

Statistical description of the collective motion of nanoparticles

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A synergetic model allowing the description of a Brownian motion of active nanoparticles within the framework of the Lorenz three-parameter system is developed. The fluctuation's influence on the transition between different motion regimes is investigated. A diagram of possible motion modes of an active nanoparticle group and the corresponding stationary values of velocity are analyzed.

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I. INTRODUCTION

Nowadays the development of nanotechnology provides an opportunity to create a variety of technical devices. In this case a special role is assigned to biotechnology, for which various micro-organisms (bacteria) are used along with proteins, enzymes, etc. It is known that micro-organisms are characterized by high sensitivity and are able to convert various kinds of energy: mechanical, magnetic, luminous, and chemical. Thus, by understanding the individual and collective behaviors of micro-organisms we can find an opportunity to develop a special technique for the logical, computational, and measuring microdevices. Such devices may be based on physical and chemical effects that govern the processes of the micro-organisms' self-organization.

It is generally known [1–4] that under certain conditions moving chaotic bacteria can form various constant as well as time-varying structures [5,6]. It is also obvious [6–9] that such systems can maintain their order only when the processes that control their behavior are nonlinear. Thus the problem of modeling the processes occurring in nonlinear biological systems (in particular biological motion) has been a topic of interest in recent years [10,11]. However, we must consider not only the nonlinear effects of the environment, but also the changes in environmental parameters due to the activity of micro-organisms.

The distinguishing feature of many bacteria (e.g., *E. Coli*, *Salmonella typhimurium*, and *Bacillus*) is the presence of specific flagella, which have a surface spreading. The bacteria are actively moving by changing periods of directional motion for the periods of so-called tumbling. Under directional motion flagella rotate and are interwoven in the spiral bundle behind the cell body. This creates a periodic force, pushing the particle forward [Fig. 1(a)]. The flagellar bundle is unwoven in the state of tumbling when a particle loses its orientation and makes abrupt chaotic rotation, which resembles trembling and tumbling [Fig. 1(b)]; however, usually there is an alternation of these movements [Fig. 1(c)]. The time of directed motion is 1–4 sec and that of tumbling is approximately 0.1 sec [12,13]. However, tumbling can be suppressed if the medium is inhomogeneous, namely, it contains the so-called attractants. This phenomenon is called chemotaxis, whereby the particles are moving in the direction of increasing concentration of attractants. In addition to modeling different devices, one

can use the phenomena of magnetotaxis, phototaxis, and the gyrotaxis. In the latter case the directional movement of particles is caused by the torque compensation.

It appears that these motion regimes are not unique to the micro-organisms, but also occur for more highly organized birds and fish. In a comprehensive survey [14] about swarm dynamics a classification of the collective motion regimes of swarms is given. It is pointed out that there are three typical modes of motion: directed motion, when the swarm moves as whole, forming a tight group; rotational motion around the empty center, forming the shape of a doughnut; and amoebalike motion, when two previous modes alternate. However, it turns out that different kinds of rotational motion are realized for particles of different levels of organization. For low-level organization (e.g., gold nanoparticles and some bacteria) the rotational motion is reduced only to the individual tumbling [15]; more highly organized micro-organisms (e.g., some colonies of bacteria, phytoplankton, and zooplankton) realize a highly organized collective rotational motion [12–14].

We generalize these types of movement to the cases of directed motion, tumbling, and run-and-tumble regime [16]. The described transition from random fluctuations to ordered movements and spatial structures is not unique to biological systems but is found in some physical ones as well [15,17]. For example, in Refs. [15,17] the so-called hot Brownian motion was investigated, which differs from ordinary Brownian motion in that the laser-heated gold nanoparticles [15] have a stock of internal energy that is converted into mechanical energy. As a result, one may observe an analogy between motion regimes of bacteria and gold nanoparticles. The investigation of analogies among living and nonliving systems has a great practical interest.

