Transport and scattering mean free paths of classical waves

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(Received 18 November 1993; revised manuscript received 16 March 1994)

Transport, as well as scattering, mean free paths are calculated within the weak-scattering approximation and within a recently developed extension of the well-known coherent-potential approximation for a random arrangement of dielectric spheres. The different theoretical results of the mean free paths are compared with experiments on scattering from dielectric spheres.

Recently, there has been 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. Approximate theories of localization using as inputs results based on the low-concentration approximation (LCA) or weak-scattering approximation (WSA) and on the coherent-potential approximation (CPA) predict frequency intervals within which localization should be observed.[1] These predictions, however, are based on results which either ignore or treat the multiple scattering from cluster improperly and, therefore, may not be justified. Recent experimental work[2] has indicated strong photon localization in the microwave regime. In addition, it was recently recognized[3] that considerable care is needed in interpreting low values of the diffusion coefficient in studies for the search of light localization. Experimental results[3] for the diffusion coefficient D and transport mean free path ℓ_t demonstrated that in the strongly scattering random regime, low values of the diffusion coefficient $D = v_E\ell_t/3$ were caused by extremely small values of the energy transport velocity v_E and not by the small values of ℓ_t, which signifies strong localization. It is, therefore, possible that in a random medium the transport velocity can be much lower than the phase velocity, which is approximately equal to the velocity of light divided by an appropriate average index of refraction. To explain this low value of the transport velocity, a transport theory was developed by Albada *et al.*,[3] in the low-concentration limit, within the Boltzman approximation. They argued[3] that their approach gives the correct transport velocity, which is the energy transport velocity v_E and confirmed the observed small values of the transport velocity. However, for high scatterer volume fraction f ($f = 0.60$), the experimental results[2] for alumina spheres show that there is no structure in the diffusion coefficient versus frequency, which suggests there is no structure in the transport velocity. It is not appropriate to calculate the transport velocity using the v_E of Albada *et al.*[3] in this high-f regime since their theory for v_E is a low-concentration theory. But if we, nevertheless, calculate[4] v_E according to Ref. 3 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[4] 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. Not surprisingly, the newly developed[4] coated CPA for low f gives a CPA phase velocity which reduces to the regular phase velocity which is higher than the velocity of light near Mie resonances. This is an undesirable feature of the CPA, which had to be fixed. Thus, for small f, the theory of Albada *et al.*[3] seems to give the correct transport velocity v_E, while for large f it is the coated CPA approach[4] which seems to give velocities consistent with experiment.[2] Soukoulis *et al.*[4] have combined these two approaches and have calculated the transport velocity for general f. In particular, they have extended[4] the approach of Albada *et al.*[3] for calculating v_E in the following way. Using the coated CPA a frequency-dependent effective dielectric function for each f is calculated. Then the energy velocity expressions of Albada *et al.*[3] were used to calculated v_E with the outside medium having the CPA effective dielectric function ϵ_e, which is frequency dependent and not equal to the host dielectric function, which can be taken to be unity for the purpose of our discussion. This approach for low f gives a v_E, which completely agrees with that of Albada *et al.*, since ϵ_e is very close to unity. As f increases the ϵ_e becomes larger than 1 and in addition, develops some frequency dependence. For $f = 0.60$, the energy transport velocity calculated this way shows little structure, agrees with the CPA phase velocity, and more importantly, agrees with experiment. This is a reasonable approach[4] in treating the very difficult regime of transport properties in the high-concentration limit.

Another important point that we want to address is that the usage of the word mean free path, in the field of propagation of classical waves in random media, has been very confusing at times. In particular, the diffusion coefficient $D = v_E\ell_t/3$, derived within the Boltzmann transport theory, depends on the transport mean free path ℓ_t. We want to stress that the transport mean free path ℓ_t is defined as the length over which momentum transfer becomes uncorrelated. This is different from the scattering mean free path ℓ which describes the decay length of the average single-particle Green's function and is easily calculated within the CPA. The transport mean free path involves an extra factor $(1-\cos\theta)$ in the calculation of the total cross section. Only in the isotropic case (or more generally, if there is no p-spherical harmonic) the

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$\cos\theta$ term averages to zero, and, therefore, $\ell \simeq \ell_t$. This is not the case in the propagation of classical waves in a random arrangement of dielectric spheres, where strong Mie resonant scattering[5] is present. In this paper, we make a detail study of the different definitions of the transport and scattering mean free paths and compare with experimental measurements.

