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NONLINEAR PHENOMENA

Tunneling Mechanism for Changing the Motion Direction of a Pulsating Ratchet. Temperature Effect

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A pulsating ratchet with a spatially periodic double-well potential profile undergoing shift fluctuations for half a period is considered. The motion direction in such a ratchet is determined by the probability of overcoming which of the barriers surrounding the shallow potential well is greater. At relatively high temperatures, in accordance with the Arrhenius law, the probabilities of overcoming the barriers are determined by their heights, and at temperatures close to absolute zero, when the ratchet moves according to the tunnel mechanism, the barrier shapes are also important. Therefore, for narrow high and low wide barriers, the overcoming mechanism may turn out to be different and, moreover, dependent on temperature. As a result, a temperature-induced change in the direction of the ratchet motion is possible. A simple interpolation theory is presented to illustrate this effect. Simple criteria are formulated for the shape of the potential relief, using which one can experimentally observe motion reversal.

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Among various properties of nanoscale systems in which a directed nanoparticle motion can be a result of the ratchet effect, one of the most impressive is the possibility to control the motion direction by changing some parameter of the system, for example, the ambient temperature or the frequency of internal or external fluctuations [1–4] (see also review [5] and the references therein). The mechanisms of obtaining the directed motion due to non-equilibrium fluctuations and asymmetry of the periodic potential profile of a nanoparticle differ significantly for the two main classes of ratchets, called ratchets with fluctuating force (rocking ratchets) and ratchets with a fluctuating (pulsating) periodic potential energy or simply pulsating (or flashing) ratchets [2, 3]. In the first of them, nonequilibrium fluctuations of the nanoparticle potential energy appear under the action of an external variable (fluctuating) force with a zero mean value, and in the second, are induced by fluctuations of the internal parameters of this potential profile itself. Different methods of introducing fluctuations into the system lead to different dependences of the particle current (average ratchet velocity) on the coefficients of spatial and temporal asymmetry of the potential energy of nanoparticles [6], as well as to different character of its dependence on the fluctuation frequency [7, 8]. The most studied is the first ratchet class, for which new properties were mainly discovered [3, 9].

Rocking ratchets are known to be easier implemented experimentally [5], e.g., by using an external electromagnetic field as the governing fluctuations. That is why the possibility of motion controlling by creating competition between the spatial and temporal asymmetry of the particle potential energy was first demonstrated for fluctuating-force ratchets [10, 11] and only after almost 10 years for pulsating ratchets [12, 13]. Accounting for quantum effects in the mechanisms of functioning of microscopic ratchets obviously goes in the same sequence.

It was shown in the pioneering work [1] that, a fluctuating-force ratchet, characterized at sufficiently high temperatures by some motion direction, can alter the motion direction at low temperatures, when the tunneling motion mechanism prevails over the classical one. This theoretical result was confirmed experimentally [14] (for details, see [15]). For pulsating ratchets, the effect of the reversal of the motion direction has been unknown so far.

This work discovers the possibility of the motion reversal due to the tunnel effect for pulsating ratchets. The model of a highly efficient Brownian motor with a periodic double-well potential profile fluctuating for

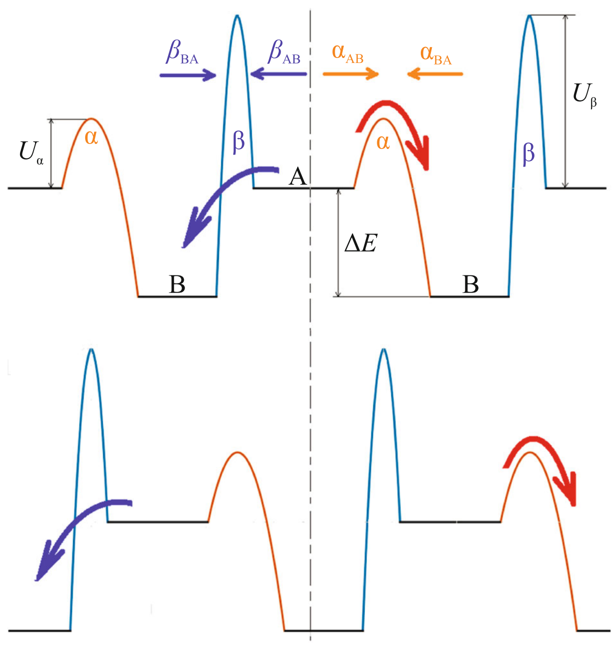

Fig. 1. (Color online) Half-period shifted double-well potential profiles of a pulsating ratchet, which are shifted by half a period relative to each other (upper and lower profiles). Shallow and deep potential wells are denoted by A and B. Wide low and narrow high barriers are denoted by α and β. The rate constants of transitions through the barriers are indicated by the corresponding barrier symbols and well indices, the sequence of which indicates the transition direction. Arrows to the right and to the left (red and blue) indicate the motion directions of the ratchet, which operates due to the thermal activation or tunneling processes, respectively.

