Quantum simulations and experiments on Rabi oscillations of spin qubits: Intrinsic vs extrinsic damping

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Electron paramagnetic resonance experiments show that the decay of Rabi oscillations of ensembles of spin qubits depends noticeably on the microwave power, and more precisely on the Rabi frequency, an effect recently called “driven decoherence.” By direct numerical solution of the time-dependent Schrödinger equation of the associated many-body system, we scrutinize the different mechanisms that may lead to this type of decoherence. Assuming the effects of dissipation to be negligible (\(T_1 = \infty\)), it is shown that a system of dipolar-coupled spins with (even weak) random inhomogeneities is sufficient to explain the salient features of the experimental observations. Some experimental examples are given to illustrate the potential of the numerical simulation approach.

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I. INTRODUCTION

Decoherence generally occurs when the phase angle associated with a periodic motion is lost due to some interaction with exterior noise. In classical mechanics, it may apply to classical waves such as sound waves, seismic waves, or sea waves, whereas in quantum mechanics, it applies to the phase angles between the different components of a system in quantum superposition. The loss of phase of a quantum system may bring it to its classical regime, raising the question of whether and how the classical world may emerge from quantum mechanics. Together with the claim that decoherence is also relevant to a variety of other questions ranging from the foundations of quantum mechanics. It is for all these reasons that the analysis of decoherence in quantum systems must make allowance and, in particular, must distinguish between decoherence induced by the imperfections of real systems and intrinsic decoherence induced by identified or hidden couplings to the environment. The different sources of decoherence can be classified in two main categories: the one-qubit decoherence coming from the coupling of individual qubits with the environment and the multiple-qubit or pairwise decoherence coming from multiple interactions between pairs of qubits.

In this paper, we take the example of paramagnetic spins because of the quality of the systems that can be elaborated (single crystals) and the possibility, offered by magnetism, to start calculations from first principles. Here, the one-qubit decoherence is, in general, associated with phonons and hyperfine couplings, which are intrinsic effects, but also with nonintrinsic effects resulting from weak disorder always present in real systems of finite size: inhomogeneous fields, g-factor distributions, and positional distributions. Multiple-qubit decoherence is generally due to pairwise dipolar interactions with distant electronic or nuclear qubits, which is an intrinsic mechanism. In the following, we shall see that, more generally, when pairwise decoherence takes place in the rotating frame, extrinsic decoherence becomes crucial by itself and also by amplifying intrinsic decoherence. In particular, by way of some examples, it will be shown that the origin of driven decoherence is of the one-qubit type, with multiple possible origins (depending on the nature of the disorder). Even if dominant sources of decoherence may sometimes be identified, the complete description of decoherence and, in particular, the discrimination of intrinsic and extrinsic decoherence, is generally not accessible to experimentalists. This is a major obstacle for the reduction of decoherence, and it holds beyond magnetism. We believe that the present, pragmatic approach should be of great help in common situations where intrinsic and extrinsic decoherence mechanisms interoperate.

Assuming that each type of decoherence has its own “signature” on the Rabi oscillations, we have started a systematic study in which the Rabi oscillations of an ensemble of spins are simulated by direct numerical solution of the time-dependent Schrödinger equation (TDSE) of the associated many-body system. These simulations are performed using a parallel algorithm implementation based on a massively parallel quantum computer simulator. The various mechanisms that may lead to decoherence of Rabi oscillations are successively implemented in Hamiltonians, leading to different types of damping, oscillation shapes, nonzero oscillation averages, and their evolutions with exterior parameters such as the microwave power and the applied static field. The comparison with measured Rabi oscillations allows us to scrutinize the
different decoherence mechanisms and to understand more basic aspects of decoherence, thereby opening a route to search for the optimal (intrinsic and extrinsic) ways to improve coherence of Rabi oscillations, i.e., the number of oscillations that is important for all applications.

This study is limited to the decoherence of Rabi oscillations, that is, the decoherence measured immediately after the application of a long microwave pulse. Following an earlier suggestion,\textsuperscript{15-17} it was shown that the microwave pulse inducing Rabi oscillations is itself an important source of decoherence in all the investigated systems ("driven decoherence\textsuperscript{18-21}"), except when the microwave power is very small, in which case the Rabi frequency is also very small.

As a consequence, the number of Rabi oscillations remains nearly constant, that is, one can not increase it by increasing the microwave power.

This observation can be quantified by comparing the damping time of Rabi oscillations (Rabi decay time \(T_R\)) with the usual spin-spin relaxation time \(T_2\). The theoretical results given in this paper are all exact. Depending on the Hamiltonian parameters, the results were obtained analytically (in simple cases) and numerically (in more general cases, including dipolar interactions) and covered the large range of possibilities, namely, from \(T_R \ll T_2\) up to \(T_R \approx 2T_2\) when dipolar interactions dominate (in the absence of disorder).

The systems used to compare the simulations results with experimental data are insulating single crystals of \(\text{CaWO}_4: \text{Er}^{3+}\), \(\text{MgO}: \text{Mn}^{2+}\), and BDPA (\(\alpha-\gamma\)-bisdiphenylene-\(\beta\)-phenylally), a free radical system often used in electron paramagnetic resonance (EPR) calibration. The latter is not a diluted system, contrary to the two others, but an antiferromagnetic single crystal (identical environments) with a Néel temperature much smaller than the temperature at which our measurements are made (between 4 and 300 K). These systems have been chosen in particular for the differences in their homogeneous and inhomogeneous EPR linewidths. Furthermore, in these systems, the relaxation time \(T_1\) is much larger than \(T_2\), as this is often the case in solid-state systems. For instance, our experiments yield a \(T_1\) that is 10 and 40 times larger than the \(T_2\), respectively. Therefore, as a first step in the theoretical modeling of these experiments, it is reasonable to neglect the effect of dissipation and focus on the decoherence only.

Rabi oscillations measurements have been performed in a Bruker Elexys 680 pulse EPR spectrometer working at about \(f = 9.6\) GHz (\(X\) band). Depending on the sample, measurements have been done at room temperature down to liquid-helium temperature (4 K). The static magnetic field has always been chosen to correspond to the middle of the EPR line. The experimental procedure is illustrated in Fig. 1. A microwave pulse \(P_R\) starts at \(t = 0\) and coherently drives the magnetization. At the end of the pulse \((t = \tau_N)\), the magnetization is recorded. Because of the dead time \((\tau_{de})\) of the spectrometer (about 80 ns), it is impossible to directly measure the magnetization right after the pulse \(P_R\). In this paper, we used two methods for the detection. The first one is simply to record the free induction decay (FID) emitted by the system when the microwave field is shut down. This method gives the value of the magnetization component \(M'\) at the end of the pulse if we take into account two important conditions: (i) \(P_R\) is a nonselective pulse (all spins of the system are excited) and under this condition, the FID signal is the Fourier transform of the EPR line. (ii) The EPR linewidth must be sharp enough. Since the FID is the Fourier transform of the EPR spectrum, a linewidth \(\gtrsim 4\) GHz will lead to a decay time of the FID less than 80 ns, and the FID will be hidden by the dead time of the spectrometer. The second method is used when the EPR line is too broad or if one wants to probe the longitudinal magnetization \(M'\). In this case, another probe sequence has to be used. After the \(P_R\) pulse, one waits a time \(T\) much longer than \(T_2\) but smaller than \(T_1\) in order that \(M'\) vanishes. After the waiting time \(T\), a standard Hahn echo sequence (\(\pi/2 - \tau - \pi - \tau - \text{echo}\)) is used to measure the longitudinal magnetization. In this paper, we do not study (a) the effects of the echo pulses on the measurements and (b) the effect of temperature. For (a), this implies that the comparison with theory is through the measured so-called free-decay time \(T_2^{*}\) (different from the usual \(T_2\)) in which a component of the total magnetization is directly measured through an induction coil, and for (b), that the measurements of \(T_2^{*}\) are done at a sufficiently low temperature, which is quite easy to realize since the \(T_2^{*}\) of \(\alpha-\gamma\)-bisdiphenylene-\(\beta\)-phenylally (BDPA) is
nearly independent of temperature and more generally the Rabi time $T_R$, most important in the context of this paper, also.

