Fractional fluctuation effects on the light scattered by a viscoelastic suspension

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We generalize fluctuating hydrodynamics to study the effect of fractional time derivatives on the light-scattering spectrum of a suspension in a viscoelastic solvent under an external density gradient. Viscoelasticity introduces additional memory effects into the fluctuating hydrodynamic equations, causing the time scales associated with the mesoscopic variables and those of the microscopic events to be no longer well separated. This situation is taken into account by introducing Caputo's fractional time derivative into the description. The structure factor of the suspension is calculated, and we find that its nonequilibrium correction is an odd function of the frequency. It exhibits a shift towards negative frequencies proportional to the magnitude of the imposed gradient. We consider solvents that are described by Maxwell's or power-law rheological equations of state. The fractional structure factor is compared with the nonfractional one, and it is found that the ratio of the former to the latter may be positive and up to two orders of magnitude for both types of viscoelasticity. This prediction of our model calculation suggests that this relative change might be measurable.

I. INTRODUCTION

In classical statistical physics the separation of the microscopic and macroscopic time scales is manifest in the central limit theorem, and this separation implies that the macroscopic dynamics can be described by the ordinary stochastic differential calculus, even if the microscopic dynamics is incompatible with the methods of ordinary calculus [1]. Perhaps the most familiar situation where this issue arises is Brownian motion, where the classical Langevin equation successfully describes the motion of the Brownian particle [2]. In this equation the Langevin force acting on the particle is a zero-centered Gaussian random force with instantaneous correlation $C(t)$. However, when the particle moves through a stationary environment like a dense fluid or fluids with internal degrees of freedom [3], the motion of the particle should be described instead with the generalized Langevin equation [4]

$$\dot{V} + \int_0^t \gamma(t - t')V(t')\, dt' + f(V) = F(t), \qquad (1)$$

where now the effect of the environment is encoded in a retarded friction kernel $\gamma(t)$ and in a noise term $F(t)$. In this case the correlation $C(t - t')$ is arbitrary, and, due to the presence of the memory kernel $\gamma(t)$, the time scales associated with the time evolution of V, and the microscopic events giving rise to it, are no longer well separated. It has been widely discussed in the literature that, strictly speaking, when this separation of time scales does not exist, the formalism of ordinary statistical physics is no longer adequate to describe the phenomenon, and the use of fractional, stochastic differential equations on the mesoscopic level should replace the conventional Langevin equation [5–7]. Actually, fractional Brownian motion has been widely used in the modeling of various physical phenomena which exhibit anomalous (non-Gaussian) diffusion and has given rise to a huge literature [8,9].

This lack of separation of time scales may also occur in the description of transport dynamics in complex systems which are governed by anomalous diffusion and nonexponential relaxation patterns. For these systems fractional equations are derived asymptotically from basic random walk models and from generalized master equations [9]. However, although these effects have been well studied for Brownian motion and diffusive systems, much less is known of their implications for hydrodynamic systems. As will be seen in this paper, this lack of separation of time scales also occurs in the description of the dynamics of fluctuations of a hydrodynamic system. Indeed, a fluctuating hydrodynamics description may comprise equations for the time evolution of velocity or density fluctuations which are governed by generalized Langevin equations with a time-dependent friction coefficient exhibiting dissipative memory due to viscoelasticity.

The Landau and Lifshitz theory of hydrodynamic fluctuations [10] close to equilibrium has been put on a firm basis within the framework of the general theory of stationary Gaussian Markov processes by Fox and Uhlenbeck [11,12]. In fluctuating hydrodynamics the usual deterministic hydrodynamic equations are supplemented with random dissipative fluxes of thermal origin, obeying a fluctuation-dissipation relation. This approach has matched the theory of Onsager and Machlup with the approach of Landau and Lifshitz, for systems where the basic state variables do not posses a definite time reversal symmetry [13–15], leading to Langevin-like stochastic equations for the evolution of the fluctuations of the state variables. In this way fluctuating hydrodynamics provides a systematic method for assessing the nature of spontaneous fluctuations induced by intrinsic thermal noise. Fox and Uhlenbeck's scheme has been applied to simple fluids and their binary mixtures [16,17]; more recently it has been also verified that fluctuating hydrodynamics can be extended to deal with thermally excited fluctuations in fluids in stationary nonequilibrium states [18–21]. In spite of the fact that the theory of fluctuations in nonequilibrium fluids was initiated in the late 1970s [22–26], and pursued by many authors [27,28],

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still nowadays several questions concerning the nature of hydrodynamic fluctuations in nonequilibrium stationary states (NESSs) are of current active interest. One of these issues is the long-range character of these fluctuations even far away from instability points. It has been shown theoretically that the existence of the so-called generic scale invariance is the origin of the long-range nature of the correlation functions [18,29]. However, in spite of the considerable interest in fluctuations around dissipative steady states of simple fluids during the last two decades, there are few similar studies for equilibrium or nonequilibrium stationary states of complex fluids [30–35].

On the other hand, models where fractional differential equations have been obtained by formally replacing first-order time derivatives by fractional ones have been proposed for several definitions of fractional derivatives, such as those of Riemann-Liouville, Grünwald-Letnikov, and Caputo [36–38]. These models describe relaxation processes occurring in the behavior of complex systems like viscoelastic fluids, glassy materials, synthetic polymers, or biopolymers. All these systems have in common that their relaxation function is nonexponential due to the large number of highly coupled elementary units responsible for the relaxation. The requirement of high cooperation among these elements leads to slower decays often modeled by empirical power laws [39–41]. In particular, fractional models have been developed to describe relaxation processes whose solutions can be represented by Fox functions [42]. Besides the relaxation function, other viscoelastic functions, such as the retardation function, the storage, and loss modulus, the relaxation time spectrum, and the retardation time spectrum, have been studied and are expressed by analytical functions. This wide class of functions offers a framework within which nonstandard relaxation processes can be discussed [43,44].

The main aim of the present paper is to consider some of these questions within the context of a simple hydrodynamic model of a viscoelastic suspension. Fractional time derivatives are introduced into the hydrodynamic equations to study their effect on the light-scattering properties of a viscoelastic suspension in a NESS. More precisely, the purpose of doing this is to investigate the effects of *fractional fluctuations* on a measurable property, the light-scattering dynamic structure factor of the suspension. To our knowledge, the study of the effects of fractional derivatives on the behavior of this property has not been considered in the literature so far, where the interest in fractional effects has been focused on Brownian motion, random walk, and anomalous diffusion. In this work we will use the Caputo fractional derivative, which, according to Ref. [45], has the following intuitive interpretation: it takes into account the past behavior of the fluctuations themselves, implying that apart from the usual dissipative memories associated with viscoelasticity, an additional memory is constructed by adding to their stationary values the successive weighted increments over time. In this sense its presence gives rise to *fractional fluctuations* in addition to the usual *thermal fluctuations* existing in the system. To assess the effect of these two different memories (fluctuations), we calculate the relative change of a quantity that is affected by them, namely, the light-scattering dynamic structure factor of the suspension. We evaluate this quantity when fractional fluctuations are present and compare it

with its value due only to thermal fluctuations. For this purpose we consider two different viscoelastic solvents, those obeying Maxwell's and a power-law rheological equations of state.

The plan of the paper is as follows. In Sec. II we recall the relevant aspects of a model, introduced in our previous work [46], for a dilute Brownian suspension under an externally imposed constant concentration gradient. Then in Sec. III we derive the fluctuating linearized hydrodynamic equations for the particles and the solvent. We define the NESSs to be considered and show that they are a solution of those equations. After introducing the Caputo fractional time derivative, we show that its presence affects only the Green functions of the ensuing fractional stochastic equations for the velocity and concentration fluctuation, arguing that the corresponding fractional fluctuation-dissipation relations may be the same as for a NESS with integer-valued time derivatives. These equations are then solved analytically by using Fourier transforms. In Sec. IV we calculate the fractional correlation function of concentration fluctuations and derive a general explicit formal expression for the equilibrium and nonequilibrium components of the fractional dynamic structure factor. This expression is valid up to first order in the external gradient, but for an arbitrary form of the frequency-dependent generalized viscosity and diffusion coefficients, as long as they have a well-defined Fourier transform. For the chosen NESS, in Sec. V we consider two specific rheological solvents, salol (*S*) which is a glass-forming liquid, and a power-law fluid, a solution of separan in glucose (E_1), which is an exceedingly elastic fluid. In Sec. VI we calculate the corresponding fractional dynamic structure factors. To quantify the difference observed in the structure factor for both fluids, we calculate the ratio of the fractional to the nonfractional spectra for frequencies and wave numbers typical of a light-scattering experiment. We find that for derivatives of fractional order α in $0 < \alpha < 1$ this ratio can be significantly large, up to two orders of magnitude. In marked contrast, we find for the range $1 < \alpha < 2$, the fractional fluctuations contribute much less to the structure factor, indicating that the former fractional fluctuations are the dominant ones. The size of this relative change suggests that these fractional effects might be measurable. However, we are not aware of any experimental results to compare with the predictions of our model, and therefore it is not possible to conclude from our analysis if this (significant) enhancement is measurable. This is an open issue that remains to be assessed. Finally, in Sec. VII we summarize the main results of our work and make some further remarks.

