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Rectified motion induced by ac forces in periodic structures

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Abstract. — A particle in a periodic potential can be set into macroscopic motion by an ac force of zero mean value if the potential is asymmetric in space or the ac force is asymmetric in time. We analyze features of the resulting complex behaviour at zero and low temperatures within the framework of a simple sawtooth potential. This allows us to suggest experiments promoting separation methods and analysis of motor protein assemblies.

Introduction.

On symmetry grounds, a particle current may be obtained in asymmetric potentials without application of any force in the direction of motion [1-3], provided that dissipation is forced one way or the other [3]. As part of a more general effort aimed at understanding how this concept could lead to the elaboration of selective pumps, we investigate in this article the behaviour of a particle at low Reynolds number, submitted to both a pinning potential and an external ac force of zero average value.

Asymmetric units such as valves and diodes have been known to act as rectifiers for quite a long time. They are « brutal » in the sense that they essentially forbid transport in one direction. A softer picture is provided by the evolution of a particle in a periodic potential under the influence of a time-periodic ac force of zero mean value, when spatial asymmetry of the potential or temporal asymmetry in the ac signal results in a rectified average macroscopic
drift. This is clear from the inspection of the generic equation describing the motion of an overdamped particle in the absence of noise:

\[
\frac{dx}{dt} = -\xi \frac{dU}{dx}(x) + \gamma \phi(t)
\]

(1)

where \(\xi\) and \(\gamma\) measure the amplitudes of the potential and ac force respectively. The temporal and spatial units \(t\) and \(x\) have been rescaled so that the periodicity conditions read:

\[ U(x + 1) = U(x) \]

and

\[ \phi(t + 1) = \phi(t) \]

(2)

(3)

then from (1), to any solution \(x(t)\) corresponds a solution \(-x(t + 1/2)\). However, as will be shown below, the average velocity must be independent of the initial conditions. Thus (2) and (3) impose a zero average velocity. Conversely a net average dc drift can be obtained when any of the two symmetries (2, 3) does not hold. The subject of this article is the quantitative analysis of the resulting velocity.

The present considerations bear some resemblance with the recent work of reference [4] in which \(\phi(t)\) is a more or less structured noise. It differs however in two ways: first we allow for the alternating force to be asymmetric in time (i.e. (3) does not hold), second we do not try to provide a paradigm for the function of motor protein assemblies [3-6], but rather want to promote experiments which take advantage of the rectifying processes we describe.

The analysis of the « phase diagrams » describing the average drift velocity as a function of the amplitudes of the pinning potential and of the ac force, reveals a surprising complexity including the existence of « Devil's staircases » and of an already observed resonant behaviour in the rectification [4]. We further show that the competing effects of spatial and temporal asymmetry can cancel the rectifying process in interesting ways.

We first analyze the case of an asymmetric sawtooth potential and a square symmetric ac force with no (or very little) noise. This allows us to extract some characteristics of these rectifying processes such as the resonant behaviour. The complementary situation of a symmetric potential with an asymmetric ac force is then shown to produce similar features. In a third stage, we illustrate how the competition of asymmetric potential and ac force can produce average drifts of variable sign. We show that antiresonance situations (zero velocity at intermediate conditions) can then be obtained. After commenting on the influence of thermal noise we end by suggesting experiments in which these rectifying processes could be of use: new separation devices are proposed, as well as investigation of motor protein assemblies.

1. Model.

The motion of a massless particle immersed in a fluid medium and submitted to both a constant periodic pinning potential \(W(x)\) and a homogeneous external force \(F(t)\) can be described by a simple Langevin equation:

\[
\xi \frac{dx}{dt} = -\frac{dW}{dx}(x) + F(t) + \Gamma(t)
\]

(4)

where the friction coefficient \(\xi\) is related to the autocorrelation function of the white noise \(\Gamma\) by an Einstein relation: \(\langle \Gamma(t)\Gamma(t') \rangle = 2\xi kT \delta(t - t')\). We write \(W(x) = AU(x/p)\) and
\( F(t) = B \phi(t/\tau) \) to exhibit the periodicities \((p \text{ and } \tau)\), shapes \((U \text{ and } \phi)\) and amplitudes \((A \text{ and } B)\) of the potential and external force. Equation (4) can then be rewritten:

\[
\frac{dx}{dt} = -\varepsilon \frac{dU}{dx}(x) + \gamma \phi(t) + \theta(t)
\]

with \( x = x/p \), \( t = t/\tau \), \( \varepsilon = A \tau / \xi p^2 \), \( \gamma = B \tau / \xi p \), \( \langle \theta(t) \theta(t') \rangle = 2 \alpha \delta(t-t') \) where \( \alpha = kT \tau / \xi p^2 \).

