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ВРАЩАТЕЛЬНЫЙ РЭТЧЕТ, УПРАВЛЯЕМЫЙ ДИХОТОМНЫМ ИЗМЕНЕНИЕМ ОРИЕНТАЦИЙ ПРИЛОЖЕННОГО ПОЛЯ

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Аннотация. Рассмотрено однонаправленное вращение полярного ротора (вращательный рэтчет) в потенциале заторможенного вращения, управляемое дихотомными флуктуациями ориентаций электрического поля. Проведен анализ симметрии, который показал отсутствие рэтчет-эффекта при нечетном количестве ям потенциала заторможенного вращения, а также при четном количестве ям потенциала заторможенного вращения, когда средний угол флуктуирующих ориентаций электрического поля совпадает с осями симметрии потенциала заторможенного вращения. Получены аналитические выражения для средней скорости вращения ротора в двухъямном потенциале заторможенного вращения в низкотемпературном адиабатическом приближении, когда происходит прыжковое вращение и в каждом состоянии дихотомного процесса успевает установиться термодинамическое равновесие, а также в высокотемпературном приближении при произвольных частотах флуктуаций, когда тепловая энергия много больше барьера переориентаций потенциала заторможенного вращения и энергии взаимодействия дипольного ротора с электрическим полем. Показано, что в обоих случаях максимальная скорость вращения достигается при больших электрических полях, флуктуирующих по знаку, и имеет колоколообразную форму в зависимости от амплитуды угловых флуктуаций, ширину которой и наличие плато можно регулировать средним углом флуктуаций.

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Зависимость скорости вращения от частоты флуктуаций также имеет колоколообразную форму, широкую для стохастических дихотомных флуктуаций и узкую для детерминистических дихотомных флуктуаций, с одинаковыми линейными низкочастотными асимптотиками.

Ключевые слова: вращательный рэтчет; заторможенное вращение; диффузионный транспорт; адиабатические броуновские моторы; дихотомный процесс; гармонические флуктуации.

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ROTATIONAL RATCHET CONTROLLED BY DICHOTOMOUS CHANGES IN APPLIED FIELD ORIENTATIONS

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Abstract. We explore the unidirectional rotation of a polar rotor (rotational ratchet) in a hindered-rotation potential (HRP), controlled by the dichotomous fluctuations of the orientation of the electric field. A symmetry analysis is carried out, which shows the absence of the ratchet effect for either an odd number of wells of the HRP or for an even number of wells if the average angle of the fluctuating orientations of the electric field coincides with any symmetry axes of the HRP. Analytical expressions are obtained for the average rotation velocity of the rotor in the double-well HRP in the low-temperature adiabatic approximation, when the hopping rotation occurs and thermodynamic equilibrium has time to be established in each state of the dichotomous process, and in the high-temperature approximation at arbitrary fluctuation frequencies, when the thermal energy is much greater than both the reorientation barrier of the HRP and the energy of the dipole rotor – electric field interaction. We showed that the maximum rotation velocity is achieved at large electric fields that fluctuate in sign, and the dependence of the rotation velocity on the amplitude of the angular fluctuations is bell-shaped, the width of which and the presence of the plateau can be tuned by the value of the average fluctuation angle. The dependence of the rotation velocity on the fluctuation frequency is also bell-shaped, wide for the stochastic fluctuations and narrow for the deterministic dichotomous ones, with the same linear low-frequency asymptotics.

Keywords: rotational ratchet; hindered rotation; diffusion transport; adiabatic Brownian motors; dichotomous process; harmonic fluctuations.

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Introduction

Ratchet systems (also called Brownian motors) include nanomachines that, under the influence of various nonequilibrium fluctuations, convert a chaotic Brownian motion into a translational, reciprocating or rotational motion [1–8]. In natural systems, ratchets provide contractile activity of tissues (muscular activity), cell motility (e. g., the motility of bacterial flagella), and intra- and intercellular transport of organelles and relatively large particles of substances (cell nutrition and waste disposal). Current knowledge shows that biomolecules that control a number of intracellular fundamental processes have one common feature, namely, they all respond to energy absorption or energy dissipation by changing their conformation and, hence, their physical shape in one plane. Cyclically repeated single-plane changes in their shapes cause a unidirectional, linear or rotational, motion in molecules and the associated processes, that are controlled by this motion; this is the basis of mechanical activity and work performed within cells [9].

Moving on to the consideration of rotational ratchets, note that the difference between molecular motors (or ratchets) and molecular rotors is that the former exhibit unidirectional rotation while the latter may randomly rotate in any direction. A molecular rotor is usually understood to mean a molecular system in which a molecule or part of a molecule (the rotor or rotator proper) rotates against another part of the molecule or against a macroscopic entity such as a surface or a solid (stator). Rotation of the rotor can occur freely (without a rotational potential) or by thermally activated jumps (hindered rotation) in the presence of an N-well rotational potential, where the value of N corresponds to the order of the rotation axis and is determined by the symmetry of the stator [10; 11]. A powerful tool for analysing the local environment of a surface molecular rotor is scanning tunneling microscopy (STM) [12]. For example, the *tert*-butyl groups on an Au(111) surface are characterised by a rotational potential with $N = 12$ [13], whereas a single acetylene (C_2H_2) rotor on an achiral PdGa(111) surface is in a potential with $N = 6$ [14]. Asymmetry in the potential energy landscape is commonly achieved by adsorbing chiral molecules on achiral surfaces or by the electric field generated by the STM tip at a certain position relative to the rotor (see reference [14] and supplementary materials for it).

