Volume 136, number 1,2 PHYSICS LETTERS A 20 March 1989

INFLUENCE OF INTERNAL REFLECTION ON DIFFUSIVE TRANSPORT IN STRONGLY SCATTERING MEDIA

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Received 23 January 1989; accepted for publication 26 January 1989
Communicated by A.A. Maradudin

We treat the effect of internal reflection of the propagation of waves in strongly scattering media. The theory will be applied to the recently discovered phenomenon of coherent backscattering of light and to the time-dependent transport of light intensity in reflection and in transmission geometries. For backscattering and for transmission through relatively thin slabs the influence of internal reflection can be very strong. For transmission through relatively thick slabs the effect can be accounted for by renormalizing the diffusion coefficient with a length-scale dependent reduction factor.

1. Introduction

At present the interest in multiple scattering of waves in random media has undergone a tremendous revival. One of the reasons for this renewed attention is the possibility of observing Anderson localization in classical systems. More generally one can say that the curiosity concerns the observation of interference effects in multiple scattering [1–3]. The observation of *enhanced backscattering* is one manner to probe the building up of multiple-scattering interference in these media [4–9]. Its connection with localization of light is so intimate that the label *weak localization* was given to it. Nowadays one refers to localization of light as *strong* localization. The study of the enhanced backscattering has become a powerful way of monitoring interference in multiple scattering in great detail [10,11].

The most direct method of observing strong localization seems the experimental determination of the diffusion coefficient. Within the framework of localization the influence of interference will bring about a reduction (and length-scale dependence) of the diffusion coefficient with respect to the bare "Boltzmann" value. The investigation of diffusive transport, either in transmission or in reflection geometries, is a direct means of inferring the diffusion coefficient from experiment [10–14]. Recently time-dependent studies of the enhanced backscattering and depolarization of light has been reported [15].

Another, elegant, way of deducing the diffusion constant from experiment is the measurement of intensity–intensity correlation functions either in the time domain or in the frequency domain [16–18].

In the majority of the novel experiments the interpretation is performed with the help of diffusion theories. Only in the work of van der Mark et al. [10] a rigorous theory beyond the diffusion assumption was developed and worked out in detail. This theory describes static transport only but the results make clear that the diffusion theory with the appropriate situation of trapping planes [13,19,20] constitutes a surprisingly good description. Light is a vector wave and recently a diffusion theory for Rayleigh scattering has been presented [21,22].

In all the models up to now the following restrictive assumption on the boundary condition was adopted: when the light diffuses to the boundary it will escape with probability one. This allows one to use the simplifying reflection principle in the calculation of Green functions [20]. The experimental optimization of the multiple scattering in these types of materials requires the maximization of the contrast in index of refraction in the medium. This results usually in a situation in which the average index of refraction in the medium differs considerably from the index of refraction of the outside world and as such causing internal (and external) reflection at the boundaries. Our estimate is that in strongly scattering media the internal reflection coefficient could be as high as 0.85. This urges the question to what extent does this finite reflectivity influence the dynamics of the propagation.

In this paper we will present a dynamic scalar diffusion theory with incorporation of the effect of internal reflection. We will first give results for the time-dependent Green function for semi-infinite and finite slabs. In addition we will use these Green functions to calculate how much coherent backscattering and diffusive transport, in reflection and transmission, is affected by the existence of internal reflection. In comparison with experiment one should realize that the simplifying assumption has been introduced of a reflectivity independent of angle. This is not true in general but this model will describe the real situation very reliably. This has been checked by using anisotropic reflectivities in computer simulations. For experiments requiring backscattering geometries, like coherent backscattering and time-resolved incoherent backscattering, it turns out that internal reflection seriously modifies the outcome. In transmission experiments the significance of the corrections depends on the thickness of the slab (in units of mean free path) and the influence disappears for very thick slabs. Of course the overall absolute intensities are always seriously affected by finite reflection coefficients. The well-known limits of zero and total internal reflection are easily recovered in our approach.

