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Heat due to system-reservoir correlations in thermal equilibrium

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The heat flow between a quantum system and its reservoir is analyzed when initially both are in a separable thermal state and asymptotically approach a correlated equilibrium. General findings are illustrated for specific systems and various classes of non-Markovian reservoirs relevant for solid state realizations. System-bath correlations are shown to be substantial at low temperatures even in the weak coupling regime. As a consequence, predictions of work and heat for actual experiments obtained within conventional perturbative approaches may often be questionable. Correlations induce characteristic imprints in heat capacities which opens a proposal to measure them in solid state devices.

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I. INTRODUCTION

Recently, the subjects of heat and work and their distributions in the quantum regime have received considerable attention \cite{1,2} with measurement protocols proposed and implemented for solid state devices \cite{3–5}. Classical thermodynamics considers a system of interest immersed in a much larger heat bath \cite{6}, where the latter one ensures thermal equilibration of the system, but can be ignored otherwise. This weak coupling assumption works extremely accurately for basically all physical realizations on a macroscopic level. For quantum systems in contact with a heat bath realized, e.g., through mesoscopic circuits, the situation is more intricate though. The nonlocality of quantum mechanical wave functions induces system-reservoir correlations and even entanglement which may have profound impact also on thermodynamical properties \cite{7}. Common wisdom is that at least in the weak coupling regime the classical-like setting of a thermal state factorizing in system and reservoir density operators, respectively, applies. Perturbative formulations for the reduced quantum dynamics of the system such as, e.g., conventional master or Lindblad equations \cite{8} rely on this separability. Quantum mechanical system-environment correlations have been discussed in the past, mainly with respect to proper initial preparations for nonequilibrium dynamics; see, e.g., Refs. \cite{7,9}. Much less attention has been paid to this subject in the context of quantum thermodynamics \cite{10–13}, where systems are kept close to thermal equilibrium and where the focus lies on quantities such as heat \cite{14} and work \cite{2}. In many theoretical studies the simplified situation of (at least initially) factorizing thermal equilibria is taken for granted \cite{15,16}. This way, predictions for work, work distributions, and heat due to the presence of weak classical driving sources have been obtained based on conventional master equations or related approaches \cite{3,17–19}. However, in actual measurements, specifically in solid state structures, quantum correlations between system and reservoir may be of relevance not only far from but also close to and in thermal equilibrium.

The goal of this paper is to contribute to this latter topic. For that purpose, we consider the situation where an initially separable thermal state asymptotically approaches a correlated equilibrium and identify the exchanged energy to establish proper system-bath correlations as heat. While an instantaneous switching-on of system-bath interactions is typically difficult to realize experimentally, it allows us, in a first step, to quantify the impact of these correlations for various types of reservoirs. We derive general expressions and discuss specific results for systems of possible experimental interest \cite{3–5,16,20}, namely, two-level systems and harmonic modes. It turns out that even in the weak coupling regime, this heat flow is substantial at low temperatures and may become comparable to typical predictions for the work based on conventional weak coupling approaches \cite{3,17–19}. It further depends sensitively on non-Markovian features of the reservoir such that the commonly made simplification of a strictly Ohmic environment \cite{8} is always unphysical.

Hence the so-defined heat is a profound measure for aspects of quantum thermodynamics beyond descriptions accounting merely for energy level quantization of otherwise separable systems and reservoirs \cite{21}. Thus, in a second step, we show that the heat capacity of an embedded system encodes this information in the form of, e.g., a characteristic temperature dependence. It is proposed to measure it by manipulating and monitoring the reservoir in a solid state circuitry by means of advanced thermometry at cryogenic temperatures.