The paper is organized as follows. In Sec. II, the quantum spin model is specified in detail and the simulation procedure is briefly discussed. Our results are presented in Sec. III. As there are many different cases to consider, to structure the presentation, the results have been grouped according to the kind of randomness, describing for each kind (i) the noninteracting case, (ii) the interacting case, and (iii) a comparison with experiments if this is possible. In Sec. IV, we present a model of “averaged local Bloch equations,” giving a complete, exact description of one-qubit decoherence and incorporates multiple-qubit decoherence phenomenologically. A summary and outlook is given in Sec. V.

II. MODEL

We consider a system of $L$ dipolar-coupled spins subject to a static magnetic field along the $z$-axis and a circular polarized microwave perpendicular to the $z$-axis. The Hamiltonian reads

$$H = -\mu_B \sum_{j=1}^{L} B_j(t) \cdot g_j \cdot S_j + \frac{\mu_0 \mu_B^2}{4\pi} \sum_{j<k} S_j \cdot g_j \cdot S_k \cdot g_j \cdot \mathbf{r}_{jk},$$

where $B_j(t) = (B'_j \cos \omega t, -B'_j \sin \omega t, B_0)$ denotes the external magnetic field, composed of a large static field $B_0$ along the $z$-axis and a circular time-dependent microwave field $B'_j$ (max \( j \left| B'_j \right| \ll \min_j \left| B_0 \right| \)), which may depend on the position of the $j$th spin, represented by the spin-1/2 operators $S_j = (S_j^x, S_j^y, S_j^z)$ with eigenvalues $\pm 1/2$. The vector $\mathbf{r}_{jk}$ connects the positions of spins $j$ and $k$. It is assumed that the $g$ tensor can be written as $g_j = g_e(1 + \Delta g_j)$, where the perturbation $\Delta g_j$ is a random matrix.

As usual in the theory of NMR/ESR, we separate the fast rotational motion induced by the large static field $B_0$ from the remaining slow motion by a transformation to the reference frame that rotates with an angular frequency determined by $B_0$. Taking the ideal, noninteracting system without fluctuations in the $g$ tensors as the reference system, we define $\omega_0 = g_e \mu_B B_0$ and assume from now on that this ideal system is at resonance, that is, the microwave frequency is given by $\omega = \omega_0$.

The transformation to the reference frame rotating with angular frequency $\omega_0$ is defined by

$$X_{RF} = \exp \left( it\omega_0 \sum_{j=1}^{L} S_j^z \right) \exp \left( -it\omega_0 \sum_{j=1}^{L} S_j^z \right).$$

where $X$ denotes any combination of spin operators. Transforming Eq. (1) to the rotating frame, we find that $H_{RF}$ contains contributions that (i) do not depend on time, (ii) have factors $e^{-i\omega_0 t}$ or $e^{-i\omega_0 t}$, or (iii) have factors $e^{2i\omega_0 t}$ or $e^{-2i\omega_0 t}$. Contributions that depend on time oscillate very fast (because $\omega_0$ is large) and, according to standard NMR/ESR theory, may be neglected, which we have confirmed for a few cases. The remaining time-independent, secular terms yield the Hamiltonian

$$H_{RF} = -\omega_0 \sum_{j=1}^{L} S_j^z - \mu_B g_e \sum_{j=1}^{L} B_j \cdot \frac{g_j^x + g_j^y}{2g_e} S_j^z$$

$$+ \frac{\mu_0 \mu_B^2 g_e^2}{4\pi} \sum_{j<k} \frac{g_j g_k \left[ 1 - 3\xi_{jk}^2/r_{jk}^2 \right]}{g_j^2 r_{jk}^3} S_j^z S_k^z$$

$$+ \frac{g_j^x g_k^z \left[ 1 - 3\xi_{jk}^2/r_{jk}^2 \right] + g_j^y g_k^z \left[ 1 - 3\xi_{jk}^2/r_{jk}^2 \right]}{2g_j^2 r_{jk}^3} \times \left( S_j^z S_k^x + S_j^x S_k^z \right).$$

From Eq. (3), it is clear that variations in $g_j^x$ have the same effect as local variations (inhomogeneities) in the static magnetic field. Inhomogeneities in the microwave field and the variations in $(g_j^x, g_j^y)$ are cumulative. Although the variations in $g_j^x, g_j^y,$ and $g_j^z$ also affect the dipolar interactions, these effects may be difficult to distinguish from the effect of the positional disorder of the spins in the solid, in particular if the spins are distributed randomly. Note that the total magnetization $M^z = \sum_{j=1}^{L} S_j^z$ commutes with the dipolar terms in Eq. (3). Therefore, neglecting the terms that oscillate with $\omega_0$ or $2\omega_0$, the longitudinal magnetization $M^z(t) = \sum_{j=1}^{L} S_j^z$ is a constant of motion in the absence of a microwave field ($B'_0 = 0$).

If all the $g$’s are the same and equal to $g_e$, the Hamiltonian (3) reduces to the familiar expression

$$H_{RF} = -\omega_0 \sum_{j=1}^{L} S_j^z + \frac{\mu_0 \mu_B^2 g_e^2}{4\pi} \sum_{j<k} \left[ 1 - 3\xi_{jk}^2/r_{jk}^2 \right] \times \left( S_j^z S_k^x + S_j^x S_k^z \right)$$

(4)

of the Hamiltonian of dipolar-coupled spins in the reference frame that rotates at the resonance frequency $\omega_0$.

A. Simulation model

We now specify the model as it will be used in our computer simulations. We rewrite the Hamiltonian (3) as

$$H_{RF}/\hbar = -2\pi F_0 \sum_{j=1}^{L} \xi_j^x S_j^x - 2\pi F_R \sum_{j=1}^{L} \frac{(1 + \xi_j^x)(2 + \xi_j^x + \xi_j^y)}{2} S_j^z + 2\pi D_0 \sum_{j<k} \left( \frac{(1 + \xi_j^x)(1 + \xi_k^x)}{r_{jk}^3} S_j^z S_k^x + \frac{(1 + \xi_j^x)(1 + \xi_k^x)}{2r_{jk}^3} \right)$$

$$+ \frac{2\pi D_0}{r_{jk}^3} \left( \frac{(1 + \xi_j^x)(1 + \xi_k^x)}{2r_{jk}^3} \right) \left( S_j^x S_k^x + S_j^x S_k^x \right),$$

where $\hbar$ denotes the reduced Planck constant.
where we take \( F_0 = \omega_0/2\pi \hbar = 9.7 \text{ GHz} \) for the Larmor frequency induced by the large static field, \( F_R = 55.96 \text{ MHz} \) denotes the Rabi frequency for an isolated spin in a microwave field of \( 1 \text{ mT} \), we introduce \( h_4 \) as a parameter to control the amplitude of the microwave pulse, \( D_0 = 51.88 \text{ GHz} \), and we express all distances in Å. With this choice of units, it is convenient to express frequencies in MHz and time in \( \mu \text{s} \). The new dimensionless variables \( \xi'_{\alpha} \) for \( \alpha = x, y, z \) and \( \zeta \) are defined by \( \xi'_x = \xi_0(1 + \xi'_y)^2 \) and \( \mu_R \xi'_y B'_y/\hbar = 2\pi h_4 F_R (1 + \zeta_j) \), respectively. For concreteness, we assume that the spins are located on the Si diamond lattice with lattice parameter \( 5.43 \text{ Å} \) (to a first approximation, the choice of the lattice is not expected to affect the results). Not every lattice site is occupied by a spin: We denote the concentration of spins (number of spins/Å\(^3\)) by \( n \). In experiment, \( n \approx 10^{-4} \ldots, 10^{-6} \).

