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Coherent-state path integral approach to the damped harmonic oscillator

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Abstract

The time evolution of a system of bilinearly coupled bosonic modes is investigated using the real-time path integral technique in the coherent-state representation. In order to get the stationary trajectories, the corresponding Lagrangian function is diagonalized and then the path integrals are evaluated by means of the stationary-phase method. The present treatment is applied to a dissipative harmonic oscillator within the Caldeira–Leggett model. The time evolution of the reduced density matrix in the basis of coherent states is given in simple analytic form for weak system–bath coupling, i.e. the so-called rotating-wave terms can be evaluated exactly but the non-rotating-wave terms only in a perturbative manner. The validity range of the rotating-wave approximation is discussed from the viewpoint of spectral equations. In contrast to many perturbative approaches, the bath degrees of freedom are treated explicitly. In addition, it is shown that systems without initial system–bath correlations can exhibit initial jumps in the population dynamics even for rather weak dissipation. Only with initial correlations can the classical trajectories for the system coordinate be recovered.

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1. Introduction

The problem of describing damping in open quantum systems has been discussed for a long time in the literature (see, for example, [1]). Among the first treatments of such problems were the quantum Langevin equation and quantum master equations which are mainly concerned with weakly damped systems in which the time of relaxation in the system is larger than the time scale of the undamped motion and larger than the relaxation time of an environment. A more general method without the restriction to weak damping is the Feynman–Vernon path integral approach [2–4] which uses the functional integral technique in a phase-space representation. This treatment allows one to investigate the quantum dynamics of dissipative

systems with arbitrarily strong friction and has been successfully applied (see, for example, [5–9]).

In this paper, we present a method for the investigation of the dynamics of bilinearly coupled bosonic modes based on the real-time path integrals in the coherent-state representation [3]. This representation has only been used as a numerical tool in a few investigations studying the propagation of wave packets [10–12] and equilibrium statistical mechanics properties [13]. In the present work, the coherent-state representation will be used in a fully analytical study. We applied this technique to the time evolution of a damped harmonic oscillator. Dissipation will be described with the well-known Caldeira–Leggett Hamiltonian [5], i.e. the full system is split into a relevant part and a thermal bath consisting of harmonic oscillators characterized by the spectral density. The system–bath coupling is assumed to be bilinear. As far as the functional integral is of Gaussian type, the stationary-phase method leads to exact results. This is the case for one or several harmonic oscillators coupled to a thermal bath consisting of further harmonic oscillators. Our goal is to diagonalize the Lagrangian allowing us to get stationary trajectories. In this way, we obtain the time evolution of the reduced density matrix while similar procedures of diagonalizing the Hamiltonian used previously [14, 15] allow one to get only the time evolution of the system operators.

The problem of a damped harmonic oscillator was already extensively discussed in the literature (see [17, 14, 16, 1] and references therein). Here we study the time evolution in the coherent-state representation to get expressions for weak system–bath coupling which are easier to evaluate than those in the previous literature and which can easily be extended to a set of coupled harmonic oscillators. Since we use the Hamiltonian in the creation/annihilation-operator representation the rotating-wave approximation (RWA) can be performed immediately by neglecting the non-rotating-wave terms. This way we get quite simple expressions for the evolution of the reduced density matrix in the basis of coherent states for uncorrelated and correlated initial system–bath conditions, which are used for the evaluation of the expectation values of the system, in particular the population dynamics for different types of initial states of the system. One important feature of the present approach is the explicit treatment of the bath modes. As is shown below, this has effects on the population dynamics already for weak system–bath interaction. The present study is quite important since the problem of coupled harmonic oscillators serves as a testbed for many dissipation theories. Such a model system is, for example, used to describe electron transfer reactions [17].

The paper is organized as follows. In section 2, we present the general expressions for the path integrals in coherent-state representation for a system of bosonic particles. In a next step, the model Hamiltonian for a damped harmonic oscillator is recapitulated in section 3. In order to evaluate the functional integral, the Lagrangian is diagonalized within and without the RWA. This procedure is presented in section 4. Together with the correct expression for the functional integral in the coherent-state representation this builds the foundation of our investigation. Next, the spectral equations for the system after diagonalization are obtained together with their reduction to the RWA. The validity range of the RWA is discussed in section 5 by using the spectral equations. The results are applied to calculate the dynamics of the reduced density matrix for non-equilibrium factorized and for correlated initial conditions. The time evolution of the energy level population and the occupation number for a damped harmonic oscillator are determined in section 6. Besides, we show the influence of the correlated initial system–bath conditions on the evolution of the mean value of the system coordinate. In the last section, a short conclusion and further possible extensions and applications of the present method are given. Atomic units are used throughout the paper and the Boltzmann constant is set to unity.

2. Path integral formalism in coherent-state representation

In this section, general expressions for the matrix elements of the time-evolution operator $\mathrm{e}^{-\mathrm{i}Ht}$ with a dissipative Hamiltonian H are obtained using the path integral technique. In contrast to the well-known Herman–Kluk method [18, 19] where coherent states are used together with a phase-space representation, here we only use the basis of coherent states for the calculation of time-evolution operators.

Following the treatment developed, for example, in [20], we obtain the functional integral defined in the coherent-state representation for an arbitrary bosonic Hamiltonian. The coherent states $|z\rangle$ are defined as usual for a harmonic oscillator [3]

$$|z\rangle = \sum_n \frac{z^n}{\sqrt{n!}}|n\rangle\,\mathrm{e}^{-|z|^2} \qquad \hat{a}|z\rangle = z|z\rangle \tag{1}$$

where $|n\rangle$ is the nth eigenstate of a harmonic oscillator, $\hat{a}$ and z are the annihilation operator and its eigenvalue, respectively. In addition, we will use the completeness relation for the coherent states

$$\int \frac{\mathrm{d}z^*\,\mathrm{d}z}{\pi}|z\rangle\langle z| = 1. \tag{2}$$

The aim is to calculate the matrix element

$$\langle\{z_\omega\}|\,\mathrm{e}^{-\mathrm{i}Ht}|\{z'_\omega\}\rangle \tag{3}$$

where the state $|\{z_\omega\}\rangle$ denotes a product of coherent states $|z_\omega\rangle$ for all bath modes with corresponding frequencies ω. The Hamiltonian H is assumed to be normal ordered. In order to evaluate this matrix element one may, as usual, divide the time interval into N infinitesimal time slices of length ε

$$\mathrm{e}^{-\mathrm{i}Ht} = \underbrace{\mathrm{e}^{-\mathrm{i}H\varepsilon}\,\mathrm{e}^{-\mathrm{i}H\varepsilon}\cdots\mathrm{e}^{-\mathrm{i}H\varepsilon}}_{N} \tag{4}$$

and insert the unity operator (2) at each dividing point. Together with the expression for the product of two coherent states

$$\langle z|z'\rangle = \exp\left[-\tfrac{1}{2}|z|^2 - \tfrac{1}{2}|z'|^2 + z^*z'\right] \tag{5}$$

it leads to the following form for the matrix element of the evolution operator:

$$\begin{aligned}\langle\{z_\omega\}|\,\mathrm{e}^{-\mathrm{i}Ht}|\{z'_\omega\}\rangle = \prod_\omega\prod_{n=1}^{N-1}\left[\int\frac{\mathrm{d}z^*_{\omega n}\,\mathrm{d}z_{\omega n}}{\pi}\right]\exp\Bigg[-\sum_\omega\Bigg(\frac{1}{2}|z_{\omega 0}|^2+\frac{1}{2}|z_{\omega N}|^2 \\ +\sum_{n=1}^{N-1}|z_{\omega n}|^2-\sum_{n=1}^{N}z^*_{\omega n}\,z_{\omega n-1}\Bigg)-\mathrm{i}\sum_{n=1}^{N}\varepsilon H(\{z^*_{\omega n}\},\{z_{\omega n-1}\})\Bigg]\end{aligned} \tag{6}$$

with the boundary conditions

$$z_{\omega 0} = z'_\omega \qquad z^*_{\omega N} = z^*_\omega. \tag{7}$$

Using the identity

$$\sum_{n=1}^{N}z^*_n\,z_{n-1}-\sum_{n=1}^{N-1}|z_n|^2 = \frac{1}{2}(z^*_N\,z_{N-1}+z^*_1\,z_0)+\frac{\varepsilon}{2}\sum_{n=1}^{N-1}\left(z_n\frac{z^*_{n+1}-z^*_n}{\varepsilon}-z^*_n\frac{z_n-z_{n-1}}{\varepsilon}\right) \tag{8}$$

and performing the continuum limit in time ($\varepsilon \to 0, N \to \infty$) one can get the following functional integral expression for the matrix element:

$$\langle\{z_\omega\}|\,\mathrm{e}^{-\mathrm{i}Ht}|\{z'_\omega\}\rangle = \prod_\tau \prod_\omega \int \frac{\mathrm{d}z^*_\omega(\tau)\,\mathrm{d}z_\omega(\tau)}{\pi} \times \exp\left[-\frac{1}{2}\sum_\omega(|z_\omega|^2 + |z'_\omega|^2) + S(\{z_\omega(\tau)\},\{z^*_\omega(\tau)\})\right]. \tag{9}$$

Here S denotes the action depending on the trajectories $z_\omega(\tau)$ and $z^*_\omega(\tau)$

$$S(\{z_\omega(\tau)\},\{z^*_\omega(\tau)\}) = \frac{1}{2}\sum_\omega(z^*_\omega(t)z_\omega(t) + z^*_\omega(0)z_\omega(0)) + \int_0^t\left[\frac{1}{2}\sum_\omega(z_\omega(\tau)\dot{z}^*_\omega(\tau) - z^*_\omega(\tau)\dot{z}_\omega(\tau)) - \mathrm{i}H(\{z^*_\omega(\tau)\},\{z_\omega(\tau)\})\right]\mathrm{d}\tau \tag{10}$$

which must be evaluated using the boundary conditions

$$z_\omega(0) = z'_\omega \qquad z^*_\omega(t) = z^*_\omega. \tag{11}$$

The form of the action (10) differs from the action sometimes used in the literature (for example in [21]) by the first two additional terms. Only with these terms are the correct results obtained if one employs the usual methods for calculating functional integrals [20]. We note in passing that the endpoints z_ω and z^*_ω are conjugates of each other but the trajectories $z_\omega(\tau)$ and $z^*_\omega(\tau)$ are not.

