Eur. Phys. J. B (2014)
DOI: 10.1140/epjb/e2014-50125-2

THE EUROPEAN
PHYSICAL JOURNAL B

Regular Article

Thermodynamic anomalies in the presence of general linear dissipation: from the free particle to the harmonic oscillator

Robert Adamietz[1], Gert-Ludwig Ingold[1,a], and Ulrich Weiss[2]

[1] Institut für Physik, Universität Augsburg, 86135 Augsburg, Germany
[2] Institut für Theoretische Physik, Universität Stuttgart, 70550 Stuttgart, Germany

Received 25 February 2014 / Received in final form 10 March 2014

Abstract. A free particle coupled to a heat bath can exhibit a number of thermodynamic anomalies like a negative specific heat, reentrant classicality or a nonmonotonic entropy. These low-temperature phenomena are expected to be modified at very low temperatures where finite-size effects associated with the discreteness of the energy spectrum become relevant. In this paper, we explore in which form the thermodynamic anomalies visible in the specific heat and the entropy of the free damped particle appear for a damped harmonic oscillator. Since the discreteness of the oscillator's energy spectrum is fully accounted for, the results are valid for arbitrary temperatures. As expected, they are in agreement with the third law of thermodynamics and indicate how the thermodynamic anomalies of the free damped particle can be reconciled with the third law. Particular attention is paid to the transition from the harmonic oscillator to the free particle when the limit of the oscillator frequency to zero is taken.

1 Introduction

The thermodynamic properties of a free particle coupled to a heat bath can exhibit several interesting effects. While the thermodynamic properties of an isolated free particle remain classical for arbitrary temperatures, the coupling to an Ohmic environment can provide a mechanism to bring the specific heat of the free damped particle down to zero in the zero-temperature limit. Thus, the validity of the third law of thermodynamics in this specific case is ensured by the particle's environment [1]. For sufficiently strong coupling, the specific heat obtained from the reduced partition function can even become negative [2]. This phenomenon may be understood in terms of a modification of the density of states of the heat bath caused by level repulsion due to coupling to the free particle [3]. Negative specific heats have also been discussed in the context of Kondo superconductors [4], quantum impurity systems [5], XY spin chains [6], two-level fluctuators [7], and energy transport in proteins [8].

Recently, it was found that the vanishing specific heat in the zero-temperature limit is specific to Ohmic heat baths [9]. Subohmic baths, i.e. baths with an increased density of low-frequency degrees of freedom with respect to an Ohmic bath, can even give rise to negative specific heats in the zero-temperature limit. In such situations, finite-size effects must be taken into account in order to save the third law of thermodynamics. Finite-size effects in the context of the present paper are effects arising when the particle is constrained to a large but finite spatial region. They should not be confused with effects arising from a finite number of particles contained in the system. In our case, the latter always consists of just one particle.

Another interesting phenomenon arises for sufficiently superohmic environments where the bath spectral density is significantly suppressed at low frequencies. With decreasing temperature, the specific heat decreases under the influence of the environmental coupling. However, at even lower temperatures, the specific heat rises again up to its classical value [9]. The low density of low-frequency bath modes renders the bath inefficient in decreasing the specific heat. Here, again finite-size effects need to be incorporated in order to obtain a correct description of the thermodynamic properties at extremely low temperatures.

With the notable exception of an Ohmic environment, dissipation is not sufficient to guarantee the validity of the third law of thermodynamics for the free particle. In general, finite-size effects due to placing the particle into a box need to be considered. They are expected to become relevant when the temperature drops below a value related to the ground state energy of the particle confined to the box. However, this temperature scale can be made arbitrarily small by making the box sufficiently large. While the finite-size effects then play their role in ensuring the validity of the third law, they do not necessarily impede the observation of the thermodynamic anomalies discussed above.

Evaluating the thermodynamic properties of a damped particle in a box is a complicated task which, in general,

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[a] e-mail: `Gert.Ingold@Physik.Uni-Augsburg.DE`

needs to be done numerically. Perturbative approaches are not sufficient for our purpose because the anomalies of interest here occur at relatively strong damping. Numerically, a damped particle on a finite chain has been treated, but there the focus was on the dissipative phase transition [10].

An analytically more tractable system is the damped harmonic oscillator for which the discreteness of the system's energy spectrum is inherent. Some results for thermodynamic low-temperature properties in the presence of non-Ohmic damping have been discussed in reference [11]. The related problem of a charged particle in a magnetic field and a harmonic potential has been widely studied [12–18]. Also, systems containing more than one harmonic degree of freedom have been discussed recently [19].

In the following, we will focus on the properties of the damped harmonic oscillator for small oscillator frequency. This will allow us to make connection to the one-dimensional free particle and in particular to decide, whether the thermodynamic anomalies found for the free particle are accessible to observation in a harmonic potential. We will start in Sections 2 and 3 by reviewing some basic relations for damped quantum systems. Specifically, in Section 2 we introduce the reduced partition function on which our evaluation of the thermodynamic quantities will be based. The explicit expressions for the reduced partition functions will give us a first idea of the transition from the damped harmonic oscillator to the damped free particle. In Section 3 we will introduce the Laplace transform of the damping kernel which within the scope of this paper is the central quantity describing the heat bath.

In Section 4 we will take the point of view of the bath [3] and study how the bath density of states is modified when a harmonic oscillator is coupled to it. The results will provide us with important information about the relation between the damped harmonic oscillator and the damped free particle. In Sections 5 and 6, we will discuss the specific heat and entropy, respectively, and we will make the connection between features found in the specific heat of a free damped particle and those found for a damped harmonic oscillator. Finally, in Section 7, we will present our conclusions.

2 Reduced partition function

As was first pointed out in reference [1], thermodynamic quantities in the quantum regime beyond the weak-coupling limit between system (S) and heat bath (B) are not uniquely defined. Here, we choose to base the derivation of thermodynamic quantities on the reduced partition function of the system

$$\mathcal{Z} = \frac{\mathcal{Z}_{\mathrm{S+B}}}{\mathcal{Z}_{\mathrm{B}}} \tag{1}$$

obtained from the partition function $\mathcal{Z}_{\mathrm{S+B}}$ of system and heat bath and the partition function $\mathcal{Z}_{\mathrm{B}}$ of the heat bath alone. In the absence of any coupling between system and bath, we have $\mathcal{Z}_{\mathrm{S+B}} = \mathcal{Z}_{\mathrm{S}}\mathcal{Z}_{\mathrm{B}}$ and the reduced partition function (1) equals the partition function of the system $\mathcal{Z}_{\mathrm{S}}$. In general, the reduced partition function will differ from the partition function of the uncoupled system, thereby describing the influence of the heat bath on the thermodynamic properties of the system.