II. GENERAL RESULTS AND PERTURBATION THEORY

We consider a system and its surrounding in a standard setting with $H = H_S + H_R + H_C$. Since neither the system part $H_S$ nor the reservoir part $H_R$ do commute with the coupling $H_C$, the canonical thermal operator of the full compound

$$W_B = \frac{e^{-\beta H}}{Z},$$

with corresponding partition function $Z$ describes system-bath correlations. The nature of these correlations has been an issue if intense research (see e.g. \cite{10,22–25}) with the general conclusion that in many system at sufficiently low temperatures they are nonclassical and related to
entanglement. This is not the case for a so-called factorized thermal state \[8\]

\[ W_f = \frac{e^{-\beta H_s}}{Z_S} \otimes \frac{e^{-\beta H_b}}{Z_R}, \]  

(2)

where \( Z_S \) and \( Z_R \) denote the partition functions of isolated system and isolated bath, respectively.

We now think of a setup where one initially starts with a factorized state \( W_f \) and then monitors the asymptotic state when the system has fully equilibrated to \( W_R \). Due to the nonequilibrium initial preparation, energy will be exchanged between system and bath such as to establish proper equilibrium correlations between them. Following the first law of thermodynamics in absence of external driving, this shift in system energy,

\[ Q_{\text{corr}} = \langle H_S(t \rightarrow \infty) \rangle - \langle H_S(0) \rangle = \langle H_S \rangle_f - \langle H_S \rangle_f, \]

(3)

can be interpreted as heat due to system-bath correlations. Here, \( \langle \cdot \rangle_f \) expectation values taken according to the distributions \( 1 \) and \( 2 \), respectively. Note that, in contrast to thermal state \[8\]

\[ \text{JOACHIM ANKERHOLD AND JUKKA P. PEKOLA PHYSICAL REVIEW B 90, 075421 (2014)} \]

where \( \beta/f \) are expectation values taken according to the density operator of the system alone according to the full Hamiltonian \( \hat{H} \). Accordingly, the reduced density operator of the system alone \( \rho(t) = \text{Tr}_R[W(t)] \) evolves from a Gibbs state into a thermal state which in general is not of Gibbs form.

A formal expression can be found based on the relation between the respective partition functions and energy expectation values, i.e.,

\[ Q_{\text{corr}} = -\frac{\partial}{\partial \beta} \ln(Z/Z_S) - \langle H_R + H_C \rangle_f. \]

(4)

While for specific systems such as harmonic oscillators, this expression can be evaluated exactly, in general, a perturbative treatment must be applied. In the sequel, we focus on the weak coupling regime and calculate the heat exchange in lowest nonvanishing order in \( H_C \) (canonical perturbation theory \[9\]). This is conveniently done by iterating the operator identity

\[ e^{-\beta H} = e^{-\beta H_0} \left( 1 - \int_0^{\beta} d\lambda e^{\lambda H} H e^{-\lambda H} \right), \]

(5)

with \( H_0 = H_S + H_R \). Since in equilibrium, the bath is assumed to exert no net force, i.e., \( \langle H_C \rangle_f = 0 \), one needs to iterate at least up to the second order in \( H_C \). This yields

\[ Z \approx Z_S Z_R \left[ 1 + \frac{1}{H_S^2} \int_0^{\beta} d\lambda \int_0^{\lambda} d\tau \langle H_C(-i\tau)H_C \rangle_f \right], \]

(6)

with Heisenberg operators taken with respect to \( H_0 \). When combined with a similar result for the not-normalized energy expectation values one finds

\[ Q_{\text{corr}} \approx \frac{1}{H_S^2} \int_0^{\beta} d\lambda \int_0^{\lambda} d\tau \langle [H_S H_C(-i\tau)H_C]_f \rangle - \langle H_S \rangle_S \langle H_C(-i\tau)H_C \rangle_f. \]

(7)

This result applies to arbitrary system operators when \( H_S \) is replaced accordingly. Here and in the sequel, \( \langle \cdot \rangle_R,S \) denote expectation values based on the individual canonical operators of reservoir and system, respectively.