Hindered rotational motion is also typical of chemisorbed polyatomic molecules or polyatomic groups tightly bound to a surface through one atom, whereas other atoms can have several equilibrium positions in the potential induced by the nearest substrate atoms. For example, for hydroxyl groups strongly bonded by an oxygen atom to a surface atom forming an oxide, the equilibrium positions of the hydrogen atom are determined by a radial distance of 1 Å relative to the oxygen atom, a polar angle of about 90°, and N values of the azimuthal angle corresponding to N symmetric wells of the hindered-rotation potential (HRP). The number N is one unit less than the valence of the surface atom forming the oxide and is equal to the number of symmetrically located oxygen atoms of the substrate closest to the hydroxyl group in question [15; 16].

First nanomachines in which unidirectional rotation occurred under the influence of light were constructed in the late 1990s by the scientific team of professor B. L. Feringa [17]. Further, the efficiency of those light-driven machines increased significantly [18]. In the experiment published in [14], a system was also considered in which the rotor was an acetylene molecule C_2H_2, and the stator was a cluster of three palladium atoms on the surface of a palladium – gallium crystal PdGa(111) with broken rotational symmetry. The unidirectional rotation of the acetylene molecule occurred due to the quantum tunneling effect in a scanning tunneling microscope. The rotational symmetry can also be broken with the fluctuating electric field itself, the orientation of which does not coincide with the symmetry axes of HRP [19]. This result was obtained using the low-temperature kinetic approach, in which the particle motion in a periodic N-well potential relief is reduced to a hopping overcoming the potential barriers. One more simplification of the consideration performed in reference [19] was the use of the second-order perturbation theory in a small alternating electric field $E(t) = E_0 \cos \omega t$ (E_0 and ω are the amplitude and frequency of the field change with time t, respectively). It turned out that, with the mentioned simplifications made, the ratchet effect occurs only at $N = 2$ and at the orientations of the electric field different from the orientations of the axes of the potential wells and barriers. The analysis of the ratchet driven by the adiabatic dichotomous arbitrary-amplitude fluctuations of the electric field $E(t) = \pm E$ characterised by an arbitrary angle φ_E to a symmetry axis of the HRP, showed the presence of the ratchet effect at even values $N > 2$ [20].

In contrast to the previous approaches to describing rotational ratchets, this article considers systems with dichotomous change in the orientations of the applied electric field themselves. The use of the low-temperature and high-temperature approximations leads to the analytical expressions for the average rotation velocity, the analysis of which provides the comprehensive information about the properties of the system not only at arbitrary, relative to the thermal energy, energies of the interaction of the rotor with the fluctuating electric field, but also at arbitrary fluctuation frequencies.

It should be noted that modern electron beam lithography [21] allows the formation of systems of closely spaced nanoelectrodes, the switching of electrical potentials between which ensures the creation of electric fields of a given intensity and direction. For example, in reference [22] the formation of structures consisting of two, four, six, and eight electrodes converging into a nanoscale region was reported. The dichotomy of the process is an essential simplification of the description and, in addition, gives a significant increase in the average rotation velocity at low frequencies of the potential switching.

N-well HRP in an external electric field

Let's consider a flat polar rotor in an N-well HRP, one of the wells of which is oriented along the axis x of a 2D coordinate system. Let the rotor be characterised by a dipole moment μ and be placed in an external electric field of a magnitude E, the orientation of which fluctuates in the plane of the rotor rotation and is specified by the values of two azimuthal angles φ_+ and φ_- (the case of dichotomous fluctuations). Then the potential energy of the rotor in the states «plus» and «minus» of the dichotomous process is represented as

$$U^{\pm}(\varphi) = -\left(\frac{\Delta U}{2}\right)\cos N\varphi - \mu E \cos(\varphi - \varphi_{\pm}), \tag{1}$$

where ΔU is the energy barrier of the HRP.

The potential energy (1) can be rearranged to the standard additive-multiplicative form of the potential energy of a pulsating ratchet:

$$U(x,\ t) = u(x) + \sigma(t)w(x), \tag{2}$$

in which the time function $\sigma(t)$ plays the role of a fluctuation variable with the zero mean $\langle\sigma(t)\rangle = 0$, while $u(x)$ and $w(x)$ are periodic functions of the particle coordinate x, that describe the average potential energy and its fluctuating component, respectively [23]. In the case of dichotomous fluctuations, the dependence $\sigma(t)$ can be represented as a switching, either random or deterministic, of its values between +1 and –1. For the rotational motion, the coordinate is the angular variable φ. Therefore, the potential energy $U(x,\ t)$ can be attributed to states «plus» or «minus» of the dichotomous process characterised by the variable $\sigma(t) = \pm 1$ and denoted as $U^{\pm}(\varphi)$. It is clear that for relations (1) and (2) to be equivalent, the functions $u(x)$ and $w(x)$ with $x = \varphi$ must be

$$u(\varphi) = -\left(\frac{\Delta U}{2}\right)\cos N\varphi - \mu E\cos\varphi_1\cos(\varphi - \varphi_0),$$
$$w(\varphi) = -\mu E\sin\varphi_1\sin(\varphi - \varphi_0), \tag{3}$$
$$\varphi_0 = \frac{\varphi_+ + \varphi_-}{2},\ \varphi_1 = \frac{\varphi_+ - \varphi_-}{2}.$$

Note that in the particular case $\varphi_1 = \frac{\pi}{2}$, the values of the angles φ_+ and φ_- differ by π. This corresponds to the sign fluctuations of the electric field oriented at an angle φ_E to the axis x. Then $\varphi_E = \varphi_+$, $\varphi_0 = \varphi_E - \frac{\pi}{2}$, $\sin(\varphi - \varphi_0) = \cos(\varphi - \varphi_E)$, and we obtain that the average potential energy $u(\varphi) = -\left(\frac{\Delta U}{2}\right)\cos N\varphi$ is independent of the applied electric field while its fluctuating part $w(\varphi) = -\mu E\cos(\varphi - \varphi_E)$ becomes equal to the interaction energy of the polar rotor with the external electric field of the orientation φ_E (the second term of equation (1)). The behaviour of rotor systems with the sign fluctuations of the applied electric field have been analysed in a number of works [13; 14]. This work differs from those previous studies by considering the general case when it is not the sign that fluctuates, but the electric-field orientations themselves, i. e. the parameter φ_1 is arbitrary.