half a period [16, 17] was chosen as the basic model and then extended to take into account the tunneling effect. The motion direction in such a ratchet is determined by which of the barriers surrounding the shallow potential well is more likely to overcome it.

In Fig. 1, a shallow potential well is bounded by a high and narrow barrier on the left and by a low and wide one on the right. At high temperatures, the probability of overcoming a barrier, according to the Arrhenius law, is determined only by its height. Therefore, the right barrier is more easily overcome, and the particle is more likely to end up in the right deep potential well. Dichotomous fluctuations of the potential profile $V(x)$ by half of its spatial period, $L/2$, mean that if, in one of the states of the dichotomous process, the potential relief is described by the function $V(x)$, then, in the other dichotomous state, it will undergo the shift by $L/2$ with complete preservation of its shape, and be described by the function $V(x \pm L/2)$. As a result of such fluctuations, the particle located in the deep well by the end of the profile lifetime will be thrown vertically into a shallow well of the shifted profile. Then the process is repeated in the shifted profile. Due to the energy acquired by the particle during the potential profile shifts, an average rightward directed motion arises (see the red arrows to the right in Fig. 1), which is the essence of the ratchet effect.

At temperatures near the absolute zero, the thermal activation motion of the ratchet freezes and tunneling begins to dominate, for which, both the height and width of potential barriers become important. With proper selection of the potential profile parameters, the probability of tunneling through the narrow high barrier will be greater than through the wide low one. Therefore, the described operating mechanism of the pulsating ratchet is realizable at low temperatures too, down to absolute zero, the motion direction reverse with the temperature decrease is being the fundamental difference here (see the blue arrows to the left in Fig. 1).

The tunneling mechanism for reversing the motion direction of a pulsating ratchet was not previously known and is the main content of this work. It differs from that given in [1] for a fluctuating force ratchet in that a small fluctuating force itself led to the necessary distortions in the shape of a single barrier on the period, while at least two barriers on the period with a certain ratio of their heights and widths are required to reverse the motion of a pulsating ratchet. Below we give a quantitative analysis of the discussed effect based on a rigorous result of the kinetic description of ratchets of the pulsating type [17, 18], using the rate constants for overcoming potential barriers. Both for high and for extremely low temperatures, these constants can be represented by simple analytical relations (the Arrhenius law and the Gamow formula, respectively [19, 20]), which are applicable to discover the conditions for the motion reversal effect. For intermediate temperatures, a simple interpolation-based description is presented, which uses the crossover temperature that is the temperature which separates the regions of the dominance of the thermally activated and tunneling mechanisms for overcoming potential barriers [21–23]. The purpose of this simplified description in the intermediate temperature range is to give a reader an understanding of the key features of the temperature dependence of the average velocity (including the motion reversal). A rigorous description of the temperature dependence of the transition rate constants [24] is beyond the scope of this article, which aims to demonstrate only the possibility of the motion reversal effect for pulsating ratchets.

Analytical solutions of the Smoluchowski equation describing the characteristics of a classical overdamped pulsating ratchet with a periodic asymmetric potential shifted by half a period are obtained in [16]. At relatively high temperatures which correspond to the thermal energy not exceeding the energy barriers, the motion of a Brownian particle can be considered as hopping and the kinetic description can be applied.

Such a description for the considered ratchet was carried out in [17] assuming a stochastic dichotomous process of potential profile fluctuations (shifts), and in [18] for deterministic fluctuations (a deterministic dichotomous process) with each profile is being characterized by a given lifetime, τ_+ or τ_-. For the stochastic dichotomous process, these lifetimes should be replaced by the average frequencies of transitions between the profiles, $\langle\tau_+^{-1}\rangle$ and $\langle\tau_-^{-1}\rangle$, the sum of which defines an important fluctuation characteristic—the inverse correlation time, Γ. In the case of the symmetric dichotomous process, $\tau_+ = \tau_-$, the value of Γ is related to the process period (average period), $\tau = \tau_+ + \tau_-$ as $\Gamma = 4/\tau$.

