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The off-resonant aspects of decoherence and a critique of the two-level approximation

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Abstract

Conditions in favour of a realistic multilevelled description of a decohering quantum system are examined. In this regard the first crucial observation is that the thermal effects, contrary to the conventional belief, play a minor role at low temperatures in the decoherence properties. The system–environment coupling and the environmental energy spectrum dominantly affect the decoherence. In particular, zero temperature quantum fluctuations or non-equilibrium sources can be present and influential on the decoherence rates in a wide energy range allowed by the spectrum of the environment. A crucial observation against the validity of the two-level approximation is that the decoherence rates are found to be dominated not by the long time resonant but the short time *off-resonant* processes. This observation is demonstrated in two stages. Firstly, our zero temperature numerical results reveal that the calculated short time decoherence rates are Gaussian-like (the time dependence of the density matrix is led by the second time derivative at $t = 0$). Exact analytical results are also permitted in the short time limit, which, consistent with our numerical results, reveal that this specific Gaussian-like behaviour is a property of the non-Markovian correlations in the environment. These Gaussian-like rates have no dependence on any spectral parameter (position and the width of the spectrum) except, in totality, the spectral area itself. The dependence on the spectral area is a power law. Furthermore, the Gaussian-like character at short times is independent of the number of levels (N), but the numerical value of the decoherence rates is a monotonic function of N. In this context, we demonstrate that leakage, as a characteristic multilevel effect, is dominated by the non-resonant processes.

The long time behaviour of decoherence is also examined. Since our spectral model allows Markovian environmental correlations at long times, the decoherence rates in this regime become exponential independently from the number of levels. The latter and the coupling strengths play the major role in the quantitative values of the rates and the rates are independent of the other spectral parameters.

The validity of the presented results is restricted only by their reliance on the Born–Oppenheimer approximation. This approximation is strongly dependent

on the external observational time and its reliability depends on an additional timescale. In the rest of the work, the crossover between the short and the long time behaviour of the density matrix of the multilevelled system is examined using an intuitive argument. It is shown that the Born approximation weakens as the resonant couplings become more effective at long times. This implies that, in calculations made with this approximation in the long time regime, a need for a justification arises for the reliability of the results. This justification is made for the present work.

1. Introduction

Two-level models are almost routinely used in recent applications concerning the decoherence and dephasing properties of open quantum systems. The well studied spin-boson [1, 2] or central spin [3] models are such examples. The general assumption is that for sufficiently low temperatures $T \ll \Delta E$, a multilevelled quantum system (MLS) is well approximated by its first two levels [1] (where ΔE is a characteristic energy scale separating the first two levels from the higher excited states of the quantum system). On the other hand, the nature of the environmental coupling and the properties of the environmental spectrum are also essential factors in the determination of the decoherence properties in an MLS. For a realistic MLS under weak environmental perturbation, it is sufficient to consider a Caldeira–Leggett-type linear coupling [4, 5] of a system coordinate to an environmental coordinate. The spectral range of the environment usually extends to energies well beyond the energy scales of the MLS. The decoherence properties of the MLS are expected to be affected at short times by the entire range of the environmental spectrum. This basic short time property of the system–environment coupling has been largely ignored in the works on decoherence until recently, due to the fact that the environmental correlations have been usually considered in the Markovian regime. A few theoretical approaches now exist on the effect of multilevels in the long time decoherence properties [6, 7], largely ignoring the short time behaviour. This long time resonant transition based approach has now become a tradition and the usual practice is to use the Fermi golden rule. The calculated decoherence rates, overall, reveal the Markovian sub-character yielding exponential time behaviour for the reduced density matrix (RDM). The latter is usually solved by analytical methods such as Bloch–Redfield [8] or Lindblad [9] operator formalisms, which in turn has the advantage of transforming the master equation into linearly coupled first-order ordinary differential equations. As the result of the Markov approximation, the corresponding Bloch–Redfield coefficients or the Lindblad operators become independent of time yielding the exponential decoherence rates influenced from the resonant sector in the system–environment coupling. These analytical techniques have the ease of being applicable to systems with arbitrarily large numbers of levels. On the other hand, approximations emphasizing the Markovian subnature and assuming weak interactions with the environment have been questioned by some authors to be restricted by the resonant transitions only [11]. From another point of view, the Markov approximation, and in turn the Bloch–Redfield formalism, is also expected to break down at observational times shorter than the environmental correlation times. Such is the case when the excitations of the decohering system into the higher levels become non-negligible at short times. Here the short time implies times shorter than the typical environmental correlation time below which the system environment coupling is allowed to remember the history of the non-Markovian time evolution. In this regard, our previous work is close to considering the full non-Markovian

scheme requiring the exact solution of the master equation of the multilevelled reduced density matrix [6]. There, we numerically solved the non-Markovian RDM including the retardation effects in which the short time and the long time mechanisms of decoherence are seen to be governed by qualitatively distinct behaviour.

In this work we expand our previous work and propose concrete counter-arguments against the validity of the resonant based approaches on decoherence: in particular, the two-level approximation (2LA). A comparison between the 2LA and the full multilevelled description is allowed by solving the master equation for an interacting multilevelled system.

In section 2 we revisit the two-level approximation and recall its basic assumptions for later reference. In section 3, we introduce the multilevel system–environment interacting model and derive the fundamental master equation governing the decoherence dynamics. In section 4, the short time decoherence is investigated, the parameters affecting it are identified and the existence of the Gaussian-like timescales is proven. In section 5, we focus our attention on the long time decoherence. For our investigation in this paper to be correct at long times, the Born–Oppenheimer approximation, as being the only approximation made in our scheme, must be valid in all decoherence timescales examined. Section 6 is devoted to examining this question.

2. The two-level approximation revisited

The 2LA has, in its essence, three assumptions:

(a) that the incoherent transitions caused by the environment in the system are generated by the resonant processes: implicit in the 2LA is the belief that the spectrum must have non-negligible couplings at the *right transition frequencies* at which the system makes transitions to higher levels;
(b) at zero or sufficiently low temperatures there are no available environmental states to couple with the system. This assumption and the notion of the *right transition frequency* is basically tempting one to neglect all parts of the spectrum $\Delta E \leqslant \omega$ because of the long standing belief that at sufficiently low temperatures the interacting part of the spectrum is in the low energies $\omega \leqslant T \ll \Delta E$ of which coupling is believed to be suppressed by the low temperature;
(c) negligible leakage of the wavefunction to higher levels: in fact this assumption has not been seriously questioned until recently. Counter-arguments against neglecting the leakage effects in the long time dynamics can be found for instance in the recent publications [7, 10].

A number of experiments have also been performed emphasizing the multilevelled nature of those systems particularly used in the context of quantum computation. For instance, current-biased Josephson junctions or rf-SQUIDs have been thoroughly investigated in the second half of [12]. Examples from solid state devices have been examined in section 7 of this paper, where references are provided.

Thsi paper presents concrete arguments against these three fundamental assumptions of the 2LA by considering an MLS interacting with a bosonic environmental bath at zero temperature through a Caldeira–Leggett-type linear and non-diagonal coupling. By considering a generic spectral noise the range of which is allowed to extend well beyond the thermal and the system's energy scales, and also adopting a model MLS, we demonstrate at zero temperature that the three basic assumptions above are neither necessary nor sufficient for the validity of the 2LA.

The method that we employ is the direct numerical solution of the non-Markovian master equation for the MLS's reduced density matrix in the Born–Oppenheimer approximation (BOA) including a large range of timescales. Since the BOA is exact at short times, our

calculations are exact in this regime and highly reliable in the intermediate times. Therefore, in the solution of the RDM, the short time and the long time behaviour are separately examined by their characteristic Gaussian-like and exponential signatures respectively. It is shown that the decoherence which exists at all timescales at different strengths is dominated by the short time dynamics induced by the off-resonant processes. The calculated Gaussian-like decoherence rates can be of the order or an order of magnitude higher than the exponential ones. The off-resonant processes are stimulated by the low temperature quantum fluctuations or by non-thermal processes and the corresponding short time decoherence rates are finite at zero temperature. This observation basically invalidates the assumptions (a) and (b) above. The rates thus found are shown to have no direct dependence on any other parameter than the spectral area, which is shown to be a square-root type, independently from the number of levels in the MLS. Similar off-resonant signatures have also been recently suggested analytically by other workers for exactly two-level systems [10]. Furthermore, we observe that in an MLS, off-resonant processes cause strong wavefunction leakage from the two-level subspace to the system's higher levels. Our numerically calculated leakage (L) rates are of the same order of magnitude as the relaxation (R) and dephasing (D) rates. Therefore the leakage, being a characteristic MLS effect, renders itself to be non-negligible at short times. The observation of strong leakage at short times invalidates assumption (c) above in that the effect of multilevels in decoherence cannot be considered, as believed, a mere renormalization of the effective two-level system parameters.