Besides the energy parameters, ΔU and μE, the potential energy (1) is characterised by the number N of azimuthal potential wells and the parameters φ_0 and φ_1, that specify the average angle of the fluctuating orientations of the electric field and the fluctuation amplitude, respectively. We should exclude from the consideration the parameters' values that make the ratchet effect impossible. First of all, we exclude the values at which there are no fluctuations (i. e. $w(\varphi) = 0$), namely, $\mu E = 0$ and $\varphi_1 = 0$; here and hereafter we do not mention the trivial values, such as, for example, the value $\varphi_1 = \pi$ that corresponds to the difference $\varphi_+ - \varphi_- = 2\pi$.

Next, we do not need the values that are prohibited by the symmetry restrictions. Such prohibitions cover the potential energies that are described by shift-symmetric or symmetric periodic coordinate functions [2; 24–27]. For periodic potential energies $U^{\pm}(x + L) = U^{\pm}(x)$ (L is the period) associated with the two states of the dichotomous process, the shift-symmetric and symmetric functions are defined by the following identities:

$$U^{\pm}_{\text{sh}}\left(x + \frac{L}{2}\right) = -U^{\pm}_{\text{sh}}(x),\ U^{\pm}_{\text{s}}(x + x_{\text{s}}) = U^{\pm}_{\text{s}}(-x + x_{\text{s}}), \tag{4}$$

where x_{s} is the location of the symmetry axis. An additional symmetry prohibition exists in ratchet systems with the time dependence of the particle potential energy (2) specified by the function $\sigma(t)$ that belongs to the universal symmetry type [26]. For a symmetric dichotomous process (equal durations of the states «plus» and «minus»), this additional symmetry property corresponds to $\sigma(t) = \pm 1$ with $\langle\sigma(t)\rangle = 0$ and is specified by the following identity:

$$U^{\pm}(x + x_{\text{s}}) = U^{\mp}(-x + x_{\text{s}}). \tag{5}$$

In our model, for which the coordinate is the angular variable φ, the functions under consideration have the natural period 2π, $L = 2\pi$. Since $U^{\pm}(\varphi + \pi) = -U^{\pm}(\varphi)$ for odd N, the first of the properties (4) leads to the absence of the ratchet effect when the number of wells of the HRP is odd.

The potential energy (1) will satisfy the second identity in the pair (4) if both $u(\varphi)$ and $w(\varphi)$ are symmetric with the same symmetry axis specified by the angle φ_{s}. Since the parameters ΔU and μE are arbitrary, the function $u(\varphi)$ is symmetric for $\varphi_s = \frac{\pi q}{N}$, $q = 0, 1, \ldots, N-1$, if either $\varphi_1 = \frac{\pi}{2}$ or $\varphi_1 \neq \frac{\pi}{2}$ with $\sin(\varphi_s - \varphi_0) = 0$. In turn, the function $w(\varphi)$ is symmetric if $\cos(\varphi_s - \varphi_0) = 0$. Since the conditions $\sin(\varphi_s - \varphi_0) = 0$ and $\cos(\varphi_s - \varphi_0) = 0$ cannot be simultaneously satisfied, the only case when the two functions $u(\varphi)$ and $w(\varphi)$ are symmetric together is specified by the equations $\varphi_1 = \frac{\pi}{2}$ and $\cos(\varphi_s - \varphi_0) = 0$.

To satisfy the identity (5), the functions $u(\varphi)$ and $w(\varphi)$ must be symmetric and antisymmetric, respectively, with the same symmetry axis φ_s. This is realised when $\sin(\varphi_s - \varphi_0) = 0$. Summarising, from the above symmetry analysis, it follows that, at arbitrary amplitude φ_1 of the orientation fluctuations, the ratchet effect is impossible for an odd number of wells of the HRP, as well as for an even number of wells, if the average angle φ_0 of the fluctuating orientations of the electric field coincides with any of the symmetry axes of the HRP $\varphi_s = \frac{\pi q}{N}$, $q = 0, 1, \ldots, N-1$, which are oriented along the extrema of this potential.

Hopping rotation in a double-well HRP

At thermal energies $k_B T$ (k_B is Boltzmann constant, T is the absolute temperature) that are low compared to the reorientation barrier ΔU, the main contribution to the unidirectional rotation of the rotor is made by the hopping rotation. The description of such a rotation is simplified by the fact that the rotation parameters include the rate constants of transitions between the particle states in neighbouring potential wells [19; 28], i. e. the rate constants for overcoming the potential barriers that separate these wells. Therefore, we can take into account not only thermally activated overcoming the potential barriers, but also tunneling, which occurs at temperatures close to the absolute zero and, thus, to study ratchet systems in a wide temperature range with both the classical and quantum descriptions [29; 30].

