

Retarded hydrodynamic fluctuations effects on the light scattering spectrum and elastic moduli of a linear viscoelastic liquidⁱ

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Fluctuating hydrodynamics is one of the theories that describes the dynamics of the fluctuations for fluids at mesoscopic scales. However, it is only valid for Markovian processes. Here we extend this approach for a viscoelastic liquid by using a generalized Langevin equation. We obtain general analytic expressions for the density fluctuations correlation function, the dynamic structure factor and the light scattering spectrum. In particular, we calculate the intermediate scattering function when the time memory and the noise correlation function are power-laws. We find that the difference in values of this quantity as functions of the viscoelasticity may vary between 56.1 - 70.7% for the time interval 510-6000 s, and might be measurable.

Keywords: Viscoelasticity; light scattering; correlation functions; stochastic equations.

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1. Introduction

The ability to describe time-dependent deformations in soft materials is important when modelling the large variety of relaxation processes that occur in many dynamic systems. These type of deformations may preclude a clear separation of time scales associated with the macroscopic transport processes and those of the microscopic ones giving rise to them. This is the case for complex systems like viscoelastic fluids [1,2], glassy materials, synthetic or reinforced polymers, and of increasing importance for biopolymers, membranes, tissues [3] and other biological systems like active systems [4], for which the associated relaxation functions are non-exponential, due to the large number of highly coupled elementary units responsible for the relaxation. For many of these systems it is the microstructure of the medium which dictates that the material response may be strongly viscoelastic. This response is manifested in the dynamics of its fluctuations and on the dynamic behavior of the correlation functions associated with many transport properties. In this work we model these retarded fluctuations correlations behavior in terms of a generalized Langevin equation (*GLE*) with long time-memory effects when both, the friction kernel and the correlation of the stochastic force obey a power-law dynamics. More specifically, for a linear, symmetric, homogeneous, viscoelastic fluid we calculate analytically its density correlation function and its intermediate scattering function (*ISF*)

or van Hove's self-correlation function [5]. A general expression for the light scattering structure factor as a function of the longitudinal elastic modulus of the fluid is also obtained. Thus, if this modulus is modeled or measured, the structure factor could be determined or, alternatively, a measure of the structure factor will yield an analytic expression for the longitudinal elastic modulus. We focus our analysis on the theoretical calculation of the van Hove's function for the case of power-law fluctuations and viscoelasticity. We find that the difference in values of the *ISF* as functions of the viscoelasticity may reach values as high as 70% [6]. Since we are not aware of experimental measurements of either of these quantities, a comparison between our theoretical results and experiment is not feasible and remains to be assessed.

2. Fluctuating hydrodynamic equations

Consider a linear, quiescent ($v_0 = 0$) viscoelastic liquid which is brought to thermodynamic equilibrium in the presence of certain macroscopic constraints at the (absolute) temperature T . In a typical light scattering experiment the deformations produced in the fluid by its interaction with an electromagnetic wave at the intensities usually employed in these experiments, do not drive the system too far from equilibrium and can be described by means of linear response theory. In this regime the most general constitutive equation for the linear stress-strain relation is of the form [7-9]

$$\sigma_{ij}(\vec{r}, t) = -p\delta_{ij} + \int_0^t dt' \left\{ K(t-t') \dot{\gamma}_{kk}(\vec{r}, t') \delta_{ij} + 2G(t-t') \left[\dot{\gamma}_{ij}(\vec{r}, t') - \frac{1}{3} \dot{\gamma}_{kk}(\vec{r}, t') \delta_{ij} \right] \right\}, \quad (1)$$

where \vec{r} is the position vector with Cartesian components $x_i = (x, y, z)$; $\sigma_{ij}(\vec{r}, t)$ is the symmetric stress tensor, $p(\vec{r}, t)$ is the pressure and $\dot{\gamma}_{ij}(\vec{r}, t) = (1/2)(\partial v_i/\partial x_j + \partial v_j/\partial x_i)$ is the rate of strain tensor. The scalar functions $K(t)$ and $G(t)$ denote, respectively, the bulk (compressional) modulus and the shear modulus, which are assumed to be spatially homogeneous quantities. Substitution of Eq. (1) into the momentum conservation equation yields the equation of motion [10]

$$\rho \left(\frac{\partial v_i}{\partial t} + v_k \frac{\partial v_i}{\partial x_k} \right) = -\frac{\partial p}{\partial x_i} + \int_0^t dt' \left\{ \left[K(t-t') + \frac{1}{3} G(t-t') \right] \frac{\partial}{\partial x_i} \nabla_k v_k(\vec{r}, t') + G(t-t') \nabla^2 v_i(\vec{r}, t') \right\}. \quad (2)$$