Similar zero temperature decoherence mechanisms have been recently verified for the mesoscopic persistent current rings experimentally [13] and studied theoretically [14]. In particular, the saturation observed in the electron dephasing rates in disordered conductors [15] has been argued in favour of the zero temperature non-resonant quantum fluctuations [16].

3. A multilevel system–environment model

We use a system–environment model where the system is represented by a spectrum with an evenly spaced large number of levels. The regularity of the energy spacing is of no importance in the short time regime where the off-resonant transitions dominate. An energetically two-level-like system and the system with regular spacing have identical decoherence properties in the shortest timescales[3]. Physically, this model can represent for instance an rf-SQUID ring under a single well flux potential [6, 17] but our general results and conclusions are not restricted by any physical model. The total Hamiltonian, in the eigenbasis of the system, can be written as [6]

$$\mathcal{H} = \sum_{n=1}^{N} E_n |n\rangle\langle n| + \int \mathrm{d}\omega\, \omega (a_\omega^\dagger a_\omega + a_\omega a_\omega^\dagger) + \sum_{n,r=1}^{N} \varphi_{nr} |n\rangle\langle r| \int \mathrm{d}\omega\, \eta_\omega (a_\omega + a_\omega^\dagger) + \sum_{n,r} \varphi_{nr}^2 |n\rangle\langle r| \int \mathrm{d}\omega\, \eta_\omega^2. \tag{1}$$

Here $E_n = n - 0.5$ for $1 < n$ are fixed system harmonic-like eigenenergies in some absolute (and irrelevant) energy scale and $|r\rangle$ indicates the rth system eigenstate. The environmental

[3] It may seem contradictory to challenge the two-level approximation using an energetically equally spaced model as in equation (1). In section 5 we demonstrate that the decoherence is a phenomenon which becomes effective at the short times and the short time decoherence is dominated by the off-resonant processes. These transitions basically ignore the information about the details of the distribution of the energy levels. Differences in the decoherence properties between different energy distributions start developing at intermediate and long timescales where the resonant transitions start dominating the decoherence. See sections 6 and 8 for details.

mode at frequency ω is annihilated (created) by the operator a_ω ($a_\omega^\dagger$). The last term usually arises in the linear coupling models which compensates for the renormalization of the system's energy levels. The correction brought by this term to the transitional amplitudes is in the second order of the dipole couplings unlike the leakage effects we are interested in (which are second order in dipole couplings). The effect of this correction term will be analysed in the next section.

An important remark in defending our point is that the positions of the E_ns qualitatively have no effect on the short and intermediate time decoherence rates until the resonant transitions become gradually effective at longer timescales. Therefore our choice of an equally spaced system should pose to threat against challenging the two-level approximation at short and intermediate times (also see remark (see footnote 3)).

We consider the system's dipole matrix elements φ_{nr} to be real, symmetric and purely off-diagonal given by

$$\varphi_{nr} = \begin{cases} 0.1\mathrm{e}^{-|n-r|/R} & \text{if } n+r = \text{odd} \\ 0 & \text{if } n+r = \text{even} \end{cases} \tag{2}$$

where the exponential coupling range is considered to be $R = 10$ number of levels. This coupling is stimulated by the parity selection rules of an even system potential and it can be generated by an rf-SQUID under a symmetric double well flux potential. In this system the range of the dipole coupling depends on the operational point of the system parameters. The frequency dependent system–environment coupling is given by η_ω. All other energy (time) scales—including the spectral parameters—are dimensionless in the same absolute energy (time) scale. For instance SQUID based systems are very popular devices used for the decoherence measurements for which typical experimental system parameters [17] yield approximately 1–10 meV or 10^{-2}–10^{-1} ps. Solid state based devices also yield similar energy scales as studied in section 7 below. The environment is characterized by a thermal spectral function $I(\omega) = \eta_\omega^2(2n_\omega+1)$ where n_ω is the bosonic distribution. Since our results are confined to zero temperature and the environment is in thermal equilibrium, the bare distribution $n_\omega = 0$, and therefore $I(\omega) = \eta_\omega^2$ for which we use a Lorentzian spectral model below. For obtaining results in regard of the pure two-level system, we set the couplings to the higher levels in (1) equal to zero (i.e. $\varphi_{1n} = \varphi_{2n} = \varphi_{nn'} = 0$ for $2 < n, n'$).

3.1. The master equation for the reduced system

The quantum system is assumed to be initially prepared as $|\psi(0)\rangle = a|1\rangle + b|2\rangle$ where $a = \sqrt{0.1}, b = \mathrm{e}^{\mathrm{i}\pi/2}\sqrt{0.9}$. The system and the environmental degrees of freedom are initially assumed to be decoupled. We solve the master equation for the MLS interacting with a zero temperature environment in the Born–Oppenheimer approximation (BOA) [18] in which the entire density matrix is assumed to be well approximated by the product form $\hat{\tilde{\rho}}(t) = \hat{\tilde{\rho}}^{(S)}(t) \otimes \hat{\tilde{\rho}}_{\mathrm{e}}(0)$. Here $\tilde{}$ denotes the interaction picture, $\tilde{\rho}^{(S)}(t)$ denotes the reduced density matrix (RDM) of the system at time t and ρ_{e} is the density matrix of the pure environment. In the short time regime BOA is nearly manifest. For the approximation to hold at long times, the equilibration time of the environment should be longer than the exponential decoherence rates. A thorough study of the validity of the BOA in this regime is provided in the section 8. The full density matrix satisfies, in the interaction picture,

$$\frac{\mathrm{d}}{\mathrm{d}t}\tilde{\rho}(t) = \mathrm{i}[\tilde{\mathcal{H}}_{\mathrm{int}}(t) + \tilde{\mathcal{H}}_{\mathrm{c}}(t), \tilde{\rho}(t)] \tag{3}$$

where $\tilde{\mathcal{H}}_{\mathrm{int}}(t)$ is the system–noise interaction (the second term) in equation (1). The effect of the compensating term $\tilde{\mathcal{H}}_{\mathrm{c}}(t)$ (the last term in the same equation) is to shift the system basis

by a unitary transformation

$$\hat{U}_t = \mathcal{T}\exp\left(-\mathrm{i}\int_0^t \mathrm{d}t'\,\tilde{\mathcal{H}}_{\mathrm{c}}(t')\right) \tag{4}$$

by which the full density matrix $\tilde{\rho}(t)$ is transformed into $\tilde{\rho}(t) \to \tilde{\rho}(t)_U$ as $\tilde{\rho}(t)_U = \hat{U}_t^{\dagger}\tilde{\rho}(t)\hat{U}_t$. Here $\mathcal{T}$ stands for the time ordering. The reduced transformed density matrix (RDM) is found as usual by tracing over the environmental degrees of freedom of which the matrix elements in the original system basis are found. After some algebra we find that

$$\frac{\mathrm{d}}{\mathrm{d}t}(\tilde{\rho}_U^{(S)})_{nm}(t) = -\int_0^t \mathrm{d}t' \sum_{r,s} K_{rs}^{nm}(t,t')(\tilde{\rho}_U^{(S)})_{rs}(t') \tag{5}$$

where

$$\begin{aligned} K_{rs}^{nm}(t,t') = \{&\mathcal{F}(t-t')[(\tilde{\varphi}_U(t)\tilde{\varphi}_U(t'))_{nr}\delta_{s,m} - (\tilde{\varphi}_U(t'))_{nr}(\tilde{\varphi}_U(t))_{sm}] \\ &+ \mathcal{F}^*(t-t')[(\tilde{\varphi}_U(t')\tilde{\varphi}_U(t))_{sm}\delta_{r,n} - (\tilde{\varphi}_U(t))_{nr}(\tilde{\varphi}_U t')_{sm}]\} \end{aligned} \tag{6}$$

is the non-Markovian system–noise kernel. In an MLS the time translational symmetry is lost by the presence of a variety of possible transitions. Here $(\tilde{\varphi}_U(t))_{nr} = (\tilde{\varphi}_U(0))_{nr}\mathrm{e}^{-\mathrm{i}(E_n-E_r)t}$. The net effect of (4) on the interacting system is therefore to transform the original system dipole matrix $\tilde{\varphi}(t))$ into

$$\tilde{\varphi}_U(t) = \hat{U}_t\tilde{\varphi}(t)\hat{U}_t^{\dagger}. \tag{7}$$

We will refer to (7) to examine the effect of the compensating term in section 4.

