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The second law of thermodynamics in the quantum Brownian oscillator at an arbitrary temperature

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Abstract. In the classical limit no work is needed to couple a system to a bath with sufficiently weak coupling strength (*or* with arbitrarily finite coupling strength for a linear system) at the same temperature. In the quantum domain this may be expected to change due to system-bath entanglement. Here we show analytically that the work needed to couple a single linear oscillator with finite strength to a bath cannot be less than the work obtainable from the oscillator when it decouples from the bath. Therefore, the quantum second law holds for an arbitrary temperature. This is a generalization of the previous results for zero temperature [1,2]; in the high temperature limit we recover the classical behavior.

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1 Introduction

The second law of thermodynamics [3] is considered one of the central laws of science, engineering, and technology. For over a century it has been assumed to be inviolable by the scientific community. Over the last 20 years, however, its absolute status has come under increased scrutiny [4]. Challenges to the second law have recently attracted big interest with consideration of the miniaturization of a system under investigation, especially at low temperatures where quantum effects are important [4,5]. In contrast to common quantum statistical mechanics which is intrinsically based on a vanishingly small coupling between system and bath ("thermodynamic limit"), the finite coupling strength between them in the quantum regime causes some subtleties that must be recognized. The *quantum* thermodynamic behaviors of small systems have theoretically been investigated intensively and extensively [1,2,4–9] and experimentally been examined [4,10,11].

The problem of a quantum linear oscillator coupled to an independent-oscillator model of a heat bath (quantum Brownian motion) has been extensively discussed [12–16]. The validity of the quantum second law has recently been questioned in this scheme at zero temperature [5,17,18] by the fact that the coupled oscillator has a higher average energy value than the free harmonic oscillator ground state, which would not be in compliance with the second law. By means of a cyclic coupling/decoupling process one might expect to extract useful work from a single bath. However, this claim turned out incorrect; the apparent excess energy in the coupled oscillator cannot be used to extract useful work, neither for the well-known Drude damping model with a cut-off frequency for the spectral density of bath modes shown by Ford and O'Connell in [1] nor for both discrete bath modes and continuous bath modes with the generalized realistic damping models [2], since the minimum value of the work to couple the free oscillator to a bath takes above and beyond this excess energy. Therefore, the quantum second law for zero temperature is inviolate.

In this paper, we would like to discuss the second law in the quantum Brownian motion at an *arbitrary* temperature. We will obtain an analytic expression for the second-law inequality which can explicitly be shown to hold for the Drude model, which is the prototype for physically realistic damping. It is known [19] that a finite frequency cut-off reflects the physical fact that the bath cannot react instantaneously to a change of the system oscillator, and that in the absence of the cut-off, some observables such as the variance of the system momentum diverge. Therefore, the physically unrealistic cutoff-free damping models considered in [2] will not extensively be considered here (cf. Sect. 5). From the result of this work, the appearance (*or* disappearance) of quantum effects versus thermal fluctuation over the different temperatures will also be seen

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explicitly. Let us begin with a brief review on the basics of the quantum Brownian motion. We will below adopt the notations used in [20].

2 Basics of quantum Brownian motion

The quantum Brownian motion under consideration is described by the model Hamiltonian

$$\hat{H} = \hat{H}_s + \hat{H}_{b-sb}, \tag{1}$$

where

$$\hat{H}_s = \frac{\hat{p}^2}{2M} + \frac{M}{2}\,\omega_0^2\,\hat{q}^2 \tag{2}$$

$$\hat{H}_{s-sb} = \sum_{j=1}^{N} \left\{ \frac{\hat{p}_j^2}{2m_j} + \frac{m_j}{2}\omega_j^2 \left(\hat{x}_j - \frac{c_j}{m_j\,\omega_j^2}\,\hat{q} \right)^2 \right\}. \tag{3}$$

The Hamiltonian $\hat{H}_{s-sb}$ splits into the bath and the coupling term,

$$\hat{H}_b = \sum_{j=1}^{N} \left(\frac{\hat{p}_j^2}{2m_j} + \frac{m_j}{2}\omega_j^2\,\hat{x}_j^2 \right) \tag{4}$$

$$\hat{H}_{sb} = -\hat{q}\sum_{j=1}^{N} c_j\,\hat{x}_j + \hat{q}^2 \sum_{j=1}^{N} \frac{c_j^2}{2m_j\,\omega_j^2}. \tag{5}$$

From the hermiticity of Hamiltonian, the coupling constants c_j are real-valued. Without any loss of generality, we assume that

$$\omega_1 \le \omega_2 \le \cdots \le \omega_{N-1} \le \omega_N. \tag{6}$$

By means of the Heisenberg equation of motion, we can derive the quantum Langevin equation

$$M\,\ddot{\hat{q}} + M\int_0^t ds\,\gamma(t-s)\,\dot{\hat{q}}(s) + M\,\omega_0^2\,\hat{q} = \hat{\xi}(t), \tag{7}$$

where we used $\hat{p} = M\dot{\hat{q}}$, and the damping kernel and the noise operator are respectively given by

$$\gamma(t) = \frac{1}{M}\sum_{j=1}^{N} \frac{c_j^2}{m_j\,\omega_j^2}\cos(\omega_j\,t)\,;\;\hat{\xi}(t) = -M\gamma(t)\,\hat{q}(0) + \sum_{j=1}^{N} c_j \left\{ \hat{x}_j(0)\cos(\omega_j\,t) + \frac{\hat{p}_j(0)}{m_j\,\omega_j}\sin(\omega_j\,t) \right\}. \tag{8}$$

Here, $\langle\hat{\xi}(t)\rangle_{\rho_{b'}} = 0$ and $\langle\hat{\xi}(t)\,\hat{\xi}(t')\rangle_{\rho_{b'}} = M\gamma(t-t')/\beta$ for the initial bath state with the shifted canonical equilibrium distribution, $\hat{\rho}_{b'} = e^{-\beta\hat{H}_{b-sb}(0)}/Z_\beta^{(b')}$ [19] where $\beta = 1/k_B T$, and $Z_\beta^{(b')}$ is the normalization constant (i.e., the partition function). The Fourier-Laplace transform of $\gamma(t)$ is [20]

$$\tilde{\gamma}(\omega) = \frac{i\omega}{M}\sum_{j}^{N} \frac{c_j^2}{m_j\,\omega_j^2}\,\frac{1}{\omega^2 - \omega_j^2}. \tag{9}$$