Guided by experimental results, we assume that the distribution of \( \xi'_n \) is Lorentzian and independent of \( \alpha \), cut off at \( \xi_0 \), and has a width \( \Gamma \):

\[
p(\xi'_n) = \frac{1}{\arctan(\xi_0/\Gamma) - \xi'_n/\Gamma^2 + \Gamma^2 \arctan(\xi_0 - |\xi'_n|)}.
\]

The reason for introducing the cutoff \( \xi_0 > 0 \) is that because the Lorentzian distribution has a very long tail, in practice, we may generate \( \xi' \)'s such that the corresponding value of \( g \) is negative, which may not be physical. Therefore, we use

\[
\xi'_n = \Gamma \tan[(2r - 1) \arctan(\xi_0/\Gamma)],
\]

to generate the random variables \( \xi'_n \) with distribution Eq. (6) from uniformly distributed random numbers \( 0 < r < 1 \). Likewise, the microwave amplitudes are given by \( B'_y = h_p (1 + \zeta_j) \), where the \( \zeta_j \)'s are random numbers with distribution

\[
p(\zeta_j) = \frac{1}{\arctan(\xi_0/\gamma)} \frac{\gamma}{\zeta_j^2 + \gamma^2} \arctan(\xi_0 - |\zeta_j|),
\]

and \( h_p \) is the average amplitude of the microwave field. Appendix A gives a summary of the model parameters that we use in our simulations.

### B. Simulation procedure

The physical properties of interest, in particular, the decay rate \( \zeta_R = 1/T_R \) of the Rabi oscillations and the intrinsic decay rate \( c_2 = 1/T_2 \), can be extracted from the time dependence of the longitudinal and transverse magnetization, respectively, and are defined by

\[
\langle M^z(t) \rangle = \sum_{j=1}^L \langle S^z_j \rangle = \sum_{j=1}^L \langle \Psi(t) | S^z_j | \Psi(t) \rangle,
\]

\[
\langle M^x(t) \rangle = \sum_{j=1}^L \langle S^x_j \rangle = \sum_{j=1}^L \langle \Psi(t) | S^x_j | \Psi(t) \rangle,
\]

respectively. We compute the time-dependent wave function \( |\Psi(t)\rangle \) by solving the TDSE

\[
i\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = H_{RF} |\Psi(t)\rangle,
\]

with \( H_{RF} \) given by Eq. (5). Numerically, we solve the TDSE using an unconditionally stable product-formula algorithm.22 For the largest spin systems, we perform the simulations using a parallel implementation of this algorithm, based on a massively parallel quantum computer simulator.14 Our numerical method strictly conserves the norm of the wave function and conserves the energy to any desired precision (limited by the machine precision).

In analogy with the experimental procedure, we carry out two types of simulations yielding the longitudinal (transverse) magnetization \( \langle M^z(t) \rangle \). From Eq. (5), it follows directly that \( d(M^z(t))/dt = -(M^z(t)) \), hence, \( \langle M^z(t) \rangle \) is directly related to \( M^z(t) \) measured in experiment. We prepare the spin system, that is, the state \( |\Psi(0)\rangle \), such that all spins are aligned along the \( z \) (x)-axis. Then, for a fixed value of the microwave amplitude \( h_p \) (\( h_p = 0 \)) and a particular realization of the random variables \( \xi'_0, \xi'_y, \xi'_z, \zeta_j \) and the distribution of the spins on the lattice, we solve the TDSE and compute Eqs. (9) and (10). This procedure is then repeated several times with different realizations of random variables and distributions of spins. Finally, we compute the average of Eqs. (9) and (10) over all these realizations and analyze its time dependence by fitting a simple, damped sinusoidal function to the simulation data. This then yields the decay rate \( \zeta_R = 1/T_R \) (intrinsic decay rate \( c_2 = 1/T_2 \) ) of the Rabi oscillations.

### III. RESULTS

In the sections that follow, we consider the various sources of decoherence separately. We also study the interplay of intrinsic decoherence due to, e.g., pairwise interactions and extrinsic decoherence due to, e.g., single spins driven by external magnetic fields when different spins have different environments (different couplings to static and microwave fields). The averaging over different spins leads to decoherence, that is, phase destruction of the electromagnetic waves generated by the spins. These two types of decoherence lead to the observed damping of Rabi oscillations, which takes place through energy exchange between the spin system and the applied microwave field. In the following, we show that energy dissipation from the spin bath to the electromagnetic bath is sufficient to explain the experimental results on the Rabi decay time. This is the reason why we neglect, in this paper, the dissipation effect of phonons, (our spin-lattice relaxation time \( T_1 \) is infinite). Note that if we turn off the microwave field, the longitudinal component of the magnetization commutes with the Hamiltonian (5) and hence does not change with time at all, showing that energy exchange with the electromagnetic bath is essential. In the following sections, we give two examples in which energy flows from the electromagnetic bath to the spin bath and from the spin bath to the electromagnetic bath.

#### A. Fixed \( g \)-factors and homogeneous fields

In the absence of randomness on the \( g \)-factors or on the microwave amplitude, the Hamiltonian is given by Eq. (5) with \( \xi^x = \xi^y = \xi^z = \xi = 0 \).
and

\[ H_{RF}/\hbar = -2\pi h_p F_R S_z^n. \]  

(12)

The time evolution of the longitudinal magnetization takes the simple form

\[ \langle \Psi(t)|S_z^n|\Psi(t)\rangle = \frac{1}{2} \cos \Omega_R t, \]  

(13)

showing that the \( z \) component of the spin performs undamped Rabi oscillations with angular frequency \( \Omega_R = 2\pi h_p F_R \). Therefore, \( T_R = \infty \). Furthermore, the transverse magnetization is conserved and, therefore, \( T_2 = \infty \). Summarizing, in the absence of randomness and dipole-dipole interactions, we have

\[ T_R = \infty, \quad T_2 = \infty. \]  

(14)

2. Dipolar-coupled spins

In Figs. 2 and 3, we present simulation results for the longitudinal and transverse magnetization, respectively, as obtained by averaging the solutions of the TDSE over 10 different distributions of 26 dipolar-coupled spins on the lattice. Our simulation results, many of them not shown, lead us to the following conclusions:

(i) For both concentrations \( n = 10^{-3} \) and \( 10^{-4} \) and for microwave amplitudes \( h_p = 0.5, 1, 2 \), the Rabi oscillations decay exponentially. Indeed, the fits are good, as indicated by the small differences between the Rabi frequency \( (F_R = 55.96) \) and the values of \( f \) obtained by the fitting procedure.

(ii) The decay rate \( c_R = 1/T_R \) increases with \( n \), with a slope of approximately 1.7 (data not shown).

(iii) Within the statistical fluctuations resulting from the random distribution of the spins on the lattice, \( c_R = 1/T_R \) does not depend on the microwave amplitude \( h_p \), but strongly depends on the concentration \( n \).

(iv) Simulations (data not shown) for \( n = 0.65 \times 10^{-4}, \ldots, 10^{-3} \) indicate that \( T_2 \propto n \), as expected theoretically.