Equation (1) corresponds to the limit of negligible noise \( \alpha \ll \varepsilon, \gamma \). One can show that the average drift velocity, \( V = \lim_{t \to x} (x(t) - x(0))/t \), does not depend on the initial position of the particle. This is easily seen for \( \alpha = 0 \):

- First, (1) being a first order differential equation, the knowledge of the location \( x_0 \) of the particle at time \( t = 0 \) entirely defines the trajectory \( \lambda(t, x_0) \). As a consequence, trajectories cannot cross, so that \( x_0 \leq \lambda_1 \) implies that \( x(t, x_0) \leq \lambda(t, \lambda_1) \) for any time \( t \).
- Second, the periodicity of the pinning potential and the homogeneity of the external force grants that \( \lambda(t, x_0 + n) = \lambda(t, \lambda_0 + n) \).

Hence, any initial condition \( \lambda_1 \) with \( x_0 = \lambda_1 \leq x_0 + 1 \) will lead to a trajectory satisfying \( \lambda(t, x_0) \leq \lambda(t, \lambda_1) \leq \lambda(t, x_0 + 1) \), therefore of the same average velocity than \( \lambda_0 \). For given shapes \( U \) and \( \phi \), the average velocity \( V \) is thus only a function of \( \gamma \) and \( \varepsilon \), but not of the initial conditions.

We will focus with some detail on the following potential and force, illustrated in figure 1:

\[
\begin{align*}
x & \in [0, a] \quad U(x) = x/a ; \\
t & \in [0, c] \quad \phi(t) = 1/2 c ; \quad t & \in [c, 1] \quad \phi(t) = -1/2(1-c)
\end{align*}
\]

where \( a, b = 1 - a \) and \( c \) are smaller than 1. As a convention we take \( a \geq b \).

2. Spatially asymmetric potential.

We start with the analysis of the action of a temporally symmetric force \( c = 1/2 \). The essence of the results in this case is summarized in figure 2. Continuous lines delineate isovelocity
Fig. 2. — a) « Phase diagram » in the \((\gamma, \varepsilon)\) plane for an asymmetric potential \((a = 0.75, \epsilon = 1/2)\). Continuous lines describe the lower contour of isovelocity domains; for sake of readability we display only the thresholds to attain velocities \(1 = 1/4, 1/3, 1/2, 1, 2, 3, \ldots\). Dashed lines correspond to the theoretical description as given by equation (8). The comparison to these lines, valid in the upper region, shows to what extent the wiggling aspect of the curves is due to the finite number of simulations \((200 \times 200)\). b) Blown-up view. Only are represented \(V = 2 - 1/2, 2 - 1/3, 2 - 1/4, 2 - 1/5, 2 - 1/6, 3 - 1/2, 3 - 1/3, 3 - 1/4, 3 - 1/5, 3 - 1/6\) and 3. The straight lines represent \(\gamma = \varepsilon/b - 2b\) and \(\gamma = \varepsilon/b\)
domains. They are obtained by a numerical resolution of equation (1). The different regimes are best understood by increasing progressively the force $\gamma$ at constant pinning potential $\varphi$ (Fig. 3). The two important force scales are clearly $\gamma_1 = \varphi/a$ and $\gamma_2 = \varphi/b$.

**a) Small forces,** $\gamma < \gamma_1$, the external perturbation is unable to extract the particle from the potential well in which it was initially. $V = 0$.