Introducing the spectral density of bath modes as a characteristic of the bath,

$$J(\omega) = \pi\sum_{j=1}^{N} \frac{c_j^2}{2m_j\,\omega_j}\,\delta(\omega - \omega_j), \tag{10}$$

we can also express the damping kernel as

$$\gamma(t) = \frac{2}{M}\int_0^\infty \frac{d\omega}{\pi}\,\frac{J(\omega)}{\omega}\cos(\omega\,t). \tag{11}$$

We now consider a response function [20], $\chi(t) = \frac{i}{\hbar}\times\,\langle[\hat{q}(t),\,\hat{q}]\rangle_\beta$, where the expectation value $\langle\cdots\rangle_\beta$ is taken with respect to the equilibrium state, $\hat{\rho}_\beta = e^{-\beta\hat{H}}/Z_\beta$ with the partition function, $Z_\beta = \mathrm{Tr}\,e^{-\beta\hat{H}}$. The Fourier-Laplace transform of $\chi(t)$ is then the dynamic susceptibility

$$\tilde{\chi}(\omega) = \frac{1}{M}\,\frac{1}{\omega_0^2 - \omega^2 - i\omega\,\tilde{\gamma}(\omega)}, \tag{12}$$

which plays important roles later. It is known [12] that the susceptibility $\tilde{\chi}(\omega)$ can be rewritten as

$$\tilde{\chi}(\omega) = -\frac{1}{M}\,\frac{\prod_{j=1}^{N}(\omega^2 - \omega_j^2)}{\prod_{k=0}^{N}(\omega^2 - \bar{\omega}_k^2)}, \tag{13}$$

where the normal-mode frequencies, $\bar{\omega}_k$ of the total system $\hat{H}$ satisfy $\omega_0^2 - \bar{\omega}_k^2 - i\,\bar{\omega}_k\,\tilde{\gamma}(\bar{\omega}_k) = 0$. Without any loss of generality, we here assume that

$$\bar{\omega}_0 \le \bar{\omega}_1 \le \cdots \le \bar{\omega}_{N-1} \le \bar{\omega}_N. \tag{14}$$

It can then be found [14,2] that

$$\bar{\omega}_0 \le \omega_1 \le \bar{\omega}_1 \le \cdots \le \omega_{N-1} \le \bar{\omega}_{N-1} \le \omega_N \le \bar{\omega}_N, \tag{15}$$

and $\bar{\omega}_0 \le \omega_0 \le \bar{\omega}_N$.

The damping function $\tilde{\gamma}(\omega)$ in (9) can be rewritten as

$$\tilde{\gamma}(\omega) = \frac{i}{M}\int_0^\infty \frac{d\omega'}{\pi}\,\frac{J(\omega')}{\omega'}\left(\frac{1}{\omega'+\omega} - \frac{1}{\omega'-\omega}\right) \tag{16}$$

$$\tilde{\gamma}(\omega)\Big|_{\omega\to\omega+i\,0^+} = \frac{J(\omega)}{M\,\omega} + \frac{i}{M}\int_0^\infty \frac{d\omega'}{\pi}\,\frac{J(\omega')}{\omega'}\,P\left(\frac{1}{\omega'+\omega} - \frac{1}{\omega'-\omega}\right), \tag{17}$$

which resulted from the Fourier-Laplace transform of (11). Here, we used the well-known formula, $1/(x + i\,0^+) = P(1/x) - i\pi\delta(x)$ for $x = \omega' - \omega$. Equation (17) is convenient for the case of a continuous distribution $J(\omega)$ of bath modes; for the simple Ohmic case, $J_0(\omega) = M\gamma_o\,\omega$ with an ω-independent constant γ_o, we easily have

$\gamma_0(t) = 2\gamma_o\,\delta(t)$, and $\tilde{\gamma}_0(\omega) = \gamma_o$ with a vanishing principal (*or* imaginary) part in (17), while for the Drude model where $J_d(\omega) = M\,\gamma_o\,\omega\,\omega_d^2/(\omega^2+\omega_d^2)$ with a cut-off frequency ω_d, we have $\gamma_d(t) = \gamma_o\,\omega_d\,e^{-\omega_d\,t}$, and

$$\tilde{\gamma}_d(\omega) = \frac{\gamma_o\,\omega_d^2}{\omega^2+\omega_d^2} + i\,\frac{\gamma_o\,\omega_d\,\omega}{\omega^2+\omega_d^2} = \frac{\gamma_o\,\omega_d}{\omega_d - i\omega}. \tag{18}$$

Here, $J_d(\omega)$ behaves like $J_0(\omega)$ for small frequencies (with $\omega_d \to \infty$).

The model Hamiltonian in (1) can also describe the classical Brownian motion (see, e.g., [21]). In considering the quantum second law below, therefore its classical counterpart will be discussed briefly in comparison.

