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Energy-density CPA: a new effective medium theory for classical waves

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Abstract

We present the framework of an effective medium theory to calculate the transport properties of classical waves in disordered media, Busch and Soukoulis (Phys. Rev. Lett. 75 (1995) 3442). It is based on the principle that the wave energy density should be uniform when averaged over length scales larger than the size of the basic scattering unit and can, therefore, be applied to electromagnetic, Busch and Soukoulis (Phys. Rev. B 54 (1996) 893); Kirchner et al. (Phys. Rev. B 57 (1998) 277) as well as elastic waves, Kafesaki and Economou (Europhys. Lett. 37 (1997) 7); Soukoulis et al. (Phys. Rev. Lett. 82 (1999) 2000). Within this energy-density CPA (ECPA) resonant scattering of the individual scatterer is treated exactly, and by using a coated sphere as the basic scattering unit, multiple scattering contributions are incorporated in a mean-field sense. In the long-wavelength limit we are able to calculate effective material properties exactly. Results for the mean-free path, transport velocity, and the diffusion coefficient for finite frequencies agree qualitatively and quantitatively with experiment for all densities of scatterers, Busch and Soukoulis (Phys. Rev. B 54 (1996) 893); Gomez et al. (Europhys. Lett. 48 (1999) 22). A study of the localization parameter $\bar{k}\ell_s$ within this effective-medium approach allows to identify the optimal parameters for localization, Kirchner et al. (Phys. Rev. B 57 (1998) 277). © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

In recent years there has been a growing interest in the study of the propagation of classical waves in random media [8] that was largely driven by the prospect of observing Anderson localization [7] of light in those systems. As a consequence, fundamental aspects of our understanding of multiple scattering of waves such as the scaling theory of localization could be addressed both experimentally and theoretically in a clean system.

Early theoretical work [9–11] indicated the existence of Anderson localization of classical waves for an intermediate frequency range. In this so-called Mie scattering regime, the wavelength λ of the wave is comparable to the scatterers extent d, leading to large scattering cross sections of the

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individual scatterer. However, soon after experimental investigations along these lines [12,13] had reported very low values of the diffusion coefficient D, it became clear that, unlike electronic systems, there exists another renormalization mechanism to the diffusion coefficient D for classical waves [14]. The presence of resonant scatterers may cause the energy transport velocity v_E to decrease sharply close to the single-scatterer resonances. This renormalized transport velocity enters the three-dimensional diffusion coefficient via $D = v_E \ell_t /3$, where ℓ_t is the transport mean-free path. The renormalization of v_E and thus of D can be regarded as a scattering delay due to temporal storage of wave energy inside the scatteres. However, the onset of Anderson localization manifests itself in low values of the transport mean-free path ℓ_t, and, therefore, considerable care had and has to be exercised when interpreting low values of the diffusion coefficient D for classical wave systems and a reliable theory for calculating transport quantities even "just" for the diffusive regime in the presence of resonant scattering is called for.

In the first part of this contribution we outline a very successful effective medium theory within the framework of which transport properties such as diffusion coefficient D, energy transport velocity v_E and transport mean-free path ℓ_s for diffusing classical waves can be calculated. The main results and predictions such as a study of the localization parameter $\bar{k}\ell_s$, $\bar{k}$ being the renormalized wavevector and ℓ_s the scattering mean-free path are summarized in the second part. At this point, we want to stress that this effective medium theory can be applied to a great variety of problems in the field of classical wave propagation.

2. The new effective medium theory

Consider a composite medium consisting of randomly placed lossless spheres with diameter $d = 2R$ and dielectric constant ε_1 embedded within a lossless host material with dielectric constant ε_2. The random medium is characterized also by f, the volume fraction occupied by the spheres. The basic idea of any effective medium theory of disordered systems is to focus on one particular scatterer and to replace the surrounding random medium by an effective homogeneous medium. The effective medium is determined self-consistently by taking into account the fact that any other scatterer could have been chosen. This procedure manifests the homogeneity of the random medium on average.