Within the kinetic approach, dichotomous shifts of a double-well potential profile by half a period are described by the antisymmetric model [18]. This model is characterized by the given set of the rate constants of transitions α_{AB}, α_{BA} and β_{BA}, β_{AB} through each potential barrier α and β in both directions (Fig. 1), as well as by the equal values of τ_+ and τ_-. In the absence of a load force, which is usually considered when calculating the energy characteristics of ratchets, the rate constants corresponding to the reverse transitions are subject to the detailed balance relation [25]

$$\alpha_{BA} = \alpha_{AB} e^{-\Delta E/k_B T}, \quad \beta_{BA} = \beta_{AB} e^{-\Delta E/k_B T}, \tag{1}$$

where k_B is the Boltzmann constant, T is the absolute temperature, and ΔE is the difference between the energies of zero-point oscillations in the potential wells A and B, which is approximately equal to the difference between the energy minima of these wells if they have close curvatures. The use of the relations (1) significantly simplifies the general expressions obtained in [18] and allows us to arrive at a simple result for the particle current, J (the average ratchet velocity $\langle v\rangle$ is related to the current as $\langle v\rangle = JL$, where L is the spatial period of the potential profile):

$$J = \frac{1}{4}\Gamma\varphi(\Gamma)\frac{\alpha_{AB} - \beta_{AB}}{\alpha_{AB} + \beta_{AB}}\tanh(\Delta E/2k_B T). \tag{2}$$

Here, the function $\varphi(\Gamma)$ of the inverse correlation time Γ is defined by the following expressions for the deterministic and stochastic dichotomous process:

$$\varphi(\Gamma) = \begin{cases} \tanh(\Sigma/\Gamma), & \text{deterministic}, \\ \Sigma(\Sigma + \Gamma)^{-1}, & \text{stochastic}, \end{cases} \tag{3}$$
$$\Sigma = (\alpha_{AB} + \beta_{AB})(1 + e^{-\Delta E/k_B T}).$$

Expression (2) has a clear physical interpretation. At $\Delta E > 0$ (that is, when the barriers β and α bound the shallow potential well on the left and right, respectively, as shown in Fig. 1), the sign of the current J is determined by the sign of the difference $\alpha_{AB} - \beta_{AB}$. This means that the ratchet motion is in the direction from the less deep well to that neighboring barrier, which is more likely to be overcome. If either barriers α and β are identical ($\alpha_{AB} = \beta_{AB}$) or the potential wells A and B are identical ($\Delta E = 0$), the ratchet effect does not occur, since the potential profile is described by a symmetric periodic function [26]. Comparison of the wells and barriers of the initial potential profile and the one shifted by half a period (Fig. 1) leads to an important observation: Directed motion occurs only with simultaneous fluctuations of the parameters of potential barriers and wells of this pulsating ratchet [27, 28].

The explicit expressions for the rate constants of transitions, α_{AB} and β_{AB}, through the right and left barriers are determined by the type of the processes which one takes into account. To optimize the representations of similar expressions for barriers α and β we will use γ to notate the rate constant of overcoming an arbitrary barrier and represent the temperature dependence of this constant in the form:

$$\gamma(T) = k_0 e^{-S_\gamma(T)}, \quad \gamma = \alpha, \beta. \tag{4}$$

Here, the pre-exponential factor k_0, which means the frequency of collisions of the particle with the potential barrier, can, with the exponential accuracy, be considered independent of temperature, so that the entire temperature dependence is contained in the exponent, the function $S_\gamma(T)$.