The theory of wave propagation in a three-dimensional (3D) weakly random medium is based on the implicit assumption that disorder modifies the phase of the unperturbed wave function. The free space wave vector k_0 is renormalized to k and at the same time gains an imaginary part $i/2\ell$, where ℓ is the phase coherence length or scattering mean free path, i.e., $k_0 \to k + i/2\ell$. As long as $k\ell \gg 1$, the effect of disorder on the amplitude of the unperturbed wave function is negligible, thus justifying the traditional approach which ignores any amplitude fluctuations. A very efficient way to obtain both k and ℓ is the so-called coherent-potential approximation (CPA). The CPA introduces a yet undetermined effective

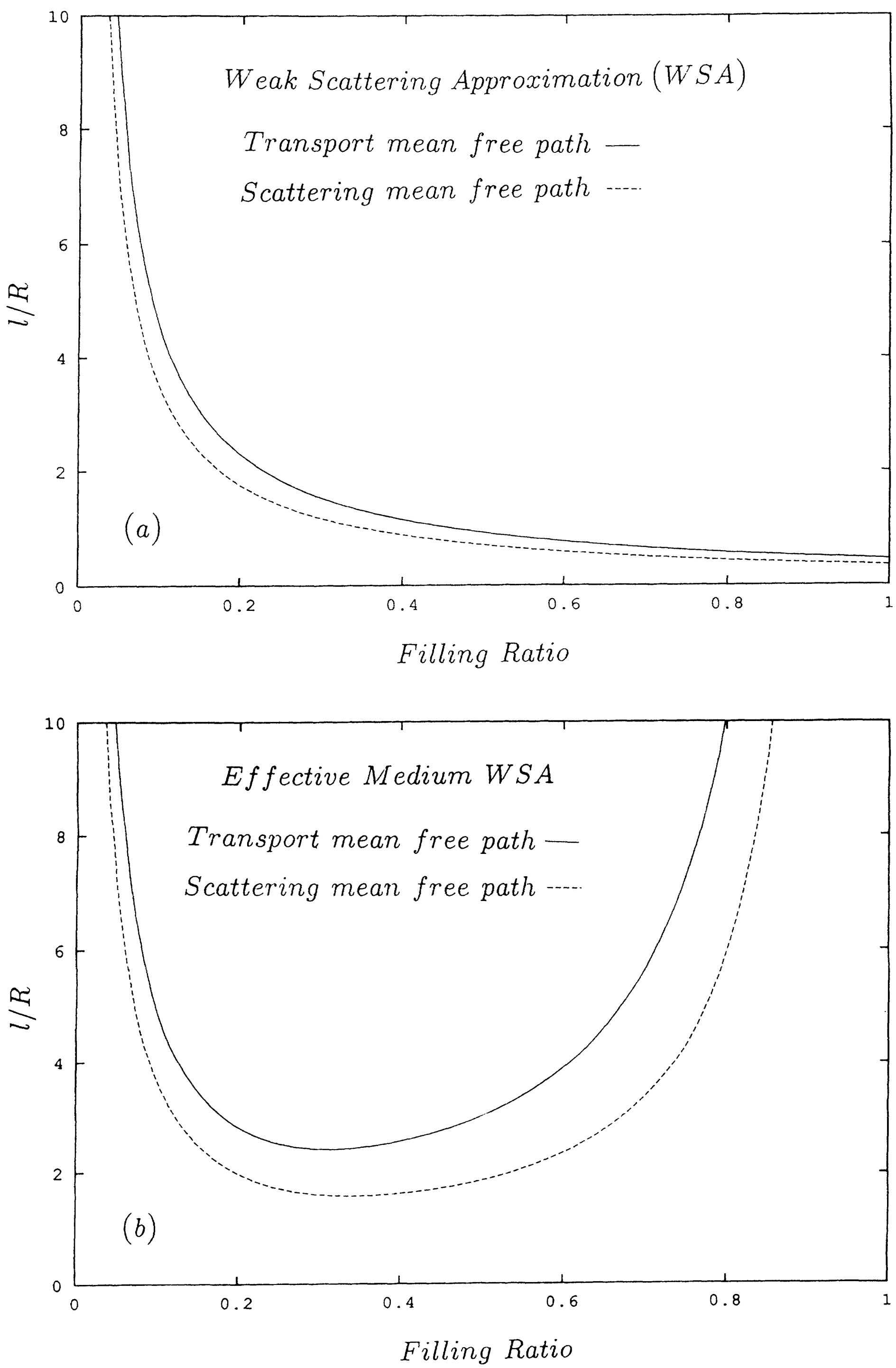

FIG. 1. The scattering mean free path ℓ (dashed line) and the transport mean free path ℓ_t, (solid line), versus the filling ratio f, for TiO_2 particles of dielectric constant ϵ=7.84 with average radius R=110 nm embedded in a host material with dielectric constant equal to 2.03. The mean free path is calculated within the weak-scattering limit (a), within the weak scattering limit (b) with the outside medium having the coated CPA effective dielectric constant, within the regular CPA (c), and within the coated CPA (d). The wavelength of light is λ=514.5 nm.

