

Brownian Refrigerator

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Onsager symmetry implies that a Brownian motor, driven by a temperature gradient, will also perform a refrigerator function upon loading. We analytically calculate the corresponding heat flow for an exactly solvable microscopic model and compare it with molecular dynamics simulations.

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Cooling techniques have a significant impact not only on our everyday life but also on the advances in science. We have come a long way from the primitive method of evaporative cooling, which our body conveniently uses when we perspire, over the evaporation by expansion of a cooling liquid in domestic refrigerators, to high-tech methods including laser cooling, magnetic cooling, radiative cooling, or quantum cooling. Temperature is a measure of thermal fluctuations, but the latter are not directly observable in macroscopic systems. Recent advances in nanotechnology and molecular biology, however, allow us to manipulate and even manufacture constructions on a molecular scale, where thermal fluctuations can no longer be ignored. To run such machines, we could copy the mode of operation from their macroscopic counterparts. An alternative, and arguably more promising approach, would be to utilize thermal fluctuations rather than fighting them. A well-documented example is the Brownian motor [1,2], which generates power through the rectification of thermal fluctuations. In this Letter, we present a novel method of microscopic cooling based on a Brownian motor in which, almost paradoxically, thermal fluctuations themselves can be harnessed to reduce the thermal jitter in one part of the system.

Our Brownian motor [3] (see Fig. 1) consists of two parts, a triangle and a flat paddle, which are rigidly linked and move as a single entity along horizontal tracks. Its motion is induced, following Newton's laws, by the random collisions with the particles of the gas in which it is embedded. Because of the random nature of these collisions, one expects a resulting rather erratic motion of the motor. The question of interest is whether, due to the asymmetry of the triangle, this random motion is characterized by a nonzero average speed in a given direction. When both motor units reside in a single compartment with a gas (or liquid) at equilibrium, we argue that such a sustained motion is impossible because it would violate the second law of thermodynamics. The construction would be an *in perpetuum mobile* of the second kind (or Maxwell demon), because the motion could be used, for

example, to lift a weight, hence extracting work out of the single heat bath. This impossibility can be understood more deeply from the fact that a system at equilibrium has a perfect time-reversal symmetry. Any movie of such a system, or of any subpart of the system (like monitoring the motion of our motor), is statistically indistinguishable when played forward or backward in time. Hence any form of systematic translation is impossible.

Having concluded that no sustained motion occurs at equilibrium, we modify the construction by inserting a perfectly insulating wall between both motor units while not hampering their joint motion as illustrated in Fig. 1(b). A nonequilibrium state is now easily realized by assuming that the particles in the separate lower and upper compartments are at a *different* temperature, respectively, T_1 and T_2 . To intuitively understand what happens in this case, we first consider the extreme case $T_1 > T_2 = 0$, corresponding to fully stationary particles (no thermal motion) in reservoir 2 (upper reservoir with the triangle unit). When a particle hits the paddle in reservoir 1, it transfers horizontal momentum to the motor. It will do so in random quantities and equally so from the left as from the right. This momentum will be dissipated as the triangle hits the stationary particles in reservoir 2. However, it is clear that the result-

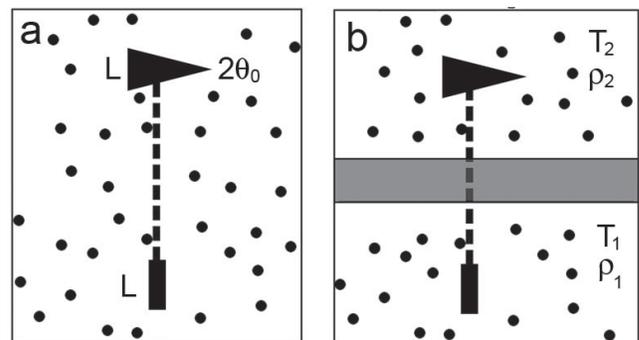


FIG. 1. An asymmetric object on horizontal tracks. (a) Performs Brownian motion with no average systematic drift when in thermal equilibrium. (b) Sustained average systematic motion when subject to a thermal gradient.

ing slowdown will be much stronger when the motion of the motor is towards the left, with the flat face of the triangle transferring large momentum to the stationary particles. When the motion is towards the right, the inclined sides of the triangle transfer less momentum allowing it to move farther, in the same way as a sharp arrow can penetrate deeper. We conclude that an average systematic motion is expected pointing to the right in the direction of the sharp angle.

