Electromagnetic Coherent Effects in Metamaterials with Randomly Rough Surfaces

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Abstract.
In this paper, we present a theory of transport based on the Bethe-Salpeter equation for a three-dimensional disordered medium with randomly rough interfaces illuminated by an electromagnetic wave. The slab is defined by a random medium composed of nanospheres randomly distributed in a layer. We study the electromagnetic coherent effects of the wave scattering inside the random volume bounded by randomly rough interfaces. Coherent effects are related for example to backscattering enhancement due to the interaction of the wave with the randomly rough boundaries or the random volume. The phenomenon manifests itself at the level of configurationally averaged mean intensity. We examine the solution by moments of the Bethe-Salpeter equation and we introduce the ladder and most-crossed contributions of the transport theory including the randomly rough boundaries of the considered structure. In this paper we are concerned with the wave transport behavior beyond the effective medium picture in taking into account the multiply scattered behavior.

1. Introduction
Metamaterials are composite structures made of small metallic or dielectric particles. These particles play the same role as atoms or molecules in ordinary materials, which makes metamaterials considered as "artificial materials". In this paper, we consider a three-dimensional disordered medium with randomly rough interfaces. For classical waves, the most interesting case is that of three-dimensional disorder. The quantity that obeys the disordered wave equation - the electric field - is a vector and not a scalar as in the Schrödinger equation. If many theoretical and numerical approaches have been developed to model transport of scalar light in random media, few or even no method at all nowadays can deal with ensembles of complex different nanoparticles in interaction between themselves and randomly rough boundaries. This aspect of the problem was never considered in detail up to now. In mesoscopic physics, multiple light scattering in disordered potentials gives rise to complex wave interference phenomena, like speckle correlations, backscattered enhancement or Anderson localization of light [1], and to applications like imaging through opaque biological tissues or photovoltaics. We present a theory of transport based on an equation for second statistical moment, i.e. the Bethe-Salpeter equation [1-6], in which the vectorial character of electromagnetic wave, the correlations between the scatterers are taken into account. We can derive the radiative transfer equation from the Maxwell equations. The problem is then reduced to find a good approximation to the solution of this equation. The Bethe-Salpeter equation is constructed in order that the medium and the boundaries are treated on the same footing [4-6]. This unified Bethe-Salpeter equation (for
details see [4,6]) enables us to obtain a general expression, whatever the choice of the scattering operators used at the boundaries. In [7-10] we describe the application of the Small Perturbation Theory and Small Approximation Theory to the scattering of electromagnetic waves from two-dimensional randomly rough interfaces and three-dimensional film bounded by random surfaces, multiple-scattering effects on the randomly rough surfaces are incorporated in these theories, this can lead to describe new coherent phenomena due to the coupling on and between the random surfaces [11]. The Quasi-Crystalline Coherent Potential Approximation (QC-CPA) [2,4,12,13] is taken into account for the contribution of the random medium, which is made of spherical nanoparticles of given permittivity in a homogeneous background medium. The boundaries are described by random functions. Mathematically, the way to study the solution of the generalized Bethe-Salpeter equation is based on the use of diagrammatic perturbation theory [1-4]. The physical process of the backscattering enhancement is obtained by adding the contribution of the so-called most-crossed diagrams [4] to the diagrammatic expansions of the transfer equation. This paper deals with the coherent effects, as the backscattered enhancement called the weak localization effect [1], which occurs when the electromagnetic intensity is scattered back to the source, it is a coherent effect experienced by waves when propagating through a disordered system despite the randomness of the system. For three-dimensional slabs with nanoscale roughness and nanoparticles, the enhanced backscattering [14] is produced by different mechanisms, wave scattering by the same boundary, wave coupling by the two boundaries or wave scattering by the randomly distributed nanoparticles. Quantum structures as metallic or dielectric nanoparticle with a size of a few nanometers exhibit behaviors different than that of the bulk [15-19] due to confinement effects which change the electronic properties. Therefore the treatment of the permittivity of such nanoparticles can not be considered without taking into account quantum effects related to the behavior of electrons in the atom cloud. We use a calculation method in which conduction electrons are modeled as a gas of free electrons. In the first part of this paper we introduce the theory and the calculation of the total specific intensity scattered by the slab illuminated by a polarized electromagnetic plane wave. In the second part of the paper, we give some numerical examples of scattering properties of random films with discrete nanoparticles. These examples of simulation of light interaction with random structures are devoted to the study of the introduction of the multiple scattering to the scattering from random media with random boundaries.

