Stochastic thermodynamics of non-harmonic oscillators in high vacuum

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We perform an analytical study on the stochastic thermodynamics of a small classical particle trapped in a time dependent single-well potential in the highly underdamped limit. It is shown that the nonequilibrium probability density function for the system’s energy is a Maxwell-Boltzmann distribution (as in equilibrium) with a closed form time dependent effective temperature and fractional degrees of freedom. We also find that the solvable model satisfies the Crooks fluctuation theorem, as it is expected. Moreover, we compute the average work in this isothermal process and characterize the optimal protocol for minimum work analytically. Surprisingly, the optimal protocol presents an initial and a final jumps which correspond to adiabatic processes linked by a smooth exponential time dependent part for all kinds of single-well potentials. Furthermore, we argue that this result connects two distinct relevant experimental setups for trapped nano-particles: the levitated particle in a harmonic trap, and the free particle in a box; as they are limiting cases of the general single-well potential and display the time-dependent optimal protocols. Finally, we highlight the connection between our system and an equivalent model of a gas of Brownian particles.

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Modeling nonequilibrium physics of very small systems requires the definition of thermodynamic quantities such as heat and work for single particle trajectories. Since the study of the original Brownian motion, stochastic thermodynamics has further evolved with the development of optical tweezers, which allowed precise trapping and cooling procedures for levitated nanoparticles and started the field of optomechanics. Applications include the experimental verification of FT in biophysics at molecular level, apparent violation of the second law of thermodynamics and evidence of Landauer’s principle. More recently, in contrast to suspended particles, experimental groups obtained measurements of particles optically trapped in high vacuum and verified several nonequilibrium results, including the ubiquitous fluctuation theorem. This experimental frontier might provide an interface to test quantum limits, non-Newtonian and quantum gravity, and the realization of feasible underdamped nanomechanical heat engines.

In this context, research is devoted to design and implement efficient thermal engines at micro- and nanoscale, which includes optimal protocols for producing minimum average work over finite time windows. In optical traps, a protocol might be obtained adjusting the laser trap frequency, which is equivalent to change the stiffness of the restoring force. How the frequency is tuned over a finite time window produces a variety of nonequilibrium thermodynamic processes, resulting in fluctuating work and heat. In this sense, optimal protocols for a time-dependent driving force in overdamped systems have been found to exhibit discontinuous jumps, where the trap frequency is required to change abruptly, followed by a smooth tuning. The same interesting feature was observed in underdamped models for optical traps in numerical simulation and also in analytical calculations. Besides the adiabatic jumps, the continuous part of the optimal protocols in such systems is exponential in time. Surprisingly, the same exponential protocol was also obtained for a free particle in a box using different methods. How general is this exponential behavior and the presence of jumps in optimal protocols is a question that remains open in stochastic thermodynamics.

In this work, we propose a solvable model for a levitated particle in the highly underdamped limit for a general single-well potential of the type $U(x) \propto x^{2n}$. Our result, which is of special experimental interest, generalizes previous results limited to the harmonic case ($n = 1$) and the particle in a box ($n \rightarrow \infty$), as well as consider finite time processes in the presence of damping, expanding previous analysis of single-well potentials. We show the model has a closed form propagator for the stochastic energy and simple expressions for averaged thermodynamic quantities. Remarkably, we find the nonequilibrium probability distribution for the energy in a isothermal process is a Maxwell-Boltzmann (MB) distribution with a time dependent effective temperature. As an application, we study the optimal protocol that produce minimum work in a isothermal process and find finite jumps in the driving protocol, whose magnitude depends on $n$, combined with a time-dependent exponential relaxation for all $n$.

The model—We consider the dynamics of a small Brownian oscillator submitted to the external potential $U(x, k_t) = k_t x^{2n}/2n$, where $k_t$ is a time-dependent generalized stiffness which can be varied by tuning the frequency of the trapping setup. Here we introduce param-
ets $k_t = m\Omega_t^2 L^{2-2n}$, where $\Omega_t$ has units of frequency (s$^{-1}$) and $L$ is a characteristic length. The particle is in contact with a thermal reservoir of temperature $T$ and it is modelled using the Langevin equation

$$\ddot{x} + \Gamma_0 \dot{x} + \Omega_t^2 L \frac{x}{L} = \frac{1}{m} F_{\text{fluc}}(t),$$