However, the position of a spheres in the medium is completely random, with the exception that they cannot overlap. This implies that the distribution $P(R)$ of spacings between neighboring spheres is sharply peaked at a distance $R_c > R$. If we approximate this distribution by a δ-function, i.e., $P(R) \propto \delta(R_c - R)$ and take into account the on-average isotropy of the random medium, we may consider a coated sphere as the basic scattering unit. The radius R_c of the coated sphere is $R_c = R/f^{1/3}$. The dielectric constants of the core and the coating are ε_1 and ε_2, respectively. Using a coated sphere as the basic scattering unit also incorporates some of the multiple scattering effects at different centers.

The use of a coated sphere as the basic scattering unit also implies that the homogeneity of the energy density is not anymore fulfilled. This fact has not been taken into account in approaches that exploit the analogy between classical and electronic wave propagation [15–17] and, as a consequence, lead to unphysical results for the transport velocity. Therefore, in the new effective medium theory [1–3] we explicitly chose the averaged energy density homogeneity as the criterion for determining the effective medium. Since we are exclusively considering lossless dielectrics the effective medium dielectric constant $\bar{\varepsilon}$ has to be real due to energy conservation. This is in contrast to the conventional approaches [15–17] and forces us to proceed in two steps: Firstly, we determine for every frequency ω the real effective dielectric constant $\bar{\varepsilon}$ by demanding the energy density to be homogeneous on scales larger than the basic scattering unit (coated sphere). Then, in a second step, the physical quantities are calculated from the (now non-vanishing) scattering cross sections. In this theory all multiple scattering effects are contained in the effective dielectric constant and, thus, we may consider the random medium consisting of independent scattering units, i.e., coated spheres, embedded in the effective medium. Fig. 1 schematically depicts

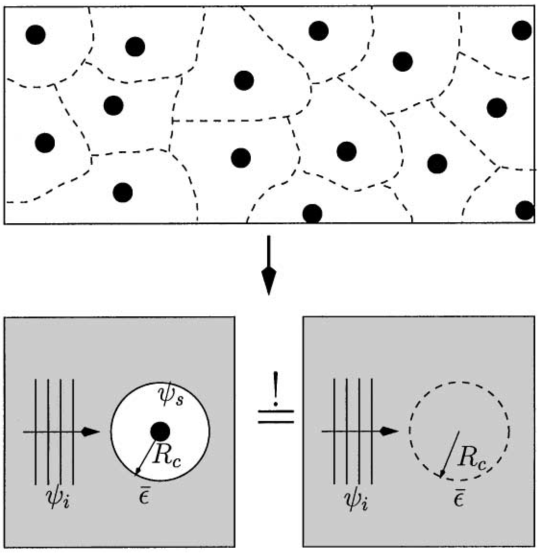

Fig. 1. In a random medium composed of spheres with dielectric constant ε_1 immersed in a host medium with dielectric constant ε_2, the basic scattering unit may be, on average, regarded as a coated sphere, as represented by the dashed lines. To calculate the effective dielectric constant, $\bar{\varepsilon}$, a coated sphere of radius $R_c = R/f^{1/3}$, is embedded in a uniform medium. The self-consistent condition for the determination of $\bar{\varepsilon}$ is that the energy of a coated sphere is equal to the energy of a sphere with radius R_c and dielectric constant $\bar{\varepsilon}$.

the reduction of the disordered medium to a description of independent coated spheres embedded in the effective medium.

The requirement that the energy content of a coated sphere embedded in the effective medium and being hit by a plane wave, should be the same as the energy stored by the plane wave in an equally sized volume of the effective medium, can be formulated quantitatively by the self-consistency equation

$$\int_0^{R_c} d^2r\, \rho_E^{(1)}(\boldsymbol{r}) = \int_0^{R_c} d^2r\, \rho_E^{(2)}(\boldsymbol{r}). \tag{1}$$

$\rho_E^{(1)}(\boldsymbol{r})$ and $\rho_E^{(2)}(\boldsymbol{r})$ are the energy densities for a coated sphere and a plane wave, respectively. Clearly, this very general principle can be applied to any kind of classical wave propagation, such as, e.g., elastic waves [4,5].