Applying the usual thermodynamic relations, our choice implies that the thermodynamic quantities of a damped quantum system like the specific heat or the entropy are actually given by the change of this quantity when the system is coupled to the environment.

For example, the specific heat of the damped system is expressed as the difference between the specific heat of system and bath and the specific heat of the bath alone

$$C = C_{\mathrm{S+B}} - C_{\mathrm{B}}. \tag{2}$$

The specific heat, as well as other thermodynamic quantities defined in this way, can thus be regarded as a property of the damped system and, alternatively, as a change in the properties of the heat bath. The latter point of view will be taken in Section 4.

For the free damped particle, the reduced partition function is given by [20]

$$\mathcal{Z} = \sqrt{\frac{\pi}{4\beta E_{\mathrm{g}}}} \prod_{n=1}^{\infty} \frac{\nu_n}{\nu_n + \hat{\gamma}\left(\nu_n\right)} \tag{3}$$

where $\beta = 1/k_{\mathrm{B}}T$ is proportional to the inverse temperature T and the Matsubara frequencies are given by $\nu_n = 2\pi n/\hbar\beta$. The Laplace transform $\hat{\gamma}(z)$ of the damping kernel will be introduced in detail below in Section 3. In the prefactor, $E_{\mathrm{g}} = \hbar^2\pi^2/2ML^2$ is the ground state energy of a particle of mass M in a one-dimensional box of width L. The finite box is merely needed for regularization. For the purpose of this paper, the width L is assumed to be so large that the discrete level structure becomes only relevant at temperatures much lower than those of interest here. All results pertaining to the free damped particle should be expected to be modified when the temperature is decreased to values of the order of E_{g}.

For a harmonic oscillator of frequency ω_0, the reduced partition function reads [20]

$$\mathcal{Z} = \frac{1}{\hbar\beta\omega_0} \prod_{n=1}^{\infty} \frac{\nu_n^2}{\nu_n^2 + \nu_n\hat{\gamma}\left(\nu_n\right) + \omega_0^2}. \tag{4}$$

The factors in front of the infinite product in equations (3) and (4) determine the behavior in the classical regime. The different powers of temperature, 1/2 for the free particle and 1 for the harmonic oscillator, are a consequence of the different number of degrees of freedom. For example, they lead to a high-temperature specific heat of $k_{\mathrm{B}}/2$ for the free particle and of k_{B} for the harmonic oscillator.

Thus, the limit $\omega_0 \to 0$ of the harmonic oscillator cannot lead to the free particle in a smooth way [1]. If we disregard the difference arising from the classical factor for the moment, there remains a difference due to the infinite products in equations (3) and (4). However, the infinite products can only differ significantly at low temperatures $k_\mathrm{B}T \ll \hbar\omega_0/2\pi$ which become arbitrarily small in the limit $\omega_0 \to 0$.

Already on the basis of the reduced partition functions (3) and (4) we can expect differences to occur between the thermodynamic quantities of damped free particle and damped harmonic oscillator which are solely determined by their classical expressions. Still, these differences can be relevant even deep into the quantum regime. In addition, below a certain temperature, the discrete energy spectrum of the harmonic oscillator will start to play a role and, in particular, will ensure the validity of the third law of thermodynamics, as we shall see.

3 Laplace transform of the damping kernel

The heat bath is described within a Caldeira-Leggett model where the system is coupled bilinearly through its position to the positions of a set of harmonic bath oscillators [20]. However, we need not be concerned with the details of this model because the only quantity of relevance for the system properties is the spectral density of the system-bath coupling $J(\omega)$. It is related to the Laplace transform of the damping kernel appearing in equations (3) and (4) by means of

$$\hat{\gamma}(z) = \frac{2}{\pi M}\int_0^\infty d\omega \frac{J(\omega)}{\omega}\frac{z}{\omega^2+z^2}. \tag{5}$$

To be specific, we choose a spectral density of the form

$$J(\omega) = M\gamma\omega^s \frac{\omega_\mathrm{c}^{2p-s+1}}{(\omega_\mathrm{c}^2+\omega^2)^p} \tag{6}$$

with $0 < s < 2p+2$. Here, M is the mass associated with the system degree of freedom and γ determines the damping strength. At low frequencies, the spectral density (6) increases proportional to ω^s where the exponent s is decisive for the low-temperature thermodynamics. In order to avoid ultraviolet divergences, we have chosen a generalized Drude-type cutoff represented by the last factor on the right-hand side of (6). It suppresses the spectral density of bath oscillators above a frequency scale determined by the cutoff frequency ω_c. Other choices for the cutoff are possible but are not expected to affect our results in an important manner.

In reference [9] it was shown for a free damped particle subject to a bath described by (6), that the specific heat approaches $C/k_\mathrm{B} = (s-1)/2$ for $s \leq 2$ as the temperature is lowered towards zero. The particular case $s = 1$ corresponds to Ohmic damping where the specific heat of the free damped particle vanishes at zero temperature. For subohmic damping, $s < 1$, the specific heat even tends to a negative value in the low-temperature limit as long as finite-size effects remain irrelevant. For superohmic damping with $s \geq 2$, the classical value $C/k_\mathrm{B} = 1/2$ is approached in the low-temperature regime.

Employing the relation (5), one finds that for the spectral density (6), the Laplace transform of the damping kernel can be expressed in terms of a hypergeometric function [9]. For our purposes, it will be sufficient to consider integer values for the exponent p appearing in the cutoff function. Then, the Laplace transform of the damping kernel can be expressed in terms of a finite sum as

$$\begin{aligned}\hat{\gamma}(z) = \frac{\gamma\omega_\mathrm{c}^{2p}}{(\omega_\mathrm{c}^2-z^2)^p}\Bigg[&\frac{(z/\omega_\mathrm{c})^{s-1}}{\sin\left(\frac{\pi s}{2}\right)}\\ &+\frac{1}{\pi}\sum_{n=1}^{p}\frac{(-1)^n}{n-\frac{s}{2}}\frac{B\left(\frac{s}{2},p+1-\frac{s}{2}\right)}{B(n,p-n+1)}\left(\frac{z}{\omega_\mathrm{c}}\right)^{2n-1}\Bigg].\end{aligned} \tag{7}$$

The beta function is defined in terms of gamma functions as $B(x,y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$. The expression (7) is not valid for even integer values of s where logarithmic terms occur.

We will mostly be interested in the Ohmic case $s = 1$ and the superohmic case $s = 3$ where analytical results can be obtained and which allow to discuss the main thermodynamic anomalies. For $s = 1$ and sufficiently strong damping, the free damped particle has a negative specific heat at low temperatures [2] while for $s = 3$ reentrant classicality is observed [9].

