Chemistry, Physics and Technology of Surface. 2016. V. 7. N 4. P. 444-452

UDC 533.723:577.352.4(086.48) doi: 10.15407/hftp07.04.444

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MOLECULAR ROTOR AS A HIGH-TEMPERATURE BROWNIAN MOTOR

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The aim of the present work is to develop a theory of high-temperature polar molecular rotors, the rotation of which is initiated by an external alternating electric field. Such rotors can be considered as Brownian motors in the sense that a unidirectional translatory motion of motor particles is similar to the unidirectional rotation of moving part of a rotor; that allows using, at calculations, the high-temperature theory of Brownian motors. The potential energy of a rotating particle, as a function of time and rotation angle, is represented in additive-multiplicative form. The approximation of the two first harmonics suffices to describe a smooth potential profile. Indeed, they are the minimum number of harmonics to simulate the asymmetry of the potential relief and to describe molecular rotors since the first harmonic is conditioned by the interaction of the rotor with the alternating field and the second one – by the potential of hindered rotation. With these conditions, the velocity of rotation of polar molecular rotors is calculated, and the analytical dependences of the velocity on the parameters of the model is analyzed for the two cases of time dependence of the applied alternating electric field, harmonic and stepped. It is shown that, at a stepped change of an alternating electric field, the values of the angular velocity are higher, for any frequency of the applied field, than those at a harmonic change, and a dichotomic mode of the electric field changing is generally more efficient one, resulting in the optimal mode of rotor operation. Moreover, there exists a qualitative difference, in the low-frequency asymptotics of the rotor velocity, between harmonic and stepwise external field: the average velocity of rotation is proportional to the square of the field modulation frequency in the former case, and it is the linear function of the frequency in the later case.

Keywords*: molecular rotor, Brownian motor, ratchet, diffusion kinetic, potential fluctuations*

INTRODUCTION

Physically adsorbed molecules are loosely bound to the surface, which provides them with relatively high mobility, and, in particular, the rotational mobility. The last is also typical for 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 [1, 2]. Recently high interest has been attracted by so-called molecular rotors artificially formed on surfaces [3–15]. They are the special case of molecular motors. These molecular engines provide an insight into the physical principles of controlled mechanical movement and friction on the nanoscale as well as the effects of random thermal movement that are inherent in nanodevices as opposed to conventional macromachinery.

Rotational movement of molecules and atomic groups on a solid surface manifests itself in a variety of experiments. Vibrational spectroscopy detects specific absorption bands in the frequency regions of both stretching and deformation (angular) vibrations, the former also giving rise to the spectral lines at combined frequencies, i.e., at sums and differences of the frequencies of original lines. In addition, rotational movement causes specific broadening of spectral lines, with its temperature dependence governed by the rotational reorientation frequencies. For instance, rotations of hydroxyl groups on oxide surfaces become possible due to relatively small reorientation barriers ($\Delta U_\varphi \approx 55$ meV) that are comparable to the characteristic thermal energy ($k_BT \approx 26$ meV at $T = 300$ K). As a result, characteristic IR absorption arises in the frequency region 100–200 cm^{-1} and a typical temperature dependence (of the Arrhenius type) is observed for the spectral bands of the stretching O–H vibrations [1].

Molecular rotor is a system in which a molecule or part of the molecule rotates relatively to the other part of the molecule or to a macroscopic object such as surface or solid. Such a device is capable of converting energy from one form to another. Molecular rotors are characterized by repetitive unidirectional rotational movement

taking place under the influence of the applied alternating electric field. Rotors may be constructed artificially. They are becoming increasingly popular because of their high sensitivity to various external influences.

Authors of [7] have studied crystalline arrays of pairs of molecular rotors. They described the experiment in which they could assign two types of Brownian rotators with different activation energies on the single rotor site. Work [8] is devoted to the study of ultra-fast molecular rotors. In work [9], authors have cooled molecular rotors from room temperature to about 4 K using an electronically-exciting broadband laser to simultaneously drive cooling resonances from many different rotational levels. In work [10], molecular rotors were described used as non-mechanical sensors of viscosity of some polymer and biological material. Authors applied this system to patients with various levels of glycosuria (a symptom of diabetes), it was found to provide excellent correlation with different clinical assessments of diabetes [10].