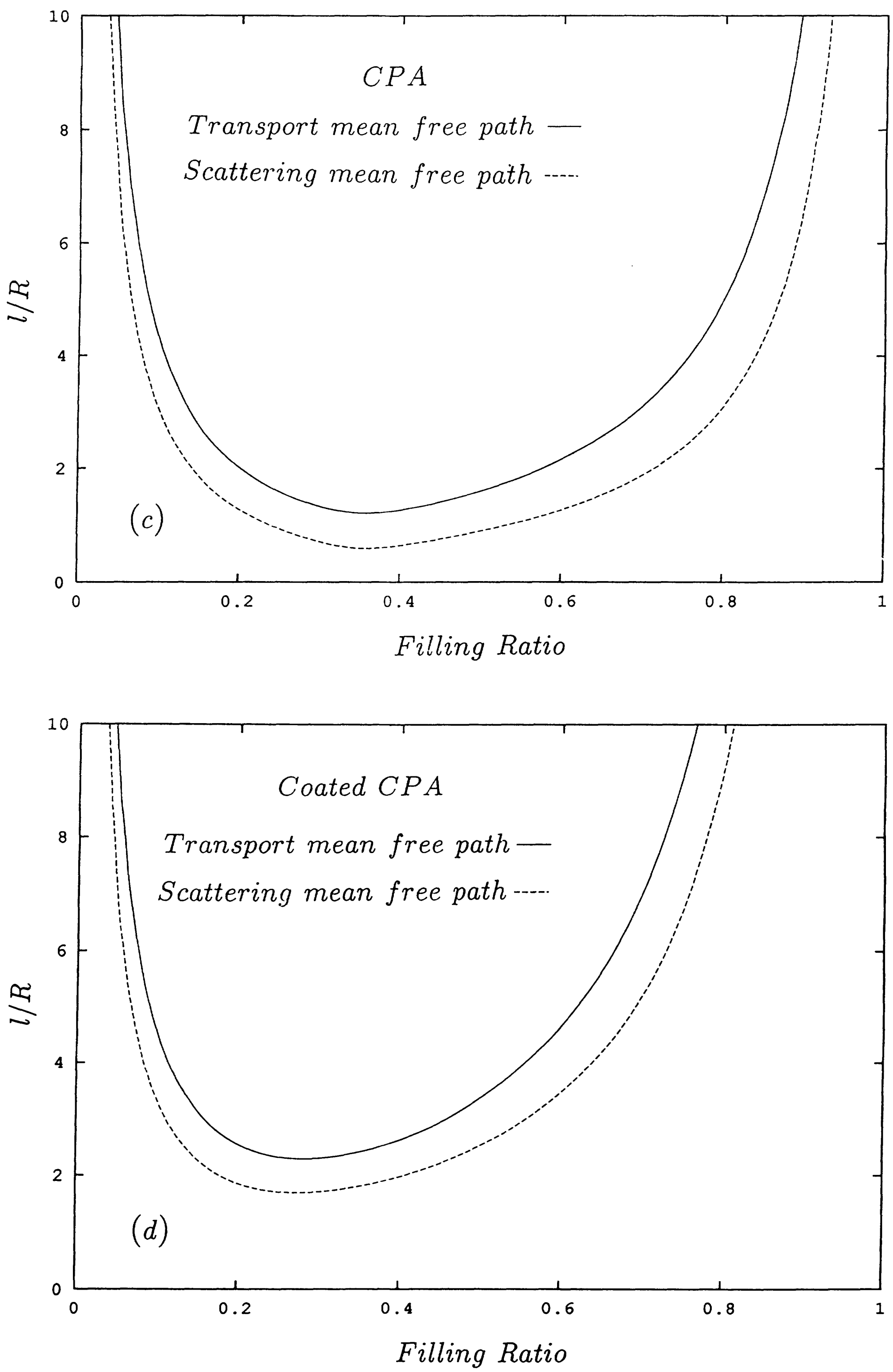

FIG. 1 *(Continued)*.

medium, characterized by an effective complex frequency dependent dielectric constant ϵ_e or equivalently an effective propagation constant q, such that

$$q = \left(\frac{\omega^2}{c^2}\epsilon_e\right)^{1/2} = k + i/2\ell. \quad (1)$$

That quantity q (or ϵ_e) is determined by the condition that the resulting scattering, when a spherical region of the effective medium is replaced by the true random medium, be equal to zero on the average. In the present case where each scatterer has a finite size, the differential cross section requires an infinite number of coefficients for its complete determination, while the effective medium is characterized by only two parameters, namely, k and ℓ. Thus, the question arises which averaged scattering quantity should be set equal to zero. We have decided[4,6] to set the so-called average forward-scattering amplitude $\langle f(0)\rangle = 0$. We have recently considered[4] as basic scattering units a coated sphere of the high dielectric material and a sphere of the host, low dielectric, material. For example, within the coated CPA,[4] one has to satisfy the following condition: $p_1 f_1(0) + p_2 f_2(0) = 0$, where p_1 and $f_1(0)$ are the probability and the forward-scattering amplitude of the coated solid sphere embedded in the effective medium with dielectric constant ϵ_e; p_2 and $f_2(0)$ are corresponding quantities of the host