In our paper we consider a stochastic dynamics of the active nanoparticle motion within the Brownian movement model [15,18]. The term active is easily understood by using an analogy to biological particles (bacteria) that do not only carry out disordered movement, but also have a stock of internal energy, which further converts into mechanical energy. In addition, the influence of noise is of particular interest for us since noise is responsible for many interesting effects and in particular noise may lead to transitions between deterministic attractors [19,20]. By extending these studies we will investigate the stochastic transitions between different motion regimes of active nanoparticles, using as noise the Ornstein-Uhlenbeck process [18].

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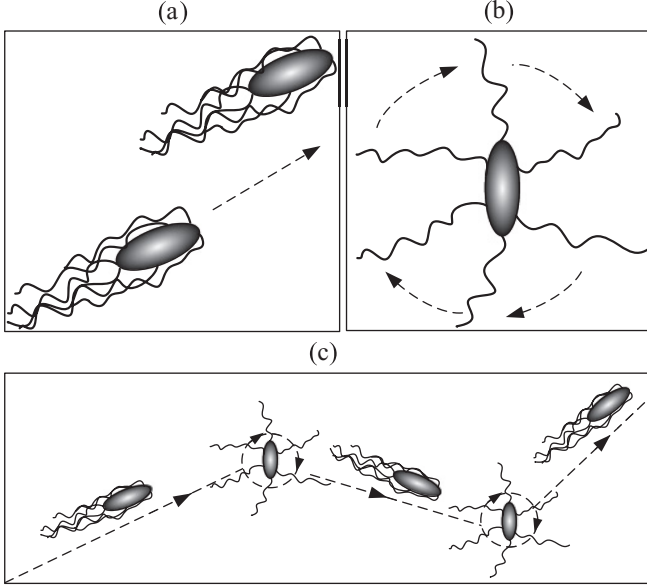


FIG. 1. Bacteria motion modes: (a) directed motion, (b) tumbling, and (c) alternation of previous regimes.

The paper is organized as follows. Section II describes the system of differential equations that form the basis of our consideration. Section III discusses the statistical analysis of the obtained equation of motion, from which we arrive at the Fokker-Planck equation, whose solution is the distribution function of nanoparticles velocity. The stationary solution of the Fokker-Planck equation and the corresponding dependencies are discussed in Sec. IV.

II. SYNERGETIC REPRESENTATION OF THE ACTIVE NANOPARTICLES' COLLECTIVE BEHAVIOR

Using the example of condensed matter [21–23], the representation of self-organizing system reduces to the self-coordinated description of the time dependencies of the order parameter, the conjugate field, and the control parameter. By using the order parameter, which distinguishes motion regimes, we will take the average velocity v of the active nanoparticles movement. By average velocity we mean the velocity of directed movement. Thus, in the case of tumbling motion, a particle is rotating and the mentioned average velocity is zero. We also need to include the interaction between the particles themselves or between the particles and the environment via some kind of field or chemical reagent [the concentration of this reagent may be represented by a field $H(r,t)$]. Thus the conjugate field reduces to the long-range force $f_h \equiv \nabla H(r,t)$. Further we will consider one-dimensional case since for the directed motion the nanoparticles always have a priority direction (for example, drug delivery by a vessel). According to Ref. [14], the active particles have another internal degree of freedom θ , which describes different activities and responses to the field and takes on only two values: ± 1 . In our consideration we will use another internal parameter ε , which describes internal energy and takes different values. However, this parameter is imposed on restrictions that are defined by the environment.

For example, for the hot Brownian motion the temperature T_e of the liquid, where heated nanoparticles are located, sets the minimum internal energy of the particle ε_e . As a result, the problem is reduced to the expression [21] of the rates of change \dot{v} , \dot{f}_h , and $\dot{\varepsilon}$ through their values v , f_h , and ε (further for simplicity the one-dimensional case is considered). Assuming that the order parameter $v(t)$ subordinates the behavior of force $f_h(t)$ and internal parameter $\varepsilon(t)$, we obtain an expression for the average acceleration in the linear form

$$\dot{v} = -\frac{v}{t_v} + a_v f_h. \quad (1)$$

The first term on the right-hand side of Eq. (1) displays a relaxation of the average velocity to zero value during the time t_v . The second term describes the linear reaction of acceleration \dot{v} on increasing force (a_v is a positive constant). The equation for the conjugate field is given by the expression

$$\dot{f}_h = -\frac{f_h}{t_f} + a_f v \varepsilon, \quad (2)$$

where the first term on the right-hand side again has a relaxational nature with the characteristic time t_f and the second term represents a positive feedback of the average velocity and the internal state parameter with a rate of change of the conjugate field (a_f is a positive constant). This correlation causes an increase of the conjugate field, which is the reason for the system's self-organization.