Simplifying the first and the second terms under the integral in equation (10), one can write

$$S(\{z_\omega(\tau)\},\{z^*_\omega(\tau)\}) = \sum_\omega z^*_\omega(t)z_\omega(t) - \int_0^t \mathcal{L}(\{z^*_\omega(\tau)\},\{z_\omega(\tau)\})\,\mathrm{d}\tau \tag{12}$$

where

$$\mathcal{L}(\{z^*_\omega(\tau)\},\{z_\omega(\tau)\}) = \sum_\omega z^*_\omega(\tau)\dot{z}_\omega(\tau) + \mathrm{i}H(\{z^*_\omega(\tau)\},\{z_\omega(\tau)\}). \tag{13}$$

As described in appendix A for the backward direction of time, i.e. for the matrix element of the Hermitian conjugated evolution operator $\mathrm{e}^{\mathrm{i}Ht}$, one has to use the action $\tilde{S}$ of the form (A.6). So the actions for forward and backward propagations, S and $\tilde{S}$, differ by the first terms depending on the end points of the trajectories and by the sign of the functional parts but are determined by one and the same Lagrangian function $\mathcal{L}$ (equation (13)).

3. Dissipative model Hamiltonian

Next we will consider the Hamiltonian for one damped harmonic oscillator coupled to a heat bath consisting of a large number of independent harmonic modes. The Hamiltonian will be of the so-called Caldeira–Leggett type [5, 6]

$$H = H_s + H_b + H_{sb} + H_r \tag{14}$$

where

$$H_s = \omega_0 a^+ a \qquad H_b = \sum_\omega \omega a^+_\omega a_\omega \tag{15}$$

represent the relevant harmonic oscillator with frequency ω_0 and the thermal bath of oscillators with frequencies ω.

The coupling between the system oscillator and the bath is considered in the coordinate–coordinate coupling model

$$H_{sb} = q \sum_{\omega} c_\omega x_\omega \tag{16}$$

where q is the coordinate of the system oscillator, x_ω are the coordinates of the bath modes and c_ω denotes the constants of interaction between the relevant oscillator and the oscillators of the bath at frequencies ω. The interaction Hamiltonian H_{sb} can be rewritten using creation and annihilation operators as

$$H_{sb} = \sum_{\omega} k_\omega (a^+ + a)\left(a_\omega^+ + a_\omega\right) \tag{17}$$

where the k_ω are connected to the c_ω by

$$c_\omega = 2\sqrt{M\omega_0\, m_\omega\, \omega} k_\omega. \tag{18}$$

M and m_ω denote the masses of the system oscillator and the oscillators in the bath, respectively.

In some of the following calculations, we will consider the RWA which consists of neglecting the terms proportional to $a^+ a_\omega^+$ and $a a_\omega$ in the interaction part of the Hamiltonian (17), i.e.

$$H_{sb}^{\mathrm{RWA}} = \sum_{\omega} k_\omega \left(a^+ a_\omega + a a_\omega^+\right). \tag{19}$$

The terms $a^+ a_\omega^+$ and $a a_\omega$ are associated with high frequencies $\omega_0 + \omega$ and are assumed to be very small. In section 5, we will discuss the criterion of validity for the RWA.

The system–bath coupling leads to the appearance of an additional contribution to the system potential. Without this additional renormalization term H_r, the system–bath coupling would lead to a dissipation-dependent shift which is undesirable. In order to cancel this contribution one introduces the renormalization term H_r

$$H_r = \sum_{\omega} \frac{k_\omega^2}{\omega} (a + a^+)^2. \tag{20}$$

In this way, the frequency of the system Hamiltonian stays almost independent of the dissipation strength.

4. Evaluation of the path integral by diagonalization of the Lagrangian

Next we will consider the Hamiltonian in the RWA (see equation (19)) and neglect the renormalization term which is very small for small dissipation strength. The corresponding Lagrangian function (13) has the form

$$\mathcal{L}(\{z_\omega(\tau)\}, \{z_\omega^*(\tau)\}, a(\tau), a^*(\tau)) = \sum_{\omega} \dot{z}_\omega(\tau) z_\omega^*(\tau) + \dot{a}(\tau) a^*(\tau)$$
$$+ \mathrm{i}\omega_0 a^*(\tau) a(\tau) + \mathrm{i} \sum_{\omega} \omega z_\omega(\tau) z_\omega^*(\tau) + \mathrm{i} \sum_{\omega} k_\omega (a(\tau) z_\omega^*(\tau) + a^*(\tau) z_\omega(\tau)) \tag{21}$$

together with the boundary conditions

$$z_\omega(0) = z_\omega' \qquad z_\omega^*(t) = z_\omega^* \tag{22}$$

$$a(0) = a' \qquad a^*(t) = a^*. \tag{23}$$

Here a (a^*) denotes the system trajectories and z_ω (z_ω^*) denotes the trajectories of the bath modes with frequency ω.

One can introduce new trajectories $A(\tau)$ and $A^*(\tau)$ using the following transformations:

$$a(\tau) = \sum_{\Omega} \alpha_{\Omega}\, A_{\Omega}(\tau) \tag{24}$$

$$a^*(\tau) = \sum_{\Omega} \alpha_{\Omega}\, A^*_{\Omega}(\tau) \tag{25}$$

$$z_{\omega}(\tau) = \sum_{\Omega} \beta_{\Omega\omega}\, A_{\Omega}(\tau) \tag{26}$$

$$z^*_{\omega}(\tau) = \sum_{\Omega} \beta_{\Omega\omega}\, A^*_{\Omega}(\tau). \tag{27}$$

The transformation for $a^*(\tau)\left(z^*_{\omega}(\tau)\right)$ must be the same as for $a(\tau)\left(z_{\omega}(\tau)\right)$ because of the symmetry of the Hamiltonian (Lagrangian) as a function of $a(\tau)$ and $a^*(\tau)$ $\left(z_{\omega}(\tau)\right.$ and $\left.z^*_{\omega}(\tau)\right)$. Substituting $a(\tau), a^*(\tau), z_{\omega}(\tau)$ and $z^*_{\omega}(\tau)$ as functions of $A(\tau)$ and $A^*(\tau)$ into the right-hand side of the Lagrangian (21) and requiring that after the transformation the Lagrangian has to have the form

$$\mathcal{L}(\{A_{\Omega}(\tau)\}, \{A^*_{\Omega}(\tau)\}) = \sum_{\Omega} \dot{A}_{\Omega}(\tau) A^*_{\Omega}(\tau) + \mathrm{i} \sum_{\Omega} \Omega A^*_{\Omega}(\tau) A_{\Omega}(\tau) \tag{28}$$

one can get the following orthogonality condition for the transformation matrix:

$$\alpha_{\Omega}\, \alpha_{\Omega'} + \sum_{\omega} \beta_{\Omega\omega}\, \beta_{\Omega'\omega} = \delta_{\Omega\Omega'} \tag{29}$$

together with the equations for the transformation matrix coefficients

$$\omega_0\, \alpha_{\Omega} + \sum_{\omega} k_{\omega}\, \beta_{\Omega\omega} = \Omega \alpha_{\Omega} \tag{30}$$

and

$$\omega \beta_{\Omega\omega} + k_{\omega}\, \alpha_{\Omega} = \Omega \beta_{\Omega\omega}. \tag{31}$$

Requiring the orthogonality of the transformation and employing equation (29) one can write the back transformation as

$$A_{\Omega}(\tau) = \alpha_{\Omega}\, a(\tau) + \sum_{\omega} \beta_{\Omega\omega}\, z_{\omega}(\tau) \tag{32}$$

$$A^*_{\Omega}(\tau) = \alpha_{\Omega}\, a^*(\tau) + \sum_{\omega} \beta_{\Omega\omega}\, z^*_{\omega}(\tau). \tag{33}$$

Then, requiring the orthogonality again for the forward transformation we can get

$$\sum_{\Omega} \alpha^2_{\Omega} = 1 \tag{34}$$

$$\sum_{\Omega} \alpha_{\Omega}\, \beta_{\Omega\omega} = 0 \tag{35}$$

$$\sum_{\Omega} \beta_{\Omega\omega}\, \beta_{\Omega\omega'} = \delta_{\omega\omega'}. \tag{36}$$

The Lagrangian (28) is now the Lagrangian of independent harmonic oscillators. Using the stationary-phase method (i.e. the condition $\delta \int_0^t \mathcal{L}\, \mathrm{d}\tau = 0$) one can get the trajectories which yield the main contribution to the functional integral of type (9)

$$A_\Omega(\tau) = A_\Omega(0)\,\mathrm{e}^{-\mathrm{i}\Omega\tau} \tag{37}$$

$$A^*_\Omega(\tau) = A^*_\Omega(0)\,\mathrm{e}^{\mathrm{i}\Omega\tau}. \tag{38}$$

Substituting this solution into the Lagrangian we see that $\mathcal{L} = 0$ on the stationary trajectories.

