Rigorous Treatment of the Speed of Diffusing Classical Waves

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Rigorous Treatment of the Speed of Diffusing Classical Waves.

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Abstract. – We present exact expressions for the transport velocity of scalar classical waves in random dielectric media in lowest order of the density of the scatterers (Boltzmann limit). This speed enters into the diffusion constant of multiply scattered classical waves. We discuss the relation with the Wigner phase delay time.

The interest in multiple scattering has undergone a dramatic revival in recent years. The main reason for this renewed interest is the discovery that, in contrast to one's intuition, dramatic interference effects can be traced even for waves that have been scattered thousands of times. This idea forms the basis of fascinating interference phenomena like weak localization and intensity fluctuations. Many of these discoveries were stimulated by the close analogy between the propagation of classical waves such as light on the one hand, and the propagation of Schrödinger particles (electrons) on the other. Novel advancements in condensed-matter research turned out to be of much wider applicability and could easily be related to fields like optics.

We have recently pointed out a fundamental difference between the multiple scattering of classical waves and electrons [1]. This difference is already present at the Boltzmann level and may have dramatic influence beyond this low-density limit. We showed that the velocity appearing in the diffusion constant for light is lowered by collisional contributions. Most surprisingly this effect is absent when Schrödinger particles scatter from potentials, the standard situation for electron impurity scattering [2]. This extremely important result, derived from a microscopic transport equation for classical waves and supported by heuristic models, was confirmed by experiments.

In our microscopic theory one well-controlled approximation regarding off-shell contributions of the scattering amplitude was introduced and was adopted in order to deal with the so-called «logarithmic derivative» in the vector Helmholtz equation for the electric-field vector. Given the consequences of these results and the doubts expressed by other workers it seems necessary to present a highly relevant case in which no approximation

whatsoever has to be invoked. The present paper shows that our original result for the energy velocity derived for vector waves is completely rigorous for scalar classical waves (*e.g.* acoustic waves). By giving apparently different but mathematically equivalent representations for the classical transport velocity, we wish to clarify the physical origin of the delay effect.

The confusion about the delay effects for classical waves was initiated by the *absence* of these effects for multiple scattering of Schrödinger particles. However, a well-known difference exists between spinless classical waves and spinless electrons at the level of the equations of motion. Contrary to scattering of Schrödinger waves from potentials, scattering of scalar waves from dielectric particles can be characterized by an «energy-dependent» potential (sometimes called an optical potential [3]). A comparison of the time-independent Schrödinger equation with potential $V(\boldsymbol{r})$ and energy $\mathcal{E}$ and the scalar-wave equation with dielectric constant $\varepsilon(\boldsymbol{r})$ at frequency E suggests to identify a «potential» $V(\boldsymbol{r}, E) = [1 - \varepsilon(\boldsymbol{r})]\, g(E)$ and an «energy» $\mathcal{E} = E^2$ ($\varepsilon_0 = c_0^2 = 1$). The energy dependence $g(E) = E^2$ is due to the second-order time derivative in the scalar-wave equation, in combination with the multiplicative nature of the dielectric interaction. Its presence signifies a conservation law different from Schrödinger scattering.

The consequences of an energy-dependent potential on the microscopic theory of multiple scattering have been given special attention lately. A Ward Identity (WI) is a diagrammatic formulation of a conservation law. A WI for classical waves different from the conventional electron impurity WI [2] was incorporated by both refs. [1, 4] and [5], but the conclusions do not seem to agree. Knowledge of the correct conservation law c.q. WI is essential for the definition of the transport velocity v_E. This transport velocity should appear in the diffusion constant $D_E = (1/3) v_E l$ for classical waves, and as such becomes subject to direct experimental verification. If the conventional (electron impurity) WI applied, the transport speed of light v_E at frequency E would have equalled the reciprocal phase velocity $k_E/E = 1/v_\mathrm{p}$, being the classical counterpart of the (shifted by disorder) Fermi velocity $\hbar k_\mathrm{F}/m_\mathrm{e}$.

