Tr. J. of Physics
23 (1999) , 887 – 894.

Electromagnetic waves in disordered dielectric media: coupled-dipole model

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Received 02.02.1999

Abstract

An effective approach to multiple scattering of electromagnetic waves by disordered dielectric media is developed. A self-consistent energy-conserving coupled-dipole model is used. Applications to the Anderson localization of electromagnetic waves are presented. A complete set of Maxwell's equations is used to describe the propagation of waves and the vector character of the electromagnetic field is fully taken into account.

1. Introduction

Disordered dielectric media with a high refractive index contrast are a vivid subject of experimental and theoretical studies. Such environments provide interesting examples of strongly scattering media where multiple scattering effects play a dominant role. Transport properties of electromagnetic waves in such media are directly affected by interference and the standard radiative transport theory must be essentially modified to agree with experiments. It should be noted that electromagnetic waves propagating in random dielectric structures exhibit many effects that are typical for the behavior of electrons in disordered semiconductors. Therefore studies of transport properties of light and microwaves in random dielectrics can benefit from the well-developed theoretical methods and concepts of solid-state physics. A striking example is the theory of electron localization in noncrystalline systems such as amorphous semiconductors or disordered insulators. According to Anderson [1, 2], an entire band of electronic states can be spatially localized in a sufficiently disordered infinite material. So-called strong localization is achieved when the diffusion constant in the scattering medium becomes zero and the Anderson transition may be viewed as a transition from particle-like behavior described by the diffusion equation to wave-like behavior described by the Schrödinger equation. Because interference is the common property of all wave phenomena, we can expect that there are some analogs of electron localization for other types of waves. Indeed, many generalizations of electron localization exist, especially in the realm of electromagnetic waves [3, 4]. So-called weak localization of electromagnetic waves manifesting itself as enhanced coherent backscattering is presently relatively well established experimentally [5, 6, 7] and understood theoretically [8, 9]. The crucial question is whether interference effects in random dielectric media can reduce the diffusion constant to zero leading to strong localization. The essential parameter is the mean free path l which should be rather short. Localization could be experimentally demonstrated if the scale dependence of the diffusion constant in disordered dielectric media would be observed. Such a demonstration has first been given for two dimensions, where strong localization takes place for arbitrarily small value of the mean free path (if the medium is sufficiently large). The strongly scattering medium has been provided by a set of dielectric cylinders randomly placed between two parallel aluminum plates on half the sites of a square lattice [10]. Only very recently a quite successful experiment dealing with localization of light in three dimensions has been performed [11]. The strongly scattering dielectric medium was provided by the semiconductor powder gallium arsenide.

Localization of electromagnetic waves is a very nontrivial effect and proper understanding of this phenomenon requires sound theoretical models. To take fully into account the vector character of electromagnetic fields, such models should be based directly on the Maxwell equations. They should also be simple enough to provide calculations without too many approximations. Although there is a temptation to immediately apply averaging procedures as soon as disorder is introduced into the considered model, such an approach can be dangerous; averaging of the scattered intensity over some random variable leads to radiative transport theory which neglects all interference effects [12].

2. General considerations

A possible definition of localized electromagnetic waves which is used in this paper resembles the definition of localized states in quantum mechanics and makes use of the analogy between the quantum-mechanical probability density and the energy density of the field. In general, the electric field $\vec{E}(\vec{r},t)$ cannot be interpreted as the probability amplitude. The correct equivalent of the quantum-mechanical probability density is rather the energy density of the field and not the squared electric field. Therefore it seems natural to say that a monochromatic electromagnetic wave

$$\vec{E}(\vec{r},t) = \mathrm{Re}\left\{\vec{\mathcal{E}}(\vec{r})e^{-i\omega t}\right\} \quad \text{and} \quad \vec{H}(\vec{r},t) = \mathrm{Re}\left\{\vec{\mathcal{H}}(\vec{r})e^{-i\omega t}\right\}, \tag{1}$$

is localized if the time-averaged energy density of the total field tends to zero far from a region of space which contains the scattering medium. In the following we will use the fact that for rapidly oscillating monochromatic electromagnetic waves (1) only time averages are measurable. The total field