II. BASIC EQUATIONS

A. Model

Consider a dilute suspension of noninteracting identical spherical particles of mass m and radius a moving through a linear, viscoelastic solvent with spatially homogeneous viscoelastic moduli, and which may flow with velocity $\overrightarrow{v}(\overrightarrow{r},t)$ [46]. Following Ref. [47], the most general constitutive equation for the linear stress-strain relation of this solvent

is of the form

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

where Einstein's summation convention for repeated indices is implied. Here $\sigma_{ij}(\overrightarrow{r},t)$ is the symmetric stress tensor, $p(\overrightarrow{r})$ is the hydrostatic pressure, and $\dot{\gamma}_{ij}(\overrightarrow{r},t)$ is the rate of strain tensor defined in terms of the velocity gradient tensor, $\dot{\gamma}_{ij} \equiv \partial v_i / \partial x_j$. $\overrightarrow{r}$ stands for the position vector with Cartesian components x_i, $i = x,y,z$. The scalar functions $G(t)$ and $K(t)$ denote the shear and the bulk (compressional) components of the stress modulus, respectively, and are assumed to be independent of $\overrightarrow{r}$.

Since at the temperatures of interest in a photon correlation experiment the thermal contribution to the density fluctuation spectrum is outside the range that can be monitored by this technique, in a first approximation the effects of thermal diffusion will be neglected and temperature fluctuations may be considered to be sufficiently small [10]. Hence, under isothermal conditions the solvent may be regarded as incompressible, and if the suspension is so diluted that the presence of the particles does not appreciable perturb the motion of the fluid, the dynamics of the solvent is described by the continuity equation

$$\overrightarrow{\nabla} \cdot \overrightarrow{v} = 0 \quad (3)$$

and the equation of motion

$$\rho(\overrightarrow{r},t)\frac{\partial}{\partial t}\overrightarrow{v}(\overrightarrow{r},t)$$
$$= -\overrightarrow{\nabla} p(\overrightarrow{r},t) - \rho(\overrightarrow{r},t)\overrightarrow{v}(\overrightarrow{r},t) \cdot \overrightarrow{\nabla}\overrightarrow{v}(\overrightarrow{r},t)$$
$$+ \int_{-\infty}^t dt' G(t-t')\nabla^2 \overrightarrow{v}(\overrightarrow{r},t'), \quad (4)$$

where $\rho(\vec{r},t)$ is the mass density. In writing this equation we have assumed that the fluid has been put in a stationary state (to be defined below) in the remote past, as indicated by the lower integration limit $-\infty$ in the retarded modulus.

Let us now consider the suspended particles. If no chemical reactions occur, the total mass of the solute is conserved, and their local number density $n(\vec{r},t)$ obeys the generalized Fick's law

$$\frac{\partial}{\partial t}n(\overrightarrow{r},t) + \overrightarrow{\nabla} \cdot \overrightarrow{J}(\overrightarrow{r},t) = 0, \quad (5)$$

where the flux of particles displays memory effects due to the viscoelasticity of the solvent

$$\overrightarrow{J}(\overrightarrow{r},t) = -\int_{-\infty}^t D(t-t')\overrightarrow{\nabla}n(\overrightarrow{r},t')\,dt'$$
$$+ n(\overrightarrow{r},t)\overrightarrow{v}(\overrightarrow{r},t). \quad (6)$$

This time memory allows the diffusion coefficient $D(t)$ to depend on times t' previous to the observation time t. The second term on the r.h.s. is the convective flux of particles arising from the flow of the solvent. Thus, the generalized

diffusion equation governing the time evolution of $n(\overrightarrow{r},t)$ is

$$\frac{\partial}{\partial t}n(\overrightarrow{r},t) + \overrightarrow{v}(\overrightarrow{r},t) \cdot \overrightarrow{\nabla}n(\overrightarrow{r},t)$$
$$= \int_{-\infty}^t dt' D(t-t')\nabla^2 n(\overrightarrow{r},t'). \quad (7)$$

Clearly, if $K(t)$, $G(t)$, and $D(t)$ are known, Eqs. (3), (4), and (7) form a complete system of hydrodynamic equations for the hydrodynamic state variables $v_i(\overrightarrow{r},t)$ and $n(\overrightarrow{r},t)$.

B. Fractional derivatives

We now generalize the hydrodynamic model described by Eq. (3), (4), and (7) by replacing formally the first-order time derivatives on the l.h.s. of these equations by fractional order derivatives. As is well known, there are many definitions for the fractional derivatives, such as those of Riemann-Liouville, Grünwald-Letnikov, and Caputo. Here we choose the left-handed Caputo fractional derivative of order α defined by [37,38],

$$_0^C D_t^\alpha f(t) = \frac{1}{\Gamma(n-\alpha)} \int_0^t \frac{f^{(n)}(\tau)}{(t-\tau)^{\alpha+1-n}}\,d\tau, \quad (8)$$

where $f^{(n)} \equiv d^n f/dt^n$ denotes the conventional n order time-derivative, being n the smallest integer greater than α, i.e., the one that satisfies $n-1 \leqslant \alpha < n$, and $\Gamma(n-\alpha)$ is the Euler gamma function. It should be noted that when $\alpha = 1$, the operator $_0^C D_t^\alpha f(t)$ reduces to the conventional derivative $df(t)/dt$, so that $_0^C D_t^{\alpha=1}\overrightarrow{v}(\overrightarrow{r},t)$ and $_0^C D_t^{\alpha=1}n(\overrightarrow{r},t)$ reduce to $\partial\overrightarrow{v}(\overrightarrow{r},t)/\partial t$ and $\partial n(\overrightarrow{r},t)/\partial t$, respectively. The operator D_t^α then converts Eqs. (4) and (7) into fractional equations. Following Ref. [45], we stress that as can be seen from Eq. (8), "the fractional derivative takes into account the past behavior of $\overrightarrow{v}(\overrightarrow{r},t)$ and $n(\overrightarrow{r},t)$, implying that these fields are constructed by adding to their initial values the successive weighted increments over time. Therefore, the values of $\partial\overrightarrow{v}(\overrightarrow{r},\tau)/\partial\tau$ at a time τ far apart from t, have smaller weights than those at times τ closer to t. These increments per unit time are represented by $f^{(n)}(\tau)$ under the integral sign in Eq. (8), while the weights are indicated by the factors $1/(t-\tau)^{\alpha+1-n}$, whose value decreases with increasing time separation from time t." In this sense the memory contained in the fractional derivative is different from the memory described by the dissipative kernels like $K(t-t')$, $G(t-t')$ and $D(t-t')$. The value of a variable is actually a weighted average of its past values, and the fractional derivative represents a different type of additional memory. Since Eqs. (4) and (7) have conventional first order time derivatives ($\alpha = 1$), it is to be expected that their fractional generalizations should have $\alpha \approx 1$, either $\alpha < 1$, (i.e., $0 < \alpha < 1$), or $\alpha > 1$ (i.e., $1 < \alpha < 2$). For the interval $0 < \alpha < 1$ Eq. (8) reduces to

$$_0^C D_t^{\alpha<1} f(t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t \frac{f^{(1)}(\tau)}{(t-\tau)^\alpha}\,d\tau, \quad (9)$$

and it describes the cumulative effect of the first derivative $f^{(1)}(\tau)$ over past times. On the other hand, in the case where $1 < \alpha < 2$, the fractional derivative involves $f^{(2)}(\tau)$, but it is still an approximation to the first derivative $f^{(1)}$. However, as will be shown later, the dominant contribution to the

dynamic structure factor will arise from the values of α in the interval $0 < \alpha < 1$. In the next section we will write fractional equations for the fluctuations of the state variables which represent *fractional fluctuations,* in addition to the usual thermal fluctuations.

Therefore, the hydrodynamic equations of the system are now the incompressibility condition Eq. (3), the equation of motion

$$\rho(\overrightarrow{r},t) {}_0^C D_t^\alpha \overrightarrow{v}(\overrightarrow{r},t)$$
$$= -\overrightarrow{\nabla} p(\overrightarrow{r},t) - \rho(\overrightarrow{r},t)\overrightarrow{v}(\overrightarrow{r},t) \cdot \overrightarrow{\nabla} \overrightarrow{v}(\overrightarrow{r},t)$$
$$+ \int_{-\infty}^t dt' G(t-t')\nabla^2 \overrightarrow{v}(\overrightarrow{r},t') \tag{10}$$

and the generalized diffusion equation

$$_0^C D_t^\alpha n(\overrightarrow{r},t) = -\overrightarrow{v}(\overrightarrow{r},t) \cdot \overrightarrow{\nabla} n(\overrightarrow{r},t)$$
$$+ \int_{-\infty}^t D(t-t')\nabla^2 n(\overrightarrow{r},t')\,dt'. \tag{11}$$

If $G(t)$ and $D(t)$ are known, Eqs. (3), (10), and (11) form a complete system of hydrodynamic equations for the hydrodynamic state variables $v_i(\overrightarrow{r},t)$ and $n(\overrightarrow{r},t)$.

C. Stationary states

The stationary solutions of Eqs. (3), (10), and (11) define nonequilibrium steady states determined by the boundary conditions imposed on the system. Here we shall consider only the stationary solution corresponding to a constant concentration gradient $\overrightarrow{\nabla} n_s$ in the direction of $\overrightarrow{r}$ corresponding to a quiescent solvent for which

$$n_s(\overrightarrow{r}) = n_0 + \overrightarrow{r} \cdot \overrightarrow{\nabla} n_s, \tag{12}$$

$$\overrightarrow{v}_s(\overrightarrow{r}) = 0, \tag{13}$$

with constant density ρ_0 and pressure

$$p_s(\overrightarrow{r},t) = \text{const.} \tag{14}$$

For later convenience we rewrite the stationary solution (12) in the form

$$n_s(\overrightarrow{r}) = n_0 + \overline{n}\sin(\overrightarrow{q} \cdot \overrightarrow{r}), \tag{15}$$

with

$$\overrightarrow{q}\,\overline{n} \equiv \overrightarrow{\nabla} n_s. \tag{16}$$

Of course, Eq. (15) reduces to Eq. (12) to first order in $\overrightarrow{q}$, but it is easier to perform the calculations using (15) and taking the limit $\overrightarrow{q} \to 0$ at the end.