**b) Intermediate forces,** $\gamma_1 < \gamma < \gamma_2$, the particle can climb uphill the smallest slope, in a direction we will call forward in the following (to the right in Fig. 1), whereas it cannot go uphill in the backward direction. An obvious «rectification» or average drift can be obtained. Clearly the average velocity is $V = n$ if during one time period, the particle moves exactly an integer number of periods $n$. This is not necessary however:

Suppose that starting at the origin (see Fig. 1), the particle travels during the first half-period (forward pulse) a distance $\gamma = (n - 1) + a + \gamma_1$. During the second half-period (reverse pulse), even though the external force tends to drive it backwards, it will move forwards as the potential gradient wins $\gamma > \varphi/b$. Two situations may then be encountered:

- **Regime 1** the particle has time to fall in the next bottom well, and stays there until the next force sign reversal. This leads to an integer average velocity $n$. The threshold to obtain this velocity in this regime corresponds to $x_1 = 0^+$, or

$$\frac{1}{2} = (n - 1) \left[ \frac{a}{\gamma - \varphi/a} + \frac{b}{\gamma + \varphi/b} \right] + \left[ \frac{a}{\gamma - \varphi/a} \right].$$

- **Regime 2**: the particle does not have time to fall all the way down to the bottom, it thus starts the next time period with a little backward shift. Upon repetition of the time periods the
Fig. 4. — Velocity $V$ as a function of ac force intensity $\gamma$ for an asymmetric potential ($a = 0.6, c = 1/2, \epsilon = 4, \alpha = 0$): Successive magnification [(a) to (c)] of the scale exhibits the « Devil's staircase » structure of the curve. (d): parabolic shape of the « staircase » obtained when the smooth potential $U(x) = (\cos (2 \pi x) - 0.5 \sin (4 \pi x))/2 \pi$, and force $\phi(r) = \cos (2 \pi r)$ are used together with parameters $\epsilon = 5, \gamma = \gamma_0 + \Delta \gamma$ ($\gamma_0 = 10.65566975$).
shifts accumulate so that after \( m \) time periods it can no longer cross the \( n \)-th hill in the subsequent forward step and falls in the \((n-1)\)-th bottom. Thus after \( m+1 \) periods the particle returns to a position similar to its starting one, its average velocity being \( V = n - 1/(m+1) \). The threshold to obtain this velocity reads:

\[
\frac{1}{2} = n \left[ \frac{a}{\gamma - v/a} + \frac{b}{\gamma + v/b} \right] - \frac{1}{m} \left[ \frac{b}{\gamma + v/b} \right] - \frac{(m-1)}{2m} \left[ \frac{v/b - \gamma}{\gamma + v/b} \right].
\]

The existence of regime 2 requires \( \gamma > \gamma_c = v/b - 2b \). Note that the \( V = n - 1/2 \) threshold curve of regime 2 is the exact analytic continuation of the \( V = n \) threshold of regime 1.
in both cases the threshold is determined by the fact that a distance \((n - 1) + a\) is travelled during the forward pulse. Upon increasing \(\gamma\), non zero velocities are obtained when \(\gamma = \epsilon/a + 2a\), with a direct jump to \(V = 1\) if \(\epsilon < 2ab\), or starting by rational values otherwise.

c) \textit{Large forces,} \(\gamma > \gamma_2\), now during the reverse pulses the particle can climb uphill backwards. The situation becomes more involved but one can still describe most of the data. Clearly, increasing \(\gamma\) favours both forward and backward motion, but the main feature is the allowance of long backward drives, and the velocity on average decreases although some local increase can be observed.

The general behaviour may be inferred from an analysis of the first, or if needed of the \(m\)-th return map. Whenever the \(m\)-th return map intersects the \(n\)-th translated diagonal, defining a stable fixed point, a rational velocity \(n/m\) is obtained. The switch from velocity \(n\) to velocity \(n - 1\) occurs now through a cascade of rational velocities, defining a « Devil's staircase », structure which becomes evident through successive blow up of the \(V(\gamma)\) curve (Fig. 4). Note that for a potential \(U\) and a force \(\phi\) less singular than those described in (6) and (7), one generically expects as shown (Fig. 4d) a square-root singularity for the envelope of the staircase instead of the straight line (finite slope) displayed (Figs. 4b, c).