2. Semi-infinite slab

It is well known that for vanishing or total internal reflection the Dirichlet or the Neumann boundary conditions apply respectively to the Green function obeying a diffusion equation with open boundary conditions [20]. To obtain the intermediate boundary condition is more complex. One expects it to be of the *mixed* type, and one would anticipate the only length scale, the (transport) mean free path λ_{mf}, to play an eminent role in this mixed boundary condition.

The simplest way to derive the boundary condition is to resort to the discrete 1D random walk and take the continuum limit, see for instance ref. [23]. The outcome is the boundary condition

$$G(z_2=0, z_1; t) = \varepsilon\lambda_{mf}\left\{\frac{\partial G(z, z_1; t)}{\partial_z}\right\}_{z=0}, \tag{1}$$

where $G(z_2, z_1; t)$ represents the Green function obeying the 1D diffusion equation in the semi-infinite medium ($z>0$), and where $\varepsilon \equiv R/(1-R) \equiv R/T$ describes the internal reflection at the $z=0^+$ boundary with reflectivity R.

The method of calculating the Green function is the standard expansion in eigenfunctions [23]. For the particular case of a semi-infinite slab the Green function can be obtained in closed form and, under the condition that $G(z_2, z_1; t=0) = \delta(z_2-z_1)\delta(t)$, is given by

$$\begin{aligned} G(z_2, z_1; t) = {} & \frac{1}{\sqrt{4\pi Dt}}\{\exp[-(z_2-z_1)^2/4Dt] \\ & + \exp[-(z_2+z_1)^2/4Dt]\} \\ & -(1/\varepsilon\lambda_{mf})\exp(\beta^2)[1-\mathrm{erf}(\beta)] \\ & \times \exp[-(z_2+z_1)^2/4Dt], \end{aligned} \tag{2}$$

in which

$$\beta \equiv (1/\varepsilon\lambda_{mf})\sqrt{Dt} + \frac{z_2+z_1}{2\sqrt{Dt}}, \tag{3}$$

where D is the diffusion coefficient and erf denotes the error function [24]. As can easily be checked in two limits $R=0$ and $R=1$, in which case the Green function will be denoted by $G^0(z_2, z_1; t)$, the "image" results are reproduced:

$$G^0(z_2, z_1; t) = G(z_2-z_1; t) \pm G(z_2+z_1; t), \tag{4}$$

where

$$G(z; t) = \frac{1}{\sqrt{4\pi Dt}}\exp(-z^2/4Dt) \tag{5}$$

represents the Green function for the infinite medium. The plus sign in eq. (4) corresponds to total internal reflection and the minus sign to vanishing internal reflection. From eq. (2) one concludes that, unless $R=1$, the long-time behavior will be proportional to $t^{-3/2}$. It is not straightforward to describe the $R \neq 0$ Green function in terms of the $R=0$ Green function with a renormalized diffusion constant. It will turn out that in the description of incoherent light transport in reflection an important quantity will be $G(\lambda_{mf}, \lambda_{mf}; t)$. Analysis of the long-time behavior of eq. (2) demonstrates that for this particular quantity the diffusion coefficient can be renormalized as

$$D^* \approx \frac{1}{(1+\varepsilon)^{4/3}} D . \tag{6}$$

The apparent reduction of the actual diffusion coefficient mimics the longer stay of the light in the sample due to reflection.

