Vol. 18, No. 2 (2019) 571-579



Revista Mexicana de Ingeniería Química

### MULTIPLICATIVE NOISE EFFECTS IN AN OPTICALLY TRAPPED AEROSOL EFECTOS DE RUIDO MULTIPLICATIVO EN UN AEROSOL CONFINADO EN UNA TRAMPA ÓPTICA

R.F. Rodríguez<sup>1</sup> and E. Salinas-Rodríguez<sup>2\*</sup>

<sup>1</sup>Instituto de Física, Universidad Nacional Autónoma de México. Apdo. Postal 20-364, 01000 México, D. F., México. Fellow of SNI, México. Also at FENOMEC, UNAM, México.

<sup>2</sup>Departamento I. P. H., Universidad Autónoma Metropolitana, Iztapalapa. Apdo. Postal 55-534, 09340 México, D. F., México. Fellow of SNI, México.

Received: March 10, 2019; Accepted: March 19, 2019

#### Abstract

We develop a stochastic model to describe the effects of external noise on the position and power spectrum fluctuations of the particles of an optically confined aerosol. We propose that externally imposed noise may induce a random frequency of the confining laser. Our analysis is based on a linear multiplicative Langevin equation and on the cumulant method. We also derive the associated Fokker-Planck equation and the mean square displacement of the particles. Our model predicts that these multiplicative fluctuations may produce a large effect on these correlations and on the corresponding fluctuations spectra which might be measurable.

Keywords: aerosols, fluctuations, stochastic processes, external noise.

#### Resumen

Desarrollamos un modelo estocástico para describir los efectos de ruido externo multiplicativo en la función de correlación y en el espectro de las fluctuaciones de la posición de las partículas de un aerosol confinado ópticamente. Proponemos que fluctuaciones impuestas externamente inducen una frecuencia aleatoria del láser confinante. Nuestro análisis se basa en una ecuación de Langevin lineal multiplicativa y en el método de cumulantes. Se construye la ecuación de Fokker-Planck asociada a esta ecuación estocástica multiplicativa y obtenemos el desplazamiento cuadrático medio de las partículas. Nuestro modelo predice que las fluctuaciones multiplicativas producen un efecto significativo en las correlaciones de posición y en el espectro de fluctuaciones correspondiente y que por lo tanto podrían ser medibles.

Palabras clave: aerosoles, fluctuaciones, procesos estocásticos, ruido externo.

### **1** Introduction

An aerosol is a multiphase system composed of a suspension of fine solid particles or liquid droplets in a gas. They occur in a broad range of important subject fields in chemical engineering such as, hydrosol or aerosol filtration, depending on whether liquid-gas suspension is involved (Tien and Ramaro, 2007). Besides water or air, systems which may be treated by granular filtration include such diverse substances as flue gas, combustion products, molten metal, petrochemical feedstocks, or polymers. While in most cases these filtration processes are carried out in the fixed-bed mode, they may also be conducted in fluidized-bed mode (Knettig and Beeckmans, 1974). More recent efforts in aerosol research in chemical engineering involve the nucleation of supersaturated droplets, the dispersion of particles as clouds, and gas cleaning (Lu *et al.*, 2005; Hrubý *et al.*, 2018). Also, understanding the process of creation of aerosols can help to design techniques for removal of aerosol after combustion; for example, the removal of particulate non-combusted material in coal fired power stations by electrostatic precipitators (Slama *et al.*, 2000). Also, the development of bioseparation techniques applicable to the separation of bioparticles such as microorganisms and macromolecules (proteins and DNA) (Garza-García and Lapizco-Encinas, 2010).

<sup>\*</sup> Corresponding author. E-mail: liz\_sabe@yahoo.com Tel. 55-58-04-46-44.

https://doi.org/10.24275/uam/izt/dcbi/revmexingquim/2019v18n2/Rodriguez issn-e: 2395-8472

In general aerosol systems, the droplet and gas phases are coupled. Transport processes and changes in chemical composition or temperature of the gas will affect the properties of the droplets and vice versa. Thus, quantification of the chemistry involved cannot be determined through studying only the bulk system. Instead one must also analyze individual aerosol droplets. In the last two decades, the technique of optical tweezers has been widely applied to measure forces on single particles and materials in physicochemical and bio-physical systems (Lang et al., 2002; Neuman and Block, 2004). Properties of the Brownian motion of optically trapped aerosol particles, such as the relation between force and displacement, depend on particle parameters like size and shape, and on the index of refraction of the bulk fluid. They depend as well, on external parameters like the wavelength and power of the trapping laser. In the regime of small particle displacements, the particle-trap interaction can be well approximated by a harmonic potential, and the particles experience a Hookean restoring force with a trapping stiffness  $\kappa$ , when displaced by Brownian stochastic forces (Denk and Webb, 1990; Yao et al., 2009: Burnham and McGloin, 2009, 2011: Burnham et al., 2010). The analysis of the random forces and position fluctuations of aerosol particles is essential to determine some properties like  $\kappa$ , which is related to the variance of the power spectrum of positions of the trapped objects (Berg-Sorensen and Flyvbjerg, 2004). Also, this power spectrum is often used to detect their position (Allersma et al., 1998), to measure forces (Denk and Webb, 1990), or to investigate colloidal dynamics (Gishlan et al., 1994).