Using the results of reference [31], the authors of reference [20] obtained an expression for the average rotation velocity Ω of the ratchet driven by the adiabatic alternation of two periodic N-well potential reliefs (states «plus» and «minus») characterised by the sets of minimum and maximum energy values $v^{\pm}_{\min,\, n}$ and $v^{\pm}_{\max,\, n}$, respectively, corresponding to the same angular coordinates $\varphi_{\min,\, n}$ and $\varphi_{\max,\, n}$ ($n = 1, 2, \ldots, N$, $0 = \varphi_{\min,\, 0} < \varphi_{\max,\, 1} < \varphi_{\min,\, 1} < \varphi_{\max,\, 2} \ldots < \varphi_{\min,\, N-1} < \varphi_{\max,\, N} < \varphi_{\min,\, N} = 2\pi$). The adiabatic alternation is understood as such an alternation when the time intervals between the switching potential reliefs (τ^+ and τ^-) are so long that the thermodynamic equilibrium has time to be established in each of them. The average angular velocity Ω is expressed through the nested sums of the functions $R^{\pm}_{\max,\, n}$ and $R^{\pm}_{\min,\, n}$ that depend on the sets of minimum and maximum energy values as follows:

$$\Omega = \frac{2\pi}{\tau}\Lambda_N,\ \Lambda_N = \sum_{n=2}^{N}\left(R^{+}_{\max,\, n} - R^{-}_{\max,\, n}\right)\sum_{m=2}^{n}\left(R^{+}_{\min,\, m} - R^{-}_{\min,\, m}\right),$$
$$R^{\pm}_{\max,\, n} = \frac{\exp\left(\beta v^{\pm}_{\max,\, n}\right)}{\sum_{l=1}^{N}\exp\left(\beta v^{\pm}_{\max,\, l}\right)},\ R^{\pm}_{\min,\, m} = \frac{\exp\left(-\beta v^{\pm}_{\min,\, m}\right)}{\sum_{l=1}^{N}\exp\left(-\beta v^{\pm}_{\min,\, l}\right)}, \tag{6}$$

where $\tau = \tau^+ + \tau^-$ is the period of the potential relief alternation, $\beta = (k_B T)^{-1}$ is the reverse thermal energy.

The analysis of the relations (6) for the rotor in the N-well HRP with dichotomous sign fluctuations of the external electric field ($\varphi_+ = \varphi_E$, $\varphi_- = \varphi_E - \pi$) showed that the ratchet effect existed for even values of N, and the average angular velocity was proportional to $\varepsilon^N \sin(N\varphi_E)$ at $\varepsilon = \beta\mu E \ll 1$ and tended to $\left(\frac{2\pi}{\tau}\right)\sin(N\varphi_E)$ at $\frac{\varepsilon}{N^2} \gg 1$. The ratchet effect of the highest intensity was associated with $N = 2$. That is why we will consider just the case $N = 2$ in our analysis of the rotational ratchet with the dichotomous fluctuations of electric-field orientations.

When the inequality $\mu E \ll \Delta U$ is satisfied, we can assume that the values of the angles $\varphi_{\min,\, n} = (n-1)\pi$ and $\varphi_{\max,\, n} = \left(n - \frac{1}{2}\right)\pi$ ($n = 1, 2$) approximately correspond to the angular locations of the minima and maxima of the potential (1) with $N = 2$, and

$$v^{\pm}_{\min,\, n} = -\frac{\Delta U}{2} + (-1)^n \mu E \cos\varphi_{\pm},\ v^{\pm}_{\max,\, n} = \frac{\Delta U}{2} + (-1)^n \mu E \cos\varphi_{\pm}. \tag{7}$$

Substituting expressions (7) into the relations (6) for $R^{\pm}_{\min,\,n}$ and $R^{\pm}_{\max,\,n}$ with $n = N = 2$ gives

$$R^{\pm}_{\min,\,2} = \left(1 + e^{2\varepsilon\cos\varphi_{\pm}}\right)^{-1},\ R^{\pm}_{\max,\,2} = \left(1 + e^{-2\varepsilon\sin\varphi_{\pm}}\right)^{-1},\ \varepsilon = \beta\mu E. \tag{8}$$

Thus, using (8) and the identity $\left(1 + e^{2a}\right)^{-1} = \dfrac{1 - \tanh a}{2}$, we finally get

$$\Lambda_2 = \frac{1}{4}\left[\tanh\left(\varepsilon\sin\varphi_+\right) - \tanh\left(\varepsilon\sin\varphi_-\right)\right]\left[\tanh\left(\varepsilon\cos\varphi_-\right) - \tanh\left(\varepsilon\cos\varphi_+\right)\right], \tag{9}$$

which is the ratchet velocity up to the constant factor $\dfrac{2\pi}{\tau}$.

Note that in the adiabatic mode of the motion, the average angular velocity does not depend on the barrier ΔU of the HRP. Therefore, the result (9), obtained for $\mu E \ll \Delta U$, can be used for both small and large values of the energy parameter ε. The quantity (9) vanishes when either $\sin\varphi_+ = \sin\varphi_-$ or $\cos\varphi_+ = \cos\varphi_-$. This means that the average orientation angle φ_0 is equal to either 0 or $\dfrac{\pi}{2}$; these values coincide with the orientations of the electric field either along the minima or along the maxima of the HRP. This result coincides with the result of the general symmetry analysis carried out at the beginning of our article.