The last task is to find the constants $A_\Omega(0)$ and $A^*_\Omega(0)$. Using boundary conditions (22) and (23), back transformation (32) and (33) and solution (37) and (38) one can obtain

$$A_\Omega(0) = \alpha_\Omega\, a' + \sum_\omega \beta_{\Omega\omega}\, z'_\omega \tag{39}$$

$$A^*_\Omega(0) = \left(\alpha_\Omega\, a^* + \sum_\omega \beta_{\Omega\omega}\, z^*_\omega\right) \mathrm{e}^{-\mathrm{i}\Omega t}. \tag{40}$$

Substituting $a(\tau)$, $a^*(\tau)$, $z_\omega(\tau)$ and $z^*_\omega(\tau)$ as functions of $A_\Omega(\tau)$ and $A^*_\Omega(\tau)$ into the action S (see equation (10)), using equations (29, 37–40) and taking into account that $\mathcal{L} = 0$ we obtain for the matrix element of the evolution operator

$$\langle\{z_\omega\}|\langle a|\,\mathrm{e}^{-\mathrm{i}Ht}|a'\rangle|\{z'_\omega\}\rangle = \exp\left[-\frac{1}{2}|a|^2 - \frac{1}{2}|a'|^2 - \frac{1}{2}\sum_\omega |z_\omega|^2 - \frac{1}{2}\sum_\omega |z'_\omega|^2 \right.$$
$$\left. + \xi(t) a^* a' + \sum_\omega \eta_\omega(t)(a^* z'_\omega + a' z^*_\omega) + \sum_{\omega,\omega'} \mu_{\omega,\omega'}(t) z^*_\omega\, z'_{\omega'}\right] \tag{41}$$

where

$$\xi(t) = \sum_\Omega \alpha^2_\Omega\,\mathrm{e}^{-\mathrm{i}\Omega t} \tag{42}$$

$$\eta_\omega(t) = \sum_\Omega \alpha_\Omega\,\beta_{\Omega\omega}\,\mathrm{e}^{-\mathrm{i}\Omega t} \tag{43}$$

$$\mu_{\omega\omega'}(t) = \sum_\Omega \beta_{\Omega\omega}\,\beta_{\Omega\omega'}\,\mathrm{e}^{-\mathrm{i}\Omega t}. \tag{44}$$

As one can see now we have an exponent of Gaussian type in expression (41) which depends on the end points only. The more general case of diagonalizing the Lagrangian beyond the RWA is given in appendix B.

5. Spectral equation and reduction to the RWA

The procedure of diagonalizing the Lagrangian of the interacting harmonic oscillators presented above allows us to transform the Lagrangian into a Lagrangian of independent harmonic modes. In order to investigate the properties of such a system, we must know the distribution of frequencies of the new harmonic oscillators. Next we will derive the equations for the eigenfrequencies of the independent modes and then, with the help of those equations we will discuss the reduction from the non-RWA to the RWA results.

First, consider the system–bath interaction within the RWA. Having equations (30) and (31) for the transformation matrix let us express $\beta_{\Omega\omega}$ from equation (31) as

$$\beta_{\Omega\omega} = k_\omega \frac{\alpha_\Omega}{\Omega - \omega} \tag{45}$$

and substitute this expression into equation (30). The equation one obtains is the equation for the eigenfrequencies of the uncoupled modes

$$\Omega - \omega_0 = \sum_{\omega} \frac{k_\omega^2}{\Omega - \omega}. \tag{46}$$

The same procedure can be performed for the transformation matrix without the RWA. Expressing $\tilde{\alpha}_\Omega$, $\beta_{\Omega\omega}$ and $\tilde{\beta}_{\Omega\omega}$ as functions of α_Ω by using equations (B.10) and (B.11) and then substituting the obtained expressions into equation (B.8) with $\Omega = \Omega'$ we can obtain the spectral equation without the RWA

$$(\Omega + \omega_0)(\Omega - \omega_0) = 4\omega_0 \sum_{\omega} \frac{k_\omega^2\, \omega}{(\Omega - \omega)(\Omega + \omega)} \tag{47}$$

which has a quadratic dependence on the frequencies in comparison to equation (46) where the dependence is linear.

The reduction to the RWA was discussed in [14] by analysis of the time evolution of the creation and annihilation operators. Here we reduce to the RWA in terms of spectral equations and transformation matrices. Let us consider the case when the k_ω are small. Then the right-hand side of equation (47) becomes important only when Ω is close to ω_0 (for simplicity we consider only the solution of the spectral equation with positive real part). Hence we can substitute $\Omega + \omega_0$ by $2\omega_0$ on the left-hand side of equation (47). On other hand, the main contribution to the sum on the right-hand side comes from ω values close to Ω. Performing all replacements we get the spectral equation (46) within the RWA.

Now consider the coefficients $\tilde{\alpha}_\Omega$ which mix the trajectories a and a^*. From equation (B.10) and the assumption that k_ω is small, one gets

$$\tilde{\alpha}_\Omega \simeq \alpha_\Omega \frac{\omega_0 - \Omega}{2\omega_0}. \tag{48}$$

Since $\omega_0 - \Omega \ll \omega_0$, $\tilde{\alpha}_\Omega$ can be neglected in equations (B.10) and (B.11). The same analysis can be done for the coefficients $\tilde{\beta}_{\Omega\omega}$. After neglecting the non-RWA coefficients $\tilde{\alpha}_\Omega$ and $\tilde{\beta}_{\Omega\omega}$, equations (B.10) and (B.11) are reduced to equations (30) and (31) for the transformation matrix in RWA. An analogous procedure can be performed for the analysis of the spectral equation solution with negative real part. In this case, the coefficients $\tilde{\alpha}_\Omega$ and $\tilde{\beta}_{\Omega\omega}$ are not small but the coefficients α_Ω and $\beta_{\Omega\omega}$ should be neglected. Actually we have to take into account both roots of the spectral equation and within the RWA we should neglect terms such as $\alpha_\Omega \tilde{\alpha}_{\Omega'}$, $\alpha_\Omega \tilde{\beta}_{\Omega\omega'}$, $\tilde{\alpha}_\Omega \beta_{\Omega\omega'}$ and $\tilde{\beta}_{\Omega\omega} \beta_{\Omega'\omega'}$ (the terms $\tilde{\Lambda}$, $\tilde{\Delta}_\omega$ and $\tilde{\Gamma}_{\omega\omega'}$ in equations (B.23–B.26)) which are non-RWA terms. As the reduction can be done only in case of weak system–bath coupling, the RWA is valid only in this case.

Next, we will obtain the renormalized spectral equation, i.e. the spectral equation for the system described by the Hamiltonian (14) with the counterterm (20). Using equations (B.10), (B.11) and (B.14) we get the spectral equation including the renormalization

$$\Omega^2 - \omega_0^2 - 4\omega_R\, \omega_0 = 4\omega_0 \sum_{\omega} \frac{k_\omega^2\, \omega}{\Omega^2 - \omega^2}. \tag{49}$$

Using the identity

$$\frac{1}{\Omega - \omega + \mathrm{i}0} = \mathcal{P}\frac{1}{\Omega - \omega} - \mathrm{i}\pi\, \delta(\Omega - \omega) \tag{50}$$

and performing the continuum limit

$$\sum_{\omega} k_\omega^2 = \int \mathrm{d}\omega\, g(\omega) \tag{51}$$

where the function $g(\omega) = \sum_{\omega'} k_{\omega'}^2\, \delta(\omega - \omega')$ is connected with the spectral density of the bath $J(\omega)$ as

$$J(\omega) = 2\pi M \omega_0\, g(\omega) \tag{52}$$

we can write equation (49) in the form

$$\omega_0^2 - \Omega^2 + 4\omega_0\, \Delta\omega_R = 2\pi \mathrm{i}\omega_0\, g(\Omega) \tag{53}$$

where the renormalized frequency shift is

$$\Delta\omega_R = \omega_R - \frac{1}{2}\int \mathrm{d}\omega\, g(\omega)\frac{1}{\Omega + \omega} + \frac{1}{2}\int \mathrm{d}\omega\, g(\omega)\mathcal{P}\frac{1}{\Omega - \omega}. \tag{54}$$

Let us assume that the spectral equation has a complex solution and the spectral density of the bath can be written in the form

$$g(\omega) = \frac{\gamma\omega}{2\pi\omega_0}\theta(\omega, \omega_c) \tag{55}$$

where $\theta(\omega, \omega_c)$ denotes the cut-off function with the cut-off frequency ω_c. In this case, we can write for the frequency shift

$$\Delta\omega_R = \frac{\gamma}{2\pi\omega_0}\mathcal{P}\int \mathrm{d}\omega\, \theta(\omega, \omega_c)\frac{\Omega^2}{\Omega^2 - \omega^2}. \tag{56}$$

The frequency shift in the unnormalized spectral equation (i.e. $\omega_R = 0$) can be written as

$$\Delta\omega = \frac{\gamma}{2\pi\omega_0}\mathcal{P}\int \mathrm{d}\omega\, \theta(\omega, \omega_c)\frac{\omega^2}{\Omega^2 - \omega^2}. \tag{57}$$

It is easy to see that $\Delta\omega$ increases on enlarging the cut-off frequency and $\Delta\omega$ becomes infinite when $\theta(\omega, \omega_c) = 1$, and the real part of the solution ω without counterterm goes to zero with increasing cut-off frequency, i.e. on enlarging the coupling to the high-frequency bath modes. But the renormalized frequency shift (56) decreases with increasing cut-off frequency. In the limiting case $\theta(\omega, \omega_c) = 1$, the renormalized frequency shift $\Delta\omega_R$ vanishes and the spectral equation which reads

$$\omega_0^2 - \Omega^2 = 2\pi \mathrm{i}\omega_0\, g(\Omega) \tag{58}$$

has the well-known solution for the classical damped harmonic oscillator

$$\Omega_0 = \pm\sqrt{\omega_0^2 - \frac{\gamma^2}{4}} - \mathrm{i}\frac{\gamma}{2}. \tag{59}$$

So, besides the physical reasoning for the renormalization term [1], it has to be introduced to avoid singularities in the damping problem.