The above handwaving arguments are confirmed by an analytic calculation, which is furthermore exact in the limit of dilute gases [3]. The average velocity of the motor is found to be

$$\bar{V} = \rho_1 \rho_2 (1 - \sin^2 \theta_0) \times \sqrt{\frac{m}{M}} \sqrt{\frac{\pi k_B}{2M}} \frac{(T_1 - T_2) \sqrt{T_1}}{[2\rho_1 \sqrt{T_1} + \rho_2 \sqrt{T_2} (1 + \sin \theta_0)]^2}. \quad (1)$$

Here m and M are the mass of gas particles and motor, respectively, and $M \gg m$ is assumed. The densities of gas particles in reservoirs 1 and 2, ρ_1 and ρ_2 , are assumed to be small. The shape of the motor units is defined by the vertical cross section L and the half apex angle of the triangle θ_0 [see Fig. 1(a)]. In agreement with the above discussion, we find for $T_1 > T_2$ a positive average velocity, i.e., a velocity to the right. At equilibrium, $T_1 = T_2$, the velocity is zero (and also for $\theta_0 = \pi/2$, in agreement with the fact that the triangle reduces to a flat paddle without preferred direction). Furthermore, note that equilibrium is a point of flow reversal: for $T_1 < T_2$, the motor has a negative velocity. Somewhat surprisingly, it then moves towards the left, i.e., in a direction opposite to the sharp angle.

The transition from a Brownian motor to a Brownian refrigerator can be made by invoking a general stability principle that appears in several branches of physics under different names: Newton's action-reaction law in mechanics, Lenz's law of magnetoinduction in electromagnetism, and, relevant to the problem under consideration here, the Le Chatelier–Braun principle in thermodynamics. The latter states that an action on a system at equilibrium induces processes that attenuate or counteract the original perturbation. This principle of negative feedback is clearly an expression of stability of the original equilibrium state. Now, consider again the above construction, but at equilibrium with $T_1 = T_2$. We perturb this state by applying an external force $F < 0$ on the motor, moving it to the left. According to the above principle, we expect that the system reacts by a process that induces an opposing motion to the right. In the above discussion we have identified such a process: we need to increase the temperature in reservoir 1 with respect to reservoir 2. This entails the appearance of a heat flow from reservoir 2 to reservoir 1, cooling down reservoir 2. Similarly, an opposite heat flow from reservoir 1 to reservoir 2 will arise when applying a force $F > 0$.

The above discussion may appear wishful and reveals neither the mechanism nor the amplitude of the process by which this miraculous heat flow appears. Fortunately, a simple but basic argument from the theory of linear irreversible thermodynamics [4] confirms the prediction and allows us to evaluate the strength of the phenomenon. We note that the Brownian motor, described above, is the result of a cross effect: a thermal gradient $\Delta T = T_1 - T_2$ produces an average motor velocity \bar{V} . In irreversible thermodynamics, one calls $J_1 = \bar{V}$ a flow and $X_2 = 1/T_2 - 1/T_1$ a thermodynamic force [5]. Since the flow is zero at equilibrium, i.e., in the absence of the thermodynamic force, we expect to find, for a small temperature difference ΔT , $T_1 = T + \Delta T/2$, $T_2 = T - \Delta T/2$, a linear relation between flow J_1 and force $X_2 = \Delta T/T^2$, namely, $J_1 = L_{12} X_2$. The value of the coefficient L_{12} is found from Eq. (1):

$$L_{12} = \rho_1 \rho_2 (1 - \sin^2 \theta_0) \times \sqrt{\frac{m}{M}} \sqrt{\frac{\pi k_B}{2M}} \frac{T^{3/2}}{[2\rho_1 + \rho_2 (1 + \sin \theta_0)]^2}. \quad (2)$$