3.2. The environmental correlator and the spectrum

Next, we introduce the environmental auto-correlation function, i.e. the noise correlator, as

$$\mathcal{F}(\tau) = \Theta(\tau)\int_{-\infty}^{\infty}\mathrm{d}\omega\,\mathrm{e}^{\mathrm{i}\omega\tau}I(\omega) \tag{8}$$

where Θ is the Heaviside step function and, $I(\omega)$ is the environmental spectrum for which we adopt a Lorentzian form

$$I(\omega) = \frac{A}{\pi}\frac{\epsilon}{(\omega-\omega_0)^2+\epsilon^2}. \tag{9}$$

The physical parameters of the Lorentizan spectrum are then the central frequency ω_0 and the spectral width ϵ of which the effects on the decoherence timescales can be independently examined. The spectral area is denoted by $A = \int_{-\infty}^{\infty}\mathrm{d}\omega I(\omega)$ which is nothing but a multiplicative factor in the system environment coupling in equation (6). The special choice of the Lorentzian is made so that the three parameters, the area A, the central frequency ω_0 and the width ϵ, can all be studied independently from each other in compliance with the purpose of this work. The model spectrum in (9) is unrealistic as a physical noise unlike the power exponential or the power Gaussian [6], which are more conventionally used for modelling the physical spectra. We could have used these more common spectral models had we given up the advantage of separating the independent parameters of the spectrum, namely the width (ϵ), the centre (ω_0) and the strength (A). The conclusion obtained from the spectrum (9) will, however, be general and applicable to the different physical models of environmental noise (with a most probable exception of $1/\omega^f$ type noise spectrum with $1 \leqslant f$ due to the divergence of the spectral area.)

Initially, the wavefunction is considered to be in the first two levels (the supposed qubit). We calculate the dephasing rate between these levels (D), the relaxation rates of the second

level (R) and the leakage rate (L) from the qubit. These are respectively found by $|\rho_{12}^{(S)}(t)|$, $\rho_{22}^{(S)}(t)$ and $\mathrm{Tr}_Q\{\tilde{\rho}^{(S)}(t)\}$ where $\mathrm{Tr}_Q\{\cdot\cdot\}$ indicates the trace over the entire RDM excluding the qubit. We first calculate in the next section the Gaussian RDL rates at short times. This is followed by the long time behaviour of the RDL rates which are read from the characteristic exponential tail of the reduced density matrix.

We use a time-iterative algorithm in these calculations. The computational time is a steep function of the product (number of levels $\times$ observational time)^2. For obtaining the Gaussian rates, a short time iteration $t \ll 1/(E_{\max} - E_1)$ is sufficient (here $E_{\max}$ is the highest resonant transition energy allowed in the system). For the exponential tail, the iteration should be continued until typical times of the resonant transitions of which the longest one is $t \simeq 1/\Delta E_{\min}$. Due to the excessive computational time in this regime we limit our current long time calculations to $N \leqslant 5$.

4. The effect of the compensating term $\mathcal{H}_c$

We have shown in the previous section that the compensating term $\mathcal{H}_c$ changes the original system basis by transforming the original dipole matrix elements as given by the relation (7). Therefore it must be ensured that the multilevel effects observed in the solution of (5) are to be interpreted correctly as the genuine property of the multistate dynamics and not merely as a consequence of the renormalized basis. In this section we examine the matrix elements of $\tilde{\varphi}_U(t)$ in the original system basis already defined in equation (1). These are

$$\tilde{\varphi}_U(t)_{nm} = \sum_{r,s} \hat{U}_{nr}(t)\tilde{\varphi}(t)_{rs}\hat{U}^{\dagger}_{sm}(t). \tag{10}$$

For perturbative dipole interaction and at short times the first few terms in the expansion of (10) are important. At the second order

$$\tilde{\varphi}_U(t) \simeq \tilde{\varphi}(t) - \mathrm{i}\int_0^t \mathrm{d}t'[\tilde{\mathcal{H}}_c(t'), \tilde{\varphi}(t)] - \frac{1}{2!}\mathcal{T}\int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2\,[\tilde{\mathcal{H}}_c(t_1), [\tilde{\mathcal{H}}_c(t_2), \tilde{\varphi}(t)]] + \cdots. \tag{11}$$

It is trivially seen that the renormalization of the initial dipole matrix elements in (2) are contributed by the terms in the expansion (11) with odd number of time integrals. The dipole elements not present in the original system, such as the even transitions $\tilde{\varphi}_U(t)_{n,n\pm 2m}$, are created by the terms with even number of integrals. For three- and four-level systems it can be seen that the corrections to all dipole matrix elements start with those terms with at least two time integrals. In the calculation of the short time decoherence rates the effect of the two small parameters, i.e. time and the dipole couplings, are multiplicative. This proves that the compensation term does not create an appreciable renormalization of the system basis in the short time limit. For a system with a small number of levels the smallness of this effect should persist even at longer times. A complete proof of the negligible contribution of the renormalization of $\tilde{\varphi}(t)$ is provided in appendix B.

There are arguments that at long times the effect of the compensating term is also negligible. See for instance [7] for details where the multistate dynamics has been examined within the Bloch–Redfield approach. An alternative proof valid at sufficiently long times is provided in appendix B.

The overall result is that it is possible to ignore the renormalizing effect of the compensating term in our calculations, which we do in the rest of the paper.

5. The short time decoherence

The density matrix usually starts with a Gaussian-like time variation unless the system–noise kernel is abruptly changing at short times, i.e. $t \to t'$. It should be remarked that those processes in the solution of (5) with a finite first derivative surviving in the limit $t \to 0$ are necessarily Markovian with delta-function-like correlations, i.e. $\mathcal{F}(t-t') \propto \delta(t-t')$. On the other hand, most physical noise sources display non-Markovian correlations at short times and the usual practice is to represent them by a power-Gaussian [6, 19] or power-exponential [1, 3]. For instance, the characteristic zero temperature environment for superconducting systems is provided by the fluctuations in the external voltage sources which is an ohmic spectrum at low frequencies with an exponential tail [27]. These non-Markovian type of correlations are usually characterized by a finite environmental equilibration timescale $\Lambda_{\mathrm{B}}^{-1}$. For such non-Markovian processes the time integral in equation (5) vanishes in the limit $to \to t'$. Therefore, the leading term in the short time limit is the second time derivative given by

$$\frac{\mathrm{d}^2}{\mathrm{d}t^2}\tilde{\rho}_{nm}^{(S)}(t=0) = -\sum_{r,s} K_{rs}^{nm}(0,0)\tilde{\rho}_{rs}^{(S)}(t=0) \tag{12}$$

provided at least one of the terms in the summation on the right-hand side is non-zero. It can also be checked by direct calculation that only the even derivatives of the RDM are non-zero at $t = 0$. Note that equation (12) is correct only at $t = 0$ and should not be interpreted as a differential equation at vanishingly short times. The decohering density matrix therefore starts with a time evolution which is Gaussian-like as

$$\tilde{\rho}_{nm}^{(S)}(t) \simeq \sum_{rs}\left[\delta_{nr}\delta_{sm} - \frac{t^2}{2}K_{rs}^{nm}(0,0)\right]\tilde{\rho}_{rs}^{(S)}(0) \tag{13}$$

