

## Transport Properties of Random Media: A New Effective Medium Theory

K. Busch<sup>1,2</sup> and C. M. Soukoulis<sup>1</sup>

<sup>1</sup>Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011

<sup>2</sup>Institut für Theorie der Kondensierten Materie, Universität Karlsruhe, 76128 Karlsruhe, Germany

(Received 11 May 1995)

We present a new method for efficient, accurate calculations of transport properties of random media. 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 scatterers. This scheme captures the effects of resonant scattering of the individual scatterer exactly, as well as the multiple scattering in a mean-field sense. It has been successfully applied to both “scalar” and “vector” classical wave calculations. Results for the energy transport velocity are in agreement with experiment. This approach is of general use and can be easily extended to treat different types of wave propagation in random media.

PACS numbers: 42.25.Bs, 05.60.+w, 41.20.Jb, 78.20.Dj

In recent years, there has been a growing interest in studies of the propagation of classical waves in random media [1]. While some of the features associated with weak localization, such as enhanced coherent backscattering, have been detected in light scattering experiments [1], the localization of classical waves in random systems has not been established beyond doubt. Recent experimental results [2] for the diffusion coefficient  $D$  and the transport mean free path  $\ell_t$  demonstrated that in a disordered medium, low values of the diffusion constant  $D = v_E \ell_t / 3$  were caused by extremely small values of the energy transport velocity  $v_E$  and not by the small values of  $\ell_t$ , which signifies strong localization. To explain this low value of the transport velocity, a theory was developed by van Albada *et al.* [2], in the low-concentration limit, of the Bethe-Salpeter equation. They argued that their approach gives the correct transport velocity observed experimentally, which is the energy transport velocity  $v_E$  and not the phase velocity  $v_p$ .  $v_p$  is approximately equal to the velocity of light  $c$  divided by an appropriate index of refraction.  $v_E$  is always less than  $v_p$ , especially close to the Mie resonances. The renormalization of the diffusion coefficient near resonances in random media has been extensively studied [3–8] after its introduction by the pioneer work of van Albada *et al.* [2]. It is by now well understood that to lowest order in density of the dielectric scatterers, the strong decrease in the transport velocity is due to the Mie resonances. Near resonances, a lot of energy is temporarily stored inside the dielectric scatterer or equivalently the wave spends a lot of time (dwell time) inside the dielectric scatterer.

Experimental results [9] for alumina spheres have shown that as the volume fraction of the scatterers  $f$  increases towards close packing ( $f \approx 0.60$ ), there is no structure in the diffusion coefficient versus frequency. This clearly suggests that there is no structure in the transport velocity. It is, therefore, inappropriate to calculate the transport velocity using the  $v_E$  of van Albada *et al.* [2] in this high- $f$  regime since their theory for  $v_E$

is a low-concentration theory. But, if we, nevertheless, calculate [10,11]  $v_E$  according to Ref. [2] for this high  $f = 0.60$ , strong structure in  $v_E$  is obtained in disagreement with the experimental results. An extension of the well-known coherent-potential approximation (CPA) was recently developed [10] and obtained a CPA phase velocity for  $f = 0.60$ , which is qualitatively consistent with experiment, in not showing any structure as a function of the frequency. The newly developed [10] coated CPA for low  $f$  gives a CPA phase velocity which is higher than the velocity of light near Mie resonances. This is an undesirable feature of the CPA that had to be fixed. Thus, for small  $f$ , the theory of van Albada *et al.* [2] seems to give the correct transport velocity  $v_E$ , while for large  $f$ , it is the coated CPA approach [10–12] that seems to give transport velocities consistent with experiment [9].