(v) The simulation data suggest that \( c_2 = 1/T_2 > c_R = 1/T_R \).

Summarizing, in the absence of local randomness but in the presence of dipole-dipole interactions, we have

\[ T_R = T_R(n) > T_2 = T_2(n). \]  

(15)

3. Experimental results: BDPA

We now compare these theoretical predictions to experiments performed on a single crystal of BDPA (\( \alpha-\gamma \)-bisdiphenylene-\( \beta \)-phenylally). With a linewidth of 0.09 mT, this system is quite homogeneous with a very narrow distribution of the \( g \)-factors. Moreover, the sample used was very tiny such that we may consider the microwave to be
homogeneous inside the sample. Results are presented in Fig. 4. They show an example of Rabi oscillations obtained from FID measurements. The Rabi oscillations fit very well to

$$M^y(t) = A_0 \sin(\Omega_R t + \phi) \exp(-t/T_R) + M^y(\infty),$$

(16)

for all microwave powers. The obtained Rabi decay time $T_R$ is clearly independent of the amplitude of the microwave field, as predicted by the model when $\Gamma = \gamma = 0$. It is also very close to $T_2^*$, the FID decay time given by the Fourier transform of the EPR linewidth. This is also in agreement with predictions when $\Gamma = \gamma = 0$ and $D_0 \neq 0$, $T_2^*$ being a coherence time fully equivalent to $T_2$. The discrepancy between $T_R$ ($\sim 140$ ns) and $T_2^*$ ($= 128$ ns) is due to a small inhomogeneous broadening (about 10%).

### B. Randomness in the microwave amplitude only

In the case of randomness in the microwave amplitude only, the Hamiltonian is given by Eq. (5) with $\xi_x = \xi^y = \xi^z = 0$. Such a randomness is inherent to finite-size cavities and becomes smaller as the size of the sample relative to the size of the cavity is reduced.

#### 1. Noninteracting spins

For noninteracting spins ($D_0 = 0$), we can readily compute the average over the distribution of $\zeta$, analytically if we neglect the cutoff of the Lorentzian distribution. As all spins are equivalent, we may drop the spin index $j$ and we obtain

$$\langle S^z(t) \rangle = \frac{\gamma}{2\pi} \int_{-\infty}^{+\infty} \frac{\cos(\Omega_R(1 + \zeta)t)}{\zeta^2 + \gamma^2} \, d\zeta = \frac{1}{2} e^{-\gamma \Omega_R t} \cos(\Omega_R t),$$

(17)

showing that the Rabi oscillations decay exponentially and that the decay time of the Rabi oscillations is given by $1/T_R = \gamma \Omega_R$. Furthermore, the transverse magnetization is conserved and therefore $T_2 = \infty$. Summarizing, in the presence of randomness in the microwave field only and in the absence of dipole-dipole interactions, we have

$$1/T_R = \gamma \Omega_R > 1/T_2 = 0,$$

(18)

showing that the decay rate of the Rabi oscillations increases linearly with the microwave amplitude $h_p$, whereas $T_2$ remains finite. This is easy to understand: $T_2$ is infinite due to the lack of pairwise intrinsic decoherence, whereas destructive interference associated with weak positional randomness in $h_p$ (the microwave field) leads to a reduction of $T_R$ when $h_p$ increases (one-qubit decoherence).

#### 2. Interacting spins: Dipole-dipole interaction

In Fig. 5, we present simulation results for systems of 12 spins with dipole-dipole interaction and randomness in $h_p$, as obtained by averaging over 100 different realizations.
FIG. 5. (Color online) Simulation results as obtained by solving the TDSE for the Hamiltonian (5) for 12 spins (concentration $n = 10^{-4}$) that interact via dipole-dipole interaction, without random fluctuations in the $g$-factors ($\Gamma = 0$) but with random fluctuations on the microwave amplitude ($\gamma = 0.01$). The results represent the average of 100 different realizations of 12-spin systems. Top left to middle right: Longitudinal magnetization for different values of $h_p$. The solid line represents the envelope $(a \pm be^{-ct})/2$ of the function $(a + be^{-ct} \cos 2\pi ft)/2$ that was fitted to the data. Bottom left: Transverse magnetization in the absence of the microwave field ($h_p = 0$). The solid line represents the function $(a + be^{-ct})/2$ that was fitted to the data. Bottom right: Bullets show the inverse relaxation time $c_R = 1/T_R$ as a function of the microwave amplitude $h_p$. The dashed line connecting the bullets is a guide to the eye only. A linear fit to the simulation data yields $c_R = 1/T_R \approx 3.69 h_p + 1.82$ and is shown by the solid line. The horizontal line represents the value of $c_2 = 1/2T_2 \approx 1.95$, estimated from the data of the transverse magnetization (see bottom left).

meaning 100 different distributions of the 12 spins on the lattice. The four upper panels of Fig. 5 show results for the longitudinal magnetization $\langle M_z(t) \rangle$.

Rabi oscillations are damped but have zero offset. The inverse Rabi time $1/T_R = c_R$, deduced from sinusoidal fits, increases linearly with the microwave field, that is, with the Rabi frequency $\Omega_R$ (bottom right). Its value at $h_p = 0$ is to good accuracy equal to $1/2T_2$ ($a_0 \approx 1.82$ for $n = 10^{-4}$).

The slope $a_1 \approx 3.69/F_R$ is related to the matrix of the gyromagnetic factor and to the root mean square of local fields resulting from the randomness in the microwave field. These results, specific to a $h_p$ distribution, agree qualitatively with recently published results of the damping time of Rabi oscillations in the limit of a large inhomogeneous linewidth.24

The results for the transverse magnetization $\langle M_x(t) \rangle$ in the absence of microwaves ($h_p = 0$) are presented in the bottom
left panel of Fig. 5. It clearly decays exponentially, as this is the case with the longitudinal magnetization. Summarizing, from Fig. 5 we conclude that, in the presence of randomness in the microwave field and of dipole-dipole interactions, we have

$$c_R = 1/T_R \approx a_1 \Omega_R + a_0, \quad a_0 \approx 1/2T_2.$$  (19)

Here, pairwise decoherence affects $T_2$, which is now finite (and proportional to $1/\nu$ as in the case without randomness, see Sec. III A) and randomness in microwave amplitude $h_p$ affects $T_R$, which is essentially proportional to $1/h_p$ at large $h_p$. As $T_R < T_2$, we can say that, in this case, energy flows from the spin bath to the electromagnetic bath, leading to energy dissipation in the spin bath.

3. Experimental results: CaWO$_4$:Er$^{3+}$ and MgO:Mn$^{2+}$

In order to show the effect of concentration on Rabi damping, we measure two samples of CaWO$_4$:Er$^{3+}$ with erbium concentration 0.01% and 0.001%, respectively. The two samples have nearly the same shape, keeping the inhomogeneity of microwave field constant. To remove the effects of zero microwave field decay (that is, $T_2$ due to multipitch or pairwise decoherence), we plot $1/T_R - 1/T_{R0}$, where $T_{R0} \approx 1/2T_2$ is the decay time at zero microwave field. The results are presented in Fig. 6. The inverse Rabi decay time fits very well to $1/T_R = 1/T_{R0} + \beta \Omega_R/2\pi$, where $\beta$ is a fitting parameter. From Fig. 6, it is clear that the Rabi decay time $T_R$ decreases with the concentration $n$, in concert with the simulation results.