(1)

for the particle position $x(t)$. The random Langevin force $F_{\text{fluc}}(t)$ is normally distributed with mean zero and its components satisfy $\langle F_{\text{fluc}}(t) F_{\text{fluc}}(t') \rangle = 2m \Gamma_0 k_B T \delta(t - t')$, where $\Gamma_0$ is a friction coefficient and $m$ is the particle mass. Considering the usual scale of observation and the suspension media involved, the system is commonly solved in the overdamped limit [3], where the inertia term is neglected, $m\dot{x}(t) \approx 0$. For levitated particles in highly diluted media [3] [4], although the mass of the particles are small, this regime imposes a small friction coefficient $\Gamma_0$ compared to the frequency $\Omega_t$, so inertial effects cannot be neglected. Here we show that using this last condition in [1] results in a solvable stochastic differential equation (SDE) for the system’s total energy given by $E(x, p) = p^2/2m + k_t x^{2n}/2n$, with momentum $p = m\dot{x}$. After a suitable combination of Itô’s Lemma [30] and the highly underdamped limit ($\Omega_t \gg \Gamma_0$) [14] [38], the energy SDE is given by

$$dE = dW - \Gamma_0 (E - \frac{f_n}{2} k_B T) dt + \sqrt{2 \Gamma_0 k_B T E} dW_t,$$

(2)

where $dW = (\partial U(x, k)/\partial k) \dot{k}_t dt$ is the increment of work [3], $dW_t$ is the increment of the Wiener process with a redefined damping constant $\Gamma_0 = (2n/(n+1)) \Gamma_0$ and it presents an effective fractional degree of freedom $f_n = (n+1)/n$. If $dW = 0$, the energy increment [2] gives us only the heat contribution. The increment of work for a particle trapped in a single-well potential is given by

$$dW = \frac{\partial U(k_t, x)}{\partial k_t} \frac{k_t}{k_t} dt = U(x, k_t) \frac{k_t}{k_t} dt,$$

(3)

where terms were rearranged in the last expression to display $U(x, k_t)$ explicitly. We consider a high frequency protocol, $\Omega_t$, that does not change the frequency appreciably during an oscillation period; i. e., $\Omega_t \delta t/\Omega_t = \Omega_t^2/\Omega_t^2 \ll 1$. By applying the highly underdamped limit to [3], we find an approximation for $dW$ by integrating the work increment over a small time interval, $\delta t$, such as $\Gamma_0^{-1} \gg \delta t = \Omega_t^{-1}$ for all $t$. In this small time interval, since dissipation and fluctuation may be neglected in [1], the particle behaves in the absence of dissipation. When averaged over a oscillation period, its potential energy is related to the mechanical energy by the virial theorem, (1/$\delta t$) $\int \dot{U}(\lambda_0, x) dt = E/(n+1)$, as a generalization of the approximations previously used for the harmonic case ($n = 1$) [11] [20] [38]. Therefore, we may integrate [6] over this small time interval, $\delta t$, and find

$$\delta W = \int_t^{t+\delta t} dW \approx \frac{k_t}{k_t} \int_t^{t+\delta t} U(x, k_t) dt = \frac{k_t}{k_t} E \frac{\delta t}{n + 1}.$$

(4)

Since the time interval, $\delta t$, is small, we rewrite the increments as differentials, $\delta W \rightarrow dW$ and $\delta t \rightarrow dt$. Finally, we replace [4] in [2] and obtain the final SDE for the energy

$$dE = -\Gamma_0 ((1 - \frac{\lambda_t}{2\lambda_0 \Gamma_0}) E - \frac{f_n}{2} k_B T) dt + \sqrt{2 \Gamma_0 k_B T E} dW_t,$$

(5)

where $\lambda_t = k_t^{2/(n+1)}$. We have omitted the index $n$ in $\lambda_t$ for clarity. It is worth noting that the SDE is identical for all potentials $n = 1, 2, \ldots$ in terms of the adjusted parameters $\Gamma_0, f_n$ and $\lambda_t$, which makes the stochastic thermodynamics of all single-well potentials equivalent in the highly underdamped limit.

Several applications emerge from [5] depending on the potential $n$ and the protocol $\lambda_t$. Notice that, in the case $n = 1$ (harmonic potential), the SDE [5] reduces to the levitated particle trapped by a laser in the highly underdamped limit [20]. We will also argue that the case $n \rightarrow \infty$ models the particle in a box [31]. Notably, intermediate cases ($1 < n < \infty$) are modeled by fractional degrees of freedom, $1 < f_n < 2$. In any case, for a constant protocol $\lambda_t = \lambda_0$ and $\lambda_t = 0$, the SDE describes the heat exchanged for a isochoric process ($dW = 0$), where the heat distribution PDF has a closed form that resembles the PDF obtained in similarly to the case $n = 1$ [38]. For completeness, we also show from [2] that the approximate work [4] satisfies the Crooks fluctuation theorem (CFT) [39] [40] for any $n$ (see the Supplementary Material).

