwell–Garnett formula:

$$\varepsilon_\mathrm{e} = \varepsilon_1 + \frac{3(\varepsilon_\mathrm{d} - \varepsilon_1)\varepsilon_\mathrm{e} f_\mathrm{vol}}{(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol} - \frac{2}{3}\mathrm{i}(K_\mathrm{vac} r_\mathrm{d})^3 \varepsilon_\mathrm{e}^{3/2} w) + 3\varepsilon_\mathrm{e}}. \tag{126}$$

Here, w is the Percus–Yevick pair distribution function. If we expand the denominator in (126) to the first order, we obtain

$$\varepsilon_\mathrm{e} = \varepsilon_1 + \frac{\varepsilon_1(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}) + 3\varepsilon_1\varepsilon_\mathrm{e} + 3(\varepsilon_\mathrm{d} - \varepsilon_1)\varepsilon_\mathrm{e} f_\mathrm{vol}}{(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}) + 3\varepsilon_\mathrm{e}}$$

$$+ 2\mathrm{i}\frac{(K_\mathrm{vac} r_\mathrm{d})^3(\varepsilon_\mathrm{d} - \varepsilon_1)^2 w f_\mathrm{vol} \varepsilon_\mathrm{e}^{5/2}}{[(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}) + 3\varepsilon_\mathrm{e}]^2}. \tag{127}$$

Equation (127) is the usual low-frequency limit of the QC-CPA approach obtained by Tsang et al. [13, 16]. Note that in the static case, the imaginary part in equation (127) is equal to zero, and if we replace the effective permittivity ε_e by ε_1 in the right-hand side of the equation, we recover the classical Maxwell–Garnett formula. Consider now an approximate solution to (127) for ε_1 and ε_d taking real values. If we assume that the real part of ε_e is larger than its imaginary part, we obtain an approximate solution for the real part of (127):

$$\mathrm{Re}\varepsilon_\mathrm{e} = {}^1\!/_6[-\{(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}) - 3\varepsilon_1 - 3(\varepsilon_\mathrm{d} - \varepsilon_1)f_\mathrm{vol}\} + \Delta^{1/2}], \tag{128}$$

where

$$\Delta = [(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}) - 3\varepsilon_1 + 3(\varepsilon_\mathrm{d} - \varepsilon_1)f_\mathrm{vol}]^2$$

$$+ 12\varepsilon_1(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}).$$

Substituting this solution of $\mathrm{Re}\varepsilon_\mathrm{e}$ into the third term of (127), we obtain an approximate value for $\mathrm{Im}\varepsilon_\mathrm{e}$:

$$\mathrm{Im}\,\varepsilon_\mathrm{e} = 2\frac{(K_\mathrm{vac} r_\mathrm{d})^3(\varepsilon_\mathrm{d} - \varepsilon_1)^2 w f_\mathrm{vol} \mathrm{Re}\,\varepsilon_\mathrm{e}^{5/2}}{[(\varepsilon_\mathrm{d} - \varepsilon_1)(1 - f_\mathrm{vol}) + 3\,\mathrm{Re}\,\varepsilon_\mathrm{e}]^2}. \tag{129}$$

We have derived these formulas to give an explicit approximate expression for the effective permittivity in the case of real permittivities for the layer and the scatterers. The general numerical process of solution of (125) consists in the following:

(i) we express equation (125), which is a function of ε_e in a complex form (ε_1 and ε_d are real or complex numbers);

(ii) we assume that the real part of ε_e is larger than its imaginary part and numerically seek a solution to $\mathrm{Re}\varepsilon_\mathrm{e}$;

(iii) we numerically solve the equation for $\mathrm{Im}\varepsilon_\mathrm{e}$ by using the previous value of $\mathrm{Re}\varepsilon_\mathrm{e}$;

(iv) these two solutions are the initial values of the iterative procedure to solve (125) and we fix numerical convergence criteria to obtain a solution to the nonlinear equation.

With this procedure, we can derive a numerical tractable solution for the effective permittivity in the case of the QC-CPA approach.

