

Quantum Brownian motion and the second law of thermodynamics

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Abstract. We consider a single harmonic oscillator coupled to a bath at zero temperature. As is well-known, the oscillator then has a higher average energy than that given by its ground state. Here we show analytically that for a damping model with arbitrarily discrete distribution of bath modes and damping models with continuous distributions of bath modes with cut-off frequencies, this excess energy is less than the work needed to couple the system to the bath, therefore, the quantum second law is not violated. On the other hand, the second law may be violated for bath modes without cut-off frequencies, which are, however, physically unrealistic models.

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1 Introduction

Thermodynamics originally developed as a purely phenomenological description of the effects caused by changes in temperature, pressure, and volume on physical systems at the macroscopic scale. At the heart of thermodynamics there are four well-known laws [1]; the zeroth law allows us to define temperature scales and thermometers while the first law is nothing else than a generalized expression of the law of energy conservation. The second law introduces the concept of thermodynamic entropy, which never decreases for an isolated system. The third law states that as a system approaches the zero temperature, the entropy of the system approaches zero. Later on, Boltzmann and his followers created and developed statistical thermodynamics by reducing the phenomenologically described thermodynamics entirely to the scheme of classical statistical mechanics. When quantum mechanics appeared, the statistical thermodynamics had to take into account additional factors offered by quantum mechanics, but the overall structure of thermodynamics, its fundamental laws, and its meaning fit for macroscopic systems remained unchanged since quantum mechanics was believed to play no roles at the macroscopic scale.

A big challenge for thermodynamics arose with the miniaturization of a system under consideration [2]; in contrast to common quantum statistical mechanics which is intrinsically based on a vanishingly small coupling between system and bath, the finite coupling strength between them causes some subtleties that must be

recognized. Recent advances in technology have enabled us to experimentally study mesoscopic systems and test various fundamental concepts. The field of nano electro-mechanical systems (NEMS) especially has emerged with a great potential, e.g., in quantum limit detection and amplification [3, 4], and *welcher-Weg* ('which-path') interferometry [5]. Here, the effects of dissipative environments that are negligible in macroscopic resonators become detrimental, and the noise is, therefore, a major limiting factor in control of NEMS resonators. Theoretically, NEMS resonators can be modeled as the simplest form in the scheme of quantum Brownian motion (see [6] for fundamental aspects of quantum Brownian motion). Such a development in various fields related to the quantum statistical and mesoscopic physics has led to considerable interest in the area of quantum and mesoscopic thermodynamics, especially with the question raised on the validity of the thermodynamic laws. Discussions about what is the meaning of quantum thermodynamics [2, 7] have started and continued up to now.

The validity of the second law was questioned in the scheme of quantum Brownian motion [2], motivated from the observation of the fact that a single harmonic oscillator coupled to a bath at zero temperature has indeed a higher average energy value than the uncoupled harmonic oscillator ground state (see also [8]), which could not be in accordance with the second law in its Kelvin-Planck form [1] that *a system operating in contact with a thermal reservoir cannot produce positive work in its surroundings* (cf. for a discussion on the validity of the quantum third law in the low temperature limit, see, e.g., Refs. [9, 10]). However, this argument has been shown to be wrong by Ford

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and O'Connell [11]; by means of the generalized Langevin equation they showed, for the well-known Drude model for the spectral density of bath modes, that the apparent excess energy in the coupled harmonic oscillator, however, cannot be used to extract useful work since the minimum value of the work to couple the free oscillator to a bath takes above and beyond this excess energy, therefore, the second law of thermodynamics is inviolate even in the quantum regime (i.e., for cases with non-negligible coupling strengths at temperature $T = 0$ without thermal fluctuation). Unfortunately they were unable to explicitly connect their result with its model-independent, deep quantum origin, thus the validity of the *quantum* second law for a more general form of the spectral density of bath modes, $J(\omega)$ would still remain an open question; actually, in the experimental study of mesoscopic systems one might be able to manipulate the spectral density $J(\omega)$, to some extent, in his own way. In this paper, we would like to discuss the second law for various damping models. We will first show the validity of the second law for a discrete distribution of bath modes by exactly proving the second-law inequality in a simple form obtained from the general treatment of the susceptibility (see Sect. 3). Subsequently, the inequality will be appropriately applied for various continuous distributions of bath modes. It is then found that for damping models with cut-off frequencies, the second law holds, whereas interestingly, we may have its violation for damping models with cut-off frequency-free $J(\omega)$, which are, however, physically unrealistic (see Sect. 4). Let us begin with a brief review on the basics of the quantum Brownian motion. We will below adopt the notations used in [12].

2 Basics and its general treatment

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

$$\hat{H} = \hat{H}_s + \hat{H}_b + \hat{H}_{sb}, \quad (1)$$

where

$$\begin{aligned} \hat{H}_s &= \frac{\hat{p}^2}{2M} + \frac{M}{2} \omega_0^2 \hat{q}^2; \quad \hat{H}_b = \sum_{j=1}^N \left(\frac{\hat{p}_j^2}{2m_j} + \frac{m_j}{2} \omega_j^2 \hat{x}_j^2 \right) \\ \hat{H}_{sb} &= -\hat{q} \sum_{j=1}^N c_j \hat{x}_j + \hat{q}^2 \sum_{j=1}^N \frac{c_j^2}{2m_j \omega_j^2}. \end{aligned} \quad (2)$$

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

$$\omega_1 \leq \omega_2 \leq \dots \leq \omega_{N-1} \leq \omega_N. \quad (3)$$

By means of the Heisenberg equation of motion for \hat{p} we can derive the quantum Langevin equation

$$M \ddot{\hat{q}} + M \int_0^t ds \gamma(t-s) \dot{\hat{q}}(s) + M \omega_0^2 \hat{q} = \hat{\xi}(t), \quad (4)$$

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

$$\begin{aligned} \gamma(t) &= \frac{1}{M} \sum_{j=1}^N \frac{c_j^2}{m_j \omega_j^2} \cos(\omega_j t); \quad \hat{\xi}(t) = -M\gamma(t)\hat{q}(0) \\ &+ \sum_{j=1}^N c_j \left\{ \hat{x}_j(0) \cos(\omega_j t) + \frac{\hat{p}_j(0)}{m_j \omega_j} \sin(\omega_j t) \right\}. \end{aligned} \quad (5)$$

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

$$J(\omega) = \pi \sum_{j=1}^N \frac{c_j^2}{2m_j \omega_j} \delta(\omega - \omega_j), \quad (6)$$

we can express the damping kernel as

$$\gamma(t) = \frac{2}{M} \int_0^\infty \frac{d\omega}{\pi} \frac{J(\omega)}{\omega} \cos(\omega t). \quad (7)$$

Let us apply the Laplace transform to equation (4) with the aid of [13,14]

$$\mathcal{L}\{\cos(\omega_j t)\}(s) = \frac{s}{s^2 + \omega_j^2}, \quad (8)$$

$$\mathcal{L}\{\sin(\omega_j t)\}(s) = \frac{\omega_j}{s^2 + \omega_j^2}. \quad (9)$$

With $s = -i\omega + 0^+ = -i(\omega + i0^+)$ we then easily obtain

$$\begin{aligned} \hat{q}_\omega &:= \mathcal{L}\{\hat{q}(t)\}(-i\omega + 0^+) \\ &= \tilde{\chi}(\omega) \left[\hat{\xi}_\omega - i\omega M \{1 + \tilde{\gamma}(\omega)\} \hat{q}(0) + M\dot{\hat{q}}(0) \right], \end{aligned} \quad (10)$$

where the Laplace-transformed damping kernel, the dynamic susceptibility, and the Laplace-transformed noise operator are, respectively, given by

$$\tilde{\gamma}(\omega) = \frac{i\omega}{M} \sum_{j=1}^N \frac{c_j^2}{m_j \omega_j^2} \frac{1}{\omega^2 - \omega_j^2}, \quad (11)$$

