Dynamics of large anisotropic spin in a sub-ohmic dissipative environment close to a quantum-phase transition

Frithjof B Anders
Institut für Theoretische Physik, Universität Bremen, PO Box 330 440, D-28334 Bremen, Germany
E-mail: anders@itp.uni-bremen.de

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Abstract. We investigate the dynamics of a large anisotropic spin whose easy-axis component is coupled to a bosonic bath with a spectral function $J(\omega) \propto \omega^s$. Such a spin complex might be realized in a single-molecular magnet. Using the non-perturbative renormalization group, we calculate the line of quantum-phase transitions (QPTs) in the sub-ohmic regime ($s < 1$). These QPTs only occur for integer spin $J$. For half-integer $J$, the low temperature fixed-point is identical to the fixed-point of the spin-boson model without quantum tunneling between the two levels. Short-time coherent oscillations in the spin decay prevail even into the localized phase in the sub-ohmic regime. The influence of the reorganization energy and the recurrence time on the decoherence in the absence of quantum tunneling is discussed.
1. Introduction

Understanding the influence of the environment on the non-equilibrium dynamics of quantum systems remains one of the challenging questions of theoretical physics. A finite number of quantum mechanical degrees of freedom, a large spin or a qubit interacting with an infinitely large bath of non-interacting bosons with a continuous energy spectrum represents a typical class of model examples for such systems. Its simplest version, the spin-boson model (SBM) \(^1\), has contributed tremendously to our understanding of dissipation in quantum systems \(^2\).

Single-molecular magnets (SMMs) based on Mn\(_{12}\)-acetate or Fe\(_8\)-complexes behave as large single spins at low temperatures. The magnetic anisotropy of the molecules prevent a simple relaxation of the spin. The resulting hysteretic behavior in the magnetization has been subject to intensive experimental \(^3\)–\(^6\) and theoretical investigations \(^7\)–\(^10\) (see \(^5\) for an overview). Using Wilson’s numerical renormalization group (NRG) method \(^11\),\(^12\), it was shown \(^13\) that the combination of quantum tunneling and antiferromagnetic coupling to a metallic substrate induces a Kondo effect in such a SMM. The recently developed time-dependent NRG (TD-NRG) method \(^14\),\(^15\) has been employed to analyze the real-time dynamics of such a large spin \(^16\).

In this paper, we will investigate the equilibrium and non-equilibrium spin dynamics of an anisotropic spin coupled to a dissipative bosonic bath. Such a spin complex might be realized in a SMM in which the physical properties depend on the type of coupling to the environment. Previously, detailed expansion of the spin-lattice relaxation \(^17\) in powers of the local spin operator have been considered to estimate the spin-relaxation rates \(^8\),\(^17\) by Fermi’s golden-rule arguments. Vorrath and Brandes have reduced the spin–bath coupling to a linear term proportional to the \(S_z\) operator \(^10\). A Weisskopf–Wigner coupling to the environment would describe the interaction with a fluctuating quantized magnetic field. This might be relevant for recent dephasing experiments \(^6\) in SMM.

Here, we will restrict ourselves to diagonally coupled SBM interaction \(^1\),\(^10\). While phase transitions are absent in a model with Weisskopf–Wigner coupling, the observation of...
a line of quantum-phase transitions (QPTs) in the sub-ohmic regime of the SBM has caused some attention [12, 18, 19]. It was a great surprise that coherent oscillations of the local spin have been found at coupling strength at the QPT and in the localized phase [20]. This apparently counterintuitive result indicates clearly that the knowledge of equilibrium properties is insufficient to understand real-time dynamics of a complex coupled system.

In this paper, we allow for the coupling of the local spin to a bath with internal structure. To illustrate the effect of some high-energy bath features on the real time dynamics, we add an additional delta-function at $\omega_0$ with spectral weight $M^2$ to the usual bath spectral function $J(\omega)$ defined below.

The real-time dynamics in the SBM is usually investigated using the blip approximation [1, 21] or by Bloch–Redfield type of approaches [22]. Recently, it has been demonstrated that the non-perturbative NRG [11, 12, 18, 19] and its extension to non-equilibrium dynamics, the TD-NRG [14, 15], is particularly suitable to access the quantum-critical region of the SBM in a sub-ohmic environment [18, 19] and is able to describe the real-time dynamics on short timescales as well as exponentially long timescales [20]. It has been shown [18, 19] that the critical coupling at which a localization–delocalization transition occurs [1] vanishes in the SBM with the bath exponent $s$ in the deep sub-ohmic regime. Therefore, a rather weak spin–bath coupling constant moves the system in the vicinity of the QPT which is not accessible to perturbative methods.

In order to gain a better understanding of the non-equilibrium dynamics, we also present the equilibrium properties of the model. We will map out the quantum-critical line separating a delocalized phase in weak coupling and a local phase at strong coupling to the dissipative sub-ohmic bath. The continuous QPTs found for $0 < s < 1$ and integer spin $J$ resemble the phase diagram previously reported for the SBM by Bulla et al [18]. No phase transition was found for half-integer spin $J$. Information on the equilibrium dynamics is provided by the equilibrium spin correlation function $C(\omega)$. The non-equilibrium spin dynamics of an anisotropic spin complex coupled to a dissipative bosonic bath is investigated in response to a sudden change of magnetic field close to QPTs.