One might think of using instead of $\hat{H}_{sb}$ in (2) its rotating wave approximation $\hat{H}_{sb}^{(r)} = \hbar\sum_j \kappa_j\,(\hat{a}\,\hat{b}_j^\dagger + \hat{a}^\dagger\,\hat{b}_j)$, which has for $\omega_0 = \omega_j$ for all j, energy-conserving terms only. Here, $\hat{a} = \sqrt{\frac{M\omega_0}{2\hbar}}\,\hat{q} + \frac{i}{\sqrt{2M\hbar\omega_0}}\,\hat{p}$ and $\hat{b}_j = \sqrt{\frac{m_j\omega_j}{2\hbar}}\,\hat{x}_j + \frac{i}{\sqrt{2m_j\hbar\omega_j}}\,\hat{p}_j$, respectively. In this case, though, we cannot observe any excess energy of the coupled oscillator at zero temperature since the system oscillator and all bath oscillators remain unchanged in their ground states, respectively, with no entanglement (see also [22,23]); from the Heisenberg equation, $i\hbar\,\dot{\hat{H}}_s^{(r)} = [\hat{H}_s,\,\hat{H}^{(r)}] = [\hat{H}_s,\,\hat{H}_{sb}^{(r)}]$, we can obtain

$$\begin{aligned}\dot{\hat{H}}_s^{(r)} &= \frac{\hbar\omega_0}{i}\sum_j \kappa_j\left(\hat{a}^\dagger\,\hat{b}_j - \hat{a}\,\hat{b}_j^\dagger\right)\\ \dot{\hat{H}}_j^{(r)} &= \frac{\hbar\omega_j}{i}\kappa_j\left(\hat{a}\,\hat{b}_j^\dagger - \hat{a}^\dagger\,\hat{b}_j\right),\end{aligned} \tag{19}$$

which yield $\dot{\hat{H}}_s^{(r)}|\psi_i\rangle = \dot{\hat{H}}_j^{(r)}|\psi_i\rangle = 0$ for the initial state $|\psi_i\rangle = |0\rangle|00\cdots\rangle$, respectively. For the full Hamiltonian $\hat{H}$ in (1), on the other hand, we have, after a fairly lengthy calculation,

$$\begin{aligned}\dot{\hat{H}}_s &= \frac{\hbar}{2i}\sqrt{\frac{\omega_0}{M}}\sum_j \frac{1}{\sqrt{m_j\omega_j}}\,(\hat{a} - \hat{a}^\dagger)\,(\hat{b}_j + \hat{b}_j^\dagger)\\ &\quad + \frac{\hbar}{2iM}\left\{(\hat{a}^\dagger)^2 - \hat{a}^2\right\}\sum_j \frac{c_j^2}{m_j\omega_j^2},\end{aligned} \tag{20}$$

which clearly gives rise to $\dot{\hat{H}}_s|\psi_i\rangle \neq 0$.

3 Formulation of the quantum second law

From the fluctuation-dissipation theorem [19], we can easily have

$$\frac{1}{2}\langle\hat{q}(t_1)\hat{q}(t_2) + \hat{q}(t_2)\hat{q}(t_1)\rangle_\beta = \frac{\hbar}{\pi}\int_0^\infty d\omega\coth\left(\frac{\beta\hbar\omega}{2}\right)\cos\{\omega(t_2-t_1)\}\mathrm{Im}\{\tilde{\chi}(\omega+i\,0^+)\}, \tag{21}$$

which immediately yields

$$\langle\hat{q}^2\rangle_\beta = \frac{\hbar}{\pi}\int_0^\infty d\omega\coth\left(\frac{\beta\hbar\omega}{2}\right)\mathrm{Im}\{\tilde{\chi}(\omega+i0^+)\} \tag{22}$$

$$\langle\dot{\hat{q}}^2\rangle_\beta = \frac{\hbar}{\pi}\int_0^\infty d\omega\omega^2\coth\left(\frac{\beta\hbar\omega}{2}\right)\mathrm{Im}\{\tilde{\chi}(\omega+i0^+)\} \tag{23}$$

and thus the energy of the coupled oscillator

$$\begin{aligned}E_s(T) := \langle\hat{H}_s\rangle_\beta &= \frac{M\hbar}{2\pi}\int_0^\infty d\omega\,(\omega_0^2+\omega^2)\coth\left(\frac{\beta\hbar\omega}{2}\right)\\ &\quad\times\mathrm{Im}\{\tilde{\chi}(\omega+i\,0^+)\}.\end{aligned} \tag{24}$$

In comparison, the internal energy of an uncoupled (*or* free) oscillator is [3]

$$e(\omega_0,T) = \hbar\omega_0\left(\frac{1}{2} + \langle\hat{n}\rangle_\beta\right) = \frac{\hbar\omega_0}{2}\coth\frac{\beta\hbar\omega_0}{2}, \tag{25}$$

where the average quantum number $\langle\hat{n}\rangle_\beta = 1/(e^{\beta\hbar\omega_0}-1)$. Its classical counterpart appears as $e_{cl}(T) = \frac{1}{\beta}$. With (25), equation (24) can now be transformed to an expression

$$E_s(T) = E_s(0) + \Delta E_s(T), \tag{26}$$

where

$$\begin{aligned}E_s(0) &= -\frac{M\hbar}{4\pi i}\oint d\omega\,\left(\omega_0^2+\omega^2\right)\,\tilde{\chi}(\omega)\\ \Delta E_s(T) &= -\frac{M\hbar}{2\pi i}\oint d\omega\,\left(\omega_0^2+\omega^2\right)\,\tilde{\chi}(\omega)\,\langle\hat{n}\rangle_\beta.\end{aligned} \tag{27}$$

Here, the integration path is a loop around the positive real axis in the complex ω-plane, consisting of the two branches, $(\infty + i\epsilon,\, i\epsilon)$ and $(-i\epsilon,\, \infty - i\epsilon)$ [24]. Therefore, $E_s(T)$ for the discrete bath modes can be exactly obtained in closed form from the residues evaluated at all poles $\{\bar{\omega}_k\}$ of $\tilde{\chi}(\omega)$ in (13) on the positive real axis. Then, equation (26) reduces to

$$E_s(T) = \frac{1}{2}\sum_{k=0}^N e(\bar{\omega}_k,T)\left\{1 + \left(\frac{\omega_0}{\bar{\omega}_k}\right)^2\right\}\frac{\prod_{j=1}^N(\bar{\omega}_k^2 - \omega_j^2)}{\prod_{\substack{k'=0\\(\neq k)}}^N(\bar{\omega}_k^2 - \bar{\omega}_{k'}^2)}. \tag{28}$$

To study the quantum second law below, we need two different Helmholtz free energies for the oscillator coupled to a bath.