Evidence of the effect of microwave field inhomogeneity on the Rabi oscillation decay has been recently given for a sample of Cr:CaWO$_4$. To provide further evidence, we took a sample of MgO doped with about 0.001% with Mn$^{2+}$ and cut the sample into a large (3.6 $\times$ 5 $\times$ 0.5 mm$^3$) and small (1 $\times$ 1 $\times$ 0.5 mm$^3$) piece. At this extremely low concentration, the dipole-dipole interaction effect on the Rabi decay is negligible, hence, disorder essentially due to the microwave field inhomogeneity inside these samples will be different. Figure 7 shows the Rabi oscillations for these two samples. All parameters (microwave power, temperature, crystal orientation) are the same for the measurements on these two samples. The effect of the inhomogeneity of the microwave field on the Rabi decay time is clearly seen as the damping in the large sample (red line) is almost two times larger than the one in the small sample (black line).

C. Randomness in the g-factors only

We assume that there are no random fluctuations in the amplitude of the microwave pulse and that the g-factors fluctuate randomly from spin to spin. This effect is generally due to weak crystal distortions, imperfections, leading to small variations of crystal-field parameters.

1. Randomness in $g^z$: Noninteracting spins

In this case, the Hamiltonian is given by Eq. (5) with $\xi_j = \xi^x = \xi^y = D_0 = 0$. As we then have a system of independent spins, we may drop the spin index $j$. In the case wherein, initially, all the spins are aligned along the z-axis, we find

$$\langle S^z(t) \rangle = 0,$$

$$\langle S^x(t) \rangle = \frac{1}{2} \int_{-\infty}^{+\infty} \frac{h_p F_R \sin[2\pi t \sqrt{(F_0 \xi^z)^2 + (h_p F_R)^2}]}{\sqrt{(F_0 \xi^z)^2 + (h_p F_R)^2}} p(\xi^z) d\xi^z + \frac{1}{2} \int_{-\infty}^{+\infty} \frac{\sin[2\pi t \sqrt{(F_0 \xi^z)^2 + (h_p F_R)^2}]}{\sqrt{(F_0 \xi^z)^2 + (h_p F_R)^2}} \frac{2\pi F_0}{\sqrt{(h_p F_R)^2 - (F_0)^2}} \left[ 0 \int_0^t J_0(2\pi h_p F_R u) \sin[2\pi(t - u)\sqrt{(h_p F_R)^2 - (F_0)^2}] du \right].$$

$$\langle S^y(t) \rangle = \frac{1}{2} \left[ \int_{-\infty}^{+\infty} \frac{(F_0 \xi^z)^2 + (h_p F_R)^2 \cos[2\pi t \sqrt{(F_0 \xi^z)^2 + (h_p F_R)^2}]}{(F_0 \xi^z)^2 + (h_p F_R)^2} p(\xi^z) d\xi^z + \frac{1}{2} \left[ \int_{-\infty}^{+\infty} \frac{-(F_0)^2 + (h_p F_R)^2 \cos[2\pi t \sqrt{(F_0 \xi^z)^2 + (h_p F_R)^2}]}{(h_p F_R)^2 - (F_0)^2} \left[ 0 \int_0^t J_0(2\pi h_p F_R u) \frac{1 - \cos[2\pi(t - u)\sqrt{(h_p F_R)^2 - (F_0)^2}]}{(h_p F_R)^2 - (F_0)^2} du \right].$$  (20)
In the case wherein, initially, all the spins are aligned along the $x$-axis, we find

$$
\langle S^x(t) \rangle = \frac{1}{2} \int_{-\infty}^{+\infty} \frac{(h_p F_R)^2 + (F_0 \xi)^2 \cos[2\pi t \sqrt{(F_0 \xi)^2 + (h_p F_R)^2}] p(\xi^z) d\xi^z}{(F_0 \xi)^2 + (h_p F_R)^2} = \\
\frac{1}{2} \left( \frac{(h_p F_R)^2 - (\Gamma F_0)^2 \cos[2\pi t \sqrt{(h_p F_R)^2 - (\Gamma F_0)^2}] }{(h_p F_R)^2 - (\Gamma F_0)^2} - 2\pi \frac{\Gamma F_0 (h_p F_R)^2 }{(h_p F_R)^2 - (\Gamma F_0)^2} \int_0^t J_0(2\pi h_p F_R u) du \right) + 2\pi \frac{\Gamma F_0 \xi^2}{(h_p F_R)^2 - (\Gamma F_0)^2} \int_0^t J_0(2\pi h_p F_R u) \cos[2\pi (t - u) \sqrt{(h_p F_R)^2 - (\Gamma F_0)^2}] du \right),
$$

(21)

Recall that we calculate the transverse magnetization for the case wherein, initially, all spins are aligned along the $x$-axis. In order to obtain the expressions in terms of elementary functions, we have ignored the cutoff of the Lorentzian distribution. We can check that for $\Gamma = 0$, Eqs. (20) and (21) reduce to

$$
\langle S^x(t) \rangle = \frac{1}{2} \cos \Omega_R t, \quad \langle S^y(t) \rangle = \frac{1}{2},
$$

(22)

while for $h_p = 0$, we find

$$
\langle S^x(t) \rangle = \frac{1}{2}, \quad \langle S^y(t) \rangle = \frac{1}{2} e^{-2\pi \Omega z},
$$

(23)

in agreement with the expressions that can be derived directly, without any averaging procedure. From Eq. (23), it follows that $1/T_2 = 2\pi \Gamma F_0$. For finite $\Gamma$, Rabi oscillations are present only if $h_p F_R > \Gamma F_0$ in both longitudinal and transverse cases.

In Fig. 8 (left), we present a typical result for the time dependence of the longitudinal magnetization with $g^z$-factor distribution (only), suggesting that the time-averaged longitudinal magnetization is nonzero, in concert with the analytical expressions

$$
\lim_{T \to \infty} \frac{1}{T} \int_0^T \langle S^x(t) \rangle dt = 0, \quad \lim_{T \to \infty} \frac{1}{T} \int_0^T \langle S^y(t) \rangle dt = 0, \quad \lim_{T \to \infty} \frac{1}{T} \int_0^T \langle S^z(t) \rangle dt = \frac{1}{2} \frac{\Gamma F_0}{h_p F_R + \Gamma F_0},
$$

(24)

The reason for this positive offset is simple: Any nonzero field in the $z$ direction tilts the plane of the Rabi oscillations away from the $(y,z)$ plane, introducing a small precession about the tilted axis superimposed on the Rabi nutation, leading to a positive long-time average. This nonzero offset effect is significant because, as we will see later, it is a unique signature of the presence of random fluctuations in the $g^z$-factor or, equivalently, of the inhomogeneity of the static magnetic field. We emphasize that this nonzero offset is due to randomness and not due to dissipation, as this paper considers the case of $T_1 = \infty$ only.

Similarly, in the case that all spins are initially along the $x$ direction, the long-time average of the transverse magnetization is given by

$$
\lim_{T \to \infty} \frac{1}{T} \int_0^T \langle S^y(t) \rangle dt = \frac{1}{2} h_p F_R \left( h_p F_R + \Gamma F_0 \right),
$$

(25)

with the long-time averages of the two other components being zero. Unlike in the case of the longitudinal magnetization, in the regime where the transverse magnetization shows oscillations ($h_p F_R > \Gamma F_0$), the transverse magnetization reaches its asymptotic value Eq. (25) already after a few oscillations (data not shown).

From Eq. (20), it is clear that we can not expect the amplitude of the Rabi oscillations to decay exponentially in a strict sense. Nevertheless, the data fit well to a function of the form $(a + be^{-\alpha \tau_\text{p}} \cos 2\pi f \tau_\text{p})/2$. The decay rate $\alpha_R$, shown in Fig. 8 (right), decreases with increasing microwave amplitude $h_p$. It seems to diverge when $h_p \to 0$, but this is never observed in experiment. This decrease is a second characteristic feature of the presence of random fluctuations in the $g^z$-factor or, equivalently, of the inhomogeneity of the static magnetic field.