2. Incoherent specific intensity
We consider harmonic waves with $e^{-i\omega t}$ dependence. We are going to study a three-dimensional slab (see Fig.1), which is composed of an incident medium of permittivity $\epsilon_0$, a first randomly rough boundary, a random medium defined by a permittivity $\epsilon_1$ which contains scatterers, a second randomly rough boundary at the distance $z = -H$ and a semi-infinite homogeneous medium with a permittivity $\epsilon_2$. The main characteristic we must study for the scattering from a random slab is the specific intensity $\mathcal{I}(\mathbf{R}, \mathbf{k})$. If we take into account the polarization of the wave, the specific intensity is defined as a Stokes vector or a tensor which gives the power flux per unit area and solid angle at the point $\mathbf{R}$ in the direction $\mathbf{k}$. Writing an energy balance equation, it can be shown shown that the specific intensity satisfies a Boltzmann type equation called radiative transfer equation. If particles are inside a slab with a permittivity different from the outside medium, boundaries condition must be added to the radiative transfer equation in order to calculate the specific intensity. For rough surfaces, these boundaries condition are expressed in function of scattering operators, where several approximate analytical expressions exist depending on the roughness of the surface [7-10]. The specific intensity can be deduced from the Wigner function of the electrical field [4,6]. After having determined the relation between the electromagnetic field and the specific intensity, we can derive the radiative transfer equation from the Maxwell equations.
The procedure is to write the Maxwell equations in an integral form with the help of Green functions and to apply the Wigner transform to the equation obtained. In our approach, we use scattering operators which are a unified way to describe how waves interact with the boundaries.

To use these operators, we have introduced two kinds of Green functions \[4\]. The first one describes the field scattered by the volume (V), which contains the scatterers, and by the rough surfaces (S). The second type of Green function is \(\mathcal{G}_S\), which gives the field scattered by a slab with rough boundaries where the scatterers have been replaced by an homogeneous medium described by an effective permittivity. With these Green functions, we can separate the contributions of the surfaces and the volume. The Quasi-Crystalline Coherent Potential Approximation (QC-CPA) \[13\] is taken into account for the contribution of the random medium, which is made of spherical nanoparticles of given permittivity in a homogeneous dielectric background medium. The main advantage of our approach is that the equation obtained are similar to the equations generally used to describe the wave scattered by an infinite random medium. For a random layer with rough interfaces, the incoherent specific intensity can be decomposed into four parts \[4,6\]:

\[
\mathcal{T}^{\text{incoh}} = \mathcal{T}^{\text{incoh}}_{L=0} + \mathcal{T}^{\text{incoh}}_{L=1} + \mathcal{T}^{\text{Ladder}} + \mathcal{T}^{\text{Crossed}},
\]

where the contributions are given by:

\[
\mathcal{T}^{\text{incoh}}_{L=0}(\mathbf{R}, \mathbf{k}) = c_0 \int_{V_0} d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} \left[ < E_{S}^{0s}(\mathbf{r} + \frac{\mathbf{r}}{2}) \otimes E_{S}^{0s}(\mathbf{r} - \frac{\mathbf{r}}{2}) >_S - < E_{S}^{0s}(\mathbf{r} + \frac{\mathbf{r}}{2}) > \otimes < E_{S}^{0s}(\mathbf{r} - \frac{\mathbf{r}}{2}) >_S \right],
\]

\[
\mathcal{T}^{\text{incoh}}_{L=1}(\mathbf{R}, \mathbf{k}) = c_0 \int_{V_0} d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} \mathcal{G}_{<S>}^{01} : \mathcal{P}^{11} : < E^{1t} \otimes E^{1t} >_S (\mathbf{R} + \frac{\mathbf{r}}{2}, \mathbf{R} - \frac{\mathbf{r}}{2}),
\]