In the present Letter, we present a new approach in calculating the transport properties of random media that takes into account the multiscattering interactions in a mean-field sense. The main new physical idea is that in a random medium *the energy density should be uniform when averaged over the correlation length of the microstructure*. This approach has been applied to *both scalar and vector* classical wave propagation in random media with many successes. For both the scalar and vector cases, we obtain results for the energy transport velocity that gives pronounced dips in  $v_E$  for low  $f$ , while as  $f$  increases the dips are smeared out, as expected and in agreement with experiment. In addition, this new energy-density CPA gives the correct long-wavelength limit for the effective dielectric constant for both the scalar and vector cases. The energy density for the vector case is calculated exactly, where both the electric and magnetic field contributions are taken into account. For the vector case, analytical as well as numerical results of this approach give that the long-wavelength dielectric constant is given by the Maxwell-Garnett formula. The formalism that has been developed in this Letter can be easily extended to treat different types of wave propagation in disordered systems and is therefore of general use.

We consider a composite medium of two lossless materials, with dielectric constants  $\epsilon_1$  and  $\epsilon_2$ . Our composite medium is assumed to consist of spheres with diameter  $d = 2R$  and dielectric constant  $\epsilon_1$  randomly placed within the host material with dielectric constant  $\epsilon_2 = 1$ . The random medium is characterized also by  $f$ , the volume fraction occupied by the spheres. We consider first the propagation of classical waves [13] in a random medium described by the wave equation for the scalar field  $\Psi$ ,  $[\nabla^2 + \omega^2 \epsilon(r)/c^2]\Psi = 0$ , where  $\epsilon(r)$  is a random variable. By correctly handling the Ward identities, van Albada *et al.* [2,7,8] reported that in the low density limit,  $D = v_E \ell_i/3$ , where  $v_E$  is given by

$$v_E = \frac{c^2}{v_p(1 + \delta)}. \quad (1)$$

The quantity  $\delta$  is given by

$$\delta = n \int d^3r |\Psi_k^+(r)|^2 [\epsilon(r) - 1], \quad (2)$$

with  $n$  the number density of the scatterers, and  $\Psi_k^+(r)$  is the one-scatterer eigenfunction with incident wave vector  $k$  for a single dielectric scatterer. Physically,  $\delta$  can be larger when the incident wave frequency coincides with an internal resonance of the scatterer. When that happens,  $|\Psi_k^+(r)|^2$  has a large magnitude inside the scatterer, leading to a large  $\delta$ , and therefore, to small values of  $v_E$ . A more convenient representation for numerical purposes of  $\delta$  for scalar waves was given by van Tiggelen [14], where  $v_E$  was written with respect to the Van de Hulst scattering coefficients of the scalar dielectric sphere [see Eq. (3.87) of Ref. [14]]. The Amsterdam group [2,7,8,14] extended their scalar results for the renormalization of  $v_E$  to the vector case by simply replacing the scalar single-scatterer  $t$  matrix with the vector  $t$  matrix. This is an oversimplified approximation of the real vector problem. The polarization of the EM waves has to be taken into account on a fully vector calculation in deriving the Boltzmann equation, starting from the Bethe-Salpeter equation. This is still the outstanding problem of the field. If indeed one makes this approximation, a  $v_E$  is obtained [see Eqs. (28) and (29) of Ref. [2]] that is much lower than  $v_p$  and it has pronounced dips close to the Mie resonances of the isolated dielectric scatterer. In addition, the long-wavelength limit of  $v_E$  and therefore of the dielectric constant  $\bar{\epsilon}$  is given by  $\bar{\epsilon} = 1 + 3f(\epsilon_1 - \epsilon_2)/(\epsilon_1 + 2\epsilon_2)$  and not by the Maxwell-Garnett theory result, which is the "correct" result for the vector case.