Figure 1 represents two families of functions $\Lambda_2\left(\varphi_1\right)$ evaluated for two different values of the average orientation angle φ_0 and corresponding to several values of ε. The highest average angular velocity can be achieved at $\varphi_0 = \dfrac{\pi}{4}$ (solid lines), when this average orientation of the fluctuating electric field provides large simultaneous fluctuations of the maxima and minima of the potential energy. The location of the maxima of each family of functions $\Lambda_2\left(\varphi_1\right)$ correspond to the amplitude of the orientation fluctuations $\varphi_1 = \dfrac{\pi}{2}$, i. e. to the sign fluctuations of the electric field.

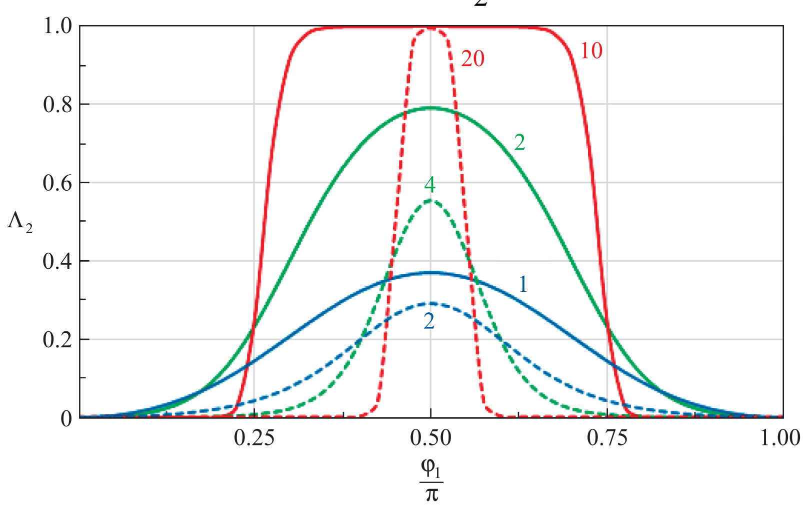

Fig. 1. The dependence of the average velocity Ω of the rotation ratchet (in units $\dfrac{2\pi}{\tau}$) on the amplitude φ_1 of the angular fluctuations of the electric field. The values of ε are indicated near the curves. The solid and dashed curves correspond to the values of the average orientation angle φ_0 equal to $\dfrac{\pi}{4}$ and $\dfrac{\pi}{20}$, respectively

Note that the value of Λ_2 is bounded from above by the value 1, i. e. the maximum possible average angular velocity is equal to $\dfrac{2\pi}{\tau}$. This value is achieved at large ε and $\varphi_1 = \dfrac{\pi}{2}$. The width of the plateau can be tuned with the parameter φ_0. The width of the plateau, which exits at large ε, decreases with the decrease of φ_0 from the value $\dfrac{\pi}{4}$.

The asymptotic behaviour of the average angular velocity at low electric fields follows from the expression (9) at $\varepsilon \ll 1$:

$$\Omega = \frac{\pi}{\tau}\varepsilon^2\sin^2\varphi_1\sin 2\varphi_0. \tag{10}$$

The angular dependences of this asymptotics turn out to be the same as in the high-temperature approximation considered in the next section.

Continuous rotation in the high-temperature mode

The high-temperature approximation implies that the potential energy of a pulsating ratchet fluctuates with coordinate and time such that the fluctuation amplitude is much less than the thermal energy $k_B T$. This helps one to obtain analytical representations for the average velocity of the ratchet in the general case of arbitrary forms of those coordinate and time dependences [32–34]. The choice of the potential energy fluctuations in the form of a spatially harmonic signal, i. e. as

$$w(x) = w_0 \cos\left[2\pi\left(\frac{x}{L} - \lambda_0\right)\right] \tag{11}$$

essentially simplifies the description. Then, assuming (11) and arbitrary functions $u(x)$ and $\sigma(t)$ (with $\langle\sigma(t)\rangle = 0$) in the additive-multiplicative potential energy (2), the average ratchet velocity can be represented as [35]

$$\langle v\rangle = 2\pi\left(\frac{L}{\tau_D}\right)\beta^3 w_0^2 \Psi\left(\frac{\Gamma}{s}\right)\operatorname{Im}\left\{u_2 e^{4\pi i\lambda_0}\right\},\ s = \frac{(2\pi)^2}{\tau_D},$$

$$\Psi\left(\frac{\Gamma}{s}\right) = s\left[\tilde{K}_\Gamma(s) + s\tilde{K}'_\Gamma(s)\right],\ \tilde{K}_\Gamma(s) = \int_0^\infty dt\, K_\Gamma(t)e^{-st},\ K_\Gamma(t) = \langle\sigma(t_0 + t)\sigma(t_0)\rangle, \tag{12}$$

$$u_q = \frac{1}{L}\int_0^L dx\, u(x)e^{-ik_q x},\ k_q = \frac{2\pi}{L}q,\ q = 0, \pm 1, \pm 2, \ldots .$$

Here $\tau_D = \dfrac{L^2}{D}$ is the characteristic diffusion time (D is the diffusion coefficient) over the potential energy period L, and $\tilde{K}_\Gamma(s)$ is the Laplace transform of the correlation function $K_\Gamma(t)$, that depends on both the current time t and the inverse correlation time Γ. Due to the spatially harmonic shape of the potential energy fluctuations $w(x)$, the average velocity of such a ratchet depends only on the second harmonic u_2 of the function $u(x)$. The importance of the result (12) is that, within the high-temperature approximation, it is valid for arbitrary time dependences of potential energy fluctuations described by the function $\sigma(t)$.