More importantly, the propagator of the probability density function (PDF) of the energy $P_t(E|E_0)$ for [5] can be found explicitly by solving its underlying Fokker-Planck equation (see Sup. Mat.). We obtain the nonequilibrium conditional PDF for the energy propagator

$$P_t(E|E_0) = \alpha_t C_t e^{-C_t (\alpha_t E + E_0 e^{-\Gamma_0 t})} \left( \frac{\alpha_t E}{E_0 e^{-\Gamma_0 t}} \right)^{q/2} \times$$

$$I_q(2C_t \sqrt{\alpha_t E_0 e^{-\Gamma_0 t}}),$$

(6)

for $E \geq 0$ and $E_0 \geq 0$, where $q = f_n/2 - 1$, $\alpha_t = (\lambda_0/\lambda_t)^{1/2}$, $C_t = \exp(\Gamma_0 t)/(\Gamma_0 k_B T m)$, $m_t = \int_0^t \exp(\Gamma_0 u) \sqrt{\lambda_0/\lambda_t} du$ and $I_q$ is the modified Bessel function of the first kind [41]. As a straightforward application of [6], we may suppose the particle is initially found in equilibrium with a reservoir of temperature $T_0$. In this case, the initial PDF for the energy is Maxwell-Boltzmann (MB), $P_0(E) = \frac{\beta_0}{E_0} E_0^{q/2} e^{-\beta_0 E_0}$, with $\beta_0 = (k_B T_0)^{-1}$. For $t > 0$, the system undergoes a protocol $\lambda_t$ in thermal contact with a reservoir at temperature $T$ for a finite time interval $[0, t]$. In this case, one may write the nonequilibrium energy distribution as the superposition of Eq. [6] over the initial conditions (MB), using $P_t(E) = \int_0^\infty P_t(E|E_0) P_0(E_0) dE_0$, which results in another MB distribution, $P_t(E) = \beta_t^{1+q}/T(q + 1) E^{q/2} e^{-\beta_t E}$, with
\[ \beta_t = (k_B T_t)^{-1}, \] for a time dependent effective temperature \( T_t \) given by
\[ T_t = e^{-\Gamma_n t} \left( \frac{\lambda_1}{\lambda_0} T_0 + \Gamma_n \int_0^t e^{\Gamma_n u} \sqrt{\frac{\lambda_0}{\lambda_u}} du \right). \] (7)

Depending on the protocol \( \lambda_t \), the effective temperature takes different forms.

For example, it is immediate to check that the constant protocol, \( \lambda_t = \lambda_0 \) for all \( t \), leads to the effective temperature \( T_t = T + (T_0 - T)e^{-\Gamma_n t} \), which represents the thermal relaxation of a system initially prepared at temperature \( T_0 \) and placed in thermal contact with a reservoir at temperature \( T \). Another example is the adiabatic protocol, with \( \lambda_t \) going from \( \lambda_0 \) to \( \lambda_\tau \) in a very brief time interval, \( \tau \Gamma_n \ll 1 \). In this adiabatic case, the effective temperature (7) results in the relation \( T_t/T = \sqrt{\lambda_t/\lambda_0} \) which can be directly related to the polytropic equation, \( T_0 V_0^{2/\gamma} = T_v V_v^{2/\gamma} \), when associating an effective volume \( V \propto \lambda^{-\gamma/\lambda} \), already obtained from previous nonequilibrium approaches \[42\] for the case \( n = 1 \). General isothermal protocols result in nontrivial time dependent effective temperatures (7). The average work in such cases can be deduced as follows. First, notice that it follows from the nonequilibrium MB distribution, \( P_t(E) \), that \( \langle E(t) \rangle = \langle f_n/2 \rangle k_B T_t \). The expression for \( \langle E(t) \rangle \) is useful for calculating the ensemble average of the work increment, \( \langle dW \rangle \), from (4). Therefore, we find the average work, \( \langle W_\tau \rangle \), over the interval \( [0, \tau] \), taking the ensemble average of (4) and integrating in time,
\[ \langle W_\tau \rangle = \int_0^\tau \langle dW \rangle = \frac{f_n}{2} k_B T_t \int_0^\tau \frac{\lambda_1 T_t}{T} dt. \] (8)

For completeness, the average heat is expressed in a similar form:
\[ \langle Q_\tau \rangle = -\frac{f_n}{2} \Gamma_n k_B T \int_0^\tau \left( \frac{T_t}{T} - 1 \right) dt. \] (9)

Equations (7), (8) and (9) represent the main algorithm for calculating the average values of work and heat in any given isothermal process. In summary, for a given protocol \( \lambda_t \), one finds the effective temperature using (7) and use it to compute the averages work (8) and heat (9).