6. Calculation of the density matrix with non-equilibrium initial conditions

6.1. Interaction in RWA

As we are interested in a system interacting with a surrounding thermal environment, we need to calculate the reduced density operator which keeps all information about the relevant system. First, we calculate the time propagation of the reduced density matrix for the damped harmonic oscillator connected to a thermal bath which is prepared independently in the initial moment of time. Physically this means that the initial excitation of the system was faster than

the time scale of the undamped motion of the system and the time of relaxation in the thermal bath.

Let us consider the density operator of the full system (system plus bath) in the Heisenberg picture

$$\rho(t) = \mathrm{e}^{-\mathrm{i}Ht}\rho(0)\,\mathrm{e}^{\mathrm{i}Ht}. \tag{60}$$

The initial density operator is assumed to be factorized

$$\rho(0) = \rho_s(0)\rho_b(0) \tag{61}$$

where ρ_s and ρ_b correspond to the density operators of system and bath, respectively. Here we want to note in passing that the form (61) of the initial density operator does not mandatorily mean that the initial state is uncorrelated [14]. Below the case of correlated initial conditions will also be investigated.

We consider an initial bath with temperature T_b

$$\langle\{z_\omega\}|\rho_b(0)|\{z'_\omega\}\rangle = \frac{1}{Z_b}\exp\left[-\frac{1}{2}\sum_\omega |z_\omega|^2 - \frac{1}{2}\sum_\omega |z'_\omega|^2 + \sum_\omega \mathrm{e}^{-\omega/T_b}\, z^*_\omega\, z'_\omega\right] \tag{62}$$

and a system with arbitrary initial conditions determined by the function $f_0(a_1^*, a_2)$

$$\langle a_1|\rho_s(0)|a_2\rangle = \exp\left[-\frac{|a_1|^2}{2} - \frac{|a_2|^2}{2} + f_0(a_1^*, a_2)\right]. \tag{63}$$

Using the identity

$$\int \frac{\mathrm{d}z\,\mathrm{d}z^*}{\pi}\,\mathrm{e}^{-\gamma|z|^2-\delta z}\, f(z^*) = \frac{1}{\gamma} f\left(\frac{\delta}{\gamma}\right) \tag{64}$$

which is valid for any integral of Gaussian type, employing our result (41) and tracing out the bath, one obtains the matrix element of the reduced density operator in the integral form

$$\begin{aligned}\langle a|\rho_s(t)|a'\rangle = F(T_b, t)\int \frac{\mathrm{d}a_1\,\mathrm{d}a_1^*}{\pi}\frac{\mathrm{d}a_2\,\mathrm{d}a_2^*}{\pi}\exp\Bigg[&-\frac{|a|^2}{2} - \frac{|a'|^2}{2} - |a_1|^2 - |a_2|^2 + f_0(a_1^*, a_2)\\ &+ \frac{\xi}{1+Q}a^*a_1 + \frac{\xi^*}{1+Q}a_2^*a' + \frac{Q}{1+Q}a^*a' + \left(1 - \frac{|\xi|^2}{1+Q}\right)a_1a_2^*\Bigg]\end{aligned} \tag{65}$$

where

$$Q(t, T_b) = \sum_\omega |\eta_\omega(t)|^2 n_\omega \qquad n_\omega = \frac{1}{\mathrm{e}^{\omega/T_b} - 1}. \tag{66}$$

The function $F(t, T_b)$ is an unknown function coming from the integration over the end points of the bath trajectories and can be determined from the normalization condition $\mathrm{Tr}\,\rho = 1$, yielding

$$\begin{aligned}1 &= F(T_b, t)(1+Q)\int \frac{\mathrm{d}a_1^*\,\mathrm{d}a_1}{\pi}\,\exp(f_0(a_1^*, a_1) - |a_1|^2)\\ &= F(T_b, t)(1+Q)\,\mathrm{Tr}(\rho_s(0)) = F(T_b, t)(1+Q).\end{aligned} \tag{67}$$

In order to evaluate the function $\xi(t)$ defined in equation (42) we will use the method proposed in [22]. Using equation (46), identity (50) and performing the continuum limit (51) we get an equation for the eigenfrequencies

$$\Omega - \omega_0 = \mathcal{P}\int \frac{\mathrm{d}\omega\, g(\omega)}{\Omega - \omega} - \mathrm{i}\pi g(\Omega). \tag{68}$$

Then, using equation (29) for $\Omega = \Omega'$ and noting that

$$1 + \sum_{\omega} \frac{k_\omega^2}{(\Omega - \omega)^2} = \frac{\mathrm{d}}{\mathrm{d}\Omega}\left(\Omega - \omega - \sum_{\omega} \frac{k_\omega^2}{\Omega - \omega}\right) \tag{69}$$

we can write $\xi(t)$ in integral form

$$\xi(t) = \sum_{\Omega} \alpha_\Omega^2 \, \mathrm{e}^{-\mathrm{i}\Omega t} = \frac{1}{2\pi \mathrm{i}} \int_{\infty}^{-\infty} \mathrm{d}\Omega \frac{\mathrm{e}^{-\mathrm{i}\Omega t}}{\Omega - \tilde{\omega}(\Omega) + \mathrm{i}\pi g(\Omega)} \tag{70}$$

where

$$\tilde{\omega}(\Omega) = \omega_0 + \mathcal{P} \int \mathrm{d}\omega \frac{g(\omega)}{\Omega - \omega}. \tag{71}$$

The residues of this integral are determined by equation (46) for the eigenfrequencies. So the function $\xi(t)$ is the Green function of the full system, and the poles of its Fourier transform determine the eigenfrequencies of the system. Using equation (65) one finds that the Green function $\xi(t)$ and its conjugate $\xi^*(t)$ determine the time evolution of the mean values of the annihilation and creation operators, respectively, as

$$\langle a(t) \rangle = \langle a(0) \rangle \frac{\xi(t)}{1 + Q(t, T_b)} \tag{72}$$

$$\langle a^+(t) \rangle = \langle a^+(0) \rangle \frac{\xi^*(t)}{1 + Q(t, T_b)}. \tag{73}$$

As was shown in section 4, the RWA leads to correct results only in the case of weak system–bath coupling. In this limit, one can replace Ω by ω_0 in the expressions for $\tilde{\omega}(\Omega)$ and $g(\Omega)$. The Green function $\xi(t)$ then reads

$$\xi(t) = \exp(-\mathrm{i}\tilde{\omega}(\omega_0)t - \pi g(\omega_0)t). \tag{74}$$

The function $Q(t, T_b)$ in this case is given by

$$Q(t, T_b) = n_{\omega_0}(1 - |\xi(t)|^2). \tag{75}$$

In order to confirm the correctness of expression (65) let us calculate the relaxation of the mean value of the occupation number for the system

$$\langle n_s(t) \rangle = Q(t, T_b) + n_s(0)|\xi(t)|^2. \tag{76}$$

If we consider ohmic dissipation $J(\omega) = M\gamma\omega$, using equation (52) we can write [22]

$$\langle n_s(t) \rangle = n_s(0) \, \mathrm{e}^{-\gamma t} + n_{\omega_0}(1 - \mathrm{e}^{-\gamma t}) \tag{77}$$

where $\gamma \ll \omega_0$ should be fulfilled.

The functions $\xi(t)$ and $\xi^*(t)$ represent the connection between the initial and final points of the trajectories in expression (65) and hence describe the process of decoherence of the system. For time $t = \infty$ using equation (74) one gets $\xi(t = \infty) = \xi^*(t = \infty) = 0$, i.e. the system loses the information about its initial state. The matrix element of the reduced density matrix at $t = \infty$ reads

$$\begin{aligned} \langle a|\rho_s(t = \infty)|a' \rangle &= \frac{1}{Z_s} \exp\left[-\frac{|a|^2}{2} - \frac{|a'|^2}{2} + \mathrm{e}^{-\omega_0/T_b} a^* a'\right] \\ Z_s &= \frac{1}{1 - \mathrm{e}^{-\omega_0/T_b}} \end{aligned} \tag{78}$$

which exactly corresponds to the system described by density operator $\rho = \frac{1}{Z_s} \mathrm{e}^{-\omega_0 a^+ a/T_b}$.