In the electromagnetic (EM) wave case, the energy density of EM waves with electric and magnetic fields, $\boldsymbol{E}(\boldsymbol{r})$ and $\boldsymbol{H}(\boldsymbol{r})$, is given by

$$\rho_E(\boldsymbol{r}) = \tfrac{1}{2}(\varepsilon(\boldsymbol{r})|\boldsymbol{E}(\boldsymbol{r})|^2 + \mu|\boldsymbol{H}(\boldsymbol{r})|^2). \tag{2}$$

Here, μ is the magnetic permeability which is taken to be the same in both materials. The specific form of the scattered fields inside the coating and the core are given in Refs. [2,3]. Eq. (1) together with Eq. (2) and the respective expressions for the fields determine the (real) dielectric constant $\bar{\varepsilon}$ of the effective medium for every frequency.

As mentioned above, the transport quantities velocity and the renormalized wave vector $\bar{k}$ can be calculated via [3,8]

$$v_E \simeq \frac{c}{\sqrt{\bar{\varepsilon}}}\sqrt{1 - \mathrm{Re}(\Sigma)/k_m^2}, \tag{3}$$

$$\ell_s = \frac{1}{\sqrt{2}\,\mathrm{Im}(\Sigma)}[(k_m^2 - \mathrm{Re}(\Sigma)) + \sqrt{(k_m^2 - \mathrm{Re}(\Sigma))^2 + (\mathrm{Im}(\Sigma))^2}]^{1/2}, \tag{4}$$

$$\bar{k} = \frac{1}{\sqrt{2}}[(k_m^2 - \mathrm{Re}(\Sigma)) + \sqrt{(k_m^2 - \mathrm{Re}(\Sigma))^2 + (\mathrm{Im}(\Sigma))^2}]^{1/2}. \tag{5}$$

The self-energy Σ is evaluated in the independent scatterer approximation:

$$\Sigma = n\, t_{\boldsymbol{kk}}(\omega), \tag{6}$$

where, $t_{\boldsymbol{kk}'}(\omega)$ denotes the t-matrix of a coated sphere embedded in the effective medium, $|\boldsymbol{k}| = k_m$ and $n = 1/R_c^3$ is the density of scatterers.

In addition, we approximate the transport mean-free path ℓ_t by the scattering mean-free path ℓ_s, i.e., $\ell_t \approx \ell_s$. Then, the three-dimensional diffusion constant D is given by $D = v_E \ell_t/3$. This approximation is supported by the fact that, as a mean-field theory, the ECPA is unable to make detailed predictions close to the Anderson transition where the distinction between scattering and transport mean-free path would become important. In addition, previous studies of the transport and scattering mean-free paths [16] obtained results consistent with this approximation.

3. Results

3.1. Long wavelength limit

In the long wavelength limit, we may define a frequency independent, long-wavelength dielectric constant ε_∞ according to

$$\varepsilon_\infty = \lim_{\omega \to 0} \left(\frac{c}{v_E(\omega)} \right)^2. \tag{7}$$

It is well known [8] that for scalar classical waves the correct result for ε_∞ is given by the volume averaged dielectric constant, whereas in the case of vector classical waves it is Maxwell–Garnett theory which gives the right answer. An analytical calculation [2,3] of ε_∞ within the new effective medium theory, according to Eq. (7), proceeds straightforwardly by computing $\bar{\varepsilon}$ for $\omega \to 0$ from Eq. (1) using a Taylor expansions of all quantities involved to extract the leading order in ω. Indeed, in the case of scalar classical waves we obtain as the long-wavelength dielectric constant the volume average of ε_1 and ε_2, i.e.,

$$\varepsilon_\infty \equiv \bar{\varepsilon} \equiv f\varepsilon_1 + (1-f)\varepsilon_2. \tag{8}$$

This result originates from the fact that for scalar waves s-wave scattering dominates in the long wavelength limit. In the case of the EM waves, however, s-wave scattering is absent and a careful analysis [2,3] of the dominant p-wave scattering for long wavelengths leads to the Maxwell–Garnett result, i.e.,

$$\varepsilon_\infty = \bar{\varepsilon} = \varepsilon_2 \left(1 + \frac{3f\alpha}{1-f\alpha} \right), \tag{9}$$

where the depolarization factor α of a sphere is given by $\alpha = (\varepsilon_1 - \varepsilon_2)/(\varepsilon_1 + 2\varepsilon_2)$.

3.2. Finite frequencies

For finite frequencies, of course, no analytical solution of Eq. (1) is possible. Fortunately, it turns out that Eq. (1) is numerically easy to deal with.