For odd integer s, the expression (7) can be written as

$$\hat{\gamma}(z) = \frac{P(z;s,p)}{(\omega_\mathrm{c}+z)^p}, \tag{8}$$

where $P(z;s,p)$ is a polynomial in z of order z^{p-1}, in which the term of zeroth order is absent for $s = 3, 5, \ldots$ Particular cases are

$$\begin{aligned}P(z;1,1) &= \gamma\omega_\mathrm{c}\\ P(z;1,2) &= \gamma\omega_\mathrm{c}\left(\omega_\mathrm{c}+\frac{z}{2}\right)\\ P(z;3,2) &= \gamma\omega_\mathrm{c}\frac{z}{2}.\end{aligned} \tag{9}$$

4 Change of bath density of states

As mentioned above, the specific heat (2) can either be viewed as specific heat of the system modified by the coupling to the heat bath or as change in the specific heat of the bath when the system degree of freedom is coupled to it. The latter point of view was taken in reference [3] where it was shown that the specific heat (2) of the damped free particle can be expressed in terms of the change in the bath density of states together with the well-known expression for the specific heat of a harmonic oscillator.

The change in the bath density of states is defined as

$$\xi(\omega) = \sum_n \left[\delta\left(\omega-\omega_n\right) - \delta\left(\omega-\omega_n^0\right)\right], \tag{10}$$

where ω_n^0 are the eigenfrequencies of the bath oscillators in the absence of the system degree of freedom and ω_n are the frequencies of the eigenmodes of system and bath coupled to each other. The density of states $\xi(\omega)$ of interest here should not be confused with the spectral density of the coupling $J(\omega)$ introduced in the context of (5) and (6).

For the harmonic oscillator (osc) and the free particle (fp) we obtain the change of the bath density of states as

$$\xi_{\mathrm{osc}}(\omega) = \frac{1}{\pi}\mathrm{Im}\frac{\partial \ln\left[\hat{\chi}(-\mathrm{i}\,\omega)\right]}{\partial\omega}, \tag{11}$$

$$\xi_{\mathrm{fp}}(\omega) = \frac{1}{\pi}\mathrm{Im}\frac{\partial \ln\left[\hat{\mathcal{R}}(-\mathrm{i}\omega)\right]}{\partial\omega}, \tag{12}$$

respectively. Here,

$$\hat{\chi}(z) = \frac{1}{z^2 + z\hat{\gamma}(z) + \omega_0^2} \tag{13}$$

is the dynamical susceptibility of the damped harmonic oscillator and

$$\hat{\mathcal{R}}(z) = \frac{1}{z + \hat{\gamma}(z)} \tag{14}$$

is the dynamical velocity response of the damped free particle. Further, Im denotes the imaginary part. Observe that $\xi_{\mathrm{osc}}(\omega)$ concurs with $\xi_{\mathrm{fp}}(\omega)$ in the limit $\omega_0 \to 0$ for any non-zero frequency ω.

With the form (8) for $\hat{\gamma}(z)$, the poles of $\hat{\chi}(z)$ are determined by the zeros z_i of the polynomial equation

$$N\left(z;\omega_0\right) \equiv \left(z^2+\omega_0^2\right)\left(z+\omega_{\mathrm{c}}\right)^p + zP(z;s,p) = 0. \tag{15}$$

Each individual pole of $\hat{\chi}(z)$ contributes a Lorentzian to the change of the bath density of states. Denoting the real and imaginary part of z by z' and z'', respectively, we then have

$$\xi(\omega) = \frac{1}{\pi}\left(-p\frac{\omega_{\mathrm{c}}}{\omega^2+\omega_{\mathrm{c}}^2} - \sum_i \frac{z_i'}{\left(\omega+z_i''\right)^2 + z_i'^2}\right). \tag{16}$$

Since equation (15) applies to a damped system, the real part of each individual zero is negative, $z_i' < 0$. Thus, the respective Lorentzians give positive contributions to the density (16).

Consider next equation (15) in the free particle limit $\omega_0 = 0$. With equations (7) and (9) we see that the polynomial $N(z;0)$ has a simple zero at the origin, $z = 0$, for $s = 1$, and a double zero at the origin for $s = 3, 5, \ldots$ These solutions of equation (15) do not contribute to the change of the bath density of states (16). As a result, for any p the density $\xi_{\mathrm{fp}}(\omega)$ has one Lorentzian less compared with $\xi_{\mathrm{osc}}(\omega)$ for $s = 1$ and two Lorentzians less for $s = 3, 5, \ldots$

The absence of one or two low-frequency peaks in the density $\xi_{\mathrm{fp}}(\omega)$ compared to $\xi_{\mathrm{osc}}(\omega)$ will lead to qualitative differences in the thermodynamic behavior of the damped free particle and harmonic oscillator. In this respect, the value of the difference of the integrated densities

$$\begin{aligned}\mathit{\Delta\Sigma}(s) &\equiv \mathit{\Sigma}_{\mathrm{osc}}(s) - \mathit{\Sigma}_{\mathrm{fp}}(s)\\ &= \int_0^\infty d\omega\left[\xi_{\mathrm{osc}}(\omega;s) - \xi_{\mathrm{fp}}(\omega;s)\right]\end{aligned} \tag{17}$$

plays a crucial role, as we shall see.

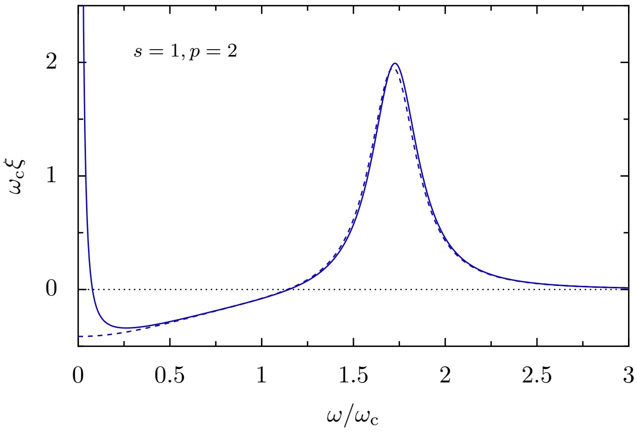

Fig. 1. Change of the density of bath oscillators $\xi_{\mathrm{osc}}(\omega)$ for a spectral density (6) characterized by $s = 1$ and $p = 2$, damping constant $\gamma = 5\omega_{\mathrm{c}}$, and an oscillator frequency $\omega_0 = 0.2\omega_{\mathrm{c}}$ (solid line). For comparison, the dashed line gives the density $\xi_{\mathrm{fp}}(\omega)$ for the free damped particle.

