

**Parity effects in spin decoherence**A. Melikidze,<sup>1</sup> V. V. Dobrovitski,<sup>2</sup> H. A. De Raedt,<sup>3</sup> M. I. Katsnelson,<sup>4</sup> and B. N. Harmon<sup>2</sup><sup>1</sup>*Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106, USA*<sup>2</sup>*Ames Laboratory, Iowa State University, Ames, Iowa 50011, USA*<sup>3</sup>*Applied Physics, Computational Physics, Materials Science Centre, University of Groningen, Nijenborgh 4, NL-9747 AG Groningen, The Netherlands*<sup>4</sup>*Department of Physics, Uppsala University, SE-751 21 Uppsala, Sweden*

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We demonstrate that decoherence of many-spin systems can drastically differ from decoherence of single-spin systems. The difference originates at the most basic level, being determined by parity of the central system, i.e., by whether the system comprises even or odd number of spin-1/2 entities. Therefore, it is very likely that similar distinction between the central spin systems of even and odd parity is important in many other situations. Our consideration clarifies the physical origin of the unusual two-step decoherence found previously in the two-spin systems.

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**I. INTRODUCTION**

Reduced dynamics of a small quantum system coupled to a bigger environment has recently become the subject of particularly active investigation. In fields like quantum optics<sup>1</sup> and quantum computation,<sup>2</sup> there is a naturally defined distinct “central” system (i.e., an atom or a qubit) which interacts with its environment, and whose dynamics is of primary importance. Similar situations are often encountered in the condensed matter physics, e.g., when considering a heavy particle tunneling in a crystal, tunneling centers in glasses,<sup>5</sup> Kondo systems,<sup>4</sup> etc. This problem is also of importance when a naturally defined central system is absent, such as in a recently developed promising approach to the theory of strongly correlated systems, the dynamical mean-field theory (for review, see Ref. 3). In this approach, the system of interacting particles in a crystal is replaced by an “effective impurity” in a self-consistently defined thermostat.

So far, quantum evolution of a single two-level system (or, equivalently, a single spin-1/2 entity) interacting with a bath of bosons<sup>5</sup> or spins<sup>6,7</sup> has been studied in much detail. In contrast, the central systems comprising several strongly interacting spins 1/2 have not been that extensively investigated. A general analysis of the two-spin central system interacting with a bath of bosons has been presented in Ref. 8, but more detailed considerations are lacking. Several interesting cases of a two-spin system coupled to a spin bath have been considered in Refs. 9 and 10, and it has been demonstrated that behavior of many-spin central systems can be very different from a single-spin case. Consideration of many-spin central systems is of particular importance for possible implementation of quantum computations which use several strongly coupled two-level systems for encoding of a single qubit.<sup>11,12</sup> This representation allows using the “decoherence-free subspaces” and error-correcting schemes developed for multispin qubits.<sup>13,14</sup>

In this work, based on an exactly solvable but realistic model, we show explicitly that decoherence of a two-spin-1/2 system can be qualitatively different from decoherence of a single spin 1/2. We demonstrate that this difference

originates at the most basic level, and is determined primarily by parity of a central system, i.e., by whether the central system comprises even or odd number of spin-1/2 entities.

It is known that the parity of the spin system is the cause of the drastically different behavior in the tunneling of magnetization in a wide class of spin systems such as magnetic nanoparticles and molecular magnets where the tunneling is due to magnetic anisotropy or magnetic field.<sup>15,16</sup> In this paper, we explore a different effect, in which the parity of the central system determines the long-time dynamics of the decoherence process. We emphasize that in the system considered here the quantum oscillations are caused by the isotropic exchange interaction and are independent of the symmetry of the crystal field and external magnetic field; thus the short-time oscillations do not depend on the parity.

Although there are many possible central systems coupled to various kinds of spin baths, the generic differences between the many-spin and the single-spin central systems can be understood based on simple models. An instructive model of a many-spin central system interacting with a spin bath, has been recently analyzed by Dobrovitski *et al.*<sup>9</sup> This model describes (at least, qualitatively) the main features of such central systems as magnetic molecules, quantum dots or impurity spins which experience decoherence from the nuclear spin bath. In these systems, the dominant interaction with the nuclear spins can be approximated by the isotropic Heisenberg interaction, since anisotropic interactions are often small. The model is defined by the Hamiltonian:

$$H = \vec{C}^2 + 2\vec{C} \sum_{k=1}^N J_k \vec{s}_k, \quad (1)$$

which describes the central system composed of two spins:  $\vec{C} = \vec{c}_1 + \vec{c}_2$ ,  $c_1 = c_2 = 1/2$ , which is coupled by Heisenberg exchange interaction to  $N$  environmental spins  $s_k = 1/2$ ,  $k = 1 \dots N$ . Note that the environmental spins do not have their own dynamics. This may be viewed as a limited case where the dynamics of the central system is much faster than that of the environment.