At relatively high temperatures, the Arrhenius law is valid, for which $S_\gamma(T) = U_\gamma/k_B T$. At temperatures tending to absolute zero, the function $S_\gamma(0)$ must follow the Gamow formula

$$S_\gamma(0) = \frac{2}{\hbar}\int_a^b dx\sqrt{2m[U_\gamma(x) - E]}, \tag{5}$$

where $\hbar$ is the Planck constant, m is the particle mass, and E is the particle energy, which sets the boundaries of the potential barrier, a and b. The temperature criterion for the boundary separating the temperature region in which tunnel transitions predominate over Arrhenius ones for a parabolic barrier has the form [21–23]:

$$T_\gamma^* = \hbar\Omega_\gamma(2\pi k_B)^{-1}, \quad \Omega_\gamma \equiv \sqrt{|U_\gamma''(x_{max})|/m}, \tag{6}$$

where x_{max} is the coordinate of the barrier maximum. Including dissipation, characterized by the friction coefficient ζ, into the description leads to the replacement in Eq. (6) of the "barrier" frequency Ω_γ by the factor [1, 19]

$$\mu = \frac{\sqrt{\zeta^2 + 4m|U_\gamma''(x_{max})|} - \zeta}{2m}, \tag{7}$$

which tends to $|U_\gamma''(x_{max})|/\zeta$ at $m|U_\gamma''(x_{max})| \ll \zeta^2$.

Thus, in a wide temperature range satisfying $T^*_\gamma \ll T < U_\gamma/k_B$, the Arrhenius law is valid and the sign of the current J is determined by the sign of the difference in the heights of the barriers surrounding the shallow potential well:

$$\operatorname{sgn} J = \operatorname{sgn}(U_\beta - U_\alpha). \quad (8)$$

Since in the region of extremely low temperatures, $T \ll T^*_\gamma$, the Gamow formula (5) can be used, then in this region,

$$\operatorname{sgn} J = \operatorname{sgn}(U_\beta/T^*_\beta - U_\alpha/T^*_\alpha). \quad (9)$$

With introducing the potential-barrier-width parameter Δx_γ, the value $S_\gamma(0)$ can be written as $S_\gamma(0) = \kappa\hbar^{-1}\Delta x_\gamma\sqrt{2mU_\gamma}$, where κ is a numerical factor depending on the barrier shape. For a rectangular barrier, κ is equal to 2, for a parabolic one, $\kappa = \pi/2$ and $S_\gamma(0) = U_\gamma/k_B T^*_\gamma$, and for a triangular one, $\kappa = 4/3$. Then the relation (9) can be represented as $\operatorname{sgn} J = \operatorname{sgn}(\Delta x_\beta\sqrt{U_\beta} - \Delta x_\alpha\sqrt{U_\alpha})$, which is valid for barriers of various shapes.

Comparing Eqs. (8) and (9), we get that the conditions for the motion reversal under the transition from the thermally activated to the tunneling mechanism of overcoming the barriers in the pulsating ratchet with a half-period shifted potential can be represented in the following two equivalent forms:

$$\left(\frac{\Delta x_\beta}{\Delta x_\alpha}\right)^2 < \frac{U_\alpha}{U_\beta} < 1, \quad \frac{T^*_\alpha}{T^*_\beta} < \frac{U_\alpha}{U_\beta} < 1. \quad (10)$$

These conditions mean that the motion reversal occurs when the ratio (which is less than unity) of the heights of the barriers surrounding the shallow potential well will exceed the ratio of either the corresponding boundary temperatures (6) or the square of the inverse ratio of the widths of these barriers.

Let us consider the temperature dependence (2) of the current in the case of the thermally activated mechanism of overcoming the potential barriers. Using the Arrhenius law for the transition rate constants leads to the dependences of the current on the inverse temperature, represented by the solid lines on the left side of Fig. 2 and in the inset. Nonmonotonicity at high temperatures is the result of the stochastic resonance, which manifests itself in nonlinear systems, when the system response has a resonance-like behavior depending on the noise level (temperature) [29].

The manifestation of the stochastic resonance in the temperature dependences of the characteristics of rotational polar ratchet systems (for which the differences between the two ratchet classes are erased) under the action of an alternating electric field was studied in detail in [28]. A characteristic feature of the stochastic resonance is an increase in the temperature