### 2. Randomness in $g^x$ and $g^z$: Noninteracting spins

In this case, the Hamiltonian is given by Eq. (5) with $\xi_j = \xi^z = D_0 = 0$ and we have

$$
\langle S^x(t) \rangle = \frac{1}{2} \int_{-\infty}^{+\infty} \cos[\Omega_R t(1 + (\xi^x + \xi^z)/2)] p(\xi^x) p(\xi^z) d\xi^x d\xi^z.
$$

(26)

Taking the cutoff $\xi_0$ to be infinity, we obtain

$$
\langle S^x(t) \rangle = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \cos[\Omega_R t(1 + \xi)] \frac{\Gamma}{\xi^2 + \Gamma^2} d\xi = \frac{1}{2} e^{-\alpha x^2} \cos \Omega_R t.
$$

(27)
Thus, we conclude that if there is randomness in $g^z$ and $g^y$ only, the Rabi oscillations will decay exponentially with a rate proportional to $\Omega_R = 2\pi h_p F_R$. In the absence of the microwave field, the transverse magnetization is a constant of motion and hence $T_2 = \infty$. Summarizing, in the presence of randomness in $g^z$ and $g^y$ only and in the absence of dipole-dipole interactions, we have

$$1/T_R = \Gamma \Omega_R > 1/T_2 = 0,$$  \hspace{2cm} (28)

showing that the decay rate of the Rabi oscillations increases linearly with the microwave amplitude $h_p$. In fact, Eq. (28) is the same as Eq. (18) with $\gamma$ replaced by $\Gamma$. Thus, we conclude that randomness in $g^z$ and $g^y$ has the same effect as randomness in the amplitude of the microwave field: The Rabi oscillations decay exponentially, with a decay rate that increases linearly with $\Omega_R = 2\pi h_p F_R$. In both cases, decoherence results from a loss of phase of superposed radiation emitted by spins in nutation leading, as a consequence, to energy transfer from the spin bath to the electromagnetic bath. Clearly enough, such dissipation does not involve the usual relaxation time $T_1$ due to dissipation by phonons. This case is very different from the one of, e.g., superconducting qubits where decoherence is dominated by $T_1$ process, as shown for example in Ref. 26.

3. Randomness in $g^x$, $g^y$, and $g^z$: Noninteracting spins

In this case, the Hamiltonian is given by Eq. (5) with $\zeta_j = D_0 = 0$. In Fig. 9 (top), we present a typical result for the time dependence of the longitudinal magnetization. It is seen that the time-averaged longitudinal magnetization is nonzero, signaling the presence of fluctuations in $g^z$ (see Sec. III C 1). Also, clearly visible is the increase of the decay rate $c_R$ of the Rabi oscillations with increasing microwave amplitude $h_p$, a signal of the presence of fluctuations in $(g^z, g^y)$ (see Sec. III C 2). Note that there is no obvious relation between the decay rate of the transverse magnetization [$c_2 \approx 60$, see Fig. 9 (bottom left)] and the values of the decay rate $c_R$ at the smallest values of $h_p$ shown in Fig. 9 (bottom right).

From the results of Secs. III C 1 and III C 2, we may expect that the decay rate $c_R$ shows a crossover from the regime in which the fluctuations on $g^z$ dominate ($c_R$ decreases with increasing $h_p$) and a regime in which the fluctuations on $(g^x, g^y)$ dominate ($c_R$ increases linearly with $h_p$). This is borne out by the data presented in Fig. 9 (bottom right) where we show the combined effect of the two different sources of decoherence, with the widths of the Lorentzian distributions for the longitudinal $(g^z, \Gamma^z)$ and transverse $[(g^x, g^y), \Gamma^x = \Gamma^y]$ fluctuations being varied independently.

4. Experimental results: MgO:Mn$^{2+}$

The combined effect of a distribution in the $g$-factors and inhomogeneities in the microwave amplitude are shown in experiments performed on single-crystalline films of MgO:Mn$^{2+}$ (see Fig. 10 where the measured Rabi decay time is plotted versus the Rabi frequency). The Mn$^{2+}$ dilution is such that dipolar interactions are negligible. Due to weak but sizable distributions of Mn$^{2+}$ local environments, we expect non-negligible and similar distributions of the three $g$-factor components. For small microwave amplitudes, the distribution in the $g^z$-factor gives the dominant, nearly constant contribution to the Rabi decay time, which compares well with Fig. 9 (bottom right). As the microwave amplitude increases, the inhomogeneities associated with transverse components take over and $1/T_R$ increases linearly on the log-log scale. Note that the slope of one-half differs from the slope one that we have for the model considered in this paper. This is because of the peculiarity of the experimental system where nutation takes place coherently over five equidistant levels of the material, an aspect that will be considered in the future. At present, we are interested in showing that the departure from the 1/$T_R$ plateau takes place more rapidly with the larger sample as expected when the effect of microwave inhomogeneities dominates over the one of $g$-factor distributions.
FIG. 9. (Color online) Top left: Time evolution of the longitudinal magnetization as obtained by numerical solution of the TDSE [see Eq. (11)] for the case wherein there are random fluctuations in all three $g$-factors only ($\hat{\mathcal{W}} = \hat{\mathcal{W}}_x = \hat{\mathcal{W}}_y = \hat{\mathcal{W}}_z = 0$). The solid line represents the envelope $(a \pm be^{-ct})/2$ of the function $(a + be^{-ct})\cos(2\pi ft)/2$ that was fitted to the data. Top right: Same as top left, except that $h_p = 10$ instead of $h_p=0.5$. Bottom left: Transverse magnetization in the absence of the microwave field ($h_p=0$). The decay rate $c_2 = 60.19$ is in excellent agreement with the analytical result $c_R = 2\pi \Gamma F_0 = 60.95$ predicted by Eq. (23). The solid line represents the function $(a \pm be^{-ct})/2$ that was fitted to the data. Bottom right: The inverse relaxation time $c_R$ as a function of the microwave amplitude $h_p$ for $\hat{\mathcal{W}}_x = \hat{\mathcal{W}}_y = \hat{\mathcal{W}}_z = 0.001$ (bullets), $\hat{\mathcal{W}}_z = 0.002$ (squares), $\hat{\mathcal{W}}_z = 0.003$ (triangles). The solid line represents the linear fit to the $\hat{\mathcal{W}}_z = 0.001$ data. The dashed lines are guides to the eye only. The number of spins in these calculations is 10000.

5. Dipolar-coupled spins

In Fig. 11 (top and middle), we present simulation results for systems of 26 spins with dipole-dipole interaction (with different concentrations $n$), with random fluctuations in the three $g$-factors and uniform microwave field amplitude. These results are obtained by averaging over 10 different realizations, meaning 10 different distributions of the 26 spins on the lattice. The striking signature of the presence of fluctuations in $g_z$, namely, the nonzero long-time average of the longitudinal magnetization, remains untouched by the effects of the dipolar interactions. For the values of $h_p$ shown in Fig. 11 (top left to middle right), the dependence of the decay rate $c_R$ is essentially the same as if the dipolar interactions were absent [see Fig 9 (bottom right)]. For large $h_p$ (data not shown), the decay rate $c_R$ linearly increases with $h_p$. Comparing Fig. 11 (bottom left) with Fig. 11 (bottom right), it follows that the value of the decay rate of the transverse magnetization is nearly independent of the concentration, hence, can not be attributed to the presence of dipolar interactions, but is mainly due to the presence of fluctuations in $g_z$.