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

$$\hat{\xi}_\omega = \sum_{j=1}^N \frac{c_j}{\omega^2 - \omega_j^2} \left\{ i\omega \hat{x}_j(0) - \frac{\hat{p}_j(0)}{m_j} \right\} - M\tilde{\gamma}(\omega) \hat{q}(0).$$

Substituting (11) into (12), we get

$$\tilde{\chi}(\omega) = \frac{-\frac{1}{M} \prod_{j=1}^N (\omega^2 - \omega_j^2)}{D_{\tilde{\chi}}(\omega)}, \quad (13)$$

where

$$D_{\tilde{\chi}}(\omega) = \prod_{j=0}^N (\omega^2 - \omega_j^2) - \frac{\omega^2}{M} \sum_{j=1}^N \frac{c_j^2}{m_j \omega_j^2} \prod_{\substack{j'=1 \\ (j' \neq j)}}^N (\omega^2 - \omega_{j'}^2). \quad (14)$$

It is known [15] that the susceptibility $\tilde{\chi}(\omega)$ in (12) has poles at the normal-mode frequencies of the total system \hat{H} in (1), $\pm \bar{\omega}_k$ with $k = 0, 1, 2, \dots, N$, so that

$$\omega_0^2 - \bar{\omega}_k^2 - i\bar{\omega}_k \tilde{\gamma}(\bar{\omega}_k) = 0. \quad (15)$$

Here, we might be able to say that a specific $k = k_0$ would represent the ‘‘system harmonic oscillator’’ with the normal-mode frequency $\bar{\omega}_{k_0}$, *uncoupled* to the ‘‘bath’’ consisting of the remaining oscillators with $\bar{\omega}_k$, where $k \neq k_0$. From equations (12), (13), and (15), we have a compact expression of the susceptibility,

$$\tilde{\chi}(\omega) = -\frac{1}{M} \frac{\prod_{j=1}^N (\omega^2 - \omega_j^2)}{\prod_{k=0}^N (\omega^2 - \bar{\omega}_k^2)}. \quad (16)$$

Without any loss of generality, we here assume that

$$\bar{\omega}_0 \leq \bar{\omega}_1 \leq \dots \leq \bar{\omega}_{N-1} \leq \bar{\omega}_N. \quad (17)$$

The damping function $\tilde{\gamma}(\omega)$ in the frequency domain has, besides equation (11), another expression which is suitable for the case of a continuous distribution of bath modes; from equations (7) and (8) we obtain

$$\tilde{\gamma}(\omega) = \frac{i}{M} \int_0^\infty \frac{d\omega'}{\pi} \frac{J(\omega')}{\omega'} \left(\frac{1}{\omega' + \omega} - \frac{1}{\omega' - \omega} \right), \quad (18)$$

$$\tilde{\gamma}(\omega) \Big|_{\omega+i0^+}^{\omega-i0^+} = \frac{J(\omega)}{M\omega} + \frac{i}{M} \int_0^\infty \frac{d\omega'}{\pi} \frac{J(\omega')}{\omega'} P \left(\frac{1}{\omega' + \omega} - \frac{1}{\omega' - \omega} \right). \quad (19)$$

We here used the well-known formula $1/(x + i0^+) = P(1/x) - i\pi\delta(x)$ for $x = \omega' - \omega$. For the simple Ohmic case $J_0(\omega) = M\gamma_0\omega$ with an ω -independent constant γ_0 , we easily have $\gamma_0(t) = 2\gamma_0\delta(t)$, and $\tilde{\gamma}_0(\omega) = \gamma_0$ with a vanishing principal (or imaginary) part in (19), while for the Drude model where $J_d(\omega) = M\gamma_0\omega\omega_d^2/(\omega^2 + \omega_d^2)$ with a cut-off frequency ω_d , we have $\gamma_d(t) = \gamma_0\omega_d e^{-\omega_d t}$, and

$$\tilde{\gamma}_d(\omega) = \frac{\gamma_0\omega_d^2}{\omega^2 + \omega_d^2} + i \frac{\gamma_0\omega\omega_d}{\omega^2 + \omega_d^2} = \frac{\gamma_0\omega_d}{\omega_d - i\omega}. \quad (20)$$

For a later purpose, it is interesting to compare $D_{\tilde{\chi}}(\omega)$ in equation (14) with the denominator of the right hand side in (16). Then, we can easily find that

$$\sum_{k=0}^N \bar{\omega}_k^2 = \sum_{j=0}^N \omega_j^2 + \gamma(0); \quad \prod_{k=0}^N \bar{\omega}_k^2 = \prod_{j=0}^N \omega_j^2. \quad (21)$$

Here, $\gamma(0) = \gamma(t)|_{t=0} \geq 0$ in equation (5). From this comparison of the denominators at $\omega = \omega_N$, we also obtain $D_{\tilde{\chi}}(\omega_N) \leq 0$ and so $\omega_N \leq \bar{\omega}_N$. Similarly, we can

acquire both $D_{\tilde{\chi}}(\omega_1) \leq 0$ for N odd and $D_{\tilde{\chi}}(\omega_1) \geq 0$ for N even, which lead to the fact that $\bar{\omega}_0 \leq \omega_1$ for any given N . Further, we can obtain the relationship, $D_{\tilde{\chi}}(\omega_j) \cdot D_{\tilde{\chi}}(\omega_{j-1}) \leq 0$ for any j . Therefore, it is found that

$$\bar{\omega}_0 \leq \omega_1 \leq \bar{\omega}_1 \leq \dots \leq \omega_{N-1} \leq \bar{\omega}_{N-1} \leq \omega_N \leq \bar{\omega}_N. \quad (22)$$

By using $D_{\tilde{\chi}}(\omega_0)$, we can also show that $\bar{\omega}_0 \leq \omega_0 \leq \bar{\omega}_N$ (see also [16]). Within this *general* treatment of the susceptibility, we would like to consider the quantum second law below.

3 General validity of the quantum second law (discrete bath modes)

The energy of the system oscillator \hat{H}_s at zero temperature can be calculated by means of the partition function $Z = \text{Tr} e^{-\beta\hat{H}}$ with $\beta = 1/k_B T$ as

$$\langle \hat{H}_s \rangle_{T=0} = \frac{\text{Tr} \left(\hat{H}_s e^{-\beta\hat{H}} \right)}{Z} \Big|_{\beta \rightarrow \infty} =: E_s(0). \quad (23)$$

It is well-known [7,17] that the system-bath entanglement induced by the coupling term \hat{H}_{sb} in (1) leads to the fact that the system oscillator \hat{H}_s , initially in a pure state (here, its ground state with the minimum energy $E_g = \hbar\omega_0/2$), is not in the pure state any longer but in a mixed state with a fluctuation in energy, and so we actually have $E_s(0) > E_g$. It was even discussed in [18] that the energy fluctuation measurements can provide entanglement information (cf. for a thermodynamical approach to quantifying entanglement in bipartite qubit states, see [19]). From the fluctuation-dissipation theorem [12], we can also easily obtain

$$E_s(0) = \frac{M\hbar}{2\pi} \int_0^\infty d\omega (\omega_0^2 + \omega^2) \text{Im} \tilde{\chi}(\omega + i0^+). \quad (24)$$