sphere. The forward-scattering amplitudes, as well as the total scattering cross sections σ of either a coated sphere or a host sphere, are given in Ref. 5. To solve the equation $\langle f(0)\rangle=0$, we have transformed[4] it into an iterative equation for the effective propagation constant q [see Eq. (1)]. After a successful convergence of q, which implies $\langle f(0)\rangle=0$, the scattering mean free path $\ell=0.5/\mathrm{Im}(q)$, the renormalization wave vector $k=\mathrm{Re}(q)$, and the effective CPA phase velocity $v_{\mathrm{CPA}} = \omega/k$ are determined. So far, in all the CPA approaches[1,4,6] the scattering mean free path was calculated, and not the transport mean free path ℓ_t. Their relation in the low-concentration limit is $\ell_t\sigma_t = \ell\sigma$, where the total scattering cross sections are given by

$$\sigma = \int d\Omega |f(\theta)|^2, \tag{2a}$$

$$\sigma_t = \int d\Omega |f(\theta)|^2 (1 - \cos\theta), \tag{2b}$$

and $f(\theta)$ is the scattering amplitude. In this manuscript, we have also calculated[7] ℓ_t by introducing the $(1-\cos\theta)$ term before we do the integration over θ and before we perform the CPA average. Before we discuss the results obtained for ℓ and ℓ_t within the coated CPA, we want to give the results for ℓ and ℓ_t in the low-concentration limit. In this limit, each scattering is treated independently from all the others and the scattering mean free path ℓ is given by