To proceed, consider the generic situation of a bath with Gaussian noise properties bilinearly coupled to a system via \( H_C = q \hat{E} \) with \( \hat{E} \) being a collective bath mode and \( q \) a dimensionless system operator. The reservoir is then completely determined by the equilibrium correlation \( L(t) = \langle \hat{E}(t)\hat{E}^\dagger \rangle_R \) so that

\[ Q_{\text{corr}} = \frac{1}{H_S^2} \int_0^{\beta} d\lambda \int_0^{\lambda} d\tau L(-i\tau) \langle [H_S q(-i\tau)q]_S \rangle - \langle H_S \rangle_S \langle q(-i\tau)q \rangle_S. \]

(8)

In imaginary time the bath correlation takes the form \[7\]

\[ L(-i\tau) = \mu : \delta(\tau) : -k(\tau), \]

(9)

where \( : \delta(\tau) : \) denotes a periodically continued \( \delta \) function beyond the interval \( \mu \) and

\[ k(\tau) = \frac{1}{\beta} \sum_{n=-\infty}^{\infty} \xi_n e^{i\nu_n \tau} \]

(10)

is periodic in \( h\beta \) and

\[ \xi_n = \left| \nu_n \right| \tilde{\gamma} \left| \nu_n \right| \]

(11)

contains the Matsubara frequencies \( \nu_n = 2\pi n / h\beta \) and the Laplace transform of the classical damping kernel

\[ \tilde{\gamma}(z) = \frac{2}{\pi} \int_0^\infty d\omega I(\omega) / \omega \frac{z}{z^2 + \omega^2}. \]

(12)

Note that this perturbative treatment is valid as long as \( Q_{\text{corr}} \) is sufficiently smaller than typical bare level spacings of the system of interest.

### III. TWO-LEVEL SYSTEM

According to common experimental realizations in superconducting circuits, we first discuss a two-level system \[5, 26\]

\[ H_S = -\frac{h\Delta}{2} \sigma_x \]

(13)

with coupling operator \( q = \sigma_x \). Performing the time integrations in \( 8 \) with \( 10 \) yields with the bare result

\[ \langle H_S \rangle_S = -\frac{h\Delta}{2} \text{tanh}(\theta/2). \]

(14)

the expression

\[ Q_{\text{corr}} = \frac{h\Delta^2}{2 \cosh^2(\theta/2)} \left[ \sum_{n \geq 1} \frac{\xi_n}{\Delta^2 + \nu_n^2} \left[ 1 - \frac{1}{\Delta^2 + \nu_n^2} \sinh(\theta) \right] \right]. \]

(15)

with the dimensionless inverse temperature \( \theta = \Delta h\beta \). Reservoir properties only appear in \( \xi_n \) so that this result allows one to analyze \( Q_{\text{corr}} \) for various spectral distributions. We start with a Drude bath

\[ I(\omega) = \eta \omega^2 e^{-\omega^2 / \omega_c^2}, \]

(16)
with coupling parameter $\eta$ and Drude frequency $\omega_c$. This then implies $\gamma(z) = \eta \omega_c/(\omega_c + z)$. Numerical results are shown in Fig. 1. At high temperatures $\theta \ll 1$, the heat grows quadratically

$$Q_{\text{corr,high}} \approx \frac{\eta \Delta}{\theta^2},$$

so that in this regime, as expected, a factorizing initial state is an accurate approximation. In contrast, at moderate and low temperatures the exchanged heat is quite substantial even for weak coupling and it tends to saturate at very low temperatures $\theta \gg 1$. For $\omega_c \gg \Delta$ the leading contributions read

$$Q_{\text{corr,low}} \approx \frac{\eta \Delta}{2\pi} \ln(\omega_c/\Delta) + \frac{\eta \hbar}{4\omega_c} - \frac{\eta \Delta \pi}{\theta^2 6}.$$

Here, the logarithmic dependence on the Drude frequency reflects the impact of zero-point fluctuations of the reservoir (Lamb shift) which have recently been measured in a circuit quantum electrodynamical setup [27]. It originates from the second term in brackets in (15) while the first one dominating weak coupling and it tends to saturate at very low temperatures $\theta \gg 1$. For $\omega_c \gg \Delta$ the exchanged heat diverges and $Q_{\text{corr}}$ exchanged between system and bath is much less pronounced in the super-classical and moderate quantum regime is exponentially suppressed. The changeover to the deep quantum domain for harmonic systems. This not only allows exact results valid also for strong system-bath coupling predictions, but may also be of experimental relevance [16,20], in solid state systems, e.g., for circuits including Josephson junctions such as superconducting quantum interference devices (SQUID). This way, we consider

