

**DIFFUSION REGIMES IN BROWNIAN MOTION
INDUCED BY THE BASSET HISTORY FORCE**

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Summary

The velocity autocorrelation and the displacement variance of a Brownian particle moving in an incompressible viscous fluid are calculated taking into account the effects of added mass and both Stokes and Basset hydrodynamic forces. These forces are known to describe the friction effects in a viscous fluid, respectively in the steady state and in the transient state of the motion, in the limit of vanishing Reynolds number.

The explicit expressions of these functions versus time are provided in terms of Mittag-Leffler functions and compared with the respective ones for the classical Brownian motion. The effect of added mass is only to modify the time scale, that is the characteristic relaxation time induced by the Stokes force. The effect of the Basset force, which is of hereditary type namely history-dependent, is to perturb the white noise of the random force and change the decay character of the velocity autocorrelation function from pure exponential to power law.

Furthermore, the displacement variance is shown to maintain, for sufficiently long times, the linear behaviour which is typical of normal diffusion, with the same diffusion coefficient of the classical case. However, for light particles, the Basset history force induces a long retarding effect in the establishing of the linear behaviour, allowing for a regime of fast anomalous diffusion.

KEY-WORDS: Diffusion, Brownian Motion, Basset Force, Fractional Calculus

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Sommario

In questo rapporto si calcolano la autocorrelazione della velocità e la varianza dello spostamento per una particella Browniana che si muove in un fluido viscoso incompressibile, prendendo in considerazione gli effetti di massa aggiunta e di entrambe le forze idrodinamiche, di Stokes e di Basset. È noto che tali forze descrivono l'attrito viscoso del fluido rispettivamente allo stato stazionario e durante la fase transiente del moto, nel limite di numero di Reynolds tendente a zero.

Per queste quantità si forniscono le espressioni esplicite in funzione del tempo tramite le funzioni speciali di Mittag-Leffler che vengono confrontate con quelle corrispondenti al moto Browniano classico. L'effetto della massa aggiunta consiste solo nell'alterazione della scala dei tempi, cioè il tempo di rilassamento caratteristico della forza di Stokes. L'effetto della forza di Basset, che è di tipo ereditario, ossia dipendente dalla storia, è di modificare il cosiddetto "rumore bianco" della forza stocastica e di cambiare il carattere del decadimento della funzione di autocorrelazione della velocità da una legge esponenziale ad una legge di potenza.

Si prova inoltre che la varianza dello spostamento mantiene, per tempi sufficientemente lunghi, la crescita lineare che è tipica della diffusione normale, con lo stesso coefficiente di diffusione del caso classico. Tuttavia, per particelle più leggere, la forza di Basset induce un lungo effetto ritardante nello stabilire il comportamento lineare, permettendo così un regime di diffusione anomala veloce.

PAROLE-CHIAVE: Diffusione, Moto Browniano, Forza di Basset, Calcolo Frazionario ■

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1. Introduction

Since the pioneering computer experiments by Alder & Wainwright (1970), which have shown that the velocity autocorrelation function for a Brownian particle in a dense fluid goes asymptotically as $t^{-3/2}$ instead of exponentially as predicted by stochastic theory, many attempts have been made to reproduce this result by purely theoretical arguments. Usually, hydrodynamic models are adopted to generalize Stokes' law for the frictional force and obtain a generalized Langevin equation, see *e.g.* Zwanzig & Bixon (1970, 1975), Widom (1971), Case (1971), Mazo (1971), Ailwadi & Berne (1971), Nelkin (1972), Hynes (1972), Chow & Hermans (1972-a,-b,-c), Hauge & Martin-Löf (1973), Dufty (1974), Bedeaux & Mazur (1974), Hinch (1975), Pomeau & Résibois (1975), Warner (1979), Reichl (1981), Paul & Pusey (1981), Felderhof (1991), Clercx & Schram (1992).

Recently, a great interest on the subject matter has been raised because of the possible connection among long-time correlation effects, (fractional) Brownian motion and anomalous diffusion, see *e.g.* Muralidar et al. (1990), Wang and Lung (1990), Wang (1992), Giona and Roman (1992).

We recall that anomalous diffusion is the phenomenon, usually met in disordered or fractal media, according to which the mean squared displacement (the variance) is no longer linear in time but proportional to a power α of time with $0 < \alpha < 1$ (slow diffusion) or $1 < \alpha < 2$ (fast diffusion), see *e.g.* Bouchaud & Georges (1990).

We also point out that Kubo (1966) introduced a generalized Langevin equation (GLE), where the friction force appears retarded or frequency dependent through an indefinite memory function. To be consistent with the *fluctuation-dissipation theorem*, in GLE the random force is no longer a white noise (as in the classical Langevin equation) with a frequency spectrum related to that of the velocity fluctuations. A critical analysis of Kubo's derivation of the fluctuation-dissipation theorem was given by Felderhof (1978). As a matter of fact, the hydrodynamic models appear as particular cases of GLE, as noted by Kubo et al. (1991).

In this report we shall revisit the Brownian motion on the basis of a generalized Langevin equation of fractional order. The purpose of our approach is to model the Brownian motion more realistically than the usual one based on the classical Langevin equation, in that it takes into account not only the Stokes viscous drag but also the retarding effects due to hydrodynamic backflow, *i.e.* the added mass and the Basset-Boussinesq history force.

The plan of the report is the following. After having reviewed in Section 2 the classical Brownian motion, in Section 3 we extend the theory according to the hydrodynamic approach. On the basis of the *fluctuation-dissipation theorem* and of the techniques of the *fractional calculus* we shall provide the analytical expressions of the autocorrelation functions (both for the random force and the particle velocity) and of the displacement variance. Consequently, the well-known results of the classical theory of the Brownian motion will be properly generalized. Significant results are shown and discussed in Section 4, where we shall point out different diffusion regimes. Finally, conclusions are drawn in Section 5.

2. The classical approach to the Brownian motion

We assume that the Brownian particle of mass m executes a random motion in one dimension with velocity $V = V(t)$ and displacement $X = X(t)$. The classical approach to the Brownian motion is based on the following stochastic differential equation (*Langevin equation*), see *e.g.* Kubo et al. (1991),

$$m \frac{dV}{dt} = F_v(t) + R(t), \quad (2.1)$$

where $F_v(t)$ denotes the *frictional force* exerted from the fluid on the particle and $R(t)$ denotes the *random force* arising from rapid thermal fluctuations, subjected to the condition $\langle R(t) \rangle = 0$. As usual, we have denoted with brackets the average taken over an ensemble in thermal equilibrium.