$$\vec{\mathcal{E}}(\vec{r}) = \vec{\mathcal{E}}^{(0)}(\vec{r}) + \vec{\mathcal{E}}^{(1)}(\vec{r}) \quad \text{and} \quad \vec{\mathcal{H}}(\vec{r}) = \vec{\mathcal{H}}^{(0)}(\vec{r}) + \vec{\mathcal{H}}^{(1)}(\vec{r}), \tag{2}$$

may be considered as the sum of the incident free field $\vec{\mathcal{E}}^{(0)}(\vec{r})$ and $\vec{\mathcal{H}}^{(0)}(\vec{r})$, which obeys the Maxwell equations in vacuum, and waves scattered by various parts of the medium [13]:

$$\vec{\mathcal{E}}^{(1)}(\vec{r}) = \vec{\nabla} \times \vec{\nabla} \times \vec{\mathcal{Z}}(\vec{r}) - 4\pi\,\vec{\mathcal{P}}(\vec{r}) \quad \text{and} \quad \vec{\mathcal{H}}^{(1)}(\vec{r}) = -ik\,\vec{\nabla} \times \vec{\mathcal{Z}}(\vec{r}). \tag{3}$$

The electric Hertz potential

$$\vec{\mathcal{Z}}(\vec{r}) = \int d^3r'\,\vec{\mathcal{P}}(\vec{r'})\,\frac{e^{ik|\vec{r}-\vec{r'}|}}{|\vec{r}-\vec{r'}|}, \tag{4}$$

is expressed by the polarization of the dielectric medium $\vec{\mathcal{P}}$. The above system of equations determines the electromagnetic field everywhere in space for a given free wave incident on the system. Analogous relationships between the stationary outgoing and the stationary incoming wave are known in general scattering theory as the Lippmann-Schwinger equations [14]. A way of dealing with bound states in the formalism of the Lippman-Schwinger equation is to solve it as a homogeneous equation, i.e., with the incoming wave equal to zero.

3. Coupled-dipoles model

Usually localization of light is studied experimentally in microstructures consisting of dielectric spheres or cylinders with typical dimensions (e.g., diameters) and mutual distances being comparable to the wavelength. It is well known, however, that in such a case the theoretical description becomes very complicated. On the other hand, the theory of multiple scattering of light by dielectric particles is tremendously simplified in the limit of point scatterers. In principle, this approximation is justified only when the size of the scattering particles is much smaller than the wavelength. Fortunately, in practical calculations many multiple-scattering effects including universal conductance fluctuations, enhanced backscattering, and dependent scattering [15] can be obtained qualitatively for coupled electrical dipoles. It is reasonable to assume that in many situations what really counts for localization is the scattering cross section and not the geometrical shape and real size of the scatterer. Therefore we will represent the dielectric particles located at the points $\vec{r}_a$ by single electric dipoles

$$\vec{\mathcal{P}}(\vec{r}) = \sum_a \vec{p}_a\,\delta(\vec{r} - \vec{r}_a), \tag{5}$$

with properly adjusted scattering properties. In 2D media it is convenient to introduce cylindrical coordinates $\vec{r} = (\vec{\rho}, z)$ and the dipole model leads to the polarization $\vec{\mathcal{P}}(\vec{r}) = \sum_a \vec{e}_z p_a\,\delta(\vec{\rho} - \vec{\rho}_a)$. Of course we should remember that the dipole model is only an approximation. Moreover, it is known that several mathematical problems emerge in the formulation of interactions of point-like dielectric particles with electromagnetic waves [15, 16]. To safely use the dipole approximation it is essential to use a representation for the scatterers that rigorously fulfills the optical theorem and conserves energy in the scattering processes. Therefore, the time-averaged field energy flux integrated over a surface surrounding arbitrary part of the medium should vanish:

$$\int d\vec{s}\cdot \vec{\mathcal{S}}(\vec{r}) = \frac{c}{4\pi}\frac{1}{2}\mathrm{Re}\int d\vec{s}\cdot \left\{\vec{\mathcal{E}}(\vec{r}) \times \vec{\mathcal{H}}^{*}(\vec{r})\right\} = 0. \tag{6}$$