To be consistent with linear response theory, Eqs. (3), (10), and (11) should be linearized in the small deviations of the state variables with respect their steady-state values (12). These quantities are defined by $\delta n(\overrightarrow{r},t) \equiv n(\overrightarrow{r},t) - n_s(\overrightarrow{r})$, $\delta p(\overrightarrow{r},t) \equiv p(\overrightarrow{r},t) - p_0$, and $\delta \overrightarrow{v}(\overrightarrow{r},t) \equiv \overrightarrow{v}(\overrightarrow{r},t)$ due to Galilei invariance. Moreover, since due to causality, $G(t)$ and $D(t)$ contain the Heaviside step function $\Theta(t - t') = 1$, for $t > t'$ and zero otherwise, we arrive at the following linearized continuity equation:

$$\overrightarrow{\nabla} \cdot \overrightarrow{v} = 0, \tag{17}$$

the equation of motion

$$_0^C D_t^\alpha \overrightarrow{v}(\overrightarrow{r},t)$$
$$= -\rho_0^{-1}\overrightarrow{\nabla}\delta p + \rho_0^{-1}\int_0^t dt' G(t-t')\nabla^2 \overrightarrow{v}(\overrightarrow{r},t'), \tag{18}$$

and the generalized linear diffusion equation

$$_0^C D_t^\alpha \delta n(\overrightarrow{r},t) + \overrightarrow{v} \cdot \overrightarrow{\nabla} n_s = \int_0^t D(t-t')\nabla^2 \delta n(\overrightarrow{r},t')\,dt'. \tag{19}$$

III. FRACTIONAL FLUCTUATING HYDRODYNAMICS

We now introduce thermal fluctuations into Eqs. (18) and (19) on the basis of Landau's fluctuating hydrodynamics [10–12]. This is accomplished by adding a random current, $\overrightarrow{J}^R(\overrightarrow{r},t)$, to the mass flux Eq. (6) and random stress tensor $\overleftrightarrow{\Pi}^R(\overrightarrow{r},t)$ into Eq. (18); they have their origin in the stochastic nature of molecular collisions and lead to

$$_0^C D_t^\alpha \overrightarrow{v}(\overrightarrow{r},t) = -\rho_0^{-1}\overrightarrow{\nabla}\delta p + \rho_0^{-1}\int_0^t dt' G(t-t')$$
$$\times \nabla^2 \overrightarrow{v}(\overrightarrow{r},t') + \overrightarrow{\nabla} \cdot \overleftrightarrow{\Pi}^R(\overrightarrow{r},t), \tag{20}$$

$$_0^C D_t^\alpha \delta n(\overrightarrow{r},t) + \overrightarrow{v} \cdot \overrightarrow{\nabla} n_s$$
$$= \int_0^t dt' D(t-t')\nabla^2 \delta n(\overrightarrow{r},t') - \overrightarrow{\nabla} \cdot \overrightarrow{J}^R(\overrightarrow{r},t). \tag{21}$$

These random terms $\overrightarrow{J}^R(\overrightarrow{r},t)$ and $\overleftrightarrow{\Pi}^R(\overrightarrow{r},t)$ are modeled by Gaussian, stationary, non-Markovian stochastic processes with zero mean

$$\langle \overleftrightarrow{\Pi}^R(\overrightarrow{r},t)\rangle = 0, \tag{22}$$

$$\langle \overrightarrow{J}^R(\overrightarrow{r},t)\rangle = 0, \tag{23}$$

and correlations given by the following fluctuation-dissipation theorems (FDTs)

$$\left\langle \Pi_{ij}^R(\overrightarrow{r},t)\Pi_{lm}^R(\overrightarrow{r'},t')\right\rangle = 2kT G(|t-t'|)\delta(\overrightarrow{r} - \overrightarrow{r'})\Delta_{ijlm}, \tag{24}$$

$$\left\langle J_i^R(\overrightarrow{r},t)J_j^R(\overrightarrow{r'},t')\right\rangle = 2n_s(\overrightarrow{r})D(|t-t'|)\delta(\overrightarrow{r} - \overrightarrow{r'})\delta_{ij}. \tag{25}$$

Here k is Boltzmann's constant, T is the temperature of the solvent, $\overleftrightarrow{1}$ is the unit tensor, Δ_{ijlm} is defined by

$$\Delta_{ijlm} \equiv \delta_{il}\delta_{jm} + \delta_{im}\delta_{jl} - \tfrac{2}{3}\delta_{ij}\delta_{lm}, \tag{26}$$

and δ denotes the Kronecker's delta. These relations are derived from the expression for the rate of change of the total entropy of the liquid and by following the general rules of fluctuation theory [48]. In Eqs. (24) and (25) the angular brackets denote an average over the NESS and actually imply that we have assumed fractional FFDTs which have the same form as those valid in generalized hydrodynamics for NESS. Admittedly, this is a strong assumption which has to be justified. We discuss the plausibility of this assumption below, after deriving Eqs. (39) and (40).

Again, if $G(t)$ and $D(t)$ are given, Eqs. (20) and (21), together with (22)–(26), constitute a closed set of linear

stochastic equations, or generalized Langevin equations, describing the dynamics of the thermal fluctuations of the system. Equation (20) can be further simplified by eliminating the gradient term by applying the operator $\vec{\nabla} \times \vec{\nabla} \times$ and using the incompressibility condition (17), with the result

$$
{}_0^C D_t^\alpha \vec{v}(\vec{r},t) = \rho_0^{-1} \int_0^t dt' G(t-t') \nabla^2 \vec{v}(\vec{r},t') \\
+ \vec{\nabla} \cdot \overleftrightarrow{\Pi}^R(\vec{r},t). \quad (27)
$$

Thus, the generalized Langevin equations (27) and (21) describe the dynamics of both fractional and thermal fluctuations.

We now define the time-space Fourier transform of an arbitrary field $A(\vec{r},t)$ by

$$
\widetilde{A}(\vec{k},s) \equiv \mathcal{F}\{A(\vec{r},t)\} \\
= \int d\vec{r} \int_{-\infty}^{+\infty} dt\, e^{i\vec{k}\cdot\vec{r}} e^{-i\omega t} A(\vec{r},t). \quad (28)
$$

In what follows the caret $\widehat{A}$ will denote the Fourier transform of A with respect to one of its variables and the tilde $\widetilde{A}$ with respect to both. To this end it will be necessary to Fourier transform the fractional derivative ${}_0^C D_t^\alpha f(t)$ in Eq. (8). It can be proven that this transform is given by

$$
\mathcal{F}\left\{{}_0^C D_t^\alpha f(t)\right\} = (i\omega)^\alpha \widehat{f}(\omega), \quad (29)
$$

which has the same form as for conventional derivatives. Although this result could have been expected, it is worth pointing out that the proof of the validity of Eq. (29) is not as straightforward as that of the well-known identity

$$
\mathcal{F}\left\{{}_{-\infty}^C D_t^\alpha f(t)\right\} = (i\omega)^\alpha \widehat{f}(\omega), \quad (30)
$$

valid when the fractional derivative ${}_{-\infty}^C D_t^\alpha f(t)$ has minus infinity in the lower limit of the integral [36,38],

$$
{}_{-\infty}^C D_t^\alpha f(t) = \frac{1}{\Gamma(n-\alpha)} \int_{-\infty}^t \frac{f^{(n)}(\tau)\, d\tau}{(t-\tau)^{\alpha+1-n}}\, d\tau. \quad (31)
$$

Note that since we have a description in terms of the diffusion coefficient which is expected to be valid only at times much longer than a molecular time, the upper limit in Eqs. (21) and (27) may be extended to $+\infty$, and definition (28) can be used. Then the fluctuating linearized equations (17), (21), and (27) in $(\vec{k},\omega)$ space read

$$
\vec{k} \cdot \widetilde{\vec{v}}(\vec{k},\omega) = 0, \quad (32)
$$

$$
\delta\overline{n}(\vec{k},\omega) = -\widetilde{g}_n(\vec{k},\omega)[\vec{\nabla} n_s(\vec{k}) \cdot \widetilde{\vec{v}}(\vec{k},\omega) \\
+ i\vec{k} \cdot \widetilde{\vec{J}}^R(\vec{k},\omega)], \quad (33)
$$

$$
\widetilde{\vec{v}}(\vec{k},\omega) \\
= -\widetilde{g}_v(\vec{k},\omega)\{(\overleftrightarrow{1} - \widehat{k}\widehat{k}) \cdot [i\vec{k} \cdot \widetilde{\overleftrightarrow{\Pi}}^R(\vec{k},\omega)]\}, \quad (34)
$$

with $0 < \alpha < 1$, where $\widehat{k}$ is the unit vector along the direction of $\vec{k}$ and

$$
\widetilde{g}_v(\vec{k},\omega) = [(i\omega)^\alpha + k^2 \widehat{G}(\omega)]^{-1}, \quad (35)
$$

$$
\widetilde{g}_n(\vec{k},\omega) = [(i\omega)^\alpha + k^2 \widehat{D}(\omega)]^{-1} \quad (36)
$$

are the fractional Green functions in $(\vec{k},\omega)$ space associated with Eqs. (34) and (33), respectively. Note that Eq. (33) shows that out of equilibrium the concentration fluctuations

are coupled to the velocity fluctuations through the density gradient $\vec{\nabla} n_s$.