3. Temporally asymmetric force.

As explained in the introduction, breaking either (2) or (3) leads to the existence of an average drift. We here briefly consider the sole breaking of the temporal symmetry, keeping the spatial one \((a = b = 1/2)\). A typical phase diagram is shown in figure 5. The structure is fairly similar to that of figure 2:

a) for \(\gamma < 4\) \(\epsilon a\), the external force is never large enough to move the particle:

![Fig. 5. « Phase diagram » in the \((\gamma, \epsilon)\) plane for a symmetric potential and an asymmetric ac force \((a = 1/2, \epsilon = 1/4)\). Continuous lines describe the lower contour of isovelocity domains corresponding to velocities \(V = 1/4, 1/3, 1/2, 1, 2, 3\).](image-url)
b) for $\gamma > 4 \varepsilon_c$, if $\gamma < 4 \varepsilon_c + 1$ the particles oscillate around a given energy minimum but macroscopic motion is still absent, $V = 0$. If $4 \varepsilon_c + 1 < \gamma < 4 \varepsilon_c (1 - c)$ the particle can only move to the right and $V$ is an increasing function of $\gamma$. The situation is similar to the spatial asymmetry case. For $\gamma < 4 \varepsilon_c (1 - c) - 1$, only integer velocities $V = n$ are encountered (regime 1), appearing at a threshold given by:

$$1 = (n - 1) \left[ \frac{1}{\gamma - 4 \varepsilon_c} + \frac{1}{\gamma + 4 \varepsilon_c} \right] + \left[ \frac{1}{\gamma - 4 \varepsilon_c} \right]$$

(10)

whereas for $4 \varepsilon_c (1 - c) - 1 < \gamma < 4 \varepsilon_c (1 - c)$ rational velocities of the form $n - 1/(m + 1)$ appear (regime 2). Note that for $c < 1/2 - 1/(4 \varepsilon_c)$, the velocity jumps directly from 0 to 1 at $\gamma = 4 \varepsilon_c + 1$, but in the opposite case motion starts with rational velocity values.

c) Upon increasing $\gamma$ above $\varepsilon (1 - \epsilon)$, the velocity will start to decrease as larger and larger backward motion is allowed, and devil staircases may be encountered.

4. Combining both asymmetries.

In the general case of both temporal and spatial asymmetry the rectifying effects can either add up or subtract. This is most easily seen for large potential barriers: $\varepsilon \gg 1$. In this limit the excursions during one time period can be large compared to the spatial period and the general trends can be understood without paying attention to the exact rational nature of the velocity values: $V$ can be estimated as $V = n_1 - n_2$ in which $n_1(n_2)$ are the number of spatial periods visited during one forward (backward) step. Focusing furthermore on $\gamma = \varepsilon b$, we get the corresponding analytical variation for $V$ as a function of $c$ (we take $a \gg 2 b$):

$$V \approx (\varepsilon/2 b) \left[ \frac{(1 - 2 c b/a)(1 + 2 c)}{(1 + 2 c (1 - b/a))} - \theta \left( c - \frac{1}{2} \right) \frac{(1 + 2 c(1 - c)b/a)(2c - 1)}{(1 - 2)(1 - c)(1 - b/a))} \right]$$

(11)

where $\theta$ is the Heavyside step function (when $c < 1/2$, no backward motion is allowed). As $c$ is varied from 0 to 1, the velocity monotonously decreases from positive values ($\approx \varepsilon/2 b$ for $c \approx 0^+$) to negative ones ($\approx -2 \varepsilon/3 a$ for $c \approx 1^-$). For $c < 1/2$ the spatial and temporal rectification processes contribute in the same (forward) direction, whereas they oppose each other for $c > 1/2$. The velocity given by equation (11) cancels for $c = c_0$ with:

$$c_0 = \frac{a}{(b + (b^2 + 2a(a - b))^{1/2})}$$

(12)

The sole measure of the temporal asymmetry leading to cancellation provides a direct estimate of the spatial asymmetry in this regime. The velocity limiting values give the scale of the pinning energies.

When excursions are of order of one or a few periods, one has of course to keep track of the detailed motion. Again as $c$ increases, $V$ decreases and changes sign, but the discontinuous nature of the velocity changes gets clearly visible.