2. Modeling of a large spin in a dissipative environment

The local Hamiltonian modeling SMMs [5]

$$\mathcal{H} = \mathcal{H}_{\text{loc}} + \mathcal{H}_1 + \mathcal{H}_{\text{bath}}$$

(1)

consists of a single large spin of size $J$ subject to an easy-axis anisotropy energy $A$ and quantum-tunneling terms [5, 13] $B_{2n}$, which induce transitions between the eigenstates of $S_z$:

$$\mathcal{H}_{\text{loc}} = -AS_z^2 + \sum_{n=1,2} B_{2n} (S_{-}^{2n} + S_{+}^{2n}) .$$

(2)

These quantum-tunneling terms stem from a reduction of the $U(1)$ easy-axis spin symmetry to the discrete symmetry group of the SMM molecule. Assuming $A \gg |B_{2n}|$ and a positive $A$, the energies of states $|J_z\rangle$ are located on an inverted energy parabola. As depicted in figure 1, there is a fundamental difference between integer and half-integer values of the spin [5, 13]: the two ground states $|\pm J\rangle$ are connected via the quantum-tunneling terms $B_{2n}$ only for integer values of $J$, whereas for half-integer values the ground states remain disconnected. Without further transition mechanism, a half-integer anisotropic spin cannot relax into the thermodynamic ground state after switching off the spin-polarizing external magnetic field.

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We use a generalization [10] of the SBM

\[ H_1 = S_z \sum_q \lambda_q \left( b_q + b_{-q}^\dagger \right), \]

\[ H_{\text{bath}} = \sum_q \omega_q b_q^\dagger b_q \]

(3) (4)

to describe the coupling to a dissipative bath. Here, \( b_q^\dagger \) creates a boson in the mode \( q \) with energy \( \omega_q \). The coupling between spin and bosonic bath is completely specified by the bath spectral function [1]

\[ J(\omega) = \pi \sum_q \lambda_q^2 \delta(\omega - \omega_q). \]

(5)

The asymptotic low-temperature behavior is determined by the low-energy part of the spectrum. Discarding high-energy details, the standard parametrization is

\[ J(\omega) = 2\pi \alpha \omega_c^{1-s} \omega^s, \quad 0 < \omega < \omega_c, \quad s > -1, \]

(6)

where the dimensionless parameter \( \alpha \) characterizes the dissipation strength. \( \omega_c \) is a cutoff energy and is used as the energy scale throughout the paper. The value \( s = 1 \) corresponds to the case of ohmic dissipation. The NRG procedure allows for an arbitrary spectral function \( J(\omega) \), in particular one with an internal structure. In order to reveal a pronounced effect by some additional high-frequency bosonic modes, we allow for a modification of the power law by an additional \( \delta \)-peak at frequency \( \omega_0 < \omega_c \) with spectral weight \( M \) such that

\[ J(\omega) \to J'(\omega) = J(\omega) + M^2 \delta(\omega - \omega_0). \]

(7)

In the usual SBM, the local spin Hamiltonian

\[ H_{\text{loc}}^{\text{SBM}} = -\epsilon \sigma_z + \frac{\Delta}{2} \sigma_s \]

(8)

is parametrized by an energy splitting \( \epsilon \) and a quantum-tunneling term \( \Delta \), where \( \sigma_i \) are Pauli matrices [1].

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3. NRG

3.1. Equilibrium NRG for a bosonic environment

In all our calculations, we have used Wilson’s NRG method. Wilson’s NRG method is a very powerful tool for accurately calculating equilibrium properties of quantum impurity models [12]. Originally the NRG was invented by Wilson for a fermionic bath to solve the Kondo problem [11, 23]. The method was recently extended to treat quantum impurities coupled to a bosonic bath [18, 19], combination of fermionic and bosonic baths [24] and real-time dynamics out of equilibrium [14, 15, 25]. The non-perturbative NRG approach has been successfully applied to arbitrary electron–bath couplings strength [18–20, 24].

At the heart of this approach is a logarithmic discretization of the continuous bath, controlled by the discretization parameter \( \Lambda > 1 \); the continuum limit is recovered for \( \Lambda \to 1 \). Using an appropriate unitary transformation [11], the Hamiltonian is mapped onto a semi-infinite tight-binding chain, defined by a sequence of finite-size Hamiltonians \( H_m \) with the impurity coupled to the open end. The tight-binding parameters \( t_m \) linking consecutive sites of the chain \( m \) and \( m+1 \) falls off exponentially as \( t_m \sim \Lambda^{-m} \). Each bosonic chain link is viewed as representative of an energy shell since its energy \( w_m \) also decreases as \( w_m \sim \Lambda^{-m} \) establishing an energy hierarchy. Both ensure that mode coupling can only occur between neighboring energy shells, which is essential for the application of the renormalization group (RG) procedure. To this end, the RG transformation \( R[H] \) reads

\[
H_{m+1} = R[H_m] = \Lambda H_m + t_m \left( a_m^\dagger a_{m+1} + a_{m+1}^\dagger a_m \right) + w_m a_{m+1}^\dagger a_{m+1},
\]  

where \( H_m \) is the Hamiltonian of a finite chain up to the site \( m \), and \( t_m = \Lambda^m t_m \). The annihilation (creation) operators of site \( m \) are denoted by \( a_m \) \( (a_m^\dagger) \) and \( w_m \) represent the energy of the bosonic mode of site \( m \). Note that the rescaling of the Hamiltonian \( H_m \) by \( \Lambda \) ensures the invariance of the energy spectrum of fixed point Hamiltonians under the RG transformation \( R[H_m] \). For a detailed review on this method see [12].