In Fourier space the fluctuation-dissipation relations (24) and (25) become

$$
\langle \widetilde{\Pi}_{ij}^R(\vec{k},\omega)\widetilde{\Pi}_{lm}^R(\vec{k}',\omega') \rangle \\
= 2(2\pi)\,\mathrm{Re}[\widehat{G}(\omega)]\delta(\omega+\omega')\delta(\vec{k}+\vec{k}')\widehat{\Delta}_{ijlm}(\vec{k}), \quad (37)
$$

$$
\langle \widetilde{\vec{J}}^R(\vec{k},\omega)\widetilde{\vec{J}}^R(\vec{k}',\omega') \rangle \\
= 2(2\pi)\,\mathrm{Re}[\widehat{D}(\omega)]\widehat{n}_s(\vec{k})\delta(\vec{k}+\vec{k}')\overleftrightarrow{1}, \quad (38)
$$

where $\widehat{n}_s(\vec{k})$ is

$$
\widehat{n}_s(\vec{k}) = (2\pi)^3 \left\{ n_0\delta(\vec{k}) + \frac{\overline{n}}{2i}[\delta(\vec{k}-\vec{q}) - \delta(\vec{k}+\vec{q})] \right\}. \quad (39)
$$

Intuitively, it may be expected the validity of the FFDTs (37) and (38) with the same structure as for a NESS. This is suggested by the fact that the fractional character of the solutions (33) and (34) shows up only in the Green functions (propagators), Eqs. (35) and (36), through their dependence on the exponent α ($0 < \alpha < 1$), the order of the fractional derivative. This in turn suggests that if after all the fractional dynamics has evolved and the action of the propagators has stopped, the final NESS is reached, it is likely that a FFDT exists, and if this is the case, it should have the same structure as the FDT in a NESS, since the fractional features of the dynamics are no longer present. This idea is further discussed in the Appendix, where we follow, adapt, and extend the method developed in Refs. [49,50] to sketch the validity of the FFDT.

IV. DYNAMIC STRUCTURE FACTOR

A. Equilibrium

When the concentration gradient vanishes ($\overline{n} = 0$), the system reaches thermodynamic equilibrium, and Eqs. (33) and (38) reduce to

$$
\delta\widetilde{n}(\vec{k},\omega) = -i\widetilde{g}_n(\vec{k},\omega)\vec{k} \cdot \widetilde{\vec{J}}^R(\vec{k},\omega), \quad (40)
$$

$$
\langle \widetilde{\vec{J}}^R(\vec{k},\omega)\widetilde{\vec{J}}^R(\vec{k}',\omega') \rangle \\
= 2(2\pi)^4 n_0\,\mathrm{Re}[\widehat{D}(\omega)]\delta(\vec{k}+\vec{k}')\delta(\omega+\omega')\overleftrightarrow{1}. \quad (41)
$$

The equilibrium density-density correlation function is then given by

$$
\langle \delta\widetilde{n}(\vec{k},\omega)\delta\widetilde{n}(\vec{k}',\omega') \rangle^{\mathrm{eq}} \\
= 2(2\pi)^4 n_0\,\mathrm{Re}[\widehat{D}(\omega)](-\vec{k} \cdot \vec{k}') \\
\times \widetilde{g}^n(\vec{k},\omega)\widetilde{g}^n(\vec{k}',\omega')\delta(\vec{k}+\vec{k}')\delta(\omega+\omega'). \quad (42)
$$

Since the only nonvanishing contribution to the structure factor is given by $\vec{k}' = -\vec{k}$ and $\omega' = -\omega$, the equilibrium structure factor $S^{\mathrm{eq}}(\vec{k},\omega)$ is

$$
S^{\mathrm{eq}}(\vec{k},\omega) \equiv \langle \delta\widetilde{n}(\vec{k},\omega)\delta\widetilde{n}(-\vec{k},-\omega) \rangle^{\mathrm{eq}} \\
= 2(2\pi)^4 n_0\delta^4(0)k^2\,\mathrm{Re}[\widehat{D}(\omega)]\widetilde{g}_n(\vec{k},\omega)\widetilde{g}_n^*(\vec{k},\omega), \quad (43)
$$

where the asterisk * denotes complex conjugation.

B. Nonequilibrium

The density fluctuations are given by Eq. (28) when the system is driven to a NESS by the presence of the density gradient $n_s(\vec{k})$. The fluctuation-dissipation theorem Eq. (36) in this case turns out to be

$$
\langle \widetilde{\overrightarrow{J}}^R(\vec{k},\omega)\widetilde{\overrightarrow{J}}^R(\vec{k}',\omega')\rangle
$$
$$
= 2(2\pi)^4 \operatorname{Re}[\widehat{D}(\omega)]\delta(\vec{k}+\vec{k}')\overleftrightarrow{1}\Bigg\{n_0\delta(\vec{k}+\vec{k}')
$$
$$
+ \frac{1}{2i}\widehat{n}(\vec{k})[\delta(\vec{k}+\vec{k}'-\vec{q})-\delta(\vec{k}+\vec{k}'+\vec{q})]\Bigg\}.
$$
(44)

The nonequilibrium density-density correlation function is constructed using Eqs. (28), (40), and (42) and the fact that $\overrightarrow{J}^F(\vec{k},\omega)$ and $\widetilde{\overrightarrow{v}}(\vec{k},\omega)$ are uncoupled. Up to first order in the density gradient this yields

$$
\langle\delta\widetilde{n}(\vec{k},\omega)\delta\widetilde{n}(\vec{k}',\omega')\rangle^{\mathrm{neq}}
$$
$$
= -\widetilde{g}_n(\vec{k},\omega)\widetilde{g}_n(\vec{k}',\omega')\Bigg\{2(2\pi)^4\operatorname{Re}[\widehat{D}(\omega)]\delta(\omega+\omega')\vec{k}\cdot\vec{k}'
$$
$$
\times \Bigg[n_0\delta(\vec{k}+\vec{k}')+\bar{n}/2i\sum_{\epsilon=\pm1}\epsilon\delta(\vec{k}+\vec{k}'-\epsilon\vec{q})\Bigg]\Bigg\}.
$$
(45)

Since the first term on the r.h.s. yields the equilibrium contribution to the density-density correlation function given by Eq. (37), we calculate only the nonequilibrium contribution proportional to $\widetilde{n}$. If in the second term on the r.h.s. of (45) we substitute $\vec{k}\to\vec{k}+\epsilon\vec{q}/2$ and $\vec{k}'\to\vec{k}'+\epsilon\vec{q}/2$, we get

$$
\langle\delta n(\vec{k},\omega)\delta n(\vec{k}',\omega')\rangle^{\mathrm{neq}}
$$
$$
= -\sum_{\epsilon=\pm1}\epsilon 2(2\pi)^4\operatorname{Re}[\widehat{D}(\omega)]\delta(\omega+\omega')
$$
$$
\times \delta(\vec{k}+\vec{k}')\frac{\bar{n}}{2i}(\vec{k}+\epsilon\vec{q}/2)\cdot(\vec{k}'+\epsilon\vec{q}/2)
$$
$$
\times \widetilde{g}_n(\vec{k}+\epsilon\vec{q}/2,\omega)\widetilde{g}_n(\vec{k}'+\epsilon\vec{q}/2,\omega').
$$
(46)

The dynamic structure factor $S(\vec{k},\omega)$ of the suspension is then obtained from Eqs. (45) and (46):

$$
S(\vec{k},\omega) = S^{\mathrm{eq}}(\vec{k},\omega)+S^{\mathrm{neq}}(\vec{k},\omega)
$$
$$
= \langle\delta\widetilde{n}(\vec{k},\omega)\delta\widetilde{n}(\vec{k}',\omega')\rangle^{\mathrm{eq}}+\langle\delta\widetilde{n}(\vec{k},\omega)\delta\widetilde{n}(\vec{k}',\omega')\rangle^{\mathrm{neq}}
$$
$$
= \frac{2(2\pi)^4\operatorname{Re}[\widehat{D}(\omega)]n_0\delta^4(0)k^2}{[(i\omega)^\alpha+\widehat{D}(\omega)k^2][(-i\omega)^\alpha+\widehat{D}^*(\omega)k^2]}
$$
$$
- \Bigg\{1\frac{2\omega\operatorname{Re}[\widehat{D}(\omega)]\vec{k}\cdot\vec{\nabla}n_s/n_0}{[(-i\omega)^\alpha+\widehat{D}(\omega)k^2][(i\omega)^\alpha+\widehat{D}^*(\omega)k^2]}\Bigg\}.
$$
(47)

This expression is general and valid for any form of $\widehat{D}(\omega)$, but in the next section we consider explicit forms of $\widehat{D}(\omega)$ which are determined by the specific viscoelasticity of the solvent. At this point, we emphasize that although $S(\vec{k},\omega)$ is a real quantity, from Eq. (47) is not clear whether it is

an odd or even function of ω. However, its parity can be established by recalling that the correlation of the stochastic current $\overrightarrow{J}^F(\vec{r},t)$ in the FDT given by Eq. (25) should be an even function in time. Consistently, $D(|t-t'|)$ should be such that $\widehat{D}(\omega)=\widehat{D}(-\omega)=\widehat{D}^*(\omega)$ and $\operatorname{Re}[\widehat{D}(\omega)]=\operatorname{Re}[\widehat{D}(-\omega)]$ is also an even function of ω. Consequently, the quantity $\widehat{\phi}(\omega)\equiv[i\omega+\widehat{D}(\omega)k^2][-i\omega+\widehat{D}^*(\omega)k^2]$ is even in ω since $\widehat{\phi}(\omega)=\widehat{\phi}(-\omega)$. Thus, the nonequilibrium part of the structure factor, $S^{\mathrm{neq}}(\vec{k},\omega)$, is an odd function of ω, and its shape is asymmetric, shifting its maximum towards negative frequencies. The magnitude of this shifting is proportional to the magnitude of the density gradient, $\vec{\nabla}n_s$, and also depends on the time scales involved in $\widehat{D}(\omega)$, which are in turn determined by the time scales of the viscoelasticity of the solvent, as we discuss below.