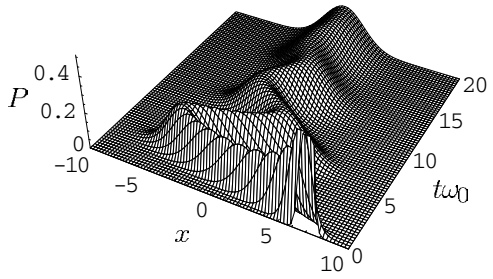

Figure 1. Evolution of $P(x,t)$ ($x_0 = 7$, $\sigma = 0.5$, $T_b = 3\omega_0$ and $\gamma = 0.2\omega_0$).

Formula (65) together with equations (74) and (75) can be used for the calculation of the mean values of observables. One has to express the initial state of the system as well as the operator of the observable via coherent states with the help of equation (1) and evaluate the integrals over the end points of the trajectories in equation (65) using equation (64). As an example, we calculate the evolution of the level populations. Assuming that the system initially is in its nth eigenstate, one gets for the evolution of the mth level

$$\langle m|\rho(t)|m\rangle = \frac{1}{n!m!}\frac{\partial^m\partial^n}{\partial\zeta_1^m\partial\zeta_2^n}\bigg|_{\zeta_1=\zeta_2=0}\frac{1}{(1+(1-\zeta_1)Q(t,T_b))(1-\zeta_2)+\zeta_2|\xi(t)|^2(1-\zeta_1)} \tag{79}$$

where the ζ_1 and ζ_2 are only auxiliary variables. The population dynamics is discussed in more detail below.

6.2. Evolution of the wave packet and population dynamics

Here we consider the relaxation in a system initially prepared in a pure state with some wavefunction $|\psi\rangle$. The initial state of the bath is an equilibrium state with temperature T_b. For the calculation of the time evolution of the system we will use equation (B.22) together with equations (B.23) to (B.32). Then, we assume the non-RWA terms (B.28, B.30, B.32) as a perturbation (i.e. the coupling strength has to be small enough). The calculations presented below have been done to first order of this perturbation. For simplicity ohmic damping, i.e. an infinite cut-off frequency, has been assumed.

First we will determine the time evolution of a Gaussian wave packet with width σ and peak position x_0, i.e.

$$\rho_s(0) = |\psi(x,t=0)\rangle\langle\psi(x,t=0)| \tag{80}$$

where

$$|\psi(x,t=0)\rangle = \frac{1}{\sqrt{\sqrt{\pi}\sigma}}\exp\left[-\frac{(x-x_0)^2}{2\sigma^2}\right] \tag{81}$$

For simplicity, we set the initial momentum to zero. The unit of measurement for x, x_0 and σ is $1/\sqrt{M\omega_0}$. In figures 1 and 2, the matrix element of the reduced density operator in coordinate representation $P(x,t) = \langle x|\rho_s(t)|x\rangle$ is shown for two different initial wave packets and temperatures. One can see that the width of the wave packet slightly oscillates with time, and the damping time for the peak position coordinate $\bar{x}$ of the wave packet is larger than the time in which the width of the wave packet changes from its initial value to the equilibrium one. Note that the wave packet keeps its Gaussian form during the time evolution.

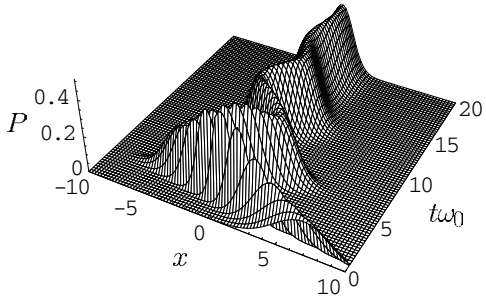

Figure 2. Evolution of $P(x,t)$ ($x_0 = 7$, $\sigma = 3$, $T_b = \omega_0$ and $\gamma = 0.2\omega_0$).

At time $t = \infty$, the function $P(x)$ can be written as

$$P(x) = \frac{1}{\sqrt{\sqrt{\pi}\sigma_f}} \exp\left(-\frac{x^2}{2\sigma_f^2}\right) \tag{82}$$

where the final width σ_f is

$$\sigma_f = \sqrt{1 + 2Q(T_b, t = \infty)}. \tag{83}$$

In the high-temperature limit of the bath, one gets $\sigma_f^2 \sim T_b$ as was shown before [7].

The time evolution of the coherent state (i.e. $\sigma = 1$) within the RWA at $T_b = 0$ has the form

$$\begin{aligned} P(x, \sigma = 1) &= \frac{1}{\pi^{1/4}} \exp\left[\frac{(x - \bar{x})^2}{2}\right] \\ \bar{x} &= x_0 \,\mathrm{Re}(\xi(t)) = x_0 \cos(\Omega_0 t) \exp(-\Omega_1 t) \end{aligned} \tag{84}$$

where Ω_0 and Ω_1 are the real and imaginary parts of the solution of the spectral equation (46), respectively. Although equation (84) implies the existence of a non-dissipative coherent state in RWA, it was shown already [14], that they can only exist at zero temperature and in systems that meet the conditions for the RWA.

Next, in figures 3 and 4 the time evolution of the populations of the system oscillator and the corresponding mean occupation numbers are shown. We considered the cases of a mixed and a pure initial state of the system. The difference between the populations with pure and mixed initial states is due to the influence of the non-RWA terms in the presence of coherence in the case of a pure initial state. The unphysical results at $t = 0^+$ in figure 4 are the consequence of jolts in the domain $0 < t < 1/\omega_c$ due to the factorized initial conditions, as was discussed by many authors (see, for example, [14] and references therein). In the case of uncorrelated initial conditions, the changes in the system at the beginning of the system–bath interaction are mostly determined by the fast decoherence of the system where the time scale of decoherence is $\tau_d \sim 1/\omega_c$. In our case, $\omega_c = \infty$ and therefore the instantaneous changes (initial jolts) of the population occur. In spite of this, the mean occupation number has the correct value at $t = 0^+$.

Finally, in figure 5 we plotted the population dynamics of a system initially prepared in a coherent state $|a\rangle$ with $|a|^2 = 1$. In spite of the presence of coherence for this initial state, one can observe relatively weak influence of non-RWA terms and an absence of the initial jolts (one can check using the formula $\rho_{m,m}(0) = \frac{|a|^{2m}}{m!} \mathrm{e}^{-|a|^2}$ for the initial state that the level populations have the correct values at $t = 0$). Since the uncertainty relation is minimized in

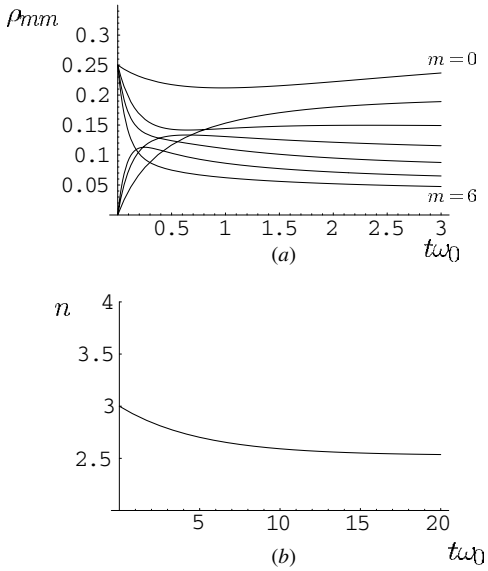

Figure 3. Evolution of the population $\rho_{mm}(t)$ of the first seven levels ($m = 0, 1, \ldots, 6$) (*a*) and the mean value of the occupation number $n(t)$ (*b*) starting from the mixed initial state of the system $\rho_s(0) = 0.25|0\rangle\langle 0| + 0.25|2\rangle\langle 2| + 0.25|4\rangle\langle 4| + 0.25|6\rangle\langle 6|$. $T_b = 3\omega_0$ and $\gamma = 0.2\omega_0$. Note that in the levels with $m > 6$ there is also some population at this value of temperature.

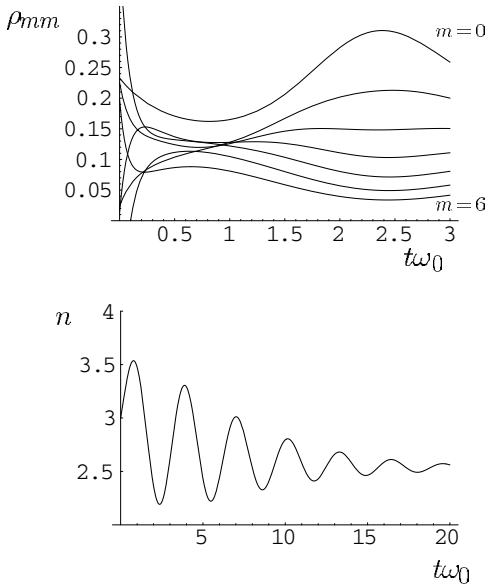

Figure 4. Evolution of the population of the first seven levels ($m = 0, 1, \ldots, 6$) and the mean value of the occupation number starting from the pure initial state of the system $|\psi(0)\rangle = 0.5|0\rangle + 0.5|2\rangle + 0.5|4\rangle + 0.5|6\rangle$. $T_b = 3\omega_0$ and $\gamma = 0.2\omega_0$.

coherent states the behaviour of the system at $t \approx 0$ is close to a classical one whereas the initial jolts caused by the fast initial decoherence appear to be purely a quantum effect.