$$\ell = 1/n\sigma \ , \tag{3a}$$

$$\ell_t = 1/n\sigma_t \tag{3b}$$

where ℓ and ℓ_t are given in units of radius R of the scattering sphere, n is the number density (i.e., concentration) of the particular type of scatterer, and σ and σ_t are the total scattering cross sections given by Eq. (2). We want to stress again that Eqs. (3) are accurate only in the limit of low scatterer concentration. In this limit the coated CPA results for ℓ and ℓ_t coincide with that given by Eqs. (3). As the concentration f of the scatterers increases, correlations among the scatterers become important and the CPA results are more reliable since they take into account some of the multiscattering effects. One way to improve the results for ℓ and ℓ_t obtained by Eqs. (3) is to employ the mixed coated CPA-LCA approach, i.e., to calculate σ and σ_t from a single scatterer with the outside medium having the coated CPA effective dielectric function ϵ_e which is frequency dependent and not equal to 1. This approach for low f gives ℓ and ℓ_t which completely agree with those given by Eqs. (3) since ϵ_e is very close to the dielectric function of the host material. As f increases ϵ_e becomes larger than 1 and in addition develops some frequency dependence. For high f, this coated CPA-LCA approach gives results in qualitatively agreement with the CPA and experiments. To check the predictions of our theory we compare the results obtained for ℓ and ℓ_t within the coated CPA and within the two versions of the low-concentration approximation (LCA) or weak-scattering approximation (WSA) with two experimental studies.[8,9] The first experimental study[8] involves the multiple scattering of light of wavelength λ=514.5 nm from TiO_2 particles of dielectric constant ϵ=7.84 with average radius R=110 nm for different concentrations f embedded in a host material with a dielectric constant equal to 2.03. In Fig. 1, we present the results for the scattering mean free path ℓ, given by a dotted line, and transport mean free path ℓ_t, given by a solid line versus the filling factor f obtained within the coated CPA, the regular CPA, the WSA, and the mixed WSA, Eq. (3), with the outside medium being the effective medium calculated within the coated CPA (coated CPA-LCA). Notice that all the mean free paths coincide

TABLE I. Experimental and theoretical values for the transport mean free path ℓ_t for TiO_2 particles of dielectric constant ϵ = 7.84 with an average diameter of 220 nm embedded in a host material with dielectric constant equal to 2.03. The wavelength of light is λ = 514.5 nm and the units of ℓ_t are in μm.

Filling ratio	0.05	0.24
Sphere diameter (nm)	220	220
ℓ_t (experimental)	1.00	0.27
ℓ_t (coated CPA)	0.99	0.26
ℓ (coated CPA)	0.74	0.19
ℓ_t (CPA)	0.97	0.18
ℓ (CPA)	0.72	0.11
ℓ_t (WSA)	1.00	0.21
ℓ (WSA)	0.77	0.16
ℓ_t (effective medium WSA)	1.03	0.28
ℓ (effective medium WSA)	0.78	0.19

TABLE II. Experimental and theoretical values for the scattering ℓ and the transport ℓ_t mean free paths for suspensions of 135 nm and 198 nm diameter latex balls of dielectric constant ϵ=2.53 embedded in water. The wavelength of light is λ= 578.5 nm and the units of ℓ and ℓ_t are in μm.

Filling ratio	0.05		0.10		0.15	
Sphere diameter (nm)	198	135	198	135	198	135
ℓ_t (experimental)	46.8	85.9	25.1	51.7	18.1	43.1
ℓ (experimental)	32.7	77.2	19.5	49.4	15.6	43.4
ℓ_t (coated CPA)	38.6	75.6	22.8	48.8	17.9	39.2
ℓ (coated CPA)	32.0	68.6	19.2	45.3	15.3	36.5
ℓ_t (CPA)	34.8	66.6	18.5	36.0	13.1	25.2
ℓ (CPA)	28.3	60.9	15.0	32.9	10.5	23.0
ℓ_t (WSA)	33.2	65.9	16.6	32.9	11.1	22.0
ℓ (WSA)	27.0	60.4	13.5	30.2	9.00	20.1
ℓ_t (effective medium WSA)	36.9	72.5	20.6	39.9	15.5	29.6
ℓ (effective medium WSA)	29.9	66.3	16.7	36.5	12.4	27.0