Assuming for the frictional force the Stokes expression for a drag of spherical particle of radius a , we obtain the classical formula

$$F_v = -\frac{1}{\mu} V(t), \quad \frac{1}{\mu} = 6\pi a \rho_f \nu, \quad (2.2)$$

where μ denotes the *mobility coefficient* and ρ_f and ν are the density and the kinematic viscosity of the fluid, respectively. If we introduce the friction characteristic time

$$\sigma = m \mu, \quad (2.3)$$

the Langevin equation (2.1) explicitly reads

$$\frac{dV}{dt} = -\frac{1}{\sigma} V(t) + \frac{1}{m} R(t). \quad (2.4)$$

We assume that the Brownian particle has been kept for a sufficiently long time in the fluid at (absolute) temperature T , so the thermal equilibrium is reached. Thus, for any t_0 in which the thermal equilibrium is maintained, the equipartition law for the energy distribution requires that

$$m \langle V^2(t_0) \rangle = kT, \quad (2.5)$$

where k is the Boltzmann constant. Consistently, we assume that the *autocorrelation functions* C_V and C_R of the stochastic processes $V(t)$ and $R(t)$,

$$C_V(t_0, t) = \langle V(t_0) V(t_0 + t) \rangle = C_V(t), \quad t \geq 0, \quad (2.6)$$

$$C_R(t_0, t) = \langle R(t_0) R(t_0 + t) \rangle = C_R(t), \quad t \geq 0, \quad (2.7)$$

do not depend on t_0 , and that the random force is uncorrelated to the particle velocity, namely

$$C_{VR}(t_0, t) = \langle V(t_0) R(t_0 + t) \rangle = 0, \quad t \geq 0. \quad (2.8)$$

Hereafter we shall assume $t_0 = 0$.

Applying the Wiener-Khintchine theorem to (2.6-7), the *power spectra* or *power spectral densities* $I_V(\omega)$ and $I_R(\omega)$, $\omega \in \mathbb{R}$, are provided by the Fourier transforms of the respective autocorrelation functions. We write

$$I_V(\omega) = \widehat{C}_V(\omega) = \int_{-\infty}^{+\infty} C_V(t) e^{-i\omega t} dt, \quad C_V(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} I_V(\omega) e^{+i\omega t} d\omega, \quad (2.9)$$

$$I_R(\omega) = \widehat{C}_R(\omega) = \int_{-\infty}^{+\infty} C_R(t) e^{-i\omega t} dt, \quad C_R(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} I_R(\omega) e^{+i\omega t} d\omega. \quad (2.10)$$

It is well known, see *e.g.* Kubo et al. (1991), that the previous assumptions lead to

$$C_V(t) = \langle V^2(0) \rangle e^{-t/\sigma} = \frac{kT}{m} e^{-t/\sigma}, \quad (2.11)$$

$$C_R(t) = \frac{m^2}{\sigma} \langle V^2(0) \rangle \delta(t) = \frac{m}{\sigma} kT \delta(t), \quad (2.12)$$

where $\delta(t)$ denotes the Dirac distribution. The result (2.11) shows that the velocity autocorrelation function decays exponentially in time with the decay constant σ , while (2.12) means that the power spectrum of $R(t)$ is to be *white*, *i.e.* independent on frequency, resulting

$$I_R(\omega) \equiv I_R = \frac{m}{\sigma} kT. \quad (2.13)$$

It can be readily shown that the mean squared displacement of a particle starting at the origin at $t_0 = 0$ (*displacement variance*) is given by

$$\langle X^2(t) \rangle = 2 \int_0^t (t - \tau) C_V(\tau) d\tau = 2 \int_0^t d\tau_1 \int_0^{\tau_1} C_V(\tau) d\tau, \quad t \geq 0. \quad (2.14)$$

For this it is sufficient to recall that $X(t) = \int_0^t V(t') dt'$, and to use the definition (2.6) of $C_V(t)$ for $t \geq t_0 = 0$. As a consequence of (2.11) and (2.14) we obtain

$$\langle X^2(t) \rangle = 2D \left[t - \sigma \left(1 - e^{-t/\sigma} \right) \right], \quad t \geq 0, \quad (2.15)$$

where

$$D = \sigma \langle V^2(0) \rangle = \int_0^\infty C_V(t) dt. \quad (2.16)$$

We note from (2.15) that

$$\langle X^2(t) \rangle = 2Dt \left[1 - (t/\sigma)^{-1} + EST \right], \quad \text{as } t \rightarrow \infty, \quad (2.17)$$

(*EST* = exponentially small terms) so that

$$D = \lim_{t \rightarrow \infty} \frac{\langle X^2(t) \rangle}{2t}. \quad (2.18)$$

Furthermore, using (2.3), (2.5) and (2.16), we recognize that

$$D = \frac{\sigma}{m} kT = \mu kT. \quad (2.19)$$

The constant D is known as the *diffusion coefficient* and the equation (2.19) as the *Einstein relation*.

3. The hydrodynamic approach to the Brownian Motion

On the basis of hydrodynamics, the Langevin equation (2.4) is not all correct, since it ignores the effects of the added mass and retarded viscous force, which are due to the acceleration of the particle, as pointed out by several authors.

The added mass effect requires to substitute the mass of the particle with the so-called effective mass, $m_e = m [1 + \rho_f / (2\rho_p)]$, where ρ_p denotes the density of the particle, see *e.g.* Batchelor (1967). Keeping unmodified the Stokes drag law, the relaxation time changes from $\sigma = m\mu$ to $\sigma_e = m_e\mu$: thus

$$\sigma_e = \sigma \left(1 + \frac{1}{2\chi} \right), \quad \text{with } \chi = \frac{\rho_p}{\rho_f}. \quad (3.1)$$

The corresponding Langevin equation has the form as (2.4), by replacing m with m_e and σ with σ_e . With respect to the classical analysis, it turns out that the added mass effect, if it were present alone, would be only to lengthen the time scale ($\sigma_e > \sigma$) in the exponentials entering the basic formulas (2.11) and (2.15) and to decrease the velocity variance $\langle V^2(0) \rangle$, consistently with the energy equipartition law (2.5) at the same temperature. Consequently, using (3.1), the diffusion coefficient is unmodified and turns out to be

$$D = \sigma_e \langle V^2(0) \rangle = \mu k T, \quad (3.2)$$

so the Einstein relation (2.19) still holds.