Work [11] describes amphidynamic crystals - formations from a combination of static components, that determine the crystalline order. These elements are associated with dynamic elements that make conformational motion along a certain direction [11]. Arrhenius analysis of the data of such structures gives information about the activation energies and pre-exponential factor for different parts of the rotor. The observed pre-exponential factors are 4–6 times greater than those of elementary site-exchange processes, indicating that the dynamics is not dictated by static energetic potentials. Instead, the activation energies for rotations in the crystals are controlled by temperature dependent local structural fluctuations and crystal fluidity [11].

Recently femtosecond laser techniques were developed, which can bring the gas molecules to extremely fast rotation in a very short time, while keeping their translational motion relatively slow [12]. It is shown that the way to equilibrium starts with the metastable "gyroscopic stage", during which the molecules maintain their fast rotation and orientation of the angular momentum through many collisions. After a certain induction time, the "gyroscopic stage" is abruptly terminated by rotational–translational energy exchange, which leads the gas to the final equilibrium [12]. Understanding the mechanism of motion in crystalline arrays of molecular rotors with complex dynamics is a key step that will contribute to development of molecular machines that can perform useful work [13]. The authors of [13] investigated the specificity of the weak links between the two rotors and the simulated energy barriers. In [14], authors discussed the artificially created rotors with "paddle-like" arms.

During the last decade, many nanomachines with controlled molecular motions have been studied on metallic surfaces, which are easy to make very clean, and that are stable over months [15]. In article [15], authors present their strategy to operate at temperatures higher than 80 K, in particular, through the use of semiconducting silicon surfaces. On metallic surfaces, the nanovehicles are molecules with two or four triptycenes as wheels and the molecular motor is built around a ruthenium organometallic center [15].

The low temperature rotor rotation velocities were studied previously [5, 16]. These works address the Brownian motion of a particle in a periodic two-well potential under the action of the external alternating electric field. In [16], it was shown that in the case when the particle energy was more than reorientation barriers of hindered rotation, the important role was played by the diffusion character of the movement, and we can apply the theory of high-temperature Brownian motors.

The novelty of the present article consists in the development of the theory of high-temperature rotors, based on the concept of Brownian motors. When using the known expression for the velocity of directed motion of nanoparticles, which are Brownian motors, direct motion of molecular rotor for two cases of time dependence of the applied alternating field: harmonic dependence $E(t) = E\cos(\omega t)$ and dichotomous process (stepped time-dependent) will be studied.

The fundamental relations of the theory of high temperature Brownian motors and their adaptation to describe the rotational motion are shown in the second section of the paper. The cases allowing simplification, and the final ratio obtained for further calculations are considered as well. The third section examines in detail the scheme of a molecular rotor work, formulates statement of the problem, and gives the potential energy of the rotating particle-rotor in the additive-multiplicative form. The final section deals with two cases of the time dependence of the alternating electric field and for each of them the analytical expressions of

angular velocity of the rotor depending on the parameters of the model have been obtained; the asymptotic behavior of the average angular velocity in the low and high frequencies is analyzed, and conclusions are formulated.

THE BASIC THEORY OF HIGH-TEMPERATURE RATIO OF BROWNIAN MOTORS

Currently existing theory of Brownian motors (ratchets) - modeling devices that convert non-equilibrium fluctuations into the directional mechanical movement of the nanoparticles – allows using different approximations to obtain analytical expressions for the average velocity of nanoparticle as a function depending on the model parameters [17].

Typically, in the theory of Brownian motors dynamics of studied particle is described by the Smoluchowski equation for the distribution functions $\rho_{\pm}(x,t)$, defining the location of a particle in one of two states with the potential profiles $U_+(x)$ or $U_-(x)$ and with additional terms, that specify the velocities of transition probabilities of the particle between $U_+(x)$ and $U_-(x)$:

$$\frac{\partial}{\partial t}\rho_{\pm}(x,t) = -\frac{\partial}{\partial x}J_{\pm}(x,t) \mp \left[\gamma_+\rho_+(x,t) - \gamma_-\rho_-(x,t)\right], \qquad J_{\pm}(x,t) = -De^{-\beta U_{\pm}(x)}\frac{\partial}{\partial x}\left[e^{\beta U_{\pm}(x)}\rho_{\pm}(x,t)\right]. \tag{1}$$

Here $D = k_BT/\zeta$ is diffusion coefficient, ζ is friction coefficient, and $\beta = (k_BT)^{-1}$ is the inverse thermal energy.