A special feature of this model, which makes it different from the ‘‘central spin’’ models considered by Garg, or Prokof’ev and Stamp,<sup>6,7</sup> is the fact that in our treatment the central system is not reduced to the doublet of lowest states. This feature is crucial to the results discussed below.

One is interested in the time evolution of the initial system-plus-environment state which is taken in the form:

$$|i\rangle = |\uparrow\rangle_{c_1} |\downarrow\rangle_{c_2} \prod_{k=1}^N |i\rangle_{s_k}. \quad (2)$$

The initial states of the environmental spins  $|i\rangle_{s_k}$  are assumed random and uncorrelated. The initial state of the system is a superposition of the singlet and triplet states of the two central spins:

$$|\uparrow\rangle_{c_1} |\downarrow\rangle_{c_2} = \frac{1}{\sqrt{2}} (|1,0\rangle_C + |0,0\rangle_C), \quad (3)$$

where we have introduced notation  $|C, C^z\rangle_C$  for the central spin. One considers the problem of the decay of this coherent singlet-triplet superposition in the central system due to its interaction and subsequent entanglement with the environmental spins. In particular, one is interested in the time dependence of the expectation value of the  $z$  component of the first spin  $\langle\sigma_1^z(t)\rangle$ , where  $\sigma_1^z$  is the Pauli matrix acting on the state of  $\vec{c}_1$ . In the absence of the coupling to the environment this quantity exhibits periodic oscillations between  $+1$  and  $-1$  caused by the first term in Eq. (1); coupling to the environment is expected to damp these oscillations.

In the work reported in Ref. 9 a numerical investigation of this problem was performed. Among many surprising features in the behavior of the above system, it was observed that after an initial fast decay of the oscillations of  $\langle\sigma_1^z(t)\rangle$  the amplitude showed a saturation at the value of  $1/3$ . Subsequently, the oscillations demonstrate a much slower decay, which is consistent with the  $1/t$  conjecture, and which leads to a complete suppression of oscillations. The main motivation of this paper was to understand the cause of the saturation and the subsequent slow decay.

While the model Eq. (1) is hard to treat analytically, we simplified it by setting all  $J_k$ ’s equal. This allowed us to solve the model exactly. The solution turned out to reproduce quantitatively several key features of the numerical results reported in Ref. 9. In fact, it reproduced the fast initial decay of the amplitude of oscillations and its subsequent saturation at  $1/3$ . It also offers a way to qualitatively understand the cause of the long-time tail. Most importantly, it answers the question: why is the decay of oscillations in our model much slower compared to a more conventional exponential decay of oscillations in, say, the spin-boson models.<sup>5</sup> The cause is the integer value of total spin of the central system.

This work shows that integer spins, in contrast to half-integer spins, may, under suitable circumstances, exhibit quantum oscillations over much longer times. From the perspective of the theory of quantum phase transitions, this work also offers a simple example of emergent power-law correlations usually associated with criticality.

## II. $J_k=J$ MODEL

To make analytical progress we consider a simplified model where we take all coupling constants  $J_k=J$  to be equal while preserving random uncorrelated initial states of the environmental spins. The Hamiltonian takes the form

$$H = \vec{C}^2 + 2J\vec{C}\vec{S} = (1-J)\vec{C}^2 + J(\vec{C} + \vec{S})^2 - J\vec{S}^2, \quad (4)$$

which describes the coupling of the central spin  $\vec{C} = \vec{c}_1 + \vec{c}_2$  to the total spin of the environment  $\vec{S} = \sum \vec{s}_k$ . We are interested in the expectation value of the  $z$  component of  $\vec{c}_1$ :  $\langle\sigma_1^z(t)\rangle$ , where  $\sigma_1^z$  is the Pauli matrix acting on the state of  $\vec{c}_1$ . Note, that the assumed initial condition Eq. (2) corresponds to the superposition of states with different  $S$ . The Hamiltonian Eq. (4) conserves  $\vec{S}^2$ , therefore the matrix element  $\langle\sigma_1^z(t)\rangle$  can be decomposed as

$$\langle\sigma_1^z(t)\rangle = \sum_S \langle S | \sigma_1^z(t) | S \rangle P(S), \quad (5)$$

where  $P(S)$  is the weight of the state with the total spin  $S$  given the random uncorrelated initial states of  $\vec{s}_k$ . We thus are led to the problem of first calculating  $P(S)$ .