\[
\mathcal{T}^{\text{Ladder}}(\mathbf{R}, \mathbf{k}) = c_0 \int_{V_0} d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} \mathcal{G}_{<S>}^{01} : \mathcal{P}^{11} : \mathcal{G}^{11}_{<SV>} : \mathcal{P}^{11} : < E^{1t} \otimes E^{1t} >_S (\mathbf{R} + \frac{\mathbf{r}}{2}, \mathbf{R} - \frac{\mathbf{r}}{2}),
\]

\[
\mathcal{T}^{\text{Crossed}}(\mathbf{R}, \mathbf{k}) = c_0 \int_{V_0} d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} \left[ \mathcal{G}_{<S>}^{11} : \mathcal{P}^{11} : \mathcal{G}_{<SV>}^{11} : \mathcal{P}^{11} : < E^{1t} \otimes E^{1t} >_S \right] \mathcal{T}_R (\mathbf{R} + \frac{\mathbf{r}}{2}, \mathbf{R} - \frac{\mathbf{r}}{2}),
\]
with \( \epsilon_0 = \epsilon_{\text{vac}} c_{\text{vac}} n_0 / 2 \). \( \epsilon_{\text{vac}} \) and \( c_{\text{vac}} \) are respectively the permittivity and the speed of light in vacuum. \( n_0 \) is the optical index of the medium 0. \( T_k \) defines the right transpose of a tensor. The superscripts of the operators and the Green functions correspond respectively to the receiver location and the source location (0 for the medium 0, 1 for the medium 1, 2 for the medium 2). The lower index \( \ll SV \gg \) represents the average over the randomly rough surfaces and the disordered medium. The lower index \( < S > \) represents the average over the randomly rough surfaces. We assume the ergodicity of the system and we look for a configurational average: i.e., the infinite-time average of the energy scattered by the slab is equivalent to its spatial average over many configurations. The fields \( \mathbf{E}^{\text{0th}}_S (r) \), \( \mathbf{E}^{\text{1st}}_S (r) \) represent respectively the field scattered by the slab with or without any interaction with the scatterers and the field transmitted in the layer before an interaction with the scatterers. The first term \( \mathcal{I}^{\text{coh}}_{L=0} \) gives the scattering by the slab with rough boundaries and defined by an effective permittivity \( \epsilon_e \) given by the QC-CPA approximation [13], we will give an overview of the formulation in the second part of the paper. In order to take into account accounting effects, quantum multiple scattering theory has been transposed in the electromagnetic case, but as a rigorous analytical answer is unreachable, several approximation schemes have been developed. One of the most advanced is the Quasicrystalline Coherent Potential Approximation (QC-CPA) which takes into account the correlation between the particles to determine \( \epsilon_e \), we use the fact that the effective medium is not spatially dispersive, it can be shown, using a diagrammatic technique, that the coherent part of the field \( < \mathbf{E} >_V \) which propagates inside an infinite random medium behaves as a wave in an homogeneous medium with a renormalized effective permittivity. The Quasi-Crystalline Approximation (QCA) is strictly valid when the particles have a fixed position, as in a crystal. The quasi-crystalline approximation is equivalent to neglect the fluctuation of the effective field on a particle located at \( r_j \), due to a position deviation of a particle located at \( r_i \) from its average position. Under the (QC-CPA) approach, for Rayleigh scatterers, our approximate formula for \( \epsilon_e \) [13] can generalize the usual Maxwell-Garnett formula and the Keller approximation. The second term \( \mathcal{I}^{\text{coh}}_{L=1} \) is related to the approximation where only one process of scattering by a particle is taken into account. The two last terms \( \mathcal{I}^{\text{incoh}}_{\text{Ladder}} \) and \( \mathcal{I}^{\text{incoh}}_{\text{Crossed}} \) correspond respectively to the ladder approximation and the most-crossed approximation (see [4,6] for the detailed expressions). Fig. 2 gives the scattering processes for the ladder approximation. The most-crossed approximation gives the physical phenomenon producing a peak in the backscattering direction due to the interference of waves following the same path but in opposite directions. The most elegant way to introduce the enhanced backscattering in the theory is to use the reciprocity principle. Heuristically, it means that when we exchange the source and the detector position and also their polarization, the field measured is the same. In the previous formulas, we can follow from the right to the left, how the scattering process is defined by the combinations of the different operators. The introduction of tensor products of matrices is related to the fact we calculate an incoherent cross-section, which depends on the square modulus of a scattering amplitude. The tensor \( \mathcal{P}^{11} \) describes the intensity scattered by the first particle the wave encounters. The expression of this tensor is given by the the Quasi-Crystalline Coherent Potential Approximation. This intensity operator is determined by using the energy conservation principle and takes into account the correlations with the other particles. Under the QC-CPA assumption, the effective permittivity satisfies a non-linear system of equations. Introducing these equation in a Ward identity, we obtain an expression, called the modified ladder approximation for the intensity operator \( \mathcal{G}^{11}_{\ll SV \gg} \) which satisfies the energy conservation [4,6]. The operator \( \mathcal{G}^{11}_{\ll SV \gg} \) is the Green tensor which contains the interaction between the scatterers and the surfaces. As we have seen, the contributions for the most-crossed and the ladder diagrams are determined by the Green tensor \( \mathcal{G}^{11}_{\ll SV \gg} \), which satisfies a Bethe-Salpeter equation. It contains the second order and the higher orders describing the scattering by particles. To calculate the previous expressions, we
need to estimate the tensor $\mathcal{G}_{SV}^{11}$, as no exact solution exists for the expression of the Green tensor, we rely on a perturbative method. In this paper, we take into account the second-order approximation. The very first results for this second-order approximation was given in [4,14].