Note that potential energy fluctuations in the form of a spatially harmonic signal were considered in references [27; 36] to describe ratchets with a sawtooth and step function $u(x)$. In this work, the function $u(\varphi)$ includes the HRP, and the function $w(\varphi)$ (see the expression (3) valid for dichotomous fluctuations of the angular variable) exactly corresponds to the spatially harmonic signal (11) with $x = \varphi$, $L = 2\pi$, $2\pi\lambda_0 = \varphi_0 - \dfrac{\pi}{2}$, $\cos\left[2\pi\left(\dfrac{x}{L} - \lambda_0\right)\right] = -\sin(\varphi - \varphi_0)$, and $w_0 = \mu E \sin\varphi_1$. The second harmonic of the function $u(\varphi)$ from equation (3) is equal to $u_2 = -\dfrac{\Delta U}{4}$, so $\operatorname{Im}\left\{u_2 e^{4\pi i\lambda_0}\right\} = -\dfrac{\Delta U}{4}\sin(4\pi\lambda_0) = \dfrac{\Delta U}{4}\sin(2\varphi_0)$. Therefore, the expression (12) for the average angular velocity of the rotational ratchet driven by the dichotomous fluctuations takes on the following form:

$$\Omega = \frac{1}{4}\beta\Delta U\varepsilon^2 s\Psi\left(\frac{\Gamma}{s}\right)\sin^2\varphi_1 \sin 2\varphi_0. \tag{13}$$

This result is valid for $\beta\Delta U \ll 1$ and $\varepsilon \ll 1$. The angular dependence (both on φ_0 and φ_1) coincides with the asymptotic behaviour (10) obtained in the adiabatic approximation.

For symmetric stochastic dichotomous fluctuations with $\langle\sigma(t)\rangle = 0$, we have $K_\Gamma(t) = \exp(-\Gamma t)$ and $\tilde{K}_\Gamma(s) = (\Gamma + s)^{-1}$. Thus,

$$\Psi\left(\frac{\Gamma}{s}\right) = \frac{\dfrac{\Gamma}{s}}{\left(1 + \dfrac{\Gamma}{s}\right)^2}. \tag{14}$$

The adiabatic approximation is valid for $\Gamma \ll s$. Then $s\Psi\left(\dfrac{\Gamma}{s}\right) \approx \Gamma$ and

$$\Omega \approx \frac{\Gamma}{4}\beta\Delta U\varepsilon^2 \sin^2\varphi_1 \sin 2\varphi_0. \tag{15}$$

If we characterise the average durations of the states of the dichotomous process by times τ_+ and τ_-, then the inverse correlation time is $\Gamma = \tau_+^{-1} + \tau_-^{-1}$. For the symmetric process, $\tau_+ = \tau_-$ and $\Gamma = \frac{4}{\tau}$, where $\tau = \tau_+ + \tau_-$ is the average period of the process. Therefore, the expression (15) includes the same factor τ^{-1} as the expression (10) does; this distinguishes the average velocities in the adiabatic approximation. Expression (10) is valid for a hopping rotation, when $\beta\Delta U \gg 1$, while the expression (15) was obtained in the high-temperature approximation with $\beta\Delta U \ll 1$. From the comparison of these expressions we conclude that the average angular velocity is proportional to $\beta\Delta U$ at high temperatures, but it saturates as the temperature decreases, and the factor $\beta\Delta U$ reaches π. That is, for the hopping motion in the adiabatic approximation, the dependence of the average angular velocity on the reorientation barrier ΔU disappears.

Let's consider time-periodic (i. e. deterministic) fluctuations $\sigma(t+\tau) = \sigma(t)$ with the period τ. Due to the periodicity, the function $\sigma(t)$ can be expanded into a Fourier series:

$$\sigma(t) = \sum_j \tilde{\sigma}_j \exp(-i\omega_j t), \; \omega_j = \frac{2\pi j}{\tau}, \; j = 0, \pm 1, \dots . \tag{16}$$

Averaging over the period yields

$$\langle \sigma(t) \rangle = \frac{1}{\tau}\int_0^\tau dt \sigma(t) = \tilde{\sigma}_0 = 0,$$

$$K_\Gamma(t) = \frac{1}{\tau}\int_0^\tau dt_0 \sigma(t_0 + t)\sigma(t_0) = \sum_{j \neq 0} \left|\tilde{\sigma}_j\right|^2 \exp(-i\omega_j t), \tag{17}$$

$$\tilde{K}_\Gamma(s) = 2s\sum_{j=1}^{\infty} \frac{\left|\tilde{\sigma}_j\right|^2}{s^2 + \omega_j^2},$$

and the desired function $\Psi\left(\frac{\Gamma}{s}\right)$ takes on the following form:

$$\Psi\left(\frac{\Gamma}{s}\right) = 4s^2 \sum_{j=1}^{\infty} \frac{\left|\tilde{\sigma}_j\right|^2 \omega_j^2}{\left(s^2 + \omega_j^2\right)^2}. \tag{18}$$

Here and in equations (16), (17) the parameter $\Gamma = \frac{4}{\tau}$ is included in the definition of the frequencies ω_j: $\omega_j = \frac{\pi}{2}\Gamma j$.