3. Finite slab

The slab will be situated between $0<z<L$. The boundary condition, as given by expression (1), has to be applied twice, at $z=0^+$ and (with opposite sign!) at $z=L^-$. The result is an eigenvalue equation for the allowed wave numbers:

$$\tan(kL) = \frac{2\varepsilon\lambda_{mf}k}{(\varepsilon\lambda_{mf}k)^2-1} . \tag{7}$$

The normalized eigenvectors are given by

$$\Psi_k(z) = A_k^{-1/2} \sin(kz+\eta_k) , \tag{8}$$

$$\tan \eta_k = \varepsilon\lambda_{mf}k , \tag{9}$$

$$A_k = \tfrac{1}{2}L - \frac{1}{2k}\cos(kL+2\eta_k)\sin(kL) . \tag{10}$$

Using an eigenfunction expansion the Green function $G(z_2, z_1; t)$ is given by

$$G(z_2, z_1, t) = \sum_k \Psi_k(z_2)\Psi_k(z_1)\exp(-Dk^2t). \tag{11}$$

The solution (11) can easily be determined numerically. Care should be taken to use at short times many eigenvectors to ensure convergence.

From summation (11) it can easily be deduced that the long-time behavior is determined by the lowest eigenvalues and the decay will eventually be described with a single exponential, corresponding to the lowest eigenvalue. It is practical to develop a perturbation expression for these eigenvalues to get an idea of the importance of the effect of internal reflection. In lowest order the result for the set of wave numbers $\{k\}$ is given by

$$k \approx \frac{n\pi}{L}\left(1-\frac{2\varepsilon\lambda_{mf}}{L}\right) . \tag{12}$$

This already indicates that a part of the influence of a finite reflectivity could be incorporated by assuming $R=0$ and renormalizing the diffusion coefficient by

$$D^* \approx D\left(1-\frac{4\varepsilon\lambda_{mf}}{L}\right) . \tag{13}$$

For the case of $R=1$ or $R=0$ the eigenvectors are known and D is independent of the slab thickness. In that case the dynamic Green functions can be presented as a simple series:

$$G^0(z_2, z_1; t) = \sum_{q=0}^{\infty} \frac{2}{L}\sin(q\pi z_2/L)\sin(q\pi z_1/L) \times \exp(-\pi^2 Dq^2t/L^2) . \tag{14}$$

This series applies to the $R=0$ case. For $R=1$ the same solution applies when replacing the sine functions by cosine functions. For numerical purpose it might be useful to apply the Poisson summation rule to this result and transform to an expansion useful for short times:

$$G^0(z_2, z_1; t) = \frac{1}{\sqrt{4\pi Dt}} \times \sum_{n=-\infty}^{\infty} \{\exp[-(z_2-z_1-2nL)^2/4Dt] \pm \exp(-(z_2+z_1-2nL)^2/4Dt]\} . \tag{15}$$

When one compares this result with eq. (4) the occurrence of multiple images is clearly demonstrated. Interestingly enough it is apparently not possible to derive the mixed situation with the method of images.

At this point it is important to pay attention to the validity of the diffusion equation. The rigorous theory describing incoherent scalar wave transport for $R=0$ is the Milne equation [10,13,19]. Applying the diffusion approximation to the Milne equation not only gives the correct diffusion result but in addition it allows optimization of the boundary conditions in the diffusion framework so as to mimic the rigorous result optimally. From this procedure one finds the presence of trapping planes not coinciding with the exact boundary but located slightly outside the physical boundary. This can be allowed for by introducing the "apparent" width $L^* \equiv L + 2z_0\lambda_{\rm mf}$. This boundary condition improves the quality of the diffusion approximation considerably. One should realize that the location of the trapping plane should depend on R. For $R=0$ the Milne equation gives $z_0 \approx 0.71$ [19,20]. At $R=1$ one expects the reflection plane to coincide with the physical boundary. The quantitative results presented in this paper are only slightly influenced by a different location of the trapping planes.

4. Enhanced backscattering

Enhanced backscattering is the phenomenon that, no matter how many multiple-scattering events are participating, at exactly the backscattering direction there will be constructive interference between time-reversed events in random media. This interference will diminish at directions away from the backscatter directions (finite perpendicular momentum transfer) and the width is determined by the mean free path, $\lambda_{\rm mf}$, and the wavelength, λ.