Consistency with linear response theory requires that Eq. (2) should be linearized in the small (deterministic) deviations of the mass density $\rho(\vec{r}, t)$ and the hydrodynamic velocity $\vec{v}(\vec{r}, t)$ with respect to the initial reference equilibrium state which will be identified by the subscript 0. These deviations are defined as $\delta\rho(\vec{r}, t) \equiv \rho(\vec{r}, t) - \rho_0(\vec{r})$, $\delta p(\vec{r}, t) \equiv p(\vec{r}, t) - p_0$ and $\delta v_i(\vec{r}, t) \equiv v_i(\vec{r}, t)$ due to Galilei invariance. However, these deviations are really random quantities, they will manifest themselves as the fluctuations in the hydrodynamic state variables and their dynamics should be described stochastically. It is important to point out that due to thermal diffusion, the fluctuations δp are an implicit function of both, $\delta\rho$ and δT . However, since at the temperatures of interest in a typical light scattering experiment using the photon correlation technique, the thermal diffusion term contributes to the density fluctuation spectrum only in the 10-100 MHz range, which is outside the dynamic range that can be monitored by this technique [11], in a first approximation the thermal diffusion may be neglected. The effects of temperature fluctuations may then be considered to be sufficiently small and [12]

$$\frac{\partial \delta p}{\partial x_i} = \left(\frac{\partial \delta p}{\partial \rho} \right)_T \frac{\partial \delta \rho}{\partial x_i} = \frac{1}{\rho_0 \chi_T} \frac{\partial \delta \rho}{\partial x_i} \quad (3)$$

where χ_T is the isothermal compressibility.

It is convenient to separate the velocity $\vec{v}(\vec{r}, t)$ field into a longitudinal (\vec{v}_L) and a transverse component (\vec{v}_T), $\vec{v} = \vec{v}_L + \vec{v}_T$, which satisfy, respectively, the conditions $\nabla \times \vec{v}_L = 0$ and $\nabla \cdot \vec{v}_T = 0$. Furthermore, if the direction of the z -axis is identified as the longitudinal component,

the linearized longitudinal part of the velocity fluctuations is $\vec{v}_L \equiv v_z \hat{z}$, where \hat{z} is a unit vector in the z -direction. In what follows we shall only consider this longitudinal component and will be denoted as $v_z \equiv v$. Accordingly, the deviations $\delta\rho$ are related to those of $\delta v(\vec{r}, t) \equiv v(\vec{r}, t)$ through the linearized continuity equation

$$\frac{\partial}{\partial t} \delta\rho(\vec{r}, t) = -\rho_0 \frac{\partial v}{\partial z} \quad (4)$$

and the equation of motion for $v(\vec{r}, t)$ reduces to

$$\rho_0 \frac{\partial}{\partial t} v(\vec{r}, t) = -\frac{1}{\rho_0 \chi_T} \nabla \delta\rho(\vec{r}, t) + \int_0^t dt' M(t-t') \frac{\partial^2}{\partial z^2} v(z, t'). \quad (5)$$

where the longitudinal modulus $M(t)$ is defined as

$$M(t) = K(t) + \frac{4}{3} G(t). \quad (6)$$

Equation (5) explicitly shows that the dynamics of the density $\delta\rho(\vec{r}, t)$ and longitudinal velocity $v_z(\vec{r}, t)$ fluctuations are coupled.

3. Density correlation function and the spectrum of fluctuations

The theory of fluctuations in fluids in an equilibrium state was initiated long ago by Einstein and Onsager, and was later reformulated in several but equivalent ways [12-14]. Here we construct an approach based on a generalized Langevin equation approach with Gaussian fluctuations, in the linear regression regime close to full equilibrium [15,16], based on the hydrodynamic Eqs. (4) and (5). If a random term, $f(t)$, is added to the r.h.s. of (5) and the Fourier transform with respect to z is taken, (5) becomes a stochastic non-Markovian equation for the longitudinal velocity fluctuations

$$\rho_0 \frac{\partial}{\partial t} \hat{v}(q, t) = -q^2 \int_0^t dt' M(t-t') \hat{v}(q, t') - \frac{iq}{\rho_0 \chi_T} \delta \hat{\rho}(q, t) + f(t). \quad (7)$$

We choose the stochastic noise term $f(t)$ to be a Gaussian, stationary, stochastic process with zero mean $\langle f(t) \rangle = 0$ and with arbitrary correlation

$$\langle f(t) f(t') \rangle = C(|t - t'|) \equiv C(\tau), \quad (8)$$

subject to the condition

$$\langle f(t) \hat{v}(q, 0) \rangle = 0, \quad (9)$$

which expresses that the noise is uncorrelated with the initial velocity fluctuations.