We first consider the Helmholtz free energy of the coupled oscillator, $F_s(T) = E_s(T) - TS_s$ [3] with its entropy $S_s = -\partial F_s/\partial T$. This can easily be solved for F_s such that [25]

$$F_s(T) = T\left(-\int_{T_0}^T \frac{E_s(T')}{T'^2}\,dT' + \mathcal{C}\right). \tag{29}$$

By requiring that the entropy

$$S_s(\beta) = k_B\,\beta\,E_s(\beta) - k_B\int_{\beta_0}^\beta E_s(\beta')\,d\beta' - \mathcal{C} \tag{30}$$

with $\beta_0 = 1/k_B T_0$ vanish at zero temperature, we can determine the constant $\mathcal{C}$ of integration; using (24) we easily obtain

$$\mathcal{C}/k_B = \frac{M\hbar}{2\pi} \int_0^\infty d\omega\, (\omega_0^2 + \omega^2)\, \mathrm{Im}\{\tilde{\chi}(\omega + i\,0^+)\}\, \mathcal{A}(\omega), \tag{31}$$

where

$$\mathcal{A}(\omega) = \left.\left(\beta \coth\frac{\beta\hbar\omega}{2} - \int_{\beta_0}^{\beta} d\beta' \coth\frac{\beta'\hbar\omega}{2}\right)\right|_{\beta\to\infty}. \tag{32}$$

The asymptotic series, $\coth z = 1 + 2\sum_{l=1}^{\infty} e^{-2lz}$ then allows equation (32) to become

$$\mathcal{A}(\omega) = \int^{\beta_0} d\beta' \coth\frac{\beta'\hbar\omega}{2} = \frac{2}{\hbar\omega} \ln\left(\sinh\frac{\beta_0\hbar\omega}{2}\right), \tag{33}$$

which clearly makes equations (29) and (30), respectively, independent of T_0 (*or* β_0) and accordingly uniquely determined.

The other Helmholtz free energy $\mathcal{F}_s(T)$ needed for the second law is described as follows; the minimum work required to couple a system oscillator at temperature T to a bath at the same temperature is equivalent to the Helmholtz free energy of the *coupled* total system minus the free energy of the *uncoupled* bath [12]. This minimum work can then be obtained as the free energy $\mathcal{F}_s(T) = -\frac{1}{\beta}\ln \mathcal{Z}_\beta$, where the canonical partition function $\mathcal{Z}_\beta = \mathrm{Tr}\, e^{-\beta\hat{H}}/\mathrm{Tr}_b\, e^{-\beta\hat{H}_b}$. Here, Tr_b denotes the partial trace for the bath alone (in the absence of a coupling between system and bath, this would exactly correspond to the partition function of the system only). By means of the normal-mode frequencies $\bar{\omega}_k$, we easily get

$$\mathcal{Z}_\beta = \frac{\displaystyle\prod_{k=0}^{N} \sum_{n_k=0} e^{-\beta\hbar\bar{\omega}_k\left(n_k+\frac{1}{2}\right)}}{\displaystyle\prod_{j=1}^{N} \sum_{n_j=0} e^{-\beta\hbar\omega_j\left(n_j+\frac{1}{2}\right)}}, \tag{34}$$

which yields

$$\mathcal{F}_s(T) = \sum_{k=0} f(\bar{\omega}_k, T) - \sum_{j=1} f(\omega_j, T) \tag{35}$$

with the free energy of an uncoupled oscillator

$$f(\omega, T) = \frac{\hbar\omega}{2} + \frac{1}{\beta}\ln\left(1 - e^{-\beta\hbar\omega}\right). \tag{36}$$

The classical counterpart of (36) is $f_{cl}(\omega, T) = \frac{\ln\beta\hbar\omega}{\beta}$ with $\hbar \ll 1$. Equation (35) can then be rewritten as [12]

$$\mathcal{F}_s(T) = \frac{1}{\pi}\int_0^\infty d\omega\, f(\omega, T)\mathrm{Im}\left\{\frac{d}{d\omega}\ln\tilde{\chi}(\omega + i0^+)\right\} \tag{37}$$

in terms of the susceptibility. We note here that for an uncoupled oscillator, $\mathrm{Im}\left\{\frac{d}{d\omega}\ln\tilde{\chi}(\omega + i0^+)\right\} \to \pi\,\delta(\omega - \omega_0)$ and thus $\mathcal{F}_s(T) \to f(\omega_0, T)$. Similarly to (37), we can also obtain the energy required to couple a system oscillator to a bath,

$$\begin{aligned}\mathcal{E}_s(T) &= \sum_{k=0} e(\bar{\omega}_k, T) - \sum_{j=1} e(\omega_j, T)\\ &= \frac{1}{\pi}\int_0^\infty d\omega\, e(\omega, T)\mathrm{Im}\left\{\frac{d}{d\omega}\ln\tilde{\chi}(\omega + i0^+)\right\}\end{aligned} \tag{38}$$

From $e(\omega, T) \geq f(\omega, T)$ (the equal sign holds for $T = 0$ only) with the frequency relationship in (15), we can easily get $\mathcal{E}_s(T) \geq \mathcal{F}_s(T)$.

We now consider a cyclic process composed of the coupling of a harmonic oscillator to a bath and then the decoupling of the oscillator from the bath (the coupling constants $c_j \to 0$). The free energy change on completion of the coupling process is $\mathcal{F}_s(T) - f(\omega_0, T)$, whereas the maximum useful work obtainable from the oscillator only in the decoupling process is the free energy difference $F_s(T) - f(\omega_0, T)$ which cannot be greater than the energy change $E_s(T) - e(\omega_0, T)$. Here it is assumed obviously that the extraction of energy from the bath is impossible. The second law can then be expressed as an inequality

$$\mathcal{F}_s(T) - f(\omega_0, T) \geq E_s(T) - e(\omega_0, T). \tag{39}$$

In obtaining (39) we used the conceptional difference between $\mathcal{F}_s(T)$ and $F_s(T)$ ("operational asymmetry") [26]. For zero temperature, this inequality, obviously, reduces to $\mathcal{F}_s(0) \geq E_s(0)$, the validity of which has been explicitly proven for the Drude damping model [1] and for the discrete bath modes, by means of $E_s(0)$ in (28) and $\mathcal{F}_s(0)$ in (35) with the frequency relationship (15), and the generalized realistic damping models of continuous bath modes [2]. For non-zero temperatures, on the other hand, it is very non-trivial to investigate the validity of inequality (39) with $E_s(T)$ and $\mathcal{F}_s(T)$ for the discrete bath modes. For the continuous bath modes, the evaluation of $E_s(T)$ and $\mathcal{F}_s(T)$ clearly depends on the parameters of the damping model considered. We will below discuss inequality (39) explicitly within the Drude model which is the prototype for physically realistic damping.