Here, we present a new approach for calculating the transport properties. Consider, for example, a random medium composed of a dispersion of spheres as shown in Fig. 1. The basic structural unit may be regarded as a coated sphere, as represented by the dashed lines in Fig. 1(a). The radius of the coated sphere  $R_c = R/f^{1/3}$ , where  $R$  is the radius of the solid sphere. Let  $k_e = \sqrt{\bar{\epsilon}} \omega/c$  characterize the effective medium, which has an average dielectric constant  $\bar{\epsilon}$  to be determined

self-consistently. The self-consistent condition for the determination of  $k_e$  or  $\bar{\epsilon}$  is that the energy content of the coated sphere embedded in the effective medium [see Fig. 1(b)] is equal to the energy content of a sphere of radius  $R_c$  with  $\bar{\epsilon}$  dielectric constant [see Fig. 1(c)] i.e.,

$$\int_0^{R_c} d^3r \rho_E^{(1)}(\vec{r}) = \int_0^{R_c} d^3r \rho_E^{(2)}(\vec{r}), \quad (3)$$

which is also shown schematically in Fig. 1.  $\rho_E^{(1)}(r)$  and  $\rho_E^{(2)}(r)$  are the energy densities for the configurations shown in Figs. 1(a) and 1(b), respectively. For scalar waves the energy density is

$$\rho_E(\vec{r}) = \frac{1}{2} [\omega^2 \epsilon(\vec{r}) |\Psi(\vec{r})|^2/c^2 + |\vec{\nabla} \Psi(\vec{r})|^2], \quad (4a)$$

whereas the energy density of the vector waves is given by

$$\rho_E(\vec{r}) = \frac{1}{2} [\epsilon(\vec{r}) |\vec{E}(\vec{r})|^2 + \mu |\vec{H}(\vec{r})|^2], \quad (4b)$$

where  $\mu$  is the magnetic permeability which is taken equal to 1 and  $\Psi(r)$  and  $\vec{E}(r)$  and  $\vec{H}(r)$  are the scattering wave function and the scattered electric and magnetic fields for a plane wave incident on a coated sphere, respectively. In Eq. (3),  $\bar{\epsilon}$  is the parameter to be determined. It should be noted that the energy density, and, therefore,  $\Psi(r)$  being a scattering wave function, implicitly depends on  $\bar{\epsilon}$ . We, therefore, have Eq. (3) as the self-consistent condition for our energy-density CPA. We find that this condition is

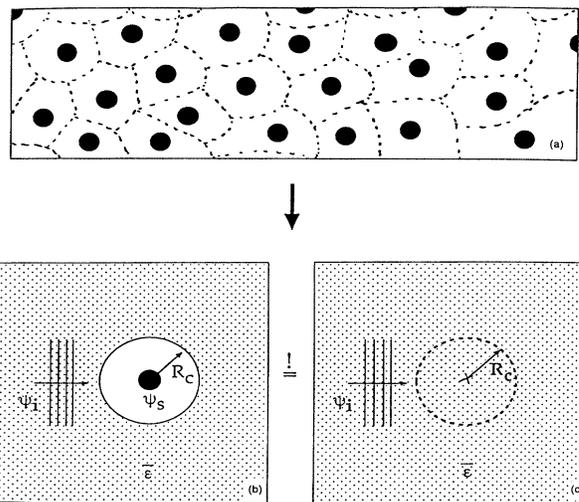


FIG. 1. (a) In a random medium composed of dielectric spheres, the basic scattering unit may be regarded as a coated sphere, as represented by the dashed lines. To calculate the effective dielectric constant  $\bar{\epsilon}$ , a coated sphere of radius  $R_c = R/f^{1/3}$  is embedded in a uniform medium.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

easily satisfied with a few numbers of iterations (of the order of 10) for all frequencies and filling ratios. This was not the case for all the previous CPAs. There the solution of the corresponding self-consistent equation can disappear or jump abruptly or even get multiple solutions. This new scheme can easily follow the unique solution of the self-consistent equation. We feel that the integration over all angles in Eq. (3) is responsible for the well behaved solution. It should be noted that  $\Psi(r)$ ,  $\vec{E}(r)$ , and  $\vec{H}(r)$ , being scattering wave functions, implicitly depend on  $\bar{\epsilon}$ . When Eq. (3) is satisfied, we have that the energy transport velocity

$$v_E = \frac{c}{\sqrt{\bar{\epsilon}}} \sqrt{1 - \text{Re}\Sigma/k_e^2}, \quad (5)$$

where the self-energy  $\Sigma = 4\pi n f(0)$  is calculated with the embedding medium characterized by  $\bar{\epsilon}$  and  $f(0)$  is the forward scattering amplitude. Equations (3), (4), and (5), together define a mean-field approach to the calculation of the transport velocity.