Fig. 2 shows a comparison between the diffusion coefficient $D = v_E \ell_t/3$ obtained within the effective medium theory and experimental values of Genack et al. [13]. Without adjustable parameters excellent agreement is obtained between experiment and the new effective medium theory. More detailed results on the energy transport velocity and mean-free paths within the ECPA can be found in [1–3,6].

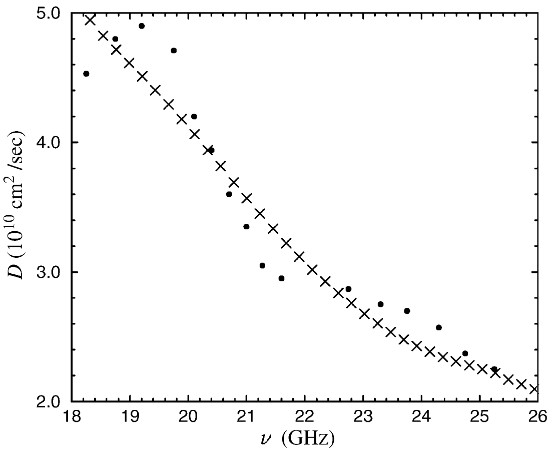

Fig. 2. The frequency dependence of the diffusion constant for a sample of $\frac{1}{2}$-in. polystyrene spheres with index of refraction 1.59 and filling ratio $f = 0.59$. The filled circles correspond to the experimental values, whereas the crosses are the results of the new effective medium theory [2].

3.3. Study of the localization parameter

The product $\bar{k}\ell_s$, where $\bar{k}$ is the renormalized wave vector and ℓ_s the scattering mean-free path, is a measure for the strength of the multiple scattering effects. Here, we wish to recall that within the effective medium theory we have $\ell_t \approx \ell_s$. For values of $\bar{k}\ell_s \simeq 1$ coherent backscattering notably renormalizes the diffusion coefficient and may ultimately lead to a change in wave functions' nature from extended to localized. This phenomenon, commonly referred to as Anderson localization [7], is a generic wave property and to date still represents a challenge to experiments: Recently, infrared localization in GaAs powders was observed [18] but the validity of these measurements has been questioned by the possibility of absorption [19]. In addition, experimental studies on similarly strong scattering Si powders [6] have failed to produce a signature of Anderson localization of EM waves.

There exist various theories which provide localization criteria for waves: if the value of $\bar{k}\ell_s$ falls below a certain value, localization is achieved. Probably, one of the most accurate among these is the potential well analogy (PWA) [20], which sets the critical value for $\bar{k}\ell_s$ to 0.844. Clearly, in a mean-field theory like the ECPA no quantitative statements as to when a wave system is crossing from extended to localized can be made. However, the value of the localization parameter $\bar{k}\ell_s$ can still be evaluated and, as a function of the system parameters, may exhibit certain trends towards parameter values optimal for localization. In this spirit, we have performed a systematic study of the localization parameter $\bar{k}\ell_s$ as a function of the dielectric contrast $\varepsilon_1/\varepsilon_2$ and filling fraction f for electromagnetic waves for the direct ($\varepsilon_1 > \varepsilon_2$) as well as for the inverse structure ($\varepsilon_2 > \varepsilon_1$). We assigned to every parameter value combination the minimum of $\bar{k}\ell_t$ as a function of frequency. In this way we were able to obtain contours of constant $\bar{k}\ell_s$ value as a function of dielectric contrast and filling fraction. Figs. 3 and 4 show the results of this study for the direct and inverse structure, respectively. We clearly obtain that localization for EM waves may be easier achieved for the inverse structure, where the optimal filling factor f is approximately around $f \approx 0.45$ for the direct and $f \approx 0.7$ for the inverse structure. The fact that for a given dielectric contrast and filling ratio the localization parameter $\bar{k}\ell_s$ is much lower in the inverse structure than in the direct structure suggests that the inverse structure is a much more strongly scattering system and may be preferred when it comes to realizing Anderson localization of EM waves. In addition, this observation is consistent with the observation that in periodic dielectric systems, the so-called photonic crystals (PCs), inverted structures much more readily exhibit complete photonic band gaps than is the case for their direct counterpart [21].