The retarded viscous force effect is due to an additional term to the Stokes drag, which is related to the history of the particle acceleration. This additional drag force, proposed by Basset and Boussinesq in earlier times, and nowadays referred to as the *Basset history force*, see *e.g.* Maxey & Riley (1983), reads (in our notation)

$$F_v^B = -\frac{1}{\mu} \sqrt{\frac{\tau_0}{\pi}} \int_{t_*}^t \frac{dV(\tau)/d\tau}{\sqrt{t-\tau}} d\tau, \quad \tau_0 = \frac{a^2}{\nu}, \quad t > t_* \geq -\infty. \quad (3.3)$$

We suggest to interpret the Basset force in the framework of the fractional calculus. In this respect, taking $t_* = 0$, we write

$$\frac{1}{\sqrt{\pi}} \int_0^t \frac{dV(\tau)/d\tau}{\sqrt{t-\tau}} d\tau = \frac{d^{1/2}}{dt^{1/2}} V(t), \quad (3.4)$$

where $d^{1/2}/dt^{1/2}$ denotes the fractional derivative of order 1/2 (in the Caputo sense), see for details Caputo (1967, 1969), Caputo & Mainardi (1971), Mainardi (1996, 1997), Gorenflo & Mainardi (1997) and Podlubny (1999). This definition of fractional derivative differs from the standard one (in the Riemann-Liouville sense) available in classical textbooks on fractional calculus, see *e.g.* Oldham & Spanier (1974), Ross (1975), Samko et al. (1993) and Miller & Ross (1993). In fact, if $f(t)$ denotes a causal function (sufficiently well-behaved) and $0 < \alpha < 1$ we have

$$\begin{aligned} \left(\frac{d^\alpha}{dt^\alpha} \right)_{RL} [f(t)] &= \frac{1}{\Gamma(1-\alpha)} \frac{d}{dt} \int_0^t \frac{f(\tau)}{(t-\tau)^\alpha} d\tau, \\ \left(\frac{d^\alpha}{dt^\alpha} \right)_C [f(t)] &= \frac{1}{\Gamma(1-\alpha)} \int_0^t \frac{df(\tau)/d\tau}{(t-\tau)^\alpha} d\tau, \end{aligned} \quad (3.5)$$

where Γ denotes the Gamma function, and the suffices *RL* and *C* refer to Riemann-Liouville and to Caputo, respectively. Recalling the Riemann-Liouville fractional derivative of the power function

$$\left(\frac{d^\alpha}{dt^\alpha} \right)_{RL} [t^\gamma] = \frac{\Gamma(\gamma+1)}{\Gamma(\gamma+1-\alpha)} t^{\gamma-\alpha}, \quad \gamma > -1, \quad t > 0, \quad (3.6)$$

we obtain

$$\left(\frac{d^\alpha}{dt^\alpha}\right)_{RL} [f(t) - f(0^+)] = \left(\frac{d^\alpha}{dt^\alpha}\right)_C [f(t)]. \quad (3.7)$$

Then, using (2.4), (3.1) and (3.3-4), the Langevin equation turns out to be

$$\frac{dV}{dt} = -\frac{1}{\sigma_e} \left[1 + \sqrt{\tau_0} \frac{d^{1/2}}{dt^{1/2}}\right] V(t) + \frac{1}{m_e} R(t), \quad (3.8)$$

where the suffix C is understood in the fractional derivative. We agree to refer to (3.8) as to the *fractional Langevin equation*.

It is worth noting that if the process is meant to be in thermodynamic equilibrium (at $t_0 = 0$), we should account for the hydrodynamic interaction long memory, and thus it is correct to integrate the Langevin equation (3.8) from $t_* = -\infty$. Basing on the observations by Dufty (1974) and Felderhof (1978), we introduce in (3.8) the random force

$$R_*(t) = R(t) - \frac{1}{\mu} \sqrt{\frac{\tau_0}{\pi}} \int_{-\infty}^0 \frac{dV(\tau)/d\tau}{\sqrt{t-\tau}} d\tau, \quad (3.9)$$

In view of the *fluctuation-dissipation theorem*, Kubo (1966) considered a generalized Langevin equation (GLE), introducing a memory function $\gamma(t)$ to represent a generic retarded effect for the friction force. In our case Kubo's GLE reads (in our notation)

$$\frac{dV}{dt} = - \int_0^t \gamma(t-\tau) V(\tau) d\tau + \frac{1}{m_e} R_*(t), \quad (3.10)$$

where $R_*(t) = R(t) - m_e \int_{-\infty}^0 \gamma(t-\tau) V(\tau) d\tau$.

For the sake of convenience, from now on we shall drop the suffix $*$ in the Langevin equations. Basing on the fundamental hypothesis (2.8). *i.e.* $\langle V(0) R(t) \rangle = 0$, $t > 0$, and using the Laplace transform,

$$f(t) \div \bar{f}(s) = \int_0^\infty e^{-st} f(t) dt, \quad s \in \mathbb{C}$$

(where the sign \div denotes the juxtaposition of a function depending on t with its Laplace transform depending on s), the *fluctuation-dissipation theorem* is readily expressed, according to Mainardi & Pironi (1996), as

$$\bar{C}_V(s) = \overline{\langle V(0) V(t) \rangle} = \frac{\langle V^2(0) \rangle}{s + \bar{\gamma}(s)}, \quad (3.11)$$

$$\bar{C}_R(s) = \overline{\langle R(0) R(t) \rangle} = m_e^2 \langle V^2(0) \rangle \bar{\gamma}(s). \quad (3.12)$$

For the proof of (3.11-12) see Appendix A. The classical results are easily recovered for $t > 0$ noting that, in the absence of added mass and history effects, we get $\bar{\gamma}(s) = 1/\sigma \div \gamma(t) = \delta(t)/\sigma$.

For our fractional Langevin equation (3.8), we note that

$$\bar{\gamma}(s) = \frac{1}{\sigma_e} \left[1 + \sqrt{\tau_0} s^{1/2} \right] \div \gamma(t) = \frac{1}{\sigma_e} \left[\delta(t) - \sqrt{\tau_0} \frac{t^{-3/2}}{2\sqrt{\pi}} \Theta(t) \right], \quad (3.13)$$

where $\Theta(t)$ is the Heaviside step function. Therefore the expression for $\gamma(t)$ turns out to be defined only in the sense of distributions. Specifically, $\delta(t)$ is the well-known Dirac delta function and $t^{-3/2} \Theta(t)$ is the linear functional over test functions, $\phi(t)$, such that

$$\langle t^{-3/2} \Theta(t), \phi(t) \rangle = \int_0^\infty \frac{[\phi(t) - \phi(0)]}{t^{3/2}} dt.$$

For more details on distributions, see *e.g.* Gel'fand & Shilov (1964) or Zemanian (1965).