Cross processes are no exception. Well-documented cases are the Seebeck, Thomson, and Peltier effects, where temperature differences induce electrical currents or vice versa. The important observation in our discussion is that Onsager [6] has identified a profound symmetry relation between any cross process and its mirror process, deriving from the time-reversal symmetry of underlying microscopic dynamics. More precisely, for any relation $J_1 = L_{12} X_2$, there is a mirror relation $J_2 = L_{21} X_1$ with an identical proportionality coefficient $L_{21} = L_{12}$. In our case, J_2 is the flow associated with the temperature gradient X_2 ; i.e., it is a heat flow $\dot{Q}_{1 \rightarrow 2}$ from reservoir 1 to reservoir 2. The thermodynamic force associated with the particle flow J_1 is a difference in chemical potential. In our setting, the latter is produced by the application of a mechanical potential generating a constant force F . More precisely, one has $X_1 = F/T$. The relation $J_2 = L_{21} X_1$, with Eq. (2), implies that the heat flow $J_2 = \dot{Q}_{1 \rightarrow 2}$ is given by

$$\dot{Q}_{1 \rightarrow 2} = \rho_1 \rho_2 (1 - \sin^2 \theta_0) \times \sqrt{\frac{m}{M}} \sqrt{\frac{\pi k_B T}{2M}} \frac{F}{[2\rho_1 + \rho_2 (1 + \sin \theta_0)]^2}. \quad (3)$$

This heat flow is quite large, namely, of the order of the power exchanged with the external force (thermal speed of the motor, $\sqrt{k_B T/M}$, times F) multiplied by $\sqrt{m/M}$. Furthermore, in agreement with our foregoing discussion based on stability principles, the direction of heat transfer depends on the direction of the force F , in such a way that it activates an opposing Brownian motor.

While heat is transferred from one reservoir to another, we have to realize that the motion induced by the force will have another quite familiar effect, namely, dissipation by

the frictional force $\gamma\bar{V}$, where γ is a frictional coefficient. The dissipation will heat up both reservoirs and thus it may forestall the cooling. This ‘‘Joule’’ energy stems from the power input $P = F\bar{V}$. But with the velocity itself proportional to the applied force $\bar{V} = F/\gamma$, the dissipation is proportional to F^2 . The quadratic law conforms with the fact that the dissipation is independent of the direction of the applied force. We conclude that the cooling effect is dominant for small forces. The application of such forces along with the use of either small motor molecules (M small) or larger substrate particles (m large) will induce a significant cooling effect in one of the compartments.

All the above results, the explicit value of the Onsager coefficients and, in particular, Onsager symmetry, are confirmed by a direct microscopic calculation. The starting point for this derivation is the following Boltzmann-Master equation for the probability distribution $P(V, t)$ for the speed of the motor:

$$\partial_t P(V, t) = -\frac{F}{M} \frac{\partial}{\partial V} P(V, t) + \int dV' [W(V|V')P(V', t) - W(V'|V)P(V, t)], \quad (4)$$

where $W(V|V')$ is a transition probability per unit time for the motor to change velocity from V' to V due to the collisions with the gas particles. This approach is exact in the limit of dilute gases. Analytic expressions of the transition probability for general convex objects are available for the dilute gases [3].

Expanding Eq. (4) in Taylor series up to second order in $\sqrt{m/M}$, we find the following explicit result (to linear order in F and ΔT):

$$J_1 = L_{11}X_1 + L_{12}X_2, \quad J_2 = L_{21}X_1 + L_{22}X_2, \quad (5)$$

with J_1, J_2, X_1 , and X_2 as defined before, and

$$L_{11} = \frac{T}{\gamma}, \quad L_{22} = \frac{k_B \gamma_1 \gamma_2 T^2}{M \gamma}. \quad (6)$$

The friction coefficients in each reservoir are given by

$$\gamma_1 = 8\rho_1 L \sqrt{\frac{k_B T m}{2\pi}}, \quad \gamma_2 = 4\rho_2 L \sqrt{\frac{k_B T m}{2\pi}} (1 + \sin\theta_0), \quad (7)$$

and the total friction by $\gamma = \gamma_1 + \gamma_2$. L_{12} is as given in Eq. (2) and Onsager symmetry, $L_{12} = L_{21}$, is confirmed.