The preceding discussion will now be illustrated by considering in more detail the important cases $s = 1$ and 3. We start with Ohmic damping, $s = 1$, and a cutoff characterized by $p = 1$. For the free particle, the non-vanishing zeros of the equation $N(z;0) = 0$, which contribute to the change of the spectral density $\xi_{\mathrm{fp}}(\omega)$, are

$$z_{1,2} = -\frac{\omega_{\mathrm{c}}}{2}\left(1 \pm \mathrm{i}\sqrt{\frac{4\gamma}{\omega_{\mathrm{c}}} - 1}\right). \tag{18}$$

These zeros supply the density $\xi_{\mathrm{fp}}(\omega)$ with two Lorentzians. Depending on whether the damping strength γ is smaller or larger than $\omega_{\mathrm{c}}/4$, they are centered around zero frequency or finite frequency with opposite signs, respectively.

Proceeding to the harmonic oscillator, we account for a small frequency ω_0. In this case, the expressions (18) receive minor corrections of order ω_0^2. The most important change, however, consists in an additional real zero acquired by $N(z;\omega_0)$. To leading order in ω_0, this zero is independent of p and located at

$$z_3 = -\frac{\omega_0^2}{\gamma}. \tag{19}$$

The Lorentzian associated with the zero (19) is centered at $\omega = 0$ and becomes a delta function in the limit of vanishing oscillator frequency, $\omega_0 \to 0$. Since this contribution is missing in the density $\xi_{\mathrm{fp}}(\omega)$, we have the concise exact relation

$$\mathit{\Delta\Sigma}(s = 1) = \frac{1}{2}. \tag{20}$$

Figure 1 compares the change of the density of bath oscillators for a damped harmonic oscillator of frequency $\omega_0 = 0.2\omega_{\mathrm{c}}$ (solid line) with that for a damped free particle (dashed line) for an Ohmic spectral density, $s = 1$. In contrast to the previous analytical considerations, we now choose a cutoff characterized by $p = 2$ in order to facilitate comparison with the superohmic case to be discussed below. Apart from the Lorentzian with negative weight given by the first term in (16), the structure of $\xi_{\mathrm{fp}}(\omega)$ for the free damped particle is dominated by a peak at finite frequency determined by a solution of $N(z;0)=0$ analogous to (18).

The main difference between free particle and harmonic oscillator concerns the behavior at low frequencies. Here, the real solution (19) of (15) for the damped harmonic oscillator leads to a peak centered around zero frequency and dominating the low-frequency behavior. As we will see later on, it is this peak which is responsible for the main difference in the low-temperature behavior of thermodynamic quantities between the free particle and the harmonic oscillator.

Consider next the case of a superohmic bath with $s=3$ and $p=2$. With respect to the free damped particle, equation (15) with $\omega_0=0$ again yields two nontrivial solutions,

$$z_{1,2}=-\omega_{\mathrm{c}}\pm \mathrm{i}\sqrt{\frac{\gamma\omega_{\mathrm{c}}}{2}}. \tag{21}$$

These account for peaks at finite frequencies $\pm\sqrt{\gamma\omega_{\mathrm{c}}/2}$, very much like in the Ohmic case.

Advancing from the free particle to the harmonic oscillator, the originally trivial double zeros $z=0$ of the equation $N(z;0)=0$ turn into a pair of complex solutions of the pole condition $N(z,\omega_0)=0$ with the leading real and imaginary parts given by

$$z_{3,4}=\pm\mathrm{i}\frac{\omega_0}{(1+\gamma/2\omega_{\mathrm{c}})^{1/2}}-\frac{\gamma}{2\omega_{\mathrm{c}}^2}\frac{\omega_0^2}{(1+\gamma/2\omega_{\mathrm{c}})^2}. \tag{22}$$

In contrast to the single peak centered at the origin in the Ohmic case, there are now two low-frequency peaks positioned symmetrically about the origin with displacement of the order of ω_0. Since these two peaks are missing in the density $\xi_{\mathrm{fp}}(\omega)$, we now have

$$\mathit{\Delta\Sigma}(s=3)=1. \tag{23}$$

The change of the density of bath oscillators $\xi(\omega)$ for the superohmic case with $s=3$, $p=2$, and damping strength $\gamma=5\omega_{\mathrm{c}}$ is displayed in Figure 2. As in Figure 1, the solid line corresponds to a damped harmonic oscillator with $\omega_0=0.2\omega_{\mathrm{c}}$ while the dashed line refers to a free damped particle. Comparison of Figures 1 and 2 clearly shows the difference in the low-frequency behavior of $\xi_{\mathrm{osc}}(\omega)$ for the damped harmonic oscillator coupled to an Ohmic and a superohmic heat bath.

A more detailed analysis of the low-frequency peak for arbitrary exponents s of the spectral density of the coupling is given in Appendix. In particular, it is shown there that the results (20) and (23) can be generalized to arbitrary s as

$$\mathit{\Delta\Sigma}(s)=\begin{cases}\dfrac{s}{2} & \text{for } 0<s<2\\ 1 & \text{for } s\geq 2.\end{cases} \tag{24}$$

We will make use of this result in the discussion of the specific heat in the next section.

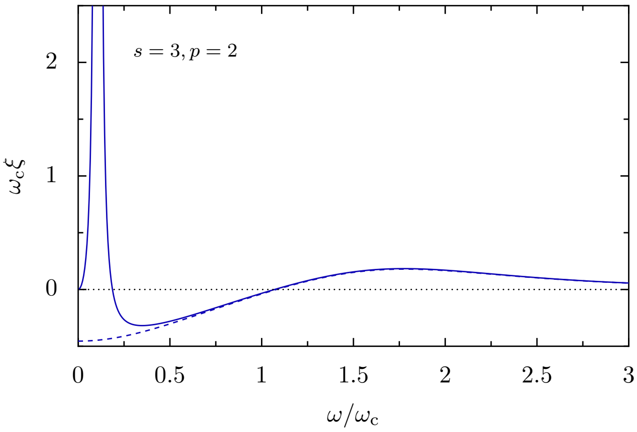

Fig. 2. Change of the density of bath oscillators $\xi_{\mathrm{osc}}(\omega)$ for a spectral density (6) characterized by $s=3$ and $p=2$, damping constant $\gamma=5\omega_{\mathrm{c}}$, and an oscillator frequency $\omega_0=0.2\omega_{\mathrm{c}}$ (solid line). For comparison, the dashed line gives the density $\xi_{\mathrm{fp}}(\omega)$ for the free damped particle.