Brownian rotors are analogues of Brownian motors in the sense that the description of the translational motion of the particles is replaced by a rotation one. In describing the rotational motion of the rotor, coordinate x in the relations for Brownian motors should be replaced by the angular variable φ, in this case, these quantities are related by ratio $x = R\varphi$, where R is the radius of the circle along which rotational movement takes place. In the theory of Brownian motors, periodicity of the potential energy of the coordinate x with the period L, $U(x+L,t) = U(x,t)$ is supposed. Time-dependence in deterministic Brownian motors is also assumed as periodic function of time, so $U(x,t+\tau) = U(x,t)$, there τ is the time period. These periodic conditions allow expanding $U(x,t)$ in a double Fourier series on the spatial and temporal variables:

$$U(x,t) = \sum_{qj} U_{qj}\exp(ik_qx - i\omega_jt),\ k_q = (2\pi/L)q,\ \omega_j = (2\pi/\tau)j,\ q,j = 0, \pm1, \pm2, ... \tag{2}$$

For rotational motion, using the analogy between the translational and rotational movement, from which qualities $x = R\varphi$ and $L = 2\pi R$ follow, the expansion of the periodic potential energy $U(\varphi,t) = U(\varphi+2\pi,t)$ in a double Fourier series on the spatial and temporal variables will take the form:

$$U(\varphi,t) = \sum_{qj} U_{qj}\exp(2\pi iq\varphi - i\omega_jt),\ \omega_j = (2\pi/\tau)j,\ q,j = 0, \pm1, \pm2, ... \tag{3}$$

It is well known that the initiation of the Brownian motion of the motor takes place in two main mechanisms: the rocking (by applying a variable external force, it is zero in average) and flashing (by fluctuations in potential profile with the saving of its spatial periodicity). Note that for the molecular rotor reviewed here, these two mechanisms are equivalent due to the natural periodicity of the angular variable.

We use the well-known expression for the average velocity of high temperature Brownian motor [17,18]:

$$\langle v\rangle = i\beta^3D^3\sum_{qj,q'j'}\frac{k_q^2k_{q+q'}^2k_{q'}U_{qj}U_{q'j'}U_{-q-q',-j-j'}}{\left(i\omega_j + Dk_q^2\right)\left(i\omega_{j+j'} + Dk_{q+q'}^2\right)}, \tag{4}$$

i is the imaginary unit. This expression is valid for small changes in the amplitude ratio of the potential relief $U(x,t)$ to the thermal energy k_BT. This expression in the important special case of the additive-multiplicative representation of the potential energy

$$U(x,t)=u(x)+\sigma(t)w(x), \qquad U_{qj}=u_q\delta_{j,0}+\sigma_j w_q, \tag{5}$$

(where $u(x)$, $\sigma(t)$, and $w(x)$ are the arbitrary periodic functions) takes such form:

$$\begin{aligned}\langle v\rangle = &-2\beta^3D^2\sum_{qq'}k_q^2k_{q'}w_qw_{q'}u_{-q-q'}\sum_{j\neq0}\frac{\omega_j\left|\sigma_j\right|^2}{\left(i\omega_j+Dk_q^2\right)\left(i\omega_j+Dk_{q'}^2\right)}+\\ &+i\beta^3D^3\sum_{qq'}k_q^2k_{q+q'}^2k_{q'}w_qw_{q'}w_{-q-q'}\sum_{\substack{jj'(\neq0)\\(j+j'\neq0)}}\frac{\sigma_j\sigma_{j'}\sigma_{-j-j'}}{\left(i\omega_j+Dk_q^2\right)\left(i\omega_{j+j'}+Dk_{q+q'}^2\right)}.\end{aligned} \tag{6}$$

