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Analytic continuation of generalized susceptibility and system–reservoir correlation effects

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Abstract. A fully non-Markovian calculation of the generalized susceptibility of a quantum system weakly coupled to a heat bath is presented. In contrast to most of the treatments we provide a complete solution in the sense that it includes the analytic continuation to the lower half-plane. Various applications are discussed including the consideration of boson and fermion heat baths. It is shown that our result takes into account system–reservoir correlations which are neglected in the Markovian approximation.

1. Introduction

Since the seminal works of Callen and Welton [1] and Kubo [2], the intimately related concepts of time correlation function and generalized susceptibility (also known as the linear response function) have been shown to be powerful starting points for a great number of successful theoretical developments in non-equilibrium statistical physics. A list of the fields of application would be almost comprehensive, covering the simplest Brownian motion to dynamic critical phenomena [3, 4] or the transport properties of dense fluids [5].

The calculations usually focus upon the time correlation function since the generalized susceptibility (GS) follows immediately from the fluctuation–dissipation theorem and the Kramers–Kronig dispersion relations [6]. A common method starts from a set of Markovian master equations which may be introduced either from phenomenological or from microscopic theories [7]. Then the time-dependent probability distribution for the set of states of the system may be obtained as the solution of such equations which immediately yields the two-time equilibrium probability distribution and hence the time correlation function. This derivation, however, is in practice limited to systems having no more than a few degrees of freedom. If this is not the case, the method must be modified in order to avoid solving directly a huge number of master equations and, in this respect, the Ising model provides a very interesting example. In fact, in such a case it is more convenient to deal with the first-order equations for the time correlation functions which are directly obtained from the set of master equations. Then the solution is exactly derived for the one-dimensional model and, for the higher-dimensional ones, it is obtained from a mean-field approximation [4].

In any case, however, if memory effects are non-negligible the above method turns out to be inapplicable, the main obstacle being that the canonical distribution of the system becomes non-stationary. In fact, this can easily be understood by taking into account the fact that the theory usually assumes vanishing initial correlations between the system and

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its environment which thus excludes a stationary state for the whole system. Nevertheless, in the Markovian approximation correlations are built on a negligible time scale, i.e. if the system is assumed to possess a canonical distribution at $t = 0$, equilibrium correlations are reached instantaneously.

The aim of this paper is to present a reformulation of the above method which, while conserving the simple input of an uncorrelated initial state, is fully non-Markovian with a unique approximation of weak coupling between the system and its reservoir. Unlike most of the treatments, we provide a complete solution in the sense that it consists of the GS together with its analytic continuation to the lower half-plane, the latter being particularly useful for practical evaluations in the time domain. In addition, we show that the analytic continuation in the form of Taylor expansions around the origin makes room for perturbative schemes, leading to simpler approximate expressions for the GS including only limited memory effects. Our expression for the GS is given in terms of matrices of the Hilbert space of the system with an implicit dependence on the reservoir degrees of freedom which is the most suitable form for an implementation of a phenomenological theory. A most important feature of our result follows by means of a comparison with the exact Kubo formula for the linear response of a quantum system. In fact, we show that the non-Markovian GS takes into account system–reservoir correlations which otherwise (i.e. in the Markovian limit) are neglected. We discuss various applications. We focus first upon a harmonic oscillator providing a detailed calculation of the phonon number GS both for boson and fermion environments. From this example it is clearly shown that the singularity spectrum of the GS brings valuable information regarding the dynamics of the system, e.g. for a highly degenerate fermion heat bath we find an infinite set of branch cuts implying a complex non-exponential time decay. As a second application we consider a particular multi-level system, namely a spin j in an external magnetic field and weakly coupled to a phonon heat bath. Here a brief analysis leads immediately to a connection with previous results which brings system–reservoir correlations into the discussion.

Finally it is worthwhile mentioning previous work by Tanimura and Kubo [8] who developed a *phenomenological Markovian* calculation of two-time correlation functions valid to all orders of the system–bath interaction that, in the lowest order, reduces to the conventional results of the master equation approach.

This paper is organized as follows. In section 2 starting from the Liouville–von Neumann equation of motion with the usual uncorrelated initial condition, the system is allowed to reach equilibrium before a driving force is turned on. Next we obtain a 'response density matrix' which yields the formula for the GS. We give detailed expressions for the two matrices appearing in the formula, provide the analytic continuation rules and discuss several points and applications. In section 3 we study the time dependence of the GS which makes room for a direct comparison with the Kubo formula and in section 4 we prove that our result reduces in the Markovian limit to the one obtained from the usual method. Finally in section 5 we state some concluding remarks.

2. Calculation of the GS

We start from a time-dependent Hamiltonian of the form

$$H(t) = H_S + H_R + H_{SR} - \lambda(t)A \tag{2.1}$$

where $H_S(H_R)$ denotes the system (reservoir) Hamiltonian and H_{SR} the interaction potential between them. A is an observable of the system which, for simplicity, is assumed to

commute with H_S and $\lambda(t)$ is a C-number external driving force which is the only time-dependent quantity. The whole system (system + reservoir) density matrix in the common basis of eigenstates of H_S, A and H_R can be written as a sum of two terms, namely a diagonal matrix ρ_0 and the remaining off-diagonal matrix ρ_C. With such definitions the Liouville–von Neumann equation can be written as follows:

$$\mathrm{i}\dot{\rho}_0 = L'_{0C}\rho_C \tag{2.2}$$

$$\mathrm{i}\dot{\rho}_C = L'_{C0}\rho_0 + (L_S + L_R - \lambda L_A + L'_{CC})\rho_C. \tag{2.3}$$

The above expressions are simply the diagonal and off-diagonal parts of the original equation of motion. In fact, if P_0 (P_C) denotes a projector onto the diagonal (off-diagonal) part, we have $L'_{0C} = P_0 L_{SR} P_C$ and similar definitions for L'_{C0} and L'_{CC}, where L_{SR} denotes the commutator of H_{SR}, $\hbar^{-1}[H_{SR}, \ ]$. Analogously, L_S, L_R and L_A denote the respective commutators of H_S, H_R and A. Note that $L'_{00} = 0$, since an off-diagonal H_{SR} was assumed without loss of generality.