4 Discussion of the second law within the Drude model

It is convenient in the Drude model to adopt, in place of $(\omega_0, \omega_d, \gamma_o)$, the parameters $(\mathbf{w}_0, \Omega, \gamma)$ through the relations [1]

$$\begin{aligned}\omega_0^2 &:= \mathbf{w}_0^2 \tfrac{\Omega}{\Omega+\gamma};\, \omega_d := \Omega + \gamma\\ \gamma_o &:= \gamma\, \tfrac{\Omega\,(\Omega+\gamma) + \mathbf{w}_0^2}{(\Omega+\gamma)^2}.\end{aligned} \tag{40}$$

Substituting equation (18) with (40) into (12), we obtain the susceptibility

$$\tilde{\chi}_d(\omega) = -\frac{1}{M}\frac{\omega + i\,(\Omega + z_1 + z_2)}{(\omega + i\Omega)(\omega + iz_1)(\omega + iz_2)}, \tag{41}$$

where $z_1 = \gamma/2 + i\mathbf{w}_1$ and $z_2 = \gamma/2 - i\mathbf{w}_1$ with $\mathbf{w}_1 = \sqrt{\mathbf{w}_0^2 - (\gamma/2)^2}$. First, we consider the overdamped case ($\gamma/2 > \mathbf{w}_0$), where z_1, $z_2 > 0$. We then have

$$\operatorname{Im}\tilde{\chi}_d(\omega) = -\frac{1}{M}\sum_{l=1}^{3}\lambda_d^{(l)}\,\frac{\omega}{\omega^2 + \underline{\omega_l}^2}, \qquad (42)$$

where $\underline{\omega_1} = \Omega$, $\underline{\omega_2} = z_1$, $\underline{\omega_3} = z_2$, and the coefficients

$$\lambda_d^{(1)} = \frac{z_1 + z_2}{(\Omega - z_1)(z_2 - \Omega)};\; \lambda_d^{(2)} = \frac{\Omega + z_2}{(z_1 - \Omega)(z_2 - z_1)}$$
$$\lambda_d^{(3)} = \frac{\Omega + z_1}{(z_2 - \Omega)(z_1 - z_2)}. \qquad (43)$$

Here, we note that

$$\sum_{l=1}^{3}\lambda_d^{(l)} = 0;\; \sum_{l=1}^{3}\lambda_d^{(l)}\,\underline{\omega_l}^2 = 0. \qquad (44)$$

To obtain an explicit expression for the energy $E_s^{(d)}(T)$ of the coupled oscillator, we first consider the integral in (21); by performing a contour integration with the aid of (42) and the identity

$$\coth\left(\frac{\beta\hbar\omega}{2}\right) = \frac{2}{\beta\hbar\omega}\left(1 + 2\sum_{n=1}^{\infty}\frac{\omega^2}{\nu_n^2 + \omega^2}\right), \qquad (45)$$

where $\nu_n = 2\pi n/\beta\hbar$, we can have

$$\frac{1}{2}\,\langle\hat{q}(0)\,\hat{q}(t) + \hat{q}(t)\,\hat{q}(0)\rangle_\beta^{(d)} =$$
$$-\frac{1}{\beta M}\sum_{l=1}^{3}\lambda_d^{(l)}\left\{\frac{e^{-\underline{\omega_l}t}}{\underline{\omega_l}} + 2\sum_{n=1}^{\infty}\frac{\nu_n\,e^{-\nu_n t} - \underline{\omega_l}\,e^{-\underline{\omega_l}t}}{\nu_n^2 - \underline{\omega_l}^2}\right\}. \qquad (46)$$

With $\hbar \to 0$, this reduces to its classical counterpart, ${}_{cl}\langle q(0)\,q(t)\rangle_\beta^{(d)} = -\frac{1}{\beta M}\sum_l \lambda_d^{(l)}\,\frac{e^{-\underline{\omega_l}t}}{\underline{\omega_l}}$. From (46) and the relation [20]

$$\langle\dot{\hat{q}}(0)\dot{\hat{q}}(t) + \dot{\hat{q}}(t)\dot{\hat{q}}(0)\rangle_\beta = -\frac{d^2}{dt^2}\langle\hat{q}(0)\hat{q}(t) + \hat{q}(t)\hat{q}(0)\rangle_\beta \qquad (47)$$

it can eventually be found that

$$E_s^{(d)}(T) = \frac{1}{\beta}\sum_{l=1}^{3}\lambda_d^{(l)}$$
$$\times\left\{\frac{\omega_0^2 - \underline{\omega_l}^2}{2\underline{\omega_l}} - \sum_{n=0}^{\infty}\frac{\omega_0^2 - \left(\nu_n^2 + \nu_n\underline{\omega_l} + \underline{\omega_l}^2\right)}{\nu_n + \underline{\omega_l}}\right\}. \qquad (48)$$