The last equation of the system evolution describes a relaxation of the internal parameter ε playing the role of the control parameter

$$\dot{\varepsilon} = \frac{\varepsilon_e - \varepsilon}{t_\varepsilon} - a_\varepsilon v f_h + \zeta(t). \quad (3)$$

In contrast to the Eqs. (1) and (2), the first term on the right-hand side of Eq. (3) presents the relaxation of the parameter ε to the final value ε_e (with t_ε the corresponding relaxation time). According to Eq. (3), negative feedback (the constant $a_\varepsilon > 0$) of the long-range force and the average velocity with the speed of change of the internal state parameter results in the reduction of this parameter, according to the Le Chatelier principle. The last term reveals the stochastic behavior of the average velocity with stochastic source $\zeta(t)$, which is defined as the Ornstein-Uhlenbeck process

$$\begin{aligned} \langle \zeta(t) \rangle &= 0, \\ \langle \zeta(t) \zeta(t') \rangle &\equiv \frac{I}{\tau} \exp\left(-\frac{|t-t'|}{\tau}\right), \end{aligned} \quad (4)$$

where I is the noise strength, τ is the correlation time of the process $\zeta(t)$, and the angular brackets mean averaging.

According to Ref. [24], the system of the synergetic equations (1)–(3) represents the elementary field scheme, which shows the main effects of the self-organization. For the analysis of this system it is convenient to take dimensionless variables for time t , average velocity v , conjugate field f , internal state parameter ε , and strength I . The corresponding

scales are

$$t_s \equiv t_v, \quad v_s \equiv (a_f a_\varepsilon t_f t_\varepsilon)^{-1/2}, \quad f_{hs} \equiv (a_v^2 t_v^2 a_f a_\varepsilon t_f t_\varepsilon)^{-1/2}, \\ \varepsilon_s \equiv (a_v a_f t_v t_f)^{-1}, \quad I_s \equiv (a_v a_f t_v t_f t_\varepsilon)^{-2}.$$

Thus the behavior of the active nanoparticle's group is presented by the dimensionless system of nonlinear equations

$$\dot{v} = -v + f_h, \quad (5)$$

$$\frac{\dot{f}_h}{\sigma} = -f_h + v\varepsilon, \quad (6)$$

$$\frac{\dot{\varepsilon}}{\delta} = (\varepsilon_e - \varepsilon) - v f_h + \zeta(t), \quad (7)$$

where the relations $\sigma^{-1} \equiv t_f/t_v$ and $\delta^{-1} \equiv t_\varepsilon/t_v$ of characteristic times are entered.

Within the framework of the mentioned parametrization, the directed regime of motion is represented as a result of spontaneous velocity and force deviations if the internal state parameter exceeds a critical value. It is presented [25] by the appearance of the minimum of the effective potential, which corresponds to the stationary value v_0 of the velocity. Therefore we will be interested in the further evolution of v .

In the general case the system (5)–(7) has no analytical solution; therefore we use the following approximation:

$$t_v \gg t_\varepsilon, \quad t_v \approx t_f. \quad (8)$$

These conditions indicate that in the course of evolution the internal state parameter ε is coordinated by velocity and force variations. As a result, the parameter $\delta \gg 1$, which allows us to assume that $\dot{\varepsilon}/\delta \approx 0$ in Eq. (7). Consequently, instead of three differential equations we obtain a system of two equations

$$\dot{v} = -v + f_h, \quad (9)$$

$$\dot{f}_h = -\sigma f_h + \sigma v[\varepsilon_e - v f_h + \zeta(t)]. \quad (10)$$