V. VISCOELASTICITY

To analyze quantitatively the behavior of the dynamic structure factor $S(\vec{k},\omega)$ it is necessary to specify first the type of viscoelasticity, by determining the explicit form of the frequency-dependent viscosity $\widehat{G}(\omega)$. One way to do this is by recalling that since the suspended particles are Brownian, the equation of motion of an isolated suspended particle of mass m is given by the Langevin equation

$$
m\frac{d}{dt}\overrightarrow{u}(t)=\overrightarrow{F}^H(t)+\overrightarrow{F}^R(t),
$$
(48)

or in the frequency representation

$$
i\omega m\widehat{\overrightarrow{u}}(\omega)=\widehat{\overrightarrow{F}^H}(\omega)+\widehat{\overrightarrow{F}^R}(\omega).
$$

Here $\overrightarrow{F}^R(t)$ is the random force originating in the internal fluctuations of the solvent described by a Gaussian stochastic process of zero mean and satisfying the fluctuation-dissipation relation

$$
\langle\overrightarrow{F}^R(t)\overrightarrow{F}^R(t')\rangle=2kT\xi(|t-t'|)\overleftrightarrow{1}
$$
(49)

or in $(\vec{k},\omega)$ space

$$
\langle\widetilde{\overrightarrow{F}}^R(\omega)\widetilde{\overrightarrow{F}}^R(\omega')\rangle=2(2\pi)kT\operatorname{Re}[\widehat{\xi}(\omega)]\delta(\omega+\omega')\overleftrightarrow{1}.
$$
(50)

These relations are obtained by finding the rate of change of the total entropy of the solvent and by applying the general rules of fluctuations [50,51]. The assumptions implicit in the Langevin equation (48) have been well established [2,3].

On the other hand, the hydrodynamic force $\widehat{\overrightarrow{F}}^H(\omega)$ on one of the suspended spherical particles of radius a moving with velocity $\overrightarrow{u}(\omega)$, is determined by the frequency-dependent viscosity $\widehat{\eta}(\omega)$ of the solvent, through a generalized Faxen theorem valid when the sphere has a time-dependent velocity and the fluid's motion (solvent) is nonstationary [52]. When the unperturbed solvent is at rest, as in the NESS defined by Eqs. (12) and (13), the force exerted by the solvent on the sphere is given by

$$
\widehat{\overrightarrow{F}}^H(\omega)=\widehat{\xi}(\omega)\widehat{\overrightarrow{u}}(\omega),
$$
(51)

where the frequency-dependent friction $\widehat{\xi}(\omega)$ is given by

$$\widehat{\xi}(\omega) \equiv 6\pi a \widehat{\eta}(\omega)\left[1 + \widehat{\alpha}(\omega)a + \tfrac{1}{9}\widehat{\alpha}^2(\omega)a^2\right], \quad (52)$$

where $\alpha(\omega)$ is the inverse penetration length

$$\widehat{\alpha}(\omega) = \left[\frac{-i\omega\rho_0}{\widehat{\eta}(\omega)}\right]^{1/2}. \quad (53)$$

The time-dependent diffusion coefficient $D(t)$ of the suspended particles is related to $\widehat{\eta}(\omega)$ through $\xi(\omega)$ by the Green-Kubo relation

$$D(t) = \tfrac{1}{3}\mathrm{Tr}[\langle \overrightarrow{u}(t)\overrightarrow{u}(0)\rangle]\Theta(t), \quad (54)$$

where $\Theta(t)$ is the Heaviside step function and Tr denotes the trace of the tensor. According to Eqs. (47)–(52) and making use of causality, it follows that the relation between diffusion and viscoelasticity is given by

$$\widehat{D}(\omega) = \frac{kT}{im\omega + \widehat{\xi}(\omega)}. \quad (55)$$

It should be emphasized that the particle velocity correlation function $\langle \overrightarrow{u}(t)\overrightarrow{u}(0)\rangle$ generally decays to zero in a short time and the description in terms of the diffusion coefficient is expected to be valid only at times much longer than a molecular time.

A. Maxwell fluid

Equation (55) is valid for any $\widehat{\eta}(\omega)$ provided that it has a well-defined Fourier transform. In previous work the case where the solvent obeys Maxwell's rheological equation of state has been considered [46],

$$\eta(t) = \frac{\eta_0}{\tau_0}e^{-t/\tau_0}\Theta(t), \quad (56)$$

τ_0 being the relaxation time and η_0 is the zero-frequency shear viscosity. The frequency dependent shear stress modulus $\widehat{G}(\omega)$ is then

$$\widehat{G}(\omega) \equiv \frac{\widehat{\eta}(\omega)}{\omega} = \frac{G_0}{(1/\tau_0)^2 + \omega^2}\,(1/\tau_0 - i\omega). \quad (57)$$

Once we have defined the type of viscoelasticity of the solvent, to determine the dependence of $\widehat{D}(\omega)$ one has to take into account that for long-time scales (low frequencies) of the order of the relaxation times of diffusion modes, the leading contribution to the frequency dependent $\widehat{D}(\omega)$ [Eq. (55)] is determined by the viscoelastic nature of the solvent through Eq. (52). If the Maxwell fluid is chosen to be liquid salol (S) for the material properties values used below in the caption of Fig. 1, it can be explicitly verified that the time scales introduced by the relaxation of the perturbations of the solvent and the time scale due to the inertia of the particle are both negligible compared to the diffusive scales, and hence $\widehat{\alpha}(\omega)a \ll 1$ in Eq. (52) and $|im\omega| \ll |\widehat{\xi}(\omega)|$ in Eq. (55). Accordingly

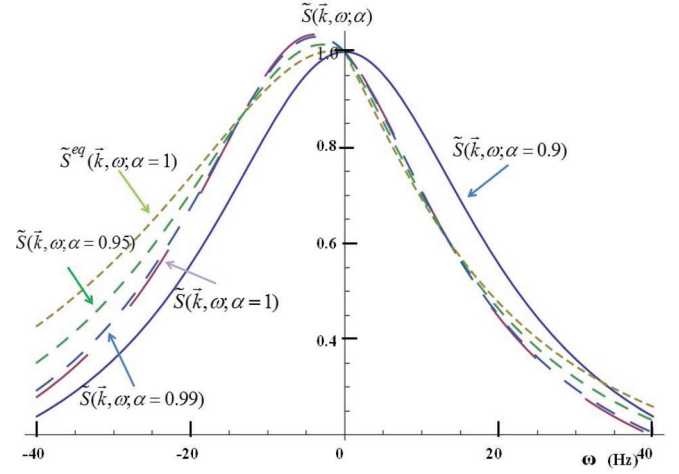$$\widehat{D}(\omega) = \frac{kT}{6\pi\eta_0 a}(1 + i\omega\tau_0) \equiv D_0(1 + i\omega\tau_0). \quad (58)$$

FIG. 1. (Color online) $\widetilde{S}(\overrightarrow{k},\omega;\alpha)$ vs ω as calculated from Eq. (65) for liquid salol assuming that it obeys Maxwell's rheological equation of state (56). We chose the following parameter values: $T = 353.2\,\mathrm{K}$, $G_0 = 1.154\,\mathrm{kg/ms}$, $\tau = 89.9 \times 10^{-6}\,\mathrm{s}$, $\rho_0 = 1.212\,\mathrm{kg/m^3}$ [53]. The different curves correspond to fractional $\widetilde{S}(\overrightarrow{k},\omega;\alpha)$ versus ω for $\alpha = 0.9, 0.95, 0.99, 1$.

B. Power-law viscoelasticity

Let us now consider the case when the viscoelasticity of the solvent is described by a power-law rheological equation of state

$$\eta(t) = \eta_0 t^{-\lambda}\Theta(t), \quad 0 < \lambda < 1, \quad (59)$$

with the complex time-Fourier transform

$$\widehat{\eta}(\omega) = \frac{\eta_0\Gamma(1-\lambda)}{\omega^{1-\lambda}}\{\cos[(1-\lambda)\pi/2] - i\sin[(1-\lambda)\pi/2]\}, \quad (60)$$

where $\Gamma(x)$ is the Gamma function. Under the same arguments on the time scales as discussed above for a Maxwell fluid, for the power-law behavior we get

$$\begin{aligned}\widehat{\xi}(\omega) &= 6\pi a\widehat{\eta}(\omega) \\ &= 6\pi a\eta_0\Gamma(1-\lambda)\{\cos[(1-\lambda)\pi/2] \\ &\quad - i\sin[(1-\lambda)\pi/2]\}\omega^{\lambda-1}, \end{aligned} \quad (61)$$

and Eq. (55) yields

$$\begin{aligned}\widehat{D}(\omega) &\simeq \frac{kT}{\widehat{\xi}(\omega)} \\ &= \frac{D_0}{\Gamma(1-\lambda)}\{\cos[(1-\lambda)\pi/2] - i\sin[(1-\lambda)\pi/2]\}\omega^{1-\lambda}. \end{aligned} \quad (62)$$

In the next section we shall use the material properties of two specific power-law rheological fluids, namely, salol (S), a glass-forming liquid, and E_1, a solution of 0.02% separan $MG500 + 2\%$ water in glucose $MCY43N$, which can be classified as an exceedingly elastic fluid [54].