where the Gaussian decoherence rate $\Lambda_{\mathrm{RDL}}^{(G)}$ enters as a sum over the square roots of the positive eigenvalues of a characteristic operator $\hat{K}(0,0)$ representing the system–noise kernel. In appendix A the positive definiteness of $\hat{K}(0,0)$ and its relation to $K_{rs}^{nm}(0,0)$ are demonstrated. This demonstrates the existence of a Gaussian-like decoherence at short times. $K_{rs}^{nm}(0,0)$ in the right-hand side of (13) is, by equation (6), proportional to the squared dipole coupling constants and $\mathcal{F}(0)$. The latter is, by (8), the total area under the spectral function. Hence, the short time decoherence rate is expected to have contributions not only from the resonant terms but from the entire spectrum as a whole. We thus expect that for all components of the RDM, $\Lambda_{\mathrm{RDL}}^{(G)} \propto (\text{spectral area})^{1/2}$. The Gaussian-like decoherence rates corresponding to the relaxation, dephasing and leakage contributions differ only in their dependence on the sum over the allowed dipole couplings $(\varphi)_{nr}$. Additionally, there is only a dependence on the spectral area with the other spectral parameters, i.e. spectral centre ω_0 and the width ϵ, being ineffective at short times (see figures 1–3).

The Gaussian decoherence rates are easily calculated numerically within a short time interval which corresponds in this work to $0 \leqslant t \leqslant 0.1$ (in units of the inverse absolute energy scale defined in (2)). The numerically found RDL rates are shown for the model system in equation (2) in figure 1(a) for their dependence on the position, and figure 1(b) for their dependence on the shape of the spectrum for three- and five-level systems. Figures 1(a), (b) provide identical decay rates independently of the central frequency ω_0 and the width ϵ for fixed spectral area. These results are already conclusive in the dominance of the off-resonant processes at short times. Before examining the dependence on the spectral area, we examine the effect of the higher levels in the system as shown in figure 2. All levels are coupled by the dipole matrix in equation (2) since we have $R = 10$. Increasing the number of levels increases

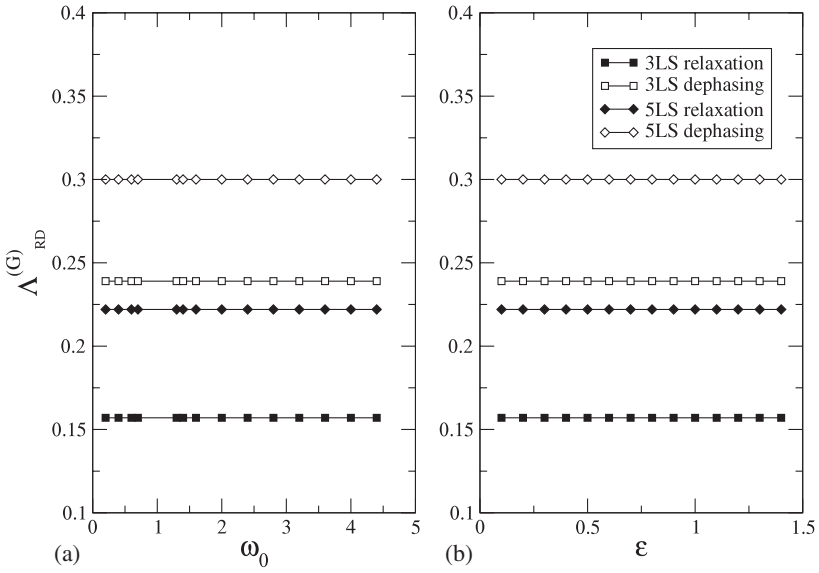

Figure 1. Gaussian RDL rates with respect to (a) central frequency ω_0 (with $A = 1$ and $\epsilon = 0.1$), and (b) spectral width ϵ (with $A = 1$ and $\omega_0 = 2.4$). Note that the results depicted in this figure are independent of the shape of the spectrum.

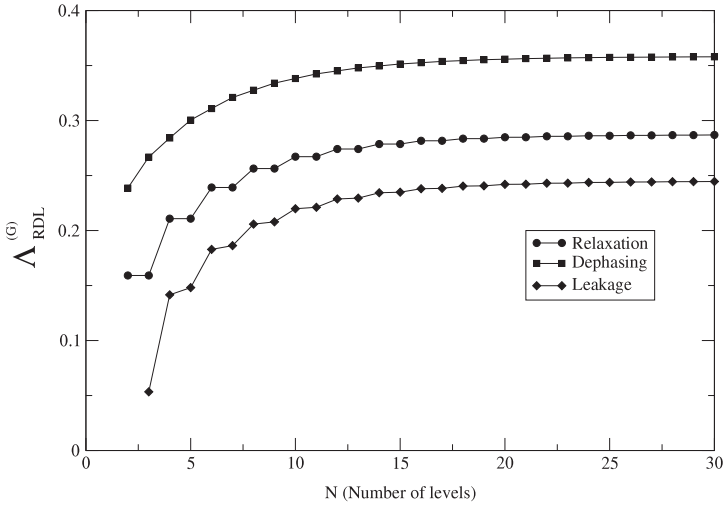

Figure 2. Gaussian RD rates as a function of the number of system levels N. The coupling range is taken to be $R = 10$. The stepwise behaviour in the rates for the first few levels is due to the forbidden (allowed) transitions by the matrix elements in equation (2). Spectral parameters are $A = 1$, $\omega_0 = 1.0$ and $\epsilon = 0.1$.

the decoherence times for $N \leqslant R$ followed by a saturation for $R < N$, which is due to the negligible contribution of the couplings out of the range.

The spectral area A strongly affects the short time decoherence. Finally, in figure 3 we observe that all RDL rates have power law dependence on the spectral area (a log–log plot indicates that $\Lambda^{(G)}_{\mathrm{RDL}} \sim \sqrt{A}$), verifying once more the dominating role of the off-resonant transitions between the levels.

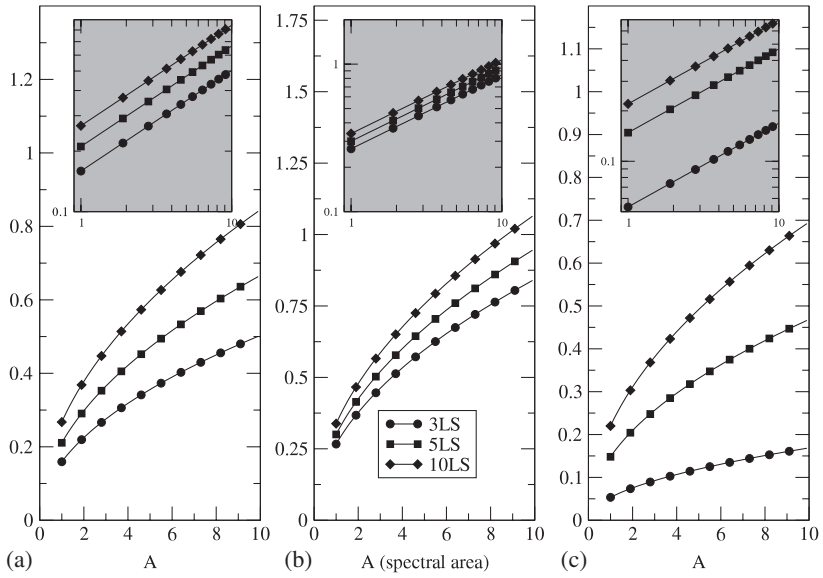

Figure 3. Dependence of the short time Gaussian rates for (a) R, (b) D, (c) L on the spectral area. On a log–log scale, as shown in the insets, the dependence is $\sim\sqrt{A}$.

From figures 1–3, it is necessary to conclude that the short time decoherence rates are unconventionally non-exponential, characterized by the short time non-Markovian correlations in the spectrum. However, the reader can always object to this conclusion and say that the decohering two-level-like system may also quickly leave the 'Gaussian-like' regime and rapidly move towards to the region where the two-level approximation is manifested. In answering such a question, all timescales, including a characteristic crossover time between the Gaussian-like and the exponential like regimes, should be compared. An answer to this crucial point will be provided in section 8.

