J. Phys. I 1 (1991) 471-491 AVRIL **1991**, PAGE 471

Classification
Physics Abstracts
42.40 — 05.40

Dynamical correlations for multiple light scattering in laminar flow

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(Received 16 October 1990, revised 20 December 1990, accepted 3 January 1991)

Résumé. — Nous présentons une étude théorique des corrélations temporelles de la lumière en régime de diffusion multiple dans un milieu désordonné. Nous considérons la situation où le fluide contenant des diffuseurs élastiques est en écoulement laminaire et stationnaire. Nous établissons une formulation générale du déphasage de la lumière diffusée en fonction du déplacement des diffuseurs dans la limite de faible diffusion $k\ell \gg 1$. Ces résultats sont appliqués aux écoulements de cisaillement et de Poiseuille aussi bien qu'à l'écoulement bouchon des fluides non newtoniens. Enfin, la validité des approximations utilisées et la possibilité d'extension de ces résultats sont discutées.

Abstract. — A theoretical study of time dependent correlation functions of the multiply scattered light in a disordered medium is presented. We consider the situation where the fluid containing the elastic scatterers is submitted to a laminar and stationary flow. We establish a general formulation for the phase variation of scattered light originating from the motion of the scatterers within the weak scattering limit $k\ell \gg 1$. These results are applied to shear and Poiseuille flow as well as to plug flow of non-Newtonian fluids. The validity of the approximations used and possible extension of these results are discussed.

1. Introduction.

The last years have seen a strong revival of the interest in the problem of the scattering of light by turbid media. New methods of investigation of the multiple scattering regime were developed which gave rise to the prediction and observation of qualitatively new phenomena. Among them, was the enhancement by a factor of two of the backscattered light within a narrow cone around the direction of the incident light [1]. More recently, it was realized [2, 3]

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that it is also possible to get new insights about the time dependent correlation function of the scattered intensity in the strongly multiple scattering regime in which the standard QELS (Quasi Elastic Light Scattering) spectroscopy does not apply anymore. It was then possible to study the dynamics of Brownian particles in dense solution at very short time scales. This « Diffusing Wave Spectroscopy » [4] was subsequently applied to study the dynamics of scatterers submitted to deterministic flow [5].

Our aim in this article is to discuss from a theoretical point of view the dynamical correlation function of the intensity for a large class of deterministic flow within different geometries. In section 2, we will give the general framework in which all these multiple scattering calculations are done. Section 3 is devoted to general behaviour of the phase of the correlation function for deterministic motions. Then, in section 4, we will apply these results to the case of shear and Poiseuille flow as well as to the case of non-Newtonian fluids flowing in a slab. Part of these results concerning the time correlation of the shear flow has already been given in a previous publication [5]. In this paper, we present a general approach of the problem of the time correlation of the multiple scattered intensity and extend the previous results to more complex situations such as Poiseuille or laminar flow of non-Newtonian fluids. We restrict ourselves to the geometry where the correlation is measured through a slab by reflection or transmission.

2. General expression of the time correlation function in the multiple scattering regime.

Our aim in this section is to study the behaviour of the multiply scattered field and then to derive a convenient expression for both the time dependent correlation function of the field and the intensity.

Suppose that the incident electric field at point $\mathbf{r}_1$ is $\mathbf{E}_i(r_1)$ and the subsequent multiply scattered field at $\mathbf{r}_2$ due to that source is $\mathbf{E}_s(r_1, r_2)$. In order to calculate $\mathbf{E}_s$, let us consider first a single scattering event on a particle of volume v described by a local fluctuation $\delta\varepsilon$ of the dielectric tensor. The scattered electric induction $\mathbf{D}_s$ satisfies the differential equation [6] :

$$\nabla^2 \mathbf{D}_s + k^2 \mathbf{D}_s = - \mathbf{rot\ rot}\ (\delta\varepsilon \cdot \mathbf{E}_i) \tag{1}$$

where $k = \dfrac{\omega}{c} = \dfrac{2\pi}{\lambda}$. The solution of equation (1) at large distance R_0 is :

$$\mathbf{D}_s = \frac{1}{4\pi}\ \mathbf{rot\ rot}\ \frac{\exp(ikR_0)}{R_0} \int_v \mathrm{d}^3 r\ \delta\varepsilon \cdot \mathbf{E}_i \exp\{- i\mathbf{k}' \cdot \mathbf{r}\} \tag{2}$$

where $\hat{\mathbf{k}}'$ is the unit vector which indicates the direction of scattered field. We suppose now, for the sake of simplicity, that $\delta\varepsilon$ is spherical and we consider an incident plane wave described by $\mathbf{E}_i = \hat{\mathbf{e}}_i E_0 \exp\{i\mathbf{k} \cdot \mathbf{r}\}$, where $\hat{\mathbf{e}}_i$ is the incident unit vector. In the far field, the relation between $\mathbf{D}_s$ and $\mathbf{E}_s$ becomes simply $\mathbf{D}_s = \varepsilon \mathbf{E}_s$ so that equation (2) gives :

$$\mathbf{E}_s = - E_0 \frac{\delta\varepsilon \cdot \exp(ikR_0)}{4\pi\varepsilon R_0} \int_v \mathrm{d}^3 r\ \hat{\mathbf{k}}' \wedge (\hat{\mathbf{k}}' \wedge \hat{\mathbf{e}}) \exp\{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{r}\}\ . \tag{3}$$

If the volume v of the scatterer is of order λ^3, then the stationary phase approximation gives :

$$\mathbf{E}_s \simeq - E_0 \frac{v\delta\varepsilon \cdot \exp(ikR_0)}{4\pi\varepsilon R_0}\ \hat{\mathbf{k}}' \wedge (\hat{\mathbf{k}}' \wedge \hat{\mathbf{e}}) \exp\{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{r}\} \tag{4}$$

where **r** defines now the position of the scatterer. Equation (4) is made of two parts. One is the phase factor $\exp\{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}\}$ which describes the change of the phase due to the scattering. The second part which is also a complex number but does not depend on direction, is nothing but the asymptotic expression of the Green function $G(k, R_0)$.

Suppose we have now a lot of such elastic scatterers in our system at points $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n$. This kind of dense solution is usually characterized by its elastic mean free path ℓ which describes the average distance between two scattering events. Usually ℓ is much larger than the average distance between scatterers. We now consider the weakly scattering regime for which $\lambda \ll \ell$. Then the distance travelled by the scattered field being very large, we can use the asymptotic expression (4) as the incident field on the forthcoming scatterer. For a given sequence $\mathcal{C}_n$ of, say, n scatterings at $\mathbf{r}_0, \mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_{n-2}$, $\mathbf{r}$ between $\mathbf{r}_0$ and $\mathbf{r}$, the scattered field can be written :

$$\mathbf{E}_s = \mathcal{A}(r_0, r, \mathcal{C}_n)\left[\prod_{i=2}^{n}\exp\{-i(\mathbf{k}_i-\mathbf{k}_{i-1})\cdot\mathbf{r}_{i-1}-i(\mathbf{k}_1-\mathbf{k})\cdot\mathbf{r}_0-i(\mathbf{k}'-\mathbf{k}_n)\cdot\mathbf{r}\}\right] \quad (5)$$

where $\mathcal{A}(r_0, r, \mathcal{C}_n)$ is the complex amplitude of the field associated with the sequence $\mathcal{C}_n$. The total electric field at point **r** is then obtained by summing over all the multiple scattering paths $\mathcal{C}_n$. This derivation can be extended to the more general case of moving scatterers $\mathbf{r}_i(t)$ so that the scattered field $\mathbf{E}_s$ can be written :

$$\mathbf{E}_s(r_0, r, t) = \sum_{\mathcal{C}_n}\mathcal{A}(r_0, r, \mathcal{C}_n(t))\, e^{i\delta_n(t)} \quad (6)$$

where the bracketed term in equation (5) defines a phase factor $\delta_n(t)$. Moreover, let us assume that the displacements $\mathbf{r}_i(t)$ are small enough in order to neglect the second order variation of the wavevectors **k**. The time correlation function of the field is therefore :

$$\langle E(r_0, r, t)\, E^*(r_0, r, 0)\rangle = \sum_{\mathcal{C}_n, \mathcal{C}_{n'}}\left\langle \mathcal{A}(r_0, r, \mathcal{C}_n(t))\, \mathcal{A}^*(r_0, r, \mathcal{C}_{n'}(0))\, e^{i(\delta_n(t)-\delta_{n'}(0))}\right\rangle . \quad (7)$$