$$H_S = \frac{p^2}{2m} + \frac{m\omega_0^2}{2} q^2,$$

which yields under a factorized equilibrium

$$\langle H_S \rangle_s = \frac{\hbar \omega_0}{2} \coth(\omega_0 h \beta/2).$$

IV. EXACT RESULTS: HARMONIC OSCILLATOR

Exact results valid also for strong system-bath coupling can be obtained for harmonic systems. This not only allows one to inspect the validity of weak coupling predictions, but may also be of experimental relevance [16,20], in solid state systems, e.g., for circuits including Josephson junctions such as superconducting quantum interference devices (SQUID).
Instead, in correlated equilibrium one has [7]
\[
\langle H_S \rangle_\beta = \frac{1}{\beta} + \frac{1}{\beta} \sum_{n \geq 1} \frac{v_n^2 + \rho_n^2}{\nu_n^2 + \omega_0^2 + \rho_n^2},
\]
(23)
with the second term describing quantum mechanical fluctuations. Hence one finds for the exchanged heat (15) the exact expression
\[
Q_{\text{corr, ex}} = \frac{1}{\beta} \sum_{n \geq 1} \frac{\rho_n \nu_n^2 - \omega_0^2}{\nu_n^2 + \omega_0^2 + \rho_n^2},
\]
(24)
In the regime of weak coupling, this reduces according to (8) to
\[
Q_{\text{corr, app}} \approx \frac{1}{\beta} \sum_{n \geq 1} \frac{\rho_n \nu_n^2 - \omega_0^2}{\nu_n^2 + \omega_0^2 + \rho_n^2},
\]
(25)
which can further be approximated for a Drude model (16). At high temperatures \( \theta \ll 1 \) one gains
\[
Q_{\text{corr, high}} \approx \frac{\eta \omega_c}{2 \pi} \theta,
\]
(26)
with a linear rise instead of a quadratic one in (17) due to an unbounded energy spectrum. The factorizing assumption seems thus to be better justified for two-level systems than for systems with an infinite number of accessible states. In the zero temperature regime and for weak coupling, the harmonic oscillator reduces to a two-level system which in turn may verify the validity of the result (18). Indeed, the exchanged heat from (25) coincides with the result (18) with \( \Delta \) replaced by \( \omega_0 \).

To analyze the accuracy of the weak coupling treatment for finite temperatures, we depict in Fig. 3 the ratio of the approximated result (25) to the exact one (24). Notably, the perturbative treatment works fairly accurately also for somewhat stronger dissipation and over the full temperature range. Its predictions tend to deviate substantially from the exact values, however, in the overdamped regime \( \eta > 1 \). In this regime, the reservoir induced level broadening \( \eta \omega_c \) even exceeds the bare level spacing so that for very strong dissipation and at low temperatures \( \eta \gg 1 \), \( \omega_c \beta \gg \omega_0 \beta \gg 1 \), one finds from (24)
\[
Q_{\text{corr, over}} \approx \frac{\eta \omega_c}{2 \pi} \ln(\eta \nu_c/\omega_0).
\]
(27)
Note that, in this latter regime, it is known that entanglement correlates system and bath [10,24].

V. HEAT CAPACITY

Heat capacity is a central experimental quantity in bulk systems. It has thus been proposed as a measure to access information about entanglement [31]. Recently, this has indeed been demonstrated for large spin ensembles [32].