The system of hydrodynamic Eqs. (4) and (7) describe the fluctuations as a non-Markovian, stationary and Gaussian process [15] and can be recast as a closed second order differential stochastic equation for density fluctuations $X(t) \equiv \delta\hat{\rho}(q, t)$,

$$\ddot{X}(t) + \int_0^t dt' \gamma(t-t') \dot{X}(t') + \omega_T^2 X(t) = f(t), \quad (10)$$

where the memory kernel is related to the longitudinal modulus, $\gamma(t) \equiv (q^2/\rho_0)M(t)$, with $\omega_T^2 \equiv (q^2/\rho_0\chi_T)$. If $X(0) = 0$, Eq. (10) has the same mathematical form as the *GLE* used in the literature to describe the non-Markovian Brownian motion of a particle in a fluid with memory in the presence of an harmonic external field [16,17]. However, our *GLE* (10) *does not describe the Brownian motion of a particle, rather, it describes the stochastic dynamics of a hydrodynamic stochastic variable, namely, the density fluctuations of the fluid.*

In Eqs. (8) and (9) the angular brackets denote an average over the realizations of the noise and over an equilibrium ensemble of initial conditions. Apart from being in the linear response regime, where thermal fluctuations are Gaussian and are dealt with a variety of standard methods, the rationale for assuming a Gaussian noise has some experimental justification. This Gaussian assumption has been experimentally shown to be adequate for other complex systems involving, for instance, the motion of tracers suspended in a fluid of swimming microorganisms [18]. In this active system the displacement of the tracers has a self-similar probability density function with a Gaussian core and exponential tails [19], a behavior of the tails which is actually a non-Gaussian spatial effect. However, since in this work we are only concerned with time memory kernels and not with spatial effects, it is a reasonable and more manageable assumption to consider a Gaussian noise with a long-time correlation.

Although $\gamma(t) \equiv (q^2/\rho_0)M(t)$ contains dissipation effects through the viscoelastic longitudinal modulus, it may have the same or different physical origin than the *noise term* $f(t)$, that is, it may be interpreted as internal or external noise. In the first case the second fluctuation-dissipation theorem is valid, whereas in the second case it has no relation with $\gamma(t)$, *i.e.*, a fluctuation-dissipation theorem does not necessarily exist. When the system is in an equilibrium state, it is expected that the viscoelastic kernel $\gamma(t)$ should be related to the correlation function of the noise $C(t)$ via the fluctuation-dissipation theorem (*FDT*) [20]. However, for non-equilibrium states the driving noise and the dissipation may have different origins and a fluctuation-dissipation relation does not necessarily holds. Although on physical grounds it is expected that the *FDT* should hold for the present model, to assume its validity is a rather strong assumption that requires justification. In previous works we have shown that this is indeed the case for the present hydrodynamic model and that the usual relation between fluctua-

tions and dissipation holds, [21,22].

$$C(t) = k_B T \gamma(t), \quad (11)$$

where k_B is Boltzmann's constant and T is the absolute temperature of the fluid.

Since only the longitudinal velocity fluctuations δv couple to the density fluctuations $\delta\rho$, to describe their effects on the structure factor of the liquid, we first calculate the (longitudinal) velocity fluctuations correlation function, $\langle X(t)X(t') \rangle$, and then we evaluate the (Rayleigh) light scattering spectrum in terms of $\gamma(t)$. The formal solutions of Eqs. (4) and (7) are, respectively, [23],

$$X(t) = \langle X(t) \rangle_{X_0 V_0} + \int_0^t H(t-t') f(t') dt', \quad (12)$$

$$V(t) \equiv \frac{dX(t)}{dt} = \langle V(t) \rangle_{X_0 V_0} + \int_0^t h(t-t') f(t') dt', \quad (13)$$

with

$$\langle X(t) \rangle_{X_0 V_0} = X_0 \left[1 - \omega_T^2 \int_0^t H(t') dt' \right] + V_0 H(t), \quad (14)$$

$$\langle V(t) \rangle_{X_0 V_0} = -\omega_T^2 \int_0^t h(t') dt' + V_0 h(t). \quad (15)$$

The relaxation functions $H(t)$ and $h(t) \equiv dH(t)/dt$ are defined in terms of the Laplace transform of the (so far arbitrary) kernel $\gamma(t)$ by

$$\widehat{H}(s) = \frac{1}{s^2 + s\widehat{\gamma}(s) + \omega_T^2}, \quad s = i\omega, \quad (16)$$

$$\widehat{h}(s) = \frac{1}{s + \widehat{\gamma}(s)}. \quad (17)$$

These expressions are general and valid for any $\gamma(t)$ and $C(t)$ with a well defined Laplace-Fourier transforms.