5 Specific heat

Since the densities of state appearing in (10) can be thought of being constituted by harmonic oscillators, the specific heat defined according to (2) can be obtained from the specific heat C_{ho} of a single harmonic oscillator with frequency ω

$$C_{\mathrm{ho}}=k_{\mathrm{B}}\left(\frac{\hbar\beta\omega}{2\sinh(\hbar\beta\omega/2)}\right)^2. \tag{25}$$

This specific heat vanishes in the zero-temperature limit and approaches k_{B} in the classical limit. Therefore, we can generally write

$$C=C_0+\int_0^\infty d\omega\,\xi(\omega)C_{\mathrm{ho}}(\omega) \tag{26}$$

$$=C_\infty+\int_0^\infty d\omega\,\xi(\omega)\left[C_{\mathrm{ho}}(\omega)-k_{\mathrm{B}}\right], \tag{27}$$

where $\xi(\omega)$ can either be the change of the density of states (11) or (12) for the damped harmonic oscillator or the free damped particle, respectively. C_0 is the specific heat of the damped system in the zero-temperature limit while C_∞ is its classical value. The two values are related by

$$C_\infty=C_0+k_{\mathrm{B}}\mathit{\Sigma}, \tag{28}$$

where $\mathit{\Sigma}$ denotes the integrated change of the density of states in agreement with the notation introduced in (17).

For the damped harmonic oscillator, we obtain from the prefactor in the partition function (4) the classical value of the specific heat $C_\infty=k_{\mathrm{B}}$. Observing that the defining expression (11) is a partial derivative and that the real part of $\hat{\gamma}(-\mathrm{i}\omega)$ is positive, we find for all s the sum rule

$$\mathit{\Sigma}_{\mathrm{osc}}=1, \tag{29}$$

and thus $C_0 = 0$. It is therefore always guaranteed that the specific heat for the damped harmonic oscillator vanishes at zero temperature. The specific heat as a function of temperature can now be expressed as

$$C = \int_0^\infty d\omega\, \xi_{\mathrm{osc}}(\omega) C_{\mathrm{ho}}(\omega). \tag{30}$$

This result can also be derived by means of $C = \partial U/\partial T$ from the expression for the internal energy obtained by Ford et al. for a damped harmonic oscillator [21,22].

For the damped free particle, the situation is a bit more complex. Again the classical value of the specific heat can be found from the prefactor of the corresponding partition function (3) and takes the value $C_\infty = k_{\mathrm{B}}/2$ as expected. For the sum rule $\Sigma_{\mathrm{fp}}(s)$ we have to distinguish the cases $0 < s < 2$ and $s \geq 2$. In the first case, we find from the expression (12) that the low-frequency behavior of the change of the bath density of states is dominated by the first term in (A.1), yielding

$$\Sigma_{\mathrm{fp}}(s) = 1 - \frac{s}{2} \qquad \text{for } 0 < s < 2. \tag{31}$$

In the low-temperature limit, making use of (28) we thus obtain [9]

$$C_0 = \frac{s-1}{2} \qquad \text{for } 0 < s < 2, \tag{32}$$

which can only be valid as long as the size of the spatial region to which the damped particle is confined is irrelevant. We conclude from (32), that in the absence of finite-size effects the specific heat approaches zero only for the special case of Ohmic damping. In the second regime, $s \geq 2$, one obtains

$$\Sigma_{\mathrm{fp}}(s) = 0 \qquad \text{for } s \geq 2. \tag{33}$$

Thus, for $s \geq 2$ the specific heat tends to its classical value at very low temperatures. This phenomenon has been termed "reentrant classicality" [9]. Clearly, the expressions (29), (31), and (33) are in correspondence with the former result (24).

In order to express the specific heat of the damped free particle for arbitrary values of s, (27) is most convenient and we get [9]

$$C = \frac{k_{\mathrm{B}}}{2} + \int_0^\infty d\omega \xi_{\mathrm{fp}}(\omega) \left[C_{\mathrm{ho}}(\omega) - k_{\mathrm{B}}\right]. \tag{34}$$

Together with the results presented in Section 4, we can now discuss how the thermodynamic anomalies of the free damped particle manifest themselves for the damped harmonic oscillator. We will mainly focus on the Ohmic environment ($s = 1$) and a superohmic environment with $s = 3$ for which the changes of the bath density of states have been shown in Figures 1 and 2, respectively. Figure 3 displays the corresponding specific heats for a damped harmonic oscillator with frequency $\omega_0 = 0.2\omega_{\mathrm{c}}$ (full line) and a free damped particle (dashed line). The middle and upper set of curves correspond to $s = 1$ and $s = 3$, respectively, and $p = 2$. The damping constant $\gamma = 5\omega_{\mathrm{c}}$ was chosen sufficiently large that the specific heat for the free particle becomes negative at low temperatures [9]. The lower set of curves corresponds to $s = 0.5$ and $p = 2$ and will be discussed at the end of this section. Note that for better comparison between the specific heat of the harmonic oscillator and the free particle, we have shifted the specific heat of the latter by $k_{\mathrm{B}}/2$, so that all curves reach a value of k_{B} in the high-temperature limit.

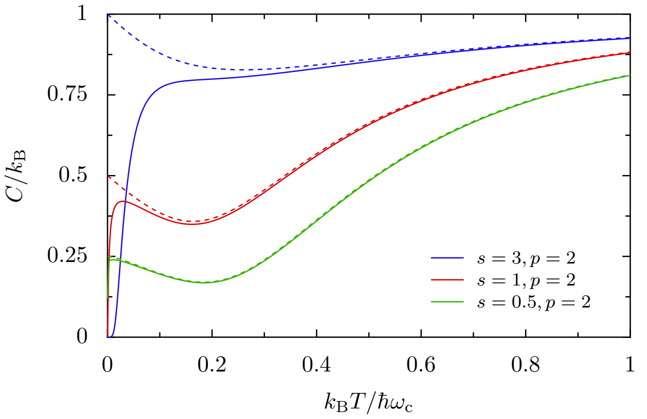

Fig. 3. Specific heat of a damped harmonic oscillator with $\omega_0 = 0.2\omega_{\mathrm{c}}$ (solid line) and a free damped particle (dashed line). For ease of comparison, $k_{\mathrm{B}}/2$ has been added to the specific heat of the free damped particle, i.e., the respective curves have been shifted upwards. The three sets correspond to the bath parameters $s = 3, 1$, and 0.5 and $p = 2$ from the upper to the lower set. The choice of $\gamma = 5\omega_{\mathrm{c}}$ implies that the specific heat of the free damped particle in the Ohmic case, $s = 1$, becomes negative at low temperatures.