Here, we follow a somewhat more ambitious route and consider the heat capacity of a single quantum degree of freedom embedded in a thermal bath. We show to what extent system-bath correlations induce deviations from bare predictions. Accordingly, we consider heat capacities \( C_\beta \) in full thermal equilibrium and compare them against those of a bare system thermal state \( C_S \). Both follow from the temperature dependence of the respective system energy expectation values (internal energy), the difference of which \( Q_{\text{corr}} \) has been calculated above. Classically or for bare systems, heat capacities can be obtained from the temperature dependence of either internal energies or partition functions. In full thermal equilibrium, however, only the energy-based definition leads to physically meaningful predictions [14]. We thus obtain
\[
\frac{C_\beta}{C_S} = 1 + \frac{\partial Q_{\text{corr}}(T)}{C_S \partial T}.
\]
(28)

For the two-level system (13), the bare textbook expression [6] reads
\[
\frac{C_S}{k_B} = \frac{\theta^2}{4 \cosh^2(\theta/2)}.
\]
(29)
This result can now be compared with the full heat capacity stemming from (15); see Fig. 4. In particular, based on (17),

FIG. 3. (Color online) Approximate value of the exchanged heat for weak coupling \( Q_{\text{corr, app}} \) for a harmonic oscillator normalized to the exact value \( Q_{\text{corr, app}} \) vs inverse temperature \( \theta \) in a Drude bath \( \omega_c/\Delta = 50 \). Depicted are results for various coupling strengths \( \eta = 0.01 \) (black, solid), 0.05 (blue, long-dashed), 0.2 (purple, short-dashed), 0.5 (green, dotted–closely spaced), and 1 (red, dotted).

FIG. 4. (Color online) Ratio of the heat capacities \( C_\beta/C_S \) of correlated and bare thermal state vs inverse temperature for a two-level system and various bath cutoff frequencies \( \omega_c/\Delta \). Depicted are results for a harmonic system are depicted (purple, dotted) for \( \omega_c/\omega_0 = 50 \). Coupling strength is \( \eta = 0.1 \).
in the high temperature limit both basically coincide
\[ \frac{C_\beta}{C_S} \approx 1 - 2\frac{\eta \omega_c}{\pi^2 \theta}. \] (30)

However, at very low temperatures, according to (18),
\[ \frac{C_\beta}{C_S} \approx \frac{\eta \omega_c \pi}{30^3 c^3 \theta}. \] (31)

One observes a strong enhancement due to the fact that the full \( C_\beta \) decays only algebraically in contrast to (29). For the full heat capacity the changeover between the classical/moderate quantum to the deep quantum regime is related to substantial suppression and enhancement, respectively, compared to the bare prediction. This feature is more pronounced for large Drude frequencies.

For harmonic systems (21), the well-known bare result
\[ \frac{C_S}{k_B} = \frac{\theta^2}{4 \sinh^2(\theta/2)} \] (32)
leads for high temperatures together with (26) to
\[ \frac{C_\beta}{C_S} \approx 1 - \frac{\eta \hbar \omega_c}{4\pi^2 \theta^2}. \] (33)

For low temperatures and weak coupling one regains the expression (31). Exact data can easily be obtained from (24). Figure 4 reveals an only weak temperature dependence of \( C_\beta / C_S \) in the range shown and the overall behavior is basically insensitive to \( \omega_c \) for \( \omega_c / \omega_0 \gg 1 \). The strong coupling domain, where perturbative results are not applicable, is addressed in Fig. 5. Interestingly, larger coupling has not always had the tendency to increase the heat capacity compared to the bare one even at moderately low temperatures. One obtains a substantial enhancement only for very low temperatures. Notably, this occurs in the regime \( \eta > 1 \) and \( \eta \omega_0 \hbar \beta \gg 1 \), also known as the quantum Smoluchowski limit [33], where the level broadening due to friction exceeds both the bare level spacing and the thermal energy scale so that quantum dynamics tends to become more classical, though, with substantial quantum fluctuations.

![Graph showing the ratio \( C_\beta / C_S \) for various inverse temperatures \( \omega_0 \hbar \beta = 0.1 \) (red, dotted), 1 (green, short-dashed), 4 (purple, long-dashed), and 6 (blue, solid).](image)

**FIG. 5.** (Color online) Ratio \( C_\beta / C_S \) of correlated and bare thermal state of a harmonic oscillator vs coupling strength for various inverse temperatures \( \omega_0 \hbar \beta = 0.1 \) (red, dotted), 1 (green, short-dashed), 4 (purple, long-dashed), and 6 (blue, solid).