The Green tensor $\mathcal{G}_{SV}^{11}$ is given by the Bethe-Salpeter equation as follows:

$$\mathcal{G}_{SV}^{11} = \mathcal{G}_{SV}^{11} + \mathcal{G}_{SV}^{11} : \mathcal{P}^{11} : \mathcal{G}_{SV}^{11}, \quad (6)$$

where the previous tensors are defined by the following tensorial products of the Green functions:

$$\mathcal{G}_{SV}^{11} = \langle \mathcal{G}_{SV}^{11} \otimes \mathcal{G}_{SV}^{11} \rangle_{SV}, \quad (7)$$

$$\mathcal{G}_{SV}^{11} = \langle \mathcal{G}_{S}^{11} \otimes \mathcal{G}_{S}^{11} \rangle_{S}, \quad (8)$$

By iterating the Bethe-Salpeter equation (6) we obtain the following perturbative development:

$$\mathcal{G}_{SV}^{11} = \mathcal{G}_{SV}^{11} + \mathcal{G}_{SV}^{11} : \mathcal{P}^{11} : \mathcal{G}_{SV}^{11} + \mathcal{G}_{SV}^{11} : \mathcal{P}^{11} : \mathcal{G}_{SV}^{11} + \cdots. \quad (9)$$

The first-order Green tensor expansion represents the double-scattering theory for the cross-section while the second-order corresponds to the third-order scattering for the cross-section. This second-order expansion is taken into account in the simulation we present in the next section.

![Figure 2](image-url)

**Figure 2.** Scattering processes for the ladder approximation. The solid and dashed lines represent respectively the propagating wave on the right and left sides of the tensorial products $\mathcal{G}_{SV}^{11} = \langle \mathcal{G}_{SV}^{11} \otimes \mathcal{G}_{SV}^{11} \rangle_{SV}$.

### 3. Scattering from film with discrete nanoparticles

The term $\mathcal{I}_{L=0}^{\text{inc}}$ contains the effective permittivity of the slab bounded by two randomly rough surfaces. In [12,13] we have introduced a formulation for the effective permittivity for different classes of spherical scatterers. In the following we take the example of two classes of scatterers $a$ and $b$. The expression of the effective permittivity is given by:

$$\epsilon_e(\omega) = \epsilon_1 + \frac{1}{K_{\text{vac}}} \frac{n_a \mathcal{T}_a^1(\omega) + n_b \mathcal{T}_b^1(\omega)}{1 - \frac{1}{K_{\text{vac}}} \left( n_a \mathcal{T}_a^1(\omega) + n_b \mathcal{T}_b^1(\omega) \right) + \frac{K_{\text{inc}}}{6\pi} \frac{\left( (w_a - 1) \mathcal{T}_a^1(\omega) + (w_b - 1) \mathcal{T}_b^1(\omega) \right)}}, \quad (10)$$
where the function $\tilde{T}_i^j(\omega)$ may be written in the form: ($i = a$ or $b$)

$$
\tilde{T}_i^j(\omega) = \frac{3 K_2^2 \epsilon_e v_i (\epsilon_i - \epsilon_1)}{(\epsilon_i - \epsilon_1)(1 - \frac{13 K_e K_2^2}{6 \pi} \epsilon_e v_i)} + 3 \epsilon_e.
$$