The significant change with respect to the classical case results from the $t^{-3/2}$ term. Not only does it imply a noninstantaneous relationship between the force and the velocity, but also it is a slowly decreasing function so that the force is effectively related to the velocity over a large time interval. The representation of the force in terms of distributions, as required by the *GLE*, is not strictly necessary since we can use the equivalent fractional form.

Let us now consider the correlation for the random force. The inversion of the Laplace transform $\bar{C}_R(s)$ yields, using (3.12-13),

$$C_R(t) = \frac{m_e^2}{\sigma_e} \langle V^2(0) \rangle \left[\delta(t) - \sqrt{\tau_0} \frac{t^{-3/2}}{2\sqrt{\pi}} \Theta(t) \right], \quad (3.14)$$

Thus, from the comparison with the classical result (2.12), we recognize that, in the presence of the history force, the random force cannot be longer represented uniquely by a white noise; an additional "fractional" or "coloured" noise is present due to the term $t^{-3/2}$ which, as formerly noted by Case (1971), is to be interpreted in the generalized sense of distributions.

Let us now consider the velocity autocorrelation. Inserting (3.13) in (3.11), it turns out

$$\bar{C}_V(s) = \frac{\langle V^2(0) \rangle}{s + [1 + \sqrt{\tau_0} s^{1/2}] / \sigma_e} = \frac{\langle V^2(0) \rangle}{s + \sqrt{\beta/\sigma_e} s^{1/2} + 1/\sigma_e}, \quad (3.15)$$

where, because of (2.2-3) and (3.1), (3.3),

$$\beta = \frac{\tau_0}{\sigma_e} = \frac{9}{2\chi + 1} = \frac{9\rho_f}{2\rho_p + \rho_f}. \quad (3.16)$$

We note from (3.16) that $0 < \beta < 9$, the limiting cases occurring for $\chi = \infty$ and $\chi = 0$, respectively. We also recognize that the effect of the Basset force is expected to be negligible for $\beta \rightarrow 0$, *i.e.* for particles which are sufficiently heavy with respect to the fluid ($\rho_p \gg \rho_f$). In this case we can assume the validity of the classical result (2.11).

Applying in (3.15) the asymptotic theorem for Laplace transform as $s \rightarrow 0$, see *e.g.* Doetsch (1974), we get as $t \rightarrow \infty$,

$$C_V(t) \sim \langle V^2(0) \rangle \sqrt{\beta/(4\pi)} (t/\sigma_e)^{-3/2}, \quad t \rightarrow \infty. \quad (3.17)$$

The presence of such a long-time tail was observed by Alder & Wainwright (1970) in a computer simulation of velocity correlation functions. Furthermore, from (3.15) it is easy to obtain the following results

$$C_V(0) = \lim_{s \rightarrow \infty} s \overline{C}_V(s) = \langle V^2(0) \rangle, \quad \int_0^\infty C_V(t) dt = \overline{C}_V(0) = \sigma_e \langle V^2(0) \rangle. \quad (3.18)$$

The explicit inversion of the Laplace transform in (3.15) can be obtained basing on Appendix B, see also Mainardi et al. (1995), Mainardi (1997), and reads

$$\frac{C_V(t)}{\langle V^2(0) \rangle} = \begin{cases} \frac{a_+ E_{1/2}(a_+ \sqrt{t}) - a_- E_{1/2}(a_- \sqrt{t})}{a_+ - a_-}, & a_\pm = \frac{-\sqrt{\beta} \pm (\beta - 4)^{1/2}}{2 \sqrt{\sigma_e}}, \\ E_{1/2}(a \sqrt{t}) [1 + 2a^2 t] + 2a \sqrt{t/\pi}, & a = -\frac{1}{\sqrt{\sigma_e}} \quad (\beta = 4), \end{cases} \quad (3.19)$$

where

$$E_{1/2}(a \sqrt{t}) = \sum_{n=0}^{\infty} \frac{a^n t^{n/2}}{\Gamma(n/2 + 1)} = e^{a^2 t} \operatorname{erfc}(-a \sqrt{t}) \quad (3.20)$$

denotes the *Mittag-Leffler function* of order 1/2 and erfc the complementary error function. For properties of the Mittag-Leffler function we refer the reader to Erdélyi (1955) and Gorenflo & Mainardi (1997).

Let us now consider the displacement variance, which is provided by (2.14). From the Laplace transform $\overline{\langle X^2(s) \rangle} = 2 \overline{C}_V(s)/s^2$, we derive the asymptotic behaviour of $\langle X^2(t) \rangle$ as $t \rightarrow \infty$, which reads

$$\langle X^2(t) \rangle = 2Dt \left\{ 1 - 2\sqrt{\beta/\pi} (t/\sigma_e)^{-1/2} + O[(t/\sigma_e)^{-1}] \right\}, \quad t \rightarrow \infty, \quad (3.21)$$

where D is the diffusion coefficient (3.2). The explicit expression of the displacement variance turns out to be, taking $\beta \neq 4$,

$$\langle X^2(t) \rangle = 2D \left\{ t - 2\sqrt{\frac{\beta \sigma_e t}{\pi}} + \frac{a_+^3 [1 - E_{1/2}(a_- \sqrt{t})] - a_-^3 [1 - E_{1/2}(a_+ \sqrt{t})]}{(a_+ - a_-)(a_+ a_-)^2} \right\}. \quad (3.22)$$

Thus, the displacement variance is proved to maintain, for sufficiently long times, the linear behaviour which is typical of normal diffusion (with the same diffusion coefficient of the classical case). However, the Basset history force, which is responsible of the algebraic decay of the velocity correlation function, induces a retarding effect ($\propto t^{1/2}$) in the establishing of the linear behaviour.

As we shall see in the next section, the Basset retarding effect is more relevant when the parameter β introduced in (3.16) is big enough, namely when $\chi = \rho_p/\rho_f$ is sufficiently small.

4. Numerical results and discussion

In order to get a physical insight of the effect of the Basset history force we exhibit some plots concerning the velocity autocorrelation (3.19) and the displacement variance (3.22), for some values of the characteristic parameter $\chi = \rho_p/\rho_f$.