The factor $\text{Im} \tilde{\chi}(\omega + i0^+)$ can be evaluated from equation (16) with $\omega \rightarrow \omega + i0^+$. By means of the technique used, e.g., in [20], equation (24) can be rewritten as

$$E_s(0) = \frac{\hbar}{2} \frac{1}{2\pi i} \oint d\omega \frac{\omega_0^2 + \omega^2}{G(\omega)}, \quad (25)$$

where $G(\omega) = -1/\{M\tilde{\chi}(\omega)\}$, and 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 $(-i\epsilon, \infty - i\epsilon)$. Therefore, $E_s(0)$ can be exactly obtained in closed form from the residues evaluated at all zeroes of $G(\omega)$ on the positive real axis. It is also interesting to note that the entanglement between any pair of the bath oscillators $\hat{H}_j = \hat{p}_j^2/2m_j + m_j\omega_j^2 \hat{x}_j^2/2$ with $j = 1, 2, 3, \dots, N$ is induced by the system-bath entanglement and the well-known entanglement swapping [21]. As a result, we must obtain an excess energy for any j , i.e.,

$\langle \hat{H}_j \rangle_{T=0} > \hbar \omega_j / 2$. However, the energy of the total system, $\langle \hat{H} \rangle_{T=0} = \sum_{k=0}^N \hbar \bar{\omega}_k / 2$ is clearly not equivalent to $\langle \hat{H}_s \rangle_{T=0} + \sum_{j=1}^N \langle \hat{H}_j \rangle_{T=0} = \langle \hat{H}_s + \hat{H}_b \rangle_{T=0}$.

The minimum work required to couple a harmonic 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 [1,11]. The Helmholtz free energy can be obtained from the canonical partition function $Z_s(\beta) = \text{Tr} e^{-\beta \hat{H}} / \text{Tr}_b e^{-\beta \hat{H}_b}$ as $F(T) = -k_B T \ln Z_s$, where 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 $\bar{\omega}_k$ the partition function can be rewritten as

$$Z_s(\beta) = \frac{\prod_{k=0} \sum_{n_k=0} e^{-\beta \hbar \bar{\omega}_k (n_k + \frac{1}{2})}}{\prod_{j=1} \sum_{n_j=0} e^{-\beta \hbar \omega_j (n_j + \frac{1}{2})}} \quad (26)$$

so that we can easily get, for $\beta \rightarrow \infty$,

$$F(0) = \frac{\hbar}{2} \left(\sum_{k=0}^N \bar{\omega}_k - \sum_{j=1}^N \omega_j \right). \quad (27)$$

With the aid of equation (21), it is evidently found that $F(0) > \hbar \omega_j / 2$ for any $j = 0, 1, 2, \dots, N$. Further, we have, from [22],

$$F(T) = \frac{1}{\pi} \int_0^\infty d\omega f(\omega, T) \text{Im} \left\{ \frac{d}{d\omega} \ln \chi(\omega + i0^+) \right\}, \quad (28)$$

where $f(\omega, T) = k_B T \ln \{ 2 \sinh(\hbar \omega / 2 k_B T) \}$. Similarly to equation (25), we can obtain an integral form of the free energy at $T = 0$,

$$F(0) = \frac{\hbar}{2} \frac{1}{2\pi i} \oint d\omega \frac{\omega G'(\omega)}{G(\omega)}. \quad (29)$$

Here, $f(\omega, 0) = \hbar \omega / 2$.

Now, we are in a position to exactly formulate the quantum second law within this general treatment; from equations (25) and (29) with (12), we easily find an expression

$$K := F(0) - E_s(0) = \frac{\hbar}{4\pi} \oint d\omega \frac{\omega^2 \tilde{\gamma}'(\omega)}{G(\omega)}, \quad (30)$$

and, for the validity of the second law, we have to get $K \geq 0$ for any N (the number of the bath oscillators) and the limit $N \rightarrow \infty$. Here, K can exactly be evaluated from all residues of the integrand on the positive *real* axis. Substituting (16) with $\tilde{\chi}(\omega) = -1/MG(\omega)$ and (11) into (30), we obtain, after a fairly lengthy evaluation of the contour integration (see Appendix A for details), the exact result

$$K = \frac{\hbar}{8M} \sum_{k=0}^N \mathcal{A}_k, \quad (31)$$

where

$$\mathcal{A}_k = \bar{\omega}_k \frac{\prod_{j=1}^N (\bar{\omega}_k^2 - \omega_j^2)}{\prod_{\substack{k'=0 \\ (k' \neq k)}}^N (\bar{\omega}_k^2 - \bar{\omega}_{k'}^2)} \times \sum_{l=1}^N \frac{c_l^2}{m_l \omega_l^2} P \left\{ \frac{1}{(\omega_l + \bar{\omega}_k)^2} + \frac{1}{(\omega_l - \bar{\omega}_k)^2} \right\}. \quad (32)$$

Considering each summand \mathcal{A}_k from $k = N$ with keeping in mind the frequency relationship in (22), we see that each of the summand is non-negative and so $K \geq 0$ indeed! Separately from this result for discrete bath modes, we will next discuss the second law for continuous bath modes. For doing this job, we will consider a continuation of the spectral density $J(\omega)$ from its original form in (6).

4 The second law for continuous bath modes

For a discussion of the second law for a continuous distribution of bath modes, we rewrite equation (30) as

$$K = \frac{\hbar}{4\pi} \left(\int_0^\infty d\omega \frac{\omega^2 \tilde{\gamma}'_-(\omega)}{G_-(\omega)} - \int_0^\infty d\omega \frac{\omega^2 \tilde{\gamma}'_+(\omega)}{G_+(\omega)} \right), \quad (33)$$

where the subscripts $+/-$ denote the branches $(\infty + i\epsilon, i\epsilon)$ and $(-i\epsilon, \infty - i\epsilon)$, respectively, so that

$$G_+(\omega) := G(\omega) \Big|_{\omega+i0^+} = \omega^2 - \omega_0^2 + i \frac{J(\omega)}{M} - \frac{\omega}{M} \int_0^\infty \frac{d\omega'}{\pi} \frac{J(\omega')}{\omega'} P \left(\frac{1}{\omega' + \omega} - \frac{1}{\omega' - \omega} \right), \quad (34)$$

and $G_-(\omega) := G(\omega) \Big|_{\omega-i0^+} = G_+^*(\omega)$. Here, we used $G(\omega) = \omega^2 - \omega_0^2 + i\omega \tilde{\gamma}(\omega)$ with equation (19) for $\tilde{\gamma}_+(\omega)$. Therefore, equation (33) easily reduces to

$$K = \frac{\hbar}{2\pi} \text{Im} \int_0^\infty d\omega \frac{\omega^2 R'_+(\omega)}{G_+(\omega)}, \quad (35)$$

where $R_+(\omega) = -i \tilde{\gamma}_+(\omega)$.

First, for the Ohmic case, $(\tilde{\gamma}_+)_{0}(\omega) = \gamma_o$, which is the prototype for damping, we easily obtain $K_0 = 0$. In fact, both $(E_s)_0(0)$ and $F_0(0)$ have the logarithmic divergence, however, the same value, namely,

$$(E_s)_0(0) = F_0(0) = \frac{\hbar \gamma_o}{2\pi} \int_0^\infty d\omega \frac{\omega(\omega^2 + \omega_0^2)}{(\omega^2 - \omega_0^2)^2 + (\gamma_o \omega)^2} \quad (36)$$

(see also the discussion in the last paragraphs of Sects. 4.1 and 4.2). However, the Ohmic model is not so realistic in its strict form because the spectral density of bath modes, $J_0(\omega) = M \gamma_o \omega$ diverges for large frequencies. We therefore introduce a cut-off frequency ω_c which leads to $J_c(\omega)$ decaying smoothly to zero for large frequencies $\omega > \omega_c$.