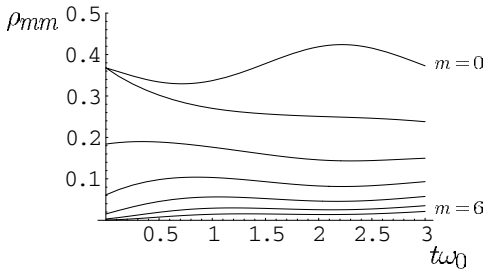

Figure 5. Evolution of the population of the first seven levels ($m = 0, 1, \ldots, 6$) starting from the coherent initial state of the system $|\psi(0)\rangle = |a\rangle$, $|a|^2 = 1$. $T_b = 3\omega_0$ and $\gamma = 0.2\omega_0$.

6.3. Influence of correlated initial system–bath conditions

In this subsection, we consider the influence of the initial correlations on the evolution of the mean value of the system coordinate and of the population dynamics. Let us suppose that the relevant system and the bath initially are in an equilibrium state with the temperature T, but the relevant vibrational mode is on a shifted harmonic potential surface, i.e.

$$V(x) = \frac{M\omega_0^2(x + x_1)^2}{2}. \tag{85}$$

Thus, the full system at the time $t < 0$ is described by the Hamiltonian

$$\tilde{H} = H + q(a^+ + a) \tag{86}$$

where $q = \omega_0 x_1/\sqrt{2}x_0$, $x_0 = 1/\sqrt{M\omega_0}$, and H is determined by equation (14). Here we will restrict ourselves to the RWA. The time evolution of the matrix element of the reduced density matrix is calculated in appendix C in the case $T = 0$. The result reads

$$\langle a|\, \mathrm{e}^{-\mathrm{i}Ht}\, \mathrm{e}^{-\lambda\tilde{H}}\, \mathrm{e}^{\mathrm{i}Ht}|a'\rangle = \exp\left[-\frac{|a|^2}{2} - \frac{|a'|^2}{2} - \varepsilon(t)a^* - \varepsilon^*(t)a' - |\varepsilon(t)|^2\right] \tag{87}$$

$$\varepsilon(t) = q\sum_{\Omega}\frac{\alpha_\Omega^2}{\Omega}\,\mathrm{e}^{-\mathrm{i}\Omega t} \tag{88}$$

$$\varepsilon^*(t) = q\sum_{\Omega}\frac{\alpha_\Omega^2}{\Omega}\,\mathrm{e}^{\mathrm{i}\Omega t}. \tag{89}$$

In order to show the influence of the initial correlations let us calculate the evolution of the mean value of the system coordinate

$$\langle x(t)\rangle = \frac{x_0}{\sqrt{2}}\langle a(t) + a^*(t)\rangle = -\frac{x_0}{\sqrt{2}}(\varepsilon(t) + \varepsilon^*(t)). \tag{90}$$

The function $\varepsilon(t)$ and its conjugate $\varepsilon^*(t)$ determine the time evolution of the annihilation and creation operators, respectively, and converge to the functions $\frac{q}{\omega_0}\xi(t)$ and $\frac{q}{\omega_0}\xi^*(t)$, respectively, if γ is very small. This can be seen by comparing equations (87) and (65) with the initial condition $f_0(a_1^*, a_2) = \frac{q}{\omega_0}(a_1^* + a_2)$ and with zero temperature of the bath. Calculating $\varepsilon(t)$ and $\varepsilon^*(t)$ in the same manner as the Green function $\xi(t)$, in the case of weak ohmic dissipation, one can get

$$\langle x(t)\rangle = -x_1\frac{\omega_0\tilde{\omega}}{\tilde{\omega}^2 + (\gamma/2)^2}\left(\cos\,\tilde{\omega}t + \frac{\gamma}{2\tilde{\omega}}\sin\,\tilde{\omega}t\right)\mathrm{e}^{-\gamma t/2} \tag{91}$$

where $\tilde{\omega} = \tilde{\omega}(\omega_0)$ is determined by equation (71). One can see that the trajectory determined by equation (91) is a classical trajectory which can be obtained for the damped harmonic oscillator only by using the correlated initial system–bath conditions [14]. Since the initial mean value of momentum is zero for the considered initial state, $\frac{\mathrm{d}\langle x(t)\rangle}{\mathrm{d}t}\Big|_{t=0} = 0$ as it should be, while for the mean value $\bar{x}$ from equation (84) obtained without initial correlations this condition is not satisfied.

7. Conclusions

In this paper, we have introduced an alternative treatment for the investigation of systems described by Hamiltonians with bilinear and quadratic terms and we applied this method to the problem of one harmonic oscillator coupled to a heat bath. In contrast to the well-known Feynman–Vernon influence-functional approach, we did not eliminate the bath but cancelled the system–bath interaction by diagonalization of the Lagrangian. In this way, one can obtain spectral equations for the system of interest which have been investigated in this work.

In contrast to a similar procedure of diagonalizing the Hamiltonian which was used in [14] and allows one to get the time evolution of the creation and annihilation operators, the present treatment gives the time evolution of the reduced density matrix and therefore leads to the full description of the relevant system. Furthermore, it allows us to consider a non-equilibrium bath and, therefore, correlated initial system–bath conditions. This consideration is normally not possible within the treatments which are based on the master equations. Besides this, our method gives a quite simple expression (65) for the reduced density matrix in RWA which can easily be used for further evaluation of the expectation values of observables, although even in the first order of non-RWA terms the calculations turned out to be quite unwieldy.

The proposed approach was tested for simple factorized initial conditions when the bath initially is in thermal equilibrium with an arbitrary temperature, and for correlated initial conditions when the relevant system initially is in thermal equilibrium with the bath but on a shifted potential. Instead of Langevin-like equations for the trajectories of the relevant system in the influence-functional method, we have a multi-dimensional integral over the end points of the trajectories. In the last section of this paper, we have shown the calculation method for such integrals in the case of weak damping. The influence of non-RWA terms such as $\tilde{\Lambda}$, $\tilde{\Delta}_\omega$ and $\tilde{\Gamma}_{\omega\omega'}$ becomes evident in the population dynamics with pure initial system conditions in the absence of initial correlations. In the first order of the perturbative treatment in the system–bath coupling, the non-RWA terms are proportional to the coupling parameter γ (in the case of ohmic dissipation $J(\omega) = M\gamma\omega$), and in the presence of coherences in the initial system state they lead to initial jolts in the population. But in the case of a system initially prepared in a coherent state, the initial jolts disappear since the system behaves almost classically at the beginning of the evolution.

The well-known Feynman–Vernon influence-functional approach is valid only in case of a bilinear system–bath coupling. One can eliminate the bath in the Feynman–Vernon manner in the case of an arbitrary system part in the interaction Hamiltonian using the diagonalization of the Lagrangian function in order to reduce the system of uncoupled harmonic oscillators to a system of one oscillator bilinearly coupled to a new effective bath and non-bilinearly coupled to the relevant system. This procedure of diagonalization is presented in our work. As this diagonalization can be done for the Hamiltonian in the creation/annihilation-operator representation, one has to use the coherent-state representation of the path integrals. After the elimination of the new bath, only integrals over the system and one harmonic mode need to be calculated. A similar procedure of normal mode transformation was used in [15] with a Hamiltonian in phase-space representation for arbitrary system–bath coupling.

Another possible application of the presented method is the description of the time evolution of electronic multilevel systems which frequently appear in electron transfer problems [17]. Using the mapping approach [23–25] the system of discrete electronic states is treated as a system of coupled harmonic oscillators in the RWA. Thus the dynamics of the electronic part of a system consisting of several electronic states can be investigated within the real-time coherent-state path integral technique developed in the present work.

Acknowledgments

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Appendix A. The matrix element of evolution operator e^{iHt}

Performing a similar procedure as for the operator e^{-iHt} in section (2), for the inverse direction of time one gets

$$\langle\{z_\omega\}|\,e^{iHt}|\{z'_\omega\}\rangle = \prod_\omega \prod_{n=1}^{N-1}\left[\int \frac{dz^*_{\omega n}\,dz_{\omega n}}{\pi}\right]\exp\left[-\sum_\omega\left(\frac{1}{2}|z_{\omega 0}|^2+\frac{1}{2}|z_{\omega N}|^2 + \sum_{n=1}^{N-1}|z_{\omega n}|^2-\sum_{n=1}^{N}z^*_{\omega n-1}\,z_{\omega n}\right)+i\sum_{n=1}^{N}\varepsilon H(\{z^*_{\omega n-1}\},\{z_{\omega n}\})\right] \tag{A.1}$$

with the boundary conditions

$$z^*_{\omega 0}=z^*_\omega \qquad z_{\omega N}=z'_\omega. \tag{A.2}$$

Then, using the identity

$$\sum_{n=1}^{N}z^*_{n-1}\,z_n-\sum_{n=1}^{N-1}|z_n|^2=\frac{1}{2}(z^*_0\,z_1+z^*_{N-1}\,z_N)+\frac{\varepsilon}{2}\sum_{n=1}^{N-1}\left(z^*_n\frac{z_{n+1}-z_n}{\varepsilon}-z_n\frac{z^*_n-z^*_{n-1}}{\varepsilon}\right) \tag{A.3}$$

and performing the continuum limit, for the matrix element of the evolution operator one can write

$$\langle\{z_\omega\}|\,e^{iHt}|\{z'_\omega\}\rangle = \prod_\tau\prod_\omega\int\frac{dz^*_\omega(\tau)\,dz_\omega(\tau)}{\pi} \times\exp\left[-\frac{1}{2}\sum_\omega(|z_\omega|^2+|z'_\omega|^2)+\tilde{S}(\{z_\omega(\tau)\},\{z^*_\omega(\tau)\})\right] \tag{A.4}$$

with the boundary conditions

$$z^*_\omega(0)=z^*_\omega \qquad z_\omega(t)=z'_\omega \tag{A.5}$$

and

$$\tilde{S}(\{z_\omega(\tau)\},\{z^*_\omega(\tau)\})=\sum_\omega z^*_\omega(0)z_\omega(0)+\int_0^t\mathcal{L}(\{z^*_\omega(\tau)\},\{z_\omega(\tau)\})\,d\tau. \tag{A.6}$$