Experimental methods based on the optical trapping of liquid aerosols (Burnham *et al.*, 2010), is a powerful tool in a variety of physico-chemical research fields, such as viscosimetry (Pesce *et al.*, 2005), or in molecular biology (Lang *et al.*, 2002). These methods may serve for studying Brownian dynamics in over and underdamped conditions. Optical trapping methods have been also used for measurements of Casimir forces (Hertlein *et al.*, 2008), to analyze the validity of fluctuation-dissipation theorems (Carberry *et al.*, 2007) and to study colloidal crystals (Lee and Grier, 2005), thermal ratchets and freezing (Chowdhury *et al.*, 1985).

For the purpose of future comparison with the results of this work, we briefly recall the usual stochastic description of aerosols. The trapped particles are microspheres with radius R, mass m, immersed in a fluid at temperature T, with dynamic viscosity  $\mu$  and mass density  $\rho$ . Its equation of motion is usually modeled by a Langevin equation with additive noise (Burnham *et al.*, 2010).

$$m\ddot{x}(t) + \gamma_0 \dot{x} + \kappa x(t) = \eta(t). \tag{1}$$

Here  $\gamma_0$  is the viscous damping of the medium (drag coefficient) and is well approximated by a constant determined by Stokes law,  $\gamma_0 = 6\pi\mu R$ . Since the phenomenological equation (1) is compatible with the local equilibrium assumption and the linear response regime, it is reasonable to assume that for near equilibrium states, the additive random force  $\eta(t)$  is a Gaussian stochastic processes. More specifically, for near equilibrium states, it is assumed to be a white noise with zero average  $\langle \eta(t) \rangle = 0$ , and an instantaneous auto-correlation

$$\langle \eta(t)\eta(t')\rangle = D\delta(t-t'),$$
 (2)

Here the parameter  $D = 2k_B T \gamma_0$  measures the strength of the stochastic force and  $k_B$  is Boltzmann's constant.

In almost all experiments with optical tweezers in a liquid environment, it is considered that the particles behave as overdamped oscillators. However, underdamped motions have also been observed (Jokutty et al., 2005). This is indeed the usual situation for optical trapping for beads and particles: They are generally in a low Reynolds number regime and strongly damped. However, there is an additional manifestation of this damping, namely, thermal fluctuations of Brownian motion. The fluctuationdissipation theorem guarantees that any system with damping, will eventually attain equilibrium and show fluctuations, whose size depends inversely on the damping. Actually, these experimental observations motivate the main purpose of this work. In contrast to the additive fluctuations described by (1), we pursue to calculate the power spectrum of position fluctuations of the optically trapped particles of an aerosol, from a stochastic model with parametric multiplicative noise. More specifically, our objective is to investigate the effects produced by multiplicative external noise in the (corner) frequency of the confining laser, on the position-position correlation function of the trapped particles and on their power spectrum. To our knowledge the effect of these type of fluctuations has not been considered in the literature for aerosols. Although there are several approaches to deal with multiplicative noise (Fox, 1972; Hänggi, 1978; Sancho and San Miguel, 1980a, 1980b, 1984; Hernández-Machado et al., 1983), here we use a description based on cumulant techniques. The practical usefullness of a direct application of these methods is appropriate to

deal with linear processes (Fox, 1972; van Kampen, 1981a; Rodríguez *et al.*, 2001, 2011).

For this purpose the work is organized as follows. In the next section 2 we discuss the stochastic model that is derived from (1) when inertial effects are neglected, but the fluctuations remain additive. Then in section 3 we generalize this model by neglecting the additive noise, and introducing parametric noise in the frequency of the confining laser. We calculate analytically the position-position correlation function and its spectral density. In section 4 we derive the Fokker-Planck equation associated to the multiplicative Langevin equation and calculate the second moment of the position to calculate the mean square displacement of the particles. We close the paper in section 5 by making some further physical remarks.