We will first consider the Drude model, where $J_d(\omega)$ is polynomially decaying for $\omega > \omega_c = \omega_d$, and next a damping model with $J_e(\omega)$ being exponentially decaying for $\omega > \omega_c = \omega_e$. For these damping models, we will be able to show that $K > 0$. Subsequently, we will also consider two different damping models without cut-off frequencies ω_c ; first, the extended Ohmic models where the spectral densities $J(\omega)$ diverge polynomially faster than $J_0(\omega)$, and secondly, the extended Drude models with $J_{d,n}(\omega)$ diverging faster or more slowly than $J_0(\omega)$. Interestingly, we will observe $K < 0$ for some of the cut-off frequency-free damping models (see Sects. 4.3 and 4.4).

4.1 Drude model (d)

We briefly review the second law in the Drude model considered in [11]; it is convenient to adopt, in place of $(\omega_0, \omega_d, \gamma_o)$, the parameters $(\mathbf{w}_0, \Omega, \gamma)$ through the relations

$$\begin{aligned} \omega_0^2 &:= \mathbf{w}_0^2 \frac{\Omega}{\Omega + \gamma}; \omega_d := \Omega + \gamma; \\ \gamma_o &:= \gamma \frac{\Omega(\Omega + \gamma) + \mathbf{w}_0^2}{(\Omega + \gamma)^2}. \end{aligned} \quad (37)$$

Substituting equation (20) with (37) into equation (12), we obtain the susceptibility

$$\tilde{\chi}_d(\omega) = -\frac{1}{M} \frac{\omega + i\omega_d}{\omega^3 + i\omega_d\omega^2 - (\omega_0^2 + \gamma_o\omega_d)\omega - i\omega_0^2\omega_d} \quad (38)$$

$$= -\frac{1}{M} \frac{\omega + i(\Omega + z_1 + z_2)}{(\omega + i\Omega)(\omega + iz_1)(\omega + iz_2)}, \quad (39)$$

where $z_1 = \gamma/2 + i\mathbf{w}_1$ and $z_2 = \gamma/2 - i\mathbf{w}_1$ with $\mathbf{w}_1^2 = \mathbf{w}_0^2 - (\gamma/2)^2$. This gives us $(G_+)_d(\omega) = -1/\{M \tilde{\chi}_d(\omega)\}$ for equation (35). By means of equation (39), we can even obtain the closed expressions for both $(E_s)_d(0)$ from (24) and $F_d(0)$ from (28). We give the detailed derivation of these expressions in Appendix B, which will also be used in Section 4.4. It has been numerically shown in [11] that $(E_s)_d(0)$ in (65) is actually greater than $E_g = \frac{\hbar \mathbf{w}_0}{2} \sqrt{\frac{\Omega}{\Omega + \gamma}}$, and $F_d(0)$ in (68) is even greater than the $(E_s)_d(0)$, i.e., $K_d > 0$. For a later purpose, we will also evaluate K_d explicitly for various pairs (ω_0, ω_d) (see Tab. 2 in Sect. 4.4).

It is noted that in the limit $\omega_d \rightarrow \infty$ (equivalently, $\Omega \rightarrow \infty$), we have $K_d \rightarrow \frac{\gamma}{\pi \mathbf{w}_0} E_g$ (see Appendix B). From the comparison between $\tilde{\gamma}_d(\omega)$ and $\tilde{\gamma}_0(\omega)$ (or, equivalently, $J_d(\omega)$ and $J_0(\omega)$), this result would be interpreted as $K_0 \rightarrow \frac{\gamma}{\pi \mathbf{w}_0} E_g$. However, it is misleading; $\tilde{\gamma}_d(\omega)$ behaves only for small frequencies, $\omega \ll \omega_d$, like in the Ohmic case, which corresponds to $\gamma_d(t) \rightarrow \gamma_0(t)$ only for large times. Actually, $\gamma_d(t) = \gamma_o \omega_d e^{-\omega_d t}$ with $\omega_d \rightarrow \infty$ does not reduce to $\gamma_0(t) = 2\gamma_o \delta(t) = \lim_{\omega_d \rightarrow \infty} \frac{2}{\sqrt{\pi}} \gamma_o \sqrt{\omega_d} e^{-\omega_d t^2}$. For the evaluation of K , however, all frequencies, $0 \leq \omega < \infty$, have to be considered. Therefore, we evidently get $\lim_{\omega_d \rightarrow \infty} K_d \gtrsim K_0 = 0$.

Table 1. K_e/E_g for various pairs (γ_o, ω_e) , where $E_g = \frac{\hbar}{2}$ (i.e., $\omega_0 = 1$); $\lim_{\omega_e \rightarrow \infty} K_e/E_g = \gamma_o/\pi$.

ω_e	$\gamma_o = 0.5$	$\gamma_o = 1$	$\gamma_o = 2$	$\gamma_o = 5$
0.5	0.04225	0.08186	0.15604	0.34038
1	0.06130	0.11838	0.22117	0.47348
5	0.10600	0.20348	0.37899	0.81614
10	0.12131	0.23326	0.43819	0.96224
50	0.14414	0.28018	0.54302	1.25567
80	0.14789	0.28896	0.56377	1.32020
∞	0.15915	0.31831	0.63662	1.59155

4.2 Exponentially decaying model (e)

We now consider a damping model with $J_e(\omega) = M \gamma_o \omega e^{-\omega/\omega_e}$ which, in the limit $\omega_e \rightarrow \infty$, clearly reduces to $J_0(\omega)$ for small frequencies. Substituting this into equation (7), we can obtain

$$\gamma_e(t) = \frac{2}{\pi} \frac{\gamma_o \omega_e}{1 + (\omega_e t)^2}. \quad (40)$$

Applying the Laplace transform [14] to equation (40) with $s = -i\omega + 0^+$, it can be found that

$$\begin{aligned} \tilde{\gamma}_e(\omega) &= \gamma_o e^{-\omega/\omega_e} \\ &+ i \frac{\gamma_o}{\pi} \left\{ e^{\omega/\omega_e} E_1\left(\frac{\omega}{\omega_e}\right) + e^{-\omega/\omega_e} \text{Ei}\left(\frac{\omega}{\omega_e}\right) \right\}, \end{aligned} \quad (41)$$

(see Appendix C for the detailed derivation). By using this with $E_1'(y) = -E_0(y) = -e^{-y}/y$, we can easily get $(R'_+)_e(\omega)$ and $(G_+)_e(\omega)$, and then introducing a dimensionless variable $\lambda = \omega/\omega_e$, we arrive at the expression

$$K_e = \frac{\hbar \gamma_o \omega_e^2}{2\pi^2} \text{Im} \int_0^\infty d\lambda \frac{f_1(\lambda)}{f_2(\lambda)}, \quad (42)$$

where

$$f_1(\lambda) = \lambda^2 \{ e^\lambda E_1(\lambda) - e^{-\lambda} \text{Ei}(\lambda) + i\pi e^{-\lambda} \}, \quad (43)$$

$$\begin{aligned} f_2(\lambda) &= \omega_e^2 \lambda^2 - \omega_0^2 - \frac{\gamma_o \omega_e}{\pi} \lambda \{ e^\lambda E_1(\lambda) + e^{-\lambda} \text{Ei}(\lambda) \} \\ &+ i\omega_e \gamma_o \lambda e^{-\lambda}. \end{aligned} \quad (44)$$

We numerically evaluate the integration in (42) for various pairs (γ_o, ω_e) to show that $K_e > 0$ (see Tab. 1).