4. Power-law rheology

In what follows we shall only consider viscoelastic fluids with a power-law rheology such that

$$M(t) = M_0 t^{-\lambda}, \quad 0 < \lambda < 1, \quad (18)$$

with the Laplace transform

$$\widehat{M}(s) = M_0 \Gamma(1 - \lambda) s^{\lambda-1}, \quad (19)$$

with M_0 being the zero-frequency longitudinal modulus and $\Gamma(x)$ the Gamma function. The parameter λ measures the degree of viscoelasticity of the flow field; low values of λ correspond to large Weissenberg numbers (W_s), which in turn, imply a weakly elastic flow field, *i.e.*, more viscoelastic than

elastic. In contrast, a large λ indicates an exceedingly elastic flow. Thus, consistency with the assumed long-time memory kernel, $\gamma(t) \equiv (q^2/\rho_0 M(t))$, we should consider also a noise with a power-law decaying correlation function of the form

$$C(t, t') = \langle f(t)f(t') \rangle = k_B T C(|t - t'|) \equiv C_\beta t^{-\beta}, \quad 0 < \beta < 1. \quad (20)$$

In this case the inverse Laplace transforms of (16) and (17) can be calculated analytically in the interval $0 < \alpha < 1$, with the results [23]

$$H(t) = t^\alpha E_{1+\alpha-\lambda, 1+\alpha} [-M_0 \Gamma(1-\lambda) t^{1+\alpha-\lambda}] \quad (21)$$

and

$$h(t) = t^{\alpha-1} E_{1+\alpha-\lambda, \alpha} [-M_0 \Gamma(1-\lambda) t^{1+\alpha-\lambda}], \quad (22)$$

where

$$E_{\mu, \nu} = \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(\nu + \mu n)} \quad (23)$$

is the generalized Mittag-Leffler function (also known as Erdelyi's function). Note that since the viscoelastic fluid in our model complies with the physical conditions assumed for the derivation of Eq. (11), from Eqs. (18)-(20) it follows that the *FDT* reads

$$C_\beta = k_B T M_0 t^{\beta-\lambda}, \quad (24)$$

with $0 < \beta < 1$ and $0 < \lambda < 1$.

The intensity distribution of the isotropic components of the light scattering detected at the scattering angle θ , is proportional to the Fourier transform of the (one-time) correlation of the density fluctuation $\delta\rho(\vec{r}, t)$ given by

$$\begin{aligned} \widehat{C}(q, t) &= \langle \widehat{X}(q, t) X_0(q) \rangle \equiv \langle \delta\widehat{\rho}(q, t) \delta\widehat{\rho}(q) \rangle \\ &\equiv \langle \langle \delta\widehat{\rho}(q, t) \rangle_{\delta\widehat{\rho}(q)} \delta\widehat{\rho}(q) \rangle^{eq}, \end{aligned} \quad (25)$$

where $\delta\widehat{\rho}(q, t)$ is the spatial Fourier transform of the fluctuations of the density from the equilibrium value ρ_0 within the scattering volume. Here the notation indicates the following: take a certain initial value $\delta\widehat{\rho}(q)$ at $t = 0$, calculate the average $\langle \delta\widehat{\rho}(q, t) \rangle_{\delta\widehat{\rho}(q)}$ conditional on the given $\delta\widehat{\rho}(q)$; multiply it by $\delta\widehat{\rho}(q)$ and average the product over the values $\delta\widehat{\rho}(q)$, as they occur in an equilibrium distribution. Following a procedure described in detail in the Appendix of Ref. [21], in the limit of long-times ($t \rightarrow \infty$) we get the result

$$\widehat{C}(q, t \rightarrow \infty) \sim \frac{q^2}{\rho_0 \omega_T^2} \frac{1}{\Gamma(-3-\lambda)} \frac{1}{t^\lambda}. \quad (26)$$

If the general form of $\widehat{C}(q, t)$, Eq. (25), is constructed from the general formal solutions (12)-(17), and calculating its