Comparing the curves for the harmonic oscillator with the shifted ones for the free particle, we find that they agree rather well as long as the temperature does not become too low. This behavior is consistent with what we had found for the change of the bath density of states in Figures 1 and 2. The important difference there was the presence of a low-frequency peak in the case of the harmonic oscillator. Its weight accounts for the difference of $k_{\mathrm{B}}/2$ in the classical values for the specific heat for harmonic oscillator and free particle. The peak is also responsible for the dramatic increase of the specific heat at low temperatures visible in Figure 3.

We now take a closer look at how the thermodynamic anomalies for the free damped particle appear in the specific heat of the damped harmonic oscillator. We start with the middle set of curves displayed in Figure 3 corresponding to Ohmic damping. Since the dashed curve has been shifted upwards by $k_{\mathrm{B}}/2$, both curves actually start at vanishing specific heat. The dashed curve thus exhibits the phenomenon of negative specific heat for the free damped particle. In the case of the damped harmonic oscillator, the Lorentzian centered at zero frequency shown in Figure 1 increases the specific heat by almost $k_{\mathrm{B}}/2$ at rather low temperatures. Then, the negative part of the change of the bath density of states shown in Figure 1 leads to a decrease

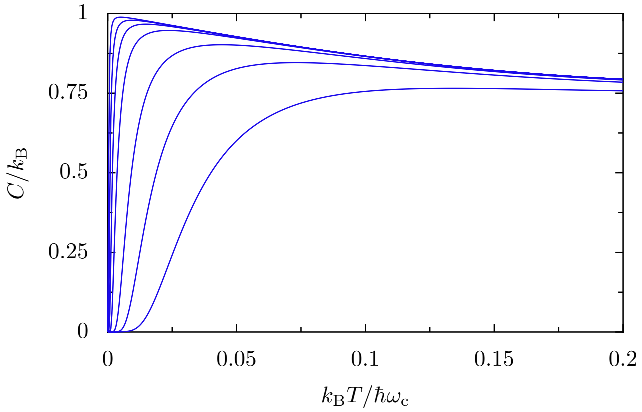

Fig. 4. Building-up of reentrant behavior for a damped harmonic oscillator with frequency $\omega_0/\omega_c = 0.2$, 0.1, 0.05, 0.02, 0.01, 0.005, and 0.002 decreasing from the lower to the upper curve. The damping strength is $\gamma = 5\omega_c$ and the bath density of states is characterized by $s = 3$ and $p = 2$.

of the specific heat with increasing temperature resulting in a pronounced dip. At higher temperatures, the classical value for the specific heat of k_B is approached from below. We can thus conclude that while for the damped harmonic oscillator, the specific heat cannot become negative, a dip may appear which has the same physical origin as the negative specific heat for the free damped particle. Such a dip in the specific heat can even be found if the heat bath is constituted by a single harmonic oscillator. Its origin was traced back to a frequency shift due to the coupling between system and bath [23]. The approach in Section 4 provides a generalization from a one-oscillator bath to a bath consisting of many harmonic oscillators.

Let us now turn to the upper set of curves in Figure 3 corresponding to a superohmic environment with $s = 3$. The dashed curve again displays the specific heat of the free damped particle shifted upwards by $k_B/2$. As we can see, the specific heat exhibits reentrant classicality in the sense that the reduction of the specific heat at intermediate temperatures disappears at low temperatures where the classical high-temperature value is approached. As already discussed, the specific heat for the damped harmonic oscillator coincides with the one of the free particle up to a shift of $k_B/2$ for sufficiently large temperatures. At lower temperatures, deviations necessarily occur because the specific heat of the damped harmonic oscillator has to approach zero in the zero-temperature limit. In order to verify that in the limit of vanishing oscillator frequency $\omega_0 \to 0$ reentrant classicality does occur, we need to consider the specific heat for smaller values of ω_0 as depicted in Figure 4. It can be seen that with decreasing oscillator frequency ω_0, the temperature range in which the specific heat drops from a value close to k_B down to zero becomes arbitrarily small. In this sense, reentrant classicality can be observed even for a damped harmonic oscillator.

We close this section by taking a look at the lower set of curves of Figure 3 corresponding to $s = 0.5$ and $p = 2$. While these curves in the view of the previous discussion provide no surprises, the curve for the damped harmonic oscillator supports the result found earlier for the free damped particle with $s < 1$ that the specific heat tends towards a negative value in the low-temperature limit. Keeping in mind the offset of $k_B/2$, for $s = 0.5$, a value of $C = -0.25k_B$ is indeed approached for small temperatures. Very much like the curve for the damped harmonic oscillator drops down to a vanishing specific heat at zero temperature, one would expect that accounting for finite-size effects would lead to a sharp increase of the specific heat of the damped free particle as zero temperature is approached.

6 Entropy

Another thermodynamic quantity of interest is the entropy. Our analysis will again be based on the reduced partition function $\mathcal{Z}$ defined in (1). By means of the usual thermodynamic relations, the entropy is then obtained as

$$S = \frac{\partial}{\partial T}\left(k_BT\ln(\mathcal{Z})\right). \tag{35}$$

In general, this entropy differs from the von Neumann entropy obtained from the reduced density matrix [24].

A nonmonotonic temperature dependence of the entropy has been discussed e.g. for Kondo systems [4,25], XY spin chains [6], and two-level fluctuators [7]. In the context of the Casimir effect, the appearance of a negative entropy due to a finite zero-frequency electric conductivity has been the subject of an extensive debate [26–30]. A relation between this problem and the damped free particle was pointed out [29] and the temperature dependence of the entropy of a damped free particle bears resemblance with curves obtained for the Casimir entropy for real metals [31].

In comparison with the specific heat, putting free damped particle and damped harmonic oscillator into relation is complicated by the fact that the classical expressions for the entropy differ by a function of temperature. For the damped harmonic oscillator, one obtains from the first factor in (4)

$$S_{\mathrm{osc,cl}} = k_B\left[\ln\left(\frac{k_BT}{\hbar\omega_0}\right)+1\right]. \tag{36}$$

For the free damped particle, one obtains instead

$$S_{\mathrm{fp,cl}} = S_0 + \frac{k_B}{2}\left[\ln\left(\frac{k_BT}{\hbar\gamma}\right)+1\right] \tag{37}$$

where

$$S_0 = \frac{k_B}{2}\ln\left(\frac{\pi}{4}\frac{\hbar\gamma}{E_g}\right). \tag{38}$$

It can be shown that S_0 is the zero-temperature value of the entropy of the free damped particle if finite-size effects are disregarded [29].