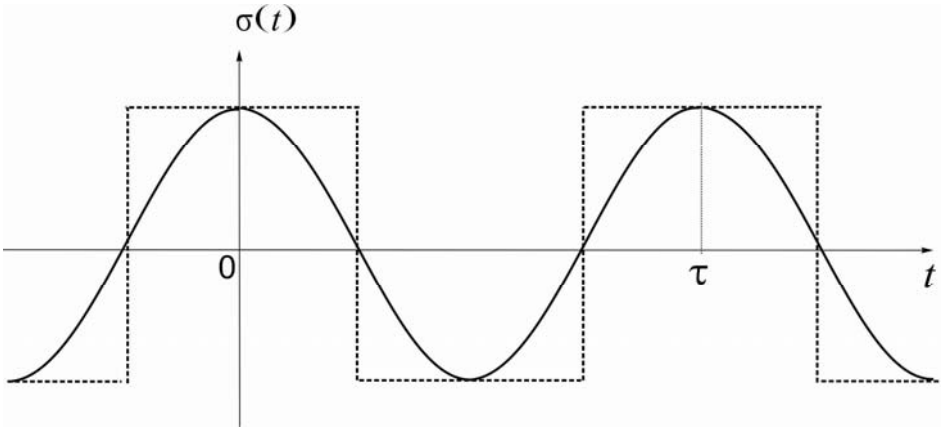


Fig. 1. Cosine (solid curve) and stepped (dashed curve) time dependence of an external electric field

Symmetrized representation of this formula for the dipole rotor in a two-well symmetric potential of hindered rotation can be expressed as:

$$\begin{aligned}\langle v\rangle = &\,2i\beta^3D^3\sum_{\substack{qq'(\neq0)\\(q+q'\neq0)}}k_qk_{q'}k_{q+q'}\left(k_q^2+k_{q'}^2\right)w_qw_{q'}u_{-q-q'}\psi\left(Dk_q^2,Dk_{q'}^2\right)-\\ &-\frac{i}{2}\beta^3D^3\sum_{\substack{qq'(\neq0)\\(q+q'\neq0)}}k_q^2k_{q'}^2k_{q+q'}w_qw_{q'}w_{-q-q'}\sum_{\substack{jj'(\neq0)\\(j+j'\neq0)}}\frac{\sigma_j\sigma_{j'}\sigma_{-j-j'}+\sigma_{-j}\sigma_{-j'}\sigma_{j+j'}}{\left(i\omega_j+Dk_q^2\right)\left(i\omega_{-j'}+Dk_{q'}^2\right)},\end{aligned} \tag{7}$$

where

$$\psi\left(a,b\right)=\sum_{j=1}^{\infty}\frac{\omega_j^2\left|\sigma_j\right|^2}{\left(\omega_j^2+a^2\right)\left(\omega_j^2+b^2\right)}. \tag{8}$$