From the fact that $\tilde{\gamma}_e(\omega)$ behaves like in the Ohmic case for small frequencies $\omega \ll \omega_e$, it is also interesting to consider the leading behavior of K_e for $\omega_e \rightarrow \infty$; from equation (42) we can easily get $\lim_{\omega_e \rightarrow \infty} K_e = \frac{\hbar \gamma_o}{2\pi} \neq 0$, which is also different from $\lim_{\omega_d \rightarrow \infty} K_d$ in Section 4.1. This confirms that these limiting values cannot reveal the Ohmic counterpart K_0 .

4.3 Extended Ohmic models (p)

Let us consider damping models with $J_p(\omega) = M \gamma_o \omega (\omega/\gamma_o)^p$ being polynomially divergent with ω .

Clearly, the case of $p = 0$ is Ohmic. First, we have $J_1(\omega) = M\omega^2$. By using the relationship $\int_0^\infty dy e^{iky} = \pi \delta(k) + iP(1/k)$, we can easily obtain $\gamma_1(t) = -\frac{2}{\pi} P \frac{1}{t^2}$, which leads to no well-defined $\tilde{\gamma}_1(\omega)$. This ($p = 1$) is, therefore, physically not acceptable. It is not difficult to show that the cases of p being odd are not acceptable.

Next, we consider the case of $p = 2$. It can be shown that $\gamma_2(t) = -\frac{2}{\gamma_o} \delta''(t)$ and $(\tilde{\gamma}_+)_2(\omega) = \frac{\omega^2}{\gamma_o} - i \frac{2\delta(0)}{\gamma_o} \omega$. By using this for equation (35), we can obtain

$$K_2 = \frac{\hbar}{2\pi} \text{Im} \int_0^\infty d\omega \frac{2\omega^2 \{-\omega + i\delta(0)\}}{\omega^3 - i\alpha\omega^2 + i\beta}, \quad (45)$$

where $\alpha = \gamma_o + 2\delta(0)$ and $\beta = \omega_0^2 \gamma_o$. The integral in (45) diverges logarithmically. This divergence is, obviously, from the fact that both $(E_s)_2(0)$ and $F_2(0)$ diverge logarithmically, however, differently from the Ohmic case, $(E_s)_2(0) \neq F_2(0)$. In fact, we find that $K_2 = -\frac{\hbar}{\pi} \{\delta(0) + \gamma_o\} \times \infty < 0$, which clearly means that the excess energy, $(E_s)_2(0)$, is greater than the minimum work (or the work in a reversible process), $F_2(0)$, required to couple a system to a bath. This violation of the second law in the reversible process may be understood to emerge from a large amount of the energy offer by the bath with $J_2(\omega)$ diverging with ω . The infinite value of K_2 suggests, however, that this model would be strictly unrealistic.

4.4 Extended Drude models (d, n)

We now consider a more general class of the spectral density than $J_d(\omega)$, which is

$$J_{d,n}(\omega) = \left(\frac{\omega}{\omega_d}\right)^n J_d(\omega) = M\gamma_o \frac{\omega^{n+1}}{\omega_d^{n-2}(\omega^2 + \omega_d^2)}. \quad (46)$$

Let us begin with n being odd. First, $n = 1$. We then have $J_{d,1}(\omega) = M\gamma_o \omega_d \omega^2 / (\omega^2 + \omega_d^2)$, which converges to a non-zero constant $M\gamma_o \omega_d$ for large frequencies. Substituting this into equation (7), we can obtain, after some calculation (see Appendix C for details),

$$\gamma_{d,1}(t) = \frac{\gamma_o \omega_d}{\pi} [\{\text{Ei}(\omega_d t) + E_1(\omega_d t)\} \sinh(\omega_d t) - \{\text{Ei}(\omega_d t) - E_1(\omega_d t)\} \cosh(\omega_d t)]. \quad (47)$$

Applying the Laplace transform [14] to this, we can get

$$(\tilde{\gamma}_+)_d(\omega) = \frac{\gamma_o \omega_d \omega}{\omega^2 + \omega_d^2} + i \frac{2}{\pi} \frac{\gamma_o \omega_d \omega}{\omega^2 + \omega_d^2} \ln \left(\frac{\omega}{\omega_d}\right). \quad (48)$$

Using equation (34) with (48), we can arrive at the expression in (35)

$$K_{d,1} = \frac{\hbar \gamma_o}{2\pi} \text{Im} \int_0^\infty d\lambda \frac{g_1(\lambda)}{g_2(\lambda)}, \quad (49)$$

Table 2. $K_d \pi / \gamma_o E_g$ from (71) versus $K_{d,1} \pi / \gamma_o E_g$ from (49) for various pairs (ω_0, ω_d) with $\gamma_o = 1$, where $E_g = \frac{\hbar \omega_0}{2}$; $(d, 0)$ denotes the Drude model.

(ω_0, ω_d)	$(d, 0)$	$(d, 1)$	(ω_0, ω_d)	$(d, 0)$	$(d, 1)$
(0.5, 0.5)	0.84275	0.50184	(1, 0.5)	1.21124	0.25468
(0.5, 1)	0.61942	0.45203	(1, 1)	0.45318	0.26521
(0.5, 5)	1.29483	0.34454	(1, 5)	0.61427	0.17800
(0.5, 10)	1.52266	0.24507	(1, 10)	0.73767	0.05454
(5, 0.5)	0.53089	0.02832	(10, 0.5)	0.28968	0.00936
(5, 1)	0.47010	0.04106	(10, 1)	0.27637	0.01345
(5, 5)	0.09790	0.05476	(10, 5)	0.13428	0.02346
(5, 10)	0.09537	0.04976	(10, 10)	0.04792	0.02347

where

$$\begin{aligned} g_1(\lambda) &= \lambda^2 \left[\frac{2\lambda_d}{\pi} \{(\lambda_d^2 - \lambda^2) \ln \left(\frac{\lambda}{\lambda_d}\right) + \lambda^2 + \lambda_d^2\} \right. \\ &\quad \left. + i\lambda_d(\lambda^2 - \lambda_d^2) \right], \\ g_2(\lambda) &= (\lambda^2 + \lambda_d^2) \left\{ (\lambda^2 - \lambda_0^2) (\lambda^2 + \lambda_d^2) \right. \\ &\quad \left. - \frac{2\lambda_d \lambda^2}{\pi} \ln \left(\frac{\lambda}{\lambda_d}\right) + i\lambda_d \lambda^2 \right\}. \end{aligned} \quad (50)$$

Here, we introduced a dimensionless variable $\lambda = \omega / \gamma_o$ with $\lambda_0 = \omega_0 / \gamma_o$ and $\lambda_d = \omega_d / \gamma_o$. We numerically evaluate $K_{d,1}$ for various pairs (λ_0, λ_d) to show that $K_{d,1} > 0$ (see Tab. 2). It is also noted that in the limit λ_d (or ω_d) $\rightarrow \infty$, the spectral density $J_{d,1}(\omega)$ with $\gamma_o = \omega_d$ reduces to $J_1(\omega)$ in Section 4.3 for small frequencies. As was discussed, however, this case $(d, 1)$ is a well-defined damping model whereas the model with $J_1(\omega)$ is not.

Let $n = 3$ next. We have $J(\omega)_{d,3} = M\gamma_o \omega_d^{-1} \omega^4 / (\omega^2 + \omega_d^2)$. After a straightforward calculation, we will obtain

$$\gamma_{d,3}(t) = -\gamma_{d,1}(t) - \frac{2\gamma_o P}{\pi \omega_d t^2}, \quad (51)$$

which indicates that this case is physically not acceptable. Similarly, we can also show that all cases for $n > 3$ being odd are not acceptable.