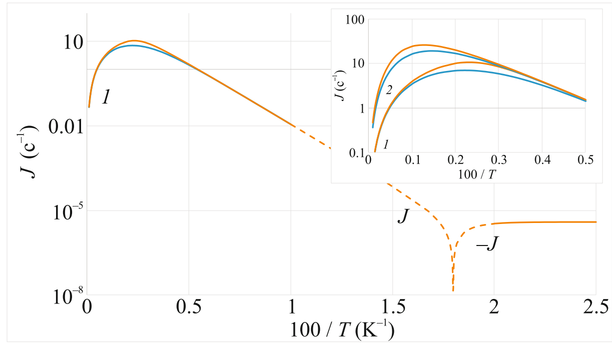

Fig. 2. (Color online) The inverse-temperature dependence of the current (logarithmic scale), calculated by Eq. (2). The solid lines in the regions of high and extremely low temperatures correspond to the Arrhenius transition rate constants and those calculated by the Gamow formula (5) for parabolic barriers. The upper and lower lines in the region of the maxima (orange and blue) correspond to deterministic and stochastic dichotomous fluctuations, respectively. The dotted line represents the schematic behavior of the current in the region of intermediate temperatures, where the motion reversal occurs. The following parameter values have been used in calculations: $U_\alpha/k_B = 1000$ K, $T^*_\alpha = 50$ K, $U_\beta/k_B = 2500$ K, $T^*_\beta =$ 140 K, $\Delta E/k_B = 500$ K, $k_0 = 1$ kHz, $\Gamma = 0.1$ kHz (line *1*), $\Gamma = 1$ kHz (line *2*).

of the maximum response of the system with an increase in the frequency of an external field. In our case, this frequency is the inverse correlation time Γ. In the inset of Fig. 2, line *2* corresponds to the value of Γ which is 10 times greater than the value of Γ for line *1*. Then, in the inverse temperature dependence of the current, the maximum shifts to the left so that the temperature of the maximum of line *2* is approximately twice the analogous temperature of line *1*. This indicates that we indeed deal with the stochastic resonance.

Only in a narrow temperature range near the maximum of the stochastic resonance one can observe the splitting of the temperature dependences of the current into the curves which describe deterministic and stochastic dichotomous fluctuations of the potential profile shifts for half a period. In the rest of the temperature range, these dependences are degenerate.

This behavior is easily explained by the form of the dependence $\varphi(\Gamma)$ (3) for the deterministic and stochastic dichotomous process. At a fixed value of Γ, in the high temperature region, $\Sigma \gg \Gamma$, the function $\varphi(\Gamma)$ is approximately equal to 1 for both processes and the current is proportional to Γ. At low temperatures, $\Sigma \ll \Gamma$, the function $\varphi(\Gamma)$ is approximately equal to Σ/Γ for both processes, and the current ceases to depend on Γ. Therefore, lines *1* and *2* merge at low temperatures, differ greatly at high temperatures, since they correspond to different values of Γ, and in the narrow region of the stochastic resonance, $\Sigma \approx \Gamma$, where the functions $\varphi(\Gamma)$ for the stochastic and deterministic processes differ, the degeneracy is removed within each pair.

Note that the applicability range of the kinetic approach used in this work does not allow consideration of the fluctuation frequency values, Γ, exceeding the characteristic frequencies of intrawell motion (the pre-exponential factor k_0 in (4)). In the frequency range $\Gamma \gg k_0$, the approximation of the hopping motion is no longer admissible, and a rigorous consideration of solutions to the Smoluchowski equation with continuous potentials leads to the disappearance of the ratchet effect as $\Gamma \to \infty$, as it should be [2] (the presence of potential jumps allows the presence of the ratchet effect in the overdamped motion mode [16]). Nevertheless, at low temperatures, when $\Sigma \ll \Gamma$, the use of the kinetic approximation is quite acceptable due to the fact that the probabilities of overcoming the barriers $\exp[-S_\gamma(T)]$ are exponentially small. Therefore, $\Sigma \sim \gamma(T) \ll k_0$ and, in the region of low temperatures, the frequency of fluctuations only needs to fall into the interval $\Sigma \ll \Gamma \ll k_0$. We also note that the phenomenon of the stochastic resonance is also inherent in fluctuating force ratchets (rocking ratchets) [28]. Nevertheless, in [1], which studied precisely this class of ratchets, stochastic resonance was not noted, since only the adiabatic (low frequency) regime of motion was considered.

The solid line in Fig. 2 in the region of extremely low temperatures corresponds to the negative ratchet tunneling current calculated by Eq. (2) with the transition rate constants (4), in which $S_\gamma(0) = U_\gamma/k_B T_\gamma^*$, where the temperature T_γ^* was determined by Eq. (6) for parabolic barriers. The parameter values were chosen such that the condition (10) of the current sign reversal with the change in temperature was satisfied.