From the expression (18) we can obtain $\Psi\left(\frac{\Gamma}{s}\right)$ for a deterministic dichotomous process if we consider a symmetric step function $\sigma(t)$ with the period τ: $\sigma(t) = 1$ for $0 < t < \frac{\tau}{2}$ and $\sigma(t) = -1$ for $\frac{\tau}{2} < t < \tau$. Then $\tilde{\sigma}_j = \frac{i}{\pi j}\left[1 - (-1)^j\right]$ and the summation in the formula (18) over odd j is performed analytically:

$$\Psi\left(\frac{\Gamma}{s}\right) = \frac{\Gamma}{s}\tanh\frac{s}{\Gamma} - \cosh^{-2}\frac{s}{\Gamma}. \tag{19}$$

In the adiabatic approximation $\Gamma \ll s$, the approximate equality $s\Psi\left(\frac{\Gamma}{s}\right) \approx \Gamma$ is again satisfied and we return to the expression (15). This means that in the adiabatic mode, deterministic and stochastic fluctuations lead to the same result. This is explained by the fact that in this mode, the lifetime of each state of the dichotomous process is much longer than the relaxation time τ_D and, in each potential profile, the equilibrium state has time to be established regardless of whether the dichotomous states are switched deterministically or stochastically.

We emphasise that the applicability of the formula (13), which contains the angles φ_0 and φ_1 representing the average over the dichotomous fluctuations and the fluctuation amplitude, strictly speaking, is limited to the consideration of a dichotomous process. At the maximum fluctuation amplitude $\varphi_1 = \frac{\pi}{2}$, the values of the angles φ_+ and φ_- differ by π. Therefore, we can assume that the magnitude of the electric field $E(t)$ itself, oriented at an angle φ_E to the axis x, fluctuates. For a dichotomous process, $E(t) = \pm E$. However, we can consider the general case with $E(t) = E\sigma(t)$, where $\sigma(t)$ is an arbitrary function of time, not necessarily equal to ± 1. For such a case, the formula (13) will be valid for a non-dichotomous process, if we put $\sin\varphi_1 = 1$ and $\sin 2\varphi_0 = -\sin 2\varphi_E$ in it. As the simplest example of a non-dichotomous periodic process, let's consider the sinusoidal periodic function $\sigma(t) = \sin\omega_1 t$, in which the only term with $j = 1$, $\left|\tilde{\sigma}_1\right|^2 = \frac{1}{4}$, remains in the sum (18), so that

$$\Psi\left(\frac{\Gamma}{s}\right) = \frac{\left(\frac{\pi\Gamma}{2s}\right)^2}{\left[1 + \left(\frac{\pi\Gamma}{2s}\right)^2\right]^2}. \tag{20}$$

Note that the low-frequency asymptotics of the expression (20) is quadratic in Γ, in contrast to the linear asymptotics in (15) for adiabatic dichotomous processes.

Figure 2 represents the graphs of the function $\Psi\left(\frac{\Gamma}{s}\right)$ calculated using relations (14), (19), and (20). The argument of this function determines the dimensionless fluctuation frequency, while the function itself is a factor in the expression (13), and therefore determines the average angular velocity of the rotor in question. The inset represents the low-frequency asymptotics, that are linear and coincident for the deterministic and stochastic dichotomous processes while quadratic for the sinusoidal fluctuations. This clearly demonstrates the advantage of the dichotomous process over the non-dichotomous one: the former provides much higher values of the average angular velocity compared to the latter. The periodic fluctuations lead to the narrow bell-shaped curves with maxima at $\frac{\Gamma}{s} \in (0.61 - 0.65)$ and the high-frequency asymptotics proportional to $\left(\frac{s}{\Gamma}\right)^2$. The stochastic dichotomous fluctuations correspond to the wide «bells» with the maximum at $\frac{\Gamma}{s} = 1$ and the high-frequency asymptotics $\frac{s}{\Gamma}$.

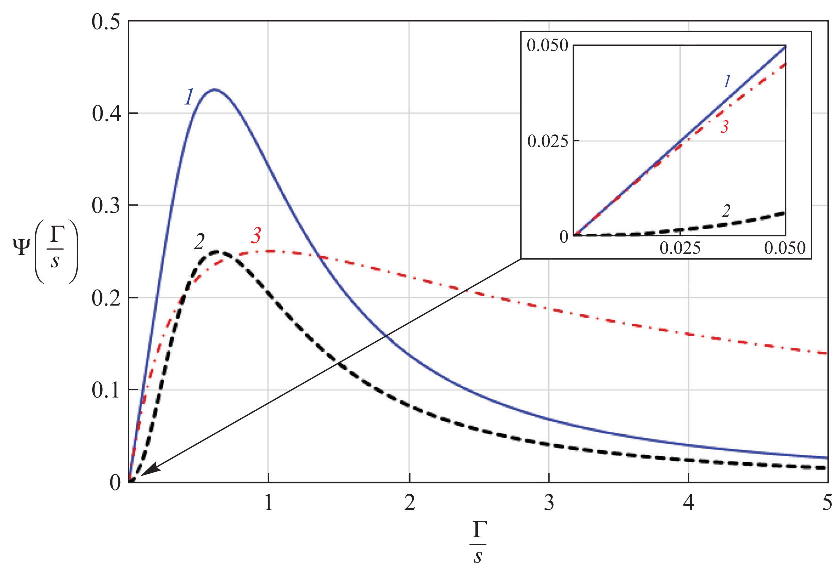

Fig. 2. The dependence of the factor Ψ, which determines, according to equation (13), the average angular velocity Ω of the rotation ratchet, on the dimensionless fluctuation frequency $\frac{\Gamma}{s}$. The curves *1*, *2*, and *3* correspond to the deterministic dichotomous, sinusoidal, and stochastic dichotomous fluctuations, given by equations (19), (18), and (13), respectively. The inset details the low-frequency behaviour of the curves *1*, *2*, and *3*