With $\hbar \to 0$, its classical counterpart easily appears as ${}_{cl}E_s^{(d)}(T) = \frac{1}{2\beta}\sum_l \lambda_d^{(l)}\,\frac{\underline{\omega_l}^2 - \omega_0^2}{\underline{\omega_l}} = \frac{1}{\beta} = e_{cl}(\omega_0, T)$. We can also express (46) and (47) at $t = 0$, respectively, in terms of the Digamma function [27]

$$\psi(y) = \frac{d\,\ln\Gamma(y)}{dy} = -c_e + \sum_{n=1}^{\infty}\frac{1}{n} - \sum_{n=0}^{\infty}\frac{1}{n+y} \qquad (49)$$

with the Euler constant c_e as

$$\langle\hat{q}^2\rangle_\beta^{(d)} = \frac{1}{M}\sum_{l=1}^{3}\lambda_d^{(l)}\left\{\frac{1}{\beta\underline{\omega_l}} + \frac{\hbar}{\pi}\psi\left(\frac{\beta\hbar\underline{\omega_l}}{2\pi}\right)\right\} \qquad (50)$$

$$\langle\dot{\hat{q}}^2\rangle_\beta^{(d)} = -\frac{1}{M}\sum_{l=1}^{3}\lambda_d^{(l)}\,\underline{\omega_l}^2\left\{\frac{1}{\beta\underline{\omega_l}} + \frac{\hbar}{\pi}\psi\left(\frac{\beta\hbar\underline{\omega_l}}{2\pi}\right)\right\} (51)$$

Here, we used (44). Equation (48) can thus be rewritten as

$$E_s^{(d)}(T) = \frac{1}{2}\sum_{l=1}^{3}\lambda_d^{(l)}\,(\omega_0^2 - \underline{\omega_l}^2)\left\{\frac{1}{\beta\underline{\omega_l}} + \frac{\hbar}{\pi}\psi\left(\frac{\beta\hbar\underline{\omega_l}}{2\pi}\right)\right\}. \qquad (52)$$

With the aid of the asymptotic expression, $\psi(y) = \ln y - \frac{1}{2y} - \sum_{n=1}^{\infty}\frac{B_{2n}}{2n\,y^{2n}}$ with the Bernoulli number B_n [27], the zero-temperature value $E_s^{(d)}(0)$, clearly, reduces to (66) derived in [2] (see Appendix).

Let us now consider the free energy $\mathcal{F}_s^{(d)}(T)$. By substituting (41) into (37) with the identity, $\ln(1 + y) = -\sum_{n=1}^{\infty}(-y)^n/n$, we can easily obtain

$$\mathcal{F}_s^{(d)}(T) = \mathcal{F}_s^{(d)}(0) + \frac{1}{\pi\beta}\sum_{n=1}^{\infty}\frac{1}{n}\,\varDelta_n(\beta), \qquad (53)$$

where

$$\mathcal{F}_s^{(d)}(0) = \frac{\hbar}{2\pi}\left\{(\Omega+\gamma)\,\ln\left(\frac{\Omega+\gamma}{\Omega}\right) + \gamma\,\ln\left(\frac{\Omega}{\mathbf{w}_0}\right)\right.$$
$$\left. + \bar{\mathbf{w}}_1\,\ln\left(\frac{\gamma/2 - \bar{\mathbf{w}}_1}{\gamma/2 + \bar{\mathbf{w}}_1}\right)\right\} \qquad (54)$$

with $\bar{\mathbf{w}}_1 = \sqrt{(\gamma/2)^2 - \mathbf{w}_0^2}$, as was derived in [2], and

$$\varDelta_n(\beta) = \sum_{\mu=0}^{3}\tau_d^{(\mu)}\int_0^\infty dy\,\frac{e^{-n\beta\hbar\underline{\omega_\mu}\,y}}{y^2+1} \qquad (55)$$
$$= \sum_{\mu=0}^{3}\tau_d^{(\mu)}\left\{\sin(n\beta\hbar\underline{\omega_\mu})\mathrm{Ci}(n\beta\hbar\underline{\omega_\mu})\right.$$
$$\left. - \cos(n\beta\hbar\underline{\omega_\mu})\mathrm{si}(n\beta\hbar\underline{\omega_\mu})\right\} \qquad (56)$$

with $\tau_d^{(0)} = 1$, $\tau_d^{(1)} = \tau_d^{(2)} = \tau_d^{(3)} = -1$, and $\underline{\omega_0} = \omega_d$ (see also Appendix). By the substituting the classical quantity $f_{cl}(\omega, T)$ into (37) with $\int_0^x dy/(y^2+1) = \arctan x$ and $\int_0^\infty dy\,\ln y/(y^2+1) = 0$ [28], we can also obtain the classical counterpart ${}_{cl}\mathcal{F}_s^{(d)}(T) = -\frac{1}{2\beta}\ln\frac{\omega_d}{\Omega} + f_{cl}(\mathbf{w}_0, T) = f_{cl}(\omega_0, T)$.

For the underdamped case ($\gamma/2 \le \mathbf{w}_0$), equations (52) and (53) can be found to hold as well, respectively, being expressed in terms of the functions with complex-valued arguments. By showing the validity of inequality (39) for underdamped and overdamped cases,

$$K_d(T) := \mathcal{F}_s^{(d)}(T) - f(\omega_0, T) - E_s^{(d)}(T) + e(\omega_0, T) \ge 0 \qquad (57)$$

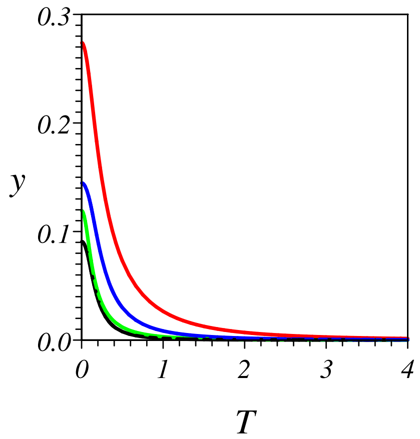

Fig. 1. $y = K_d(T)/\hbar\mathbf{w}_0$ versus temperature T; from bottom to top: ($\Omega = 1$ and $\gamma = 3/2$ underdamped), ($\Omega = 1$ and $\gamma = 4$ overdamped), ($\Omega = 5$ and $\gamma = 3/2$ underdamped), and ($\Omega = 5$ and $\gamma = 4$ overdamped); here, $\hbar = k_B = \mathbf{w}_0 = 1$; in the high temperature limit, we have the classical behavior, $y \to 0^+$.

as in Figure 1, we see that there is no violation of the quantum second law; in fact, $K_d(T)$ vanishes asymptotically with the increase of T.