Now, let n be even. We can then find that

$$\gamma_{d,2m}(t) = (-1)^m \left\{ \gamma_d(t) - \sum_{j=1}^m \frac{\gamma_0^{\{2(j-1)\}}(t)}{\omega_d^{2(j-1)}} \right\}, \quad (52)$$

where $m = 1, 2, \dots$, and $\gamma_0^{\{2(j-1)\}}(t)$ represent $2(j-1)$ -time derivatives of $\gamma_0(t)$. We begin with a simple case ($m = 1$) with $J_{d,2}(\omega) = M\gamma_o \omega^3 / (\omega^2 + \omega_d^2)$. This case is particularly interesting because $J_{d,2}(\omega)$ diverges for large frequencies, however, more slowly than $J_0(\omega)$ for the Ohmic case, whereas all $J_{d,2m}(\omega)$ for $m > 1$ diverge faster than $J_0(\omega)$; $J_{d,2}(\omega)$ may be said to be of *weak* divergence. Due to the fact that $K_{d,0} > K_{d,1} > 0$ seen from Table 2, we would like to pose a question if we will here obtain $K_{d,1} > K_{d,2} > 0 = K_0$ or $K_{d,1} > 0 \geq K_{d,2}$. In fact, we have an interesting relation $(\tilde{\gamma}_+)_d(\omega) = -\tilde{\gamma}_d(\omega) + \tilde{\gamma}_0(\omega)$

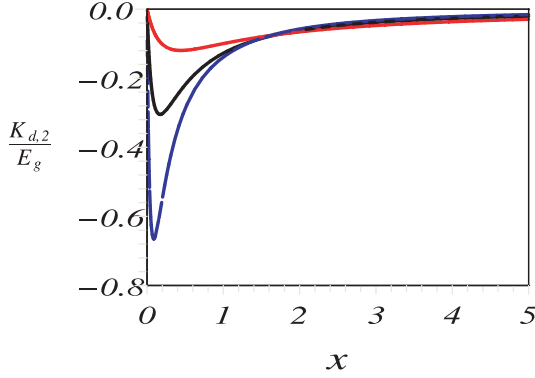


Fig. 1. $K_{d,2}/E_g$ versus $x = \gamma/\mathbf{w}_0$, where the ground state energy $E_g = \frac{\hbar}{2} \sqrt{\Omega\gamma + \mathbf{w}_0^2}$; for $\Omega = 2\mathbf{w}_0$ (dot), $5\mathbf{w}_0$ (solid), $10\mathbf{w}_0$ (dashed) from top to bottom.

from equation (52), and so $(R'_+)'_{d,2}(\omega) = -(R'_+)'_d(\omega)$ for equation (35). Introducing the parameters $(\mathbf{w}_0, \Omega, \gamma)$ defined as the relations

$$\omega_d \mathbf{w}_0^2 := \Omega \mathbf{w}_0^2; \omega_d + \gamma_o := \Omega + \gamma; \omega_0^2 := \Omega\gamma + \mathbf{w}_0^2 \quad (53)$$

(note that these differ from the relations in (37)), we can easily obtain

$$\begin{aligned} (\tilde{G}_+)'_{d,2}(\omega) &= \frac{\omega^3 + i(\gamma_o + \omega_d)\omega^2 - \omega_0^2\omega - i\omega_0^2\omega_d}{\omega + i\omega_d} \\ &= \frac{(\omega + i\Omega)(\omega + iz_1)(\omega + iz_2)}{\omega + i\Omega\mathbf{w}_0^2/(\Omega\gamma + \mathbf{w}_0^2)}, \end{aligned} \quad (54)$$

where $z_1 = \frac{\gamma}{2} + i\mathbf{w}_1$ and $z_2 = \frac{\gamma}{2} - i\mathbf{w}_1$ with $\mathbf{w}_1^2 = \mathbf{w}_0^2 - (\frac{\gamma}{2})^2$. By using equations (35) and (54), we arrive at the expression

$$K_{d,2} = \frac{\hbar}{2\pi} \frac{\Omega\mathbf{w}_0^2}{(\Omega\gamma + \mathbf{w}_0^2)(\Omega\gamma - \Omega^2 - \mathbf{w}_0^2)} C(\mathbf{w}_0, \Omega, \gamma) \quad (55)$$

where

$$\begin{aligned} C(\mathbf{w}_0, \Omega, \gamma) &= \gamma(\mathbf{w}_0^2 - \Omega^2) \frac{1}{\mathbf{w}_1} \arctan \frac{2\mathbf{w}_1}{\gamma} \\ &\quad + (\Omega^2 + \mathbf{w}_0^2 - \Omega\gamma) \ln(\Omega\gamma + \mathbf{w}_0^2) \\ &\quad - 2(\Omega^2 + \mathbf{w}_0^2) \ln \mathbf{w}_0 + 2\Omega\gamma \ln \Omega. \end{aligned} \quad (56)$$

In case that \mathbf{w}_1 is complex-valued ($\mathbf{w}_0 < \gamma/2$), i.e., for the overdamped case, this has to be understood in terms of the relation, $\frac{1}{\mathbf{w}_1} \arctan \frac{2\mathbf{w}_1}{\gamma} = \frac{1}{2\mathbf{w}_1} \ln \left(\frac{\gamma + 2\mathbf{w}_1}{\gamma - 2\mathbf{w}_1} \right)$, where $\bar{\mathbf{w}}_1^2 = (\frac{\gamma}{2})^2 - \mathbf{w}_0^2$.

Interestingly enough, we can here observe $K_{d,2} < 0$ (see Fig. 1), which would allow us to have a violation of the second law in the reversible process for this damping model. This negativity may be understood from the comparison, with the aid of the relation $(R'_+)'_{d,2}(\omega) = -(R'_+)'_d(\omega)$, between

$$\begin{aligned} K_{d,2} &= \frac{\hbar}{2\pi} \omega_d \gamma_o \operatorname{Im} \int_0^\infty \frac{d\omega}{\omega^2} \\ &\quad \times \frac{1}{(\omega + i\omega_d)(\omega + i\Omega)(\omega + iz_1)(\omega + iz_2)} \end{aligned} \quad (57)$$

with $(\mathbf{w}_0, \Omega, \gamma)$ in equation (37) and $\omega_d = \Omega + \gamma$, and

$$\begin{aligned} K_d &= -\frac{\hbar}{2\pi} \omega_d \gamma_o \operatorname{Im} \int_0^\infty \frac{d\omega}{\omega^2} \\ &\quad \times \frac{1}{(\omega + i\omega_d)(\omega + i\Omega)(\omega + iz_1)(\omega + iz_2)} > 0 \end{aligned} \quad (58)$$

with $(\mathbf{w}_0, \Omega, \gamma)$ in (53) and $\omega_d = \mathbf{w}_0^2 \Omega / (\Omega\gamma + \mathbf{w}_0^2)$. In fact, we can also show, by using equation (28) with (54), that $F_{d,2}(0) > F_d(0)$.

Next, we briefly consider the case of $(d, 4)$. We then have $(\tilde{\gamma}_+)'_{d,4}(\omega) = \tilde{\gamma}_d(\omega) - \gamma_o + (\gamma_o \omega^2 - 2i\gamma_o \delta(0)\omega) / \omega_d^2$ from equation (52). After a fairly lengthy calculation with this, we can eventually obtain an explicit expression for $K_{d,4} = \frac{\hbar}{2\pi} \int_0^\infty d\omega f_{d,4}(\omega)$, where $f_{d,4}(\omega) \rightarrow -2\{\delta(0) + \omega_d^2/\gamma_o\}/\omega$ for large ω . From this asymptotic form, we easily see that $K_{d,4} \rightarrow -\infty$, which indicates the violation of the second law. This infinity of $K_{d,4}$ suggests, however, that this case would be strictly unrealistic.