The RG transformation (9) is used to setup and diagonalize iteratively the sequence of Hamiltonians \( H_m \). In the first step, only the large spin coupling to the single bosonic site \( m = 0 \) is considered. It turns out to be sufficient [12, 18, 19] to include only the \( N_b \) lowest lying bosonic states, where \( N_b \) takes typical values of 8–12. The reason for that is a quite subtle one: the coupling between different chain links decays exponentially and is restricted to the nearest-neighbor coupling by construction, both essential for the RG procedure. In each successive step (i) a finite number of \( N_b \) bosonic states of the next site \( m+1 \) are added, (ii) the Hamiltonian matrices are diagonalized and (iii) only the lowest \( N_s \) states are retained in each iteration. The discarding of high energy states is justified by the Boltzmannian form of the equilibrium density operator when the temperature is lowered simultaneously in each iteration step to the order \( T_m \sim \Lambda^{-m} w_c \).

Denoting the set of low-lying eigenstates by \( |r\rangle_N \) and the corresponding eigenvalues \( E_r(N) \sim O(1) \) at iteration \( N \), the equilibrium density matrix \( \rho_0 \) is given [12] by

\[
\hat{\rho}_0 = \frac{1}{Z_N} \sum_r e^{-\hat{\beta} E_r^N} |r\rangle_N \langle r|,
\]
where $Z_N = \sum_r e^{-\beta E_r^N}$ and $\tilde{\beta}$ are of the order $O(1)$, such that $T_N = w_c \Lambda^{-N} / \tilde{\beta}$. The thermodynamic expectation value of each local observable $\hat{O}$ is accessible at each temperature $T_N$ by the trace

$$\langle \hat{O} \rangle_{eq} = \text{Tr} \left[ \hat{\rho}_0 \hat{O} \right] = \frac{1}{Z_N} \sum_r e^{-\beta E_r^N} \langle r | \hat{O} | r \rangle_N. \tag{11}$$

The procedure described above turns out to be very accurate because the coupling $t_m$ between the bosonic sites along the chain is falling off exponentially so that the rest of the semi-infinite chain contributes only perturbatively [11, 12] at each iteration $m$, while contributions from the discarded high-energy states are exponentially suppressed by the Boltzmann factor.

### 3.2. TD-NRG

While the equilibrium properties are fully determined by the energy spectrum of the Hamiltonian, the non-equilibrium dynamics requires two conditions: the initial condition encoded in the many-body density operator $\hat{\rho}_0$ and the Hamiltonian $\hat{H}$ which governs its time-evolution. For a time-independent Hamiltonian, the density operator evolves according to $\hat{\rho}(t > 0) = e^{-i\hat{H}t} \hat{\rho}_0 e^{i\hat{H}t}$. All time-dependent expectation values $\langle \hat{O} \rangle(t)$ are given by

$$\langle \hat{O} \rangle(t) = \text{Tr} \left[ \hat{\rho}(t) \hat{O} \right] = \text{Tr} \left[ e^{-i\hat{H}t} \hat{\rho}_0 e^{i\hat{H}t} \hat{O} \right], \tag{12}$$

where we set $\hbar = 1$.

We obtain the density operator $\hat{\rho}_0$ from an independent NRG run using a suitable initial Hamiltonian $\hat{H}$. For instance, by choosing a large local magnetic field in $\hat{H}$, we can prepare the system such that $S_z$ is fully polarized. To investigate decoherence we choose $\mathcal{H}_\text{loc}^i$ such that

$$|s\rangle = \frac{1}{2J + 1} \sum_m |m\rangle \tag{13}$$

is an eigenstate of an appropriate $\mathcal{H}_\text{loc}^i$ with the lowest eigenenergy.