The above result also supplies us with the explicit expression for Joule heating in each reservoir:

$$\dot{Q}_{J_i} = \gamma_i \bar{V}^2 = \gamma_i F^2 / \gamma^2 \quad (i = 1, 2). \quad (8)$$

The ratio of the heat transferred from reservoir 2 to reservoir 1 ($\dot{Q}_{2 \rightarrow 1} = -\dot{Q}_{1 \rightarrow 2}$) over the Joule heat dissipated in reservoir 2 is thus found to be

$$\frac{\dot{Q}_{2 \rightarrow 1}}{\dot{Q}_{J_2}} = -2(1 - \sin\theta_0) k_B T \frac{m}{M} \frac{L \rho_1}{F}. \quad (9)$$

If this ratio is larger than 1, reservoir 2 will cool down. The following condition ensues in terms of the applied small (negative) force:

$$0 > F > -2(1 - \sin\theta_0) k_B T \frac{m}{M} L \rho_1. \quad (10)$$

A similar condition applies for the range of small positive forces, inducing cooling in reservoir 1.

Besides the issue of Joule heating, we also need to address the fact that the Brownian motor itself conducts heat [7]. From Eqs. (5) and (6) we obtain the Fourier law, $L_{22}X_2 = \kappa\Delta T$ with $\kappa = L_{22}/T^2$. Assuming there is no other conductivity leak involved, one concludes from Eqs. (2), (5), and (6) that total heat flow ($J_2 = 0$) will vanish at a relative temperature gradient $\Delta T/T$ of the order of the ratio of the drift speed F/γ of the motor over the thermal speed $\sqrt{k_B T/m}$ of the gas particles. Note that this result is independent of the mass M of the motor due to the fact that both heat currents, originating from thermal fluctuations, share the same $1/M$ dependence.

To test the above predictions in a more realistic model, we have performed extensive two-dimensional hard disk molecular dynamics simulations (diameter $\sigma = 1$). The system comprises two separate rectangles (size = 1200×300), with periodic boundary conditions in both directions, each containing 3600 hard disks (density $\rho_1 = \rho_2 = 0.01$). The motor unit is as represented in Fig. 1 ($L = 10$ and $\theta_0 = 10^\circ$). Averages are taken over 30 000 realizations. In Fig. 2, we show the evolution of the temperatures in both reservoirs, starting with $T_1 = T_2 = 1$ (dimensionless units $k_B = m = 1$ and $M = 2$ are used). We apply a weak force $F = -0.007$ to the motor. The cooling effect in reservoir 2 is clearly observed. To compare the theoretical

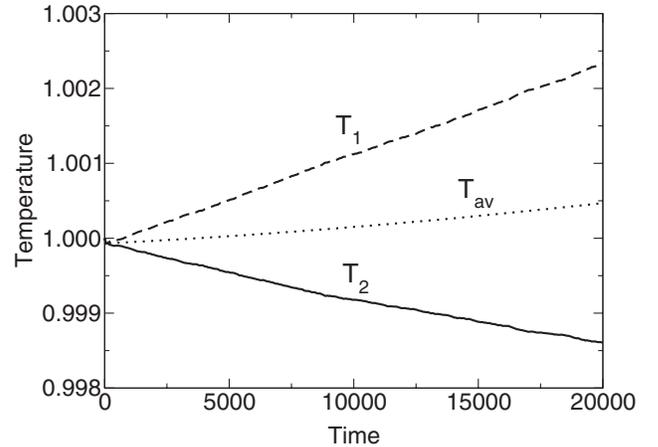


FIG. 2. The motor of mass $M = 2m$ is driven by a constant force satisfying Eq. (10), namely, $F = -0.007$. The temperature of two reservoirs, T_1 (dashed line) and T_2 (solid line), are initially equal to 1. Clearly the temperature of reservoir 2 (T_2) is decreasing, while reservoir 1 is heating up. Note that the average temperature $T_{av} = (T_1 + T_2)/2$ increases due to the Joule heat dissipated in both reservoirs.