Thus, on average, the energy radiated by the medium must be equal to the energy given to the medium by the incident wave. Using this energy conservation condition and assuming that the dielectric particles modeled by the dipoles are spherically symmetrical, we arrive at the following field-dipole coupling [17]:

$$\frac{2}{3}\, ik^3\, \vec{p}_a = \frac{e^{i\phi}-1}{2}\, \vec{\mathcal{E}}'(\vec{r}_a). \tag{7}$$

Thus, to provide conservation of energy, the dipole moments $\vec{p}_a$ must be coupled to the electric field of the incident wave $\vec{\mathcal{E}}'(\vec{r}_a)$ by complex polarizability $(e^{i\phi}-1)/2$. We get for the 2D model the same kind of coupling, with the only difference being the constant in the front of the dipole moment: $i\pi k^2$ [18]. To get some insight into the physical meaning of the parameter ϕ from Eq. (7) let us observe that it is directly related to the total scattering cross-section σ of an individual dielectric sphere represented by the single point-like dipole in 3D. Indeed, within our formalism, the explicit formula for the total scattering cross section of a dipole reads: $2k^2\,\sigma = 3\pi(1-\cos\phi)$. A very similar relationship holds also for the total scattering cross section of a dielectric cylinder modeled by a 2D dipole: $k\,\sigma = 2(1-\cos\phi)$. In both cases ϕ is a function of frequency and the physical parameters describing the scatterer (dielectric constant and radius of a sphere or a cylinder).

The field acting on the ath dipole,

$$\vec{\mathcal{E}}'(\vec{r}_a) = \vec{\mathcal{E}}^{(0)}(\vec{r}_a) + \sum_{b\neq a} \vec{\mathcal{E}}_b(\vec{r}_a), \tag{8}$$

is the sum of the free field and waves scattered by all other dipoles:

$$\vec{\mathcal{E}}_a(\vec{r}) = \tilde{g}(\vec{r}-\vec{r}_a)\cdot \vec{p}_a. \tag{9}$$

In 2D the Green function $\tilde{g}(\vec{\rho}) = 2k^2 K_0(-ik|\vec{\rho}|)$ is given by the Bessel function of the second kind [18]. In 3D $\tilde{g}$ denotes the proper Green tensor

$$\tilde{g}(\vec{r}) = k^2\,\frac{e^{ik|\vec{r}|}}{|\vec{r}|}\left\{\left(\frac{3}{(k|\vec{r}|)^2} - i\frac{3}{k|\vec{r}|} - 1\right)\frac{\vec{r}\,\vec{r}}{|\vec{r}|^2} - \left(\frac{1}{(k|\vec{r}|)^2} - i\frac{1}{k|\vec{r}|} - 1\right)\right\}. \tag{10}$$

Inserting Eq. (9) into (8), and using (7), it is easy to finally obtain the system of linear equations:

$$\sum_b M_{ab}\cdot \vec{\mathcal{E}}'(\vec{r}_b) = \vec{\mathcal{E}}^{(0)}(\vec{r}_a), \tag{11}$$

determining the field $\vec{\mathcal{E}}'(\vec{r}_a)$ acting on each dipole for a given free field $\vec{\mathcal{E}}^{(0)}(\vec{r}_a)$. If we solve it and use again Eqs. (7) and (9), we are able to find the electromagnetic field everywhere in space outside of the dipoles. Nonzero solutions $\vec{\mathcal{E}}'(\vec{r}_a) \neq 0$ of Eq. (11) for the incoming wave equal to zero $\vec{\mathcal{E}}^{(0)}(\vec{r}_a) \equiv 0$ may be interpreted as localized waves [17]. Let us stress that perfectly localized waves exist only in infinite systems of dipoles.