as one approaches the low-concentration limit f. Notice also that the WSA results for ℓ, given by a dotted line in Fig. 1(a), and ℓ_t, given by a solid line in Fig. 1(a), are monotonically decreased as the concentration f increases and fail completely to describe the high-f regime, as expected. However, these WSA results change their monotonic behavior, if the outside medium is replaced by the coated CPA effective medium. The effective medium scattering mean free path ℓ, given by a dashed line in Fig. 1(b), decreases as f increases up to $f \simeq 0.3$ but then ℓ starts to increase as f is further increased. Similar nonmonotonic behavior is seen for the effective transport mean free path ℓ_t, given by a solid line in Fig. 1(b). The only important difference is that ℓ_t is larger than ℓ for all the concentrations. We also present the coated CPA results for ℓ, given by a dotted line in Fig. 1(d), and ℓ_t, given by a solid line in Fig. 1(d). We again see that indeed the transport mean free path ℓ_t is larger than the scattering mean free path ℓ for almost all f's. In addition, our coated CPA results give values of ℓ_t and ℓ that increase as a function of f, provided that f is higher than 30%. This is a nice feature of the coated CPA which, by taking into account some of the short-range order, predicts that the mean free path will increase drastically as one approaches close packing. For completeness we also present in Fig. 1(c) the results for ℓ and ℓ_t within the regular CPA. Our detailed studies of the different mean free paths suggest that indeed the transport mean free path ℓ_t obtained within the coated CPA is the one that agrees best with the experimental results.[8] From Table I, it is obvious that ℓ_t obtained within the coated CPA agrees extremely well with the experimental results for the two concentrations we have considered, with the coated CPA-WSA coming a close second. To further check the predictions of the coated CPA for ℓ and ℓ_t, we compare it with experiments of Watson *et al.*[9] In their experiment, photon transport parameters (i.e., ℓ and ℓ_t) were obtained for suspensions of 0.135 and 0.198 μm diameter latex balls of dielectric constant ϵ=2.53 with volume fractions f ranging from 0.1% to 20%, embedded in water. In Table II, we compare our theoretical results for ℓ_t and ℓ with the experiments. Notice that ℓ_t obtained within the coated CPA agrees well with the experimentally obtained transport and scattering mean free paths.

Finally, we would like to comment on the validity of the approximate theories we have used. The low-concentration approximation (LCA) or weak-scattering approximation (WSA) results for ℓ and ℓ_t presented in Fig. 1(a) are valid only in the low-concentration limit f ($f < 0.10$). They fail completely to describe the high-f regime, as expected, since they are low-concentration theories. As the concentration f of the scatterers increases, correlations among the scatterers become important and the CPA results are more reliable since they take into account some of the multiscattering effects. It is well known that CPA is an interpolation theory between low and high concentrations and gives reasonable results for a relatively large regime of concentrations. We expect that our coated CPA and CPA results [Figs. 1(c) and 1(d)] to be inaccurate for $f > 0.64$ (the random close-packed limit) and completely inapplicable for $f > 0.74$ (the fcc close-packed limit).

In Fig. 2, we present the results for the phase velocity,[10] the energy transport velocity v_E, given by Eqs. (28) and (29) of Ref. 3, and the coated CPA results for the effective velocity $v_{\rm CPA}$ and the CPA energy transport velocity v'_E. The CPA phase velocity is defined as ω/k [see Eq. (1)]. The CPA energy velocity v'_E is obtained[4] by extending the approach of Ref. 3 for calculating v_E the following way. The coated CPA is used to calculate an effective dielectric function ϵ_e; then the energy transport velocity is calculated with this ϵ_e as the outside medium. The phase velocity $v_{\rm ph}$ is much higher than all the other velocities and in some cases can be