## 2 Underdamped regime with additive noise

With the purpose of future comparison with our future results for the multiplicative case, we recall the analytic expressions for the position correlation function and its corresponding fluctuation spectrum for the case of additive noise. It is experimentally well established that a micro-sized colloidal particle optically trapped in a viscous fluid has a characteristic time for loss of energy through friction given by,  $t_{inert} = m/\gamma_0 \approx 10$  ns. Since this time is far shorter than the experimental time resolution,  $t_{exp} \approx 20\mu$ s, its motion is extremely overdamped and the inertial term in (1) can be dropped leaving (Denk and Webb, 1990; Burnham, 2009),

$$\gamma_0 \dot{x}(t) + \gamma_0 \omega_c x(t) = \eta(t), \tag{3}$$

where  $\omega_c = \kappa / \gamma_0$  is the corner frequency, defined as the frequency at which the power reaches half its low frequency asymptotic value.  $\eta(t)$  is a zero averaged,  $\langle \eta(t) \rangle = 0$ , and stationary Gaussian stochastic processes with auto-correlation function

$$\langle \eta(t)\eta(t')\rangle = C(|t-t'|). \tag{4}$$

When the system described by Eq. (3) is in an equilibrium state, the functions  $\omega_c$  and C(t) are related to each other by means of the fluctuation-dissipation theorem (Kubo, 1966),

$$C(t) = 2k_B T \gamma_0, \tag{5}$$

where  $k_B$  is the Boltzmann constant. In this case the random force is sometimes referred to as internal noise. However, in nonequilibrium systems the driving noise and the dissipation may have different physical origin and no fluctuation-dissipation relation holds. In such a case the driving noise will be referred to as external noise.

To calculate the fluctuation spectrum  $S(\omega)$  of the position x(t) in this overdamped additive regime, we have to compute first the one-time position autocorrelation function  $\chi(t)$  defined by

$$\chi(t) \equiv \left\langle x_0 \langle x(t) \rangle_{x_0} \right\rangle^{eq}.$$
 (6)

Here the notation indicates the following: take a certain initial value  $x_0$  at t = 0, calculate the average  $\langle x(t) \rangle_{x_0}$  conditional on the given  $x_0$ ; multiply it by  $x_0$  and average the product over the values  $x_0$ , as they occur in an equilibrium distribution. From (3) the additive normalized position autocorrelation function turns out to be

$$\chi_w^a(t) \equiv \frac{\left\langle x_0 \langle x(t) \rangle_{x_0} \right\rangle^{eq}}{x_0^2} = e^{-\omega_c t}.$$
 (7)

According to the Wiener-Kintchine theorem the power spectrum of positions of the trapped objects,  $S^{a}(\omega)$ , is given by (van Kampen, 1981)

$$S_{w}^{a} = \frac{2}{\pi} \int_{0}^{\infty} \chi^{a}(\tau) \cos(|\omega\tau|) d\tau = \frac{2}{\pi} \frac{1}{\omega^{2} + \omega_{c}^{2}}$$
(8)

In the following section we derive the corresponding expressions for multiplicative noise and compare them with  $\chi^a_w(t)$  and  $S^a_w(\omega)$ .

# 3 Non-inertial limit with multiplicative noise

It is well-known that the fluctuations existing in open systems may be conveniently classified into internal and external fluctuations. The former are those self-originated in the system, while the latter are determined by the environment. Internal fluctuations are a consequence of the large number of degrees of freedom averaged out in a macroscopic description; they scale with the size of the system and therefore vanish in the thermodynamic limit, except at a critical point where long range order is established. Their study is an important and well known part of statistical mechanics (Landau, 1970). In contrast, external fluctuations exist when a system is under the influence of noise caused by a natural or induced randomness of the environment of the system. These fluctuations play the role of an external field driving the system and they do not scale with the system size. Thus, if external noise is present in a macroscopic system it will dominate over internal fluctuations. Among others, physico-chemical systems where the effect of external noise has been considered include fluids (Gollub and Steinman, 1980); lasers and optical systems (Arecci, and Politi, 1979); chemical reactions (De Kepper and Horsthemke, 1978), and liquid crystals (Rodríguez et al., 1997). In these applications external noise is usually considered as a stochastic process that is introduced into the parameters of the deterministic equations which describe the macroscopic behavior of a system. In any case, in this work the term fluctuations in a state variable or in a system's parameter, shall be identified with the random deviations from its average near equilibrium values.

Let us now change the physical situation of the previous section by assuming that the state of the surrounding fluid is modified by an external noise. Multiplicative stochastic equations arise frequently in the description of systems under the influence of external or parametric noise. The description of their dynamics is usually given in terms of a phenomenological equation which describe the system in the absence of noise. Then an appropriate parameter in this equation is replaced by a stochastic process and is allowed to fluctuate with prescribed statistics. If the fluctuating parameter enters linearly in the deterministic equation, one is led to a stochastic equation of the Langevin type with multiplicative noise (Horsthemke, 1981).