The set of multiple scattering paths $\mathcal{C}_n(t)$ at time t coincides with the set $\mathcal{C}_{n'}(0)$ at time $t = 0$ (ergodic hypothesis). Then the amplitudes $\mathcal{A}$ are now stationary and independent of the phases $\delta_n(t)$. Moreover, the calculation of

$$\sum_{\mathcal{C}_n, \mathcal{C}_{n'}}\langle \mathcal{A}(r_0, r, \mathcal{C}_n)\, \mathcal{A}^*(r_0, r, \mathcal{C}_{n'})\rangle$$

can be done within the multiple scattering theory [7] and gives within the weak scattering limit $k\ell \gg 1$, the Green function $G(r_0, r) = [4\pi D|r_0 - r|]^{-1}$ of the stationary diffusion equation in 3-dimensions. This Green function has a very convenient expansion [7] in terms of random walk paths of length n : $G(r_0, r) = \sum_n G(r_0, r, n)$. $G(r_0, r, n)$ is the Green function of the time dependent diffusion equation where the role of the time t is played by the number n of steps of random walk. Equation (7) can then be written :

$$\langle E(r_0, r, t)\, E^*(r_0, r, 0)\rangle = \sum_n G(r_0, r, n)\left\langle e^{i(\delta_n(t)-\delta_n(0))}\right\rangle . \quad (8)$$

The $\langle \ldots \rangle$ which appears in equation (8) describes an average over all the diffusion paths of length n and eventually an average over a possibly random motion [2] of scatterers

$r_i(t)$. Finally, to compare with experimental results, we have to relate the field correlation function given by equation (8) to the correlation function $C_2(r_0, r, t) = \langle |E(r_0, r, t)|^2 \, |E(r_0, r, 0)|^2 \rangle$ of the intensities. In the weak scattering limit, one can show [3] that :

$$C_2(r_0, r, t) = |\langle E(r_0, r, t) \, E^*(r_0, r, 0) \rangle|^2 \tag{9}$$

then C_2 reduces to the calculation of equation (8). We clearly see now that the time correlation function of the field needs two basic ingredients to be calculated. One is the Green function $G(r_0, r, n)$ which essentially describes effects related to the geometry of the system. The second is $\langle \exp(i \, \Delta\phi_n(t)) \rangle$; $(\Delta\phi_n(t) = \delta_n(t) - \delta_n(0))$ which depends essentially upon the motion of the scatterers.

3. General expression of the phase for deterministic motions in the stationary regime.

We concentrate in this part on the general features of the phase $\langle \exp(i \, \Delta\phi_n(t)) \rangle$. One knows that :

$$\langle \exp(i \, \Delta\phi_n(t)) \rangle = \exp\left(\sum_{q=1}^{\infty} i^q M_q \right) \tag{10}$$

where M_q is the q^{th} cumulant :

$$M_1 = \langle \Delta\phi_n(t) \rangle \; ; \quad M_2 = \frac{1}{2} \left[\langle \Delta\phi_n^2(t) \rangle - \langle \Delta\phi_n(t) \rangle^2 \right] ; \ldots$$

Let us now consider the stationary flow [8]. The change of position $\mathbf{\Delta r}(\mathbf{R}_\nu, t)$ of the ν^{th} scatterer with time can therefore be written :

$$\mathbf{\Delta r}(\mathbf{R}_\nu, t) = \mathbf{V}(\mathbf{R}_\nu) \, t \tag{11}$$

where $\mathbf{V}$ is the local velocity. The position of the ν^{th} scatterer is defined as follows : $\mathbf{R}_\nu = \sum_{i=1}^{\nu-1} \Lambda_i(0) \, \hat{\mathbf{e}}_i$; $(\nu > 1)$, where $\hat{\mathbf{e}}_i$ is the emergent unit vector after i^{th} scattering and $\Lambda_i(0) \equiv |\mathbf{R}_{i+1} - \mathbf{R}_i|$. $\mathbf{R}_\nu$ is therefore defined by the variables of previous $(\nu - 1)$ scatterings.

From equations (5) and (6), the phase $\Delta\phi_n(t)$ associated with a given multiple scattering path $\mathbf{r}_0, \mathbf{r}_1, \ldots, \mathbf{r}_{n-2}, \mathbf{r}$ can be written :

$$\Delta\phi_n(t) = \sum_{\nu=1}^{n} \mathbf{k}_\nu \cdot [\mathbf{\Delta r}(\mathbf{R}_\nu, t) - \mathbf{\Delta r}(\mathbf{R}_{\nu+1}, t)] \tag{12}$$

where we have neglected the phase variation associated with the first and the last scattering events. According to equation (11), we can write equation (12) as :

$$\Delta\phi_n(t) = kt \sum_{\nu=1}^{n} \hat{\mathbf{e}}_\nu \cdot [\mathbf{V}(\mathbf{R}_\nu) - \mathbf{V}(\mathbf{R}_{\nu+1})] \tag{13}$$

where $\hat{\mathbf{e}}_\nu$ is the emergent unit vector after the ν^{th} scattering. Note that decorrelation only occurs through the relative motion of particles. In the particular case of uniform flow, no decorrelation occurs. The positions $\mathbf{R}_\nu$ and $\mathbf{R}_{\nu+1}$ of the ν^{th} and the $(\nu + 1)^{\text{th}}$ scatterers respectively are separated on average by a distance of the order of the elastic mean free path. For the velocity field which varies slowly at length scale ℓ i.e. $\|\nabla V\|/\|\Delta V\| > \ell$ we can then expand to first order the bracket in equation (13) :

$$\Delta\phi_n(t) \simeq - kt \sum_{\nu=1}^{n} \Lambda_\nu(0)\ (\hat{\mathbf{e}}_\nu \cdot \nabla)(\hat{\mathbf{e}}_\nu \cdot \mathbf{V}(\mathbf{R}_\nu)) . \tag{14}$$

By introducing the symmetric tensor $A_{q\ell}(\nu) = (\hat{\mathbf{e}}_\nu \cdot \hat{\mathbf{e}}_q)(\hat{\mathbf{e}}_\nu \cdot \hat{\mathbf{e}}_\ell)$ with $q, l = x, y, z$; the phase $\Delta\phi_n(t)$ can now be expressed as :

$$\Delta\phi_n(t) = - kt \sum_{\nu=1}^{n} \Lambda_\nu(0)\ A^{q\ell}(\nu)\ (\partial_x V)_{q\ell} . \tag{15}$$

he average value $\langle \Delta\phi_n(t) \rangle$ over all the possible paths of length n is proportional to $\langle \Lambda_\nu(0)\ A^{q\ell}(\nu)(\partial_x V)_{q\ell} \rangle$. This average can be factorized since $A^{q\ell}(\nu)$ depends only on the angles at ν^{th} scattering ; $\langle A_{q\ell}(\nu) \rangle = \frac{1}{3}\delta_{q\ell}$ while $\langle \Lambda_\nu(0) \rangle = \ell$ by definition. $(\partial_x V)_{q\ell}$ depends only on the variables of the $(\nu - 1)^{\text{th}}$ scattering. Therefore :

$$\langle \Delta\phi_n(t) \rangle = - \frac{1}{3} k\ell t \sum_{\nu=1}^{n} \langle \operatorname{div} \mathbf{V}(\mathbf{R}_\nu) \rangle . \tag{16}$$

The variation of the phase is then zero on average for incompressible fluids or when $(\partial_x V)_{xx} = (\partial_x V)_{yy} = (\partial_x V)_{zz} = 0$. This is the situation we will consider throughout this article. Consider now $\Delta\phi_\nu(t) = - kt\ \Lambda_\nu(0)\ A^{q\ell}(\nu)(\partial_x V)_{q\ell}$. The correlation function of the elementary phase is :

$$\langle \Delta\phi_\nu\, \Delta\phi_{\nu'} \rangle = (kt)^2 \langle \Lambda_\nu(0)\ \Lambda_{\nu'}(0) \rangle \langle A^{ij}(\nu)\ A^{q\ell}(\nu') \rangle \langle (\partial_x V)_{ij}\ (\partial_x V)_{q\ell} \rangle . \tag{17}$$

But $\langle A^{ij}(\nu)\ A^{q\ell}(\nu') \rangle \propto \delta_{\nu\nu'}\,\delta_{iq}\,\delta_{j\ell}$. Then the phases $\Delta\phi_\nu$ are independent random variables of zero average so that the central limit theorem applies.