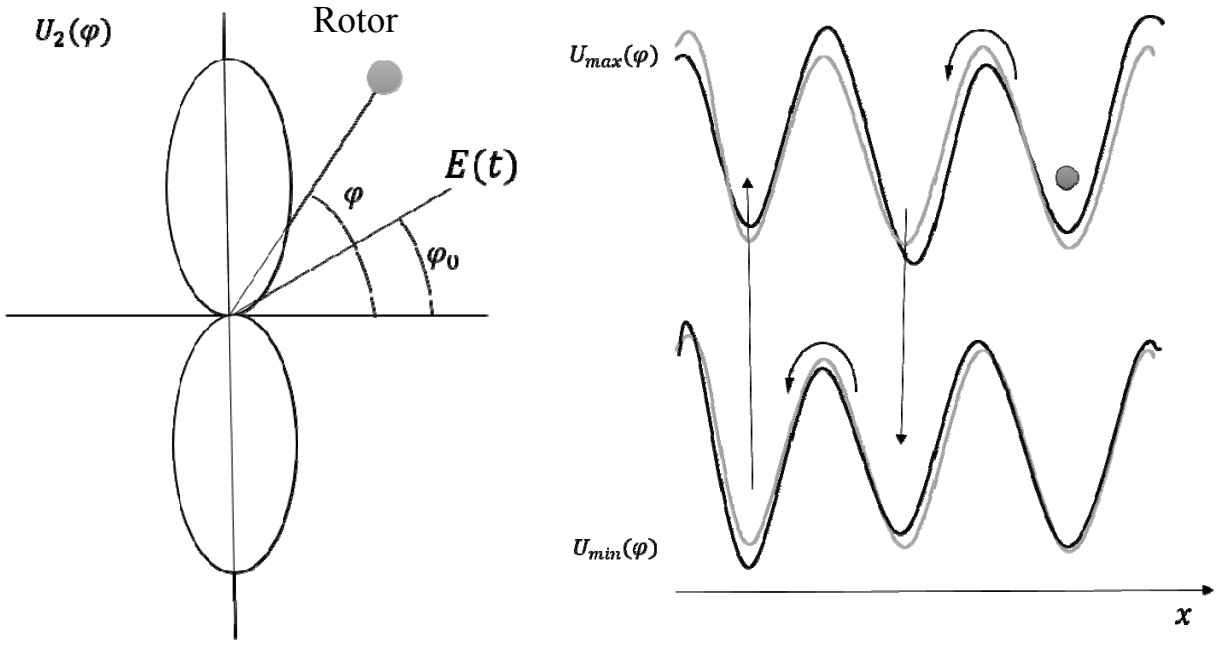
In certain cases molecular rotors [for example, cosine and stepped time-dependence of the applied alternating field (Fig. 1)], the second sum in (7) is missing. In addition, if the potential relief is sufficiently smooth, we can use the approximation of first two harmonics: the minimum number of harmonics that can describe the asymmetry of the potential profile that is sufficient to describe the molecular rotors. Given these conditions, we can rewrite the expression for the average velocity (7) as:

$$\begin{aligned}\langle v\rangle = &-16\beta^3D^3k_1^5\left[\psi\left(Dk_1^2,Dk_1^2\right)\mathrm{Im}\left(u_2^*w_1^2\right)+\right.\\ &\left.+5\psi\left(Dk_1^2,Dk_2^2\right)\mathrm{Im}\left(u_1w_1w_2^*\right)\right]\end{aligned} \tag{9}$$

Expression (9) will be used below in calculating the average velocity of rotor rotation.

FORMULATION OF THE PROBLEM

In [5], a theoretical analysis of *n*-well potentials has shown that unidirectional rotation is possible only in two-well potential case. We shall confine ourselves to this case. The alternating electric field $E(t)$ is oriented at the angle φ_0, and the rotor itself is oriented at the angle φ to the plane of rotation (see Fig. 2 *a*). At Fig. 2 *b*, the mechanism of occurrence of unidirectional rotation is represented in a symmetric two-well periodic potential (dashed curves), with the wells and barriers modulated with an alternating external field that is switching between two opposite directions. The asymmetry of hindered rotation potentials modified by the field makes a particle jump from the shallow well into the deep one. Change in the field polarity occurring with time causes vertical transitions of the particle from the deep to the shallow well [5].



a *b*

Fig. 2. The mechanism of the molecular rotor movement: (*a*) azimuthal potential of hindered rotation with two wells and two barriers presented in polar coordinates. The alternating electric field $E(t)$ is oriented under the angle φ_0, and rotor is oriented under the angle φ to the plane of rotation; (*b*) the mechanism for the occurrence of unidirectional rotation in a periodic symmetric two-well potential (thin curves). Wells and barriers are modulated with an alternating external field that is switching between two opposite directions (thick curves at the top and bottom)

It is convenient to represent the potential energy of a rotating particle-rotor as a function of the angle of rotation and time in additive-multiplicative form similar to equation (5) for the forward movement:

$$U(\varphi,t) = u(\varphi) + \sigma(t)w(\varphi). \qquad (10)$$

For a rotor having the symmetry axis of the second order, the function $u(\varphi)$ may be represented as hindered rotation potential:

$$u(\varphi) = (1/2)\Delta U(1 - \cos 2\varphi), \qquad (11)$$

and the multiplicative part of the potential energy as

$$\sigma(t)w(\varphi) = E(t)\mu\cos(\varphi - \varphi_0), \qquad (12)$$

where the time dependence $E(t)$ is given by the modulation of the applied alternating field, μ is absolute value of the dipole moment of the rotor, and φ_0 is the angle between the reference axis of the potential wells and the applied field. At this representation, the spatial part of the potential energy is the sum of the first two harmonics (the first harmonic of the rotor due to the interaction

with the alternating field, and the second is due to the potential of hindered rotation). This allows to use the high-temperature theory of Brownian motors to study rotors rotation velocity depending on the type of $E(t)$.