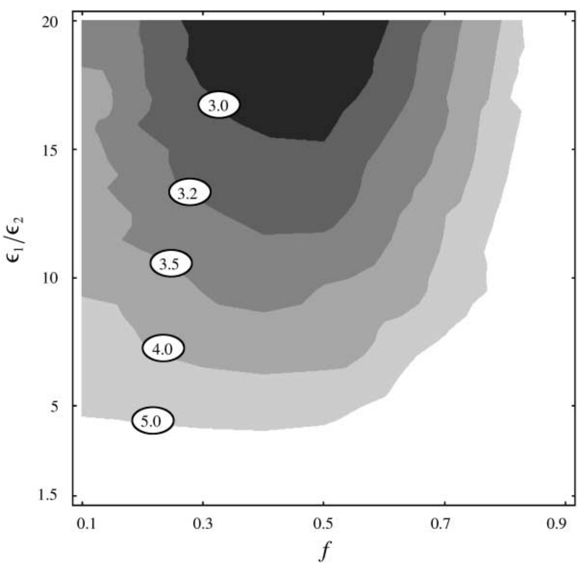

Fig. 3. Contour plot of the localization parameter $\bar{k}\ell_s$ for electromagnetic waves in three dimensions in the direct structure ($\varepsilon_1 > \varepsilon_2$) for various filling factors f and dielectric contrasts.

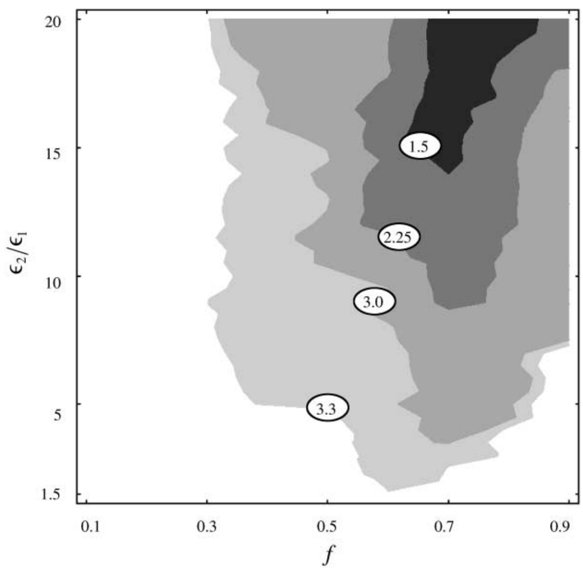

Fig. 4. Contour plot of the localization parameter $\bar{k}\ell_s$ for electromagnetic waves in three dimensions in the inverse structure ($\varepsilon_2 > \varepsilon_1$) for various filling factors f and dielectric contrasts.

The above-mentioned difference in localization behavior between direct and inverse structure may also account for the apparent absence of Anderson localization in the Si powder experiment of Gomez Rivas et al. [6] as compared to the GaAs powder experiment of Wiersma et al. [18]: Despite the fact that the refractive indices of Si and GaAs are almost equal and the volume fraction as well as the size of the scatterers in the Si and GaAs sample are comparable (and close to the optimal value predicted by ECPA), there still is a different connectivity between the particles. The shape of the GaAs particles is not nearly as spherical as the shape of the Si particles. Therefore, the GaAs sample might be much closer to an inverse structure than is the Si sample. This possible explanation of the differences seen in experiments [6] and [18] is certainly not diminishing the importance of having to separate the effects of localization and absorption [19]. To this end, novel forms of disorder, such as the recently suggested thermally disordered liquid crystal director field in otherwise isotropic silicon-based inverse opal PC [22] may have to be investigated.

4. Discussion

In summary, the effective medium theory discussed in this work, the so-called ECPA, allows to reliable calculate transport properties of disordered classical wave systems. It can be applied to a wide variety of wave propagation problems. In the long-wavelength limit well-known results are rediscovered. Without adjustable parameters, excellent qualitative as well as quantitative agreement with experiment is obtained. A study of the localization parameter $\bar{k}\ell_s$ predicts optimal parameter ranges for classical localization depending on the type of wave (scalar or vector) and type of structure (direct and inverse configuration). Generally, localization is favored in the inverse structure, since the localization parameter $\bar{k}\ell_s$ takes on significantly lower values in those structure.

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