Before proceeding with the actual calculation an important comment is in order. Since Eq. (5) looks like an average over all possible initial orientations of the environmental spins one might interpret the above quantity  $\langle\sigma_1^z(t)\rangle$  as an ensemble-averaged expectation value. Quite importantly, in the case where the number of environmental spins is large the actual weight of the state with total spin of the environment  $S$  tends to the ensemble-averaged quantity  $P(S)$ . Therefore, in this limit Eq. (5) describes well the evolution of the central system in a single realization of the experiment.

In the basis where  $\vec{s}_k$  are good quantum numbers the initial density matrix is, by assumption, a  $2^N \times 2^N$  matrix:

$$\rho_i^S = 2^{-N} I, \quad (6)$$

where  $I$  is a unit matrix. Let us make a unitary transformation to the basis spanned by the eigenstates of  $\vec{S}^2$ . There are  $N$  different values that  $\vec{S}^2$  can take. To preserve the dimensionality of the Hilbert space we conclude that some (in fact almost all) of these latter states are degenerate. A unitary transformation will leave the initial density matrix unchanged. This means that

$$P(S) = 2^{-N} G(S) (2S+1), \quad (7)$$

where  $G(S)$  is the degeneracy of the state with total spin  $S$  (with  $S_z$  fixed). To calculate  $G(S)$  we change variables and introduce  $g(k) = G(N/2 - k)$ . The state with the maximum total spin  $S = N/2$  is unique and is the state where all  $\vec{s}_k$ ’s point up (we choose  $S_z = S$ ), therefore  $g(0) = 1$ . Next, a state with  $S = S_z = N/2 - 1$  should be a superposition of the states with  $N-1$  spins up and one spin down. There are  $C_1^N = N$  such states [ $C_M^N = N! / M!(N-M)!$  is the binomial coefficient]. However, among such states there are  $g(0) = 1$  states with  $S = N/2$  and  $S_z = N/2 - 1$  which have to be excluded. Generalizing to arbitrary  $k$  we get:

$$g(k) = C_k^N - \sum_{i=0}^{k-1} g(i) = C_k^N - C_{k-1}^N = C_k^N \left( \frac{N-2k+1}{N-k+1} \right). \quad (8)$$

We thus have the result for the weight of the state with spin  $S$ :

$$P(S) = 2^{-N} C_{N/2-S}^N \frac{(2S+1)^2}{N/2+S+1} \approx \frac{8S^2/N}{\sqrt{2\pi D}} e^{-S^2/2D}, \quad D = N/4, \quad (9)$$

where we have used a well-known approximation for the binomial distribution described by the first two factors above. One can easily check that  $\int_0^\infty P(S) dS = 1$ , i.e., the approximations we made preserve the normalization of the probability.

We have thus reduced the problem to finding the time evolution of the initial state:

$$|f\rangle = e^{-iHt}|i\rangle, \quad (10)$$

$$|i\rangle = \frac{1}{\sqrt{2}}(|1,0\rangle_C + |0,0\rangle_C)|S, S^z\rangle_S, \quad (11)$$

estimating the spin polarization  $\langle f|\sigma_1^z|f\rangle$ , and averaging the result with respect to  $S^z$  (trivial) and  $S$  [according to Eq. (9)]. There are two circumstances that greatly simplify the calculation. First, the Hamiltonian acting on the singlet state  $|0,0\rangle_C$  gives zero, therefore the evolution of the second term in Eq. (11) is trivial. Second, the symmetry of the Hamiltonian with respect to  $\vec{c}_1$  and  $\vec{c}_2$  implies that given the above initial condition we have  $\langle f|\sigma_1^z|f\rangle = -\langle f|\sigma_2^z|f\rangle$ . Thus, we can calculate the expectation value of  $\sigma^z = (\sigma_1^z - \sigma_2^z)/2$  instead. For this operator we have:  $\sigma^z|1,0\rangle_C = |0,0\rangle_C$ ,  $\sigma^z|0,0\rangle_C = |1,0\rangle_C$ ,  $\sigma^z|1, \pm 1\rangle_C = 0$ . Taking all this into account, we see that

$$\langle f|\sigma^z|f\rangle = \text{Re}\langle t|e^{-iHt}|t\rangle, \quad (12)$$