In general, the initial density operator $\hat{\rho}_0$ contains states which are most likely superpositions of excited states of $\hat{H}$. For the calculation of the real-time dynamics of the large spin it is therefore not sufficient to take into account only the retained states of the Hamiltonian $\hat{H}$ obtained from an NRG procedure. The recently developed TD-NRG [14, 15] circumvents this problem by including contributions from all states. It turns out that the set of all discarded states eliminated during the NRG procedure form a complete basis set [14, 15] of the Wilson chain which is also an approximate eigenbasis of the Hamiltonian. Using this complete basis, it was shown [14, 15] that equation (12) transforms into the central equation of the TD-NRG for the temperature $T_N$

$$\langle \hat{O} \rangle(t) = \sum_{m=0}^{N} \sum_{r,s} \sum_{m=0}^{\text{trun}} e^{i(E_r^m - E_s^m)t} O_{r,s}^m \rho_{s,r}^\text{red}(m), \tag{14}$$

where $O_{r,s}^m = \langle r; m | \hat{O} | s; m \rangle$ are the matrix elements of any operator $\hat{O}$ of the electronic subsystem at iteration $m$, and $E_r^m$, $E_s^m$ are the eigenenergies of the eigenstates $| r; m \rangle$ and $| s; m \rangle$ of $\mathcal{H}_m$. At each iteration $m$, the chain is formally partitioned into a 'system' part on which the Hamiltonian $\mathcal{H}_m$ acts exclusively and an environment part formed by the bosonic sites.
$m + 1$ to $N$. Tracing out these environmental degrees of freedom $e$ yields the reduced density matrix [14, 15]

$$\rho^\text{red}_{s,r}(m) = \sum_e \langle s, e; m | \hat{\rho}_0 | r, e; m \rangle$$ (15)

at iteration $m$, where $\hat{\rho}_0$ is given by (10) using $\mathcal{H}_i$. The restricted sum $\sum_{r,s}^{\text{trun}}$ in equation (14) implies that at least one of the states $r$ and $s$ is discarded at iteration $m$. Excitations involving only kept states contribute at a later iteration and must be excluded from the sum.

As a consequence, all energy shells $m$ contribute to the time evolution: the short time dynamics is governed by the high energy states, whereas the long time behavior is determined by the low lying excitations. Dephasing and dissipation are encoded in the phase factors $e^{i(E_r^m - E_s^m)t}$ as well as the reduced density matrix $\rho^\text{red}_{s,r}(m)$.

Discretization of the bath continuum will lead to finite-size oscillations of the real-time dynamics around the continuum solution and deviations of expectation values from the true equilibrium at long timescales. In order to separate the unphysical finite-size oscillations from the true continuum behavior, we average over different bath discretization schemes using Oliveira’s $z$-averaging (for details see [15, 26]). We average over $N_z = 16$ different bath discretizations in our calculation.

Previously, the TD-NRG has been successfully applied to the simple SBM [15, 20], the SMM coupled to a fermionic bath [16] and electron-transfer in a dissipative environment [27].

4. Equilibrium and QPTs

4.1. Integer spin $J$ and QPTs

In figure 2(a), we plot the zero-temperature phase diagram in the sub-ohmic regime for a moderate value of an integer spin ($J = 3$). Depicted is the rescaled critical coupling strength $\alpha_c J^2$ as a function of the bath power-law exponent in the sub-ohmic regime in the absence of an additional $\delta$-peak in $J(\omega)$ ($M_2 = 0$). The line marks the QPT between a localized and a delocalized phase. We have obtained the critical coupling strength by investigating the NRG level flow which shows that the QPT is associated with a distinct fixed point. For $s > 1$, no QPTs exist [18, 19].

A fixed point Hamiltonian $H^*$ remains invariant under the RG transformation equation (9), i.e. $H_{m+1} = R[H^*_m] = H^*$. As a consequence, the energy spectrum of the eigenstates of $H^*$ does not change after the iteration $m \rightarrow m + 1$. Therefore, the fixed-point spectra can be identified by plotting the eigenenergies as function of the iteration $m$. Each fixed point is characterized by a constant eigenenergy spectrum, while different fixed points will have different eigenenergy spectra.

In order to illustrate how the critical couplings $\alpha_c$ were determined in figure 2(a), figure 2(b) depicts the NRG level flow for three different coupling constants $\alpha = 0.0002 < \alpha_c$, $\alpha_c$ and $\alpha = 0.0025 > \alpha_c$ for $s = 0.7$ and $J = 3$. For large $m$, three different fixed points are visible. The energy level flow to the black flow for all $\alpha < \alpha_c$ and to the blue flow for $\alpha > \alpha_c$, while $\alpha_c$ is obtained via bisection of the coupling constant $\alpha$. Since the energy level is two-fold degenerate in the localized phase $\alpha > \alpha_c$, the same fixed amount of ten energy levels map onto five visible levels in figure 2(a), while the finite renormalized tunneling rate induces a level splitting in the delocalized phase.
Figure 2. Zero temperature phase diagram (a) in the sub-ohmic regime for $J = 3$. The critical coupling strength $\alpha_c(2J)^2$ is plotted versus the exponent $s$ in $J(\omega)$ for $M = 0$, $A = -0.1$, $B_2 = B_4 = 10^{-3}$ and $H = 0$. NRG energy level flow as function of the iterations for $s = 0.7$ and three different coupling constants $\alpha = 0.002, \alpha_c$ and 0.0025, where $\alpha_c = 0.002 228 55$. NRG parameters: $\Lambda = 2$, $N_s = 100$ and $N_b = 8$.

Analyzing the NRG fixed-point spectra [11], the same qualitative picture emerges for all integer values of $J$. We noted that the critical coupling strength depends (i) on the value of the spin $J$ and (ii) the values of the quantum tunneling rates $B_{2n}$. Only for $s \to 1$, the critical coupling becomes independent of $B_{2n}$ and always approaches

$$\lim_{s \to 1} \alpha_c(s) \approx 1/(2J)^2,$$

which can be understood from the analytical form of $H_1$.