In our model an external noise is superimposed on the corner frequency  $\omega_c$  of the confining laser. This parameter then fluctuates around its average (deterministic) value with fast and small fluctuations,

$$\omega_c \to \omega_c + \zeta(t). \tag{9}$$

Here  $\zeta(t)$  is a so far unspecified external noise which, as before, is assumed to be a stationary Gaussian process, with zero mean and arbitrary finite auto-correlation

$$\left\langle \zeta(t)\zeta(t')\right\rangle = C(|t-t'|). \tag{10}$$

Under these assumptions (3) can be rewritten as the linear multiplicative stochastic equation

$$\dot{x} = A(t)x \equiv [A^{(0)} + \alpha A^{(1)}(t)]x, \qquad (11)$$

where we have identified

$$A^{(0)} \equiv -\omega_c, \quad A^{(1)}(t) \equiv \zeta(t).$$
 (12)

Furthermore,  $A^{(1)}(t)$  is assumed to have a finite autocorrelation time  $\tau_c$ , in the sense that for any two times  $t_1$ ,  $t_2$ , such that  $|t_1 - t_2| \ge \tau_c$ , one may treat  $A^{(1)}(t_1)$  as statistically independent of  $A^{(1)}(t_2)$ . It is also convenient to assume that  $A^{(1)}(t)$  is a stationary stochastic process, so that  $\langle A^{(1)}(t) \rangle$  can be incorporated in  $A^{(0)}$  by setting  $A^{(0)\prime} = A^{(0)} + \alpha \langle A^{(1)}(t) \rangle$  and  $A^{(1)\prime}(t) =$  $A^{(1)}(t) - \langle A^{(1)}(t) \rangle$ , so that  $\langle A^{(1)\prime}(t) \rangle = 0$ . Assuming that this has been done, in what follows we shall suppress the primes and treat Eq. (11) with  $\langle A^{(1)}(t) \rangle =$ 0. As mentioned before, for near equilibrium states the fluctuations are known to be small from statistical mechanics, therefore it is reasonable to assume that the parameter  $\alpha$ , which measures the magnitude of the fluctuations in the coefficient  $A_1(t)$ , is small with a finite correlation time  $\tau_c$ . These assumptions are conveniently expressed in terms of the Kubo number,  $\alpha \tau_c$ , which is thus assumed to be small,  $\alpha \tau_c \ll 1$ .

There are few approaches to solve a multiplicative stochastic equation like Eq. (11) (Hänggi, 1978; Grabert *et al.*, 1980). Among them the functional methods, based on short  $\tau_c$  expansions, are often used in nonlinear stochastic equations (Sancho and San Miguel, 1980a, 1980b). However, the cumulant techniques (Fox, 1972; van Kampen, 1981a) are more appropriate for linear multiplicative equations. Within this last approach and using a heuristic derivation, van Kampen has derived the following non-stochastic differential equation for the expectation value  $\langle x(t) \rangle$ , (van Kampen, 1981a)

$$\frac{d}{dt}\langle x(t)\rangle = \left[A^{(0)} + \alpha^{2} \int_{0}^{\infty} \left\langle A^{(1)}(t)e^{\tau A^{(0)}}A^{(1)}(t-\tau) \right\rangle e^{-\tau A^{(0)}}d\tau \right] \langle x(t)\rangle, \quad (13)$$

This equation reinforces the statement that the free motion of x(t) is slow compared to the fluctuations in  $A_1(t)$ . This heuristic expression can be also rigorously derived by using the cumulants theory (van Kampen, 1981a). In this case (13) reduces to

$$\frac{d}{dt}\langle x(t)\rangle = \left\{-\omega_c + \alpha^2 c_0(t)\right\}\langle x(t)\rangle, \qquad (14)$$

where we have used (10) and

$$c_0(t) = \int_0^\infty \langle \zeta(t)\zeta(t-\tau) \rangle d\tau.$$
 (15)

www.rmiq.org

Thus, for given  $x_0$  we get

$$\langle x(t) \rangle_{x_0} = x_0 \exp\left\{-[\omega_c - \alpha^2 c_0(t)]t\right\}$$
(16)

From (6) it follows that the normalized positionposition correlation function is

$$\chi^m(t) = \exp\left\{-[\omega_c - \alpha^2 c_0(t)]t\right\}.$$
 (17)

and for the special case of white noise where  $C(t) = \beta \delta(t - t')$  and  $c_0(t) = \beta$ ,

$$\chi_{w}^{m}(t) = \exp\left\{-\frac{1}{\gamma_{0}}(\kappa - \alpha^{2}\gamma_{0}\beta)t\right\},$$
 (18)