However, the expression $v_E = 1/v_\mathrm{p}$ was never used. Instead, on the basis of physical intuition, one always believed the phase velocity v_p to be the speed in the diffusion constant of classical waves. Our theory predicts the transport velocity v_E to be (approximately) equal to the phase velocity v_p *only* away from resonances. In the regime of resonant scattering both the transport velocity and the Boltzmann diffusion constant become an order of magnitude smaller than what was usually taken for granted, the amount of reduction being proportional to the number density of the scatterers.

A WI signifies the presence of a conserved quantity in an indirect way. Let us first circumvent the debate on which WI to use for classical waves and give a derivation of v_E by looking *directly* at the conserved quantity. For scalar classical waves the energy density before averaging is $W = (1/2)\varepsilon(\boldsymbol{r})|\partial_t \psi|^2 + (1/2)|\nabla\psi|^2$ [6], with corresponding «Poynting vector» $\boldsymbol{S} = -\mathrm{Re}(\partial_t \psi)(\nabla\psi)^*$. After averaging over realizations and cycles, both terms in W represent an equal amount of energy. For monochromatic modes $\psi \sim \psi_E \exp[-iEt]$ we arrive at

$$\langle W_E \rangle = E^2 \left\{ \langle |\psi_E|^2 \rangle + \langle [\varepsilon(\boldsymbol{r}) - 1] |\psi_E|^2 \rangle \right\}, \tag{1a}$$

$$\langle \boldsymbol{S}_E \rangle = E \boldsymbol{k}_E \langle |\psi_E|^2 \rangle = \hat{\boldsymbol{k}} \frac{E^2}{v_\mathrm{p}} \langle |\psi_E|^2 \rangle . \tag{1b}$$

The brackets denote ensemble averaging in the thermodynamic limit, but can (on the basis of ergodicity) be replaced by space averaging. The second term in $\langle W_E \rangle$ is non-zero only inside

the scatterers and is therefore at least first order in density. Since it is *not* present for Schrödinger scattering, this term gives concrete form to the different conserved quantity mentioned earlier. The current S_E is essentially identical to the Schrödinger current. We used $k_E \approx E/v_p$ with v_p given by Sheng and Zhang [7] to describe the impact of disorder on this current. By normalizing ψ_E we equate the first term in eq. (1*a*) within braces to unity, and the second to $\Delta(E)$. Hence $\langle W_E \rangle = E^2[1 + \Delta(E)]$. The quantity $E^2\Delta(E)$ represents the «potential energy» inside the scatterers. In the case of elastic light scattering from systems with internal degrees of freedom (*e.g.* the radiating dipole) a similar parameter shows up [8, 9] and equals the energy of internally excited (vibration) states of the oscillators. *By definition* the transport velocity is given by [9]

$$v_E \equiv \frac{\langle S_E \rangle}{\langle W_E \rangle} = \frac{1}{v_p} \frac{1}{1 + \Delta(E)} . \tag{2}$$

The second equality makes use of eqs. (1*a*) and (1*b*). The quantity $\Delta(E)$ can be calculated into lowest order of the number density $n = N/V$ of the scatterers by inserting for ψ_E the normalized eigenfunction $\psi_E^+ / \sqrt{V}$ of one single scatterer S. We get

$$\Delta(E) = n \int_S \mathrm{d}\boldsymbol{r}\, [\varepsilon(\boldsymbol{r}) - 1] |\psi_E^+(\boldsymbol{r})|^2 + \mathcal{O}(n^2) . \tag{3}$$

We observe that *always* $\Delta \geq 0$ for $\varepsilon(\boldsymbol{r}) \geq 1$. In all known cases this guarantees that the causality requirement $v_E < c_0$ is obeyed. The phase velocity v_p as well as the group velocity v_g is often seen to be larger than c_0 near resonances [8]. The group velocity can be argued to describe the transport of the *coherent* wave $|\langle \psi_E \rangle|^2$, and emerges from the averaged amplitude. Near resonances is $|\psi_E^+| \gg 1$ inside the scatterers so that $1 + \Delta$ is much larger than the effective-medium dielectric constant $\langle \varepsilon \rangle$.