From the above results for cases (d, n) including the case $(p = 2)$ in Section 4.3, we may be able to say that aside for physically unacceptable damping models, the divergence (weak or strict) of the spectral density $J(\omega)$ for large frequencies could lead to the violation of the second law. It is also interesting here to note that $\operatorname{Re}(\tilde{\gamma}_+)'_{d,2}(\omega) = \gamma_o \omega^2 / (\omega^2 + \omega_d^2) > 0$ and $\operatorname{Re}(\tilde{\gamma}_+)'_{d,4}(\omega) = \gamma_o \omega_d^{-2} \omega^4 / (\omega^2 + \omega_d^2) > 0$. It is known [23] that a violation of the positivity for $\operatorname{Re} \tilde{\gamma}_+(\omega)$ is tantamount to a violation of the second law in the thermodynamic limit (where a coupling strength between system and bath vanishes). We see here, however, that this positivity would not be a sufficient condition for the second law in the quantum regime (with a non-negligible finite coupling strength between system and bath).

5 Conclusions

In summary, we have extensively studied the second law in the scheme of quantum Brownian motion. It has been observed that from the system-bath entanglement, a system oscillator coupled to a bath at zero temperature has a higher average energy value than the ground state of an uncoupled harmonic oscillator. For a damping model with arbitrarily discrete bath modes and damping models with continuous bath modes with cut-off frequencies, however, this apparent excess energy has actually been found to be less than the minimum work to couple a system to a bath. Therefore, the second law holds in the quantum regime. We also found, on the other hand, that the violation of the second law may happen for some cut-off frequency-free damping models, which are, however, physically unrealistic; especially the case $(d, 2)$ in Section 4.4, with a less diverging spectral density of bath modes than the Ohmic model being the prototype for damping, has a finitely negative value of $K_{d,2}$. The further question about the validity of the quantum second law for a broader class of quantum systems than the quantum Brownian motion considered here, particularly non-linear systems coupled to a bath, clearly remains open.

$$\int_0^\infty d\omega \operatorname{Im}\chi_d(\omega) = \begin{cases} \frac{1}{M} \frac{(\mathbf{w}_0^2 + \Omega^2 - \gamma^2/2) \arccos(\gamma/2\mathbf{w}_0) - \gamma \mathbf{w}_1 \ln(\Omega/\mathbf{w}_0)}{\mathbf{w}_1 (\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} & \text{for } \mathbf{w}_0 > \gamma/2 \\ \frac{1}{M} \frac{(\gamma^2/2 - \mathbf{w}_0^2 - \Omega^2)/2 \cdot \ln\left(\frac{\gamma/2 - \bar{\mathbf{w}}_1}{\gamma/2 + \bar{\mathbf{w}}_1}\right) - \gamma \bar{\mathbf{w}}_1 \ln(\Omega/\mathbf{w}_0)}{\bar{\mathbf{w}}_1 (\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} & \text{for } \mathbf{w}_0 \leq \gamma/2 \end{cases} \quad (63)$$

$$\int_0^\infty d\omega \omega^2 \operatorname{Im}\chi_d(\omega) = \begin{cases} \frac{1}{M} \frac{(\mathbf{w}_0^4 + \mathbf{w}_0^2 \Omega^2 - \Omega^2 \gamma^2/2) \arccos(\gamma/2\mathbf{w}_0) + \Omega^2 \gamma \mathbf{w}_1 \ln(\Omega/\mathbf{w}_0)}{\mathbf{w}_1 (\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} & \text{for } \mathbf{w}_0 > \gamma/2 \\ \frac{1}{M} \frac{(\Omega^2 \gamma^2 - 2\mathbf{w}_0^4 - 2\mathbf{w}_0^2 \Omega^2)/4 \cdot \ln\left(\frac{\gamma/2 - \bar{\mathbf{w}}_1}{\gamma/2 + \bar{\mathbf{w}}_1}\right) + \Omega^2 \gamma \bar{\mathbf{w}}_1 \ln(\Omega/\mathbf{w}_0)}{\bar{\mathbf{w}}_1 (\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} & \text{for } \mathbf{w}_0 \leq \gamma/2 \end{cases} \quad (64)$$

One of us (I.K.) is grateful to G.J. Iafrate for some interesting remarks.

Appendix A: detailed derivation of equation (31)

By substituting equation (16) with $\tilde{\chi}(\omega) = -1/MG(\omega)$ and equation (11) into K in (30), we immediately have

$$K = \frac{-i\hbar}{4\pi M} \oint d\omega \omega^2 \frac{\prod_{j=1}^N (\omega^2 - \omega_j^2)}{\prod_{k=0}^N (\omega^2 - \bar{\omega}_k^2)} \times \sum_{l=1}^N \frac{c_l^2}{2m_l \omega_l^2} \left\{ \frac{1}{(\omega_l + \omega)^2} + \frac{1}{(\omega_l - \omega)^2} \right\}, \quad (59)$$

where the integration path over ω is a loop around the *positive real axis* in the complex ω -plane, consisting of the two branches, $(\infty + i\epsilon, i\epsilon)$ and $(-i\epsilon, \infty - i\epsilon)$. By using the residues at all poles $\omega = \bar{\omega}_k$ of the integrand, we can evaluate the contour integration. In doing so, we do not have any residue at $\omega = \bar{\omega}_{k'}$ when $\omega_l = \bar{\omega}_{k'}$. Accordingly, we can finally obtain

$$K = \frac{\hbar}{4M} \sum_{k=0}^N \mathcal{A}_k; \quad \mathcal{A}_k = \mathcal{A}_k^{(1)} \cdot \mathcal{A}_k^{(2)}, \quad (60)$$

where

$$\mathcal{A}_k^{(1)} = \bar{\omega}_k \frac{\prod_{j=1}^N (\bar{\omega}_k^2 - \omega_j^2)}{\prod_{\substack{k'=0 \\ (\neq k)}}^N (\bar{\omega}_k^2 - \bar{\omega}_{k'}^2)}; \quad (61)$$

$$\mathcal{A}_k^{(2)} = \sum_{l=1}^N \frac{c_l^2}{2m_l \omega_l^2} P \left\{ \frac{1}{(\omega_l + \bar{\omega}_k)^2} + \frac{1}{(\omega_l - \bar{\omega}_k)^2} \right\}.$$

Here, $\mathcal{A}_k^{(2)}$ is, obviously, positive-valued. Therefore, the non-negativeness of \mathcal{A}_k can be completely determined by the factor $\mathcal{A}_k^{(1)}$. Keeping in mind the frequency relationship in (22), we first consider $\mathcal{A}_N^{(1)}$. This is clearly non-negative. Next, for $k = N - 1$, we have

$$\mathcal{A}_{N-1}^{(1)} = \bar{\omega}_{N-1} \frac{\prod_{j=1}^{N-1} (\bar{\omega}_{N-1}^2 - \omega_j^2)}{\prod_{k=0}^{N-2} (\bar{\omega}_{N-1}^2 - \bar{\omega}_k^2)} \times \frac{\bar{\omega}_{N-1}^2 - \omega_N^2}{\bar{\omega}_{N-1}^2 - \bar{\omega}_N^2}. \quad (62)$$

The first factor on the right hand side is non-negative, and so is the second factor whose numerator and denominator are negative-valued, respectively. Therefore, we get $\mathcal{A}_{N-1} \geq 0$. Along the same line, we can straightforwardly show that each summand \mathcal{A}_k with $k = N - 2, N - 3, \dots, 0$ is non-negative, which will yield $K \geq 0$.