(11)

and

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

(12)

In these equations, $n_i$ is the number of particles of type $i$, $v_i$ their volume and $f_{vol}^i = n_i v_i$ the volume fraction, $K_2^2 = \epsilon_e K_2^{vac}$. The equation (10) is a non linear equation connecting the effective permittivity $\epsilon_e$ and the permittivities $\epsilon_1$ of the layer, and $\epsilon_a, \epsilon_b$ of the scatterers. Its derivation is obtained with the assumptions (QC-CPA) and gives a tractable formula for the effective permittivity. The permittivity of a nanoparticle is given by a quantum expression where conduction electrons are modeled as a gas of free electrons. The relation between the specific intensity and the incoherent cross-section is given by a product of two tensors:

$$
4 \pi \cos \theta \mathbf{T}^{\text{incoh}}(R, k) = \frac{\gamma^{\text{incoh}}(\hat{k}|\hat{k}_0)}{\mathbf{J}^0_i(\hat{k}_0)} : \mathbf{J}^{0_i}(\hat{k}_0),
$$

(13)

where

$$
\mathbf{J}^{0_i}(\hat{k}_0) = \epsilon_0 \mathbf{E}^{0_i}(\hat{k}_0) \otimes \mathbf{E}^{0_i*}(\hat{k}_0).
$$

(14)
Eq.14 is a tensorial product of two vectors. We give two examples of calculation of the scattering from two thin films. The wavelength is given by $\lambda = 0.8 \mu m$. The four terms of the incoherent cross-section are taken into account in these computations. We consider that the incident medium is vacuum ($\epsilon_0 = 1$). We compute the incoherent cross-section $\gamma^{\text{incoh}}(\theta)$ for the four polarizations in the plane of incidence for a slab, which is defined as follows. The layer depth is $4.5 \mu m$, the optical index of the layer is $n_1 = 1.49$. The upper layer boundary is defined as a slightly randomly rough surface defined by a root mean square height of $200 \text{nm}$ and a correlation length of $462 \text{nm}$, we consider a Gaussian distribution for the distribution of surface heights and for the correlation function. The lower boundary is a flat surface, which separates the layer from a semi-infinite medium of optical index $n_2 = 1.5$. Inside the medium we consider nanoobjects. In the first example, we consider two types of spherical nanoholes with diameters of 10 and $100 \text{nm}$. The filling rate is 4% with a volume fraction of $1/2$. The permittivity of the holes is that of the vacuum. In the second example, the medium contains two types of spherical metallic particles defined as Rayleigh spherical scatterers with the same parameters as for medium with nanoholes. We obtain the total incoherent cross-section for an incident angle $\theta = 8^\circ$, for the co and cross-polarization components in the case a medium with nanoholes (Fig.3) and a medium with metallic nanospheres (Fig.4). Despite the size of the metallic particles and the filling ratio, their influence on the scattering process is important as demonstrated by the patterns of the two curves.

**Figure 4.** Incoherent cross-section in function of scattering angle, medium with metallic nanoparticles.
4. Conclusion
This paper was devoted to a theory of transport based on a Bethe-Salpeter equation. We presented a multiple scattering theory for a random slab with discrete nanoscale scatterers and bounded with randomly rough surfaces. By starting from the Maxwell equations, we are able to give an unambiguous definition of the specific intensity as a function of the electric field. Furthermore, we can take into account the correlations between the scatterers. We use a new theory of effective permittivity, which has been extended to random media with different types of particles and bounded with randomly rough surfaces. Finally, we have also incorporated the most-crossed contributions in our theory, with which we can estimate the enhanced backscattering phenomenon and a quantum expression of the permittivity of the metallic nanosphere. We gave two example of scattering calculation for spherical nano-holes and metallic nanospheres.

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