Appendix B. Diagonalization of the Lagrangian without the RWA

Here we will evaluate the path integral (9) with the Hamiltonian without the RWA (17). First the renormalization term H_r will be neglected and reintroduced in the second part. The action (10) has the form

$$S(\{z_\omega(\tau)\},\{z_\omega^*(\tau)\}) = \frac{1}{2}\left[a^*(t)a(t) - a^*(0)a(0) + \sum_\omega z_\omega^*(t)z_\omega(t) - \sum_\omega z_\omega^*(0)z_\omega(0)\right] - \int_0^t \mathcal{L}(a(\tau),a^*(\tau),\{z_\omega^*(\tau)\},\{z_\omega(\tau)\})\,\mathrm{d}\tau \tag{B.1}$$

where the Lagrangian $\mathcal{L}$ is given by

$$\begin{aligned}\mathcal{L}(a(\tau),a^*(\tau),\{z_\omega^*(\tau)\},\{z_\omega(\tau)\}) &= \frac{1}{2}\dot{a}(\tau)a^*(\tau) - \frac{1}{2}\dot{a}^*(\tau)a(\tau) + \frac{1}{2}\sum_\omega \dot{z}_\omega(\tau)z_\omega^*(\tau) - \frac{1}{2}\sum_\omega \dot{z}_\omega^*(\tau)z_\omega(\tau)\\ &\quad + \mathrm{i}\omega_0 a^*(\tau)a(\tau) + \mathrm{i}\sum_\omega z_\omega^*(\tau)z_\omega(\tau) + \mathrm{i}\sum_\omega k_\omega(a^*(\tau)+a(\tau))(z_\omega^*(\tau)+z_\omega(\tau))\end{aligned} \tag{B.2}$$

which should be taken with the boundary conditions (22) and (23).

Going beyond the RWA we have to use an extended transformation, since the non-RWA terms in the interaction part of the Hamiltonian mix the operators a with a^+. The transformation which should be used in this case is

$$a = \sum_\Omega \alpha_\Omega A_\Omega + \sum_\Omega \tilde{\alpha}_\Omega A_\Omega^* \tag{B.3}$$

$$a^* = \sum_\Omega \alpha_\Omega A_\Omega^* + \sum_\Omega \tilde{\alpha}_\Omega A_\Omega \tag{B.4}$$

$$z_\omega = \sum_\Omega \beta_{\Omega\omega} A_\Omega + \sum_\Omega \tilde{\beta}_{\Omega\omega} A_\Omega^* \tag{B.5}$$

$$z_\omega^* = \sum_\Omega \beta_{\Omega\omega} A_\Omega^* + \sum_\Omega \tilde{\beta}_{\Omega\omega} A_\Omega. \tag{B.6}$$

Below we will follow a similar procedure as within the RWA. Substituting the trajectories a, a^*, z_ω and z_ω^* as functions of A_Ω and A_Ω^* into the Lagrangian (B.2) we require the Lagrangian to have diagonal form after the transformation, i.e.

$$\begin{aligned}\mathcal{L}(a(\tau),a^*(\tau),\{z_\omega^*(\tau)\},\{z_\omega(\tau)\}) &= \frac{1}{2}\sum_\Omega \dot{A}_\Omega(\tau)A_\Omega^*(\tau) - \frac{1}{2}\sum_\Omega \dot{A}_\Omega^*(\tau)A_\Omega(\tau) + \mathrm{i}\sum_\Omega \Omega A_\Omega^*(\tau)A_\Omega(\tau).\end{aligned} \tag{B.7}$$

It leads to the following orthogonality relations for the transformation matrices:

$$\alpha_\Omega\,\alpha_{\Omega'} - \tilde{\alpha}_\Omega\,\tilde{\alpha}_{\Omega'} + \sum_\omega \beta_{\Omega\omega}\,\beta_{\Omega'\omega} - \sum_\omega \tilde{\beta}_{\Omega\omega}\,\tilde{\beta}_{\Omega'\omega} = \delta_{\Omega\Omega'} \tag{B.8}$$

$$\alpha_\Omega\,\tilde{\alpha}_{\Omega'} - \tilde{\alpha}_\Omega\,\alpha_{\Omega'} + \sum_\omega \beta_{\Omega\omega}\,\tilde{\beta}_{\Omega'\omega} - \sum_\omega \tilde{\beta}_{\Omega\omega}\,\beta_{\Omega'\omega} = 0 \tag{B.9}$$

and to the equations for the transformation matrix coefficients

$$\alpha_\Omega(\omega_0 - \Omega) = \tilde{\alpha}_\Omega(\omega_0 + \Omega) = -\sum_\omega k_\omega(\beta_{\Omega\omega} + \tilde{\beta}_{\Omega\omega}) \tag{B.10}$$

$$\beta_{\Omega\omega}(\omega - \Omega) = \tilde{\beta}_{\Omega\omega}(\omega + \Omega) = -k_\omega(\alpha_\Omega + \tilde{\alpha}_\Omega). \tag{B.11}$$

One may also diagonalize the Lagrangian including renormalization (20). The renormalized Lagrangian $\mathcal{L}_R$ corresponding to the Hamiltonian (14) is the Lagrangian (B.2) with the additional term

$$\mathcal{L}_r = \mathrm{i}\omega_R((a^*)^2 + a^2 + 2a^*a) \tag{B.12}$$

with the renormalization frequency

$$\omega_R = \sum_\omega \frac{k_\omega^2}{\omega}. \tag{B.13}$$

It is obvious that the orthogonality relations (B.8) and (B.9) remain the same. Equation (B.10) for the transformation matrix coefficients changes and is now given by

$$\alpha_\Omega(\omega_0 - \Omega) = \tilde{\alpha}_\Omega(\omega_0 + \Omega) = -\sum_\omega k_\omega(\beta_{\Omega\omega} + \tilde{\beta}_{\Omega\omega}) - 2\omega_R(\alpha_\Omega + \tilde{\alpha}_\Omega). \tag{B.14}$$

Equation (B.11) remains the same.

The back transformation reads

$$A_\Omega = \alpha_\Omega\, a - \tilde{\alpha}_\Omega\, a^* + \sum_\omega \beta_{\Omega\omega}\, z_\omega - \sum_\omega \tilde{\beta}_{\Omega\omega}\, z_\omega^* \tag{B.15}$$

$$A_\Omega^* = \alpha_\Omega\, a^* - \tilde{\alpha}_\Omega\, a + \sum_\omega \beta_{\Omega\omega}\, z_\omega^* - \sum_\omega \tilde{\beta}_{\Omega\omega}\, z_\omega. \tag{B.16}$$

The final step is to find the additional orthogonality relations for the transformation matrices. Substituting A_Ω and A_Ω^* as functions of a, a^*, z_ω and z_ω^* (B.15) and (B.16) into equation (B.3) to (B.6) and using the orthogonality of the transformation, one can obtain

$$\sum_\Omega \left(\alpha_\Omega^2 - \tilde{\alpha}_\Omega^2\right) = 1 \tag{B.17}$$

$$\sum_\Omega (\alpha_\Omega\, \beta_{\Omega\omega} - \tilde{\alpha}_\Omega\, \tilde{\beta}_{\Omega\omega}) = 0 \tag{B.18}$$

$$\sum_\Omega (\alpha_\Omega\, \tilde{\beta}_{\Omega\omega} - \tilde{\alpha}_\Omega\, \beta_{\Omega\omega}) = 0 \tag{B.19}$$

$$\sum_\Omega (\beta_{\Omega\omega}\, \beta_{\Omega\omega'} - \tilde{\beta}_{\Omega\omega}\, \tilde{\beta}_{\Omega\omega'}) = \delta_{\omega\omega'} \tag{B.20}$$

$$\sum_\Omega (\beta_{\Omega\omega}\, \tilde{\beta}_{\Omega\omega'} - \tilde{\beta}_{\Omega\omega}\, \beta_{\Omega\omega'}) = 0. \tag{B.21}$$

The Lagrangian of form (B.7) describes a system of independent harmonic oscillators. Applying the stationary-phase method we get the stationary trajectories (37) and (38) and the Lagrangian is equal to zero at the stationary point.