6. The long time decoherence

We now examine the decoherence rates of an MLS in the exponential regime. For non-Markovian systems the exponential behaviour can be reached after a significant decoherence has already taken place at short and intermediate times. Here we should also remark that although a Lorentzian (or Drude) type noise spectrum has Markovian correlations in sufficiently long observational times, there are also physical spectra without a Markovian limit. For instance, the widely used Rubin model which represents a bosonic environment with Einstein phonons has power law dependence of the noise correlations at long times [20]. This indicates that the long time decoherence, in contrast to the short time decoherence, depends on the type of physical processes generating the spectrum.

First, we depict the dependence of the exponential rates on the spectral centre ω_0 in figure 4. The marked contrast to figure 1 is clearly seen. The decoherence rates in this regime are identified by the dominance of the resonant transitions. These rates tend to increase significantly at the resonance frequencies, i.e. the system experiences strong long time decoherence only near the resonant frequencies. Therefore there is no surprise in this regime, which the majority of the literature on decoherence considers either by the Fermi golden rule, or equivalently, by directly looking at the Markovian limit (Bloch–Redfield and

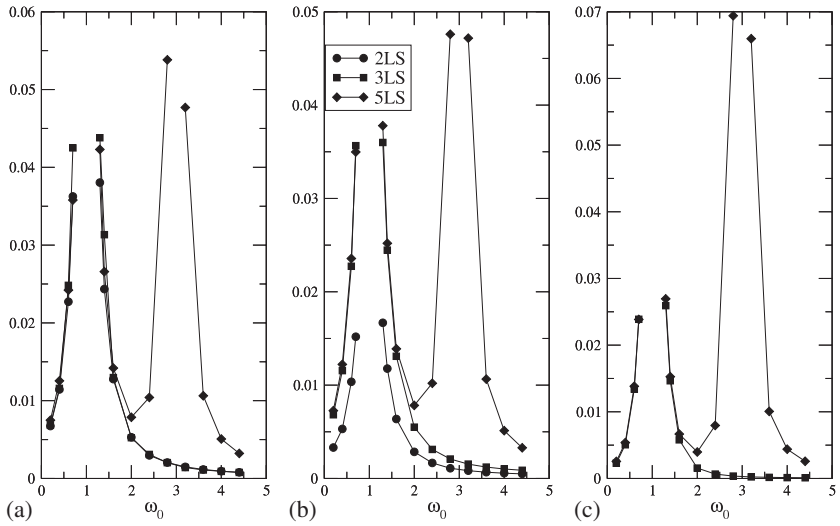

Figure 4. Long time exponential rates for (a) R, (b) D, (c) L mechanisms as a function of the spectrum centre ω_0. The legends in (b) are applied to all figures. Note that for 2LS $\omega_0^{(2LS)} = 1$, $\omega_0^{(3LS)} = 1, 2$ and $\omega_0^{(5LS)} = 1, 2, 3, 4$. The $\omega_0 = 2$ and the $\omega_0 = 4$ peaks are unobservable due to the transition selection rules in equation (2). The other parameters in these results are $A = 1$ and $\epsilon = 0.1$. As ω_0 gets closer to an allowed resonant transition the rates rapidly increase. In the close vicinity of a resonance we observe numerical instabilities. These unstable artefacts are avoided in the figures.

Lindblad formalisms) of the density matrix. The results obtained here confirm that the major contribution to long time decoherence originates from the resonant transitions induced by the environment. In an MLS the exponential decoherence rates are contributed by all resonant frequencies available in the environmental spectrum. For a 5LS, for instance, the frequencies $\omega_0 = 2$ and 4 are not observable as odd–odd and even–even parity transitions are forbidden. But the effect of the second resonant frequency $\omega_0 = 3$ is significant, as shown in figure 4. Dephasing and leakage rates are also similar, which are also depicted in figures 4(b) and (c).

7. Leakage effects

The leakage is a characteristic multilevel effect which can be deduced by looking at the deviations in the response of the system from a two-level one under a controllable coupling to an external field. Here we report three recent experiments where anomalies have been reported in the Rabi oscillation dynamics indicating a clear breakdown of the two-level approximation and strong *short time leakage* effects. In the first experiment by Zrenner *et al* [21], single self-assembled excitonic q-dots are used to create excitons by a strong Rabi field which are then tunnelled out and transformed into a photocurrent. The anomaly appears in the damping of the Rabi induced oscillations in the pulse averaged photocurrent as the area under the Rabi pulse is increased. This effect within the fixed time of the short 1 ps pulse (dephasing and relaxation times are reported to be approximately 500 ps) could not be observed in a purely two-level system as it would violate the fundamental principle of unitarity. The pulse width is shorter than the decoherence time scale by three orders of magnitude and it is clear that the observed damping does not originate from decoherence. The short timescale of the effect indicates the influence of the higher excitonic states. In a simple approach it has been shown

that the damping of the oscillations as the intensity of the short pulse is increased is due to the off-resonant leakage into biexcitonic levels [22]. Taking [22] as the basis of a physical model for this experiment, we show that the results are the manifestation of short time non-resonant processes.

To model the experiment, a Rabi coupling term is added to equation (1) as

$$\mathcal{H}_{\mathrm{R}} = \sum_{nr=1}^{N} \varphi_{nr} |n\rangle\langle r| (\alpha(t) \mathrm{e}^{-\mathrm{i}\Omega_{\mathrm{R}} t} + \mathrm{h.c.}) \tag{14}$$

where $\alpha(t)$ and Ω_{R} are the complex pulse amplitude and the frequency of the coherent Rabi field respectively. We use rectangular and real pulse

$$\alpha(t) = \alpha \begin{cases} 1 & \text{for } t \leqslant t_{\mathrm{P}} \\ 0 & \text{for } t_{\mathrm{P}} < t, \end{cases} \qquad 0 \leqslant t. \tag{15}$$

To compare with the experimental results we consider $t_{\mathrm{P}} = 1$ in units of the fundamental energy scale in (1) which physically corresponds to Zrenner *et al*'s experimental range $t_{\mathrm{P}} \simeq 1$ ps when $E_2 - E_1 \simeq 1$ meV. The master equation for the RDM including this coupling with the Rabi field acquires the additional term, yielding

$$\frac{\mathrm{d}}{\mathrm{d}t} \tilde{\rho}^{(S)}_{nm}(t) = \mathrm{i}\langle n|[\tilde{\mathcal{H}}_{\mathrm{R}}(t), \tilde{\rho}^{(S)}(t)]|m\rangle - \int_0^t \mathrm{d}t' \sum_{r,s} K^{nm}_{rs}(t, t') \tilde{\rho}^{(S)}_{rs}(t'). \tag{16}$$