To simplify the system it is necessary to express f_h via v . The equation for f_h is determined by time differentiation of Eq. (9). Substituting expressions for f_h and \dot{f}_h from Eq. (9) into Eq. (10), we get the canonical form of the motion equation for the nonlinear stochastic oscillator:

$$\ddot{v} + \gamma(v)\dot{v} = f(v) + g(v)\zeta(t), \quad (11)$$

where the friction function $\gamma(v)$, effective force $f(v)$, and noise intensity $g(v)$ are defined by the expressions

$$\gamma(v) = 1 + \sigma(1 + v^2), \\ f(v) = \sigma(\varepsilon_e - 1)v - v^3, \quad (12) \\ g(v) = v.$$

III. STATISTICAL ANALYSIS OF THE SYNERGETIC REPRESENTATION

Our task is to find a distribution function $P(v, \dot{v}, t)$ of the system in the phase space formed by the velocity v and acceleration \dot{v} depending on time t . For this purpose we represent the Euler equation (11) within the framework of the Hamilton formalism

$$\dot{v} = a, \quad \dot{a} = -\gamma(v)a + f(v) + g(v)\zeta(t). \quad (13)$$

As a result, the required probability density $P(v, a, t)$ is reduced to the distribution function $\rho(v, a, t)$ of the solutions of the system (13),

$$P(v, a, t) = \langle \rho(v, a, t) \rangle_\zeta. \quad (14)$$

Here $\langle \dots \rangle_\zeta$ stands for averaging over noise ζ . The continuity equation is constructed in a standard manner:

$$\left[\frac{\partial}{\partial t} + \hat{L}(v, a) \right] \rho(v, a, t) = -g(v)\zeta(t) \frac{\partial}{\partial a} \rho(v, a, t), \quad (15)$$

where the operator

$$\hat{L}(v, a) = -\gamma(v) \frac{\partial}{\partial a} a + a \frac{\partial}{\partial v} + f(v) \frac{\partial}{\partial a} \quad (16)$$

is introduced. According to the method of cumulant expansion in terms of Van Kampen [26,27], the equation for the averaging of function $\rho(v, a, t)$ takes the form of the integral-differential equation

$$\left[\frac{\partial}{\partial t} + \hat{L}(v, a) \right] \langle \rho(v, a, t) \rangle \\ = -\frac{\partial}{\partial a} g(v) \int_0^t \langle \zeta(t)\zeta(t') \rangle e^{-\hat{L}(t-t')} g(v) \frac{\partial}{\partial a} \langle \rho(v, a, t) \rangle dt'. \quad (17)$$

We now proceed to the initial distribution $P(v, a, t)$ by replacing the operator on the left-hand side of Eq. (17) with \hat{N} :

$$\left\{ \frac{\partial}{\partial t} + \hat{L}(v, a) \right\} P(v, a, t) = \hat{N}(v, a, t) P(v, a, t). \quad (18)$$

Since we consider the long time and smoothed behavior according to Ref. [27], Eq. (11) can be summarized as

$$\ddot{v}_i + \gamma(v_i)\dot{v}_i = f(v_i) + g(v_i)\zeta_i(t) \quad (19)$$

for a set of variables v_1, v_2, \dots, v_n and $\zeta_1, \zeta_2, \dots, \zeta_n$ (summation over repeated indices is implied). Analyzing Eq. (19) instead of the integro-differential operator \hat{N} , we arrive at the sum

$$\hat{N}(v, a, t) = \sum_{n=0}^{\infty} \hat{N}_n = g(v) \frac{\partial}{\partial a} \sum_{n=0}^{\infty} M_n \hat{L}_n(v, a). \quad (20)$$

The moments of the correlation function are defined as follows:

$$M_n(t) = \frac{1}{n!} \int_0^{\infty} t^n \langle \zeta(t)\zeta(0) \rangle dt. \quad (21)$$

The use of Eq. (4) gives the relations for the zeroth and first moments

$$M_0(t) = I, \quad M_1(t) = I\tau. \quad (22)$$

The operators \hat{L}_n are defined by the rule [27]

$$\hat{L}_n = [\hat{L}_{n-1}, \hat{L}], \quad \hat{L}_0 = g(v) \frac{\partial}{\partial a}, \quad (23)$$

where the square brackets mean the commutator $[A, B] = AB - BA$.