We shall work under a weak-coupling approximation which consists of neglecting L'_{CC} in equation (2.3). Then, assuming the simplest initial condition $\rho_C(t=0) = 0$, we have

$$\dot{\rho}_0(t) = -\int_0^t \mathrm{d}t'\, L'_{0C} \exp[-\mathrm{i}(L_S + L_R)t'] \exp\left[\mathrm{i}\int_{t-t'}^t \mathrm{d}t''\, \lambda(t'')L_A\right] L'_{C0}\rho_0(t-t') \tag{2.4}$$

which follows from solving equation (2.3) in ρ_C and replacing the result in (2.2). The formal solution of equation (2.4) follows immediately via Laplace transformation:

$$\begin{aligned}\rho_0(t) = {} & \frac{\mathrm{i}}{2\pi}\int_{-\infty+\mathrm{i}0}^{+\infty+\mathrm{i}0} \mathrm{d}z\, \mathrm{e}^{-\mathrm{i}zt}[z + \Psi(z)]^{-1}\rho_0(t=0) \\ & + \frac{\mathrm{i}}{(2\pi)^2}\int_{t_0}^{+\infty}\mathrm{d}t'\,\lambda(t')\int_{-\infty}^{+\infty}\frac{\mathrm{d}\omega}{\omega}\int_{-\infty+\mathrm{i}0}^{+\infty+\mathrm{i}0}\mathrm{d}z\,\mathrm{e}^{\mathrm{i}(\omega t' - zt)}[z+\Psi(z)]^{-1} \\ & \times[\Omega(z-\omega) - \Omega(z)][z - \omega + \Psi(z-\omega)]^{-1}\rho_0(t=0)\end{aligned} \tag{2.5}$$

where we have retained only first-order terms in the driving force λ. The two operators appearing in (2.5) are

$$\Omega(z) = L'_{0C}(L_S + L_R - z)^{-1}L_A L'_{C0} \tag{2.6}$$

and the so-called collision operator in the weak-coupling approximation [9–11],

$$\Psi(z) = L'_{0C}(L_S + L_R - z)^{-1}L'_{C0}. \tag{2.7}$$

We have assumed a driving force which vanishes for $t < t_0$ and tends to zero for $t \to +\infty$ or, more precisely, has a regular Laplace transform at the origin. The next step consists of tracing in equation (2.5) over the reservoir degrees of freedom in order to obtain the diagonal component of the system density matrix. Recalling that $\rho_0(t=0)$ is assumed to be of the form $\rho_S(0)\rho_R(0)$, we see that the task reduces to averaging the operators in the integrands of (2.5) over the initial state of the reservoir. In this respect, we have recently given a useful property of the collision operator valid in the thermodynamic limit, namely [11]

$$\langle[\Psi(z)]^n\rangle_R = [\langle\Psi(z)\rangle_R]^n \qquad (n = 2, 3, \ldots)$$

where $\langle O\rangle_R = \mathrm{Tr}_R[O\rho_R(0)]$ denotes the average of an operator O over the initial state of the reservoir. This decoupling property might, at first sight, seem surprising; we refer the reader to the appendix in [11] for details of the proof. Here we shall only mention that the key point of such a proof is quite simple since it rests upon equalities such as

$$\langle a_\beta a_\beta^\dagger a_\alpha^\dagger a_\alpha\rangle_R = \langle a_\beta a_\beta^\dagger\rangle_R\langle a_\alpha^\dagger a_\alpha\rangle_R$$

for creation and annihilation operators of reservoir particles. However, it is important to remark that the cases for which the above equation does not hold, i.e. $\alpha = \beta$, become a null measure set in the thermodynamic limit (continuum limit of reservoir labels). In the present problem we shall need the following generalization of this lemma which, fortunately, can be shown in complete analogy with the previous derivation:

$$\langle \Psi_{A_1} \Psi_{A_2} \dots \Psi_{A_n} \rangle_R = \langle \Psi_{A_1} \rangle_R \langle \Psi_{A_2} \rangle_R \dots \langle \Psi_{A_n} \rangle_R \tag{2.8}$$

where $\Psi_{A_i} = L'_{0C} A_i L'_{C0}$, A_i being an operator which is a function of L_S, L_R and L_A. Thus using (2.8) the averages in (2.5) are easily extracted, leading to

$$\begin{aligned} \rho_S(t) = \frac{\mathrm{i}}{2\pi} \int_{-\infty+\mathrm{i}0}^{+\infty+\mathrm{i}0} \mathrm{d}z\, \mathrm{e}^{-\mathrm{i}zt} [z + \phi(z)]^{-1} \rho_S(t=0) \\ + \frac{\mathrm{i}}{(2\pi)^2} \int_{t_0}^{+\infty} \mathrm{d}t'\, \lambda(t') \int_{-\infty}^{+\infty} \frac{\mathrm{d}\omega}{\omega} \int_{-\infty+\mathrm{i}0}^{+\infty+\mathrm{i}0} \mathrm{d}z\, \mathrm{e}^{\mathrm{i}(\omega t' - zt)} [z + \phi(z)]^{-1} \\ \times [\varphi(z-\omega) - \varphi(z)] [z - \omega + \phi(z-\omega)]^{-1} \rho_S(t=0) \end{aligned} \tag{2.9}$$

where $\phi(z) = \langle \Psi(z) \rangle_R$ and $\varphi(z) = \langle \Omega(z) \rangle_R$ are reduced operators acting only upon the system degrees of freedom.