5. Towards strong localization

It seems reasonable to expect that each electromagnetic wave localized in a system of dipoles (5) usually corresponds to a certain curve on the plane $\{\omega, \phi\}$. Nevertheless, in the case of random and infinite system of dipoles there can exist an entire continuous band of spatially localized states corresponding to a region in the plane $\{\omega, \phi\}$. After choosing a point (ω, ϕ) from this region a localized wave of frequency (arbitrarily near) ω exists in almost any random distribution of the dipoles described by the scattering properties ϕ. To illustrate this statement we have to study the properties of finite systems for increasing number of dipoles N (while keeping the density constant). For each distribution of the dipoles $\vec{r}_a$ placed randomly inside a sphere with the uniform density of $n = 1$ dipole per wavelength cubed (in 2D they are placed randomly inside a square with the uniform density $n = 1$ dipole per wavelength squared) we have diagonalized numerically the M matrix from Eq. (11) and obtained the lowest eigenvalue:

$$\Lambda(\phi) = \min_j |\lambda_j(\phi)|. \tag{12}$$

The resulting probability distribution $P_\phi(\Lambda)$, calculated from 10^3 different distributions of N dipoles, is normalized in the standard way: $\int d\Lambda\, P_\phi(\Lambda) = 1$. Let us now compare the contour plots of $P_\phi(\Lambda)$ (treated as a function of two variables ϕ and Λ) calculated for systems consisting of $N = 300$ three dimensional and two dimensional dipoles. They are presented in Figs. 1 and 2, respectively. It is seen from inspection of these plots that there is a region of ϕ values (this region strongly depends on dimensionality) for which the probability distribution $P_\phi(\Lambda)$ is apparently closer to the $\Lambda = 0$ axis than for other values of ϕ. In 3D this happens only for values of $|\phi|$ that are sufficiently greater than zero but smaller than π. Our numerical investigations indicate that for increasing size of the system the variances of $P_\phi(\Lambda)$ decrease. In the limit of an infinite medium, the probability distributions presented in these figures seem to tend to the delta function

$$\lim_{N\to\infty} P_\phi(\Lambda) = \delta(\Lambda), \quad \text{for} \quad |\phi| \geq \phi_{\rm cr}. \tag{13}$$

Therefore, for some ϕ the function $\Lambda(\phi)$ is an example of a self-averaging quantity. This means that for almost any random distribution of the dipoles $\vec{r}_a$, the equation $\lambda_j(\phi) = 0$ is satisfied. Thus, as we expected, a localized wave described by the corresponding eigenvector of the M matrix, exists. Similarly, localized electronic states in solids appear always at discrete energies only. However, in the case of a disordered and unbounded system a countable set of energies corresponding to localized states becomes dense in some finite interval. It is a signature that the Anderson localization occurs. However it is always difficult to distinguish between the allowed energies which may be arbitrarily close to each other. Therefore, physically speaking, an entire continuous band of spatially

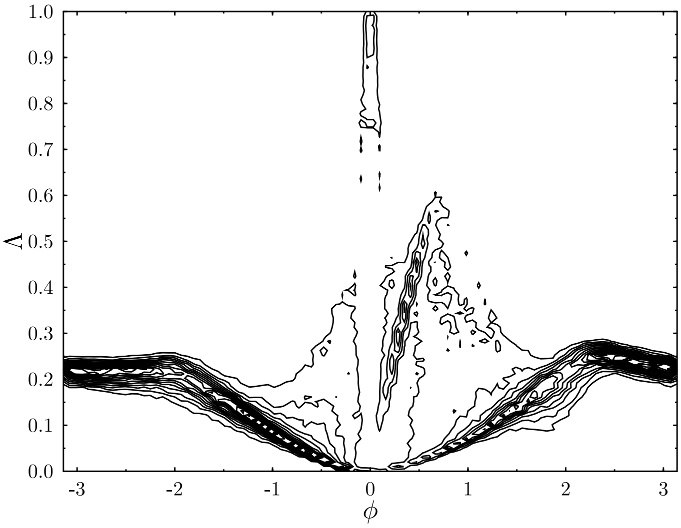

Figure 1. Contour plot of the probability distribution $P_\phi(\Lambda)$ calculated for 10^3 systems of $N = 300$ three dimensional dipoles distributed randomly in a sphere with the density $n = 1$ dipole per wavelength cubed.