For the discussion of the entropy, we focus on an Ohmic heat bath with standard Drude cutoff, i.e. $s = 1$ and $p = 1$.

In this special case, the reduced partition functions (3) and (4) can be expressed as

$$\mathcal{Z} = \mathcal{Z}_{\rm cl} \frac{\prod_n \Gamma\left(1 - \dfrac{\hbar\beta z_n^0}{2\pi}\right)}{\Gamma\left(1 + \dfrac{\hbar\beta\omega_{\rm c}}{2\pi}\right)} \tag{39}$$

where $\Gamma(x)$ is the gamma function. $\mathcal{Z}_{\rm cl}$ is the classical contribution to the reduced partition function, i.e. the factor in front of the infinite product in (3) and (4). The quantities z_n^0 are obtained as zeros of the polynomial

$$N(z;\omega_0) = z^3 + z^2\omega_{\rm c} + z\left(\gamma\omega_{\rm c} + \omega_0^2\right) + \omega_0^2\omega_{\rm c} \tag{40}$$

for the damped harmonic oscillator and

$$\frac{1}{z}N(z;0) = z^2 + z\omega_{\rm c} + \gamma\omega_{\rm c} \tag{41}$$

for the damped free particle.

Using standard relations from thermodynamics, the entropy can then be expressed as

$$S = S_{\rm cl} + k_{\rm B}\left[-f\left(\frac{\hbar\beta\omega_{\rm c}}{2\pi}\right) + \sum_n f\left(-\frac{\hbar\beta z_n^0}{2\pi}\right)\right], \tag{42}$$

where $S_{\rm cl}$ for the harmonic oscillator and free particle is given by (36) and (37), respectively. Furthermore,

$$f(x) = \ln[\Gamma(1+x)] - x\psi(1+x) \tag{43}$$

with the digamma function $\psi(x)$.

Figure 5a displays the entropy for a damped harmonic oscillator with frequency $\omega_0 = 0.2\omega_{\rm c}$ and damping strengths $\gamma/\omega_{\rm c} = 1$, 10, 100, and 1000 increasing from the lower to the upper curve. The straight-line behavior for large temperatures on this semilogarithmic plot reflects the classical behavior (36) of the entropy. For sufficiently strong damping, a crossover to a logarithmic behavior characteristic of the classical free particle is observed as the temperature is lowered. At even smaller temperatures, the oscillator frequency becomes relevant again to ensure that the entropy goes to zero in accordance with the third law of thermodynamics as temperature goes to zero.

Correcting for the difference in the classical behavior between harmonic oscillator and free particle, as explained in the context of the reduced partition function in Section 2, we can define an entropy

$$\tilde{S} = S_{\rm osc} - S_{\rm osc,cl} + S_{\rm fp,cl} \tag{44}$$

which should agree with the entropy of the damped free particle except for not too low temperatures. This correction for the classical behavior corresponds to the shift by $k_{\rm B}/2$ which we had applied to the specific heat of the free damped particle in Figure 3. In Figure 5b we have taken the entropy data displayed in the upper panel to obtain together with (36) and (37) the entropy $\tilde{S}$ defined in (44) represented as solid lines. The damping strength increases here from the upper to the lower curve.

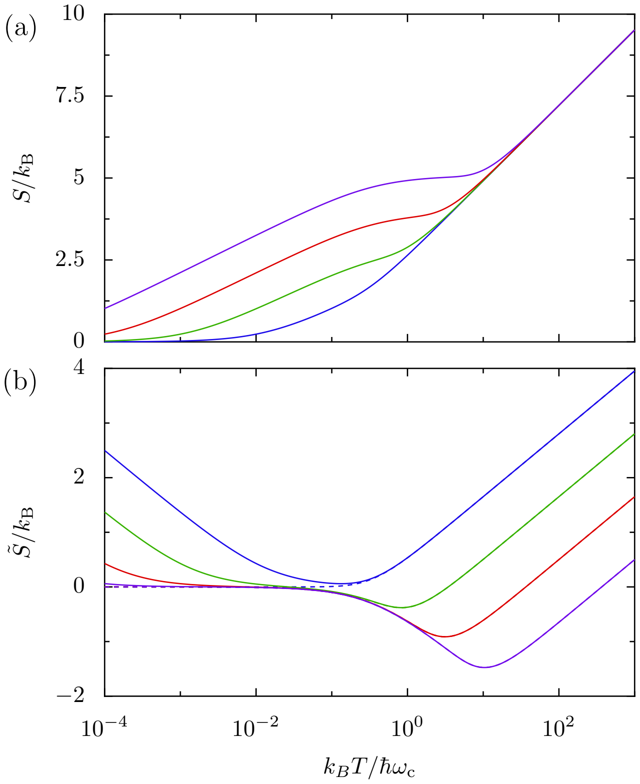

Fig. 5. (a) Entropy of a damped harmonic oscillator with $\omega_0 = 0.2\omega_{\rm c}$ coupled to an Ohmic environment ($s = 1$) with Drude cutoff ($p = 1$) and varying damping strength $\gamma/\omega_{\rm c} = 1$, 10, 100, and 1000 from the lower to the upper curve. (b) The solid lines represent the entropy $\tilde{S}$ defined in (44) and obtained from the data represented in panel (a) by means of (36) and (37). The values of the damping strengths coincide with those chosen for panel (a), but here they increase from the upper to the lower curve. The dashed lines represent the entropy $S - S_0$ of the free damped particle disregarding finite-size effects. Because the entropy constant (38) has been subtracted off all data in this panel, the dashed lines approach zero in the limit of vanishing temperature.

For comparison, the dashed lines in Figure 5b show the results for the entropy of the free damped particle. The larger the damping strength, the longer $\tilde{S}$ agrees with the entropy $S - S_0$ of the free particle as the temperature is lowered. Therefore, at intermediate temperatures, a dashed curve is only visible for $\gamma/\omega_{\rm c} = 1$. Only for even lower temperatures, the finite frequency of the oscillator used in determining $\tilde{S}$ becomes relevant and a dashed curve appears close to the zeroline. Note that in the curves shown in Figure 5b, the constant (38) has been subtracted. Its dependence on the damping strength is responsible for the reversed order of the curves in Figures 5a and 5b.

It was already shown in reference [29] that the entropy of the damped free particle at finite temperature can be smaller than its zero-temperature value as long as finite-size effects are disregarded. As we can see from Figure 5, this effect is visible in the temperature dependence of the entropy of the damped harmonic oscillator in the form of a plateau separating the regime dominated by the classical behavior and the one dominated by the free particle classical behavior. This thermodynamic scenario complements the scenario for the dynamics of an overdamped harmonic oscillator where free particle behavior is observed for not too long times [32].