Now recall that the system must have reached equilibrium before the driving force is turned on, i.e. in (2.9) we must take in the limit $t \to +\infty$ keeping $\tau = t - t_0$ as a constant, which is easily achieved via the following change of variables:

$$\begin{aligned} \theta &= (t + t')/2 \qquad \tau' = t - t' \\ u &= (z + \omega)/2 \qquad v = z - \omega. \end{aligned} \tag{2.10}$$

Thus one gets

$$\rho_S(\tau) = \rho_S^{\mathrm{eq}} + \int_0^{\tau} \mathrm{d}\tau'\, \lambda(\tau') \rho_{\mathrm{resp}}(\tau - \tau') \tag{2.11}$$

where

$$\rho_S^{\mathrm{eq}} = \lim_{z \to +\mathrm{i}0} z [z + \phi(z)]^{-1} \rho_S(t=0) \tag{2.12}$$

is the equilibrium distribution of the system and ρ_{resp} is a 'response density matrix' with a Laplace transform given by

$$\tilde{\rho}_{\mathrm{resp}}(z) = -[z + \phi(z)]^{-1} \left[\frac{\varphi(z) - \varphi(+\mathrm{i}0)}{z} \right] \rho_S^{\mathrm{eq}}. \tag{2.13}$$

Here we want to analyse a possible incidence in our derivation of the question of positivity breaking of the system density matrix which has recently received attention from several authors [12]. To this aim, let us focus upon equation (2.11); on the one hand, assuming a reservoir in a canonical or macrocanonical equilibrium at $t = 0$, ρ_S^{eq} will be simply the canonical distribution of the system (an explicit proof is given in the appendix). Now, regarding the second term of (2.11), from the infinitesimal nature of λ we conclude that the positivity of $\rho_S(\tau)$ could only be broken for vanishing temperatures which will therefore be excluded from our treatment.

Finally the mean value of A is thus

$$\begin{aligned} \langle A(\tau) \rangle &= \mathrm{Tr}_S [A \rho_S(\tau)] \\ &= \langle A \rangle_{\mathrm{eq}} + \int_0^{\tau} \mathrm{d}\tau'\, \lambda(\tau') \alpha(\tau - \tau') \end{aligned} \tag{2.14}$$

with $\alpha(\tau - \tau')$ the function for which the Laplace transform is just the GS:

$$\tilde{\alpha}(z) = -\mathrm{Tr}_S\{A[z + \phi(z)]^{-1}\Pi(z)\rho_S^{\mathrm{eq}}\} \tag{2.15}$$

where we have defined

$$\begin{aligned}\Pi(z) &= [\varphi(z) - \varphi(+\mathrm{i}0)]/z \\ &= \sum_{k\geqslant 0} \varphi^{(k+1)}(0) z^k/(k+1)!\end{aligned} \tag{2.16}$$

having assumed in the second line the holomorphy of $\varphi(z)$ at the origin to be discussed below. The result (2.15) deserves several comments; the operators appearing in it are just matrices of the Hilbert space of the system. The elements of $\varphi(z)$ are:

$$\begin{aligned}\varphi_{s_1 s_2}(z) = &\sum_{s'}\sum_{R'R} |\Lambda_{s'R';s_1R}|^2 \\ &\times\left[\frac{f(\omega_{s'}) - f(\omega_{s_1})}{\omega_{s'} - \omega_{s_1} + \omega_{R'} - \omega_R - z} + \frac{f(\omega_{s_1}) - f(\omega_{s'})}{\omega_{s_1} - \omega_{s'} + \omega_R - \omega_{R'} - z}\right]\rho_R\delta_{s_1 s_2} \\ &-\sum_{R'R} |\Lambda_{s_1R';s_2R}|^2 \\ &\times\left[\frac{f(\omega_{s_1}) - f(\omega_{s_2})}{\omega_{s_1} - \omega_{s_2} + \omega_{R'} - \omega_R - z} + \frac{f(\omega_{s_2}) - f(\omega_{s_1})}{\omega_{s_2} - \omega_{s_1} + \omega_R - \omega_{R'} - z}\right]\rho_R\end{aligned} \tag{2.17}$$

where $s(R)$ are system (reservoir) indices, $\hbar\Lambda_{sR;s'R'}$ are the matrix elements of H_{SR} in the common basis of eigenstates of H_S, H_R and A, $\hbar\omega_s$ ($\hbar\omega_R$) denotes the energy levels of H_S (H_R) and ρ_R are the (diagonal) elements of $\rho_R(0)$. For simplicity we have assumed an observable A which is a direct function of H_S, i.e. $A = \hbar f(H_S/\hbar)$. The matrix elements of the reduced collision operator $\phi(z)$ are obtained from (2.17) by making the replacement $f(\,) - f(\,) \to 1$ [11]. It is important to bear in mind, however, that equation (2.17), as it stands, is only valid for $\mathrm{Im}\, z > 0$. Therefore to achieve a complete solution useful for practical applications we should also provide the analytic continuation of the GS to the lower half-plane. In a recent article [11], we have reported analytic continuation and Taylor expansion formulae for the collision operator $\phi(z)$ which were obtained by taking into account that in the thermodynamic limit (a reservoir with a continuous energy spectrum) the matrix elements of $\phi(z)$ become Cauchy integrals of the form

$$C(z) = \frac{1}{2\pi\mathrm{i}}\int_{-\infty}^{+\infty}\frac{\mathrm{d}\omega}{(\omega - z)}c(\omega) \qquad (\mathrm{Im}\, z > 0) \tag{2.18}$$