To describe weak localization the Green function should allow for perpendicular momentum transfer. The relevant 3D Green function for enhanced backscattering [9,10,25]

$$G(z_2, z_1, \boldsymbol{k}_\perp; t) = G(z_2, z_1; t)\exp(-Dk_\perp^2 t), \qquad (16)$$

in which $\boldsymbol{k}_\perp$ is the perpendicular momentum transfer. For the present purpose it is sufficient to limit the discussion to increased backscattering for a semi-infinite slab. Using eq. (2) and introducing trapping planes results after integrating eq. (16) over time in

$$G(z_2, z_1, \boldsymbol{k}_\perp) = \frac{1}{2Dk_\perp} \times \left(\frac{\varepsilon\lambda_{\rm mf}k_\perp - 1}{\varepsilon\lambda_{\rm mf}k_\perp + 1} \exp(-k_\perp |z_2 + z_1 + 2z_0|) + \exp(-k_\perp |z_2 - z_1|) \right). \qquad (17)$$

This equation reproduces the correct $R=1$ and $R=0$ results.

To obtain the intensity for the enhanced backscattering one has to sum the contributions of time-reversed events (most-crossed or cyclical diagrams) and one has to account for the incoming and outgoing waves. This has been treated in detail in the recent literature [9,10,21,22,25]. The result for the bistatic coherent scattering coefficient related to enhanced backscattering [12] is given by

$$\gamma_{\rm c}(\mu_{\rm i}, \mu_{\rm s}) = (1-R)(1-\tilde{R})\frac{c}{\lambda_{\rm mf}^2} \times \frac{1}{\mu_{\rm i}} \int_0^\infty {\rm d}z_2 \int_0^\infty {\rm d}z_1\, G(z_2, z_1, \boldsymbol{k}_\perp) \times \cos[k_0(\mu_{\rm i} - \mu_{\rm s})(z_2 - z_1)] \times \exp\left[-\frac{1}{2\lambda_{\rm mf}} \left(\frac{1}{\mu_{\rm s}} + \frac{1}{\mu_{\rm i}} \right)(z_2 + z_1) \right], \qquad (18)$$

in which c is the speed of light, $k_0 \equiv 2\pi/\lambda$, and $\mu_{\rm i}$, $\mu_{\rm s}$ are the cosines of respectively the incoming direction $\theta_{\rm i}$ and outgoing (scattered) direction $\theta_{\rm s}$, and $\tilde{R}$ is the (external) reflectivity for incoming waves. Substituting eq. (17) in this expression leads after some algebra to the following result, assuming $\theta_{\rm i}=0$ and $\theta_{\rm s} \equiv \theta$:

$$\gamma_{\rm c}(1, \mu_{\rm s}) = (1-R)(1-\tilde{R})\frac{3}{2\alpha v}[(\alpha+v)^2 + u^2]^{-1} \times \left(\alpha + v[1 - \exp(-2\alpha z_0)] + \frac{2\alpha v}{\varepsilon + \alpha}\exp(-2\alpha z_0) \right),$$

$$v \equiv \frac{1}{2}\left(1 + \frac{1}{\cos\theta}\right), \quad u \equiv k_0\lambda_{\rm mf}(1-\cos\theta),$$

$$\alpha \equiv k_0\lambda_{\rm mf}|\sin\theta| = k_\perp\lambda_{\rm mf}. \qquad (19)$$

In fig. 1 we show the intensity as a function of angle around the backscattering direction ("backscatter cones") for varying internal reflection. In fig. 2 the dependence of the width (fwhm) of the backscatter cone is given as a function of the reflectivity. To show the mild dependence on the location of the trapping plane we have presented the results for two different locations of the trapping plane ($z_0=0.71$, appropriate for $R=0$ and $z_0=0$, appropriate for $R=1$). All the results show a dramatic influence on the presence of internal reflection.