VI. RESULTS AND DISCUSSION

To examine the behavior of $S(\overrightarrow{k},\omega)$ given by Eq. (47) as a function of ω for different values of α, it is convenient to

define the dimensionless fractional structure factor

$$\widetilde{S}(\vec{k},\omega;\alpha) \equiv \frac{S(\vec{k},\omega)}{S^{eq}(\vec{k})}, \tag{63}$$

where $S^{eq}(\vec{k})$ is

$$
\begin{aligned}
S^{eq}(\vec{k}) &\equiv S(\vec{k},\omega=0) \\
&= \frac{2(2\pi)^4 D_0 n_0 \delta^4(0)}{D_0^2 k^2}.
\end{aligned}
\tag{64}
$$

Then

$$\frac{\widetilde{S}(\vec{k},\omega;\alpha)}{\widetilde{S}^{eq}(\vec{k},\omega;\alpha)} = 1 - \frac{2\omega\widetilde{\nabla}n_s}{[(i\omega)^\alpha + \widehat{D}(\omega)k^2][(-i\omega)^\alpha + \widehat{D}^*(\omega)k^2]}, \tag{65}$$

where we have defined the dimensionless concentration gradient $\widetilde{\nabla}n_s$ as

$$\widetilde{\nabla}n_s \equiv \frac{\vec{k}\cdot\vec{\nabla}n_s/n_0}{k^2(1+D_0 k^2\tau)}. \tag{66}$$

We now plot $\widetilde{S}(\vec{k},\omega;\alpha)$ versus ω parametrized by α, assuming that it obeys Maxwell's rheological equation of state.

The symmetric curve $\widetilde{S}^{eq}(\vec{k},\omega;\alpha=1)$ is the nonfractional equilibrium dimensionless structure factor. The asymmetric curves result from the properties of $\widetilde{S}^{neq}(\vec{k},\omega;\alpha)$, defined by Eqs. (47) and (63), which is an odd function of ω with its maximum shifted towards negative frequencies; the size of the shift is proportional to the magnitude of the dimensionless density gradient, $\widetilde{\nabla}n_s$, and also depends on the time scales determined by the viscoelasticity of the solvent. It is clear from these curves that for low frequencies ($\sim\omega \leqslant \pm 15$ Hz) the nonfractional structure factor $\widetilde{S}(\vec{k},\omega;\alpha=1)$ is larger than the fractional ones $\widetilde{S}(\vec{k},\omega;\alpha\neq 1)$. However, as ω increases this behavior is inverted, and the fractional structure factors become larger than the nonfractional ones.
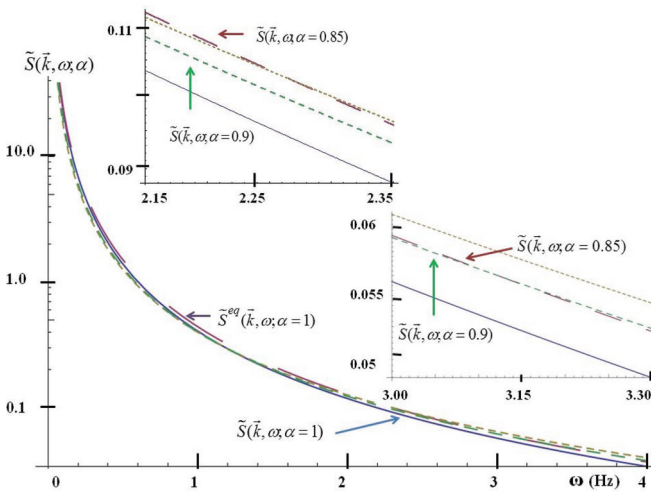

FIG. 2. (Color online) The same as in Fig. 1 but for E_1 when it is described as a power-law fluid by Eq. (59) with $\lambda = 0.2$. The following material and parameter values were used, $T = 292$ K, $G_0 = 17.3$ kg/ms, $\rho_0 = 1414$ kg/m³, $\tau = 0.18$ s.
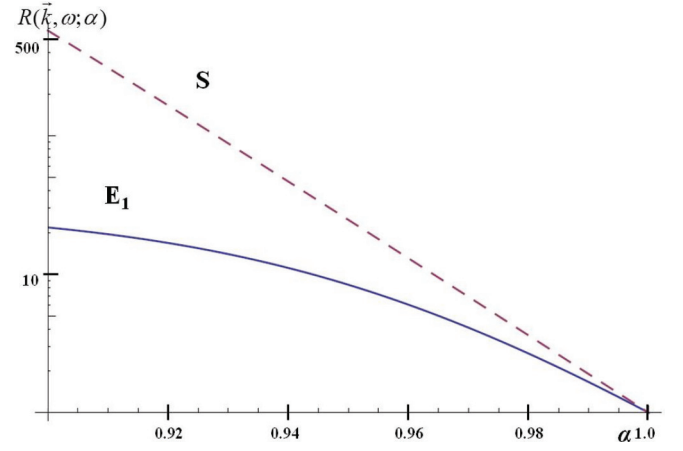

FIG. 3. (Color online) The ratio $R(\vec{k},\omega;\alpha)$ as defined by Eq. (67), for $\omega \sim 10^{14}$ Hz and $k \sim 10^7$ m^{-1}. The discontinuous line refers to S and the continuous one to E_1 when both are considered to be Maxwell fluids.

Now consider E_1 assuming that it obeys the power-law rheological equation of state [Eq. (59)]. The behavior of the corresponding $\widetilde{S}(\vec{k},\omega;\alpha)$ is shown in Fig. 2 and exhibits the same trend as observed in Fig. 1 for S; i.e., for low frequencies the nonfractional values $\widetilde{S}(\vec{k},\omega;\alpha=1)$ are larger than the fractional ones $\widetilde{S}(\vec{k},\omega;\alpha\neq 1)$, but ranges of values of ω for which the latter become larger than the former can be clearly identified. These are shown in the two insets in Fig. 2.

This behavior clearly suggests that fractional effects may be significant for $\widetilde{S}(\vec{k},\omega;\alpha\neq 1)$, which is a measurable property; however, from Figs. 1 and 2 it is difficult to quantify these differences. In order to do so we plot the ratio

$$R(\vec{k},\omega;\alpha) \equiv \frac{\widetilde{S}(\vec{k},\omega;\alpha\neq 1)}{\widetilde{S}^{eq}(\vec{k},\omega;\alpha=1)} \tag{67}$$

as a function of α for fixed values of $\vec{k}$ and ω. Note that since we have an analytical expression for $\widetilde{S}(\vec{k},\omega;\alpha)$ valid for any value of $\vec{k}$ and ω, we calculate the value of $R(\vec{k},\omega;\alpha)$ for the
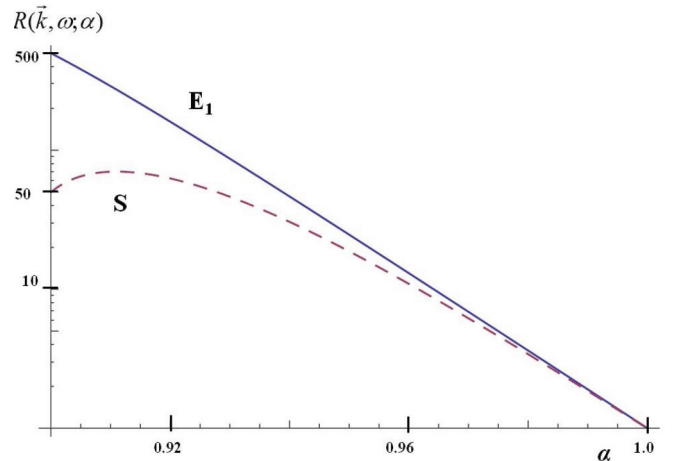

FIG. 4. (Color online) The same as in Fig. 3 when both S and E_1 obey the power-law rheological equation of state Eq. (59). Same parameter values as in Figs. 1 and 2.
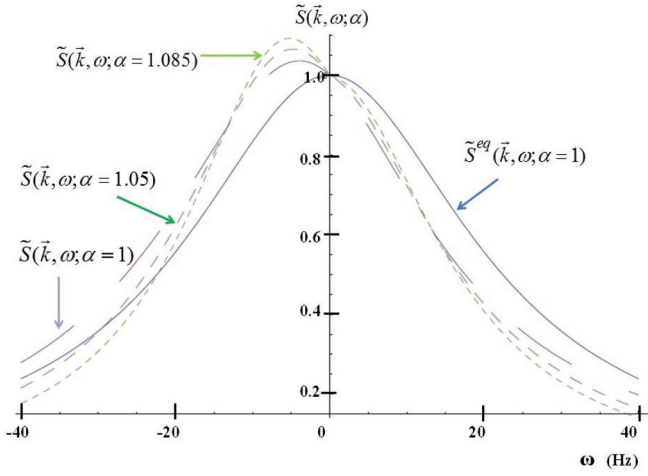
FIG. 5. (Color online) The same as in Fig. 1 for $\alpha = 1, 1.05, 1.085$.

values $\omega \sim 10^{14}$ Hz and $k \sim 10^7$, which are typical for a light-scattering experiment [55]. This yields the curves shown in Fig. 3 when both S and E_1 are considered to be Maxwell fluids.