The Wiener-Kintchine theorem then leads to

$$S_{w}^{m}(\omega) = \sqrt{\frac{2}{\pi}} \frac{\gamma_{0}(\kappa - \alpha^{2}\gamma_{0}\beta)}{(\kappa - \alpha^{2}\gamma_{0}\beta)^{2} + \gamma_{0}^{2}\omega^{2}} = \sqrt{\frac{2}{\pi}} \frac{\omega_{0}}{\omega^{2} + \omega_{0}^{2}},$$
(19)

with  $\omega_0 \equiv (\kappa - \alpha^2 \gamma_0 \beta)/\gamma_0$ . A plot of Eqs. (17) and (7) is shown in Fig. 1, where  $\chi_w^a(t)$  and  $\chi_w^m(t)$  are functions of *t* for different values of  $\alpha$ . The material aerosol parameters were taken as  $\kappa = 1pN/\mu m$ ,  $\mu = 2 \times 10^{-5}$ kg/ms and  $R = 5\mu m$ , which correspond to a microsphere in water (Burnham, 2009). The frequency of the laser is  $\omega_c = 531 s^{-1}$ . Figure 1 shows that the intensity of  $\chi_w^m(t)$  is always larger than that of  $\chi_w^a(t)$ , and that the multiplicative position correlation decays more slowly than the additive one. This behavior shows that multiplicative noise may produce a significant, and perhaps measurable effects, on the position correlation function of the aerosol particles.

This comparison between  $\chi_w^a(t)$  and  $\chi_w^m(t)$  can be better quantified by plotting the ratio,  $R(t) \equiv \chi^m(t)/\chi^a(t)$ , as a function of *t*. This is displayed in Fig. 2 and the curves show that, indeed, multiplicative noise in the frequency of the confining laser may increase significantly the fluctuations of the positions of the trapped particles. Note that the quantitative effect of R(t) depends on the values of both parameters  $\alpha$  and  $\beta$ . The multiplicative correlation function can be one order of magnitude larger than the additive one.



Fig. 1. The correlation functions  $\chi_w^a(t)$  and  $\chi_w^m(t)$ , given by Eqs. (7) and (18), as functions of time.



Fig. 2. Plot of the ratio  $R(t) = \chi_w^m / \chi^a(t)$  as a function of time for the noise parameter values  $\alpha^2 \beta = (2, 2.6, 3) \times 10^2 \text{ s}^{-1}$ .



Fig. 3. The power spectra of position fluctuations,  $S_w^a(\omega)$  and  $S_w^m(\omega)$ , for the trapped particles, as functions of  $\omega$ .



Fig. 4. Power spectra of position fluctuations neglecting inertia and calculated from Eqs. (8) and (19) for the same parameter values as in Figs. 1-3.

The calculated power spectra of position fluctuations of the trapped particles,  $S_w^a(\omega)$  and  $S_w^m(\omega)$ , as functions of  $\omega$ , are shown in Fig. 3. Note that the curves show the same behavior as the one observed in Fig. 1 for the position correlations functions, namely, the multiplicative spectrum may be ~ 100% larger than the additive one in the interval between  $\omega \approx \pm 372$  Hz. Below these values there is a crossover, and  $S_w^m(\omega)$  decreases in ~ (30 – 40)%. This effect can be more easily appreciated in the log-log plot of  $S_w^a(\omega)$  and  $S_w^m(\omega)$  shown in Fig. 4. Both curves show a plateau and the spectra show a characteristic tail  $\omega^{-2}$ ;  $S_w^a(\omega)$  and  $S_w^m(\omega)$  show a low frequency plateau of amplitudes  $\gamma_0/\kappa^2$  and  $\gamma_0/(\kappa - \alpha^2 \gamma_0 \beta)^2$ . Note that although they have the same form, the multiplicative one has an *effective* lower trapping stiffness  $\kappa - \alpha^2 \gamma_0 \beta$ .

## 4 Fokker-Plank equation and mean square displacement

For the purpose of calculating the mean square displacement (MSD),

$$\langle [\Delta x(t)]^2 \rangle \equiv \langle x^2(t) \rangle - \langle x(t) \rangle^2, \tag{20}$$

of the trapped particles for multiplicative non-inertial position fluctuations, it is necessary to calculate the second moment  $\langle x^2(t) \rangle$ . This quantity can be calculated from the associated Fokker-Planck equation to (11). This equation can be derived by noting first that (13) is actually an equation for the entire probability density P(x,t), rather than just for the average  $\langle x(t) \rangle$ , because according to van Kampen's lemma, the following identification is valid (van Kampen, 1981),

$$\langle x(r) \rangle = P(x,t)$$
 (21)

To see the consequences of this relation in more detail, note that for all noise realizations the stochastic equations for x(t) describe a flow of trajectories in x-space with density Q(x,t) which obeys the continuity equation

$$\frac{\partial Q(x,t)}{\partial t} = -\frac{\partial}{\partial x} [xQ(x,t)]$$
(22)

If x now runs over all its realizations with their appropriate probabilities and we use (11), then (22) can be rewritten in the general form of a linear stochastic differential equation for Q(x,t),

$$\frac{\partial Q(x,t)}{\partial t} = \left\{ -Q^{(0)} \left( 1 + x \frac{\partial}{\partial x} \right) - \alpha^2 Q^{(1)}(t) \left( 1 + x \frac{\partial}{\partial x} \right) \right\} Q(x,t) \quad (23)$$