with an even kernel $c(\omega) = c(-\omega)$. In view of the analogy between the matrix elements of $\phi(z)$ and $\varphi(z)$, it is easy to show that the latter are also Cauchy integrals, but this time with an odd kernel. The analytic continuation and Taylor expansion formulae for the cases $c(\omega)$ even and [odd] are [13]

$$C(z) = -[+]C(-z) + c(z) \qquad (\mathrm{Im}\, z < 0) \tag{2.19}$$

$$C(z) = \frac{1}{2\pi\mathrm{i}}\mathcal{P}\int_{-\infty}^{+\infty}\frac{\mathrm{d}\omega}{(\omega - z)}c(\omega) + c(z)/2 \qquad (z \text{ real}) \tag{2.20}$$

$$C(z) = \sum_{n\geqslant 0} C^{(n)}(0) z^n/n! \tag{2.21}$$

with

$$C^{(n)}(0) = c^{(n)}(0)/2 \qquad n \text{ even [odd]} \tag{2.22}$$

$$C^{(n)}(0) = \frac{n!}{2\pi\mathrm{i}}\int_{-\infty}^{+\infty}\frac{\mathrm{d}\omega}{\omega^{n+1}}\left[c(\omega) - \sum_{k=0}^{n-1} c^{(k)}(0)\omega^k/k!\right] \qquad n \text{ odd [even]} \tag{2.23}$$

and $\mathcal{P}$ indicates the Cauchy principal part. In the above expressions it was assumed that the kernel $c(z)$ is an analytic function along the whole real axis which, as we shall see later through various examples, is a commonly encountered situation. Note that equation (2.19) shows the important fact that the singularities of $C(z)$ are those of the kernel $c(z)$.

As is obvious from (2.15), practical evaluations of the GS will generally require a numerical diagonalization of the matrix $\phi(z)$ since only a few model systems such as a harmonic oscillator [14] or a spin [10] have been shown to admit an analytical solution for such a problem. However, (2.15) may be approximated by expressions that are easier to handle and to see this we anticipate a result to be discussed in the following section, namely that the Markovian limit of the GS corresponds to the approximation $\phi(z) \approx \phi(0)$ and $\Pi(z) \approx \Pi(0)$. This immediately suggests the possibility of working with simpler approximate expressions for the GS which take limited memory effects into account by the procedure of retaining a finite number of terms in the Taylor expansions of $\phi(z)$ and $\Pi(z)$. A similar method has previously been employed for generating non-Markovian corrections to the quantum Langevin equation [15] and also to quantum Fokker–Planck equations [16]. The simplest non-Markovian effect should then arise from retaining only linear or second-order terms and as an interesting application it is worthwhile mentioning the problem of an electron in the blackbody radiation field. In fact, it has been shown that the first- and second-order terms are the leading ones of such a problem since they respectively give rise to the renormalization of the electron mass and to the radiation reaction force in a quantum stochastic version of the Abraham–Lorentz equation of motion (see [15] and for a more complete treatment [17]).

As a first example of the application of the present formalism we shall analyse the GS of a harmonic oscillator, focusing upon its phonon number since it is the simplest observable which commutes with the Hamiltonian. The linear response functions of coordinate and momentum are well known for this system, particularly in the case that the heat bath consists of a set of harmonic oscillators [18, 19]. The present master equation formalism is evidently unsuitable for such calculations, the quantum Langevin equation or path integrals being, for instance, the correct tools to be employed. However, if other kinds of heat bath, i.e. fermionic, are to be considered, such formalisms become rather cumbersome, allowing only an analysis which usually does not go beyond the formal level. In contrast to such drawbacks, now we shall see that the method we propose can readily be applied to both kinds of reservoir. To this aim let us consider the linear interaction,

$$H_{SR} = \Lambda O x \tag{2.24}$$

where $x = a + a^\dagger$ denotes the adimensionalized coordinate of the harmonic oscillator (a and $a^\dagger$ are, respectively, the annihilation and creation operators), O denotes an unspecified observable of the heat bath and Λ is the parameter that measures the interaction strength. The Hamiltonian (2.24) leads to a collision operator $\phi(z)$ represented by a tridiagonal matrix which has been fully diagonalized in [14]. This allows a direct calculation of the GS (2.15) which for $A = a^\dagger a$ is given by an expression of the form

$$\tilde{\alpha}(z) = (1 - \mathrm{e}^{-\hbar\Omega/k_B T})^{-1} \frac{[P_+(z)\mathrm{e}^{-\hbar\Omega/k_B T} - P_-(z)]}{z + \mathrm{i}[V_+(z) - V_-(z)]} \tag{2.25}$$

where $\mathrm{e}^{-\hbar\Omega/k_B T}$ is the usual Boltzmann's factor of the harmonic oscillator, $\mathrm{i}[V_+(z) - V_-(z)]$ denotes the first non-vanishing eigenvalue of $\phi(z)$ and

$$P_\pm(z) = [Q_\pm(z) - Q_\pm(+\mathrm{i}0)]/z \tag{2.26}$$

where the functions $Q_\pm(z)$ define the matrix elements of $\varphi(z)$. Next we shall specialize the above functions for both kinds of reservoir. For the boson heat bath we have, as usual,

an observable of the form $O = \sum_\alpha f_\alpha x_\alpha$ in equation (2.24), f_α being a parameter that measures the relative contribution to the interaction of the harmonic oscillator of frequency ω_α and adimensionalized coordinate $x_\alpha = a_\alpha^\dagger + a_\alpha$. On the other hand, for the fermion reservoir we assume a number-conserving operator $O = \sum_\alpha \sum_\mu b_\mu^\dagger b_\alpha$, where $b_\mu^\dagger(b_\alpha)$ is the fermion creation (annihilation) operator of the single-particle state $|\mu\rangle(|\alpha\rangle)$ [14, 16]. Thus, denoting by $Q_\pm^{(b)}(z)$ and $Q_\pm^{(f)}(z)$ the corresponding boson and fermion functions $Q_\pm(z)$ of (2.26) we have