In addition to this motion imposed by the fluid, the scatterers can also experience a Brownian motion. The time correlation function associated with it was recently extensively studied [2]. Here we are mainly interested in the deterministic motion of the scatterers. This imposes to work at time scales such that the relative displacement of two scatterers is larger than their Brownian displacement. This corresponds to $t \gg D_B/(\Gamma_{\text{typ}}\,\ell)^2$ where D_B is the Brownian diffusion constant, Γ_{typ} a typical velocity gradient of flow and ℓ the elastic mean free path of light. In recent experiment [5] the characteristic values of the parameters are : $D_B \simeq 10^{-8}\ \text{cm}^2.\text{s}^{-1}$, $\Gamma_{\text{typ}} \simeq 50\ \text{s}^{-1}$ and $\ell \simeq 100\ \mu\text{m}$. Then the deterministic regime corresponds to the time range $t > 40$ ns. In the following, we will adopt these values for estimating the various time scales.

4. Laminar flow in slab.

We consider a slab of width L perpendicular to the z-axis, within which flows a non compressible fluid with a distribution of velocities :

$$\mathbf{V}(\mathbf{R}) = V_z(x)\ \hat{\mathbf{e}}_z \tag{18}$$

$\mathbf{V}(\mathbf{R})$ obeys the Navier-Stokes equation with specified boundary conditions. According to equation (17), the fluctuation of the phase is :

$$\langle \Delta\phi_n^2(t) \rangle = k^2 \ell^2 t^2 \sum_{\nu=1}^{n} \left\langle (A^{xz}(\nu))^2 \left(\frac{\partial V_z}{\partial x} \right)^2 \right\rangle . \tag{19}$$

Since $\dfrac{\partial V_z}{\partial x}$ does not depend on $\hat{\mathbf{e}}_\nu$, we can factorize in equation (19) the angular coordinates, and calculate $\langle (A^{xz}(\nu))^2 \rangle = \langle \cos^2(\theta_\nu) \sin^2(\theta_\nu) \cos^2(\psi_\nu) \rangle = \dfrac{1}{15}$ — for the isotropic diffusion — so that :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{1}{15} k^2 \ell^2 t^2 \sum_{\nu=1}^{n} \left\langle \left(\frac{\partial V_z}{\partial x} \right)_\nu^2 \right\rangle . \tag{20}$$

Within the diffusion approximation considered here, we can write :

$$\left\langle \left(\frac{\partial V_z}{\partial x} \right)_\nu^2 \right\rangle = \int_v P_{n,\nu}(x) \left(\frac{\partial V_z}{\partial x} \right)^2 \mathrm{d}x \tag{21}$$

where $P_{n,\nu}(x)$ is the probability density to reach the point x after ν steps in a random walk involving n steps without touching the boundaries. The calculation of $P_{n,\nu}(x)$ is very reminiscent of the related statistical mechanics of polymers as considered long ago by Edwards [9] and de Gennes [10]. We will therefore only give the main lines of the calculation in Appendix B. Then, if we know the Green function $G(r_0, r, n)$ of the diffusion equation and the density $P_{n,\nu}(x)$, the set of equations (8), (20) and (21) defines completely the field and the intensity correlation functions. We will now apply it to the study of some characteristic flow.

4.1 SHEAR FLOW. — The expression of the velocity in that case is $\mathbf{V}(\mathbf{R}) = \dfrac{V_{\max}}{L} x\hat{\mathbf{e}}_z$ so that $\dfrac{\partial V_z}{\partial x} = \dfrac{V_{\max}}{L} \equiv \Gamma$ is constant. From equation (20) we obtain for the fluctuation of the phase

$$\langle \Delta\phi_n^2(t) \rangle = 2 \left(\frac{t}{\tau_s} \right)^2 n \tag{22}$$

where we have defined $\tau_s^{-1} = \dfrac{1}{\sqrt{30}} k\ell\Gamma$. $\tau_s(\simeq 70\ \mu\text{s})$ [5] the characteristic time needed by a pair of scatterers initially distant from ℓ to move a relative distance λ due to a shear flow of time constant Γ^{-1}. τ_s is then the decorrelation time and for $t > \tau_s$ the field correlation function involves only a single scattering event. To enter the multiple scattering regime, we must therefore consider times $t < \tau_s$. From equation (8) we obtain for the field correlation function :

$$\langle E(r_0, r, t)\, E^*(r_0, r, 0) \rangle = \sum_{n=1}^{\infty} G(r_0, r, n) \exp\left\{ -n \left(\frac{t}{\tau_s} \right)^2 \right\} \tag{23}$$

and for a slab of width L with the two points r_0 and r inside the medium, we can evaluate the Green function $G(r_0, r, n)$ (cf. appendix A) so that :

$$\langle E(r_0, r, t) E^*(r_0, r, 0)\rangle = \frac{1}{\sqrt{3}} \frac{\tau_s}{t} \frac{\sinh\left[\sqrt{3}\frac{L}{\ell}\frac{t}{\tau_s}\left(1-\frac{x}{L}\right)\right]\sinh\left[\sqrt{3}\frac{x_0}{\ell}\frac{t}{\tau_s}\right]}{\sinh\left[\sqrt{3}\frac{L}{\ell}\frac{t}{\tau_s}\right]} \tag{24}$$

where x_0 and x are respectively the projections of r_0 and r on the x-axis. In these relations, r_0 describes the point where the incident light coming from the source is impinging onto the system, then [7] we will consider $x_0 \simeq \ell$. Moreover, in the limit $t \to 0$, we find $\langle |E(r_0, r)|^2\rangle = 1 - \frac{x}{L}$ which, indeed, coincides with the reflection coefficient for $x = \gamma\ell$ and with the transmission coefficient for $x = L - \gamma\ell$. The coefficient γ introduced above depends on the boundary conditions we have chosen for the diffusion equation [7] ; but it is always of the order of unity.

We are now in a position to calculate the normalized time correlation function both in reflection and in transmission. The reflected part $C_1^R(t)$ is obtained from equation (24), taking $x = \gamma\ell$:

$$C_1^R(t) = \frac{1}{\left(1-\frac{\gamma\ell}{L}\right)} \frac{\sinh\left[\sqrt{3}\frac{L}{\ell}\frac{t}{\tau_s}\left(1-\frac{\gamma\ell}{L}\right)\right]}{\sinh\left[\sqrt{3}\frac{L}{\ell}\frac{t}{\tau_s}\right]}. \tag{25}$$

In the limit $L \to \infty$ describing a semi-infinite system, we obtain for $t < \tau_s$:

$$C_1^R(t) = \exp\left\{-\gamma\sqrt{3}\frac{t}{\tau_s}\right\} \simeq 1 - \gamma\sqrt{3}\frac{t}{\tau_s}. \tag{26}$$

The linear decrease of $C_1^R(t)$ has to be compared with the case of pure Brownian motion of the scatterers for which the correlation function [2] is :

$$C_1^R(t) \simeq 1 - \gamma\sqrt{\frac{6t}{\tau_0}}, \qquad \text{where} \qquad \tau_0^{-1} = k^2 D_B (\simeq 1 \text{ ms}).$$

The two laws in t and $\sqrt{t}$ define a characteristic time t^* for which both correlations are equal. This characteristic time is similar to the previous one since $t^* \simeq D_B/(\Gamma\ell)^2$. Beyond this characteristic time, shear flow varying in $1 - t$ is more efficient than the Brownian motion of the scatterers. It may be interesting to note that the slope at the origin $\left.\frac{\partial C_1^R(t)}{\partial t}\right|_{t=0} = -\frac{\gamma\sqrt{3}}{\tau_s}$ gives immediately the characteristic time τ_s. This expression (Eq. (25)) and the definition of time correlation τ_s are nearly the same ([1]) as those obtained by Pine *et al.* [5].

([1]) The only difference resides in the factor $\sqrt{6}$ in their time correlation function instead of $\sqrt{3}$ here.

The transmitted part $C_1^{\mathrm{T}}(t)$ is obtained from equation (24), taking $x = L - \gamma \ell$

$$C_1^{\mathrm{T}}(t) = \frac{L}{\gamma \ell} \frac{\sinh\left[\gamma \sqrt{3} \frac{t}{\tau_s}\right]}{\sinh\left[\sqrt{3} \frac{L}{\ell} \frac{t}{\tau_s}\right]}. \tag{27}$$

From equation (27), two regimes have to be distinguished. For times $t \ll \tau_s \frac{\ell}{L}$ $\left(\tau_s \frac{\ell}{L} \simeq 7\ \mu\text{s for } L = 10\ \ell\right)$, the normalized transmitted time correlation becomes :

$$C_1^{\mathrm{T}}(t) \simeq 1 - \left(\frac{kL\Gamma}{\sqrt{30}}\right)^2 t^2. \tag{28}$$

It is a surprising result ! In spite of the fact we are in a strongly multiple scattering regime, $C_1^{\mathrm{T}}(t)$ given by equation (28) does not depend either on the mean free path ℓ nor on the coefficient γ describing the effect of the boundaries. It is therefore a very interesting regime to study in order to obtain Γ.