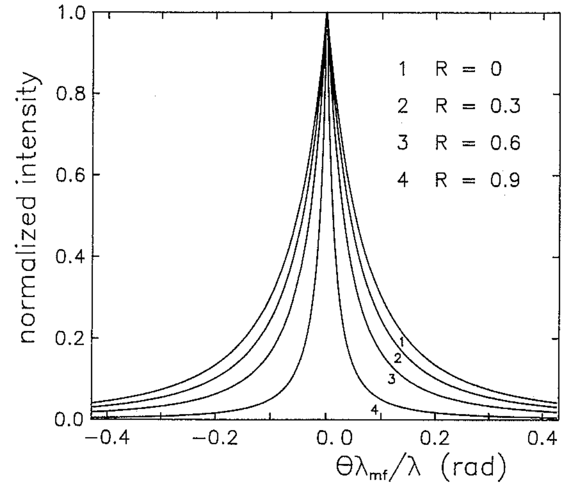

Fig. 1. Computed angular dependence of the (normalized) enhanced backscattering intensity for various values of the internal reflectivity.

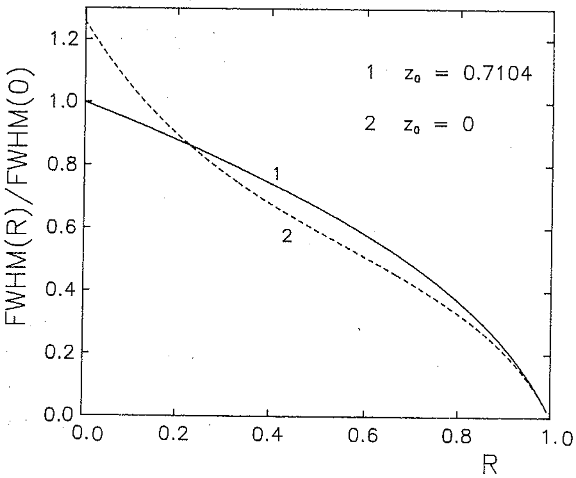

Fig. 2. Computed width (fwhm) of the enhanced backscattering as a function of the internal reflectivity R. The fwhm for $R=0$ is $\approx 0.7\lambda/2\pi\lambda_{\text{mf}}$ [10].

The fact that the cone becomes narrower on increasing R can easily be understood. The influence of finite reflectivity is to force the light paths in the sample to be longer. But longer light paths get out of phase easier and consequently the backscattering cone becomes narrower.

5. Transport in reflection

The description of time-dependent incoherent transport ('ladder diagrams') in reflection from a semi-infinite slab can easily be done within the framework of the diffusion theory. The spreading of an initial delta-function pulse is easily shown to be given by

$$\gamma_{\ell}(\mu_{\text{i}}, \mu_{\text{s}}; t) = \frac{c}{\mu_{\text{i}}\lambda_{\text{mf}}^2}(1-R)(1-\tilde{R}) \times \int_0^\infty \mathrm{d}z_2 \int_0^\infty \mathrm{d}z_1\, G(z_2, z_1, \boldsymbol{k}_\perp = 0; t) \times \exp\left(-\frac{z_1}{\mu_{\text{i}}\lambda_{\text{mf}}} - \frac{z_2}{\mu_{\text{s}}\lambda_{\text{mf}}}\right), \tag{20}$$

in which the time-dependent propagation of the incoming wave has been neglected. Akkermans et al. [9] have shown that the double integral in this equation can be approximated by

$$\lambda_{\text{mf}}^2 G(\lambda_{\text{mf}}, \lambda_{\text{mf}}, \boldsymbol{k}_\perp = 0; t) \approx \lambda_{\text{mf}}^2 (1 + 1/\varepsilon)^2 G(0, 0, \boldsymbol{k}_\perp = 0; t)\,. \tag{21}$$

The expression in the r.h.s. of eq. (21) was obtained by using a Taylor expansion. The introduction of these simplifications is not absolutely necessary but as we have checked that the more rigorous theory does not give rise to important corrections we will stick to these appealing simplifications.