We now prove that our original, microscopic derivation of v_E is in complete agreement with the result in eq. (3). In ref. [1, 4] we indicated that the proper transport equation is obtained when an expansion in the frequency domain is carried out. In this approach one defines the quantity

$$\frac{\delta_1(E)}{n} \equiv - \frac{\partial \operatorname{Re} t_{pp}(E)}{\partial E^2} + \int \mathrm{d}\Omega \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \frac{\partial \phi}{\partial E} - \frac{4\pi}{E} \int \frac{\mathrm{d}^3\boldsymbol{p}}{(2\pi)^3} \operatorname{Im} t_{pp}(E) \frac{\partial \operatorname{Re} G_p^0(E)}{\partial E^2} . \tag{4}$$

All derivatives are to be performed at constant momenta, $G_p^0(E) = (E^2 - p^2 + i0)^{-1}$, the free-space Green's function and $t_{pp}(E)$, the off-shell (*i.e.* $E^2 \neq p^2$) t-matrix. In terms of δ_1 the transport velocity is now given by $v_E v_p = 1/[1 + \delta_1]$. By a WI is $\delta_1 = 0$ for Schrödinger potential scattering [2]. For classical waves $\delta_1 \neq 0$ by the presence of the energy-dependent potential. In our original derivation the third term in eq. (4) has not been invoked. In ref. [5] it was argued that this term makes the resonance effect disappear. However, since the derivative in this third term does not suffer from the extra energy dependence of the potential, it is already part of the cancellation theorem for Schrödinger waves and on balance does not contribute to δ_1.

Given definitions (3) and (4), we now prove the «Ward Identity» $\Delta - \delta_1 = 0$ for scalar waves. To this end, we first write for the potential $V(\boldsymbol{r}, E) = V_0 f(\boldsymbol{r}) g(E)$, and use the identity

$$\frac{\partial}{\partial E^2} = \frac{\partial}{\partial E^2}\bigg|_g + \frac{V_0}{g} \frac{\mathrm{d}g}{\mathrm{d}E^2} \times \frac{\partial}{\partial V_0} . \tag{5}$$

Applying the WI for energy-independent potentials at energy E^2 (which then states that $\delta_1 = 0$), the derivatives at constant g and momenta $\boldsymbol{p}$ must cancel in eq. (4). We obtain

$$\frac{\delta_1(E)}{n} = \frac{1}{E^2}\frac{\mathrm{d}g}{\mathrm{d}E^2} \times \left\{ \frac{V_0 E^2}{g} \left[- \frac{\partial \operatorname{Re} t_{pp}(E)}{\partial V_0} + 2E \int \mathrm{d}\Omega \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \frac{\partial \phi}{\partial V_0} \right] \right\}. \tag{6}$$

We now have to establish the equivalence of $\delta_1(E)$ as obtained in eq. (6) to the parameter $\Delta(E)$ found earlier in eq. (3). The second factor between curly brackets in the r.h.s. of (6) is the (3D generalization of the) derivative of the phase shift with respect to the potential, which is the definition of the «dwell time» of a wave in the scattering region [10]. It is known from literature [10, 11] that the integral of the square of the eigenfunction over the *scatterer region* is an equivalent definition of dwell time. This implies that eqs. (3) and (6) coincide perfectly, which is our desired result. The exact algebraic equivalence has been established elsewhere [11, 12].

It is tempting to associate δ_1 to the Wigner phase delay time. The relation of the dwell time c.q. δ_1 to the Wigner phase delay time becomes evident if one realizes that the latter is known to be expressible by the same integral over *whole of the space* [13]. Near resonances of scattering, $|\psi_E^+|^2$ is sharply confined within the scatterer, so that the dwell time is the dominating part in the phase delay time. Therefore, as far as classical waves are concerned, identifying δ_1 with the Wigner phase delay time is numerically accurate on resonances [1].

It may come as a surprise that neither the Wigner time nor the dwell time enter into the transport velocity of electrons in the presence of impurities, where particle conservation guarantees unambiguously that $\Delta(E)$ vanishes, a fact which can be expressed in a Ward identity for electron impurity scattering. Associating $\delta_1(E)$ or $\Delta(E)$ with either Wigner time or dwell time is in this case again tempting but false, since neither of these times vanish.