For times $\tau_s \gg t > \tau_s \frac{\ell}{L}$, we obtain instead :

$$C_1^{\mathrm{T}}(t) \simeq 2\sqrt{3} \frac{\gamma t}{\tau_s} \exp\left[-\sqrt{3} \frac{L}{\ell} \frac{t}{\tau_s}\right]. \tag{29}$$

Here again, it is worthwhile to notice that the exponential decay is driven by ℓ and τ_s which are independent quantities.

4.2 POISEUILLE FLOW. — A Poiseuille flow is characterized by the distribution of velocities :

$$\mathbf{V}(\mathbf{R}) = \frac{4\, V_{\max}}{L^2} (Lx - x^2)\, \hat{\mathbf{e}}_z \tag{30}$$

where L is the width of the slab and x the projection of $\mathbf{R}$ on the x-axis. Then the velocity gradient is $\frac{\partial V_z}{\partial x} = \frac{\Gamma}{L}(L - 2x)$ where $\Gamma \equiv \frac{4\, V_{\max}}{L}$. It must be noted that expression (30) takes into account the cancellation of the velocity on the boundaries of the slab at $x = 0$ and $x = L$. The fluctuation of the phase is given by equation (20) which leads to :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{1}{15} k^2 \ell^2 \Gamma^2 t^2 \left[n - \frac{4}{L} \sum_{\nu=1}^{n} \langle x_\nu \rangle + \frac{4}{L^2} \sum_{\nu=1}^{n} \langle x_\nu^2 \rangle \right]. \tag{31}$$

The qualitative difference between the Poiseuille flow considered here and the shear flow or even the Brownian motion of the scatterers clearly appears here. In the previous cases, each contribution of scatterers at the phase variation was independent of their positions. Now, in Poiseuille flow as in more complex situations, the contribution depends on the positions of the scatterers. Hence for each diffusion path of equal length n but geometrically different, one expects a different contribution to the phase variation. According to equation (31) the fluctuation function $\langle \Delta\phi_n^2(t) \rangle$ is obtained by an average over all diffusion paths of the same length n, starting at point x_0 and reaching the point x after ν steps. We have then to specify the probability density $P_{n,\nu}(x)$. Due to its dependence on the boundaries (cf. appendix B), the expression of $\langle \Delta\phi_n^2(t) \rangle$ will be different in reflection and in transmission. Using equation (B2), we obtain :

$$\langle x_\nu \rangle = \frac{4}{\sqrt{3\pi}} \ell \sqrt{\frac{\nu(n-\nu)}{n}}$$

and

$$\langle x_\nu^2 \rangle = \frac{4}{9} \ell^2 \frac{\nu(n-\nu)}{n}$$

so that in reflection we have :

$$\langle \Delta\phi_n^2(t) \rangle = 2\left(\frac{t}{\tau_s}\right)^2 n \left[1 + \frac{8}{27}\frac{\ell^2}{L^2} n - 2\sqrt{\frac{\pi}{3}}\frac{\ell}{L}\sqrt{n}\right] \tag{32}$$

where the characteristic time τ_s associated with the Poiseuille flow in reflection is the same as for the shear flow i.e. defined by $\tau_s^{-1} = \frac{1}{\sqrt{30}} k\ell\Gamma$. Equation (32) shows that $\langle \Delta\phi_n^2(t) \rangle$ depends on the parameter $n(\ell/L)^2$. It can be rewritten $\mathcal{L}/\mathcal{L}_{typ}$ where $\mathcal{L} = \ell n$ is the total length of the random walk of n steps while $\mathcal{L}_{typ} = L^2/\ell$ is the typical value of the length of a random walk crossing the slab of width L. To obtain the total correlation function in reflection, we must now convoluate this result with the Green function $G(r_0, r, n)$. For a semi-infinite system the contribution of the short paths to $G(r_0, r, n)$ with $\mathcal{L} \ll \mathcal{L}_{typ}$ is dominant, so that in reflection equation (32) reduces to :

$$\langle \Delta\phi_n^2(t) \rangle \simeq 2\left(\frac{t}{\tau_s}\right)^2 n \tag{33}$$

which is formally identical to equation (22) obtained for the shear flow. Figure 1 shows the

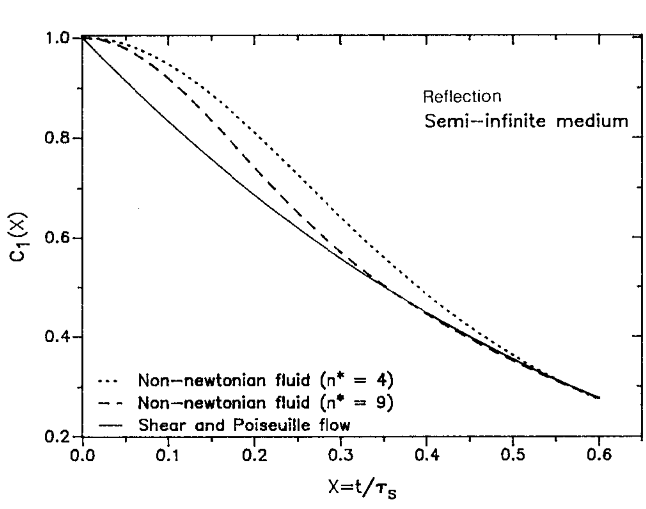

Fig. 1. — Normalized time correlation function $C_1(x)$ for reflection for a semi-infinite medium *versus* $x = t/\tau_s$ for $\gamma = 1$. For this geometry, shear and Poiseuille flow give the same curve (see 4.2). Plug flow of non-Newtonian fluid contributes to $C_1(x)$ with two values of $n^* = \frac{3}{4}\left(\frac{\xi}{\ell}\right)^2$.

correlation function for the reflection in this approximation. Shear and Poiseuille flow are identical since the light diffusion paths explore mainly the space near the plane of the incident light. However for the short time the important light diffusion paths are large and can approach the opposite plane of the slab. For this regime, the light will see the complete velocity gradient profile and a difference between the various laminar regimes considered here is expected. This complex regime calls for a special treatment which is in progress. The identity between equations (22), (33) occurs only in reflection for the Poiseuille flow while in transmission, the situation is somewhat different.

The probability density $P_{n,\nu}(x)$ in transmission has been calculated in appendix B. Since, in this case, the distribution of n is peaked around the value $n_0 \equiv \frac{3}{\pi^2}\frac{L^2}{\ell^2}$, we consider the approximation $n \simeq n_0$. Then we find a straightforward calculation (cf. appendix C) :

$$\langle \Delta\phi_n^2(t)\rangle = 2\left(\frac{t}{\tau_p}\right)^2\left(n + \frac{\pi^2\alpha}{3}n_0\right) \qquad (34)$$

for the expression of the phase fluctuation in transmission. The characteristic time τ_p ($\simeq 180$ μs) associated with the Poiseuille flow is now defined by $\tau_p^{-1} = k\ell\Gamma\sqrt{\frac{1}{30}\left(\frac{1}{3}-\frac{2}{\pi^2}\right)}$ and the constant α is given by 0.093 approximately for $L/\ell = 10$ (see appendix C). The total correlation function is then obtained by using the expression of the Green's function in transmission (see appendix A) :

$$\langle E(r_0, r, t)\, E^*(r_0, r, 0)\rangle = \frac{\sinh\left[\gamma\sqrt{3}\,\frac{t}{\tau_p}\right]}{\sinh\left[\sqrt{3}\,\frac{L}{\ell}\,\frac{t}{\tau_p}\right]}\exp\left[-\frac{\pi^2\alpha}{3}\,n_0\,\frac{t^2}{\tau_p^2}\right]. \qquad (35)$$

Here again as for the shear flow (Eq. (27)) we can make the distinction between short time $\left(t \ll \frac{\ell}{L}\tau_p\right)$ or long time $\left(t > \frac{\ell}{L}\tau_p\right)$ regimes and write the normalized correlation function $C_1^T(t)$ in transmission as :

$$C_1^T(t) = 1 - \left(\frac{1}{2}+\alpha\right)\left(\frac{tL}{\ell\tau_p}\right)^2 \qquad t \ll \frac{\ell}{L}\tau_p \qquad (36a)$$

$$C_1^T(t) = 2\sqrt{3}\,\frac{\gamma t}{\tau_p}\exp\left\{-\sqrt{3}\left(\frac{Lt}{\ell\tau_p}\right) - \alpha\left(\frac{Lt}{\ell\tau_p}\right)^2\right\} \qquad \frac{\ell}{L}\tau_p < t < \tau_p\,. \qquad (36b)$$

In figure 2a is must be noticed first that shear flow decorrelates more efficiently than Poiseuille and plug flow. The explanation is simple : instead of a constant velocity gradient for the shear flow, Poiseuille flow involves a vanishing gradient in the central part of the slab (this effect is still enhanced in the plug flow of non-Newtonian fluid — see below). In the short time regime, we notice (as previously) that only the combination $\ell\tau_p$ appears which depends neither on the mean free path ℓ nor on γ.