We now agree to take *non-dimensional* quantities, by scaling the time with the decay constant σ of the classical Brownian motion and the displacement with the diffusive scale $(D\sigma)^{1/2}$. With these scales the asymptotic equation for the displacement variance reads $\langle X^2(t) \rangle \sim 2t$.

In Figs 1 and 2 we consider the velocity autocorrelation normalized with its initial value $\langle V^2(0) \rangle$ and the displacement variance normalized with its asymptotic value $2t$, assuming $\chi = 0.1, 0.5, 1, 2$. We compare versus time the functions C_V and $\langle X^2 \rangle/(2t)$ provided by our full hydrodynamic approach (added mass and Basset force), in continuous line, with the corresponding ones, provided by the classical analysis, in dashed line, and by the only effect of the added mass, in dotted line. For large times we also exhibit the asymptotic estimations (3.17) and (3.21), in dotted line, in order to recognize their range of validity.

The correlation plots exhibit the well-known algebraic tail. The time necessary to reach the asymptotic behaviour increases as the density ratio χ decreases. By comparing the two figures, it appears that the variance approaches the asymptotic regime as the autocorrelation becomes sufficiently small, independently on its time dependence. In the time interval necessary to reach the asymptotic behaviour the displacement variance exhibits a marked deviation from the standard diffusion.

Because this time interval turns out to be orders of magnitude longer than the classical one, it appears relevant to discuss about various diffusion regimes.

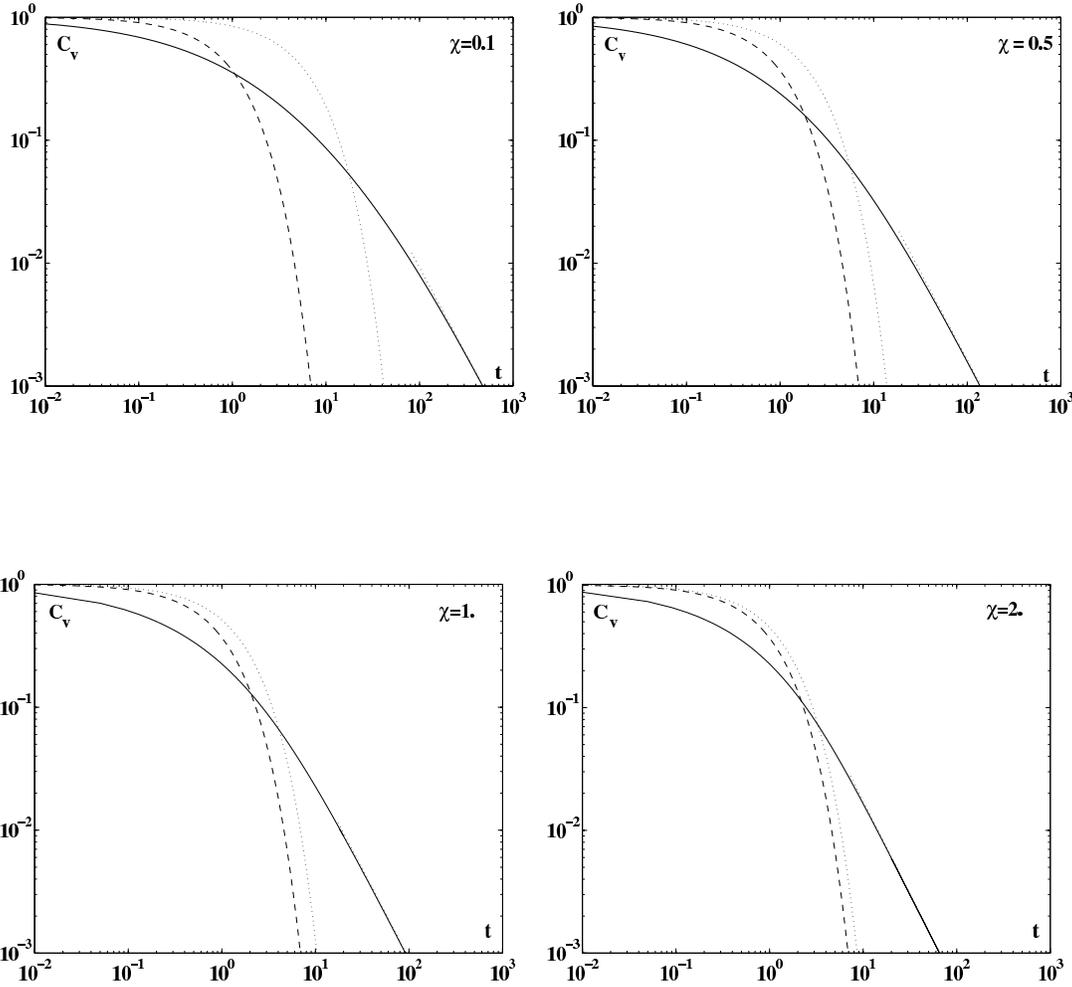


Fig. 1. The velocity autocorrelation versus time for $\chi = 0.1, 0.5, 1, 2$: full hydrodynamic —; added mass \cdots ; classical — — —.