D. Randomness in the $g$-factors and the microwave amplitude

1. Noninteracting spins

In Fig. 12, we present a few representative results for the case wherein there are random fluctuations in both the
FIG. 11. (Color online) The Rabi oscillations of the longitudinal magnetization as obtained by solving the TDSE with the Hamiltonian (5) for 26 spins that interact via dipole-dipole interaction, for different concentrations \( n \), with random fluctuations in the three \( g \)-factors (\( \hat{W} = 0.001 \)) and without random fluctuations in the microwave amplitude (\( \gamma = 0 \)). Top left: \( n = 10^{-3} \). Top right to middle right: \( n = 10^{-4} \). The solid line represents the envelope \((a \pm be^{-cRt})/2\) of the function \((a + be^{-cRt} \cos 2\pi ft)/2\) that was fitted to the data. Bottom: Time evolution of the transverse magnetization for \( n = 10^{-3} \) (left) and \( n = 10^{-4} \) (right). The solid line represents the function \((a + be^{-czt})/2\) that was fitted to the data.

microwave amplitude and in the \( g \)-factors, as obtained by solving the TDSE for the Hamiltonian (5) with \( D_0 = 0 \). In essence, the results are very similar to those of the case where there are fluctuations in all three \( g \)-factors only. This is easy to understand from Eq. (5): Fluctuations in \((g^x, g^y)\) or (exclusive) in the microwave amplitude have the same effect on the decay of the Rabi oscillations. With both types of fluctuations present, our numerical results show that this contribution does not significantly alter the dependence of \( c_R \) on \( h_p \).

As before, the presence of fluctuations in \( g^z \) (see Sec. III C 1) is signaled by the time-averaged longitudinal magnetization being nonzero and by a contribution to the decay rate \( c_z \) of the transverse magnetization, which is in excellent agreement with the analytical result \( c_z = 2\pi \gamma F_0 \) predicted by Eq. (23) (data not shown). Thus, in this case, we obviously have \( c_z > c_R \), which is the same as \( T_R > T_2 \) where \( T_2 \) is reduced by the fluctuations in \( g^z \).

2. Dipolar-coupled spins

In Fig. 13, we present simulation results for systems of 12 spins with dipole-dipole interaction, as obtained by averaging the solution of the TDSE over 100 different distributions of the 12 spins on the lattice, for the case that there are random
fluctuations in the microwave amplitude and in all three
$g$-factors. The four upper panels of Fig. 13 show results for
the longitudinal magnetization. The decay of the longitudinal
magnetization is exponential to good approximation. The
signature of the presence of fluctuations in $g^z$, namely, the
nonzero long-time average of the longitudinal magnetization
is clearly visible. For the values of $h_p$ shown in Fig. 13 (bottom
left), the linear dependence of the decay rate $c_R$ is essentially
the same as if the dipolar interactions were absent [see Fig. 9
(bottom right)].

A linear fit to the data of $c_R$ yields $\lim_{t \to 0} c_R \approx 4.43$.
This value should be contrasted with the result $c_2 \approx 63.74$
for the transverse magnetization in the absence of microwaves
($h_p = 0$) [see Fig. 13 (bottom right)]. Such a large $c_2$ (small $T_2$)
resulting from both dipolar interactions and fluctuations
on all $g$-factors is effectively caused by the effect of $g^z$
fluctuations, in concert with the results shown in Fig. 11
(bottom) that demonstrate that the concentration dependence
is weak, implying that the effect of the dipolar interactions is
small compared to that of the presence of fluctuations in $g^z$.

According to theory, the total decay rate of the transverse
magnetization is the sum of the decay rates due to the dipolar
interactions only and the combined decay rate due to field
inhomogeneities only. From Fig. 5, the former is given by $c_2 \approx 3.90$. In the absence of dipolar interactions, the latter is
given by $c_2 = 2\pi \Gamma' F_0 = 60.95$ (see Sec. III C 3 and Fig. 9,
yielding $c_2 \approx 60.19$ for $\gamma = 0$). Therefore, we have $c_2^{\text{total}} \approx 64.85$, in very good agreement with the value $c_2 = 63.74$
extracted from the simulation (see bottom right panel of Fig. 13).

### IV. PHENOMENOLOGICAL MODEL

The simulations of the dipolar-coupled spin systems are
rather expensive in terms of computational resources. For
instance, one simulation of a single realization of a 26-
spin system takes about 20 hours, using 512 CPUs on
an IBM BlueGene/P. Such relatively expensive simulations
are necessary to disentangle the various mechanisms that
may cause decoherence but are not useful as a daily tool
for analyzing experiments. Therefore, it is of interest to
examine the possibility as to whether a simple phenomeno-
logical model can capture the essence of the physics of
the full microscopic model. Based on our results, pre-
{

![Figure 12](image_url)
spin components is modified according to

$$\frac{\partial}{\partial t} \langle S(t) \rangle = \begin{pmatrix} -1/T_2 & 2\pi \xi^z F_0 & 0 \\ -2\pi \xi^z F_0 & -1/T_2 & \pi h_p (1 + \zeta)(2 + \xi^z + \xi^\gamma) F_R \\ 0 & -\pi h_p (1 + \zeta)(2 + \xi^z + \xi^\gamma) F_R & -1/T_1 \end{pmatrix} \langle S(t) \rangle,$$

(29)
where we adopt the same notation as the one used in Sec. II A. The phenomenological aspect enters in the introduction of the decay times $T_1$ and $T_2$.

Equation (29) has the same structure as the Bloch equation, but there is a conceptual difference and a practical consequence. The former comes from the introduction of g-factor and microwave field amplitude distributions and the latter offers the possibility to calculate numerically the effects of one-spin decoherence to a high degree of accuracy. As we showed in this paper, one-spin decoherence plays an essential role when several qubits act at the same time. It is then natural to start from the well-known equation of motion of a spin $S = 1/2$, add disorder through distribution probabilities (here of g-factors and microwave field amplitude), and average over the solutions. This leads to the exact knowledge of corresponding one-spin decoherence, namely, to Eq. (29) without the $T_1$ and $T_2$ terms. If we now want to make a link with the Bloch equations, we have just to add the phenomenological damping times $T_1$ and $T_2$ as it is done in the original Bloch equations. The difference between Eq. (29) and the original Bloch equations is that, in the latter, $T_1$ and $T_2$ include all damping contributions, i.e., many-spin and one-spin damping, whereas in the former, $T_1$ and $T_2$ include many-spins damping only, with one-spin damping being calculated exactly.

Before assessing the usefulness of Eq. (29) by comparing its results to the numerical solution of the TDSE of the interacting spin system, it is instructive to analyze the case $\xi^x = \xi^y = \xi^z = \zeta = T_1 = 0$. Then, the solution of Eq. (29) reads as

\[
(S^i(t)) = e^{-i/T_2}(S^i(0)),
\]

\[
(S^i(t)) = e^{-i/2T_2} \cos(2\pi h_p F_R \sqrt{1 - (1/4\pi h_p F_R T_2)^2})(S^i(0)),
\]

where, for simplicity, we have assumed that $(S^i(0)) = 0$. From Eq. (30), it follows that the transverse and longitudinal magnetization decays exponentially with a relaxation time $T_2$ and $2T_2$, respectively. In other words, in the absence of randomness and for $T_1 = 0$, Eq. (29) predicts a factor of 2 between the relaxation time of the Rabi oscillations and the relaxation time of the transverse magnetization, in qualitative (and almost quantitative) agreement with our simulation results of dipolar-coupled spin-1/2 systems with randomness. Thus, model Eq. (29) may give a simple explanation as to why in our simulations we find that extrapolation of $c_R$ to $h_p = 0$ gives, in the presence of dipolar interactions, precisely $c_2/2$ if there is no distribution of g-factors ($D_0 \not= 0, \Gamma = 0$) and a value larger than $c_2$ if there is a distribution of g-factors ($\Gamma > 0$).