Appendix B: No violation of the second law in the Drude model

In the Drude model, we can even evaluate the system energy $(E_s)_d(0)$ and the free energy $F_d(0)$ explicitly and in closed form. From equation (39) we see that for $\mathbf{w}_0 > \gamma/2$ (*underdamped* case), z_1 and z_2 are conjugate complex numbers to each other, while for $\mathbf{w}_0 \leq \gamma/2$ (*overdamped* case), both z_1 and z_2 are real-valued. Therefore, $\operatorname{Im}\{(R'_+)_d(\omega)/(G_+)_d(\omega)\}$ in (35), and thus the explicit expressions of K_d , for both cases would differ from each other in parameters $(\mathbf{w}_0, \Omega, \gamma)$. By using equation (39) we can obtain

see equations 63 and 64 above

where $\mathbf{w}_1 = \sqrt{\mathbf{w}_0^2 - (\gamma/2)^2}$ and $\bar{\mathbf{w}}_1 = -i\mathbf{w}_1$.

From equations (24), (63), and (64), we have an exact expression

$$(E_s)_d(0) = \frac{\hbar}{2\pi} \{A(\mathbf{w}_0, \Omega, \gamma) + B(\mathbf{w}_0, \Omega, \gamma)\}, \quad (65)$$

$$A(\mathbf{w}_0, \Omega, \gamma) = \begin{cases} \frac{(\mathbf{w}_0^2 + \Omega^2)(2\Omega\mathbf{w}_1^2 + \mathbf{w}_0^2\gamma) - \Omega^2\gamma^3/2}{\mathbf{w}_1(\Omega + \gamma)(\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} \arccos(\gamma/2\mathbf{w}_0) \\ \frac{(\mathbf{w}_0^2 + \Omega^2)(\Omega\gamma^2/4 - \mathbf{w}_0^3 - \mathbf{w}_0^2\gamma/2) - \Omega\gamma^2/2 \cdot (\mathbf{w}_0^2 - \Omega\gamma/2)}{\bar{\mathbf{w}}_1(\Omega + \gamma)(\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} \ln\left(\frac{\gamma/2 - \bar{\mathbf{w}}_1}{\gamma/2 + \bar{\mathbf{w}}_1}\right) \end{cases} \quad (66)$$

where

see equation 66 above

for $\mathbf{w}_0 > \gamma/2$ and $\mathbf{w}_0 \leq \gamma/2$, respectively, and

$$B(\mathbf{w}_0, \Omega, \gamma) = \frac{\Omega\gamma(\Omega^2 + \Omega\gamma - \mathbf{w}_0^2)}{(\Omega + \gamma)(\mathbf{w}_0^2 - \Omega\gamma + \Omega^2)} \ln(\Omega/\mathbf{w}_0). \quad (67)$$

Similarly, the free energy $F_d(0)$ in equation (28) can be exactly evaluated as

$$F_d(0) = \begin{cases} \frac{\hbar}{2\pi} \left\{ (\Omega + \gamma) \ln\left(\frac{\Omega + \gamma}{\Omega}\right) + \gamma \ln\left(\frac{\Omega}{\mathbf{w}_0}\right) + 2\mathbf{w}_1 \arccos\left(\frac{\gamma}{2\mathbf{w}_0}\right) \right\} & \text{for } \mathbf{w}_0 > \gamma/2 \\ \frac{\hbar}{2\pi} \left\{ (\Omega + \gamma) \ln\left(\frac{\Omega + \gamma}{\Omega}\right) + \gamma \ln\left(\frac{\Omega}{\mathbf{w}_0}\right) + \bar{\mathbf{w}}_1 \ln\left(\frac{\gamma/2 - \bar{\mathbf{w}}_1}{\gamma/2 + \bar{\mathbf{w}}_1}\right) \right\} & \text{for } \mathbf{w}_0 \leq \gamma/2. \end{cases} \quad (68)$$

Clearly, $(E_s)_d(0)$ and $F_d(0)$ for the underdamped case are identical to equations (10) and (14) in [11], respectively. These expressions can also be applied for the overdamped case ($\mathbf{w}_1 \notin \mathbb{R}$) with the aid of the complex-valued expression, $\arccos(y) = \frac{\pi}{2} + i \ln(iy + \sqrt{1 - y^2})$ and actually equivalent to those for the overdamped case derived here. Then, we can obtain $E_s(0) < F(0)$ for both cases (see Fig. 2 in [11]).

In the limit $\Omega \rightarrow \infty$ (equivalently, $\omega_d \rightarrow \infty$), we get from equations (65–68)

$$K_d \approx \frac{\hbar\Omega}{2\pi} \ln\left(\frac{\Omega + \gamma}{\Omega}\right) \approx \frac{\hbar\gamma}{2\pi} \left(1 - \frac{\gamma}{2\Omega}\right) + \mathcal{O}\left(\frac{1}{\Omega^2}\right) \quad (69)$$

which reduces to

$$\begin{aligned} \frac{\gamma}{\pi\mathbf{w}_0} E_g &= \frac{\hbar\gamma}{2\pi} \frac{1}{\sqrt{1 + \gamma/\Omega}} \\ &\approx \frac{\hbar\gamma}{2\pi} \left(1 - \frac{\gamma}{2\Omega}\right) + \mathcal{O}\left(\frac{1}{\Omega^2}\right) \end{aligned} \quad (70)$$

(cf. Eq. (15) in [11]). Lastly, we explicitly give an explicit expression for K_d in original parameters ($\omega_0, \omega_d, \gamma_o$),

$$\begin{aligned} K_d &= \frac{\hbar\gamma_o}{2\pi} \\ &\times \int_0^\infty d\lambda \frac{2\lambda_d^2\lambda^5 - (2\lambda_0^2 + \lambda_d)\lambda_d^2\lambda^3}{\{(\lambda^2 + \lambda_d^2)(\lambda^2 - \lambda_0^2) - \lambda_d\lambda^2\}^2 + (\lambda_d^2\lambda)^2}, \end{aligned} \quad (71)$$

where a dimensionless parameter $\lambda = \omega/\gamma_o$ with $\lambda_0 = \omega_0/\gamma_o$ and $\lambda_d = \omega_d/\gamma_o$. Here, we see that the non-zero value of K_d for $\lambda_d \rightarrow \infty$ arises from the competition between two terms of the numerator of the integrand for large λ . Equation (71) is also used in Table 2 for comparison with $K_{d,1}$ in equation (49).

Appendix C: Details for equations (41) and (47)

In derivation of equation (41), we used the relationship [24] $E_1(-y \pm i0^+) = -\text{Ei}(y) \mp i\pi$, where the exponential integrals $E_1(y) = \int_1^\infty dz e^{-yz}/z$ and $\text{Ei}(y) = P \int_{-\infty}^y dz e^z/z$. Also, for equation (47), we employed, first [25],

$$\int_0^\infty dy \frac{\cos(ay)}{y+b} = -\sin(ab)\text{si}(ab) - \cos(ab)\text{Ci}(ab), \quad (72)$$

where the sine integral $\text{si}(y) = -\int_y^\infty dz \frac{\sin(z)}{z} = -\frac{\pi}{2} + \text{Si}(y)$ with $\text{Si}(y) = \int_0^y dz \frac{\sin(z)}{z}$, and the cosine integral $\text{Ci}(y) = -\int_y^\infty dz \frac{\cos(z)}{z} = c_e + \ln y + \int_0^y dz \frac{\cos(z)-1}{z}$ with the Euler constant $c_e = 0.5772156649\dots$; secondly, we used the relations $\text{Si}(iy) = \frac{i}{2}\{\text{Ei}(y) + E_1(y)\}$ and $\text{Ci}(iy) = \frac{1}{2}\{\text{Ei}(y) - E_1(y)\} + \frac{\pi}{2}i$ [24].

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