Discussion and conclusions

This article is the first to consider a rotational ratchet controlled by a dichotomous change in the orientations of the applied electric field. The average angle φ_1, which describes the amplitude of the orientation fluctuations, is an arbitrary-valued parameter, in contrast to the previously considered case with $\varphi_1 = \frac{\pi}{2}$ corresponding only to the sign fluctuations of the electric field, i. e. to the angle between the orientations of the fluctuating field equal to π [19; 20]. The ratchet effect is very sensitive to changes in parameters of the ratchet system, so the presence of one more parameter always provides an additional control over the average rotation velocity. The performed symmetry analysis showed that the ratchet effect is absent for an odd number of the wells of the HRP, as well as for an even number of the wells, if the average angle φ_0 of the fluctuating orientations of the electric field coincides with any symmetry axes of the HRP. The latter result can be associated with the well-known property according to which the existence of the ratchet effect requires synchronous fluctuations of the depths of the wells and the heights of the barriers of the periodic potential relief [3; 19]. Since the symmetry axes of the HRP pass through its minima and maxima, the coincidence of φ_0 with these orientations leads to fluctuations of either only the depths of the wells or only the heights of the barriers. There are no synchronous fluctuations, and therefore no ratchet effect.

Analytical expressions for the average rotation velocity have been obtained in the low-temperature and high-temperature approximations. At low temperatures, when the thermal energy is much less than the reorientation barrier, the hopping rotation occurs. The description of this rotation is the simplest in the adiabatic approximation, i. e. at low-fluctuation frequencies Γ, since in each of the states of the adiabatic process, equilibrium has time to be established. It does not matter whether these states are switched deterministically or stochastically. For the hopping motion, the relaxation time is determined by the inverse rate constant for overcoming the barrier ΔU of the HRP, that, according to the Arrhenius law, is equal to $k = k_0 \exp\left(-\frac{\Delta U}{k_B T}\right)$, where k_0 is the characteristic frequency of the angular oscillations of the rotor in the potential well. Therefore, the applicability of the adiabatic mode of the hopping motion is specified by the inequalities $k_B T < \Delta U$ and $\Gamma \ll k$. In this mode, the average rotation velocity Ω is proportional to Γ and independent of ΔU. The main ratchet parameters are the average angle φ_0 of the field orientations and the amplitude φ_1 of the orientation fluctuations, as well as the dimensionless electric field strength $\varepsilon = \frac{\mu E}{k_B T}$, which can take arbitrary values. Note that the hopping rotation can be considered outside the adiabatic approximation as well, at frequencies Γ satisfying $k \ll \Gamma \ll k_0$. Then Ω will depend on both ΔU and the mechanism of switching the states of the dichotomous process, and the value of Ω will saturate with the growth in Γ and be limited in order of magnitude to the value of k (this follows from the general description of the properties of flashing ratchets within the framework of the kinetic approach [37]). For example, for the hydroxyl groups of an oxide surface, the values k at room temperatures are of the order of 100 GHz [15], which is the upper limit of the rotation frequency of most rotor systems [10; 11]. When $\Gamma \gg k_0$, in the calculations of Ω, one should take into account the intrawell motion, that, when considered correctly, must lead to the general ratchet property: $\Omega \to 0$ when $\Gamma \to \infty$ [2].

Analytical expression (9), obtained for the average hopping-rotation velocity in the low-temperature approximation, and the corresponding families of the dependencies φ_1 of the average velocity show that the maximum ratchet effect is associated with large electric fields fluctuating in sign, i. e. with $\varepsilon \gg 1$ and $\varphi_1 = \frac{\pi}{2}$. In this case, the width of the plateau, that means the largest velocity, is maximum when the average angle φ_0 of the fluctuating orientations of the electric field is the bisector of the angles of the symmetry axes of the HRP, i. e. $\varphi_0 = \frac{\pi}{4}$, and the fluctuations of the barriers and wells of this potential are most correlated.

The high-temperature approximation assumes that the thermal energy is much greater not only than the reorientation barrier of the HRP, but also the interaction energy of the dipole rotor with the electric field μE, i. e. $\varepsilon \ll 1$. Therefore, the angular dependence of the average rotation velocity (13), obtained in this approximation, coincides with the dependence (10), obtained for the adiabatic hopping motion at $\varepsilon \ll 1$. The difference is that the expression (13) contains also the small factor $\beta \Delta U$ and the function $\Psi\left(\frac{\Gamma}{s}\right)$ which distinguishes stochastic and deterministic dichotomous angular fluctuations outside the adiabatic approximation. At the maximum amplitude of dichotomous angular fluctuations ($\varphi_1 = \frac{\pi}{2}$), one can replace these fluctuations by arbitrary-time dependences of the electric field $E(t)$ with $\langle E(t) \rangle = 0$, oriented at the angle φ_E to the axis x. Then the formula (13) with $\varphi_1 = \frac{\pi}{2}$ and $\varphi_0 = \varphi_E - \frac{\pi}{2}$ will be valid for arbitrary fluctuations $E(t)$.

The performed analysis of the dependence of the average rotation velocity Ω on the type of electric-field fluctuations showed that the low-frequency asymptotic behaviour of the velocity is proportional to the fluctuation frequency Γ for a dichotomous process with a jump-like change in either the magnitude or direction of the field. For continuous changes in the field strength with time, the low-frequency asymptotics of the velocity is proportional to the fluctuation frequency squared. Deterministic fluctuations lead to a bell-shaped frequency dependence $\Omega(\Gamma)$, the width of which is much narrower than that for a stochastic dichotomous process. The high-frequency asymptotics of the velocity is proportional to Γ^{-2} for the deterministic fluctuations and to Γ^{-1} for stochastic ones. Both frequency dependences tend to zero in the high-frequency limit, as it should be for ratchet systems.

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