The fixed-point spectra always agree with those of the SBM. This establishes a mapping of our model at low temperatures onto an effective SBM. This mapping becomes intuitively clear by inspecting figure 1(b). It shows a coupling between the two lowest states $|\pm J\rangle$ on the parabola. For $T \to 0$, it is expected that the model can be replaced by an effective SBM with a tunneling rate $\Delta$ which is a complicated function of parameters $A$ and $B_{2n}$. Therefore, it is not surprising that our calculations for the anisotropic large spin model reveal the similarities to the results of Bulla et al [19], who established such a QPT in the SBM. Since the coupling of the two states $|\pm J\rangle$ to the bosonic bath is proportional to $J_z$, we find that $\alpha_c(s)(2J)^2$ resembles the phase boundary of the SBM. The critical coupling $\alpha_c$ is proportional to $1/J^2$. 

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Such a behavior is also observed in the equilibrium spin–spin correlation function $C(\omega) = \Im m \ll S_z |S_z \gg (\omega - i\delta)$. The correlation function $C(\omega)$ is calculated using the sum-rule conserving algorithm [28, 29]. We plot $C(\omega)$ on a log–log scale for a series of coupling constants and the sub-ohmic regime $s = 0.5$ in figure 3(a). The high-energy feature stemming from the easy-axis energy splitting proportional to $A$ is clearly visible. At lowest coupling strength $\alpha = 10^{-4}$, the second peak at $\omega \approx 3 \times 10^{-3}$ is well pronounced, originating from the weak effective quantum tunneling rate which is a function of parameters of $H_{loc}: A, B_2, B_4$ as well as $J$. For $\alpha < \alpha_c$, $C(\omega)$ vanishes according to $C(\omega) \propto \omega^s$, while $C(\omega)$ diverges as $\omega^{-s}$ for $\omega \rightarrow 0$ and $\alpha > \alpha_c$.

4.2. Half-integer spin $J$

A completely different picture emerges for half-integer spins $J$. Independent of the bath exponent $s$, no phase transition is found. As seen already in figure 1, the quantum tunneling matrix elements $B_2$ and $B_4$ cannot connect the two states $|\pm J \rangle$. The model can be mapped onto a SBM with a vanishing tunneling rate $\Delta$ at low temperatures. To illustrate this, we compare the NRG level flow [12] of the large spin model with the NRG level flow of a simple SBM and $\Delta = 0$. The fixed-point spectra for $m \rightarrow \infty$ is identical for both models as depicted in figure 4. The coupling constant $\alpha$ determines the crossover from high-temperature to the low-temperature fixed point: this crossover scale accidentally coincides in both models for our choice of coupling constants. $C(\omega)$ depicted in figure 3(b) is similar to the results of figure 3(a) at high energies: the peak due to the easy-axis splitting $A$ is also present. However, a pronounced peak at the effective quantum-tunnel energy is absent for any half-integer $J$.

5. Non-equilibrium dynamics

For the investigation of the non-equilibrium dynamics, the local spin is prepared in a maximal polarized initial state with the expectation value $S_z(0) = J$ by applying a strong local magnetic field in the $z$-direction. The external magnetic field is switched off at $t = 0$. 

![Figure 3](http://www.njp.org/)
Figure 4. NRG level flow of the lowest eigenenergies $E^m$ for $J = 7/2$, $\alpha = 0.0012$ in the sub-ohmic regime $s = 0.5$ in comparison with the level flow of the SBM for $\Delta = 0$ and $\alpha = 0.05$. Parameters as in figure 2.

Figure 5. Real-time dynamics of the spin $S_z$ initially prepared in $S_z(0) = J = 4$ for different values of $\alpha$ in the ohmic regime $T = 3 \times 10^{-5}$. Parameters: $A = 0.1$, $B_2 = 10^{-3}$, $B_4 = 2 \times 10^{-3}$, $\Lambda = 2$, $N_s = 160$, $N_b = 8$ and $N_z = 16$.

Figure 5 displays the real-time dynamics of $S_z(t)$ for a series of coupling strength $\alpha$ and an ohmic bath ($s = 1$), $J = 4$ and the easy-axis splitting $A = 0.1$. A weak quantum tunneling was enabled by $B_2 = 10^{-3}$ and $B_4 = 2 \times 10^{-3}$. As a reference, the dynamics of a decoupled spin ($\alpha = 0$) is included in the graph. For small $\alpha$, damped coherent oscillations with a superimposed short-time dynamics of small amplitude are clearly visible. The oscillation frequency decreases with increasing $\alpha$ and vanishes at a finite coupling $\alpha$. Above that value—here $\alpha \approx 0.0075$—only a spin decay is observed. This corresponds to the regime $0.5 < \alpha < 1$ of the SBM. When approaching the critical coupling $\alpha_c(J = 4) \approx 0.016$, $S_z(t)$ only decays insignificantly on a very short timescale of the order $O(1)$ and remains constant for $t \to \infty$, as seen in figure 5(a). This behavior occurs in the localized phase of the model where the effective tunneling rate is renormalized to zero.
Figure 6. Real-time dynamics of the spin $S_z$ initially prepared in $S_z(0) = J = 5$ for different values of $\alpha$ in the sub-ohmic regime $s = 0.5$, $T = 10^{-6}$. The upper panel (a) shows the short time dynamics on a linear timescale, while (b) resolves the real-time dynamics on a logarithmic timescale. For couplings $\alpha > \alpha_c \approx 0.00061$, the finite value of $S_z(t \to \infty)$ is a consequence of the QPT into a localized phase. Parameters: $A = 0.1$, $B_2 = 10^{-3}$, $B_4 = 2 \times 10^{-3}$, $\Lambda = 2$, $N_s = 160$, $N_b = 8$ and $N_z = 16$.