The results based on the entropy of the damped harmonic oscillator shown as solid lines in Figure 5b indicate how the entropy of a free damped particle constrained to a finite-size region behaves as a function of temperature. The entropy can indeed be a nonmonotonic function of the temperature. However, as the deviations between $\tilde{S}$ and the entropy of the free particle show, there will be a potentially very small temperature interval on which the entropy changes its value from S_0 to zero as required by the third law of thermodynamics when zero temperature is approached.

7 Conclusions

Thermodynamic quantities of a damped free particle cannot directly be obtained from those of the damped harmonic oscillator by taking the limit of vanishing oscillator frequency. Nevertheless, it is possible to identify correspondences between thermodynamic anomalies of the damped free particle and features in the temperature dependence of the respective quantities for the damped harmonic oscillator. Negative specific heats for the free damped particle correspond to dips in the specific heat of the damped harmonic oscillator and minima of the entropy of the free damped particle can be related to plateaus in the entropy of the damped harmonic oscillator. For superohmic environments with $s \geq 2$, reentrant classicality can be found both for the damped free particle and the damped harmonic oscillator when the oscillator frequency tends to zero.

From the bath point of view, the main difference between the two damped systems consists in an additional peak in the change of the bath density of states either at zero frequency or at a low but finite frequency. This peak is relevant for the behavior of thermodynamic quantities in the classical regime and continues to describe the difference between damped free particle and damped harmonic oscillator down to low temperatures before finite-size effects set in.

The results for the damped harmonic oscillator show that the transition to vanishing specific heat and entropy as temperature goes to zero can occur within a rather small temperature range. It can be expected that the regularization of the thermodynamic quantities of a free damped particle due to confinement to a finite-size region behaves in a very similar way to ensure the validity of the third law of thermodynamics.

One of us (GLI) would like to thank Peter Hänggi, Peter Talkner, Astrid Lambrecht, and Serge Reynaud for useful discussions. He also gratefully acknowledges the hospitality of the Laboratoire Kastler Brossel in Paris during the preparation of this manuscript. UW has received financial support from the Deutsche Forschungsgemeinschaft through SFB/TRR 21.

Appendix: Low-frequency peak in the change of the bath density of states for arbitrary s

In Section 4 we have restricted the discussion of the low-frequency peak in the change of the bath density of states to the Ohmic case, $s = 1$, as well as the superohmic case $s = 3$. In this Appendix, we provide the generalization to arbitrary values of s.

To obtain the low-frequency peak of $\xi_{\rm osc}(\omega)$ for non-integer values of s, we assume $\omega \ll \omega_{\rm c}$ so that we may truncate the expression (7) to the form

$$\hat{\gamma}(z) = \frac{\gamma}{\sin\left(\frac{\pi s}{2}\right)}\left(\frac{z}{\omega_{\rm c}}\right)^{s-1} + \mu z. \tag{A.1}$$

Here, $\mu = \Delta M/M$ is the relative mass contribution of the bath [9]

$$\mu(s,p) = \frac{2}{\pi}\frac{\gamma}{\omega_{\rm c}}\frac{pB\left(\frac{s}{2}, p+1-\frac{s}{2}\right)}{s-2}. \tag{A.2}$$

The first term in (A.1) has a branch cut at $z = 0$.

In the regime $0 < s < 2$ and $\omega \ll \omega_{\rm c}$, the first term in (A.1) is the leading one. A low-frequency approximation of the dynamical susceptibility of the damped harmonic oscillator and the dynamical velocity response of the damped free particle is obtained by inserting this leading term into (13) and (14), respectively. By means of the expressions (11) and (12), we find that $\xi_{\rm osc}(\omega)$ and $\xi_{\rm fp}(\omega)$ differ by a low-frequency peak of the form

$$\xi_{\rm osc}(\omega) - \xi_{\rm fp}(\omega) = \frac{s}{\pi}\left(\frac{\omega}{\omega_{\rm c}}\right)^{s-1}\frac{\Gamma_{\rm r}}{\Gamma_{\rm r}^2 + \left(\frac{\omega^s}{\omega_{\rm c}^{s-1}} + \Omega_{\rm r}\right)^2} \tag{A.3}$$

with the parameters

$$\begin{aligned}\Gamma_{\rm r} &= \frac{\omega_0^2}{\gamma}\sin\left(\frac{\pi s}{2}\right)^2,\\ \Omega_{\rm r} &= \frac{\omega_0^2}{2\gamma}\sin(\pi s).\end{aligned} \tag{A.4}$$

The expression (A.3) diverges at $\omega = 0$ for subohmic damping, $s < 1$, while it goes to zero and displays a maximum at finite frequency in the superohmic regime $1 < s < 2$. The two regimes adjoin at the Ohmic point $s = 1$ where $\xi_{\rm osc}(0) - \xi_{\rm fp}(0) = \gamma/\pi\omega_0^2$. For the parameters used in Figure 1 the peak reaches $39.8/\omega_{\rm c}$ and is too high to be shown.

The expression (A.3) with (A.4) is a reasonable approximation in the regime $0 < s \lesssim 1.5$. As s approaches 2, cut-off dependent contributions, which are absent in (A.4), become increasingly relevant in the expressions for $\Gamma_{\rm r}$ and $\Omega_{\rm r}$.

In the superohmic regime $s > 2$ also the second term in (A.1) is relevant. Using the form (A.1) for $\hat{\gamma}(z)$, the low-frequency behavior of the density is found as

$$\xi_{\mathrm{osc}}(\omega) - \xi_{\mathrm{fp}}(\omega) = \frac{1}{\pi} \sum_{r=\pm 1} \frac{\varGamma}{(\omega - r\varOmega)^2 + \varGamma^2}. \tag{A.5}$$

The parameters $\varGamma$ and $\varOmega$ read

$$\begin{aligned} \varGamma &= \frac{\gamma}{2} \frac{(\omega_0/\omega_{\mathrm{c}})^{s-1}}{(1+\mu)^{(1+s)/2}} \\ \varOmega &= \frac{\omega_0}{\sqrt{1+\mu}}, \end{aligned} \tag{A.6}$$

where terms of order $(\omega_0/\omega_{\mathrm{c}})^2$ and $(\omega_0/\omega_{\mathrm{c}})^{s-2}$ have been omitted. As s approaches 2 from above, these terms become increasingly important. The expressions (A.6) effectively apply in the regime $s \gtrsim 2.5$.

From (A.3) and (A.5), we find as an important result the expressions given for $\varDelta\varSigma(s)$ in (24).

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