Note that for salol $R(\vec{k}, \omega; \alpha)$ can be as large as two orders of magnitude for $\alpha \sim 0.9$, indicating that the relative change in $\widetilde{S}(\vec{k}, \omega; \alpha)$ due to the presence of fractional derivatives may be quite significant even for Maxwell's rheology. On the other hand, if both fluids obey the power-law rheological equation of state Eq. (59), the plot of $R(\vec{k}, \omega; \alpha)$ versus α is shown in Fig. 4, where the larger increment corresponds to E_1.

Again, in this case the value of $R(\vec{k}, \omega; \alpha)$ can also be considerable large (~ 500) for power-law rheology (E_1), indicating that fractional effects may be significant.

It is worth emphasizing that all the previous results were obtained for $0 < \alpha < 1$, but since α can also take values in $1 < \alpha < 2$, we now examine their effect on the same quantities plotted in Figs. 1–3. If we plot $\widetilde{S}(\vec{k}, \omega; \alpha)$ versus
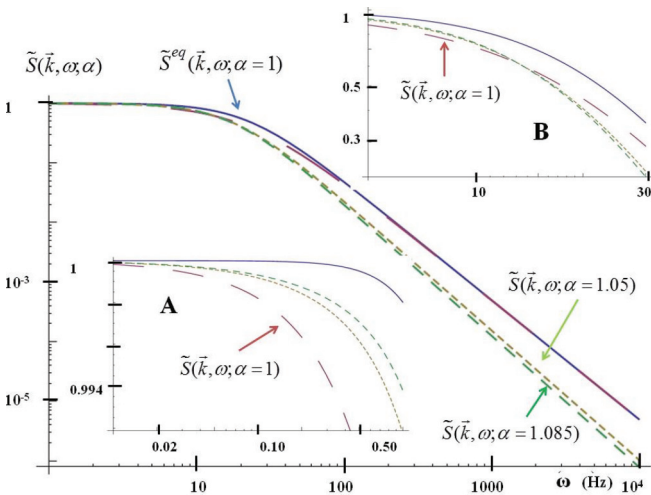

FIG. 6. (Color online) The same as in Fig. 5 for large frequencies. Inset A shows a frequency range where $\widetilde{S}(\vec{k}, \omega; \alpha = 1.05)$ and $\widetilde{S}(\vec{k}, \omega; \alpha = 1.085)$ are below $\widetilde{S}^{eq}(\vec{k}, \omega; \alpha = 1)$ and above $\widetilde{S}(\vec{k}, \omega; \alpha = 1)$. Inset B shows a frequency range where both fractional structure factors are below $\widetilde{S}(\vec{k}, \omega; \alpha = 1)$ and $\widetilde{S}^{eq}(\vec{k}, \omega; \alpha = 1)$.
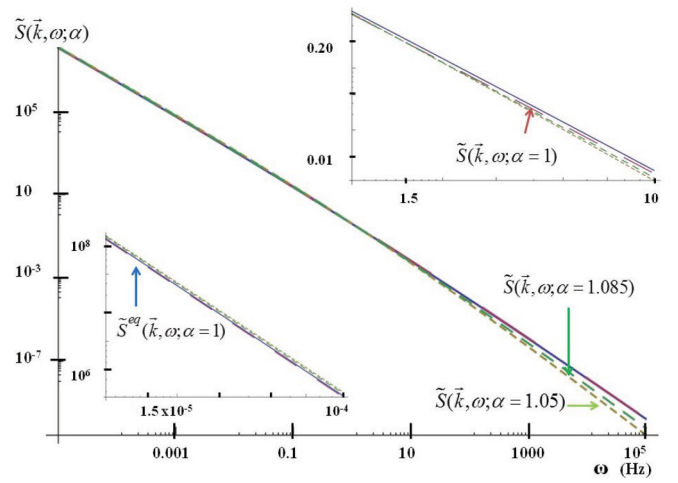

FIG. 7. (Color online) The same as in Fig. 2 for $\alpha = 1, 1.05, 1.085$.

ω parametrized by $\alpha = 1.05$ and 1.085, considering salol as a Maxwell fluid and for the same material properties as those indicated in Fig. 1, we get the curves shown in Fig. 5.

Note that in contrast to Fig. 1, the maxima of the curves $\widetilde{S}(\vec{k}, \omega; \alpha = 1.05)$ and $\widetilde{S}(\vec{k}, \omega; \alpha = 1.085)$ in Fig. 6 are located above the maximum value of $\widetilde{S}(\vec{k}, \omega; \alpha = 1)$. These relative positions are maintained around $\omega \sim 0$, but as ω increases this situation changes. For instance, inset A shows a frequency range where $\widetilde{S}(\vec{k}, \omega; \alpha = 1.05)$ and $\widetilde{S}(\vec{k}, \omega; \alpha = 1.085)$ are below $\widetilde{S}^{eq}(\vec{k}, \omega; \alpha = 1)$ and above $\widetilde{S}(\vec{k}, \omega; \alpha = 1)$, whereas inset B shows a frequency range where both fractional structure factors are below $\widetilde{S}(\vec{k}, \omega; \alpha = 1)$ and $\widetilde{S}^{eq}(\vec{k}, \omega; \alpha = 1)$. Afterwards both fractional curves are always below $\widetilde{S}^{eq}(\vec{k}, \omega; \alpha = 1)$.

A similar behavior occurs for power law fluids as shown in Fig. 7.

This behavior already indicates that contrary to the behavior found for $\alpha < 1$, for the interval $1 < \alpha < 2$ the fractional effects on the structure factor are lower and less significant than in the nonfractional case. This feature is quantified more
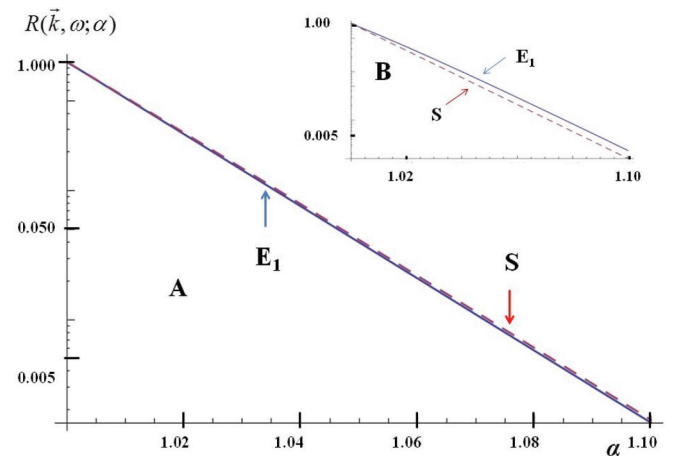

FIG. 8. (Color online) Equation (67), for $\omega \sim 10^{14}$ Hz and $k \sim 10^7$ m^{-1}. The discontinuous line refers to S and the continuous one to E_1 when both are considered to be power law fluids. The inset shows the same behavior when S and E_1 are Maxwell fluids.

precisely in Fig. 8, where the ratio $R(\vec{k},\omega;\alpha)$ is plotted as a function of α for the typically experimental values $\omega \sim 10^{14}$ Hz and $k \sim 10^7$.

This figure shows that in marked contrast to the behavior of Figs. 3 and 4, the ratio $R(\vec{k},\omega;\alpha)$ is always lower than 1 in the interval $1 < \alpha < 2$, indicating that the effect of fractional fluctuations is much less significant on $\widetilde{S}(\vec{k},\omega;\alpha)$ than those for the interval $0 < \alpha < 1$, which are the dominant ones. This result is in accordance with the common use in the literature of fractional derivatives, which usually replaces first-order derivatives by fractional ones with a fractional order α in the interval $0 < \alpha < 1$ [56].

VII. CONCLUDING REMARKS

We collect the following conclusions commenting briefly on each one.

In this work we have proposed a model to analyze the effect produced on the light-scattering properties of a nonequilibrium dilute suspension of solid particles in a viscoelastic solvent, when fractional time derivatives are introduced into the time evolution equations for the density and velocity fluctuations. The lack of separation of the time scales due to viscoelasticity is taken into account by the inclusion of Caputo's fractional time derivative in the stochastic equations. We showed that this operator does not change the basic structure of the usual fluctuating hydrodynamic description, due to the fact that the Fourier transform of this fractional time derivative is well defined and the fractional character of the dynamics is manifest in the propagators (35) and (36). In the Appendix we discuss that it is reasonable to expect that the FFDTs [Eqs. (37) and (38)] have the same structure as for a NESS. These features allow to generalize the usual fluctuating hydrodynamic description to the fractional case.

It should be pointed out that strictly speaking, not only the conventional stochastic calculus should be replaced by a fractional one, but also the white Gaussian noise of the fluctuating currents has to be modified as well. This issue was not considered in this work, but it deserves further attention. It should be stressed as well, that our approach and conclusions are applicable only to the model analyzed here and to the class of nonequilibrium states we have considered in the present work. Also, we did not addressed the issue of replacing the spatial derivatives in the dynamic equations by fractional derivatives; this is a difficult problem which to our knowledge has not been explored.