Furthermore, if we identify the operators

$$\hat{Q}^{(0)} \equiv \omega_c \left( 1 + x \frac{\partial}{\partial x} \right), \tag{24}$$

$$\hat{Q}^{(1)} \equiv -\zeta(t) \left( 1 + x \frac{\partial}{\partial x} \right), \tag{25}$$

from van Kampen's lemma (21), we can rewrite (23) as a time evolution equation for the probability density P(x,t)

$$\frac{\partial P(x,t)}{\partial t} = \{\hat{Q}^{(0)} + \alpha^2 \int_0^\infty d\tau \left\langle \hat{Q}^{(1)}(t) e^{\tau \hat{Q}^{(0)}} \hat{Q}^{(1)}(t-\tau) \right\rangle$$
$$e^{-\tau \hat{Q}^{(0)}} \} P(x,t)$$
(26)

The right hand side (r.h.s) is evaluated by calculating the action of the different operators over P(x,t), either by Fourier transforming or by developing the operators  $e^{\pm \tau Q^{(0)}}$  in a Taylor series around the point (x,t). In this way one arrives at the following nonlinear Fokker-Planck equation (NLFP) or the time evolution of P(x,t), which in its canonical form reads

$$\frac{\partial}{\partial t}P(x,t) = -\frac{\partial}{\partial x}\{[-\omega_c + \alpha^2 c_0(t)]\}P(x,t) + \frac{1}{2}\frac{\partial^2}{\partial x^2}[2\alpha^2 c_0(t)x^2P(x,t)]$$
(27)

### 4.1 Second moment

The time evolution equation for the second moment  $\langle x^2 \rangle$  is obtained by multiplying Eq. (27) by  $x^2$  an integrating over all the values of *x*,

$$\frac{d}{dt}\langle x^2 \rangle = -2[\omega_c - 2\alpha^2 c_0(t)]\langle x^2 \rangle \qquad (28)$$

where  $c_0(t)$  is given by Eq. (15). For given  $x_0^2$  its solution reads

$$\langle x^2(t) \rangle = x_0^2 \exp\{-2[\omega_c - 2\alpha^2 c_0(t)]t\},$$
 (29)

and is valid for an arbitrary (Gaussian) noise with a well defined auto-correlation  $c_0(t)$ , For multiplicative non-inertial fluctuations the mean square displacement (*MS D*) is obtained from Eqs. (16) and (29),

$$\frac{1}{x_0^2} \langle [\Delta x(t)]^2 \rangle = \exp\{-2[\omega_c - 2\alpha^2 c_0(t)]t\} [\exp(2\alpha^2 c_0(t)t) - 1] \quad (30)$$

Note that the explicit time dependence of the MSD as given by (30), depends on the explicit form of  $c_0(t)$ ,

www.rmiq.org

which in terms depends on the type of noise. Thus, the type of diffusion of the aerosol confining particles depends on the nature of the external noise.

## Conclusions

In this work we have developed a stochastic model for the position-position correlation function and its power fluctuation spectrum of an optically trapped aerosol when the frequency of the confining laser fluctuates. To elaborate on these results the following comments may be useful. It should be stressed that in this paper we have reported model calculation results of some properties which may be measurable. Our calculations were based on the multiplicative stochastic linear equation (11), valid when inertial effects on the stochastic motion of the particles are neglected. As mentioned in the Introduction, underdamped motions of aerosol particles have been observed (Jokutty et al., 2005). This is indeed the usual situation for optical trapping for beads and particles, since they are generally in a low Reynolds number regime and strongly damped. More specifically, we show that this situation may be generalized by considering multiplicative parametric noise in the frequency of the trapping laser, instead of the usual thermal additive noise. This noise in the signal may be produced with a noise generator, or it can be produced by the action of external agents. These fluctuations were described by a multiplicative Langevin equation with white noise. Explicit analytic expressions were obtained for the corresponding additive  $(\chi_w^a(t))$  and multiplicative  $(\chi_w^m(t))$  position correlations functions for external white noise, given by (7) and (18), respectively. We showed that the effect produced by externally imposed multiplicative fluctuations on the frequency of the confining laser may produce a large effect (~one order of magnitude) on these correlations and on the corresponding fluctuations spectra  $S_w^a(\omega)$ and  $S_w^m(\omega)$ , Eqs. (8) and (19), respectively. This effect could be also manifested in transport properties like the diffusion coefficient for different types of noise, and will determine the nature of the diffusion, which could be anomalous. This issue will be discussed elsewhere. The difference between  $\chi_w^a(t)$  and  $\chi_w^m(t)$ were quantified and our results show that the inclusion of multiplicative external noise may have significant effects on  $\chi_w^m(t)$ . This quantity is always larger in intensity than  $\chi^a_w(t)$  and decays more slowly. This behavior shows that multiplicative noise may