In that respect, besides the α-dependence, it is formally similar to the result (Eq. (28)) obtained for shear flow. It is then remarkable that at very short time the correlation function is quasi-universal. This relative insensitivity to the different types of flow is seen in figure 2b where the three types of flow are indistinguishable. This originates in the expression of the

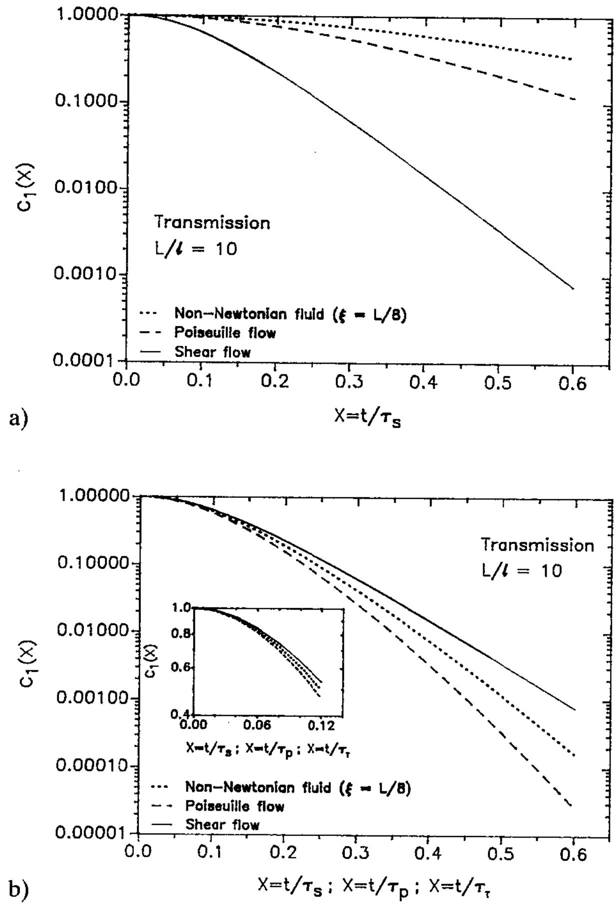

Fig. 2. — Normalized time correlation function $C_\mathrm{I}(x)$ for transmission in a slab of thickness $L = 10\,\ell$. The three curves are obtained for $\gamma = 1$. We have $\alpha \simeq 0.093$ for Poiseuille flow and $\beta \simeq 0.043$ for plug flow of non-Newtonian fluid. Shear flow corresponds to α or $\beta = 0$. a) $C_\mathrm{I}(x)$ *versus* $x = t/\tau_\mathrm{s}$ exhibit the relative efficiency of the different types of flow to the decorrelation. b) $C_\mathrm{I}(x)$ *versus* reduced variables of each type of flow i.e. $x = t/\tau_\mathrm{s}$, $x = t/\tau_\mathrm{p}$ and $x = t/\tau_\mathrm{T}$. This show the functional difference between shear flow and the two others at long time. Insert : shows the same behaviour of the correlation function for the three types of flow.

probability density $P_{n,\nu}(x) \simeq \frac{2}{L}\sin^2\left[\frac{\pi x}{L}\right]$ as a first harmonic of the mathematical series (B4) which reflects the dominant contribution of the long diffusion paths. But at long time regime corresponding to $(Lt/\ell\tau_\mathrm{p}) \gg 1$, $C_\mathrm{I}^\mathrm{T}(t)$ is driven by the part of the exponential involving α and is therefore qualitatively different from equation (29) obtained for the shear flow. The correlation starts to depend on the nature of flow as exhibited in figure 2b. This originates from equation (21). In shear flow case, $\frac{\partial V_z}{\partial x} = \text{constant}$, keeping in (B4) only the $m = 1$ term so that $\alpha = 0$. In contrast, for Poiseuille and plug flow (see below), the integral in equation (21) involves higher harmonics and therefore $\alpha \neq 0$. This new ponderation of the velocity gradient brings about a characteristic time distribution as compared to shear flow. This explains the relative positions of the curves in figure 2b where the correlations are expressed in reduced time variables t/τ_s and t/τ_p. Finally, it is worthwhile to notice again here that the exponential decay of $C_1^T(t)$ is also independent of ℓ and γ which appear only in the prefactor $\gamma t/\tau_p$.

4.3 PLUG FLOW OF NON-NEWTONIAN FLUID FLOWING IN A SLAB. — Several fluids of practical interest like the polymer solutions or colloidal suspensions are non-Newtonian fluids. Let us recall that a non-Newtonian fluid has a shear viscosity which depends on the velocity. It was shown first by Eyring *et al.* [11] that the viscosity of typical non-Newtonian fluid is well described by the relation :

$$\eta = \eta_0 \frac{\sinh^{-1}\left[\tau \dfrac{\partial V}{\partial x}\right]}{\partial x} \tag{37}$$

where η_0 is reference viscosity and τ a relaxation time.

We assume that the direction of flow is along the z-axis perpendicular to the boundaries at $x = 0$ and $x = L$. The applied pressure gradient is assumed to be in the direction of the flow and constant. The momentum balance equation for a steady flow is then given by :

$$\frac{\partial}{\partial x}\left[\eta \frac{\partial V_z}{\partial x}\right] = \frac{\partial p}{\partial z} \tag{38}$$

where $\dfrac{\partial p}{\partial z}$ is the pressure gradient.

By integrating (38) with η given by (37) and assuming vanishing velocities on the walls $x = 0$ and $x = L$, one finds :

$$\mathbf{V}(\mathbf{R}) = V_{\max} \frac{\sinh\left[\delta \frac{x}{L}\right] \sinh\left[\delta\left(1 - \frac{x}{L}\right)\right]}{\sinh^2\left[\frac{\delta}{2}\right]} \hat{\mathbf{e}}_z \tag{39}$$

with :

$$\delta = \frac{\tau L}{2\,\eta_0} \frac{\partial p}{\partial z}$$

δ describes the departure from the Newtonian fluid. For $\delta \ll 1$, the fluid is nearly Newtonian and in our case the velocity field is of the Poiseuille type (cf. 4.2). For $\delta > 1$ the fluid is non-Newtonian and the velocity field gradient depends on x :

$$\Gamma(x) = \frac{\delta V_{\max}}{L} \frac{\sinh\left[\delta(1 - 2\,x/L)\right]}{\sinh^2\left[\delta/2\right]} = \Gamma \frac{\sinh\left[\delta(1 - 2\,x/L)\right]}{\sinh\left[\delta\right]} \tag{40}$$

with $\Gamma = \dfrac{\delta V_{\max}}{L} \dfrac{\sinh[\delta]}{\sinh^2[\delta/2]}$.

Let us consider first the correlation in the reflection. Suppose that $\left(\dfrac{L}{\ell}\right)^2 \gg n$ like in the semi-infinite medium, the expression (40) is written as :

$$\Gamma(x) \simeq \frac{2\,\delta V_{\max}}{L} \exp\left\{-\frac{2\,\delta x}{L}\right\}. \tag{41}$$

Let us define $\xi = \dfrac{L}{2\,\delta}$, the characteristic length beyond which the velocity field is constant. Then :

$$\Gamma(x) = \Gamma \exp\left\{-\frac{x}{\xi}\right\} \tag{42}$$

where Γ is the velocity gradient at the boundary $x = 0$. To be in the multiple scattering regime we must verify the inequalities $1 < \delta \leqslant \dfrac{L}{2\,\ell}$.