$$|t\rangle = |1,0\rangle_C|S, S^z\rangle_S. \quad (13)$$

From Eq. (4) it is clear that the above matrix element can be easily calculated after going to the basis with well defined total spin  $\vec{L} = \vec{C} + \vec{S}$ . The necessary Clebsch-Gordan decomposition (in the limit  $S \gg 1$  of interest to us) is:

$$|t\rangle \approx \sqrt{\frac{1 - (S^z/S)^2}{2}} (|S+1, S^z\rangle_L - |S-1, S^z\rangle_L) + \frac{S^z}{S} |S, S^z\rangle_L, \quad (14)$$

where we have introduced the notation  $|L, L^z\rangle_L$ . In this basis we easily calculate using Eq. (4):

$$\langle f|\sigma_1^z|f\rangle = \cos 2(1-J)t \left[ \left[ 1 - \left( \frac{S^z}{S} \right)^2 \right] \cos 2JSt + \left( \frac{S^z}{S} \right)^2 \right]. \quad (15)$$

Finally, we have to average this result over  $S^z$  and  $S$ . The first average is done trivially using the fact that (in the same limit

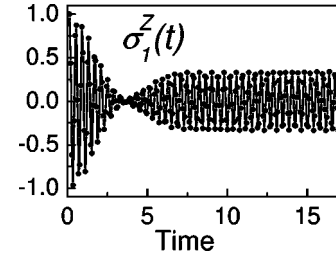


FIG. 1. Numerical simulation of 13 spins with  $J_0=8$ ,  $J_k=J=0.128$ . The figure shows the expectation value of  $\sigma_1^z$  as a function of time.

$S \gg 1$ )  $\langle (S^z)^2 \rangle = S^2/3$ . The second average is calculated using Eq. (9) which leads to a Gaussian integral. The result is:

$$\langle \sigma_1^z \rangle = A(t) \cos 2(1-J)t, \quad (16)$$

$$A(t) = \frac{1}{3} + \frac{2}{3}(1 - NJ^2 t^2) e^{-NJ^2 t^2/2}. \quad (17)$$

It should be stressed that this result is *exact* in the limit  $N \gg 1$  ( $S \gg 1$ ). We see that an initial exponential decay of the amplitude of the oscillations is followed by a transient and an eventual leveling at  $A(t) = 1/3$ .

To check the above results we have performed a direct numerical solution of the Schrödinger equation corresponding of the system with a Hamiltonian  $H = J_0 \vec{C}^2 + 2\vec{C} \sum_{k=1}^N J_k \vec{S}_k$ ,  $J_k = J$ , which can be reduced to Eq. (1) by rescaling  $J_0 \rightarrow 1$ ,  $J \rightarrow J/J_0$ ,  $t \rightarrow tJ_0$ . Exact diagonalization was used to find the time evolution. An example of the results is shown in Fig. 1. It shows the expectation value of  $\sigma_1^z$  as a function of time. The parameters are: the number of spins  $N=13$ ,  $J_0=8$ ,  $J_k=J=0.128$ . This can be compared with the analytic result for the same quantity which is given (after rescaling) by Eq. (16) and is shown in Fig. 2. The numerical and analytical results show excellent agreement.

Absence of the decay of the amplitude of oscillations at long times is quite an unexpected result. Therefore it is worth explaining it in more detail.

### III. DISCUSSION OF THE RESULTS: SIMPLE PICTURE

One trivial situation where the oscillation of the central spin does not decay is that of no interaction between the central spin and the set of environmental spins. In the presence of such an interaction, however, one may still ask what

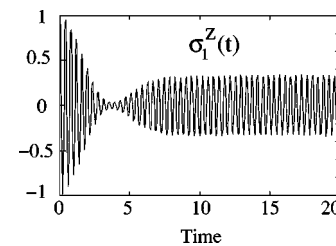


FIG. 2. Analytical result for  $\sigma_1^z(t)$  with the same parameters as those used in numerical simulations.

are the conditions under which this interaction is ineffective in damping the oscillations. A natural suggestion is to try to find a state  $|\Psi\rangle$  of the combined system in which  $\langle\Psi|H_{\text{int}}|\Psi\rangle=0$ . Since in our case  $H_{\text{int}}=2J\vec{C}\vec{S}$ , classically such a state  $|\Psi\rangle$  would correspond to vectors  $\vec{C}$  and  $\vec{S}$  being orthogonal. The condition  $\vec{C}\vec{S}=0$  defines a plane in 3D space, therefore one could argue that for the case of random initial orientation of  $\vec{S}$  the probability of being in the state with  $H_{\text{int}}=0$  is zero. Remarkably, the quantum nature of spins proves the result to be quite different.