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**VI. EXPERIMENTAL DETECTION**

We now propose a scheme to retrieve information about the impact of system-bath quantum correlations by measuring the heat capacity of the embedded system. The latter one is taken as a solid state implementation of a two-level system, e.g., in the form of a biased SQUID coupled inductively to a resistor (reservoir). In these devices, at cryogenic temperatures, electronic degrees of freedom (Fermi gas) are very weakly coupled to the underlying phonon background with relaxation times on the order of 100 \( \mu s \), while electron-electron interaction leads to equilibration in the Fermi gas within a few nanoseconds. It is this latter heat bath which dominantly interacts with the SQUID. By means of rf thermometry [34–36], it is now possible to monitor the actual temperature of the Fermi gas of a mesoscopic metallic island on sub-\( \mu s \) time scales which in turn allows one to monitor its relaxation dynamics after a heating pulse has been applied.

We imagine a situation where both SQUID and its electronic environment are in thermal equilibrium at a temperature \( T_1 \). A short and weak heating pulse of duration \( \tau_p \) is sent to the reservoir, where it leads within a few nanoseconds to a temperature \( T_2 \) of the electron gas. The coupling between the system and this reservoir is assumed to be weak such that the coupling rate \( \Gamma \) depending on their correlations obeys \( \tau_p \approx 1/\Gamma_c \ll 1/\Gamma_\text{eq} \), with \( \Gamma_c \) (\( \Gamma_\text{eq} \)) being the equilibration rate of the Fermi gas (Fermi gas and phonon bath). Accordingly, on a much shorter time scale than \( 1/\Gamma_\text{eq} \), the electronic reservoir will transfer energy to the two-level system and eventually equilibrate with it. This loss of energy of the electronic reservoir corresponds to a heat flow \( Q_R = C_R \delta T \), where \( C_R \) is the heat capacity of the bare reservoir and \( \delta T \) is the temperature drop \( T_2 - T_1 \). The heat flow balances the heat flow received by the system when it heats up from \( T_1 \to T_2 - \delta T \), i.e., \( C_\beta(T_2 - \delta T - T_1) \) with \( C_\beta \) being the heat capacity of the embedded system. Hence one arrives at
\[ \frac{C_\beta}{C_R} \approx \frac{\delta T}{T_2 - T_1 - \delta T}, \] (34)

where the small portion of heat lost to the phonon bath during the equilibration between system and reservoir has been neglected. It can be estimated to be on the order of \( \Gamma_\text{ep} \). A specific advantage of this protocol is that the heat capacity \( C_R \) (known for typical bulk materials) can be extracted *in situ* from the known energy carried in the initial heating pulse \( E_m \) via \( C_R = E_m/(T_2 - T_1) \). Experimental data for varying \( T_1 \) and \( T_2 \) (but still small \( T_2 - T_1 \)) can then be compared with predictions for the bare system according to the results of the previous section.

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**VII. CONCLUSION**

The quantum correlations between a system and its reservoir are analyzed in terms of the heat exchange during the equilibration when starting initially with a separable thermal state. Specific results are discussed for systems for which recent theoretical predictions of work, work distributions, and heat have been made based on factorized thermal states. System-bath correlations substantially influence heat...
capacities in the low temperature regime which may open a way to detect them in solid state devices.

As we have shown, system-bath correlations induce deviations in the weak coupling quantum regime when compared to predictions based on separable thermal states on the order of 10% of the bare level splitting. When a weak external driving is exerted to the system, according to the first law of thermodynamics, part of its energy is deposited into the bath in the form of heat. Weak coupling approaches such as master equations obtain work and heat based on separable bath in the form of heat. For example, for a monochromatic pulse with frequency \( \Omega \) and amplitude \( \lambda_0 \) in resonance with a two-level system, they are applicable as long as work and heat are on the order of \( (\lambda_0/\hbar\Omega)^2 \ll 1 \) which basically matches the impact of system-bath correlations. For actual realizations at low temperatures, their predictions may thus be of limited reliability. System-bath correlations may be substantial not only in theoretical approaches to understand work and heat at the quantum level but also to analyze experimental data.

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