This is solved below numerically for $N = 3$ considering the model multilevel coupling (2). Since for a few ps pulse the decoherence effects can be eliminated due to the much longer decoherence times, we totally neglect the environment in (16) for the response of the MLS within a single pulse width. Then the time dependence becomes a function of the total pulse area αt. The result is then averaged over the pulse as in the experiment. In figure 5 the pulse-averaged occupation of this non-resonantly coupled third level (i.e. $\langle \rho_{33}(t) \rangle_{\mathrm{pulse}} = 1/t_{\mathrm{P}} \int_0^{t_{\mathrm{P}}} \mathrm{d}t\, \rho_{33}(t)$) is shown as a function of the pulse area when the Rabi field is resonant with the first two levels for three different third-level energies. In the figure, all three curves indicate pulse-averaged third-level occupation, i.e. $\langle \rho_{33}(t) \rangle_{\mathrm{pulse}}$, for the third-level energies $E_3 = 2, 7.5, 10$. In the first one, E_3 is degenerate with $E_2 = 2$, and, for the last one, $E_3 = 10$ is supposedly close to an ideal two-level system $(E_2 - E_1) \ll (E_3 - E_2)$. The occupation of the third level is clearly observed to be largely unaffected by the large range of the energies for the third level. Another point is that the peak position occurs at $t_{\mathrm{P}} \leqslant 1/\alpha$. Using (2), this implies $t_{\mathrm{P}} \leqslant \varphi_{12} T_{\mathrm{R}}/(2\pi)$, where T_{R} is the Rabi oscillation period; therefore $t_{\mathrm{P}} \simeq 0.1 T_{\mathrm{R}}$, concluding that the third level is already occupied maximally before the completion of a single Rabi period. This short time effect is counterintuitive from the traditional way of thinking in terms of the long time resonant transitions. It must be remarked that this is an exact result. It appears that a multilevelled system decides to act like so at very short times in comparison with typical resonant timescales. Thus figure 5, in confirmation of the earlier theoretical calculations [22], manifests the effect of the strong influence of the non-resonant processes on leakage. A second experiment supporting strong non-resonant leakage at short times has been recently made on semiconductor NMR devices by Yusa *et al* [23] where off-resonant multiple quantum coherence effects between levels separated by more than one quantum of nuclear spin quantum number were observed. This experiment is the four-level version of the quantum dot experiment by Zrenner *et al* done with stable nuclei with total spin 3/2 in which the short time multilevel dynamics can be probed. For the spin 3/2 system, As^{75}, Ga^{69} or Ga^{71} were used, which are all four-fold degenerate spin systems conforming to the nuclear shell model but additionally split by an electric-quadrupolar crystal field as well as a much larger external Zeeman coupling by a controllable DC magnetic field at 5 T. A weak RF-magnetic

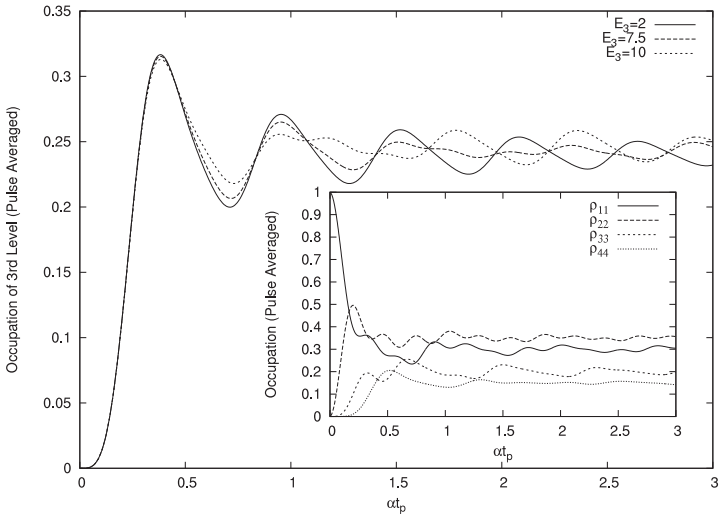

Figure 5. The time average over the Rabi pulse of the third-level occupancy is shown as a function of αt_{P}. Three curves refer to three different third-level resonant energy $E_3 = 10, 7.5, 2$ with $E_2 = 2$ and $E_1 = 1$. The Rabi field is in resonance with the first two levels. In the inset the four level occupancies are shown for equally spaced levels with a Rabi field resonantly coupling the neighbouring levels.

field of strength 10^{-4} T with a tunable frequency and pulse width 0.1–1 ms are also applied perpendicular to the DC field. We used the model Rabi pulse in equation (14) to regenerate the multilevel dynamics for $N = 4$. The pulse-averaged occupation of each of the four levels is shown in the inset of figure 5. The wavefunction is initially confined at the ground level. The first two levels are already nearly fully occupied for $t_{\mathrm{P}} \simeq 0.2/\alpha$ when the third level is 10% full and the fourth level is empty. Within another short time, i.e. at $t_{\mathrm{P}} \simeq 0.5/\alpha$, the third and the fourth levels are already saturated at about 20%.

The third experiment we report is by Claudon *et al* [24], done on the DC-SQUID system in which they found strong deviation from the strict linear law between the Rabi frequency and the field strength. This deviation is explained by the increasing number of multilevels participating in the dynamics by the increasing field strength at fixed microwave frequency.

In this section, we have examined two experiments in which the effect of the multistate dynamics is clearly manifested. The results indicate that leakage is a short time effect mainly induced by non-resonant couplings between the levels. A similar experiment for a decohering MLS with a clear indication of the multilevel dynamics with short time effects so clearly distinguished is much harder to accomplish. The results in this section are, however, in support of the previous sections in that leakage cannot be ignored at short timescales.

8. The limitations of the Born–Oppenheimer approximation

The essence of the Born–Oppenheimer approximation is in neglecting the back influence of the quantum system on the environment. At sufficiently short observational times, in contrast with the typical environmental equilibration time, this approximation holds well and our results within those observational times are reliable. The crucial question is whether the BOA also holds at intermediate as well as exponential regimes, so that the results of section 4 can also be claimed to be correct.

For a rigorous check of the BOA one has to solve the equations without this approximation and compare the two results with and without the approximation. This can be done by using other techniques, for instance like the Nakajima–Zwanzig projection operator method or time convolutionless projection operator (TCL) formalism [25] by calculating each perturbation series independently. Another alternative is analytic real time renormalization group calculations [26]. However, these analytic calculations are practically applicable only to two-level systems.

There is, nevertheless, a very intuitively simple method for estimating the range of the BOA. For a multilevelled system with a large number of resonant frequencies, the smallest resonance energy defines the onset of the exponential behaviour. In our case, this energy scale is $\Lambda_c \simeq \Delta E = 1$ and for times $1 = 1/\Lambda_c \ll t$ the exponential behaviour should be manifest. The BOA continues to hold in the exponential regime if the Born–Oppenheimer time (a typical reaction–equilibration timescale of the environment which is of the order of the inverse width of the spectrum $1/\epsilon$) is much larger than the transition time $1/\Lambda_c$ to the exponential regime.

An estimate for the critical region can be made by looking at the crossover between the short and the long time behaviour. Assume that, after some crossover time t_c, the Gaussian-like amplitudes are of the same order of magnitude as the exponential ones. This amounts to

$$e^{-(\Lambda_c^{-1}+t_c)\Lambda^{(E)}} \simeq e^{-(\Lambda^{(G)})^2 t_c^2} \tag{17}$$

Solving this equation for the exponents, one obtains for t_c

$$t_c = \frac{\Lambda^{(E)}}{2\Lambda^{(G)^2}}\left[1 + \sqrt{1 + 4\Lambda^{(G)^2}/(\Lambda^{(E)}\Lambda_c)}\right] \tag{18}$$

as an estimate for the crossover time between the intermediate and the exponential regions. Crudely speaking, for $t \ll t_c$ the Gaussian-like and for $t_c \ll t$ the exponential behaviour are manifest. However, for the BOA to be accurately valid in the exponential regime, an additional constraint has to be satisfied: the Born equilibration time $1/\epsilon$ has to be much longer than the crossover time t_c to the exponential behaviour. Thus

$$t_c \ll \frac{1}{\epsilon}. \tag{19}$$

By defining a test parameter $\mu = t_c\epsilon$ and using equation (19) we find that the Born approximation holds well in the exponential regime if $\mu \ll 1$ and it is unreliably poor for $1 \ll \mu$. Within the ranges of parameters investigated in this paper, we calculated the μ parameter from the corresponding RDL rates. The results are tabulated in table 1 for 2LS, 3LS and 5LS. Thus the μ parameter is influenced by two independent timescales, that is the short and the long interaction timescales. In the table, there are certain regions where the BOA is threatened. For instance, close to a resonant frequency μ increases towards unity and for the opposite case of off-resonant frequencies, $\mu \ll 1$. The basic mechanism is that the contribution of the long time resonant coupling tends to increase the exponential rates. This increases μ. In this regime a possibility arises in that the environmental back reaction takes place before the onset of the exponential behaviour. On the other hand, and independently from the decohering system, the contribution of the off-resonant parts of the spectrum is to increase the Gaussian rates. This in turn decreases the μ parameter by the relations (18) and (19).