$$Q_\pm^{(b)}(z) = -i(\Lambda/\hbar)^2 \sum_\alpha f_\alpha^2 \left\{ n_\alpha \left[\frac{[i/\hbar]}{\mp\omega_\alpha - \Omega - z} + \frac{[-i/\hbar]}{\pm\omega_\alpha + \Omega - z} \right] + (n_\alpha + 1) \left[\frac{[i/\hbar]}{\pm\omega_\alpha - \Omega - z} + \frac{[-i/\hbar]}{\mp\omega_\alpha + \Omega - z} \right] \right\} \tag{2.27a}$$

$$Q_\pm^{(f)}(z) = -i(\Lambda/\hbar)^2 \sum_\alpha \sum_\mu \begin{Bmatrix} \rho_\mu(1-\rho_\alpha) \\ \rho_\alpha(1-\rho_\mu) \end{Bmatrix} \left[\frac{[i/\hbar]}{\omega_{\alpha\mu} - \Omega - z} + \frac{[-i/\hbar]}{\Omega - \omega_{\alpha\mu} - z} \right] \tag{2.27b}$$

where $n_\alpha = (\exp(\hbar\omega_\alpha/k_B T) - 1)^{-1}$ in (2.27*a*) denotes the mean number of phonons of frequency ω_α, $\rho_\gamma = \{1 + \exp[(\epsilon_\gamma - \epsilon_F)/k_B T]\}^{-1}$ in (2.27*b*) denotes the Fermi occupation number for a state $|\gamma\rangle$ of energy ϵ_γ, $\omega_{\alpha\mu}$ is the difference $(\epsilon_\alpha - \epsilon_\mu)/\hbar$ and the factors in square brackets $[\pm i/\hbar]$ are to be replaced by unity in the corresponding expressions for $V_\pm(z)$. Now, turning to the continuum limit, the summations in (2.27) become integrals according to

$$\sum_\alpha f_\alpha^2 \to \int_0^\infty f(\omega)\, d\omega \tag{2.28a}$$

$$\sum_\mu \to (L/2\pi)^3 \int d^3 k_\mu \tag{2.28b}$$

where L^3 denotes the volume of the fermion reservoir and the summation over α in (2.27*b*) can be trivially performed taking into account momentum conservation, i.e. $k_\alpha = k_\mu + q$, $\hbar q$ being the phonon momentum. Thus, the functions which define the z-dependence of the GS (2.25) turn in the upper half-plane into Cauchy integrals, namely

$$Q_+(z)\exp(-\hbar\Omega/k_B T) - Q_-(z) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{d\omega}{(\omega - z)} F(\omega) \tag{2.29}$$

$$V_+(z) - V_-(z) = \frac{1}{2\pi i} \int_{-\infty}^{+\infty} \frac{d\omega}{(\omega - z)} \nu(\omega) \tag{2.30}$$

with

$$F(\omega) = R(\omega)\frac{n(\Omega + \omega)}{n(\omega)} - R(-\omega)\frac{n(\Omega - \omega)}{n(-\omega)} \tag{2.31}$$

$$\nu(\omega) = \frac{\hbar}{i}[R(\omega) + R(-\omega)] \tag{2.32}$$

$$n(\omega) = [\exp(\hbar\omega/k_B T) - 1]^{-1} \tag{2.33}$$

where the function $R(\omega)$ depends on the heat bath according to

$$R^{(b)}(\omega) = \frac{2\pi i}{\hbar} \left(\frac{\Lambda}{\hbar} \right)^2 f(\Omega + \omega) \tag{2.34a}$$

$$R^{(f)}(\omega) = \frac{i}{2\pi}\left(\frac{L}{\hbar}\right)^3\left(\frac{\Lambda}{\hbar}\right)^2\left(\frac{2m^3\epsilon_F}{3}\right)^{1/2}\frac{k_BT}{\hbar\Omega}\ln\left[\frac{1+\exp\left\{\frac{\epsilon_F}{k_BT}\frac{1}{3\Omega^2}[3\Omega^2-(\omega+\Omega_-)^2]\right\}}{1+\exp\left\{\frac{\epsilon_F}{k_BT}\frac{1}{3\Omega^2}[3\Omega^2-(\omega+\Omega_+)^2]\right\}}\right] \tag{2.34b}$$

m being the fermion mass and $\Omega_\pm$ the shifted frequency $\Omega(1 \pm 3\hbar\Omega/4\epsilon_F)$. For the boson reservoir we have assumed that $f(\omega)$, which characterizes its continuum limit, is an odd-parity function, analytic along the whole real axis, e.g. the Drude [19] or Ullersma [18] model,

$$f(\omega) \sim \frac{\omega}{\omega^2+\eta^2}. \tag{2.35}$$

On the other hand, we have assumed a highly degenerate Fermi gas so that the details of the calculation leading to (2.34*b*) are the same as those discussed in [16].

Finally, the singularity spectrum of the GS can easily be extracted by focusing upon (cf equation (2.19)) the singularities of the kernels (2.31) and (2.32). Thus one finds the following poles.

(i) The infinite set of poles $\pm\Omega - in2\pi k_BT/\hbar$ $(n = 1, 2, \ldots)$ arise from the factors $n(\Omega \pm \omega)$ in (2.31), giving rise to time-decaying oscillations on the well known [18, 19] thermal time scale $\hbar/k_BT$.