Comments deserve here. In the classical treatment both sides of inequality (39) vanish, namely, ${}_{cl}K_d(T) = 0$, which clearly means that no work is required to couple a linear system to a bath at the same temperature, and no energy change in the system is obtained during the decoupling (and the coupling). In the quantum treatment, on the other hand, $E_s^{(d)}(T)$ and $\mathcal{F}_s^{(d)}(T)$ depend on the damping parameters, respectively. Therefore, while both sides of (39) become vanishing in the high temperature limit (equivalently, $\hbar \to 0$), they actually do not vanish especially in the low temperature regime. This non-vanishing behavior stems from the system-bath entanglement induced by the finite coupling strength between them, which leads to the deviation from $\hat{\rho}_\beta^{(s)} = e^{-\beta \hat{H}_s}/Z_\beta^{(s)}$ for the reduced density matrix $\hat{\rho}_s^{(d)}(T)$ being, clearly, damping-parameter dependent.

In fact, we have $K_d(T) > 0$ especially in the low temperature regime (see Fig. 1). This strict irreversibility over a single cycle composed of the coupling and decoupling process appears from the fact that the system-bath entanglement induces the entanglement between any pair of infinitely many bath oscillators ("entanglement swapping" [29]), which cannot completely removed over the system-bath decoupling process. Therefore, we essentially cannot recover the original state of the bath, $\hat{\rho}_\beta^{(b)}$ and thus that of the system, $\hat{\rho}_\beta^{(s)}$. As a result, $\mathcal{F}_s^{(d)}(T) - f(\omega_0, T)$, being the minimum work required for the entangling in the coupling process, is greater than the energy change $E_s^{(d)}(T) - e(\omega_0, T)$, which can necessarily not be less than the free energy change, $F_s^{(d)}(T) - f(\omega_0, T)$ being the maximum useful work obtainable from the system only in the decoupling process. With the increase of T, however, the strict irreversibility shrinks ($K_d(T) \to 0^+$) since the thermal effect dominates the quantum effect. In the classical case, on the other hand, this operational asymmetry, introduced in the last paragraph of Section 3, disappears at an arbitrary temperature, namely ${}_{cl}\mathcal{F}_s^{(d)}(T) = {}_{cl}F_s^{(d)}(T) = f_{cl}(\omega_0, T)$.

5 Comparison with the Ohmic model

Let us briefly consider the Ohmic model (as a cutoff-free damping model) for an arbitrary temperature to compare with the Drude model considered in Section 4. For zero temperature it is known [2] that $K_o(0) = \mathcal{F}_s^{(o)}(0) - E_s^{(o)}(0)$ vanishes, where

$$\mathcal{F}_s^{(o)}(0) = E_s^{(o)}(0) = \frac{\hbar\gamma_o}{2\pi} \int_0^\infty d\omega \, \frac{\omega\,(\omega^2 + \omega_0^2)}{(\omega^2 - \omega_0^2)^2 + (\gamma_0\omega)^2} \tag{58}$$

diverges logarithmically, while its Drude-model counterpart $K_d(0) \to E_g\,\gamma/\pi\mathbf{w}_0$ in the limit $\omega_d \to \infty$ (equivalently, $\Omega \to \infty$) where E_g is the ground state energy of a free oscillator.

For the overdamped case ($\gamma_o/2 > \omega_0$), the susceptibility in (12) appears as

$$\tilde{\chi}_o(\omega) = -\frac{1}{M} \frac{1}{(\omega + i\omega_1)(\omega + i\omega_2)}, \tag{59}$$

where $\omega_1 = \gamma_o/2 - \bar{\mathbf{w}} > 0$ and $\omega_2 = \gamma_o/2 + \bar{\mathbf{w}} > 0$ with $\bar{\mathbf{w}} = \sqrt{(\gamma_o/2)^2 - \omega_0^2}$. Substituting (59) into (21), we can obtain

$$\frac{1}{2}\,\langle \hat{q}(0)\,\hat{q}(t) + \hat{q}(t)\,\hat{q}(0)\rangle_\beta^{(o)} = -\frac{1}{2\beta\bar{\mathbf{w}}M} \sum_{j=1}^{2} \lambda_o^{(j)} \left\{ \frac{e^{-\omega_j t}}{\omega_j} + 2\sum_{n=1}^{\infty} \frac{\nu_n\, e^{-\nu_n t} - \omega_j\, e^{-\omega_j t}}{\nu_n^2 - \omega_j^2} \right\} \tag{60}$$

with $\lambda_o^{(1)} = -1$ and $\lambda_o^{(2)} = 1$ [30], from which, similarly to (50),

$$\langle \hat{q}^2 \rangle_\beta^{(o)} = \frac{1}{2\bar{\mathbf{w}}M} \sum_{j=1}^{2} \lambda_o^{(j)} \left\{ \frac{1}{\beta\,\omega_j} + \frac{\hbar}{\pi}\psi\left(\frac{\beta\hbar\omega_j}{2\pi}\right) \right\}. \tag{61}$$

Substituting (60) into (47), we can also get

$$\langle \dot{\hat{q}}^2 \rangle_\beta^{(o)} = -\frac{1}{2\bar{\mathbf{w}}M} \sum_{j=1}^{2} \lambda_o^{(j)}\,\omega_j^2 \left(\frac{1}{\beta\omega_j} + \frac{\hbar}{\pi}\left\{ \psi\left(\frac{\beta\hbar\omega_j}{2\pi}\right) + c_e - \sum_{n=1}^{\infty} \frac{1}{n} \right\} \right), \tag{62}$$

which diverges logarithmically for an arbitrary temperature (note that $\lim_{n\to\infty}(\sum_{k=1}^{n} \frac{1}{k} - \ln n) = c_e$); compare this with $\langle \dot{\hat{q}}^2 \rangle_\beta^{(d)}$ in (51) being convergent. From (61) and (62), the energy of the coupled oscillator is