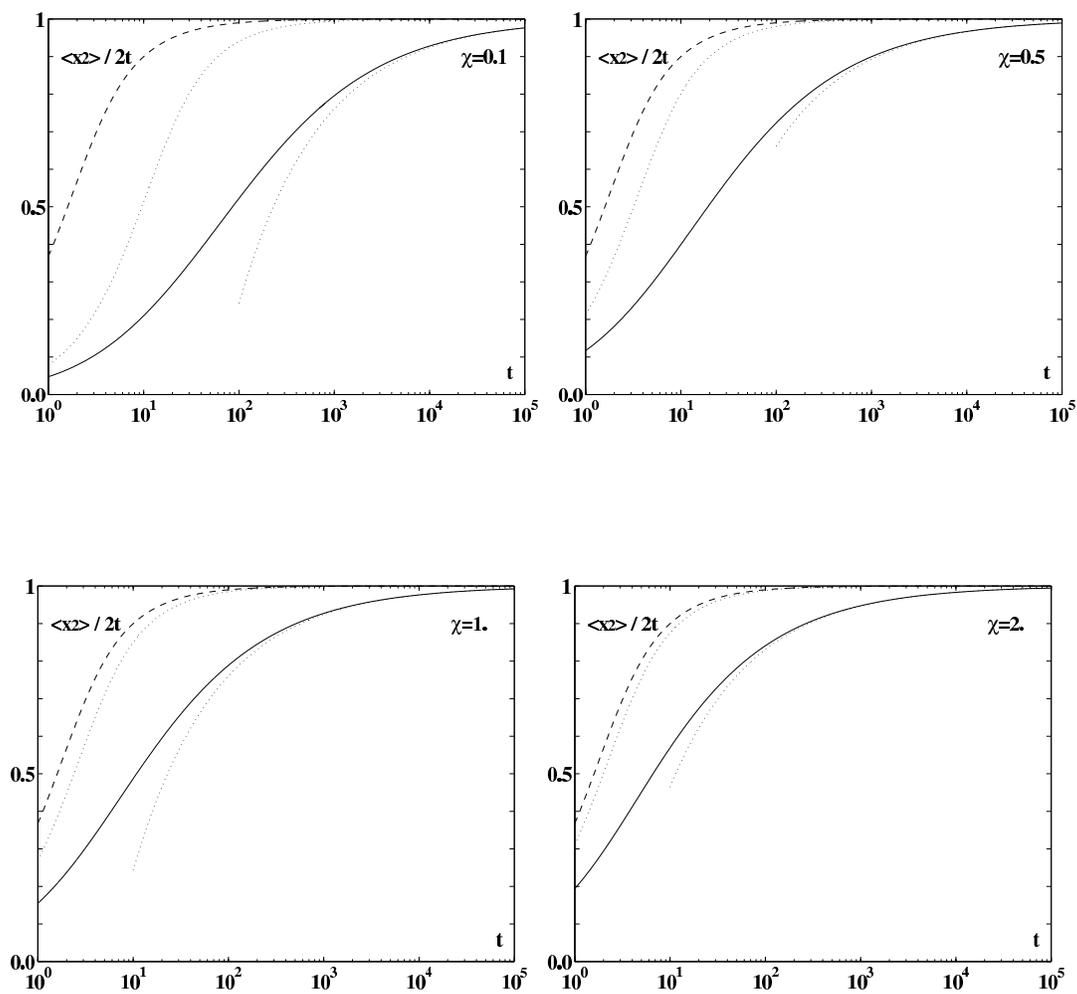


Fig. 2. The displacement variance versus time for $\chi = 0.1, 0.5, 1., 2.:$
 full hydrodynamic —; added mass ...; classical - - -.

In order to characterize these regimes, we consider a time interval (say two decades) starting when the classical analysis foresees the establishment of the asymptotic linear behaviour for the displacement variance, and we look a law of anomalous diffusion $\langle X^2(t) \rangle \sim 2 a t^\alpha$. Evaluating the parameters of the anomalous diffusion, a and α , with a best fit based on the least squared method, we find $0 < a < 1$ and $1 < \alpha < 2$. It results that this law can well approximate the exact behaviour provided by the full hydrodynamic model (3.22) in the selected time range.

We recognize a regime of fast anomalous diffusion; in particular, the diffusion is faster as χ is smaller, with parameters $a \rightarrow 0^+$ and $\alpha \rightarrow 2^-$ as $\chi \rightarrow 0^+$. Of course, the normal diffusion is recovered as $\chi \rightarrow \infty$, since $a \rightarrow 1^-$ and $\alpha \rightarrow 1^+$, and the anomalous effect is significant only for $\chi < 1$.

For increasing values of χ , *i.e.* $\chi = 0.01, 0.05, 0.1, 0.5$, we show in Fig. 3 the function $\langle X^2 \rangle / 2$ corresponding either to our analysis in continuous line (below), or to the classical analysis in dashed line (above). While the classical curve is practically coincident with the linear one, in dotted line, our curve is fitted with a power-law curve, in dashed line, with an exponent $\alpha > 1$.

The best fit values of a and α are reported both in Fig. 3 and in Table I. We recognize that as χ increases the values of a and α increase and decrease, respectively.

χ	a	α
0.001	0.023	1.47
0.05	0.094	1.31
0.1	0.15	1.25
0.5	0.35	1.14

Table I

The best fit values of a and α for increasing values of χ .

We note that, in the time range where the diffusion shows the anomalous behaviour, the displacement variance reads in *dimensional* quantities as

$$\langle X^2(t) \rangle \sim 2 D_a t^\alpha, \quad D_a = a D \sigma^{1-\alpha}; \quad 0 < a < 1, \quad 1 < \alpha < 2. \quad (4.1)$$