(ii) The poles of $R(\omega)$ should not be singular points of $\tilde{\alpha}(z)$ since they appear equally in both the numerator and denominator of expression (2.25); this is the case for the poles $\Omega \pm i\eta$ of (2.34*a*)–(2.35). On the other hand, the infinite set of branch-cuts issuing from the singularities of (2.34*b*), which has been studied in [16], form part of the singularity spectrum of the fermion GS which therefore is expected to exhibit a highly complex non-exponential time decay due to the low temperatures.

(iii) Poles arise from the zeros of the denominator $z + i[V_+(z) - V_-(z)]$ in (2.25). At this point it is interesting to consider a low-frequency expression $\tilde{\alpha}^{(lf)}(z)$ for the GS, valid for $|z|$ much less than any characteristic frequency of the singularities discussed above in (i) and (ii). Within such a range, the functions $V_\pm(z)$ and $P_\pm(z)$ should be well approximated by the values at $z = 0$, then taking into account (2.22) it is easy to obtain

$$\tilde{\alpha}^{(lf)}(z) = \frac{\hbar R(0)}{k_BT}\frac{e^{\hbar\Omega/k_BT}[n(\Omega)]^2}{z+\hbar R(0)}. \tag{2.36}$$

From the above expression and (2.34) we see that for a small enough coupling parameter Λ, the imaginary Markovian [16] pole $z = -\hbar R(0)$ will be the most important singularity of the GS. In the case of stronger couplings (but still within a weak-coupling approximation) the location of this pole can be corrected as in [16] by considering several terms of the Taylor expansion of $V_+(z) - V_-(z)$ (cf equations (2.21)–(2.23)) which therefore amounts to including limited memory effects.

Now we shall briefly consider as a second example of the application of the theory the case of a particular multi-level system consisting of a spin j in a static magnetic field and weakly coupled to a phonon reservoir. The non-Markovian dynamics of this system has recently been studied [10, 11] showing that the collision operator $\phi(z)$ is completely defined through a pair of complex functions $W_\pm(z)$ which were investigated both in frequency and time domains, but unfortunately an exact diagonalization of $\phi(z)$ was only carried out for limited cases, namely $j = \frac{1}{2}, 1$ or in a high-temperature limit [10]. Nevertheless, we shall see that important information can be obtained from this example. In fact, taking as the observable A the spin component along the static magnetic field, it is not difficult to show that $\varphi(z)$ follows simply from $\phi(z)$ through the replacement $W_\pm(z) \to \mp B_\pm(z)$, where the

functions $B_{\pm}(z)$ are given in equation (3.8) of [11]. The point we want to make here is that such functions have been shown to be intimately related to the system–reservoir correlations [11], a feature that will be better understood in the following sections.

3. Time-dependent GS and comparison with the Kubo formula

Now we shall display the non-Markovian character of our GS by focusing upon its time dependence. To this aim let us first consider the free system evolution (i.e. without a driving force) that is given by setting in equation (2.9) $\lambda(t') = 0$. Under such conditions the Laplace transformed mean value of A is

$$\langle \tilde{A}(z) \rangle = \mathrm{i}\,\mathrm{Tr}_S\{A[z + \phi(z)]^{-1}\rho_S(t=0)\}. \tag{3.1}$$

On the other hand, from the Heisenberg picture one has

$$\langle \tilde{A}(z) \rangle = \mathrm{Tr}_S[\langle \tilde{A}(z) \rangle_R \rho_S(t=0)]. \tag{3.2}$$

Thus the above equations and the arbitrariness of $\rho_S(t=0)$ imply

$$P_0\langle \tilde{A}(z) \rangle_R = \mathrm{i}A[z + \phi(z)]^{-1} \tag{3.3}$$

where P_0 denotes projection onto the diagonal part. The replacement of this result in (2.15) finally yields the time representation of the GS:

$$\alpha(\tau) = \int_0^{\tau} \mathrm{d}\tau'\ \mathrm{Tr}_S\left[\langle A(\tau - \tau') \rangle_R P(\tau')\rho_S^{\mathrm{eq}}\right] \tag{3.4}$$

where $P(\tau')$ denotes the operator with Laplace transform $\mathrm{i}\Pi(z)$. Note that since $P(\tau')\rho_S^{\mathrm{eq}}$ is a diagonal matrix, only the diagonal component of $\langle A(\tau - \tau') \rangle_R$ will take part in the calculation. Now it is easy to identify the Markovian limit; it arises when $P(\tau')$ has a very short lifetime in comparison with the characteristic times of $\langle A(\tau) \rangle_R$. In such a case it has been shown [10, 11] that the collision operator is well approximated by its value at the origin and the same must occur for $\Pi(z)$, yielding

$$\alpha_M(\tau) = \mathrm{i}\,\mathrm{Tr}_S[\langle A(\tau) \rangle_R^{\mathrm{M}} \Pi(0)\rho_S^{\mathrm{eq}}] \tag{3.5}$$

where the superscript 'M' indicates the Markovian approximation of $\langle A(\tau) \rangle_R$, i.e. obtained from replacing $\phi(z)$ by $\phi(0)$. From a more general point of view, $\alpha_M(\tau)$ may be regarded as an asymptotic approximation valid for not too short (or large) times, whose frequency-dependent counterpart given by

$$\tilde{\alpha}^{(\mathrm{lf})}(z) = -\mathrm{Tr}_S\left\{A[z + \phi(0)]^{-1}\Pi(0)\rho_S^{\mathrm{eq}}\right\} \tag{3.6}$$

is a low-frequency approximation such as that of equation (2.36). The expression (3.5) may be written in a more convenient form via the following equality:

$$\Pi(0)\rho_S^{\mathrm{eq}} = -\frac{1}{k_B T}\phi(0)\left[A\rho_S^{\mathrm{eq}}\right] \tag{3.7}$$

where the brackets on the right-hand side indicate that the matrix $\phi(0)$ is acting upon the column vector corresponding to the product of the diagonal matrices A and ρ_S^{eq}. The proof of the above equation requires an explicit calculation in the thermodynamic limit that is displayed with some detail in the appendix. In addition, from (3.3) one has

$$P_0\,\langle \dot{A}(\tau) \rangle_R = \int_0^{\tau} \mathrm{d}\tau'\ P_0\langle A(\tau - \tau') \rangle_R \chi(\tau') \tag{3.8}$$

where $\chi(\tau')$ has as its Laplace transform $i\phi(z)$. Finally from (3.5), (3.7) and (3.8) in the Markovian limit one gets

$$\alpha_M(\tau) = -\frac{1}{k_B T} \mathrm{Tr}_S \left[\langle \dot{A}(\tau) \rangle_R^M A \rho_S^{eq} \right]. \tag{3.9}$$

This expression makes room for a direct comparison with the Kubo exact formula for linear response, which can be written as [20]

$$\alpha(\tau) = -\int_0^{1/k_B T} d\beta \, \langle e^{\beta H} A e^{-\beta H} \dot{A}(\tau) \rangle_{eq} \tag{3.10}$$

where H is, in our problem, $H_S + H_R + H_{SR}$ and $\langle\ \rangle_{eq}$ indicates the average over the equilibrium distribution of the whole system. Therefore from (3.9) and (3.10) it becomes clear that the Markovian approximation neglects system–reservoir correlations of equilibrium. The interesting point, however, is that our non-Markovian expression (2.15) does indeed take into account such correlations. To see this, let us consider the Laplace transform of Kubo formula (3.10) with vanishing correlations:

$$\tilde{\alpha}_{n-c}(z) = -\frac{1}{k_B T} \mathrm{Tr}_S \left[\langle \tilde{\dot{A}}(z) \rangle_R A \rho_S^{eq} \right]. \tag{3.11}$$

On the other hand, Laplace transforming (3.4) and (3.8) one gets

$$\tilde{\alpha}(z) = \mathrm{Tr}_S \left[\langle \tilde{\dot{A}}(z) \rangle_R \phi^{-1}(z) \Pi(z) \rho_S^{eq} \right] \tag{3.12}$$

which differs from (3.11) except in the Markovian limit (cf (3.7)); we postpone a further comparison to the final section 5.

4. Comparison to the usual Markovian calculation

In this section we shall prove that our Markovian GS leads to the same time correlation function that arises from the usual method. The Markovian master equation is, in our notation,

$$\dot{\rho}_S(t) = i\phi(0)\rho_S(t) \tag{4.1}$$

and its solution is given by (2.9) with $\lambda(t') = 0$ and $\phi(z) = \phi(0)$. Then calling $\rho_{s/s'}(t)$ the conditional probability of finding the system in the state s at time t provided it has been in the state s' at time zero, we have,

$$\rho_{s/s'}(t) = \frac{i}{2\pi} \int_{-\infty+i0}^{+\infty+i0} dz \, e^{-izt} [z + \phi(0)]^{-1}_{ss'} \tag{4.2}$$

where the subindices ss' on the right-hand side indicate the corresponding matrix element. Now, the joint probability of finding the system at equilibrium in the state s at time t and in the state s' at time zero, $\rho_{ss'}(t, 0)$, is

$$\rho_{ss'}(t, 0) = \rho_{s/s'}(t) \rho_{s'} \tag{4.3}$$

where $\rho_{s'}$ denotes the $s's'$ element of ρ_S^{eq}. At this point it is important to recall that the canonical distribution of the system becomes non-stationary outside the Markovian limit. So, if we were interested in calculating the two-time equilibrium distribution through the formula (4.3) in a general case, it would be necessary to construct a more elaborated non-Markovian theory which takes into account initial correlations (see for instance [19]) so that the canonical distribution of the system remains stationary.

Now let us consider the time correlation function $C(t)$ of an observable A, which is defined as the average of $[A_0(t)A_0(0) + A_0(0)A_0(t)]/2$ in the equilibrium state, being $A_0 = A - \langle A\rangle_{eq}$. $C(t)$ must be an even function so, taking into account (4.3) and (4.2), we can find its Fourier transform

$$\bar{C}(\omega) = 2i\,\mathrm{Tr}_S\left\{A[-\omega + \phi(0)]^{-1}[\omega + \phi(0)]^{-1}\phi(0)\left[A\rho_S^{eq}\right]\right\}. \tag{4.4}$$

On the other hand, a straightforward application of the fluctuation–dissipation theorem [1, 6] to our GS (2.15) yields

$$\bar{C}_{nM}(\omega) = \hbar\coth\left(\frac{\hbar\omega}{2k_BT}\right)\mathrm{Im}[\tilde{\alpha}(\omega)] \tag{4.5}$$

where $\bar{C}_{nM}(\omega)$ denotes the Fourier transform of the non-Markovian time correlation function. The Markovian limit of (4.5) is obtained from (3.5) and (3.7), namely

$$\bar{C}_M(\omega) = i\frac{\hbar\omega}{k_BT}\coth\left(\frac{\hbar\omega}{2k_BT}\right)\mathrm{Tr}_S\left\{A[-\omega + \phi(0)]^{-1}[\omega + \phi(0)]^{-1}\phi(0)\left[A\rho_S^{eq}\right]\right\}. \tag{4.6}$$

Finally a comparison of (4.4) and (4.6) clearly shows that in the usual method the thermal decay time $\hbar/k_BT$ is neglected.