In order to schematically describe the characteristic behavior of the temperature dependence of the current in the region of the sign change, we took a somewhat rough representation, which is nevertheless often used, of the total rate constants of overcoming potential barriers as the sum of the Arrhenius and tunneling contributions (see Eq. (9.1) and the discussion of the simple quantum theory of transition states in [19]). The reason for the use of such a representation is that it correctly reproduces the values of the transition rate constants in the regions of high and low temperatures: At high temperatures, the smallness of the tunneling contribution compared to the Arrhenius one justifies it, while, at low temperatures, the exponentially fast vanishing of it. The dotted line in Fig. 2 represents the result of such a description. The current sign reversal (the ratchet stopping point appearance) occurs at a temperature $T \approx 56$ K which is within the interval (T_α^*, T_β^*) and close to the value $T_\alpha^* = 50$ K.

Note that for a comprehensive description of the temperature dependence of the current in the region of the sign change as well as for determining the temperature of the ratchet stopping point, one should take into account that the particle tunneling rate constant depends on particle mass, the parameters of the barrier to be overcome, the properties of the medium, and temperature. Several mechanisms are known for the exponential dependence of the rate constant on temperature (4). For example, for the electron transfer in a polar medium, the temperature dependence is determined by the energy of its reorganization; for the tunneling of atoms and ions, the main role is played by the intermolecular vibrations that change the magnitude and shape of a potential barrier, as well as the reorganization of the medium [23, 24, 30–34]. The energy dissipation in the process of tunneling at zero and arbitrary temperatures also plays a significant role [19, 35–39].

In this work, to analyze the temperature dependence of the current (including the analysis of the possibilities for its reversal) in a highly efficient pulsating ratchet with a periodic double-well potential profile fluctuating for half a period, Eq. (2) is proposed. This formula, subject to the detailed balance condition (the absence of load forces or concentration gradients), is a simplification of the result known for the antisymmetric ratchet model with dichotomous switching of two states with two reaction channels [18]. The value and attraction of Eq. (2) is in its structure, the product of several factors, each of which reflects various important properties of the ratchet in question.

In Eq. (2) for the current, the fluctuation frequency-dependent factor $\Gamma\varphi(\Gamma)$, defined by Eq. (3), firstly, distinguishes a pulsating ratchet from a fluctuating-force one in that it gives the proportionality of the current to the frequency Γ in the adiabatic mode of the motion, secondly, it demonstrates the existence of the stochastic resonance outside the adiabatic mode, and thirdly, it describes the difference between the currents induced by a stochastic dichotomous process and the deterministic one, which occurs only in a narrow temperature range near the maximum of the stochastic resonance.

The remaining factors in Eq. (2) reflect the symmetry of the model under consideration, and also contain rate constants for overcoming potential barriers, the explicit form of which makes it possible to include into the description both classical thermal activation and quantum tunneling processes. The potential profile asymmetry is provided by the difference between the depths of the potential wells, ΔE, and the velocity rate constants α_{AB} and β_{AB}, so that the sign of the current is determined by the signs of ΔE and $(\alpha_{AB} - \beta_{AB})$. For classical and quantum processes, we have $\mathrm{sgn}(\alpha_{AB} - \beta_{AB}) = \mathrm{sgn}(U_\beta - U_\alpha)$ and $\mathrm{sgn}(\alpha_{AB} - \beta_{AB}) = \mathrm{sgn}(\Delta x_\beta \sqrt{U_\beta} - \Delta x_\alpha \sqrt{U_\alpha})$, respectively, where U_γ and

Δx_γ are the heights and widths of the right ($\gamma = \alpha$) and left ($\gamma = \beta$) barriers surrounding the shallow potential well ($\Delta E > 0$). Therefore, under the conditions (10), the motion reversal is realized which is schematically shown in Fig. 2 by the dotted line.

Note that, in contrast to a fluctuating-force ratchet, the analytical description of tunneling of which turned out to be possible only in the adiabatic (low frequency) regime [1], the analysis of the motion reversal of the pulsating ratchet carried out in this article is free from this limitation. It also revealed the stochastic resonance and the difference of its characteristics for deterministic and stochastic fluctuations of the potential reliefs. To observe experimentally the predicted motion reversal of a pulsating ratchet of the described type, it is required to create a periodic potential relief, with a certain asymmetric shape, fluctuating for half a period. The source of such a relief, for example, in organic pulsating electron ratchets [40] might be finger electrodes fabricated by the focused-ion-beam-assisted deposition. In this case, its half-period fluctuations can be realized by switching the potentials applied to these electrodes.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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