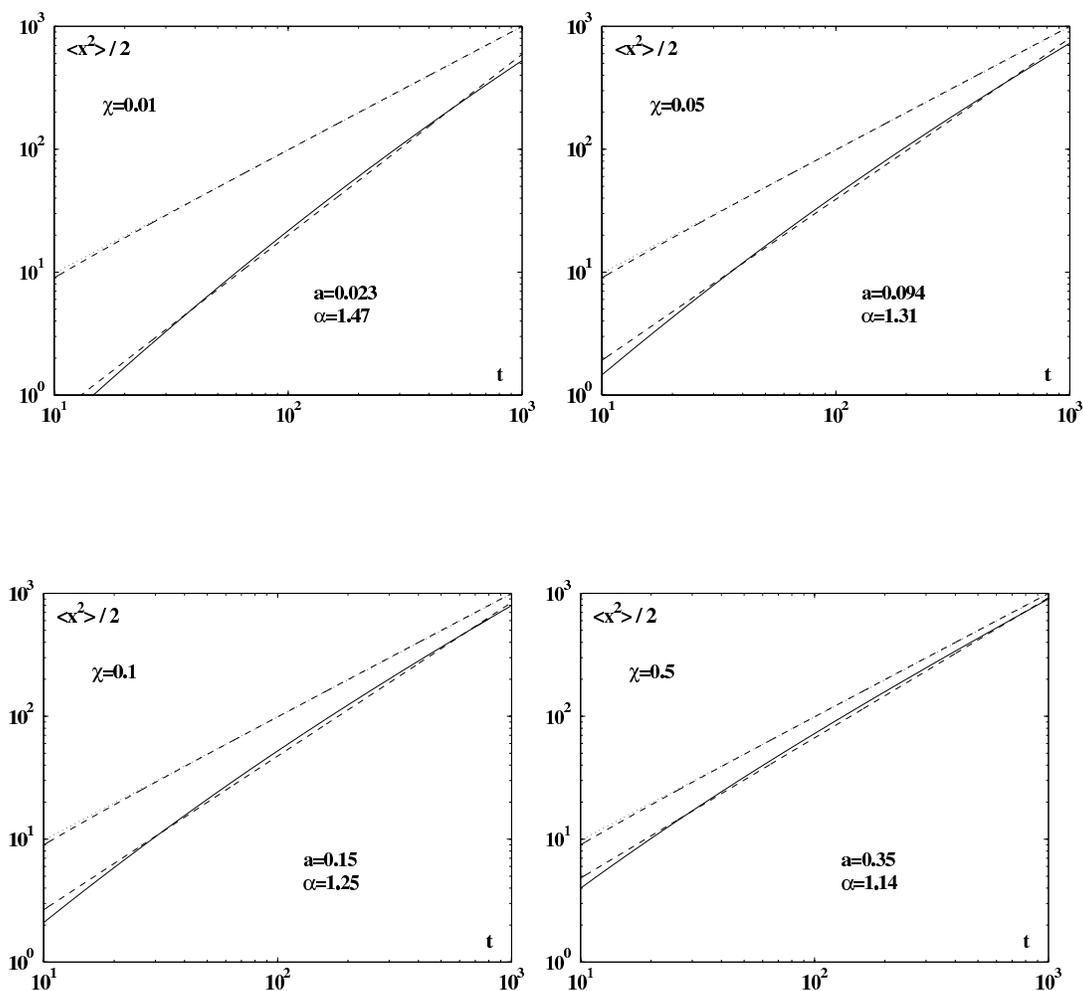


Fig. 3. The displacement variance at large times for $\chi = 0.001, 0.005, 0.01, 0.05$; full hydrodynamic: regime of anomalous diffusion (below), classical: regime of normal diffusion (above).

5. Conclusions

We can summarize our analysis of the Brownian motion based on the hydrodynamic model as follows.

The random force is shown to be represented by a superposition of the usual white noise with a "fractional" noise.

The velocity autocorrelation function $C_V(t)$ is no longer expressed by a simple exponential but by a combination of Mittag-Leffler functions of order $1/2$. As a consequence, one can recognize for $C_V(t)$ a slower decay, proportional to $t^{-3/2}$ as $t \rightarrow \infty$, which indeed is more realistic.

Finally, the displacement variance is shown to maintain, for sufficiently long times, the linear behaviour which is typical of normal diffusion, with the same diffusion coefficient D of the classical case, *i.e.* $\langle X^2(t) \rangle \sim 2Dt$. However, the Basset history force, which is responsible of the algebraic decay of the velocity correlation function, induces a retarding effect in the establishing of the linear behaviour.

The above effect is seen to be evident for "light" Brownian particles, *i.e.* less dense than the fluid. In these cases, if one considers the variance versus time in the interval corresponding to the establishment of the classical linear behaviour, a best fit is obtained with the law $\langle X^2(t) \rangle \sim 2D_a t^\alpha$, where D_a is a sort of effective coefficient of anomalous diffusion with exponent $1 < \alpha < 2$. Thus, the characteristic exponent being greater than 1, the resulting effect appears such as a manifestation of *fast anomalous diffusion*. Less dense are the Brownian particles with respect to the fluid, greater is the exponent α , *i.e.* faster is the diffusion, where $\alpha \rightarrow 2$ as $\rho_p \rightarrow 0$.

In conclusion, if an observer investigates the time evolution of a cloud of light Brownian particles, he recognizes that the normal diffusion is preceded by a regime of fast anomalous diffusion, which lasts for long time. If the observation interval is not sufficiently long, he may be induced to trust in the occurring of fast anomalous diffusion.

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Appendix A

Let us consider the *generalized Langevin equation* (3.10), that we write as

$$R(t) = m_e \left[\dot{V}(t) + \gamma(t) * V(t) \right], \quad (\text{A.1})$$

where \cdot denotes time differentiation and $*$ time convolution. The assumption of stationarity for the stochastic processes along with the following hypothesis

$$\langle R(t) \rangle = 0, \quad \langle V(0) R(t) \rangle = 0, \quad t > 0, \quad (\text{A.2})$$

allows us to derive, by using the Laplace transforms, the two *fluctuation-dissipation theorems*

$$\overline{C}_V(s) := \overline{\langle V(0) V(t) \rangle} = \frac{\langle V^2(0) \rangle}{s + \overline{\gamma}(s)}, \quad (\text{A.3})$$

and

$$\overline{C}_R(s) := \overline{\langle R(0) R(t) \rangle} = m_e^2 \langle V^2(0) \rangle \overline{\gamma}(s). \quad (\text{A.4})$$

Our derivation is alternative to the original one by Kubo (1966) who used Fourier transforms; furthermore, it appears useful for the treatment of our *fractional Langevin equation*.

Multiplying both sides of (A.1) by $V(0)$ and averaging, we obtain

$$\langle V(0) \dot{V}(t) \rangle + \gamma(t) * \langle V(0) V(t) \rangle = 0. \quad (\text{A.5})$$

The application of the Laplace transform to both sides of (A.5) yields

$$s \overline{\langle V(0) V(t) \rangle} - \langle V^2(0) \rangle + \overline{\gamma}(s) \overline{\langle V(0) V(t) \rangle} = 0, \quad (\text{A.6})$$

from which we just obtain (A.3).

Multiplying both sides of (A.1) by $R(0)$ and averaging, we obtain

$$C_R(t) := \langle R(0) R(t) \rangle = m_e^2 \left[\langle \dot{V}(0) \dot{V}(t) \rangle + \gamma(t) * \langle \dot{V}(0) V(t) \rangle \right]. \quad (\text{A.7})$$

Noting that, by the stationary condition,

$$\langle \dot{V}(0) V(0) \rangle = 0, \quad \langle \dot{V}(0) V(t) \rangle = -\langle V(0) \dot{V}(t) \rangle, \quad (\text{A.8})$$

the application of the Laplace transform to both sides of (A.7) yields

$$\overline{C}_R(s) = m_e^2 \left\{ s \overline{\langle \dot{V}(0) V(t) \rangle} - \overline{\gamma}(s) \left[s \overline{\langle V(0) V(t) \rangle} - \langle V^2(0) \rangle \right] \right\}. \quad (\text{A.9})$$

Since

$$\overline{\langle \dot{V}(0) V(t) \rangle} = -\overline{\langle V(0) \dot{V}(t) \rangle} = -s \overline{\langle V(0) V(t) \rangle} + \langle V^2(0) \rangle, \quad (\text{A.10})$$

we get

$$\overline{C}_R(s) = m_e^2 \left\{ s \left[-s \overline{C}_V(s) + \langle V^2(0) \rangle - \overline{\gamma}(s) \overline{C}_V(s) \right] + \overline{\gamma}(s) \langle V^2(0) \rangle \right\}, \quad (\text{A.11})$$

from which, accounting for (A.3), we just obtain (A.4).