produce a significant and perhaps measurable effect on the position correlation function. The curves in Fig. 3 show the Lorentzian character of the power spectra of position fluctuations  $S_w^a(\omega)$  and  $S_w^m(\omega)$ . The multiplicative spectrum is ~ 100% larger than the additive one in the interval between  $\omega \approx \pm 372$ Hz, but below these values there is a crossover and  $S_w^m(\omega)$  decreases in ~ (30-40)%. Both spectra show a plateau of the same form in similar frequency ranges. The multiplicative plateau is of the same form as for the additive case, but with a smaller effective trapping stiffness  $\kappa - \alpha^2 \gamma_0 \beta$ . In this sense our results reduce to those reported in the literature (Burnham, 2009; Yao et al., 2009) and validate the model. It should be emphasized that in this work we have proposed a stochastic model to describe multiplicative noise effects on the fluctuations of aerosol particles which is based on a simple model calculation. The eventual verification of these predictions and their validity can only come from experiments, but this is, to our knowledge, an open question that remains to be assessed.

### References

- Allersma, M.W., Gittes, F., de Castro, M.J., Stewart, R.J., Schmidt, C.F. (1998). Two-dimensional tracking of ncd motility by back focal plane interferometry. *Biophysical Journal* 74, 1074-1085. doi.org/10.1016/S0006-3495(98)74031-7
- Arecci, F.T. and Politi, A. (1979). Generalized Fokker-Planck equation for a nonlinear Brownian motion with fluctuations in the control parameter. *Optics Communications 29*, 361-363. doi.org/10.1016/0030-4018(79)90118-4
- Berg-Sorensen, K. and Flyvbjerg, H. (2004). Power spectrum analysis for optical tweezers. II: Laser wavelength dependence of parasitic filtering, and how to achieve high bandwidth. *Review of Scientific Instruments* 75, 594-612. doi.org/10.1063/1.2204589
- Burnharm, D. R. (2009). Microscopic applications of holographic beam shaping and studies of optically trapped aerosols. Ph. D. Dissertation, University of St. Andrews, U.K.
- Burnham D.R. and McGloin D. (2009). Radius measurements of optically trapped aerosols

through Brownian motion. *New Journal* of *Physics 11*, 063022. doi:10.1088/1367-2630/11/6/063022

- Burnham, D.R., Reece P.J. and McGloin, D. (2010). Parameter exploration of optically trapped liquid aerosols. *Physical Review E* 82, 051123. doi.org/10.1103/PhysRevE.82.051123
- Carberry, D.M., Baker, M.A.B., Wang, G.M., Sevick, E. M. and Evans, D.J. (2007). An optical trap experiment to demonstrate fluctuation theorems in viscoelastic media. *Journal of Optics A 9*, S204- S214. doi.org/10.1088/1464-4258/9/8/S13
- Chowdhury, A., Ackerson, B.J. and Clark, N.A. (1985). Laser-induced freezing. *Physical Review Letters* 55, 833-836. doi.org/10.1103/PhysRevLett.55.833
- De Kepper, P. and Horsthemke W. (1978). Etude d'un réaction chimique périodique. Influence de la lumière et transitions induites par un bruit externe. *Comptes Rendus Hebdomadaires des Séances de l'Académie des Sciences C 287*, 251-254.
- Denk, W. and Webb, W.W. (1990). Optical measurement of picometer displacements of transparent microscopic objects. *Applied Optics* 29, 2382-2391. doi.org/10.1364/AO.29.0 02382
- Fox, R.F. (1972). Contributions to the theory of multiplicative stochastic processes. *Journal of Mathematical Physics* 13, 1196-1207. doi.org/10.1063/1.1666123
- Garza.García; L.D. and Lapizco-Encinas, B. H. (2010). State of the art on protein manipulation employing dielectrophoresis. *Revista Mexicana de Ingeniería Química 9*, 125-137.
- Gishlan, L.P., Switz, N.A., Webb, W.W. (1994). Measurement of small forces using an optical trap. *Review of Scientific Instruments* 65, 2762-2768. doi.org/10.1063/1.1144613
- Gollub, J.P. and Steinman J.F. (1980). External noise and the onset of turbulent convection. *Physical Review Letters* 45, 551-554. doi.org/10.1103/PhysRevLett.45.551
- Grabert, H., Hänggi, P. and Talkner, P.J. (1980). Microdynamics and nonlinear stochastic processes of gross variables.