A different picture emerges in the sub-ohmic regime. The real-time dynamics of a large anisotropic spin with $J = 5$ is depicted for different coupling strength in figure 6. The upper panel shows the dynamics on a linear time axis. In the weak coupling regime, coherent high- and low-frequency oscillations are clearly visible, in addition to a slow decay of the amplitude. At a critical coupling of $\alpha = 0.00061$, we found a QPT in the equilibrium NRG calculations. As discussed in section 4.1, the quantum-critical point separates a delocalized from a localized phase. As seen in figure 6, coherent oscillations prevail in the short-time dynamics even in the localized phase close to the QPT. A finite time is required before the strong correlations can build up. In the long-time limit, $S_z(t)$ approaches a finite value in the localized phase which should be proportional to the order parameter for the QPT. These results are very similar to the dynamics reported in the sub-ohmic SBM [20]. In our case, larger number of eigenstates of $H_{\text{loc}}$ yields additional coherent oscillations absent in the simple model.

In figure 7, we used the same model parameters as in figure 6, but add an additional $\delta$-peak with a weight $M^2 = 0.02\omega_0^2$ to illustrate the effect of the coupling to an additional single high-energy mode at $\omega_0 = 0.0\omega_c$. We have benchmarked the TD-NRG for such a modified $J(\omega)$ (18) by finding excellent agreement between the exact analytical dephasing curves of
Figure 7. Real-time dynamics of the spin $S_z$ initially prepared in $S_z(0) = J = 5$ for different values of $\alpha$ in the sub-ohmic regime $s = 0.5$ as in figure 6. However, an additional sharp mode at $\omega_0 = 0.9\omega_c$ with a spectral weight of $M^2 = 0.02\omega_c^2$ has been added to $J(\omega)$.

The SBM as described in equation (18) with the prediction of the TD-NRG [15]. Therefore, the TD-NRG is capable of describing the changes in the short-time dynamics in the presence of such a $\delta$-peak. Clearly visible are the additional short time beats stemming from the interference with this additional mode. Since the effective coupling has increased, we also observe a shift of the coherent oscillations to lower frequencies. Note, however, that the long time behavior remains almost unaltered, despite the dominating coupling to the $\omega_0$ mode since $\int d\omega J(\omega) = 2\pi\alpha\omega_c^2/(1 + s) \approx 4.1\alpha\omega_c^2 \ll M^2\omega_c^2$. This clearly confirms our knowledge on the spin-boson-type models [1] that additional internal structure of the bath spectral function only influences the short time response, but has a rather weak impact on the long time dynamics and the occurrence of QPTs. $\alpha_c$ is very weakly dependent on this additional mode $\omega_0$.

We expect that coherent oscillations prevail to longer timescales with decreasing exponent $s$. Simultaneously, the critical coupling $\alpha_c$ reduces strongly with decreasing exponent $s$, as shown in figure 2.

6. Decoherence

Decoherence is caused by the interaction of a subsystem with its environment. The concept of decoherence [30, 31] provides insight into how a coherence quantum state $|s\rangle$ of the subsystem
is destroyed by the entanglement with the environment. The reduced density matrix \[ (17) \]

\[ \rho^\text{red}_{i,j}(t) = \sum_e \langle i,e|\hat{\rho}(t)|j,e\rangle \]
evolves from a matrix describing such a pure state \( |s\rangle \) into a matrix describing an ensemble. In an appropriate local basis \( |i\rangle \), the off-diagonal matrix elements of \( \rho^\text{red}_{i,j} \) must vanish for \( t \to \infty \) \[ 30, 31 \]. In a diagonally coupled SBM as described by the interaction \( \mathcal{H}_I \), equation (4), this would be the eigenstates of \( S_z \) \[ 30 \]. In the absence of quantum tunneling, \( S_z \) also commutes with \( \mathcal{H}_\text{loc} \) and no energy is exchanged by the coupling \( \mathcal{H}_I \) to the environment. Decoherence is induced by the dephasing with continuum of the bath modes each contributing a different phase shift. For \( J = 1/2 \), this is a well-studied problem, and it was shown analytically \[ 33, 34 \] that the off-diagonal component of the reduced density matrix for the spin can be written as \( \rho_{\uparrow,\downarrow}(t) = e^{-\Gamma(t)}\rho_{\uparrow,\downarrow}(0) \), where \( \Gamma(t) \) is given by the exact analytic expression \[ (18) \]

\[ \Gamma(t) = \frac{1}{\pi} \int_0^\infty d\omega J(\omega) \coth \left( \frac{\omega}{2T} \right) \frac{1 - \cos(\omega t)}{\omega^2}. \]