We have systematically calculated the energy content of the sphere and coated sphere for the scalar and vector [15] cases. It is very remarkable that the energy stored in a dielectric sphere or coated sphere for both the scalar and vector cases are given by these relatively simple forms, which are very convenient for numerical calculations. In Fig. 2, we present the result obtained for the frequency dependence of energy transport velocity for scalar waves and for three different filling ratios. We presented the frequency as  $d/\lambda_i$ , where  $d$  is the diameter of the dielectric sphere and  $\lambda_i = 2\pi c/\omega\sqrt{\epsilon_1}$  is the wavelength inside the sphere. Notice that the  $v_E$  exhibits pronounced dips as a function of frequency only at low scatterer concentrations, in agreement with the results of the Amsterdam group [2,7,8] and experiments [9]. At around  $f = 0.10$  the Boltzmann theory of the Amsterdam group starts to deviate from our results. At higher scatterer concentrations, the variation with frequency is reduced, as physically expected. Remember that the low density approximation of the Amsterdam group gives negative values of  $v_E$  for  $f = 0.60$ . For the scalar case, we also calculated analytically the long-wavelength limit of  $\bar{\epsilon}$ , and indeed we find that  $\bar{\epsilon} = f\epsilon_1 + (1-f)\epsilon_2$ , as expected. In Eq. (3), if one replaces the energy density  $\rho_E^{(2)}(r)$  of the effective medium by  $\rho_E^{(1)}(r)$ , one obtains the self-consistent equation [13,14]

$$\int_0^{R_c} d^3r \rho_E^{(1)}(r) [\epsilon(r) - \bar{\epsilon}] = 0$$

This equation is exactly equal to Eq. (1), provided that the background dielectric constant is  $\bar{\epsilon}$  and not unity.

This new energy-density CPA scheme can be easily applied to the vector case. The energy density for the vector case is given in Eq. (4b), and contains the contribution from both the electric and magnetic fields. Up to now all the low density limit theories, as well as the CPA, were developed by just transferring all the

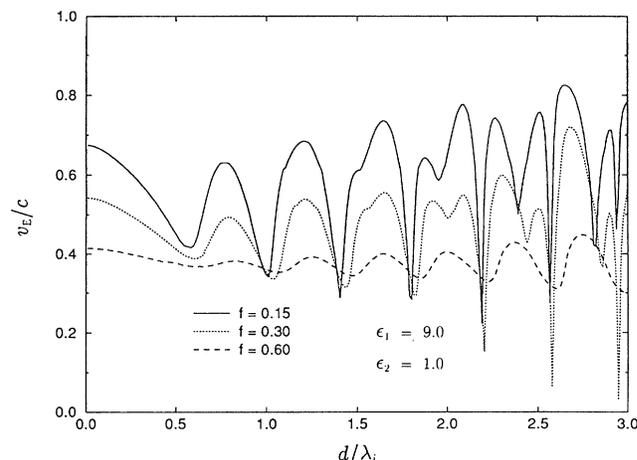


FIG. 2. The energy transport velocity  $v_E$  for the *scalar* waves calculated by the energy-density CPA vs  $d/\lambda_i$  for alumina spheres with dielectric constant 9.0 for different values of filling ratios.