5. Influence of a small noise.

When the temperature is small but finite, noise has to be taken into account. For $\alpha \ll \varepsilon$, the particle can occasionally make a thermal hop from one well of the potential to one of its neighbours. the typical hopping rate following a Kramers-like formula: $(\varepsilon^2/\alpha) \exp(-\varepsilon/\alpha)$. If an external force characterized by $\gamma$ is applied, it will bias this hopping
motion, so that even at low values of \( \gamma \) a macroscopic velocity is obtained if asymmetry is present. Indeed for \( \gamma < \alpha < \epsilon \), we find:

\[
V = \frac{\epsilon}{\alpha^2} \exp \left( -\frac{\epsilon}{\alpha} \right) \left[ \frac{a^2 - b^2}{4 \epsilon (1 - \epsilon)} \left( \frac{\gamma}{\alpha} \right)^2 - \frac{a^2 + b^2}{8 \epsilon^2 (1 - \epsilon)^2} (1 - 2 \epsilon) \left( \frac{\gamma}{\alpha} \right)^3 + \cdots \right] \tag{13}
\]

which is non-zero in the case of a spatial (temporal) asymmetry owing to the first (second) term of the r.h.s. The low frequency behaviour at finite temperature was also analyzed in [4] for a spatial asymmetry.

Note that an experimental study in this regime can lead to both a measure of the anisotropy \((a - b)\) via the cancellation point, and a measure of the pinning energy compared to \(kT\).

More generally, the singularities of the \(V(\epsilon, \gamma)\) diagram are progressively washed out as \(\alpha\) is increased (Fig. 3).


Although rectification processes have been known for a long time, we show in this analysis that they take interesting characteristics in the periodic structures we consider in this article. They could lead to a new generation of separation techniques, and to new tools to probe and analyze asymmetric systems such as motor protein assemblies.

Indeed, most separation techniques up to now rest upon the use of an external continuous field, that induces the migration of particles at a speed which characterizes them. There are only a few exceptions to this general scheme [7]. Separation techniques such as Force Flux Fractionation (FFF) [8] could be renewed by the use of asymmetric structures and alternating fields. Let us just give here two examples: in one of the versions of FFF, small non brownian particles are set into motion, by either a hydrodynamic flow or a d.c. electric field, parallel to a homogeneous horizontal wall on which they are confined by gravity. Surfaces such as blazed gratings and alternating electric fields could be used, allowing for the selection of monodisperse particles. This technique would work well for particles in the 10-100 \(\mu\m\) range. A second example is adapted to the selection of polyelectrolytes of a given molar mass. Currently, one of the processes uses FFF in a mode where the confinement is provided by a temperature gradient (Soret effect) and the driving field is of electric origin. Again, selection would be greatly enhanced by the use of a blazed grating type of surface and ac electric fields. In a similar way, symmetric structures such as zeolites, driven by appropriate asymmetric electric field sequences, could be used as selective ionic pumps. Note that the difference in electrochemical potential for a given ion, which could be forced across such a structure, would be a fraction of the applied alternating voltage times the charge: this number can be large compared to the thermal energy \(kT = 1.40\) eV even with reasonable voltages. Therefore such pumps should be extremely efficient in maintaining sizeable concentration differences.

Let us finally point out that motor protein assemblies have precisely the symmetry considered in this article. These assemblies are part of the cytoskeleton and allow efficient directed transport in the cytoplasm along a network of filaments. Each of these filaments is a linear assembly of tubulin molecules which possesses an intrinsic local polarity (broken symmetry). Motor proteins (kinesins or dyneins) "walk" along them in a directed way when fed with Adenosine Triphosphate (ATP). Different models have been recently proposed to describe the motion generation [3-6], making use of the fact that the "adsorption" potential that a tubulin filament exerts on a dynein or kinesin molecule is flat on average but with inherent local asymmetry. Rather than trying to provide clues as to the motor activity, we here propose to use external alternating fields in motility assays [9, 10] to investigate the main
characteristics of the adsorption potential. Varying amplitude, frequency and asymmetry of the driving ac field should allow us to get information about the amplitude, asymmetry and shape of this potential, but also about the friction of the molecules on the filament.

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References

[7] Pulsed fields methods are widely used for separation of large DNA fragments, see e.g. Carle G. F., Olson M. V., Methods in Enzymology 155 (1987) 468-482.