RESULTS AND DISCUSSION

Let us consider the case of a cosinusoidal alternating electric field dependence (Fig. 1, solid curve). In this case only time harmonics with $j = \pm 1$, $\sigma_1 = \sigma_{-1} = 1/2$, $\omega_1 = -\omega_{-1} = 2\pi/\tau$ are not equal to zero. Thus, using (9), we obtain an expression for the angular velocity of the dipole rotor:

$$\Omega_G = -\Omega_0 \sin 2\varphi_0 \frac{\tilde{\omega}^2}{4(1+\tilde{\omega}^2)^2}, \tag{13}$$

where

$$\Omega_0 = \beta^3 \mu^2 E^2 \Delta U \frac{D}{R^2}, \ \tilde{\omega} = \frac{R^2}{D}\omega, \ \omega = \frac{2\pi}{\tau}. \tag{14}$$

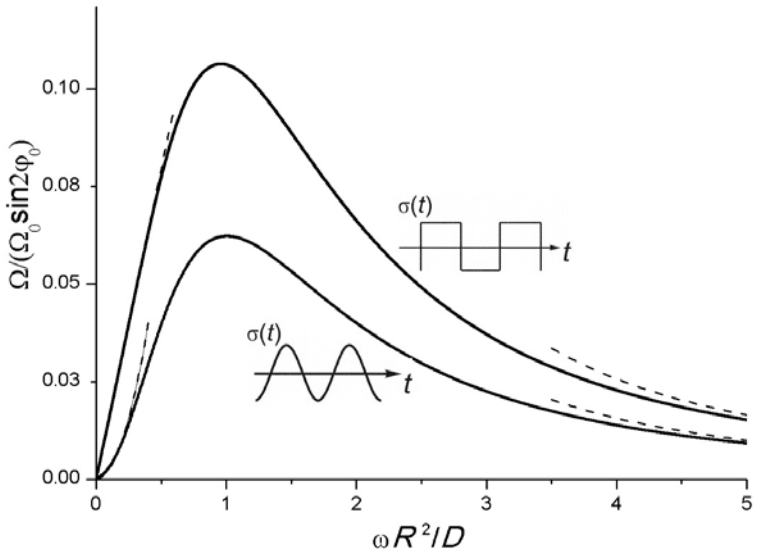


Fig. 3. The dependence of the angular velocity of the rotor in the dimensionless units in the case of harmonic change of the alternating field (lower solid curve) and the stepped change of the field (the upper solid curve) on the dimensionless parameter $\tilde{\omega} = \omega R^2/D$, that is proportional to the modulation frequency of the applied field. The dashed lines show the asymptotes for low and high frequency areas for each dependence

In the Fig. 3 the lower solid line corresponds to the expression (13) for the harmonic alternating field changing. Dashed lines show asymptotes to it: $\Omega_G \approx -\tilde{\omega}^2/4$ is for low frequencies and $\Omega_G \approx -1/(4\tilde{\omega}^2)$ is for high frequencies.