We should stress that following Ref. [45], in this work we have interpreted a fractional fluctuation as a superposition of conventional thermal fluctuations with successive weighted increments over time. As a consequence, the values of $\partial \vec{v}(\vec{r},\tau)/\partial \tau$ at a time τ far apart from t have smaller weights than those at times τ closer to t. Although we do not have a clear interpretation of the action of fractional fluctuations and of their possible relation with the coherent or incoherent nature of the solutions of the fractional equations, some features of our results may be interpreted along this direction. Indeed, the asymmetry, the size of the maximum, and the shift of the peak of the spectrum (for conventional time derivatives) is a typical nonequilibrium effect [46]. However, as α becomes less than

1, the presence of fractional fluctuations tend to decrease the magnitude of the peak and the size of its width, as well as the size of the shift. As a consequence, the spectrum becomes slightly less asymmetric, but this occurs only for low frequencies (left part of Fig. 1). As ω increases, there is a crossover, and these fractional effects become larger than those for $\omega \sim 0$ (right part of Fig. 1). In contrast, if $\alpha < 1$, the behavior near $\omega \sim 0$ is the opposite, and there is also a crossover which as t grows makes the fractional spectrum smaller than that for $\alpha = 1$. This behavior has the implication that the ratio is positive and considerably larger than 1 for $\alpha < 1$ (Figs. 3 and 4) and almost vanishes for $\alpha > 1$ (Fig. 8). Since the spectrum is constructed from the solutions (34) and (33), this behavior of the correlations that produce the spectrum should reflect the behavior of the solutions and the nature of the equations. If $\alpha > 1$, according to Ref. [45], the fractional fluctuations can be interpreted as a superposition of normal derivatives of order higher than one; therefore, the second time derivative in the equation becomes more important, indicating that the the effect of second derivatives appear in the equation (wave equation) with (coherent) propagating wave solutions. When $\alpha < 1$ the presence of fractional fluctuations does not introduce higher order derivatives, and the equation remains a relaxation (diffusive) equation.

However, a deeper explanation of the action of a fractional fluctuation is necessary. In a recent publication [57] alternative explanations of the extensions resulting from replacing integer-valued derivatives by fractional derivatives are made [58]. This interpretation makes use of the notion of fluctuating trajectory and considers fractional models as averages over an ensemble of these trajectories. The authors of this work propose a physical interpretation of Caputo's fractional derivative based on the assumption that a single trajectory generated by the ordinary fractional calculus moves according to operational time, and that at a given time t there exists a distribution of operational times, over which to evaluate an average over the single trajectory, converted in this way into a Gibbs set of trajectories. The essential issue that this interpretation arises is that fractional calculus generates decorrelation rather than friction, as is usually assumed. Moreover, what should be the physical interpretation of the use of fractional calculus acting on trajectories that have an (apparently) infinite memory, as was assumed to arrive at Eqs. (34) and (33) is still lacking. It should be emphasized though, that the lower and upper limits in Eqs. (20) and (21) were extended to $-\infty$ and $+\infty$, respectively, due to the assumption that the system was prepared in the remote past in the given NESS and assuming that it did not age and was preserved in this state till the observation time interval. This assumption justifies the existence of the Fourier transform (29); however, the same result could have been reached by using Laplace transforms with a finite memory and with vanishing initial conditions.

It should be mentioned that in the literature there have been proposed other approaches involving fractional derivatives for anomalous diffusion in systems of particles and clusters, possessing a viscoelastic response describable by a power-law memory function in the evolution equation for the probability density [59]. This equation is constructed by using a thermokinetic approach in which the usual integer valued derivatives in this equation are not replaced by fractional derivatives. Instead, the assumed memory function leads to a scaling behavior

describable by a fractional (Riemann-Liouville) derivative, giving rise to a fractional diffusion equation for the probability density. Whether this approach and the one used in the present work may lead to similar results is an interesting question that might be explored, perhaps, by calculating the structure factor of the particles from the probability distribution function obtained in Ref. [59] and by comparing the result with our results. Also, an equivalent fractional Fokker-Planck equation to our generalized Langevin equations could be constructed and compared with the above mentioned kinetic equation.

Finally, from the time evolution equations for the fluctuations, we calculated analytically the concentration correlation function and the light-scattering dynamic structure factor for the suspension, with and without the presence of fractional fluctuations. We found that the fractional fluctuations in the interval $0 < \alpha < 1$ enhance the value of the structure factor of the suspension. The relative change in the values of the structure factor when fractional effects are present may be significantly larger, up to two orders of magnitude, than its value with conventional thermal fluctuations only. In contrast, we found that fractional fluctuations corresponding to the interval $1 < \alpha < 2$ contribute much less to the structure factor. However, we are not aware of any experimental results to compare with the predictions of our model; therefore it is not possible to conclude from our analysis if this (significant) enhancement is measurable. This is an open question that remains to be assessed.

APPENDIX

The assumed validity of the FFDT, given by Eqs. (24) and (25) or (37) and (38), when fractional time derivatives are used is a strong assumption which requires justification. To exhibit its plausibility, in this appendix we follow, adapt, and extend the method developed in Refs. [48,49] to sketch the validity of the FFDT.

To be specific and to simplify the notation used in the main text, let $A(t)$ denote one component of the fluctuations of the velocity $\overrightarrow{v}(\overrightarrow{r},t)$ whose time evolution is given by (20). Taking the space-Fourier transform of the resulting equation yields

$$\prescript{C}{0}{D}_t^\alpha A(t) = \int_0^t dt' \Gamma(t - t') A(k,t') + F(k,t), \quad \text{(A1)}$$

where we have defined

$$\Gamma(t - t') \equiv -k^2 \rho_0^{-1} G(t - t'), \quad \text{(A2)}$$

$$F(k,t) \equiv [-i \overrightarrow{k} \cdot \widehat{\overrightarrow{\Pi}}^R (\overrightarrow{k},t)]_{\text{component}}. \quad \text{(A3)}$$

Since we have assumed a quiescent solvent with $\overrightarrow{v}_s(\overrightarrow{r}) = 0$, if we take the Laplace transform of (A1) with $A(t = 0) = 0$,

we arrive at

$$A^f(z) = \frac{F(z)}{z^\alpha + \Gamma(z)} \equiv R(z) F(z), \quad \text{(A4)}$$

where $z \equiv i\omega$ is the Laplace transform variable. The upper index f has been introduced to emphasize that it is a fractional variable (fluctuation). From this definition of $R(z)$ it follows that

$$\frac{d}{dt} R(t) = -\int_0^t \Gamma(t - t') R(t') \, dt'. \quad \text{(A5)}$$

On the other hand, when only conventional time derivatives are considered for NESS, it is accepted in the literature that the noise correlation $C_F(t - t')$ obeys the FDT of generalized hydrodynamics, which has the same structure as that in equilibrium, but with the local values of the corresponding Onsager coefficients. For the random stresses this is expressed by (24)

$$[C_F(t - t')]_{ijlm} \equiv \left\langle \Pi_{ij}^R(\overrightarrow{r},t) \Pi_{lm}^R(\overrightarrow{r'},t') \right\rangle$$
$$= 2kTG(|t - t'|)\delta(\overrightarrow{r} - \overrightarrow{r'})\Delta_{ijlm}$$
$$\equiv \langle A^2 \rangle^{\text{NESS}} \Gamma(|t - t'|), \quad \text{(A6)}$$

with

$$\langle A^2 \rangle^{\text{NESS}} \equiv \Omega_{ijlm} \equiv 2kT\delta(\overrightarrow{r} - \overrightarrow{r'})\Delta_{ijlm}. \quad \text{(A7)}$$

Since for our argument only the time dependence of C_F is relevant, we have further simplified the notation by omitting the tensorial character, thus

$$C_F(t - t') = \Omega\Gamma(|t - t'|). \quad \text{(A8)}$$

Now, from (A4) it follows that

$$A^f(t) = \int_0^t R(t - t') F(t') \, dt', \quad \text{(A9)}$$

and therefore

$$\langle A^{f2}(t) \rangle = 2\int_0^t R(t') \int_0^{t'} C_F(t' - t'') R(t'') \, dt'' \, dt'. \quad \text{(A10)}$$

It should be stressed that the fractional character of this expression is contained only in $R(t)$ and not in $C_F(t' - t'')$. Furthermore, since it is well known that around equilibrium states the form of the FDT is the same as in (A6) but with $G(|t - t'|)$ replaced by the zeroth frequency dependent shear stress modulus G_0, the explicit form of $C_F(t - t')$ will not affect $R(t)$ and, as a first approximation, $C_F(t - t')$ may be expressed in terms of the the zero-frequency shear modulus G_0. This is equivalent to making a Taylor series expansion of $C_F(t - t')$ around a reference time t_0 and keeping only the first term of the expansion. In this case (A10) may be rewritten as

$$\langle A^{f2}(t) \rangle = 2\Omega \int_0^t R(t') \left[\int_0^{t'} \Gamma(t' - t'') R(t'') \, dt'' \right] dt'$$
$$= -2\Omega \int_0^t R(t') \frac{d}{dt'} R(t') \, dt'$$
$$= \langle A^2 \rangle^{\text{NESS}} [1 - R^2(t)]. \quad \text{(A11)}$$

To arrive at this expression we have used that

$$R(t) \equiv \frac{\langle A^f(t) A^f(0) \rangle}{\langle A^{f2}(0) \rangle} \equiv \frac{C_A(t)}{C_A(0)}, \quad \text{(A12)}$$

which follows from (A4), and the property

$$\lim_{t \to \infty} R(t) = 0, \tag{A13}$$

which is a consequence of the Tauberian theorem

$$\lim_{t \to \infty} C_A(t) = \lim_{z \to 0} z C_A(z) = 0. \tag{A14}$$

The result (A11) implies that in the limit of long times

$$\lim_{t \to \infty} \langle A^{f 2}(t) \rangle = \langle A^2 \rangle^{\text{NESS}}, \tag{A15}$$

which expresses that in this limit a fractional system may be driven to a NESS, a result which is the essential content of the fractional fluctuation-dissipation theorem.

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