scalar wave formalism to the vector case, without taking the vector character of the wave function into account. The vector character was used only in calculating the  $t$  matrix or the forward-scattering amplitude for the dielectric scatterer. In Fig. 3, we present the results for the frequency dependence of  $v_E$  for the vector case, for three values of  $f$ . Notice that for  $f = 0.15$ ,  $v_E$  has pronounced dips close to the Mie resonances, but these dips become weaker or disappear altogether as  $f$  increases. These results capture the correct physics, by exhibiting dips near resonant frequencies for low  $f$  and no structure for high  $f$ , in agreement with experiment. We have also analytically calculated the long-wavelength

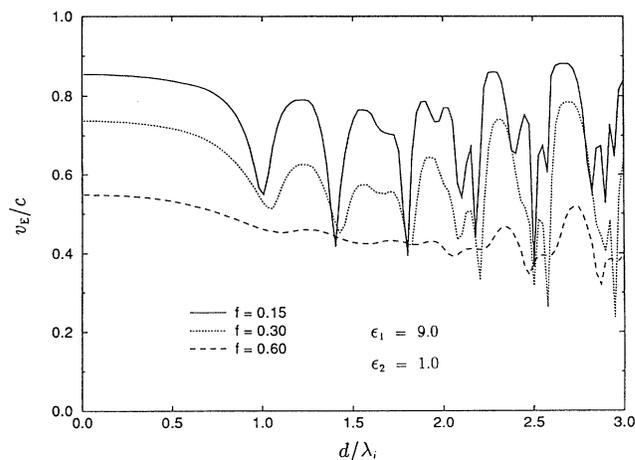


FIG. 3. The energy transport velocity  $v_E$  for *vector* waves calculated by the energy-density CPA vs  $d/\lambda_i$  for alumina spheres with dielectric constant 9.0 for different values of filling ratios.

limit results for  $\bar{\epsilon}$  and found that

$$\bar{\epsilon} = \epsilon_2 \left( 1 + \frac{3f\alpha}{1-f\alpha} \right) \quad \text{where } \alpha = \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}, \quad (6)$$

which is the Maxwell-Garnett theory result. Finally, we want to mention that within the energy-density CPA, we can also calculate [13] the scattering mean path  $\ell_s$  from the following expression:

$$\ell_s = \frac{1}{\sqrt{2} \text{Im}\Sigma} \left[ \left( k_e^2 - \text{Re}\Sigma \right) + \sqrt{\left( k_e^2 - \text{Re}\Sigma \right)^2 + \left( \text{Im}\Sigma \right)^2} \right]^{1/2}. \quad (7)$$

Preliminary results for  $\ell_s$  agree with the weak scattering results for low  $f$  and low frequency. At higher  $f$ , there is structure in the frequency dependence of  $\ell_s$ , near the Mie resonances, which becomes weaker as  $f$  increases.

We want to thank Ping Sheng for discussing an energy homogenization idea as applied to the scalar wave case in Chap. 8 of his book (Ref. [13]), and E.N. Economou and P. Wölfle for useful discussions. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG-82. This work was supported by the Director of Energy Research, Office of Basic Energy Sciences, and NATO Grant No. CRG 940647.

- 
- [1] For a review, see *Scattering and Localization of Classical Waves in Random Media*, edited by Ping Sheng (World Scientific, Singapore, 1990); *Photonic Band Gaps and Localization*, edited by C.M. Soukoulis (Plenum, New York, 1993).
- [2] M.P. van Albada, B.A. van Tiggelen, A. Lagendijk, and A. Tip, Phys. Rev. Lett. **66**, 3132 (1991); B.A. van Tiggelen, A. Bart, A. Lagendijk, P.M. van Albada, and A. Tip, Phys. Rev. B **45**, 12 233 (1992).
- [3] E. Kogan and M. Kaveh, Phys. Rev. B **46**, 10 636 (1992).
- [4] G. Cwilich and Y. Fu, Phys. Rev. B **46**, 12 015 (1992).
- [5] J. Kroha, C.M. Soukoulis, and P. Wölfle, Phys. Rev. B **47**, 11 093 (1993).
- [6] Yu.N. Barabanenkov and V. Ozrin, Phys. Rev. Lett. **69**, 1364 (1992).
- [7] B.A. van Tiggelen, A. Lagendijk, and A. Tip, Phys. Rev. Lett. **71**, 1284 (1993).
- [8] B.A. van Tiggelen and A. Lagendijk, Europhys. Lett. **23**, 311 (1993).
- [9] N. Garcia, A.Z. Genack, and A.A. Lisyansky, Phys. Rev. B **46**, 14 475 (1992); A.A. Lisyansky *et al.*, in *Photonic Band Gaps and Localization*, edited by C.M. Soukoulis (Plenum, New York, 1993), p. 171.
- [10] C.M. Soukoulis, S. Datta, and E.N. Economou, Phys. Rev. B **49**, 3800 (1994).