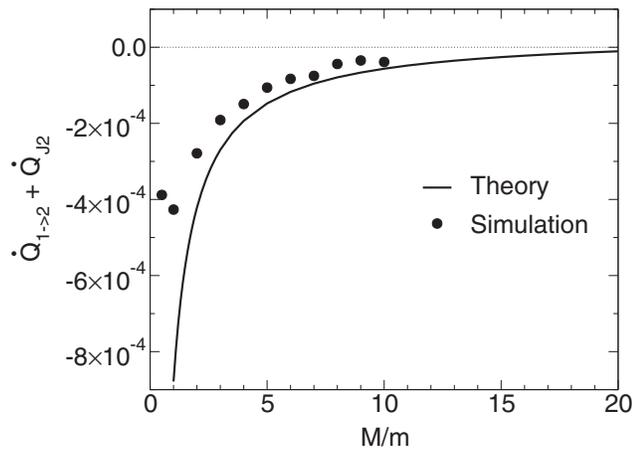


FIG. 3. The rate of energy change in reservoir 2 as a function of the mass of the motor upon application of a force $F = -0.007$. The energy change encompasses both the heat transfer from reservoir 2 into reservoir 1 as well as the Joule heating in reservoir 2. Cooling is clearly observed.

results with the simulations, we plot in Fig. 3 the net heat flow injected into reservoir 2, $\dot{Q}_{1 \rightarrow 2} + \dot{Q}_{J2}$. The agreement between theory and simulation is, as expected, very good for large values of M/m , but even for small values of M/m the order of magnitude is correctly predicted by the theory.

Moving beyond the academic discussion, we need to address a number of issues of more experimental or technological relevance. Under which conditions could the above discussed heat flow be observed or applied? The explicit result given in Eq. (3) was derived for an ideal gas. Molecular dynamics carried out at larger densities, however, show that the order of magnitude of the heat flow remains the same. We can therefore use the above result to estimate the effect, even when we have in mind motors operating in an aqueous or a lipid environment. Consider a macromolecule embedded in lipid bilayers, under physiological conditions, $T \approx 300$ K. For a typical value of its mass $M = 10^5 m$ (m being typically the mass of a water molecule) and for a force of 0.1 pN, Eq. (3) predicts ($\rho_1 = \rho_2$) a heat flow of the order of 5×10^{-17} J/s. On the other hand, using a typical value for the drag coefficient of a protein $\gamma \sim 100$ pN \cdot s/m, the Joule heat is found to be of the same order, and both heat effects are comparable. Hence, a negative net heat flow of this order of magnitude will appear for F slightly smaller or for M slightly larger. Such a flow will cool down an aqueous reservoir of diameter 0.1 μm by 1° in about 1 min.

We finally note that an alternative construction with rotational instead of translational motion of the motor would appear easier to realize. One can imagine a chiral molecule protruding on both sides of a membrane that is

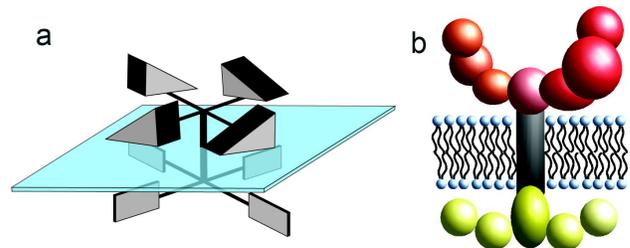


FIG. 4 (color online). A rotational version of the proposed refrigerator. (a) A chiral rotor is placed across an insulating wall. (b) A chiral biological molecule will channel heat across a membrane when a small external torque is applied.

set into rotational motion by some polarizing force; see Fig. 4. As we have revealed above, such a construction could, in principle, function as a Brownian refrigerator, cooling down one side and heating up the other. Putting such constructions in parallel would amplify the heat current. Putting them in series would allow one to channel heat out of the system following a predesigned pathway. Aside from the possible application in molecular biology, the aforementioned advances in nanotechnology should allow one to construct carefully designed devices of the type described above. As a final comment, we stress that the Le Chatelier–Braun principle implies the appearance of a Brownian refrigerator under very general conditions, whenever an opposing thermal Brownian motor can be activated in response to an applied external or thermodynamic force.

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