In fig. 3 we display several of our results for the time-dependent incoherent backscattering intensity (spreading in time of an incoming wave) for various values of the internal reflection coefficient. Clearly can be seen that a finite reflectivity induces a much stronger (relative) intensity at longer times. The $R \neq 0$ results can be fit numerically into $R=0$ theory results over the whole time domain by introducing a renormalization of the diffusion coefficient. In table 1 we give the results obtained with this overall fit and compare them with the results obtained from perturbation theory (see eq. (6)), supposed to be valid for long times. The apparent reduction of the diffusion constant can easily be understood. The effect of internal reflection is to keep the light longer in the medium. In an $R=0$ theory this can only be achieved by reducing the diffusion coefficient.

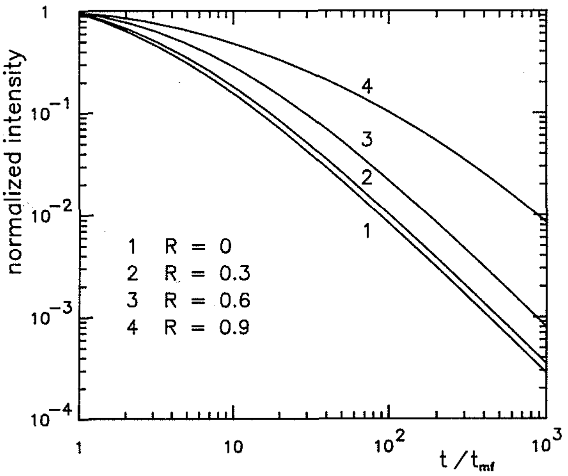

Fig. 3. Calculated dynamic pulse profile in reflection from a semi-infinite slab for various values of the internal reflection coefficient. The instantaneous response at $t=0$ should be convoluted in practice with the incoming pulse profile. Time is in units of $t_{mf} \equiv \lambda_{mf}/c$.

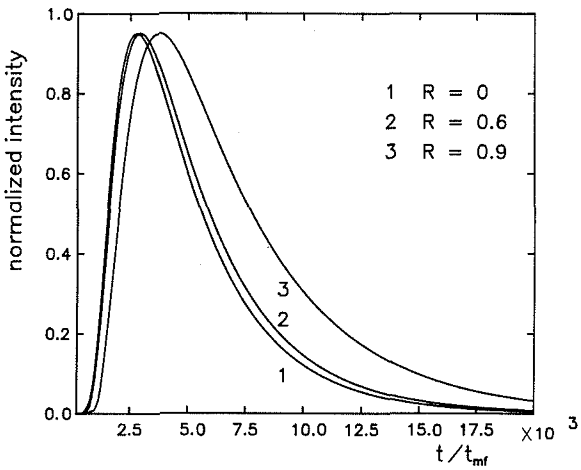

Fig. 4. Calculated dynamic pulse profile in transmission for various values of the internal reflectivity. Time is in units of $t_{mf} \equiv \lambda_{mf}/c$.

Table 1
Renormalization of diffusion constant in reflection.

L/λ_{mf}	R	D^*/D [a]	D^*/D [b]
100	0	1	1
100	0.3	0.8	0.62
100	0.6	0.3	0.30
100	0.9	0.07	0.046
500	0	1	1
500	0.3	0.8	0.62
500	0.6	0.3	0.30
500	0.9	0.07	0.046

[a] Numerical fit.
[b] Perturbation (see eq. (6)).