- [11] K. Busch, C.M. Soukoulis, and E.N. Economou, Phys. Rev. B **50**, 93 (1994).
- [12] X. Jing, P. Sheng, and M. Zhou, Phys. Rev. A **46**, 6513 (1992); Physica (Amsterdam) **207A**, 37 (1994).
- [13] Ping Sheng, *Introduction to Wave Scattering, Localization and Mesoscopic Phenomena*, (Academic, New York, 1995), Chaps. 3, 4, and 8.
- [14] B.A. van Tiggelen, Ph.D. thesis, University of Amsterdam, 1992.
- [15] The energy content of a coated sphere in the vector case is given by  $E = E_s + E_c$  where

$$E_s = \frac{1}{2} \frac{\omega^2}{c^2} \epsilon_1 \sum_{l=1}^{\infty} \frac{1}{k_1^3} \int_0^{k_1 R_i} \rho^2 d\rho \times (|c_l|^2 + |d_l|^2) W_l^{(v)}(j_l, j_l),$$

$$E_c = \frac{1}{2} \frac{\omega^2}{c^2} \epsilon_2 \sum_{l=1}^{\infty} \frac{1}{k_2^3} \int_{k_2 R_i}^{k_2 R_c} \rho^2 d\rho \times [ (|c_l|^2 \phi_l^2 + |d_l|^2 \gamma_l^2) \times W_l^{(v)}(j_l, j_l) + (|c_l|^2 \xi_l^2 + |d_l|^2 \eta_l^2) \times W_l^{(v)}(n_l, n_l) + 2(|c_l|^2 \phi_l \xi_l + |d_l|^2 \gamma_l \eta_l) \times W_l^{(v)}(j_l, n_l) ],$$

$$W_l^{(v)}(z_l, \bar{z}_l) = (2l + 1) z_l(\rho) \bar{z}_l(\rho) + (l + 1) z_{l-1}(\rho) \bar{z}_{l-1}(\rho) + l z_{l+1}(\rho) \bar{z}_{l+1}(\rho),$$

$$\phi_l = \psi_l(k_1 R) \chi_l'(k_2 R) - (k_2/k_1) \psi_l'(k_1 R) \chi_l(k_2 R),$$

$$\xi_l = \psi_l(k_2 R) \psi_l'(k_1 R) - (k_2/k_1) \psi_l'(k_2 R) \psi_l(k_1 R),$$

$$\gamma_l = (k_2/k_1) \chi_l(k_2 R) \psi_l'(k_1 R) - \chi_l'(k_2 R) \psi_l(k_1 R),$$

$$\eta_l = (k_2/k_1) \psi_l(k_2 R) \psi_l'(k_1 R) - \psi_l'(k_2 R) \psi_l(k_1 R),$$

where  $k_i = \epsilon_i^{1/2} \omega/c$  and  $i = 1, 2$ .  $\psi$  and  $\chi$  denote the Ricatti-Bessel functions of first and second kind, respectively. The  $c_l$  and  $d_l$  are the scattering coefficients for the field inside the core. Similar expressions have been obtained for the scalar case.