The correct way of treating  $H_{\text{int}}$  is, of course, to rewrite it in the following form:

$$H_{\text{int}}=J(\vec{C}+\vec{S})^2-J\vec{S}^2-J\vec{C}^2. \quad (18)$$

Adding spin  $C=1$  with spin  $S$  results in possible values of total spin  $C+S$  being:  $S-1$ ,  $S$ , and  $S+1$ . It is the second case in which the first two terms in Eq. (18) cancel each other. The remaining last term does not depend on  $S$  and, therefore, does not suppress the oscillation amplitude when the averaging over  $S$  is performed and only shifts the oscillation frequency of the central spin [this effect is reflected in Eq. (16)]. The condition “ $C+S=S$ ” is, thus, the closest analog of the classical condition  $\vec{C}\vec{S}=0$ . But, unlike in the classical case, a simple Clebsch-Gordan algebra (see previous section) shows that the probability of being in the subspace “ $C+S=S$ ” is actually finite and is equal to  $1/3$ .

One can easily see now that this effect can only occur if the central system has integer spin. Indeed, the condition “ $C+S=S$ ” can never be satisfied if  $C$  is half-integer. These considerations allow us to formulate the main result of the paper: Based on a particular model of a central spin interacting with randomly oriented environmental spins we have been able to show that the decay of the oscillations of the central spin is essentially different for *integer* central spins: the decay is no longer exponential, instead the amplitude of the oscillations saturates at a constant value.

Moreover, the results presented in this work make clear the physical origin of the unusual two-step decoherence found in Ref. 9, where the generic model Eq. (1) has been considered with all  $J_k$  being different. The first step of decoherence, associated with the initial decay of oscillations to the value of  $1/3$ , has been described in Ref. 9 using a mean-field-like treatment of the spin bath, by replacing the interaction part of the Hamiltonian with a random classical static field having Gaussian distribution. However, such a treatment fails to describe the second step of decoherence, i.e., the long-time slow decay of oscillations. As the results above demonstrate, the representation of a bath as a static random field corresponds to the case of all  $J_k$  being equal to  $J$ . This stems from the fact that the total spin of the bath  $S^2$  commutes with the Hamiltonian (4), so that the bath dynamics in the case  $J_k=J$  is trivial, and can be removed completely by a transformation into the rotating coordinate system. Then, in the rotating coordinate system the effect of the bath on the central spins is equivalent to the action of a random static field. Therefore, the initial decoherence is similar to the

“adiabatic decoherence” by a static spin bath, considered, e.g., in Ref. 17.

Correspondingly, the second step of the decoherence process, i.e., the long-time slow decay of quantum oscillations, can be caused only by an internal evolution of the bath. For all  $J_k$  being different,  $S^2$  does not commute with the interaction part of the Hamiltonian (4), and, as a result, the system-bath coupling induces a nontrivial dynamics inside the bath. It is not surprising that the spin bath possessing a complex dynamics cannot be represented as a random static magnetic field acting on the system. Understanding this “minimally nonadiabatic” decoherence regime represents a challenge for future investigations.<sup>18</sup>

Summarizing, in this work we have demonstrated that decoherence of many-spin systems can drastically differ from decoherence of single-spin systems. This difference originates at the most basic level, and is determined by parity of the central system, i.e., whether the system comprises even or odd number of spin-1/2 entities. Therefore, it is very likely that similar distinction between the central spin systems of even and odd parity is important in many other situations. Moreover, our consideration clarifies the origin of the unusual two-step decoherence found numerically in Ref. 9. The exactly solvable model allows a clear demonstration that the initial step of decoherence (associated with the saturation of oscillations at the value of  $1/3$ ) is caused by “adiabatic decoherence” by a static spin bath, while the subsequent long-time slow decay is induced by a nontrivial internal dynamics of the spin bath. The model is applicable to the qualitative analysis of a range of experimental systems such as magnetic molecules and shallow impurity spins in semiconductors, which experience decoherence from the nuclear spin bath. In these cases, the dominant interaction with the nuclear spins is well approximated by the isotropic Heisenberg interaction (anisotropic interactions are often small).

*Note added in proof.* Recently, we learned of an interesting development in the problem studied here. E. Yuzbashyan *et al.*, cond-mat/0407501, have shown that the quasi-classical equations of motion of the spins are integrable. This work raises two major questions: (1) Can one reproduce the parity effect in decoherence found in the model by using the quasi-classical approximation and studying the asymptotics of the solution? (2) Is the full quantum-mechanical model itself integrable? Answers to these questions can provide better understanding of the decoherence in multi-spin systems.

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