Let us consider a dichotomic mode of the electric field changing which is described by the stepped time dependence of $\sigma(t)$ represented by dotted line in Fig. 1 (symmetric deterministic dichotomous process). For such a symmetrical stepped function, Fourier components of the function $\sigma(t)$ are equal to $\sigma_j = 2i/(\pi j)$ and are not equal to zero only for odd values j. Using (9) and performing the summation in (8) for a function $\psi(a,b)$ with a known table-valued expression for sum [19]:

$$\sum_{n=0}^{\infty} \frac{1}{\left[(2n+1)^2 + a^2\right]^2} = \frac{\pi}{8a^3}\tanh\frac{\pi a}{2} - \frac{\pi^2}{16a^2}\cosh^{-2}\frac{\pi a}{2}, \tag{15}$$

we obtain the final expression for the angular velocity in this case:

$$\Omega_S = -\Omega_0 \sin 2\varphi_0 \left(\frac{\tilde{\omega}}{2\pi}\tanh\frac{\pi}{2\tilde{\omega}} - \frac{1}{4}\cosh^{-2}\frac{\pi}{2\tilde{\omega}}\right), \tag{16}$$

where Ω_0 and $\tilde{\omega}$ are determined in (14).

In Fig. 3, expression (16) corresponds to the upper solid curve characterized by asymptotic $\Omega_S \approx -\tilde{\omega}/(2\pi)$ for low frequencies and $\Omega_S \approx -\pi^2/(24\tilde{\omega}^2)$ for high frequencies (see the dotted lines adjacent to the curve).

Let us discuss the results. As seen in Fig. 3 at the stepped modeling of the alternating field, the absolute values of the angular velocity is high for any frequency of the applied field. In addition, in adiabatic limit for the low frequencies, the character of asymptotic dependence of the angular velocity on the frequency differs: in the case of harmonic external field, it is quadratic dependence, $\Omega_G \approx -\tilde{\omega}^2/4$, and for stepped one, it is linear, $\Omega_S \approx -\tilde{\omega}/(2\pi)$. Thus, we can conclude that the optimal mode of operation of the rotor with the highest rotation velocity, is the stepped mode of the applied field, in accordance with the general conclusion of [18], which is valid for Brownian motors. At the low frequencies of the field changing, this conclusion agrees with the fact that the linear law of increase of the rotational velocity exceeds the quadratic. In an arbitrary frequency range, this conclusion can be justified by the fact that the efficiency of the Brownian motor increases with the fluctuation of the potential energy, average values are significantly greater for a stepped change of the field (see Fig. 1).

CONCLUSIONS

In the present work we examined the unidirectional rotation of the polar molecular rotors, arising under the influence of an external alternating electric field. Time-dependences of the applied field were studied for harmonic and stepped mode of field changing. We used theory of high-temperature Brownian motors to obtain the analytical dependence of the rotation velocity for each of these cases. It is shown that, at a stepped change of an alternating electric field, the values of the angular velocity are higher, for any frequency of the applied field, than those at a harmonic change, and a dichotomic mode of the electric field changing produces more efficient regime of rotor operation. Moreover, there exists a qualitative difference, in the low-frequency asymptotics of the rotor velocity, between harmonic and stepwise external field: it is a quadratic one in the former case while linear in the latter case.

Молекулярний ротор як високотемпературний броунівський мотор

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Розраховано швидкість обертання полярних молекулярних роторів з використанням теорії високотемпературних броунівських моторів. Досліджено аналітичні залежності швидкості обертання від параметрів моделі для двох випадків залежності прикладеного змінного електричного поля від часу: гармонійної та ступінчастої. Середня швидкість обертання ротора в першому випадку пропорційна квадрату частоти модуляції поля, а в другому – лінійно залежить від частоти. Показано, що оптимальним режимом роботи ротора є дихотомний режим зміни електричного поля.

***Ключові слова**: молекулярний ротор, броунівський мотор, ретчет, дифузійна динаміка, флуктуації потенціалу*

Молекулярный ротор как высокотемпературный броуновский мотор

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Рассчитана скорость вращения полярных молекулярных роторов с использованием теории высокотемпературных броуновских моторов. Изучены аналитические зависимости скорости вращения от параметров модели для двух случаев временной зависимости приложенного переменного электрического поля: гармонической и ступенчатой. Средняя скорость вращения ротора в первом случае пропорциональна квадрату частоты модуляции поля, а во втором – линейно зависит от частоты. Показано, что оптимальным режимом работы ротора является дихотомный режим изменения электрического поля.

***Ключевые слова**: молекулярный ротор, броуновский мотор, рэтчет, диффузионная динамика, флуктуации потенциала*

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Received 25.07.2016, accepted 03.11.2016