Expressions for \hat{N}_0 and \hat{N}_1 allow us to define Eq. (18) as a Kramers equation, whose solution is a function $P(v, a, t)$. Since the integral

$$\mathcal{P}(v, t) = \int_a P(v, a, t) da \quad (24)$$

is of our practical interest [more than $P(v, a, t)$], it is necessary to consider the moments of the initial distribution

$$\mathcal{P}_n(v, t) = \int_a a^n P(v, a, t) da. \quad (25)$$

In this case a zeroth moment will be the required one: $\mathcal{P}_0(v, t) = \mathcal{P}(v, t)$. After multiplying the Kramers equation by a^n and integrating over acceleration a , one can obtain a recurrent relation [27,28], which implies a hierarchical system of equations

$$\begin{aligned} \frac{\partial \mathcal{P}}{\partial t} &= -\frac{\partial \mathcal{P}_1}{\partial v}, \\ \gamma \mathcal{P}_1 &= f \mathcal{P} - \frac{\partial \mathcal{P}_2}{\partial v} - M_1 \left[g \frac{\partial g}{\partial v} \mathcal{P} + g^2 \frac{\partial \mathcal{P}}{\partial v} \right], \\ \gamma \mathcal{P}_2 &= g^2 (M_0 - M_1 \gamma) \mathcal{P}. \end{aligned} \quad (26)$$

For brevity, the dependence of the velocity v is omitted. By solving the cyclic system of equations (26), we arrive at the Fokker-Planck [29] equation

$$\frac{\partial \mathcal{P}}{\partial t} = -\frac{\partial}{\partial v} (D_1 \mathcal{P}) + \frac{\partial^2}{\partial v^2} (D_2 \mathcal{P}), \quad (27)$$

where

$$D_1 = \frac{1}{\gamma} \left[f - M_0 \frac{g^2}{\gamma^2} \frac{\partial \gamma}{\partial v} + M_1 g \frac{\partial g}{\partial v} \right], \quad (28)$$

$$D_2 = M_0 \frac{g^2}{\gamma^2} \quad (29)$$

are the drift and diffusion coefficients, respectively.

IV. STATIONARY SOLUTION

Equation (27) may be presented as a continuity equation for the probability distribution $\mathcal{P}(v, t)$,

$$\frac{\partial \mathcal{P}}{\partial t} + \frac{\partial S}{\partial v} = 0, \quad (30)$$

where $S(v, t)$ is interpreted as the probability current

$$S = D_1 \mathcal{P} - \frac{\partial}{\partial v} (D_2 \mathcal{P}). \quad (31)$$

In a stationary case the probability distribution $\mathcal{P}(v, t)$ does not depend on time and the probability current (31) takes a constant value ($S = \text{const}$). As a result we get a stationary distribution

$$\begin{aligned} \mathcal{P}(v) &= \frac{1}{D_2(v)} \exp \int_0^v \frac{D_1(v')}{D_2(v')} dv' \\ &\times \left[N - S \int_0^v \exp \left(- \int_0^{v'} \frac{D_1(v'')}{D_2(v'')} dv'' \right) dv' \right], \end{aligned} \quad (32)$$

where N is a normalization constant

$$\begin{aligned} N &= \left(\int_0^\infty \frac{dv}{D_2(v)} \exp \int_0^v \frac{D_1(v')}{D_2(v')} dv' \right)^{-1} \\ &\times \left\{ 1 + S \int_0^\infty \left[\exp \left(\int_0^v \frac{D_1(v')}{D_2(v')} dv' \right) \right. \right. \\ &\left. \left. \times \int_0^v \exp \left(- \int_0^{v'} \frac{D_1(v'')}{D_2(v'')} dv'' \right) dv' \right] \frac{dv}{D_2(v)} \right\}. \end{aligned} \quad (33)$$