We also examined the BOA as a function of the number of levels. Considering the behaviour of μ with respect to N, $\Lambda^{(G)}$ and $\Lambda^{(E)}$ are both monotonically increasing functions with N for $N \leqslant R$. Therefore, using (18), and the fact that ϵ is independent of N, the overall dependence is

$$\mu \sim 1/N. \tag{20}$$

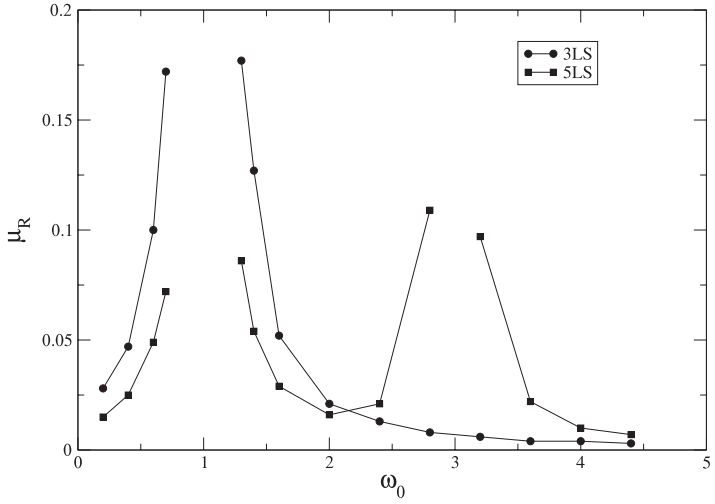

Figure 6. μ_R for 3LS and 5LS as a function of spectrum centre ω_0. The figure is an illustration of the data in the table 1 and clearly shows the weakening of the Born approximation near the resonant transitions.

Table 1. The μ parameter of the RDL processes for 2LS, 3LS and 5LS against varying ω_0. The other spectral parameters are $\epsilon = 0.1$ and $A = 1$.

	2LS		3LS			5LS		
ω_0	μ_R	μ_D	μ_R	μ_D	μ_L	μ_R	μ_D	μ_L
0.2	0.027	0.006	0.028	0.010	0.080	0.015	0.008	0.012
0.4	0.046	0.009	0.047	0.016	0.177	0.025	0.014	0.024
0.6	0.092	0.018	0.100	0.032	0.470	0.049	0.026	0.063
0.7	0.147	0.027	0.172	0.050	0.837	0.072	0.039	0.109
1.3	0.154	0.029	0.177	0.051	0.909	0.086	0.042	0.123
1.4	0.099	0.021	0.127	0.034	0.514	0.054	0.028	0.070
1.6	0.052	0.011	0.052	0.018	0.203	0.029	0.015	0.030
2.0	0.021	0.005	0.021	0.008	0.055	0.016	0.009	0.018
2.4	0.012	0.003	0.013	0.004	0.023	0.021	0.011	0.036
2.8	0.008	0.003	0.008	0.003	0.013	0.109	0.053	0.316
3.2	0.006	0.002	0.006	0.002	0.008	0.097	0.052	0.301
3.6	0.005	0.001	0.004	0.002	0.006	0.022	0.012	0.046
4.0	0.004	0.001	0.004	0.001	0.005	0.010	0.006	0.020
4.4	0.003	0.001	0.003	0.001	0.004	0.007	0.004	0.012

Hence, as N increases the BOA becomes increasingly more reliable and least reliable results are obtained for two-level systems. This behaviour is illustrated in figure 6 for $N \leqslant 5$ and can be studied for larger N values provided sufficient computing power. An important corollary of equation (20) is that the crossover time has a scaling with the inverse number of decoherence channels (number of levels). Consider now a two-level system and a multilevelled system interacting with the same environment. The fact that $t_c \sim 1/N$ implies that the initial short time decoherence is expected to take a longer time in the smaller system. Although one may obtain a few exceptions to this result using uncommon environmental spectra, this result should hold to be generally true.

9. Conclusions

It is shown that for the most conventionally used system–environment couplings of linear coordinate type, decoherence has different characteristic signatures in different time domains, and a short time decoherence exists independently and it may even be more important than the Markovian decoherence.

At short times, the entire interacting environmental spectrum affects the decoherence rates, indicating the dominating role of the off-resonant transitions. The short time rates are largely ignorant about the shape and the centre of the spectrum, whereas the total area under the spectrum and the number of interacting levels play an important role.

It is confirmed that the long time behaviour shows a complementary picture to that at short times. The initial decoherence started by the off-resonant transitions is gradually taken over by the resonant ones. The suppression of the transitions corresponding to the resonant frequencies forbidden by the selection rules may be seen as a confirmation of the use of the Fermi golden rule if the coupling is sufficiently weak.

Motivated by recent experiments probing the Rabi dynamics of multilevelled systems, we showed that the phenomenon of leakage is observable in the pulse-averaged Rabi oscillations. We demonstrated that these effects are created by the off-resonant excitations.

We finally examined the Born–Oppenheimer approximation, which is widely used in the master equation formalism. For the resonant parts of the spectrum, the BOA is shown to be poor, whereas for the off-resonant contributions the approximation holds more accurately. In the literature, the BOA is often used without justification in the long time regime and assuming Markovian effects. Our work indicates that the reliability of this approximation is affected not only by the long time but also the short time dynamics, which should also be examined.

Overall, we have shown by counter-examples that the two-level approximation is not reliable in issues related to decoherence and the conditions assumed by it are not sufficiently general. Our calculations comprise one of the few works where the two-level approximation is explicitly checked by solving the master equation of a system with variable number of levels interacting with an environment.

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Appendix A. Proof of the reality and the positivity of Λ^G

Let us define a Hermitian dipole transition operator $\hat{\tilde{\varphi}}_t$ such that

$$(\hat{\tilde{\varphi}}_t)_{jk} = \langle j|\hat{\tilde{\varphi}}_t|k\rangle = \mathrm{e}^{\mathrm{i}(E_j - E_k)t}\langle j|\hat{\tilde{\varphi}}_0|k\rangle. \tag{21}$$

By using (21) it is trivially checked that the system–noise kernel can be written at $t = t' = 0$ as a non-negative real operator

$$\hat{K}(0,0) = \mathcal{F}(0)|\hat{\tilde{\varphi}}_0 \otimes \mathbf{I} - \mathbf{I} \otimes \hat{\tilde{\varphi}}^{\mathrm{T}}{}_0|^2 \tag{22}$$

where $\mathcal{F}(0) = A$ is the spectral area, which is real by definition and T denotes transpose. Hence equation (22) describes a real non-negative operator in the two-folded Hilbert space $\{|n\rangle \times |m\rangle$

where $n, m = 1, N$. By using equation (22), it can be shown easily that the system–noise kernel in equation (6) is equivalently written as

$$K_{rs}^{nm}(0,0) = \langle n| \times \langle m|\hat{K}(0,0)|r\rangle \times |s\rangle. \tag{23}$$

Defining a vector $|V\rangle$ such that

$$\hat{\tilde{\rho}}_{nm}^{(S)}(0) = \langle n| \times \langle m|V\rangle \tag{24}$$

we can write (12) in the implicit operator form as

$$|\ddot{V}\rangle = -\hat{K}(0,0)|V\rangle \tag{25}$$

where the left-hand side clearly implies the second derivative of the density matrix at $t = 0$. Since $\hat{K}(0,0)$ is real and non-negative, its eigenvalues are real and non-negative; hence a real Gaussian-like short decoherence timescale exists.

Another analytic exact result is that the Gaussian-like short decoherence timescales are proportional to the square root of the spectral area. This follows from the fact that the matrix elements of $\hat{K}(0,0)$ have, in common, a term proportional to $\mathcal{F}(0)$ which is just the spectral area by equation (8). The solution of equation (25) then indicates that the short time decoherence rates scale with the square root of the spectral are (see figure 3 for the numerical manifestation of this result).