$$E_s^{(o)}(T) = \frac{1}{4\bar{\mathbf{w}}} \sum_{j=1}^{2} \lambda_o^{(j)} \left\{ (\omega_0^2 - \omega_j^2) \left\{ \frac{1}{\beta\omega_j} + \frac{\hbar}{\pi}\psi\left(\frac{\beta\hbar\omega_j}{2\pi}\right) \right\} - \frac{\hbar\omega_j^2}{\pi} \left(c_e - \sum_{n=1}^{\infty} \frac{1}{n} \right) \right\}, \tag{63}$$

which clearly diverges. With $\hbar \to 0$, its classical counterpart, however, reduces to ${}_{cl}E_s^{(o)}(T) = e_{cl}(\omega_0, T)$ (it can also be found [9] that $E_s^{(o)}(T)$ is identical to the energy $\mathcal{E}_s^{(o)}(T)$ obtainable from (38)). Further, similarly to (53), we can easily obtain

$$\mathcal{F}_s^{(o)}(T) = \mathcal{F}_s^{(o)}(0) + \Delta\mathcal{F}_s^{(o)}(T), \tag{64}$$

where

$$\Delta\mathcal{F}_s^{(o)}(T) = -\frac{1}{\pi\beta} \sum_{n=1}^{\infty} \frac{1}{n} \sum_{j=1}^{2} \{ \sin(n\beta\hbar\omega_j)\,\mathrm{Ci}(n\hbar\beta\omega_j) - \cos(n\beta\hbar\omega_j)\,\mathrm{si}(n\beta\hbar\omega_j) \}. \tag{65}$$

The free energy $\mathcal{F}_s^{(o)}(T)$ also diverges. With $\hbar \to 0$, we get ${}_{cl}\mathcal{F}_s^{(o)}(T) = f_{cl}(\omega_0, T)$. Equations (63) and (64) can be found to hold, respectively, for the underdamped case as well.

For comparison with the Drude model, we take the limit Ω (*or* ω_d) $\to \infty$ in (4) so that $\lambda_d^{(1)} \to 0$, $\lambda_d^{(2)} \to 1/(z_1 - z_2)$, and $\lambda_d^{(3)} \to -\lambda_d^{(2)}$. Then, it can easily be shown that $E_s^{(d)}(T) \nrightarrow E_s^{(o)}(T)$ and $\mathcal{F}_s^{(d)}(T) \nrightarrow \mathcal{F}_s^{(o)}(T)$; the second term on the right hand side of (53) reduces to $\Delta\mathcal{F}_s^{(o)}(T)$ in (64), however, the first term $\mathcal{F}_s^{(d)}(0) \nrightarrow \mathcal{F}_s^{(o)}(0)$ as discussed in [2]. As a result, the second-law inequality (39) for the Drude model with $\omega_d \to \infty$ is not equivalent to that for the Ohmic model. Whereas the classical counterpart ${}_{cl}K_o(T) = 0$, both sides of (39) come to diverge differently so that it is non-trivial to explicitly evaluate $K_o(T)$ for this *unrealistic* damping model.

6 Conclusions

In summary, we have studied the second law in the scheme of quantum Brownian motion at an *arbitrary* temperature. It is clearly a generalization of the previous works for zero temperature by Ford and O'Connell [1] and by the authors of the present paper [2]. It has been shown for the physically realistic damping model that the work needed to couple a system oscillator to a bath at the same temperature cannot be less than the work obtainable from the oscillator only when it is extracted from the bath; especially in the low temperature regime the apparent irreversibility, $K_d(T) > 0$, stemming from the system-bath entanglement was found, which is different from the behavior of its classical counterpart, ${}_{cl}K_d(T) = 0$. Therefore, the quantum second law holds for an arbitrary temperature. The question about the validity of the quantum second law for a broader class of quantum systems than the quantum Brownian motion considered here, especially non-linear systems coupled to a bath, clearly remains open.

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Appendix A: Mathematical supplements

It has been shown in [2] that for the overdamped case ($\mathbf{w}_0 \le \gamma/2$),

$$E_s^{(d)}(0) = \frac{\hbar}{2\pi} \{ A(\mathbf{w}_0, \Omega, \gamma) + B(\mathbf{w}_0, \Omega, \gamma) \}, \tag{66}$$

where

$$A(\mathbf{w}_0, \Omega, \gamma) = \frac{(\mathbf{w}_0^2 + \Omega^2)(\Omega\gamma^2/4 - \Omega\mathbf{w}_0^2 - \mathbf{w}_0^2\gamma/2) + \Omega^2\gamma^3/4}{\bar{\mathbf{w}}_1(\Omega + \gamma)(\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} \ln\left(\frac{\gamma/2 - \bar{\mathbf{w}}_1}{\gamma/2 + \bar{\mathbf{w}}_1}\right) \tag{67}$$

with $\bar{\mathbf{w}}_1 = \sqrt{(\gamma/2)^2 - \mathbf{w}_0^2}$, and

$$B(\mathbf{w}_0, \Omega, \gamma) = \frac{\Omega\,\gamma\,(\Omega^2 + \Omega\,\gamma - \mathbf{w}_0^2)}{(\Omega + \gamma)\,(\mathbf{w}_0^2 - \Omega\,\gamma + \Omega^2)} \ln(\Omega/\mathbf{w}_0). \tag{68}$$

In derivation of equation (56) from (55), we used [28]

$$\int_0^{\infty} dy\, \frac{e^{-ay}}{y^2 + b^2} = \frac{1}{b} \{\sin(ab)\mathrm{Ci}(ab) - \cos(ab)\mathrm{si}(ab)\}, \tag{69}$$

where a, $b > 0$; the sine integral $\mathrm{si}(y) = -\int_y^{\infty} dz\, \frac{\sin(z)}{z} = -\frac{\pi}{2} + \mathrm{Si}(y)$ with $\mathrm{Si}(y) = \int_0^y dz\, \frac{\sin(z)}{z}$, and the cosine integral $\mathrm{Ci}(y) = -\int_y^{\infty} dz\, \frac{\cos(z)}{z} = c_e + \ln y + \int_0^y dz\, \frac{\cos(z) - 1}{z}$ with the Euler constant $c_e = 0.5772156649\cdots$.

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