In the limiting case $\xi \leqslant \ell$, the approximation of the slow variation of the velocity field at the scale of ℓ is no longer valid. We will suppose therefore that $\xi \gg \ell$. In this regime the total phase correlation function is (see appendix D) :

$$\langle \Delta\phi_n^2(t)\rangle = 2\left(\frac{t}{\tau_s}\right)^2 n \qquad n \ll n^* \tag{43a}$$

$$= 2\left(\frac{t}{\tau^*}\right)^2 \qquad n > n^* \tag{43b}$$

where τ_s is the characteristic time for the shearing flow since, in this regime, only the initial gradient is relevant. The most probable value of n in order to travel a distance ξ in a random walk of step ℓ is $n^* = \dfrac{3}{4}\dfrac{\xi^2}{\ell^2}$. Then :

$$\tau^* \simeq \frac{\tau_s}{\sqrt{n^*}} \tag{44}$$

this time τ^* ($\simeq 13$ µs for $\xi \simeq 6\,\ell$ and $\tau_s \simeq 70$ µs) is the correlation time corresponding to n^* scatterings which decorrelate the electrical field by a factor $\exp\left\{-\left(\dfrac{t}{\tau_s}\right)^2\right\}$ at each step. For $n \ll n^*$, the phase fluctuation function is proportional to the number of scatterings and contains only the initial velocity gradient. Actually only this initial gradient contributes to the correlation function in reflection. And the linear dependence on n comes from the quasi-linear dependence of velocity field. For $n > n^*$, the velocity field is nearly constant and the gradient vanishes. Then the fluctuation function becomes independent on n. For the semi-infinite medium, one finds :

$$C_1^{\mathrm{R}}(t) \simeq 1 - \left(\frac{t}{\tau^*}\right)^2 \qquad t < \tau^* \tag{45a}$$

$$\simeq 1 - \gamma\,\sqrt{3}\,\frac{t}{\tau_s} \qquad \tau^* < t < \tau_s\,. \tag{45b}$$

Equation (45a) presents the same behaviour than the equations (28), (36a) discussed above for transmission. Hence ξ plays the same role as L in the transmission expression. Indeed, because the velocity gradient acts only within a distance ξ form the interface, $C_1^{\mathrm{R}}(t)$ decreases as t^2 at short time. All reflected diffusion paths longer than ξ^2/ℓ give the same contribution (independent of the length) to the decorrelation.

In transmission, the total correlation of phase are given by the expression (D15) in the appendix D :

$$\langle \Delta\phi_n^2(t)\rangle = 2\left(\frac{t}{\tau_{\mathrm{T}}}\right)^2 \cdot \left\{n + \frac{\pi^2\beta}{3}\,n_0\right\} \tag{46}$$

where :

$$\tau_{\mathrm{T}}^{-1} = \frac{2\,\pi}{\sqrt{30\left(1 + \frac{\pi^2 \xi^2}{L^2}\right)}} \left(\frac{\xi}{L}\right)^{3/2} k\ell\Gamma \tag{47}$$

τ_{T} ($\simeq 270\ \mu\mathrm{s}$ for $\xi \simeq L/8$) corresponds to a characteristic time of the same type as τ_{s} but with an average velocity gradient less than Γ by a factor $\frac{2\,\pi}{\sqrt{1 + \frac{\pi^2 \xi^2}{L^2}}} \left(\frac{\xi}{L}\right)^{3/2}$. In terms of this new time τ_{T} one obtains an expression of $C_{\mathrm{I}}^{\mathrm{T}}(t)$ very similar to the previous one for Poiseuille flow :

$$C_{\mathrm{I}}^{\mathrm{T}}(t) = \frac{L}{\gamma\ell} \frac{\sinh\left[\gamma\sqrt{3}\,\frac{t}{\tau_{\mathrm{T}}}\right]}{\sinh\left[\sqrt{3}\,\frac{L}{\ell}\,\frac{t}{\tau_{\mathrm{T}}}\right]} \exp\left\{-\beta\left(\frac{Lt}{\ell\tau_{\mathrm{T}}}\right)^2\right\}. \tag{48}$$

5. Conclusion.

Let us summarize the main results of this article. We have developed a theoretical analysis of the dynamical correlation of the light multiply scattered by a disordered medium. We considered the case for which the scatterers have a dynamical deterministic motion imposed by the underlying fluid in which there are immersed. In section 2, we have derived a general expression for the time correlation function in the multiple scattering regime. It is based on an expression for the scattered electric field written as a summation over all the possible random walk paths of an accumulated phase. This phase describes the motion of each scatterer with time. This approach was introduced and used by Maret and Wolf [2] in their study of the Brownian motion of the scatterers. Here, we tried to give some more details about the delicate transition between a description of the multiple scattering paths in terms of usual so called path integrals to a description based on classical random walk paths weighted by a phase term. We emphasize here that such an approach is derived within the weak scattering limit $k\ell \gg 1$ and describes the random walk paths as the Markovian process of a stationary diffusion problem.

All the relevant information about the characteristics of the flow is contained in the phase variation $\Delta\phi_n(t)$ associated with paths of length n. Then in section 3, we have evaluated it carefully for the case of stationary flow. We restricted ourselves to the case of incompressible fluids for which the average $\langle \Delta\phi_n(t) \rangle$ over the diffusion paths is identically zero.

In section 4, we have applied our general formula to some characteristic motions such as laminar shear and Poiseuille flow in a slab. For each case, we have evaluated the time correlation function of electric field both in transmission and in reflection. The behaviour at short times in the strongly multiple scattering regime reveals to be quantitatively different from the case of the Brownian motion. This is a signature of a deterministic flow. The case of Poiseuille flow clearly marks a qualitative difference compared to previous cases. It is the simplest case where the fluctuation $\langle \Delta\phi_n^2(t) \rangle$ of the phase involves the exact position of the visited scatterers instead of only the number n of the steps of the random walk. We had therefore to introduce (in order to calculate the average) the probability density $P_{n,\nu}(x)$ giving the probability reach the point x after ν steps in a random walk of n steps. Our evaluation of this very important function $P_{n,\nu}(x)$ was based on similar calculations used long ago by Edwards [9] and De Gennes [10] in statistical mechanics of polymeric chains. Then, it gave rise to a marked differentiation between the expressions of $\langle \Delta\phi_n^2(t) \rangle$ in reflection or in transmission and therefore for the corresponding correlation functions. We have also shown that for all these flow, the transmitted correlation function at very short times becomes independent of the elastic mean free path as well as from other characteristics of disorder. We believe that this remarkable fact could be very helpful experimentally and it will be possible to obtain quantitative information about flow even under conditions of strong multiple scattering.

In section 4, we considered the case of plug flow of non-Newtonian fluid flowing in a slab. Since within a large portion of the space the velocity field amplitude is constant, no dephasing occurs and the range of the correlation function is increased beyond the Poiseuille flow value as shown in figures 1 and 2.

Some of the results exposed here were already derived theoretically and tested experimentally [5]. This is the case of shear flow. But concerning for instance Poiseuille flow, we think that we developed a refined and powerful approach which extends beyond the cases where it coincides with shear. It gives rise to new predictions which could be tested. More generally, our approach of the problem might be the starting point for the study of more difficult situations dealing with viscous, correlated or even turbulent flow.

Acknowledgements.

We are pleased to acknowledge G. Maret for fruitful discussions and E. Wolf for a critical reading of the manuscript. One of us (E. Akkermans) acknowledges support from the Lady Davies foundation.

Appendix A.

Expression of the Green function of the diffusion equation in a slab.