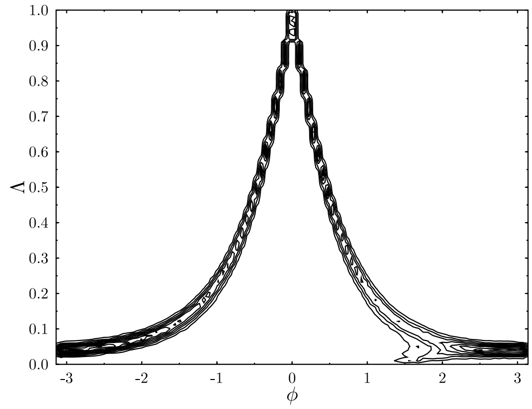

Figure 2. Contour plot of the probability distribution $P_\phi(\Lambda)$ calculated for 10^3 systems of $N = 300$ two dimensional dipoles distributed randomly in a square with the density $n = 1$ dipole per wavelength squared.

localized electronic states exists. In 3D the total scattering cross section of individual particles σ must exceed some critical value $\sigma_{\rm cr} = \sigma(\phi_{\rm cr})$ before localization will take place in the limit $N \to \infty$. This fact is in perfect agreement with the scaling theory of localization [19]. Moreover, our preliminary calculations indicate that in three dimensions the value of $k^2\,\sigma_{\rm cr}$ may decrease with n but slower than n^{-2}. Within our approach internal resonances of scatterers can be modeled by $|\phi| \simeq \pi$. Our calculations do not exclude the possibility that in infinite three dimensional medium the band of localized waves may appear in this region of ϕ. However, in all experiments we can investigate only systems confined to certain finite regions of space. And, as follows from Fig. 1, in 3D the band of localized waves appears faster with increasing size of the system when $\phi_{\rm cr} \le |\phi| \ll \pi$, i.e., when the frequency is not tuned to the internal resonances of individual scatterers. Let us emphasize that this result is specific for 3D random media. In both one and two dimensions, macro- and microscopic resonances appear at the same frequencies. To illustrate this fact, in Fig. 2 we have prepared the plot analogous to Fig. 1 but calculated for 10^3 configurations of $N = 300$ dielectric cylinders modeled by 2D dipoles [18]. In this case, the band of localized waves does appear faster for $|\phi| \simeq \pi$; in 2D the parameters of the single scatterers that give the internal and global resonances coincide and matching the internal resonances helps to establish localization. In our opinion this could be the main reason that the convincing experimental demonstration of strong localization of microwave radiation has first been given for two dimensions [10], although also more practical reasons as, e.g., polydispersity of the actual 3D samples (leading to phase shifts) play an important role. Results obtained from our model for 2D media (which seem to agree with experimental results) prove that the surprising features of localization we observed for 3D random media are not the artifacts produced by the model.

6. Summary

We have developed a quite realistic coupled-dipole model describing multiple scattering of electromagnetic waves by a disordered dielectric medium. Its relative simplicity allowed us to discover some new features of the Anderson localization of electromagnetic waves in 3D dielectric media without using any averaging procedures. Within our theoretical approach one can easily see how localization "sets in" for increasing size of the system. Very striking universal properties of random matrices describing the scattering from a collection of randomly distributed point-like scatterers have been observed. Self-averaging of the lowest eigenvalue emerging in the limit of infinite medium has been discovered numerically and the appearance of the band of localized electromagnetic waves in 3D was demonstrated. Possible relationships between this phenomenon and the dramatic inhibition of the propagation of electromagnetic waves in spatially random dielectric media are currently under investigation. It can be understood as a counterpart of Anderson transition in solid state physics.

Acknowledgments

We are grateful to Edward Kapuścik and Andrzej Horzela for their kind hospitality extended to us in Kraków. This work was supported by the Polish Committee for Scientific Research (KBN) under Grant No. 2 P03B 108 12. We thank the Interdisciplinary Center for Mathematical and Computational Modeling (ICM) of Warsaw University for providing us with their computer resources.

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