For the matrix element of the evolution operator, one gets

$$\langle\{z_\omega\}|\langle a|\,\mathrm{e}^{-\mathrm{i}Ht}|a'\rangle|\{z'_\omega\}\rangle = \exp\left[-\frac{1}{2}|a|^2 - \frac{1}{2}|a'|^2 - \frac{1}{2}\sum_\omega |z_\omega|^2 - \frac{1}{2}\sum_\omega |z'_\omega|^2 + \frac{1}{2}\left(a^*(t)a(t) - a^*(0)a(0) + \sum_\omega z^*_\omega(t)z_\omega(t) - \sum_\omega z^*_\omega(0)z_\omega(0)\right)\right]. \tag{B.22}$$

Using the boundary conditions (22) and (23) and the solution for the stationary trajectories (37) and (38), we can find the connection between the end points of the trajectories $a(\tau)$, $a^*(\tau)$, $z_\omega(\tau)$ and $z^*_\omega(\tau)$ at the moments of time $\tau = 0$ and $\tau = t$

$$a(0) = \Lambda(t)a(t) + \tilde{\Lambda}(t)a^*(t) + \sum_\omega \Delta_\omega(t)z_\omega(t) + \sum_\omega \tilde{\Delta}_\omega(t)z^*_\omega(t) \tag{B.23}$$

$$z_\omega(0) = \Delta_\omega(t)a(t) - \tilde{\Delta}^*_\omega(t)a^*(t) + \sum_{\omega'} \Gamma_{\omega\omega'}(t)z_{\omega'}(t) + \sum_{\omega'} \tilde{\Gamma}_{\omega\omega'}(t)z^*_{\omega'}(t) \tag{B.24}$$

$$a^*(0) = \Lambda^*(t)a^*(t) + \tilde{\Lambda}^*(t)a(t) + \sum_\omega \Delta^*_\omega(t)z^*_\omega(t) + \sum_\omega \tilde{\Delta}^*_\omega(t)z_\omega(t) \tag{B.25}$$

$$z^*_\omega(0) = \Delta^*_\omega(t)a^*(t) - \tilde{\Delta}_\omega(t)a(t) + \sum_{\omega'} \Gamma^*_{\omega\omega'}(t)z^*_{\omega'}(t) + \sum_{\omega'} \tilde{\Gamma}^*_{\omega\omega'}(t)z_{\omega'}(t) \tag{B.26}$$

where

$$\Lambda(t) = \sum_\Omega \left(\alpha^2_\Omega\,\mathrm{e}^{\mathrm{i}\Omega t} - \tilde{\alpha}^2_\Omega\,\mathrm{e}^{-\mathrm{i}\Omega t}\right) \tag{B.27}$$

$$\tilde{\Lambda}(t) = \sum_\Omega (\tilde{\alpha}_\Omega\,\alpha_\Omega\,\mathrm{e}^{-\mathrm{i}\Omega t} - \alpha_\Omega\,\tilde{\alpha}_\Omega\,\mathrm{e}^{\mathrm{i}\Omega t}) \tag{B.28}$$

$$\Delta_\omega(t) = \sum_\Omega (\alpha_\Omega\,\beta_{\Omega\omega}\,\mathrm{e}^{-\mathrm{i}\Omega t} - \tilde{\alpha}_\Omega\,\tilde{\beta}_{\Omega\omega}\,\mathrm{e}^{\mathrm{i}\Omega t}) \tag{B.29}$$

$$\tilde{\Delta}_\omega(t) = \sum_\Omega (\tilde{\alpha}_\Omega\,\beta_{\Omega\omega}\,\mathrm{e}^{-\mathrm{i}\Omega t} - \alpha_\Omega\,\tilde{\beta}_{\Omega\omega}\,\mathrm{e}^{\mathrm{i}\Omega t}) \tag{B.30}$$

$$\Gamma_{\omega\omega'}(t) = \sum_\Omega (\beta_{\Omega\omega}\,\beta_{\Omega\omega'}\,\mathrm{e}^{\mathrm{i}\Omega t} - \tilde{\beta}_{\Omega\omega}\,\tilde{\beta}_{\Omega\omega'}\,\mathrm{e}^{-\mathrm{i}\Omega t}) \tag{B.31}$$

$$\tilde{\Gamma}_{\omega\omega'}(t) = \sum_\Omega (\tilde{\beta}_{\Omega\omega}\,\beta_{\Omega\omega'}\,\mathrm{e}^{-\mathrm{i}\Omega t} - \beta_{\Omega\omega}\,\tilde{\beta}_{\Omega\omega'}\,\mathrm{e}^{\mathrm{i}\Omega t}). \tag{B.32}$$

Expression (B.22) for the matrix element of the evolution operator, depending on the end points of the trajectories together with equations (B.23) to (B.32) will be used for the calculation of the time evolution of the reduced density matrix.

Appendix C. Calculation of the evolution of the density matrix with correlated initial conditions

The purpose is to calculate the matrix element of the reduced density matrix

$$\frac{1}{Z_{sb}}\langle a|\,\mathrm{e}^{-\mathrm{i}Ht}\,\mathrm{e}^{-\lambda\tilde{H}}\,\mathrm{e}^{\mathrm{i}Ht}|a'\rangle \tag{C.1}$$

with $Z_{sb} = \mathrm{Tr}\,\mathrm{e}^{-\lambda\tilde{H}}$ and λ being the inverse temperature. Writing the matrix element of the initial density operator $\mathrm{e}^{-\lambda\tilde{H}}$ as an imaginary-time path integral and using the procedure of diagonalization of the Lagrangian presented above, one gets

$$\langle z_\omega|\langle a|\,\mathrm{e}^{-\lambda\tilde{H}}|a'\rangle|z'_\omega\rangle = \exp\left[-\frac{|a|^2}{2} - \frac{|a'|^2}{2} - \sum_\omega \frac{|z_\omega|^2}{2} - \sum_\omega \frac{|z'_\omega|^2}{2} + a^* a' \tilde{\xi}(\lambda) + \sum_\omega \tilde{\eta}(\lambda)(a^* z'_\omega + a' z^*_\omega) + \sum_{\omega\omega'} \tilde{\mu}_{\omega\omega'}(\lambda) z^*_\omega z'_{\omega'} - \varphi_1(\lambda)a^* - \varphi_2(\lambda)a' - \sum_\omega \chi_{1\omega}(\lambda) z^*_\omega - \sum_\omega \chi_{2\omega}(\lambda) z'_\omega + \chi_0(\lambda)\right] \tag{C.2}$$

where

$$\tilde{\xi}(\lambda) = \sum_\Omega \alpha_\Omega^2\,\mathrm{e}^{-\Omega\lambda} \tag{C.3}$$

$$\tilde{\eta}_\omega(\lambda) = \sum_\Omega \alpha_\Omega\,\beta_{\Omega\omega}\,\mathrm{e}^{-\Omega\lambda} \tag{C.4}$$

$$\tilde{\mu}_{\omega\omega'}(\lambda) = \sum_\Omega \beta_{\Omega\omega}\,\beta_{\Omega\omega'}\,\mathrm{e}^{-\Omega\lambda} \tag{C.5}$$

$$\varphi_1(\lambda) = q\sum_\Omega \alpha_\Omega^2 \int_0^\lambda \mathrm{e}^{-\Omega(\lambda - s)}\,\mathrm{d}s \tag{C.6}$$

$$\varphi_2(\lambda) = q\sum_\Omega \alpha_\Omega^2 \int_0^\lambda \mathrm{e}^{-\Omega s}\,\mathrm{d}s \tag{C.7}$$

$$\chi_{1\omega}(\lambda) = q\sum_\Omega \alpha_\Omega\,\beta_{\Omega\omega} \int_0^\lambda \mathrm{e}^{-\Omega(\lambda - s)}\,\mathrm{d}s \tag{C.8}$$

$$\chi_{2\omega}(\lambda) = q\sum_\Omega \alpha_\Omega\,\beta_{\Omega\omega} \int_0^\lambda \mathrm{e}^{-\Omega s}\,\mathrm{d}s \tag{C.9}$$

$$\chi_0(\lambda) = q^2\sum_\Omega \alpha_\Omega^2 \int_0^\lambda \int_0^s \mathrm{e}^{-\Omega(s - s')}\,\mathrm{d}s'\,\mathrm{d}s. \tag{C.10}$$

Below we will consider the case $T = 0\,(\lambda = \infty)$. Hence $\tilde{\xi}(\lambda) = \tilde{\eta}_\omega(\lambda) = \tilde{\mu}_{\omega\omega'}(\lambda) = 0$. Then, using equation (C.2) together with our result for the matrix element of the evolution operator (41) and integrating over the end points of the trajectories one obtains for the matrix element (C.1)

$$\langle a|\,\mathrm{e}^{-\mathrm{i}Ht}\,\mathrm{e}^{-\lambda\tilde{H}}\,\mathrm{e}^{\mathrm{i}Ht}|a'\rangle = \tilde{F}(t)\exp\left[-\frac{|a|^2}{2} - \frac{|a'|^2}{2} - \varepsilon_1(t)a^* - \varepsilon_2(t)a'\right] \tag{C.11}$$

$$\varepsilon_1(t) = \xi(t)\varphi_1 + \sum_\omega \eta_\omega(t)\chi_{1\omega} \tag{C.12}$$

$$\varepsilon_2(t) = \xi^*(t)\varphi_2 + \sum_\omega \eta^*_\omega(t)\chi_{2\omega} \tag{C.13}$$

where the pre-exponential factor $\tilde{F}(t)$ is an unknown function of time coming from the integration over the end points of the bath trajectories. Using the normalization condition $\mathrm{Tr}\,\rho(t) = 1$, one obtains

$$\tilde{F}(t) = \mathrm{e}^{-\varepsilon_1(t)\varepsilon_2(t)}. \tag{C.14}$$

Finally, using equations (C.3) to (C.10) for zero temperature ($\lambda = \infty$) and introducing $\varepsilon = \varepsilon_1$ ($\lambda = \infty$) and $\varepsilon^* = \varepsilon_2$ ($\lambda = \infty$) one gets the result (87).

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