In ref. [5] a very interesting formula for $\delta_1(E)$ is found. However, the final outcome of the calculation $\delta_1(E) = -n \operatorname{Re} t(E)/E^2$, so that $v_E = v_\mathrm{p}$, is not positive-definite as it should be on the basis of eq. (3), and must therefore be wrong. In view of eq. (1a) this result is based on an unjustified decoupling of the average into the (amplitude-)averaged fluctuation of $(\varepsilon(\boldsymbol{r}) - 1)$ times the averaged intensity. This procedure neglects correlations between both quantities, that are manifestly present. With regard to eq. (6) one has apparently assumed that the extra energy dependence of the t-matrix enters in a linear way, which is another way of ignoring correlations. Only in the Born approximation (the Rayleigh limit for classical waves) and for some non-resonant point scatterer models [14], the identity $\delta_1(E) = -n \operatorname{Re} t(E)/E^2$ can be established. As a matter of fact, it is the on-shell (far-field) assumption of ref. [5] that makes the resonant effect disappear. As is evident from eq. (3), the delay effect is caused by the off-shell field (1).

In the rest of this paper we will clarify two additional details. First we discuss the physical meaning of the third term in eq. (4). Next we obtain a third convenient expression for the «classical defect» δ_1.

The first term of eq. (4) represents the contribution of the group velocity, the second term is a collision contribution. As we will point out now, the third term is a correction to the group velocity. We will calculate the third term within the «Wigner» approximation employed in our previous papers. By definition, this approximation is obtained if the $\boldsymbol{p}$-dependence of all

(1) In the final expression for δ_1 in ref. [5] an (off-shell) principal-value integration is missed. The WI is nevertheless correct.

t-matrices in eq. (4) is ignored. We set $t_{pp}(E) = t(E)$. Such a procedure replaces finite-size scatterers by point scatterers with same on-shell ($E^2 = p^2$) t-matrix. This third term now becomes equal to zero. One can generalize the third term beyond the Boltzmann approximation, a difficult regime in which many corrections to the first two terms have to be incorporated as well. The impact of the generalized third term in second order of the density can be obtained when G_p^0 is replaced by its averaged counterpart $G_p = [E^2 - p^2 - nt(E)]^{-1}$ and equals $\mathrm{d}/\mathrm{d}E^2[1/4l_s^2)$, where $l_s = 1/n\sigma$ is the scattering mean free path. This, in turn, can be recognized as a correction to the *group velocity*. By solving the complex dispersion law $(k + i/2l_s)^2 = E^2 - nt(E)$, we get $k^2(E) = E^2 - n\,\mathrm{Re}\,t(E) + 1/4l_s^2$. The group velocity is then

$$v_G \equiv \frac{\mathrm{d}E}{\mathrm{d}k} = \frac{1}{v_p}\left(\frac{\mathrm{d}k^2}{\mathrm{d}E^2}\right)^{-1} = \frac{1}{v_p}\left[1 - n\frac{\mathrm{d}\,\mathrm{Re}\,t(E)}{\mathrm{d}E^2} + \frac{\mathrm{d}}{\mathrm{d}E^2}\frac{1}{4l_s^2}\right]^{-1}. \tag{7}$$

The extra term in the group velocity coincides with the third term mentioned above. This notion fits nicely into the picture of ref. [1, 9] that the transport velocity can be decomposed into the group velocity and a collision contribution.

The most elegant, and for numerical purposes most convenient representation for $\delta_1(E)$ emerges after applying the Cauchy-Riemann equations to eq. (6) [12]. This yields $\delta_1(E)$ as a derivative of the absorption cross-section with respect to the *imaginary* part of the potential,

$$\delta_1(E) = n\frac{V_0 E}{g}\frac{\mathrm{d}g}{\mathrm{d}E^2}\frac{\mathrm{d}\sigma_{\mathrm{abs}}}{\mathrm{d}(\mathrm{Im}\,V_0)}, \qquad \mathrm{Im}\,V_0 \to 0. \tag{8}$$

For a medium filled at random with scalar Mie spheres (radius r_0, index of refraction $m + im_i$ and size parameter $x = Er_0$) this reduces to

$$\delta_1^{\mathrm{Mie}}(E) = f \times \frac{3}{8}\frac{m^2 - 1}{mx}\lim_{m_i \to 0}\frac{Q_{\mathrm{abs}}}{m_i}, \tag{9}$$

f being the packing fraction. The right-hand side is proportional to the path length of the wave in one individual Mie sphere. In ref. [4] we presented this formula as a successful heuristic model. It is, quite satisfactorily, seen to be an exact solution for scalar waves. In fig. 1 we compare this exact solution to the Wigner time approximation employed in