Appendix B. Summing up the effects of the compensating term

It is possible to sum up the infinite series generated in equation (11) to all orders. The general term in the series is given at the operator level by

$$\tilde{\varphi}_U(t) = \tilde{\varphi}(t) + \mathcal{T}\sum_{n=1}^{\infty}\frac{(-\mathrm{i}A)^n}{n!}\int_0^t \mathrm{d}t_1 \ldots \int_0^t \mathrm{d}t_n[\tilde{\varphi}^2(t_n), \ldots, [\tilde{\varphi}^2(t_1), \tilde{\varphi}(t)]\ldots] \tag{26}$$

where we used $\tilde{\mathcal{H}}_c(t) = A\tilde{\varphi}^2(t)$ explicitly. The last term includes an n-fold commutator and $\mathcal{T}$ is the time ordering operator. Since we are interested in the short time part of the proof, we consider the observation time t in the limit $t \to 0$. Consider now one of the $n!$ possibilities of the order of the internal times $t_1, \ldots, t_n \leqslant t$ such as $t_n \leqslant t_{n-1} \leqslant \cdots t_2 \leqslant t_1 \leqslant t$. It is clear that

$$\int_0^t \mathrm{d}t_1 \ldots \int_0^t \mathrm{d}t_n 1 \simeq \frac{t^n}{n!}. \tag{27}$$

Hence for sufficiently short times,

$$(\tilde{\varphi}_U(t)) = \mathrm{e}^{-\mathrm{i}At\tilde{\varphi}^2(0)}\varphi(t)\mathrm{e}^{\mathrm{i}At\tilde{\varphi}^2(0)} \tag{28}$$

which proves that all corrections to the system dipole transitions occur in powers of the square of the transition matrix elements. It can be seen easily that equation (28) becomes an appreciable renormalization only when a sufficiently large number of levels are involved in the non-resonant transitions. For instance, using equation (2) it can be shown that, for a system with $N \leqslant 3$, equation (28) yields no renormalization of the dipole matrix. Hence the effect of the compensating term vanishes for $N \leqslant 3$ and in the short time limit.

To examine the effect of the compensation at long times, we consider the limit $t \to \infty$ in (11) as

$$\tilde{\varphi}_U(t) \simeq \tilde{\varphi}(t) - \mathrm{i}\int_0^{\infty}\mathrm{d}t'[\tilde{\mathcal{H}}_c(t'), \tilde{\varphi}(t)] - \frac{1}{2!}\mathcal{T}\int_0^{\infty}\mathrm{d}t_1\int_0^{t_1}\mathrm{d}t_2\,[\tilde{\mathcal{H}}_c(t_1), [\tilde{\mathcal{H}}_c(t_2), \tilde{\varphi}(t)]] + \cdots. \tag{29}$$

The first term does not include a time ordering and therefore can be easily calculated:

$$\text{First term in}(29) \propto -\mathrm{i}A \sum_{s}[\delta(E_n - E_s)\tilde{\varphi}^2(0)_{ns}\tilde{\varphi}(t)_{s,m} - \delta(E_s - E_m)\tilde{\varphi}(t)_{n,s}\tilde{\varphi}^2(0)_{s,m}]. \tag{30}$$

In the limit $t \to \infty$, $\tilde{\varphi}(t)_{n,s}$ is a sharply peaked function for $n = s$, otherwise highly rapidly oscillating. We approximate this term by $\delta_{n,s}$ at $t \to \infty$ and with this approximation it vanishes. Likewise, the next term includes a time ordering which can also be calculated exactly. In this term, the non-zero contributions are restricted to even more energetic conditions and the second term also vanishes in the limit $t \to \infty$. Moreover, all the terms have, in common, different matrix elements of $\tilde{\varphi}(t)$. In the $t \to \infty$ limit these contributions average out anything slower to zero, making it possible to ignore them all.

The overall result is that it is possible to ignore the renormalizing effect of the compensating term in our calculations.

References

[1] Leggett A J, Chakravarty S, Dorsey A T, Matthew P A F, Garg A and Zwerger W 1987 *Rev. Mod. Phys.* **59** 1
Loss D and DiVincenzo D P 2003 Exact Born approximation for the spin-boson model *Preprint* cond-mat/0304118
Vorrath T, Brandes T and Kramer B 2001 Dynamics of a large spin-boson system in the strong coupling regime *Preprint* cond-mat/0111220
Grifoni M, Paladino E and Weiss U 1999 *Eur. Phys. J* B **10** 719
Leggett A J 1987 Quantum mechanics at the macroscopic level *Chance and Matter* (*Les Houches Session XLVI*) ed J Vannimenus and R Stora (Amsterdam: Elsevier) p 1986

[2] Anastopoulos C and Hu B L 2000 *Phys. Rev.* A **62** 033821

[3] Dube M and Stamp P C E 2001 *Preprint* cond-mat/0102156
Stamp P C E and Tupitsyn I S 2004 *Chem. Phys.* **296** 281
Fedichkin L, Fedorov A and Privman V 2003 *Proc. SPIE* **5105** 243

[4] Caldeira A O and Leggett A J 1981 *Phys. Rev. Lett.* **46** 211
Caldeira A O and Leggett A J 1983 *Ann. Phys.* **149** 374

[5] Leggett A J and Anupam G 1985 *Phys. Rev. Lett.* **54** 857

[6] Hakioğlu T and Savran K 2005 *Phys. Rev.* B **71** 115115

[7] Burkard G, Koch R H and Di Vincenzo D P 2004 *Phys. Rev.* B **69** 064503

[8] Bloch F 1957 *Phys. Rev.* **105** 1206
Redfield A G 1957 *IBM J. Res. Dev.* **1** 19

[9] Lindblad G 1976 *Commun. Math. Phys.* **48** 119

[10] DiVincenzo D P and Loss D 2005 *Phys. Rev.* B **71** 035318

[11] Ford G W and O'Connell R F 2000 *Opt. Commun.* **179** 451

[12] Voss R V and Webb R A 1981 *Phys. Rev. Lett.* **47** 265
Martinis J M, Devoret M H and Clarke J 1985 *Phys. Rev. Lett.* **89** 207601
Friedman J R, Patel V, Chen W and Lukens J E 2000 *Nature* **406** 43

[13] Natelson D, Willett R L, West K W and Pfeiffer L N 2001 *Phys. Rev. Lett.* **86** 1821

[14] Cedraschi P, Ponomarenko V V and Büttiker M 2000 *Phys. Rev. Lett.* **84** 346

[15] Lin J J and Giordano N 1987 *Phys. Rev.* B **35** 1071
Pookr D M *et al* 1989 *J. Phys.: Condens. Matter* **1** 3289

[16] Mohanty P, Jariwala E M Q and Webb R A 1997 *Phys. Rev. Lett.* **78** 3366
Mohanty P and Webb R A 1997 *Phys. Rev.* B **55** R13452
Mohanty P and Webb R A 2003 *Phys. Rev. Lett.* **91** 066604

[17] Hakioğlu T, Anderson J and Wellstood F 2002 *Phys. Rev.* B **66** 115324
Everitt M J, Clark T D, Stiffell P, Prance H, Prance R J, Vourdas A and Ralph J F 2001 *Phys. Rev.* B **64** 184512

[18] Born M and Oppenheimer J R 1927 *Ann. Phys.* **84** 457
Bohm A and Loewe M 2001 *Quantum Mechanics* (Berlin: Springer)

[19] Hu B L, Paz J P and Zhang Y 1992 *Phys. Rev.* D **45** 2843

[20] Weiss U 1999 *Quantum Dissipative Systems* (Singapore: World Scientific)

[21] Zrenner A, Beham E, Stufler S, Findeis F, Bichler M and Abstreiter G 2002 *Nature* **418** 612
[22] Villas-Boas J M, Ulloa S E and Govorov A O 2005 *Phys. Rev. Lett.* **94** 057404
Villas-Boas J M, Govorov A O and Ulloa S E 2004 *Phys. Rev.* B **69** 125342
[23] Yusa G, Muraki K, Takashina K, Hashimoto K and Hirayama Y 2005 *Nature* **434** 1001
[24] Claudon J, Balestro F, Hekking FW J and Buisson O 2004 *Phys. Rev. Lett.* **93** 187003
[25] Breuer H P and Petruccione F 2002 *The Theory of Open Quantum Systems* (Oxford: Oxford University Press)
[26] Keil M and Schoeller H 2001 *Phys. Rev.* B **63** 180302
[27] Snirman A, Schön G and Hermon Z 1997 *Phys. Rev. Lett.* **79** 2371