5. Concluding remarks

The first point we wish to make in this section is the fundamental connection existing between memory and correlations. In fact, we have shown that the inclusion of the former is equivalent to considering the equilibrium correlations between the system and its environment to be non-negligible. In this respect it is worthwhile recalling that the Markovian approximation has been shown to be valid in extremely weak-coupling situations [11, 16], which is therefore consistent with negligible correlations. Another insight into this matter follows from equations (3.11) and (3.12), which clearly exhibit the link between memory and correlations. In fact, we observe that the uncorrelated expression (3.11) does not show the additional z dependence of (3.12) which, in turn, implies additional memory. Finally it is worth mentioning previous studies on the dynamics of correlated initial states by means of functional integral techniques [19] which, however, do not seem to reflect the connection with memory we are stressing here.

On the other hand, regarding our result (2.15)–(2.16) which depends on the two matrices $\phi(z)$ and $\varphi(z)$, the former being a reduced version of the well known collision operator, one could give the latter the name of 'correlation operator'. In fact, it has been shown that the uncorrelated Markovian GS does indeed depend only upon the collision operator (cf (3.7)) and also, for the special case of a spin j, the close connection existing between $\varphi(z)$ and the operator governing spin-bath correlations has already been mentioned [11].

Finally we remark that our present treatment which, for simplicity, has been restricted to the case of a single observable A can easily be generalized if several commuting observables A_i are considered simultaneously (e.g. the set of spins of an Ising model). In fact, a straightforward generalization of (2.15) yields

$$\tilde{\alpha}_{jk}(z) = -\mathrm{Tr}_S\left\{A_j[z + \phi(z)]^{-1}\Pi_k(z)\rho_S^{eq}\right\} \tag{5.1}$$

where $\tilde{\alpha}_{jk}(z)$ denotes, briefly speaking, the susceptibility of A_j due to the force acting upon A_k and $\Pi_k(z)$ is obtained by means of the replacement $A \to A_k$ in $\Omega(z)$ (equation (2.6)).

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Appendix A. Derivation of equation (3.7)

First, from (2.16) and (2.17) a straightforward calculation yields the following ss element of the diagonal matrix $\Pi(0)\rho_S^{eq}$:

$$\frac{2\pi}{i}\sum_{s'}\sum_{R'R}|\Lambda_{sR;s'R'}|^2\delta'(\omega_s-\omega_{s'}+\omega_R-\omega_{R'})[f(\omega_s)-f(\omega_{s'})][\rho_s\rho_R-\rho_{s'}\rho_{R'}] \tag{A.1}$$

where ρ_s denotes an ss element of ρ_S^{eq}. The next step consists of writing the above expression in the thermodynamic limit and, to this aim, it is convenient to separate the total energy of the heat bath from the remaining quantum numbers, i.e. to replace R by ω_R, R_r in the summation over the reservoir labels R and R' in (A.1), then the passage to the thermodynamic limit can be done according to

$$\sum_{\omega_R}\to\int_0^\infty g(\omega_R)\,d\omega_R \tag{A.2}$$

with such prescriptions (A.1) becoming (note that the contribution arising from derivatives of g and $|\Lambda|^2$ identically vanishes)

$$\frac{2\pi\hbar}{ik_BTZ_S\Xi_R}\sum_{N_R}\sum_{s'}\sum_{R_r'R_r}\int_0^\infty d\omega_R|\Lambda_{s,\omega_R,R_r;s',\omega_R+\omega_s-\omega_{s'},R_r'}|^2g(\omega_R)g(\omega_s-\omega_{s'}+\omega_R)$$
$$\times[f(\omega_s)-f(\omega_{s'})]\exp\{[\mu N_R-\hbar(\omega_s+\omega_R)]/k_BT\} \tag{A.3}$$

where Z_S denotes the canonical partition function of the system and Ξ_R the macrocanonical partition function of the heat bath. In (A.3) we have assumed a system with a fixed number of particles and a reservoir for which such a number, N_R, is conserved, e.g. the Fermi gas of section 2. However, this is often not the case as, for example, in a phonon reservoir which, nevertheless, can be considered as a special case of (A.3) with a vanishing chemical potential μ.

On the other hand, we have seen that the matrix elements of the collision operator $\phi(z)$ arise from (2.17) by means of the replacement $f(\,)-f(\,)\to 1$; using this it is easy to show that $-[k_BT]^{-1}\phi(0)[A\rho_S^{eq}]$ is a diagonal matrix whose ss element is

$$\frac{2\pi\hbar}{ik_BT}\sum_{s'}\sum_{R'R}|\Lambda_{sR;s'R'}|^2\,\delta(\omega_s-\omega_{s'}+\omega_R-\omega_{R'})[f(\omega_s)\rho_s\rho_R-f(\omega_{s'})\rho_{s'}\rho_{R'}]. \tag{A.4}$$

Finally, taking the thermodynamic limit of the above expression one gets (A.3), which proves equation (3.7).

For completeness we shall also provide a brief derivation that ρ_S^{eq} is indeed canonical. To this aim we first note that equation (2.12) is equivalent to

$$\phi(0)\rho_S^{eq}=0. \tag{A.5}$$

The ss component of the left-hand side of this matrix equation is obtained by means of the replacement $\hbar f(\,)/k_B T \to -1$ in equation (A.4). Now seeking the solution to order zero in Λ we find the condition

$$\rho_s \rho_R = \rho_{s'} \rho_{R'} \tag{A.6}$$

to be fulfilled for all the states connected by a non-vanishing $\Lambda_{sR;s'R'}$. Finally, taking into account the equilibrium distribution of ρ_R and the δ factor in (A.4), the canonical nature of ρ_S^{eq} is demonstrated.

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