Appendix B

In this Appendix we report the detailed manipulations necessary to obtain the result (3.19) as Laplace inversion of (3.15). For this purpose we need to consider the Laplace transform

$$\bar{N}(s) = \frac{1}{s + b s^{1/2} + 1}, \quad b = \sqrt{\beta}, \quad (B.1)$$

and recognize that

$$\frac{C_V(t)}{\langle V^2(0) \rangle} = N(t/\sigma_e) \div \sigma_e \bar{N}(\sigma_e s) = \frac{1}{s + \sqrt{\beta/\sigma_e} s^{1/2} + 1/\sigma_e}, \quad (B.2)$$

where we have used the sign \div for the juxtaposition of a function depending on t with its Laplace transform depending on s . The required result is obtained by expanding $\bar{N}(s)$ into partial fractions and then inverting. Considering the two roots λ_{\pm} of the polynomial $P(z) \equiv z^2 + b z + 1$ with $z = s^{1/2}$, we must treat separately the following two cases: *i*) $0 < b < 2$, or $2 < b < 3$, and *ii*) $b = 2$, which correspond to two distinct roots ($\lambda_+ \neq \lambda_-$), or two coincident roots ($\lambda_+ \equiv \lambda_- = -1$), respectively. We obtain

i) $b \neq 2 \iff \beta \neq 4, \chi \neq 5/8,$

$$\bar{N}(s) = \frac{1}{s + b s^{1/2} + 1} = \frac{A_+}{s^{1/2} (s^{1/2} - \lambda_+)} - \frac{A_-}{s^{1/2} (s^{1/2} - \lambda_-)}, \quad (B.3)$$

with

$$\lambda_{\pm} = \frac{-b \pm (b^2 - 4)^{1/2}}{2} = \frac{1}{\lambda_{\mp}}, \quad A_{\pm} = \frac{\lambda_{\pm}}{\lambda_+ - \lambda_-}; \quad (B.4)$$

ii) $b = 2 \iff \beta = 4, \chi = 5/8,$

$$\bar{N}(s) = \frac{1}{s + 2 s^{1/2} + 1} = \frac{1}{(s^{1/2} + 1)^2}. \quad (B.5)$$

The Laplace inversion of (B.3) and (B.5) turns out, see below,

$$N(t) = \begin{cases} i) & A_+ E_{1/2}(\lambda_+ \sqrt{t}) - A_- E_{1/2}(\lambda_- \sqrt{t}), \\ ii) & (1 + 2t) E_{1/2}(-\sqrt{t}) - 2 \sqrt{t/\pi}, \end{cases} \quad (B.6)$$

where

$$E_{1/2}(\lambda \sqrt{t}) = \sum_{n=0}^{\infty} \frac{\lambda^n t^{n/2}}{\Gamma(n/2 + 1)} = e^{\lambda^2 t} \operatorname{erfc}(-\lambda \sqrt{t}) \quad (B.7)$$

denotes the *Mittag-Leffler function* of order 1/2 and erfc denotes the complementary error function. In view of (B.1-2), equation (B.6) is equivalent to (3.19).

Let us first recall the essentials of the generic Mittag-Leffler function in the framework of the Laplace transforms. The Mittag-Leffler function $E_\alpha(z)$ with $\alpha > 0$, so named from the great Swedish mathematician who introduced it at the beginning of this century, is defined by the following series representation, valid in the whole complex plane,

$$E_\alpha(z) = \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(\alpha n + 1)}, \quad \alpha > 0, \quad z \in \mathbb{C}. \quad (B.8)$$

It turns out that $E_\alpha(z)$ is an *entire function*, of order $\rho = 1/\alpha$ and type 1, which provides a generalization of the exponential function.

The Mittag-Leffler function is connected to the Laplace integral through the equation

$$\int_0^{\infty} e^{-u} E_\alpha(u^\alpha z) du = \frac{1}{1-z}, \quad \alpha > 0. \quad (B.9)$$

This integral is fundamental in the evaluation of the Laplace transform of $E_\alpha(\lambda t^\alpha)$ with $\lambda \in \mathbb{C}$ and $t \geq 0$. Putting in (B.9) $u = st$ and $u^\alpha z = \lambda t^\alpha$, we get the following Laplace transform pair

$$E_\alpha(\lambda t^\alpha) \div \frac{s^{\alpha-1}}{s^\alpha - \lambda}, \quad \text{Re } s > |\lambda|^{1/\alpha}. \quad (B.10)$$

We note that, up to our knowledge, in the handbooks containing tables for the Laplace transforms, the Mittag-Leffler function is ignored so that the transform pair (B.10) does not appear if not in the special case $\alpha = 1/2$. In fact, in this case we recover from (B.10) the basic Laplace transform pair

$$\frac{1}{s^{1/2}(s^{1/2} - \lambda)} \div E_{1/2}(\lambda \sqrt{t}), \quad (B.11)$$

where the Mittag-Leffler function can be expressed in terms of known functions, as shown in (B.7). As an exercise we can derive from (B.11) the following transform pairs and consequently the result (B.6):

$$\frac{1}{s^{1/2} - \lambda} = \frac{1}{s^{1/2}} + \frac{\lambda}{s^{1/2}(s^{1/2} - \lambda)} \div \frac{1}{\sqrt{\pi t}} + \lambda E_{1/2}(\lambda \sqrt{t}), \quad (B.12)$$

$$\frac{1}{s^{1/2}(s^{1/2} - \lambda)^2} = -2 \frac{d}{ds} \left(\frac{1}{s^{1/2} - \lambda} \right) \div 2 \sqrt{\frac{t}{\pi}} + 2 \lambda t E_{1/2}(\lambda \sqrt{t}), \quad (B.13)$$

$$\begin{aligned} \frac{1}{(s^{1/2} - \lambda)^2} &= \frac{1}{s^{1/2}(s^{1/2} - \lambda)} + \frac{\lambda}{s^{1/2}(s^{1/2} - \lambda)^2} \\ &\div 2 \lambda \sqrt{\frac{t}{\pi}} + (1 + 2 \lambda^2 t) E_{1/2}(\lambda \sqrt{t}). \end{aligned} \quad (B.14)$$

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