If we choose a constant S as the origin of the probability current ($S = 0$), the extremum condition for the distribution (32) reduces to the expression

$$D_1(v) - \frac{\partial}{\partial v} D_2(v) = 0. \quad (34)$$

Substituting the corresponding dependencies in Eq. (34), we obtain a cubic equation

$$\begin{aligned} x^3 - \left(\alpha - \frac{2}{\sigma} \right) x^2 - \left(\frac{2\alpha}{\sigma} - \frac{1}{\sigma^2} \right) x \\ + 2I \left(1 + \frac{1}{\sigma} \right) - \frac{\alpha}{\sigma^2} = 0, \end{aligned} \quad (35)$$

where $x \equiv 1 + v^2$ and $\alpha \equiv \varepsilon_e + I\tau\sigma$. According to Eq. (35) the distribution mentioned above realizes a maximum value at the point $v = 0$ if the internal state parameter does not exceed the critical value

$$\varepsilon_{ec} = 1 - I\tau\sigma + \frac{2I\sigma}{1 + \sigma}. \quad (36)$$

Since, according to the approximation (8), t_v and t_f are approximately equal, the parameter $\sigma = t_v/t_f$ can be equated to one. Therefore, to simplify the calculations in all following expressions, we assume that $\sigma = 1$.

To determine the conditions under which a directed motion is possible, we find (after excluding the trivial solution $v = 0$) a solution to Eq. (35),

$$v_\pm^2 = \frac{1}{2} [\alpha - 5 \pm \sqrt{(\alpha - 1)(\alpha + 7)}], \quad \sigma = 1. \quad (37)$$

To find the conditions for the existence of the solution (37), it is necessary to equate to zero the discriminant of Eq. (35). As a result, we arrive at the equation

$$\varepsilon_{e1} = 3\sqrt[3]{I} - I\tau - 1. \quad (38)$$

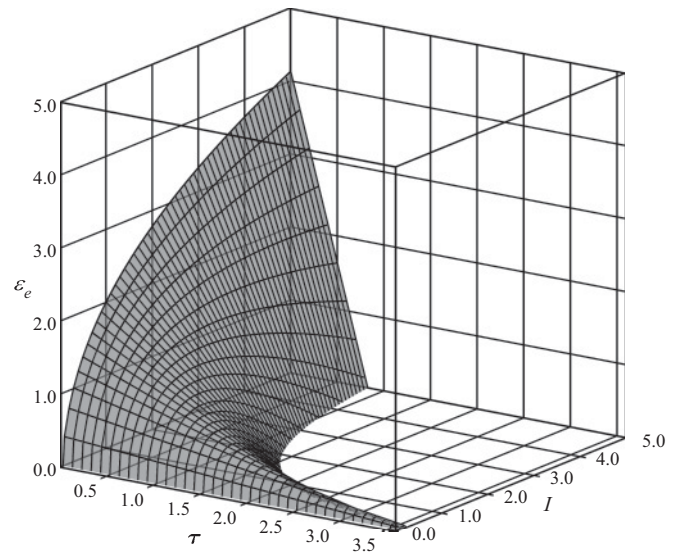


FIG. 2. The surface corresponds to the characteristic value ε_{e1} , which is given by the equation (38). The domain of the existence of only trivial solution ($v = 0$) is located under the surface. Above this surface a nonzero solution appears.