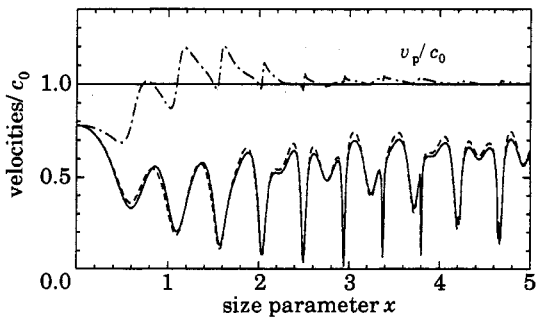

Fig. 1. – Exact solution for the transport velocity (bold line) and the Wigner approximation (dashed line) for a collection of scalar Mie spheres with volume fraction $f = 10\%$ and index of refraction $m = 2.73$. The phase velocity is shown as a dash-dotted curve.

ref. [1, 4]. The figure demonstrates that this Wigner approximation (dashed curve) and the exact solution (bold curve) practically coincide, proving that the p-dependence of t-matrices in eq. (4) is insignificant indeed. The phase velocity is seen to be larger than c_0 near resonances. In the regime of resonant multiple scattering the transport velocity becomes very small.

Our theory of the speed of light in dielectric media is consistent with the approach for radiating dipoles [9]. The exact similarity between both cases has been described elsewhere [15]. The smallness of the transport velocity has been confirmed by several independent light scattering experiments [1, 16], and is also known to occur in resonant spin systems [17].

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REFERENCES

[1] VAN ALBADA M. P., VAN TIGGELEN B. A., LAGENDIJK A. and TIP A., *Phys. Rev. Lett.*, **66** (1991) 3132.
[2] MAHAN G. D., *Many Particle Physics* (New York, Plenum) 1981, p. 617.
[3] JOACHAIN C. J., *Quantum Collision Theory* (North-Holland, Amsterdam) 1975.
[4] VAN TIGGELEN B. A., LAGENDIJK A., VAN ALBADA M. P. and TIP A., *Phys. Rev. B*, **45** (1992) 12233.
[5] BARABANENKOV YU. N. and OZRIN V. D., *Phys. Rev. Lett.*, **69** (1992) 1364; *Phys. Lett. A*, **154** (1991) 38.
[6] MORSE P. M. and FESHBACH H., *Methods of Theoretical Physics* (McGraw-Hill, New York, N.Y.) 1953, p. 304.
[7] SHENG P. and ZHANG Z. Q., *Phys. Rev. Lett.*, **57** (1986) 1879.
[8] LOUDON R., *The Quantum Theory of Light*, first edition (Oxford University Press, Oxford) 1973, p. 61.
[9] LOUDON R., *J. Phys. A*, **3** (1970) 233.
[10] BÜTTIKER M., in *Electronic Properties of Multilayers and Low Dimensional Semiconductor Structures*, edited by J. M. CHAMBERLAIN *et al.* (Plenum Press, New York, N.Y.) 1990.
[11] MARTIN PH. and SASSOLI DE BIANCHI M., *J. Phys. A*, **25** (1992) 3627.
[12] VAN TIGGELEN B. A., TIP A. and LAGENDIJK A., *J. Phys. A*, **26** (1993) 1731.
[13] JAUCH J. M. , SINHA K. B. and MISRA B. N., *Helv. Phys. Acta*, **45** (1972) 398.
[14] KROHA J., SOUKOULIS C. M. and WÖLFLE P., *Phys. Rev. B*, **47** (1993) 11093.
[15] NIEUWENHUIZEN TH. M., LAGENDIJK A. and VAN TIGGELEN B. A., *Phys. Lett. A*, **169** (1992) 191.
[16] DE BOER J. F. , VAN ALBADA M. P. and LAGENDIJK A., *Phys. Rev. B*, **45** (1992) 658; ERBACHER F. A., LANKE R. and MARET G., *Europhys. Lett.*, **21** (1993) 551; GENACK A. Z. , *Phys. Rev. B*, **46** (1992) 14475.
[17] SHIREN N. S., *Phys. Rev.*, **128** (1962) 2103.