Laplace transform with respect to t , one arrives at

$$\begin{aligned} \widetilde{C}(q, s) &\equiv \frac{\langle \langle \delta\widehat{\rho}(q, s) \rangle_{\delta\rho_0} \delta\rho^*(q) \rangle}{\langle |\delta\rho_0|^2 \rangle} \\ &= \frac{[s + q^2 \rho_0^{-1} \widehat{M}(s)]}{s [s + q^2 \rho_0^{-1} \widehat{M}(s)] + \omega_T^2}. \end{aligned} \quad (27)$$

Furthermore, by setting $s = i\omega$ and taking the real part of the r.h.s. of Eq. (27) we arrive at [24]

$$\begin{aligned} \widetilde{S}(q, \omega) &= \left[\frac{\text{Re}}{\pi} \widetilde{C}(q, s) \right]_{s=i\omega} \\ &= \frac{\text{Re}}{\pi} \left\{ \frac{[s + q^2 \rho_0^{-1} \widehat{M}(s)]}{s [s + q^2 \rho_0^{-1} \widehat{M}(s)] + \omega_T^2} \right\}_{s=i\omega}, \end{aligned} \quad (28)$$

which can also be expressed in terms of the complex longitudinal modulus $\widehat{M}(s)$

$$i\omega \widehat{M}(i\omega) = i\omega \int_0^\infty M(t) e^{-i\omega t} dt = M'(\omega) + i\omega M''(\omega), \quad (29)$$

where $M'(\omega)$ and $M''(\omega)$ are, respectively, the real and imaginary parts of $\widehat{M}(i\omega)$. Furthermore, the scattering light spectrum $I(q, \omega)$ is given by [25]

$$\begin{aligned} I(q, \omega) &= \frac{k_B T}{\pi} \widehat{S}(q) \left[\widetilde{S}(q, s) \right]_{s=i\omega} = \frac{k_B T}{\pi} \widehat{S}(q) \\ &\times \frac{q^2 \rho_0^{-1} \omega_T^2 M''(\omega)/\omega}{[\omega_T^2 - \omega^2 + q^2 \rho_0^{-1} M'(\omega)]^2 + (q^2 \rho_0^{-1})^2 [M''(\omega)]^2}. \end{aligned} \quad (30)$$

This Eq. (30) allows us to calculate the light scattering spectrum as a function of an arbitrary longitudinal modulus $M(t)$, as long as it has a well-defined Laplace transform.

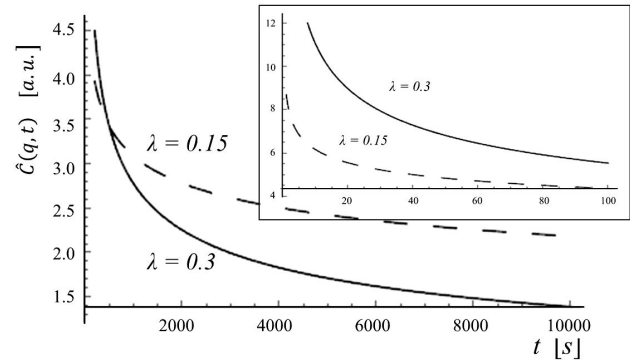


FIGURE 1. The behavior of $\widehat{C}(q, t \rightarrow \infty)$, as given by Eq. (26), for the material properties of liquid salol: $T = 353.2$ K, $M_0 = 3.2G_0$, $G_0 = 1.154$ Kg/ms, $\rho_0 = 1.212$ Kg/m³. The curves correspond to $\lambda = 0.3$ and $\lambda = 0.15$ [26].

5. Results and discussion

The long time behavior of (26) is shown in Fig. 1 as a function of t and λ for the values $\lambda = 0.15$ and $\lambda = 0.3$. The inset shows that for the time interval (4 s, 100 s) $\widehat{C}(q, t \rightarrow \infty, \lambda = 0.3)$ is always larger than $\widehat{C}(q, t \rightarrow \infty, \lambda = 0.15)$; however, for longer times, ($10^3 - 10^4$ s), there is a crossover and the latter correlation is always greater than the former. The difference between these quantities was quantified for liquid

Salol for the material values given in the Fig. 1 caption. As shown in the figure, this difference can be quite significant, for instance, for $t \sim 10^4$ s it is $\sim 62\%$, and it may vary between 56.1-70.7% for the time interval 510-6000 s, and it might be measurable. However, ultimately this behavior has its origin in the microscopic processes generating the mesoscopic dynamics that we are modeling and to describe it is certainly outside the scope of the present work.

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- i.* This article is dedicated to the memory of the late Professor G. Cocho as a token of admiration to his work and his strong influence on current developments in physics and in many interdisciplinary fields.
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