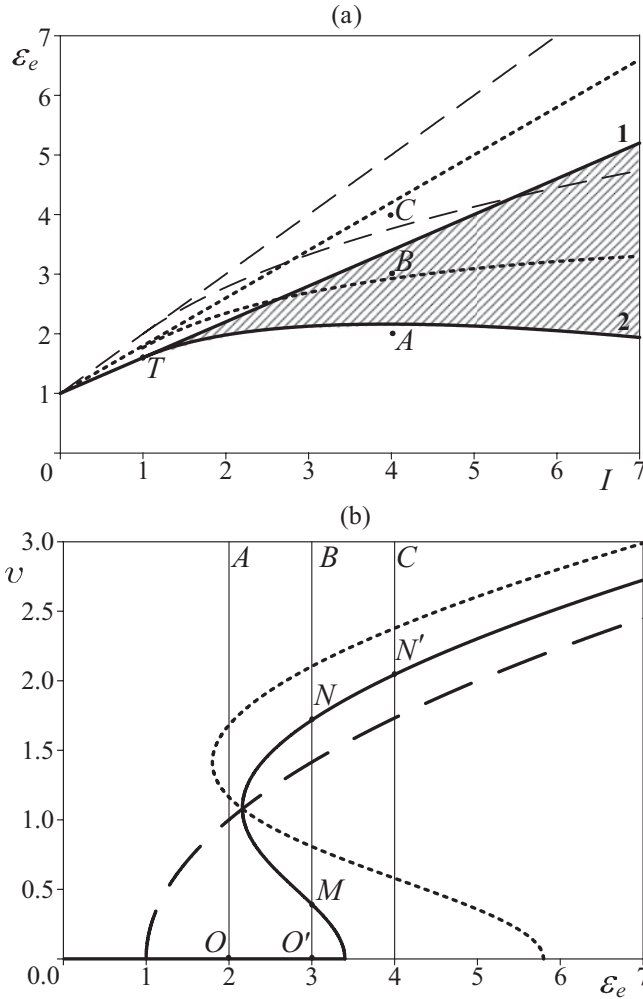


FIG. 3. (a) Phase diagram of the system for $\tau = 0.4$. Line 1 is given by Eq. (36) and curve 2 is given by Eq. (38). The dashed lines correspond to $\tau = 0.0$ and dotted lines correspond to $\tau = 0.2$. (b) Dependence of the stationary velocity for $I = 4$ and $\tau = 0.4$. The dashed curve corresponds to $I = 0.0$ and the dotted curve corresponds to $I = 8$. Rays A, B, and C meet the appropriate points of the phase diagram.

The corresponding dependence is shown in Fig. 2. If the internal state parameter ε_e exceeds the value ε_{e1} , then Eq. (35) has three real solutions; otherwise it has only one solution. This is easily seen by comparing the corresponding points of the phase diagram [Fig. 3(a)] with the velocity dependence [Fig. 3(b)].

The diagram shows that if the internal state parameter ε_e takes values less than ε_{ec} the zero solution is realized. If

$\varepsilon_e > \varepsilon_{e1}$ we have a nonzero solution. A shaded region between line 1 and curve 2 [Fig. 3(a)] corresponds to the coexistence of zero and nonzero velocity (i.e., the so-called stick-slip motion regime). It is easy to see from Fig. 3(b) that for different domains of the phase diagram a different number of points (values of the stationary velocity) is realized. For example, ray A meets on the dependence of the stationary velocity only one point O, which corresponds to the tumbling mode ($v = 0$). For another ray B there are three points O' , M, and N. Point O' has the same meaning $v = 0$, point N corresponds to the directed mode $v \neq 0$, and point M is an intermediate point, which can be considered a minimum, separating the maxima O' and N of the corresponding distribution function. This situation describes the coexistence of two phases (tumbling and directed) or the so-called stick-slip regime. The last ray C is characterized by the presence of only one point N' , which has the same meaning as N, but unlike the previous case for these parameters only directed movement is realized. The coordinates of the critical point T (indicated in Fig. 3(a)),

$$I_T = 1, \quad \varepsilon_{eT} = 2 - \tau, \quad (39)$$

determine the connection point of three diagram regions.

V. CONCLUSION

For many experiments in nanotechnology it is necessary to understand the motion features of nanoparticles for exact control of the corresponding technique (for example, optical tweezers). In our paper the simplest self-consistent scheme, which allows one to describe the motion of nanoparticles in liquid, was considered. Taking as a basis a synergistic Lorenz system and taking into account the fluctuations of the control parameter in the form of the Ornstein-Uhlenbeck process, we were able to describe the transition between three types of motion of the nanoparticles. In addition, the parameters defining the behavior of the system were determined and values of the stationary velocity were analyzed.

Note that the results described above do not claim to be a general theory of synergy and cannot replace the study of other real systems. However, these results may provide additional information and different ideas in the investigation of both living and nonliving systems. Alternative theory (more advanced with respect to accounting for interactions between particles themselves and between particles and the environment) will monitor and control the movement of nanoparticles in the liquid. The application of this theory is quite promising in the field of medical research, where nanoparticles are used as markers or for drug delivery.

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