We consider the geometry of an infinite slab of width L perpendicular to the x-axis and solve the time-dependent diffusion equation :

$$\frac{\partial n}{\partial t} = D \nabla^2 n(r, t) \tag{A1}$$

where the diffusion constant $D = \dfrac{\ell^2}{3\tau}$. Moreover we impose to the field $n(r, t)$ the Dirichlet boundary conditions $n(x = 0, t) = n(x = L, t) = 0$. These boundary conditions are adapted to the problem of propagation of light we consider here. Indeed, we assume that light is lost for the medium when it arrives within one mean free path from the boundaries [7]. The Green equation of equation (A1) is :

$$\frac{\partial G}{\partial t} - D \nabla_r^2 G(r_0, r, t) = \delta(r - r_0)\, \delta(t) \tag{A2}$$

with a source placed at r_0. $G(r_0, r, t)$ obeys the Dirichlet boundary conditions $G(r_0, x = 0, t) = G(r_0, x = L, t) = 0$. The solution of equation (A2) is :

$$G(\rho, x_0, x, t) = \frac{1}{4\pi D t} \exp(-\rho^2/4Dt) \sum_{m=1}^{\infty} \psi_m(x_0)\, \psi_m(x) \exp\left(-\frac{\pi^2 m^2 D^2 t^2}{L^2}\right) \tag{A3}$$

where $\boldsymbol{\rho}$ is the projection of $\mathbf{r} - \mathbf{r}_0$ on the $y - z$ plane and x_0 and x are respectively the projections of $\mathbf{r}_0$ and $\mathbf{r}$ on the x-axis. Finally, in equation (A3) the eigenfunctions $\psi_m(x)$ are given by :

$$\psi_m(x) = \sqrt{\frac{2}{L}} \sin\left(\frac{m\pi x}{L}\right) . \tag{A4}$$

Due to our choice of the geometry and since we are considering only incident plane waves, all the quantities we calculated are integrated over the cross-section. Thus, the Green function of interest to us is :

$$G(x_0, x, t) = \int_0^\infty 2\pi\rho \, d\rho \, G(\rho, x_0, x, t) . \tag{A5}$$

In random walk theory, $G(x_0, x, n)$ can also be considered as the probability density for a Brownian particle starting at x_0 to arrive at point x after n steps, provided we make the change $n = t/\tau$ in equation (A5). This is exactly the meaning we give to the quantity appearing in equation (8), thus :

$$G(x_0, x, n) = \frac{2}{L} \sum_{m=1}^{\infty} \sin\left(\frac{m\pi x_0}{L}\right) \sin\left(\frac{m\pi x}{L}\right) \exp\left\{-\frac{m^2\pi^2\ell^2}{3L^2} n\right\} . \tag{A6}$$

In the limit $L \to \infty$, equation (A6) gives the well-known result for a semi-infinite medium :

$$G(x_0, x, n) = \sqrt{\frac{3}{4\pi\ell^2 n}} \left\{\exp\left(-\frac{3(x-x_0)^2}{4n\ell^2}\right) - \exp\left(-\frac{3(x+x_0)^2}{4n\ell^2}\right)\right\} . \tag{A7}$$

Appendix B.

Expression of the probability density $P_{n,\nu}(x)$ in a slab.

In this appendix, our aim is to calculate the probability density $P_{n,\nu}(x)$ defined as the probability for a random walk of n steps starting at point x_0 to reach point x after ν steps. This quantity was extensively studied in statistical mechanics of polymers by Edwards [9] and De Gennes [10]. They considered exclusively the problem of non correlated monomers so that the Green function of the underlying random walk has the property of a Markov process. It is then possible to break the random walk paths into pieces so that :

$$P_{n,\nu}(x) = \frac{G(x_0, x, \nu)\, G(x, x_f, n-\nu)}{G(x_0, x_f, n)} \tag{B1}$$

where $G(x_1, x_2, p)$ is the Green function defined in equation (A6). In equation (B1), $P_{n,\nu}(x)$ also depends on the initial and the final points (x_0 and x_f respectively) of the n-steps random walk. In evaluating $P_{n,\nu}(x)$ we then have to specify which conditions are used :

i) *Reflection.*

The point source x_0 as well as the escaping point x_f are within a mean free path ℓ from the boundaries. Moreover, the distribution to $P_{n,\nu}(x)$ is dominated by the short paths so that we can use expression (A6) to compute it in the limit of $L \to \infty$. This gives :

$$P_{n,\nu}(x) = \frac{4}{\sqrt{\pi}} x^2 \left[\frac{3n}{4\ell^2 \nu(n-\nu)}\right]^{3/2} \exp\left\{-\frac{3nx^2}{4\ell^2\nu(n-\nu)}\right\} . \tag{B2}$$

ii) *Transmission.*

For transmission we take $x_0 = \ell$ and $x_f = L - \ell$ and we use equation (A6). Moreover, the distribution of n in transmission is strongly peaked around $n_0 \equiv \frac{3 L^2}{\pi^2 \ell^2}$. Then in the Green function $G(x_0, x_f, n)$ appearing in equation (B1) we retain only the $m = 1$ harmonic, all the subsequent ones being strongly attenuated. Then, we can write :

$$P_{n,\nu}(x) = \frac{2}{L} \frac{\exp\left(\frac{n}{n_0}\right)}{\sin^2\left(\frac{\pi\ell}{L}\right)} \sum_{m, m'=1}^{\infty} (-1)^{m'+1} \sin\left(\frac{m' \pi \ell}{L}\right) \sin\left(\frac{m\pi\ell}{L}\right) \sin\left(\frac{m' \pi x}{L}\right) \times$$
$$\times \sin\left(\frac{m\pi x}{L}\right) \times \exp\left\{-\frac{\nu}{n_0}(m^2 - m'^2) - \frac{m'^2 n}{n_0}\right\} \quad \text{(B3)}$$

$\frac{\nu}{n_0}(m^2 - m'^2)$ can be very small even for high harmonics but the term $\frac{nm'^2}{n_0}$ gives rise to an exponentially small contribution unless $m' = 1$. Thus, we obtain :

$$P_{n,\nu}(x) = \frac{2}{L} \frac{\sin\left(\frac{\pi x}{L}\right)}{\sin\left(\frac{\pi\ell}{L}\right)} \sum_{m=1}^{\infty} \sin\left(\frac{m\pi\ell}{L}\right) \sin\left(\frac{m\pi x}{L}\right) \exp\left\{-\frac{(m^2-1)\nu}{n_0}\right\} \quad \text{(B4)}$$

which does not depend on n anymore for $n \simeq n_0$.

Appendix C.

Calculation of the fluctuation $\langle \Delta\phi_n^2(t) \rangle$ of the phase for Poiseuille flow in transmission.

From equations (20), (30) we have :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{1}{15} k^2 \ell^2 \Gamma^2 t^2 \sum_{\nu=1}^{n} \int_0^L P_{n,\nu}(x) \left[1 - 4\frac{x}{L} + 4\frac{x^2}{L^2}\right] \mathrm{d}x \quad \text{(C1)}$$

where the probability density is given in equation (B4). The expression of $\langle \Delta\phi_n^2(t) \rangle$ involves the integral :

$$\int_0^L \sin\left(\frac{\pi x}{L}\right) \sin\left(\frac{m\pi x}{L}\right) \left[1 - 4\frac{x}{L} + 4\frac{x^2}{L^2}\right] \mathrm{d}x$$
$$= \left(\frac{1}{3} - \frac{2}{\pi^2}\right) \frac{L}{2} \qquad m = 1 \quad \text{(C2a)}$$
$$= \left[1 - (-1)^m\right] \frac{8\, mL}{\pi^2 (m^2-1)^2} \qquad m \neq 1 \quad \text{(C2b)}$$

so that :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{1}{15} k^2 \ell^2 \Gamma^2 t^2 \left\{ \left(\frac{1}{3} - \frac{2}{\pi^2}\right) n + \frac{48\, L^2}{\pi^4 \ell^2} \sum_{m=3}^{\infty} \frac{\sin\left(\frac{m\pi\ell}{L}\right)}{\sin\left(\frac{\pi\ell}{L}\right)} \cdot E(m) \right\} \quad \text{(C3)}$$

$$E(m) = [1 - (-1)^m] \frac{m}{(m^2-1)^3} \left[\exp\left(-\frac{(m^2-1)}{n_0} \right) - \exp\left(-\frac{(m^2-1)\,n}{n_0} \right) \right].$$

Since in transmission, we consider $n \simeq n_0$, the second exponential in the bracket is negligible compared to the first one, thus defining :

$$S(L/\ell) \equiv \sum_{m=3}^{\infty} [1 - (-1)^m] \frac{m}{(m^2-1)^3} \frac{\sin\left(\frac{m\pi\ell}{L}\right)}{\sin\left(\frac{\pi\ell}{L}\right)} \exp\left(-\frac{(m^2-1)}{n_0} \right) \tag{C4}$$

and

$$\alpha \equiv \frac{144\, S(L/\ell)}{\pi^2(\pi^2-6)}$$

we can rewrite $\langle \Delta\phi_n^2(t) \rangle$ as :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{1}{15} \left(\frac{1}{3} - \frac{2}{\pi^2} \right) \left\{ n + \frac{\pi^2 \alpha}{3} n_0 \right\} k^2 \ell^2 \Gamma^2 t^2 . \tag{C5}$$

The series $S(L/\ell)$ is obviously uniformly converging even in the limit $n_0 \to \infty$ and $\lim_{L/\ell \to \infty} S(L/\ell) \simeq 0.04$. For $L/\ell = 10$, $S(L/\ell) \simeq 0.0246$ so that $\alpha \simeq 0.093$. Equation (C5) is the equation (34) used in the text.