Optional protocol— We are interested in the protocol that minimizes the average work \( \langle W_\tau \rangle \), with boundary conditions \( \lambda_0 \) at \( t = 0 \) and \( \lambda_\tau \) at \( t = \tau \), for a system initially in equilibrium with the reservoir \( T_0 = T \). For simplicity, take \( \lambda_\tau > \lambda_0 \). This is the nonequilibrium analogue of a isothermal compression. Replacing the effective temperature (7) with \( T_0 = T \) in (8), we obtain a functional of the protocol \( \lambda_t \). The optimization is not trivial, because \( T_t \) depends on the integral of \( \lambda_t \), as noticed for the harmonic case \( n = 1 \) for different boundary conditions \[29\]. We are able to rewrite the work functional (8) as
\[ \langle W_\tau \rangle = \frac{f_n}{2} k_B T \int_0^\tau \left( 1 - \frac{\dot{h}_t}{\Gamma_0 \dot{h}_t} \right) \Gamma_n \dot{h}_t \Delta_t \Gamma_\tau dt, \] (10)

with the auxiliary variable \( h_t = 1+\Gamma_n \int_0^t e^{\Gamma_n u} \sqrt{\lambda_0/\lambda_u} du \), with boundary conditions \( h_0 = 1 \), \( \dot{h}_0 = \dot{h}_0 \), and \( \dot{h}_\tau = \Gamma_n e^{\Gamma_n \tau} \sqrt{\lambda_0/\lambda_\tau} \). Using Euler-Lagrange (EL) equations to minimize (10), one obtains \( \dot{h}h = \dot{h}^2 \), with solution \( h_t = Ae^{bt} \), for constants \( A, b \). But the solution fails to satisfy all boundary conditions for finite \( \tau \). This situation of broken extrema prevents the optimal protocol to be smooth everywhere \[13\]. It suggests the optimal protocol should include discontinuous jumps in the values of \( \lambda = t = 0 \) and \( t = \tau \). For simplicity, take \( \lambda_\tau \) constant protocol, \( \lambda_\tau \), as noticed for the harmonic case \( n = 1 \) for different boundary conditions \[12\]. In our setup, we find the optimal protocol starts with a jump \( \lambda_0 \rightarrow \lambda_1 \), followed by a smooth exponential part,
\[ \lambda_t = \lambda_1 (\lambda_2/\lambda_1)^{\lambda_\tau}, \] (11)
in close analogy to the particle in a box \[31\]. Finally, a second jump takes place, \( \lambda_2 \rightarrow \lambda_\tau \). Both jumps are related from the the boundary conditions of the problem,
\[ 1 - \frac{1}{\Gamma_n \tau} \log \sqrt{\frac{\lambda_2}{\lambda_1}} = \sqrt{\frac{\lambda_0}{\lambda_1}}, \] (12)

and the average work (8) can be written in terms of \( \lambda_1 \) as follows
\[ \frac{\langle W_\tau \rangle}{\langle f_n/2 \rangle k_B T} = \left( \Gamma_n \tau \sqrt{\frac{\lambda_1}{\lambda_0} - 1} + \sqrt{\frac{\lambda_0}{\lambda_1}} \Gamma_n \tau \sqrt{\frac{\lambda_0}{\lambda_1}} - 1 \right), \] (13)

which now can be minimized with respect to \( \lambda_1 \), leading to the closed form expression for the jump
\[ \frac{\lambda_1}{\lambda_0} = \frac{(\Gamma_n \tau/2)^2}{W((\lambda_0/\lambda_\tau)^{1/4} e^{\Gamma_n \tau/2}(\Gamma_n \tau/2))}. \] (14)