Journal of Statistical Physics 22, 537-552. doi.org/10.1007/BF01011337

- Hänggi, P. (1978). Correlation functions and master equations of generalized (non-Markovian) Langevin equations. *Zeitschrift für Physik B* 32, 407-416.
- Hertlein, C., Helden, L., Gambassi, A.S., Dietrich, S. and Bechinger, C. (2008). Direct measurement of critical Casimir forces. *Nature* 451, 172-175. doi.org/10.1038/nature06443
- Hernández-Machado, A., Sancho, J. M., San Miguel, M. and Pesquera L. (1983). Joint probability distribution of nonmarkovian stochastic differential equations. *Zeitschrift für Physik B* 52, 335-343. doi.org/10.1007/BF01307403
- Horsthemke W. (1981). Stochastic Nonlinear Systems in Physics, Chemistry and Biology. Synergetics Series. Springer-Verlag, Berlin.
- Hrubý, J., Duška, M., Němec, T., Kolovratník, M. (2018). Nucleation rates of droplets in supersaturated steam and water vapourcarrier gas mixtures between 200 and 450 K. Proceedings of the Institution of Mechanical Engineers, Part A: Journal of Power and Energy 232, 536-549. doi.org/10.1177/0957650918770939
- Jokutty, J., Mathur, V., Venkatarraman, V. and Natarajan, V. (2005). Direct measurement of the oscillation frequency in an opticaltweezers trap by parametric excitation. *Physical Review Letters* 95, 193902. doi.org/10.1103/PhysRevLett.95.193902
- Knettig, P. and Beeckmans, J.M. (1974). Capture of monodispersed aerosol particles in a fixed and in a fluidized bed. *The Canadian Journal of Chemical Engineering* 52, 703-706. doi.org/10.1002/cjce.5450520602
- Kubo, R. (1966). The fluctuation-dissipation theorem. *Reports on Progress in Physics* 29, 255-284. doi.org/10.1088/0034-4885/29/1/306
- Landau, L.D. and Lifshitz E.M. (1970). *Statistical Physics*. Addison-Wesley, Reading, USA.
- Lang, M.J., Asbury, C.L., Shaevitz, J.W. and Block, S.M. (2002). An automated twodimensional optical force clamp for single molecule studies. *Biophysical Journal 83*, 491-501. doi.org/10.1016/S0006-3495(02)75185-0

- Lee, S. and Grier D.G. (2005). One-dimensional optical thermal ratchets. *Journal of Physics 17*, S3685-S3695. doi.org/10.1088/0953-8984/17/47/003
- Lu, S., Pugh, R. and Forssberg, E. (2005). *Interfacial Separation of Particles*, Volume 20, 1st Edition, Elsevier Science, Amsterdam, The Netherlands.
- Neuman K. C. and Block S. M. (2004). Optical trapping. *Review of Scientific Instruments* 75, 2787-2809. doi.org/10.1063/1.1785844
- Pesce, G., Sasso, A. and Fusco, S. (2005). Viscosity measurements on micron-size scale using optical tweezers. *Review* of Scientific Instruments 76, 115105. doi.org/10.1063/1.2133997
- Rodríguez, R.F., Olivares, J.A. and Díaz-Uribe, R. (1997). Parametric noise induced birrefringence in nematic liquid crystals. *Revista Mexicana de Física 43*, 93-107.
- Rodríguez, R.F., Salinas- Rodríguez, E., Hayashi, A, Soria, A., Zamora, J.M. (2001). Simple stochastic model of spontaneous imbibition in Hele-Shaw cells. *AIChE Journal* 47, 1721-1730. doi.org/10.1002/aic.690470804
- Rodríguez, R.F., Salinas-Rodríguez, Е., Maldonado, A., Hernández-Zapata Е., Cocho, G. (2011).Criticality and biological supradiffusion in membranes: effect of transverse multiplicative The fluctuations. Physica A 390, 1198-1208. doi.org/10.1016/j.physa.2010.11.022
- Sancho, J.M. and San Miguel M. (1980a). External non-white noise and nonequilibrium phase

transitions. *Zeitschrift für Physik B 36*, 357-364. doi.org/10.1007/BF01322159

- Sancho, J. M. and San Miguel M. (1980b). Fokker-Planck approximation for N-dimensional nonmarkovian Langevin equations. *Physics Letters A* 76, 97-100. doi.org/10.1016/0375-9601(80)90579-4
- Slama Lighty J., Veranth J.M. and Sarofim A.F. (2000). Combustion aerosols: Factors governing their size and composition and implications to human health. *Journal of the Air & Waste Management Association 50*, 1565-1618. doi.org/10.1080/10473289.2000.104641 97
- Tien, C. and Ramaro, B.V., Editors. (2007). *Granular Filtration of Aerosols and Hydrosols*. Elsevier Science & Technology Books. 2nd ed. Syracuse, New York.
- van Kampen, N. G. (1981). *Stochastic Processes in Physics and Chemistry*. 3rd edition. North-Holland, Amsterdam.
- van Kampen, N. G. (1981a). *Stochastic Processes in Physics and Chemistry*. 3rd edition. Chapter XVI Sections 2 and 3. North-Holland, Amsterdam.
- Yao, A.M., Keen, S.A.J., Burnham, D.R., Leach, J., Di Leonardo, R., McGloin, D. and Padgett, M.J. (2009). Underdamped modes in a hydrodynamically coupled microparticle system. *New Journal of Physics 11*, 053007. doi.org/10.1088/1367-2630/11/5/053007