Table 1 presents some numerical results for the solution to the nonlinear equation (125). We can compare the results for one and two types of scatterers at different permittivities of the layer. Note that an increase in the volume fraction $f_{\mathrm{vol}\,1}$ by a factor 100 results in an increase in three orders of magnitude of the imaginary part of the permittivity (line 3), and the real part is also affected by the multiple scattering. Adding an imaginary part to the permittivity ε_d1 has the effect of increasing $\mathrm{Im}\,\varepsilon_\mathrm{e}$ by three orders of magnitude. Nanoscatterers introduce an imagery part in the permittivity, which can be significant (especially for metallic nanoparticles, line 5). The presence of second type scatterers in the layer with different permittivities or radii is shown in lines 8–12. Let us analyse the influence of the nanoparticles on the effective permittivity. One of the principal effects of dielectric nanoparticles with a low concentration is the introduction of an imaginary part in the permittivity. The results of this analysis show that we must take into account the scattering by nanoparticles at a

Table 1. Effective permittivity ε_e for scatterers of type 1 ($r_\mathrm{d1}, f_{\mathrm{vol}\,1}, \varepsilon_\mathrm{d1}$) and type 2 ($r_\mathrm{d2}, f_\mathrm{vol2}, \varepsilon_\mathrm{d2}$) and the permittivity ε_1 of the layer.

№	r_d1	$f_{\mathrm{vol}\,1}$	ε_d1	r_d2	f_vol2	ε_d2	ε_1	ε_e
1	0.035λ	1×10^{-4}	2.0	–	–	–	1.2	$1.2+\mathrm{i}\,1.1\times10^{-8}$
2	0.070λ	1×10^{-4}	2.0	–	–	–	1.2	$1.2+\mathrm{i}\,8.87\times10^{-7}$
3	0.035λ	1×10^{-2}	2.0	–	–	–	1.2	$1.207+\mathrm{i}\,1.03\times10^{-5}$
4	0.035λ	1×10^{-4}	5.0	–	–	–	1.2	$1.2+\mathrm{i}\,8.84\times10^{-7}$
5	0.035λ	1×10^{-4}	$\mathrm{i}\infty$	–	–	–	2.0	$2.0+\mathrm{i}\,1.2\times10^{-5}$
6	0.035λ	1×10^{-4}	$5.0+\mathrm{i}0.5$	–	–	–	2.0	$2.0+\mathrm{i}\,2.35\times10^{-5}$
7	0.035λ	1×10^{-4}	5.0	–	–	–	$2.0+\mathrm{i}0.5$	$2.0+\mathrm{i}\,0.49$
8	0.035λ	5×10^{-5}	2.0	0.070λ	5×10^{-5}	2.0	1.2	$1.2+\mathrm{i}\,4.98\times10^{-7}$
9	0.035λ	5×10^{-5}	2.0	0.035λ	5×10^{-5}	5.0	1.2	$1.2+\mathrm{i}\,4.97\times10^{-7}$
10	0.035λ	5×10^{-5}	2.0	0.035λ	5×10^{-5}	$5.0+\mathrm{i}0.5$	2.0	$2.0+\mathrm{i}\,1.17\times10^{-5}$
11	0.035λ	9×10^{-3}	2.0	0.035λ	1×10^{-3}	5.0	1.2	$1.2+\mathrm{i}\,1.79\times10^{-5}$
12	0.035λ	5×10^{-5}	2.0	0.035λ	5×10^{-5}	$5.0+\mathrm{i}0.5$	$2.0+\mathrm{i}\,5\times10^{-5}$	$2.0+\mathrm{i}\,6.17\times10^{-5}$

low concentration in a medium especially when we design optical components, which can transmit or scatter the optical field with specified angular, spatial or spectral properties.

6. Conclusions

The theory of the effective permittivity has been extended to random media (bounded by rough surfaces) with different types of particles. The expression of the scattered coherent field can be obtained using the transition operators defined in this paper. We have derived a new formula for the effective permittivity, which characterises the coherent part of an electromagnetic wave propagating in a random medium. The starting point of our theory has been the quasi-crystalline coherent potential approximation which takes into account the correlation between the particles. Our formulation contains the corrections to the effective permittivity due to the randomly rough surfaces. We express these corrections by Green's functions of the rough surface scattering. The accuracy of the effective permittivity is greatly improved under the CPA-QCA approach since an approximate formula can be derived from the multiple scattering theory, which is a generalisation of the conventional Maxwell–Garnett formula. Numerical calculations of the effective permittivity under the QC-CPA approach can be performed for a thin layer. One can also obtain an approximate formula for the effective permittivity, which at the same time contains the Maxwell–Garnett formula and the Keller approximation [23]. The Keller formula can be obtained in considering the QC-CPA approach in the scalar case [13, 16]. The equations are identical to the equations obtained previously under conditions that the dyadic Green's function is replaced by the scalar Green's function.

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