Here, \( T \) is the temperature. Note that decoherence of the pure quantum state \( |s\rangle \) does not follow a simple exponential form. Approximating the dephasing by a function \( \propto \exp(-t/T_{\text{deph}}) \), which defines a single ‘dephasing’ timescale \( T_{\text{deph}} \), might be insufficient even in very simple models such as the SBM, since \( \Gamma(t) \) cannot be replaced by \( t/T_{\text{deph}} \).

We initially prepared the spin system in a pure state \( |s\rangle \) comprising a linear combination of all \( S_z \) eigenstates

\[ |s\rangle = \frac{1}{2J+1} \sum_{m=-J}^{J} |m\rangle \]

by an appropriate choice of the initial Hamiltonian \( \mathcal{H}_\text{loc}^i \).

We can gain some information on the decoherence of a large anisotropic spin by measuring the time-dependent expectation value \( S_z(t) = \langle \hat{S}_z \rangle(t) \), which only depend on the off-diagonal matrix elements of \( \rho^\text{red}_{i,j}(t) \). The quantum-tunneling parameters were set to \( B_{2m} = 0 \) in the calculations. Figure 8 shows the dynamics of \( S_z(t) \) on a rescaled dimensionless time axis \( \tau \alpha \) for an integer \( (J = 5) \) and a half-integer \( (J = 9/2) \) value of the spin in the \( s = 0.5 \) sub-ohmic regime. In both cases, we observe an oscillatory decay of \( S_z \) different from the result for the SBM, equation (18). Here, the finite coupling \( \alpha \) introduces an effective \( A \) via the reorganization energy \( E_{1g} \)

\[ E_{1g} = \sum_q \frac{\lambda_q^2}{\omega_q} = \int_0^\infty d\omega \frac{J(\omega)}{\pi \omega} = \frac{2\alpha \omega_c}{s}, \quad s > 0. \]

The effective energy \( \tilde{E}_m \propto (A - E_{1g}) m^2 \) of eigenstates \( |m\rangle \) of \( S_z \) causes quantum-oscillations whose oscillation frequency is proportional to \( \alpha \) for \( A = 0 \). This \( \alpha \) dependence of the frequency is revealed by plotting \( S_z(t) \) versus the dimensionless timescale \( \tau \alpha \) so that the oscillation beats coincide and occur on a timescale of \( \tau \alpha \approx O(1) \). Since all eigenenergies \( \tilde{E}_m \) are related to each other by integer values proportional to \( A - E_{1g} \), the local dynamics is characterized by a recurrence time proportional to \( 1/(A - E_{1g}) \) at which the original local state would reappear in the absence of a spin–environment coupling. However, the envelope
Figure 8. $S_s(t)$ versus $t*\alpha$ for three different coupling strength $\alpha = 10^{-4}$, $10^{-3}$ and $10^{-2}$ for $J = 5$ (a) and $J = 9/2$ (b) values of the spin in the $s = 0.5$ sub-ohmic regime. $S_s(t)$ measures the decoherence of the off-diagonal matrix elements of $\rho_{i,j}^{\text{red}}$. Parameters: $T = 10^{-6}$, $A = B_{2n} = 0$. (c) shows $S_s(t)$ versus $t*\alpha$ for the same values of $\alpha$ as (b) but with $A = 2\alpha\omega_c/s$, $B_{2n} = 0$. NRG parameters as in figure 5.

function describing the time dependence of the recurrence amplitude decreases on a timescale proportional to $1/\alpha$.

We can prolong this recurrence time by an appropriate counter-term in the Hamiltonian. The curves in figure 8(c) are obtained for the same parameters as in figure 8(a) and by setting $A = 2\alpha\omega_c/s$. Due to the discretization of the bath continuum, the cancellation is not perfect, but the oscillation frequency is lowered by almost two decades.

7. Conclusion

The equilibrium and non-equilibrium dynamics of an anisotropic large spin coupled to a dissipative sub-ohmic bosonic environment has been investigated using the non-perturbative NRG. Such a large spin might be realized in a SMM. The coupling of the bath modes to the easy-axis component of the spin enhances the tendency for localization. A QPT exists only for integer spin values in the sub-ohmic regime, and the quantum-critical point is associated with a new fixed point. These results generalized the work of Bulla et al [18] for the SBM. The spin–spin correlation function $C(\omega)$ diverges approximately as $|\omega|^{-s}$ at and above the critical
coupling $\alpha_c$. The critical coupling for $s \to 1$ is related to the critical coupling of the SBM by $\alpha_c = \alpha_c^{SBM}/(2J)^2$. We have shown that the fixed point of the half-integer model is identical to the $S = 1/2$ SBM without quantum tunneling $\Delta$. No QPT is found for half-integer $J$ due to the lack of quantum tunneling between the two states $|\pm J\rangle$.