Appendix D.

Calculation of the fluctuations $\langle \Delta\phi_n^2(t) \rangle$ of the phase for plug flow of non-Newtonian fluid flowing in a slab.

The fluctuation of the phase is :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{1}{15} k^2 \ell^2 t^2 \sum_{\nu=1}^{n} \int_a^b P_{n,\nu}(x) \left[\frac{\partial V_z}{\partial x} \right]^2 \mathrm{d}x . \tag{D1}$$

Due to the sensitivity of $P_{n,\nu}(x)$ to the boundaries, we will distinguish the reflection and in transmission in the calculation.

i) *Reflection.*

Let us consider a semi-infinite medium $\left(n \ll \frac{L^2}{\ell^2} \right)$, the boundaries of the integral over x in equation (D1) are $a = 0$ and $b = \infty$. For the calculation, we use equation (42) for $\frac{\partial V_z}{\partial x}$ and (B2) for $P_{n,\nu}(x)$. As defined in the previous appendix B, $P_{n,\nu}(x)$ is the probability density for a random walk of n steps starting at point x_0 to reach the point x after ν steps. In reflection, the most probable distance L_ν between x and x_0 is defined as :

$$L_\nu^2 = \frac{4}{3} \frac{\nu(n-\nu)}{n} \ell^2 \tag{D2}$$

the typical number of steps n^* for a random walk over a distance ξ with $L_\nu = \xi$, is then : $n^* \simeq \frac{3}{4}\left(\frac{\xi}{\ell}\right)^2$. Equation (D1) is written :

$$\langle \Delta\phi_n^2(t)\rangle = \frac{1}{15} k^2 \ell^2 \Gamma^2 t^2 \frac{4}{\sqrt{\pi}} \sum_{\nu=1}^{n} \frac{1}{L_\nu^3} \int_0^\infty x^2 \exp\left[-\frac{x^2}{L_\nu^2} - \frac{2x}{\xi}\right] \mathrm{d}x \tag{D3}$$

we find [12] :

$$\langle \Delta\phi_n^2(t)\rangle = \frac{4 k^2 \ell^2 \Gamma^2 t^2}{15\sqrt{\pi}} \times \\ \times \sum_{\nu=1}^{n} \left\{ -\frac{L_\nu}{2\xi} + \frac{\sqrt{\pi}}{4}\left(\frac{2 L_\nu^2}{\xi^2} + 1\right)\left(1 - \Phi\left(\frac{L_\nu}{\xi}\right)\right) \exp\left[\left(\frac{L_\nu}{\xi}\right)^2\right]\right\} \tag{D4}$$

where $\Phi(y)$ is the error-function.

When $L_\nu \ll \xi$, means $n \ll n^*$, equation (D4) becomes :

$$\langle \Delta\phi_\nu^2(t)\rangle = \frac{1}{15} k^2 \ell^2 \Gamma^2 t^2 \sum_{\nu=1}^{n} \left\{1 - \frac{4}{\sqrt{\pi}}\frac{L_\nu}{\xi} + \cdots\right\}. \tag{D5}$$

Equation (D5) appears as an expansion into the parameter $\frac{L_\nu}{\xi}$ which is smaller than one in this case. $\langle \Delta\phi_n^2(t)\rangle$ is driven essentially by zero order term of the expansion, the others give rise to a small contribution. Then, equation (D5) reduces to :

$$\langle \Delta\phi_n^2(t)\rangle \simeq 2\left(\frac{t}{\tau_s}\right)^2 n \tag{D6}$$

where $\tau_s^{-1} = \frac{1}{\sqrt{30}} k\ell\Gamma$ is the characteristic time correlation in reflection. For $L_\nu > \xi$, equation (D4) takes the following form :

$$\langle \Delta\phi_n^2(t)\rangle \simeq \frac{1}{15\sqrt{\pi}} k^2 \ell^2 \Gamma^2 t^2 \sum_{\nu=n^*}^{n-n^*} \left[\frac{\xi}{L_\nu}\right]^3 \tag{D7}$$

from equation (D2) and equation (D7) we obtain :

$$\langle \Delta\phi_n^2(t)\rangle \simeq 2\left(\frac{t}{\tau^*}\right)^2 \tag{D8}$$

where $\tau^* \simeq \frac{\tau_s}{\sqrt{n^*}}$ is the time correlation associated to the multiple scattering in a slab of width ξ, corresponding to the region within which the velocity gradient is non-zero. We can resume :

$$\langle \Delta\phi_n^2(t)\rangle \simeq 2\left(\frac{t}{\tau_s}\right)^2 n \qquad n \ll n^* \tag{D9a}$$

$$\langle \Delta\phi_n^2(t)\rangle \simeq 2\left(\frac{t}{\tau^*}\right)^2 \qquad n > n^*. \tag{D9b}$$

ii) *Transmission.*

Now, the boundaries are $a = 0$ and $b = L$ in equation (D1). The probability density $P_{n,\nu}(x)$ is given by equation (B4). The fluctuation of the phase is then :

$$\langle \Delta\phi_n^2(t) \rangle = \frac{k^2 \ell^2 t^2}{15} \frac{\Gamma^2}{\sinh^2[\delta]} \sum_{\nu=1}^{n} \int_0^L P_{n,\nu}(x) \sinh^2\left[\delta - \frac{x}{\xi}\right] \mathrm{d}x \tag{D10}$$

the expression of $\langle \Delta\phi_n^2(t) \rangle$ involves the integral :

$$\int_0^L P_{n,\nu}(x) \sinh^2\left[\delta - \frac{x}{\xi}\right] \mathrm{d}x = -2 + \frac{2\pi^2 \xi^3 \sinh[2\delta]}{L^3} \left\{ \frac{1}{1 + \frac{\pi^2 \xi^2}{L^2}} + \frac{1}{8} \sum_{m=3}^{\infty} E_\nu(m) \right\} \tag{D11}$$

$$E_\nu(m) = [1 - (-1)^m] \frac{m}{1 + \frac{(m^2+1)\pi^2 \xi^2}{2L^2} + \frac{(m^2-1)^2 \pi^4 \xi^4}{16 L^4}} \frac{\sin\left[\frac{m\pi\ell}{L}\right]}{\sin\left[\frac{\pi\ell}{L}\right]} \times \exp\left\{-\frac{(m^2-1)\nu}{n_0}\right\}.$$

The sum of equation (D11) over ν from 1 to n, retaining the dominant term, gives :

$$\sum_{\nu=1}^{n} \text{equation (D11)} = \frac{2\pi^2 \xi^3}{L^3} \frac{\sinh[2\delta]}{1 + \frac{\pi^2 \xi^2}{L^2}} n + \frac{3\xi^3}{4 L \ell^2} \sinh[2\delta] \, S(\xi, L/\ell) \tag{D12}$$

where :

$$S(\xi, L/\ell) = \sum_{m=3}^{\infty} \frac{\sin\left[\frac{m\pi\ell}{L}\right]}{\sin\left[\frac{\pi\ell}{L}\right]} \frac{E_1(m)}{m^2 - 1} \tag{D13}$$

which, for the greater value of ξ ; $\xi_{\max} = \frac{L}{2}$, tends to :

$$\lim_{L/\ell \to \infty} S(\xi_{\max}, L/\ell) \simeq 0.0717$$

and for the smaller value $\xi_{\min} = \ell$, behaves as :

$$\lim_{L/\ell \to \infty} S(\xi_{\min}, L/\ell) \simeq 0.275 \frac{L}{\ell}.$$

Let us call :

$$\beta(\xi, L/\ell) = \beta = \frac{3\left(1 + \frac{\pi^2 \xi^2}{L^2}\right)}{8\pi^2} S(\xi, L/\ell) \tag{D14}$$

and $\tau_{\mathrm{T}}^{-1} = \dfrac{2\pi}{\sqrt{30\left(1+\dfrac{\pi^2\xi^2}{L^2}\right)}}\left(\dfrac{\xi}{L}\right)^{3/2} k\ell\Gamma$ the characteristic time correlation in transmission, then the fluctuation of the phase can be written as:

$$\langle\Delta\phi_n^2(t)\rangle = 2\left(\frac{t}{\tau_{\mathrm{T}}}\right)^2\left\{n+\frac{\pi^2\beta}{3}n_0\right\}. \tag{D15}$$

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