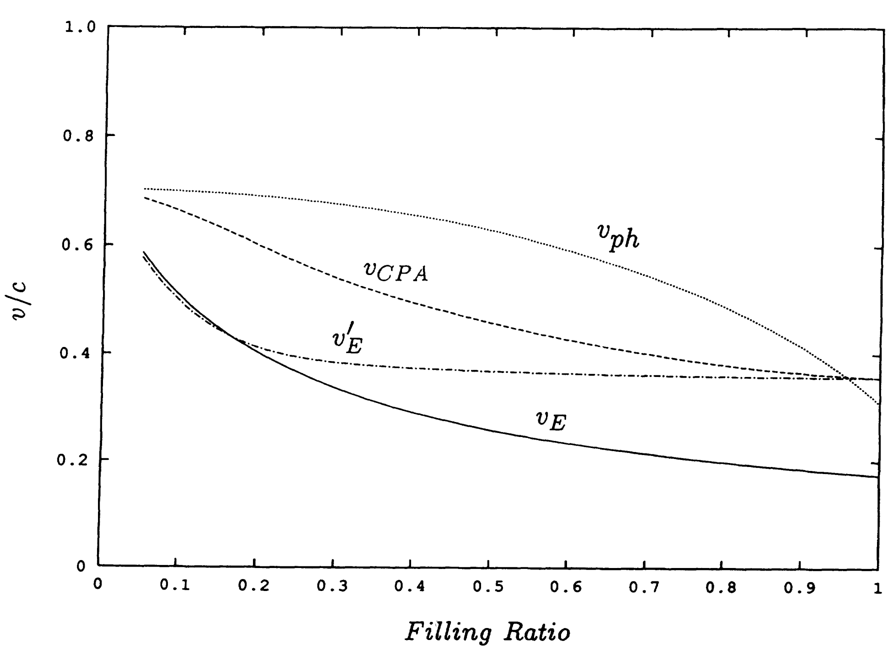

FIG. 2. The phase velocity $v_{\rm ph}$, the energy transport velocity v_E, the coated CPA effective phase velocity $v_{\rm CPA}$, and the CPA energy transport velocity v'_E (with the outside medium having the CPA effective dielectric constant) versus the filling ratio f for the TiO_2 spheres of Fig. 1.

higher[3,4] than the velocity of light, especially close to Mie resonances. However, the energy transport velocity v_E, the CPA phase velocity $v_{\rm CPA}$, and the CPA energy transport velocity v'_E give results which are comparable to each other, for the case we have studied. In general, the CPA energy transport velocity v'_E gives correct results for most of the cases[4] and extrapolates smoothly between low to high concentrations. This is clearly seen in Fig. 2, where $v'_E \simeq v_E$ for low f and $v'_E \simeq v_{\rm CPA}$ for high f.

In conclusion, we have calculated the transport and the scattering mean free path for propagation of classical waves in a random arrangement of dielectric spheres. The transport mean free path calculated within the coated CPA gives results in very good agreement with experiments. Results for ℓ and ℓ_t within the weak-scattering approximation can be greatly improved by using as an outside medium one having the coated CPA effective dielectric constant.

K. Busch acknowledges the financial support of a "DAAD Doktorandenstipendium aus Mitteln des zweiten Hochschulsonderprogramms HSPII/AUFE." We want to thank G. H. Watson for providing us with the experimental results. Ames Laboratory is operated by the U.S. Department of Energy by Iowa State University under Contract No. W-7905-ENG-82. This work was supported by the Director of Energy Research, Office of Basic Energy Sciences and NSF Grant No. INT-9117356. Support by the European Union Grant Nos. SCC CT-90-0020 and ERBCHRXCT930136 is also acknowledged.

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7 In Ref. 5, the different scattering cross sections for scattering by a sphere and coated sphere are given. The scattering cross section σ is given by Eq. (2a) and is equal to $\sigma = (2\pi/k^2)\sum_n (2n+1)(|a_n|^2 + |b_n|^2)$, where a_n and b_n are the scattering coefficients. The extinction cross section $\sigma_{\rm ext} = (2\pi/k^2)\sum_n (2n+1){\rm Re}(a_n + b_n)$, and the asymmetry cross section $\sigma_{\rm asym} = \int d\Omega |f(\theta)|^2 \cos\theta = (2\pi/k^2)\sum_n \left[\frac{2n+1}{n+1}{\rm Re}(a_n b_n^*) + \frac{n(n+2)}{n+1}{\rm Re}(a_n a_{n+1}^* + b_n b_{n+1}^*)\right]$. So σ_t given in Eq. (2b) is equal to $\sigma_t = \sigma - \sigma_{\rm asym}$. When we did our CPA or the coated CPA calculation, we decided to set not $\langle\sigma_t\rangle$=0 but $\langle\sigma_{\rm pr}\rangle$=0, where $\sigma_{\rm pr} = \sigma_{\rm ext} - \sigma_{\rm asym}$.

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