We investigated the spin decay by switching off an external easy-axis magnetic field at $t = 0$. In the ohmic regime ($s = 1$), the real-time dynamics of $S_z(t)$ is governed by several local frequencies stemming from the easy-axis splitting $A$ and the quantum-tunneling matrix elements $B_{2n}$. With increasing coupling $\alpha$, the quantum tunneling is reduced, but the splitting $A$ is increased by the reorganization energy $E_1g$. The low-frequency coherent oscillations vanish at some value $\alpha$ but the high-frequency coherent oscillations prevail with a very small amplitude. Further increasing of $\alpha$ beyond the critical coupling $\alpha_c$ drives the system in the localized phase. Here, the spin decays weakly on a very short timescale of the order of the reciprocal cutoff $1/\omega_c$.

A completely different picture emerges in a sub-ohmic environment. Oscillatory response is found at short and intermediate timescales even in the localized phase at coupling strength $\alpha > \alpha_c$. The coupling of the large spin to an additional strong mode at $\omega_0$ modifies the short-time response. The long-time response remains only very weakly affected by such an internal structure of a bath: the long-time response as well as the physics of the QPTs are governed by the power law of the bath at low frequencies $\omega \to 0$.

In the calculation of the decoherence through dephasing, we identified the occurring long-time oscillation frequency with recurrence frequency which is dependent on the reorganization energy. These oscillations can be reduced and suppressed by adding a counter-term in the original Hamiltonian. We have demonstrated that the non-perturbative RG approach can describe such equilibrium and non-equilibrium dynamics close to a QPT.

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References

[1] Leggett A J, Chakravarty S, Dorsey A T and Fisher M P A 1987 Rev. Mod. Phys. 59 1
[2] Weiss U 1999 Quantum Dissipative Systems (Singapore: World Scientific)
[3] Thomas L, Caneschi A and Barbara B 1999 Phys. Rev. Lett. 83 2398–401
[4] Chiorescu I, Giraud R, Jansen A G M, Caneschi A and Barbara B 2000 Phys. Rev. Lett. 85 4807
[5]Gatteschi D and Sessoli R 2004 Anzgew. Chem. Int. Ed. Engl. 42 268
[6]Keren A, Shimshoni O S E, Marvauv D, Bachschildt A and Long J 2007 Phys. Rev. Lett. 98 257204
[7] Prokof’ev N V and Stamp P C E 1998 Phys. Rev. Lett. 80 5794
[8] Leuenberger M N and Loss D 1999 Europhys. Lett. 46 692
[9] Fernández J F and Alonso J J 2003 Phys. Rev. Lett. 91 047202
[10] Vorrath T and Brandes T 2005 Phys. Rev. Lett. 95 070402
[11] Wilson K G 1975 Rev. Mod. Phys. 47 773
[12] Bulla R, Costi T and Pruschke T 2008 Rev. Mod. Phys. 80 395
[13] Romeike C, Wegewijs M R, Hofstetter W and Schoeller H 2006 Phys. Rev. Lett. 96 196601
[14] Anders F B and Schiller A 2005 Phys. Rev. Lett. 95 196801
[15] Anders F B and Schiller A 2006 Phys. Rev. B 74 245113

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[16] Roosen D, Wegewijs M R and Hofstetter W 2008 Phys. Rev. Lett. 100 087201
[17] Politi P, Rettori A, Hartmann-Boutron F and Villain J 1995 Phys. Rev. Lett. 75 537
[18] Bulla R, Tong N and Vojta M 2003 Phys. Rev. Lett. 91 170601
[19] Bulla R, Lee H J, Tong N H and Vojta M 2005 Phys. Rev. B 71 045122
[20] Anders F B, Bulla R and Vojta M 2007 Phys. Rev. Lett. 98 210402
[21] Nesi F, Paladino E, Thorwart M and Grifoni M 2007 Europhys. Lett. 80 40005
[22] Argyres P N and Kelley P L 1964 Phys. Rev. A 134 98
[23] Krishna-murthy H R, Wilkins J W and Wilson K G 1980 Phys. Rev. B 21 1003
[24] Glossop M T and Ingersent K 2007 Phys. Rev. B 75 104410
[25] Costi T A 1997 Phys. Rev. B 55 3003
[26] Yoshida M, Whitaker M A and Oliveira L N 1990 Phys. Rev. B 41 9403
[27] Tornow S, Bulla R, Anders F B and Nitzan A 2008 Phys. Rev. B 78 035434
[28] Peters R, Pruschke T and Anders F B 2006 Phys. Rev. B 74 245114
[29] Weichselbaum A and von Delft J 2007 Phys. Rev. Lett. 99 076402
[30] Zurek W H 2003 Rev. Mod. Phys. 75 715–75
[31] Schlosshauer M 2005 Rev. Mod. Phys. 76 1267–305
[32] Feynman R P 1972 Statistical Mechanics, A Set of Lectures (Reading, MA: Benjamin)
[33] Unruh W G 1995 Phys. Rev. A 51 992
[34] Palma G M, Suominen K A and Ekert A K 1996 Proc. R. Soc. Lond. A 452 567

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