6. Transmission

Transmission experiments are very important as it is to be expected that the experimental evidence for localization will come from this type of experiments. The canonical experiment is to propagate a short pulse through the medium and to measure the spreading of the transmitted pulse in the time domain. The diffusion coefficient can be inferred from this experiment. The neglect of finite internal reflection could lead to serious errors in the determination of the actual diffusion coefficient and as such could lead to wrong conclusions about the proximity of the Anderson transition. For this reason understanding of the present effect is crucial. Having the Green functions at our disposal the transmitted intensity is easily shown to be given by

$$\begin{aligned}\gamma_{\ell}(1,-1;t)&\\ &\approx c(1-R)(1-\tilde{R})G(\lambda_{mf}, L-\lambda_{mf}, \boldsymbol{k}_{\perp}=0;t)\\ &\approx c(1-R)(1-\tilde{R})(1+1/\varepsilon)^2 G(0,L,\boldsymbol{k}_{\perp}=0;t)\,.\end{aligned} \tag{22}$$

This equation is derived under the same conditions as those of eq. (21). In fig. 4 we display several of our results for the time-dependent incoherent transmitted intensity (spreading in time of an incoming wave) for various values of the internal reflection coefficient. Clearly can be seen that a finite reflectivity induces a much stronger (relative) intensity at longer times. The $R\neq 0$ results can be fit into $R=0$ results by introducing a renormalization of the diffusion coefficient. In table 2 we give the results which compare favorably with the results obtained from perturbation theory (see eq. (13)).

7. Absorption

Up to now we have neglected the presence of absorption. The inclusion in the theory is rather trivial. In the time-dependent version one only includes an

Table 2
Renormalization of diffusion constant in transmission.

L/λ_{mf}	R	D^*/D [a]	D^*/D [b]
100	0	1	1
100	0.3	0.98	0.98
100	0.6	0.94	0.94
100	0.9	0.74	0.64
500	0	1	1
500	0.3	1.00	1.00
500	0.6	0.99	0.99
500	0.9	0.93	0.93

[a] Numerical fit.
[b] Perturbation (see eq. (13)).

exponential damping factor $\exp(-t/\tau_a)$, containing the inelastic time τ_a. In the static Green function the result can easily be modified as to include absorption. One can replace α in the appropriate equations by

$$(\alpha^2+\lambda_{mf}^2/D\tau_a)^{1/2}\equiv[\alpha^2+3(1-a)]^{1/2},$$

in which a is the albedo. The fitting of our results by introducing reduced diffusion coefficients has shown that these results are only slightly influenced by the presence of absorption, although the overall curve is of course strongly influenced by the presence of absorption.

8. Consequences

The theory presented in this paper leads to important corrections to earlier approaches when sizeable internal reflection occurs. The occurrence of internal reflection can be circumvented by matching of index of refraction. Experiments with liquid samples usually involves the use of transparent glass windows in order to contain the sample and let the light through. For these experiments the corrections for internal reflection is probably not important. On the other hand experiments with strongly scattering solid samples, like titania, can be seriously affected by the effect of internal reflection. For these materials the interpretation of static coherent backscattering experiments and of measurements with pulses both in backscattering [15] and in transmission through thin slabs (for an estimate of how thin see eq. (13)) should be performed with the consideration of influence of internal reflection. Also in other experiments where reflection geometries are used or where transmission through relatively thin samples is studied, including fluctuation experiments, the influence of internal reflection could be sizeable.

9. Conclusion

In this paper we have shown that in experiments where transport of waves in strongly scattering media is studied the presence of finite internal reflectivity can be very important. We have calculated all the relevant Green functions and have computed the influence for typical experimental conditions. The result is effectively a reduction of the diffusion coefficient. In the case of transmission this reduction depends on the thickness of the slab and thus leads to a (trivial) length-scale dependence of the diffusion constant. With the theory presented in this paper other experimental situations can easily be considered as the numerical calculations are straightforward.

Acknowledgement

This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (Foundation for the Fundamental Research on Matter) and was made possible by financial support of the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organization for the Advancement of Research). We would like to thank Roos Eisma, Adriaan Tip, Meint van Albada and Martin van der Mark for valuable discussions.

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