where \( W \) is the Lambert \( W \) function. Finally, the protocol takes a second jump \( \lambda_2 \rightarrow \lambda_\tau \) at \( t = \tau \). From (12), the final condition for the smooth part of the protocol, \( \lambda_2 \), satisfies the relation
\[ \lambda_2/\lambda_\tau = \lambda_0/\lambda_1, \] (15)

where \( \lambda_2 \) is found in terms of \( \lambda_1 \) from (14). We show the optimal protocol \( \lambda_t \) as a function of time using (11) in Fig. 1, with jumps \( \lambda_1 \) and \( \lambda_2 \) given by equations (14) and (15), respectively, for different values of \( \Gamma_n \tau \) and \( \lambda_\tau/\lambda_0 = 2 \), valid for all values of \( n \). We also use the magnitude of the first jump \( \lambda_1 \) from (14) to compute the optimal work using (13), depicted in Fig. 2, for different values of \( \Gamma_n \tau \) and \( \lambda_\tau/\lambda_0 \).

Therefore, the compression protocol that minimizes the dissipated work for a fixed time duration \( \tau \) and bounded values for the trapping parameters \( \lambda_0, \lambda_\tau \) is given by an initial fast adiabatic compression, followed by an exponential time-dependent isothermal drive, and ending with another fast adiabatic jump. Intuitively, one might understand this optimal protocol as follows: the system needs to be initially heated through a jump
By using kinetic theory considerations and the single-well potential work solution,

$$E_all = \frac{E}{V}$$

we write the stochastic pressure as a function of the kinetic energy $E$ as the usual identity $P = (2/f)E/V$. This approach yields to the expression for the increment of work of the Brownian gas $dW = -E(2V f/ fV) dt$. It is worth noting that, using the single-well potential work

$$E_all = \frac{E}{V}$$

and the effective volume as $V \propto \lambda^{-1/2}$ results in

$$\lambda / (2 \lambda_t) = -(2/f) V_t / V_t.$$  

Finally, upon combining the increments of heat and work, one obtains the same form of the SDE [3], proving the analogy between the highly underdamped limit of the Langevin system and the gas of Brownian particles. Therefore, all the results derived for the SDE [3] also apply to the Brownian gas: the Newton’s law of cooling and the heat fluctuation theorem [33], the propagator for the stochastic energy $P_t (E|E_0)$ from Eq. (6), Crooks FT, the nonequilibrium MB distribution with effective temperature given by Eq. (7), and the optimal work protocol found in Eq. (8). It is interesting to notice that the same exponential optimal work protocol of Eq. (9) was rigorously found for the linear regime in the case of a single particle in a box [34, 35], but without the adiabatic jumps [14] and [15]. Moreover, the box could also be modeled from [1] by making the limit $n \to \infty$, which in turn makes the potential $U(x) = 0$ for $|x| < L$ and $U(x) \to \infty$ otherwise, leading us to the same conclusions.

Conclusions— Usually in thermodynamics, processes are either too fast (e. g. adiabatic) with $\Gamma_0 \tau \to 0$, or too slow (quasistatic) with $\Gamma_0 \tau \to \infty$. A solvable stochastic thermodynamics framework as [5] has the advantage of providing a description of different nonequilibrium processes for finite time intervals. In this paper, we consider a classical particle submitted to a generalized single-well potential in high vacuum and derive the time dependent probability density function for the energy [5] explicitly, which allows the computation of far from equilib-
rium thermodynamics quantities. As a matter of fact, in our system the nonequilibrium information is encoded in a general time dependent effective temperature \[^7\] and fractional degrees of freedom \( f_a \). We showed the system satisfies the Crooks FT on its more general form. In addition, as a relevant application, we have found the optimal protocol \( \lambda_t \), for fixed values of \( \lambda_0 \) and \( \lambda_\tau \), that produces minimum average work over a finite time window \([0, \tau]\). The optimal protocol always has adiabatic jumps (at \( t = 0 \) and \( t = \tau \)) and a smooth exponential part (for \( t > 0 \)) for all kinds of single-well potentials. This finding sheds light on the analytic description of general thermal engines, as discontinuous protocols are likely to appear. Notice that the adiabatic, isochoric and isothermal processes are important parts of the description of nanoscopic thermal engines. The calculations of power and efficiency for different sorts of protocols requires dealing with the nonequilibrium thermodynamical observable quantities obtained in this paper and can be addressed in future works. As a final remark, it is relevant to mention that the highly underdamped limit SDE for the energy \[^5\] presents a form that resembles the thermodynamics of free Brownian particles, with similar exponential optimal protocols for long time isothermal processes \[^31\].

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