If we set $\xi^x = \xi^y = \xi^z = \zeta = 0$, which in principle we should do if we strictly adopt the Bloch-equations approach, we can never recover the linear dependence of the decay rate $1/T_R$ on the microwave amplitude $h_p$. However, if we average over the $\xi$’s and/or $\zeta$ and set $T_2 = \infty$, the results are the same as those obtained from the direct solution of the TDSE of the spin-1/2 system.

In Appendix B, we give a simple, robust, unconditionally stable algorithm\textsuperscript{57} to solve Eq. (29). In Fig. 14, we present some representative results. We used the same parameters for $\Gamma$, $\gamma$, and $h_p$ and changed the phenomenological parameter $T_2$ until we found a fair match with the data of the corresponding interacting system. Taking into account that we did not attempt to make a best fit to these data, the agreement is excellent. In both cases shown in Fig. 14 (and in many others cases not shown), this simple procedure seems to work quite well. This suggests that the simple model Eq. (29) may be very useful for the analysis of experimental data, including the effects of the pulse sequence and pulse shapes, effects that are rather expensive to analyze using the large-scale simulation approach adopted in this paper.

V. SUMMARY AND OUTLOOK

The main results of this paper may be summarized as follows:

(i) The noninteracting spin model can account for the $\Omega_R$ dependence of the decay of the Rabi oscillations if we introduce randomness in the g-factors (all three) and/or in the amplitude of the microwave field. In the case of $g^z$ randomness, the long-time average of the longitudinal

![Image](image_url)
magnetization deviates from zero. This deviation increases as the Rabi frequency decreases and reaches its maximum (1/2) when $h_pF_R/\Gamma F_0 \to 0$. The effect of the $g^z$ distribution on the value of $c_R$ at zero microwave field ($h_p = 0$) is simply related to the value of $\langle M^z(t = \infty) \rangle$, suggesting that this decoherence effect comes from the combination of different spin precessions about the $z$ axes and the nutational motion of spins.

(ii) The dipolar-coupled spin system without randomness in all three $g$-factors and without randomness in the amplitude of the microwave field can not account for the $\Omega_R$ dependence of the Rabi oscillation decay rate, observed in experiment. The decay rate of the Rabi oscillations increases as the concentration of magnetic moments increases, as one naively would expect.

(iii) The dipolar-coupled spin system without randomness in $g^z$ but with randomness in the amplitude of the microwave field and/or randomness in $(g^x, g^y)$ can account for the $h_p$ dependence of the Rabi oscillation decay rate and also for the concentration dependence of this decay rate, just as in the case of noninteracting spins.

(iv) The dipolar-coupled spin system with randomness in all three $g$-factors and with or without randomness in the amplitude of the microwave field can account for the $h_p$ dependence of the Rabi oscillation decay rate and also for the concentration dependence of this decay rate. A salient feature of the presence of fluctuations on $g^z$ (or, equivalently, on inhomogeneities in the static field) is that the long-time average of the longitudinal magnetization deviates from zero, as in the case of noninteracting spins.

For future work, we want to mention that the effects on the decay of the Rabi oscillations of the measurement by the spin-echo pulses themselves may be studied by the simple phenomenological model described in Sec. IV. Among other aspects, not touched upon in this study, are the case where motional narrowing is important or where dipolar interactions are strong enough to induce decoherence by magnons, as recently shown in the Fe$_8$ single molecular magnet. These cases can be treated by the simulation approach adopted in this paper, and we plan to report on the results of such simulations in the near future.

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APPENDIX A: OVERVIEW OF THE MODEL PARAMETERS

For convenience, we list the parameters of our model:

(i) The Larmor frequency $F_0 = \omega_0/2\pi\hbar = 9700$ MHz, which is fixed.

(ii) The Rabi frequency at a microwave amplitude of 1 mT is $F_R = 55.96$ MHz, which is fixed.

(iii) The amplitude of the microwave pulse, controlled by the parameter $h_p$. By convention, if $h_p = 1$, a single isolated spin will perform Rabi oscillations with a frequency of $F_R = 55.96$ MHz. The Rabi pulsation in the microwave field $h_p$ is $\Omega_R = 2\pi F_R h_p$.

(iv) The width $\gamma$ of the Lorentzian distribution of the random fluctuations of the amplitude of the microwave pulse $h_p$.

(v) The width $\Gamma$ of the Lorentzian distribution of the random fluctuations of $g^x$, $g^y$, and $g^z$. Unless mentioned explicitly, we assume that $g^x$, $g^y$, and $g^z$ share the same distribution.

(vi) The dipole-dipole coupling strength $D_0 = 51.88$ GHz, which is fixed.

(vii) The concentration $n$ of magnetic impurities on the diamond lattice.

APPENDIX B: NUMERICAL SOLUTION OF THE PHENOMENOLOGICAL MODEL

As in the case of the Bloch equations, if the relaxation time $T_1$ is finite, it is useful to be able to specify both the initial value $\langle S(t = 0) \rangle = \langle S(0) \rangle$ of the magnetization and its stationary-state value $\langle S(t = \infty) \rangle = \langle S \rangle_0$. Therefore, we extend Eq. (29) to

$\frac{d}{dt}\langle S(t) \rangle = \mathbf{A}\langle S(t) \rangle + \mathbf{b}$, \hspace{1cm} (B1)

where

$\mathbf{A} = \begin{pmatrix}
-1/T_2 & 2\pi\xi^z F_0 & 0 \\
-2\pi\xi^z F_0 & -1/T_2 & \pi h_p(1 + \xi)\xi^z F_R \\
0 & -\pi h_p(1 + \xi)(2 + \xi^x + \xi^y) F_R & -1/T_1
\end{pmatrix}$, \hspace{1cm} (B2)

and $\mathbf{b}^T = \langle S \rangle_0/T_1$. The formal solution of Eq. (B1) reads as

$\langle S(t) \rangle = e^{\mathbf{A}t} \langle S(0) \rangle + \int_0^t e^{\mathbf{a}u} \mathbf{b} du$

$= e^{\mathbf{A}t} \langle S(0) \rangle + \mathbf{A}^{-1}(1 - e^{\mathbf{A}t})\mathbf{b}$. \hspace{1cm} (B3)
We integrate Eq. (B1), that is, we compute $e^{tA}$, using the product formula $e^{tA} = (e^{tA_1/2}e^{tA_2}e^{tA_1/2})^m + O(t^3)$, where $t = \tau/m$, $A = A_1 + A_2$, and

$$A_1 = \begin{pmatrix} -1/T_2 & 0 & 0 \\ 0 & -1/T_2 & 0 \\ 0 & 0 & -1/T_1 \end{pmatrix},$$

$$A_2 = \begin{pmatrix} 2\pi \xi^2 F_0 & 2\pi \xi^2 F_0 & 0 \\ -2\pi \xi^2 F_0 & 0 & \pi \hbar \rho(1 + \xi)(2 + \xi^2 + \xi^6)F_R \\ 0 & -\pi \hbar \rho(1 + \xi)(2 + \xi^2 + \xi^6)F_R & 0 \end{pmatrix}.$$  \hfill (B4)

In detail, we have

$$e^{tA_1} = \begin{pmatrix} e^{-t/T_2} & 0 & 0 \\ 0 & e^{-t/T_2} & 0 \\ 0 & 0 & e^{-t/T_1} \end{pmatrix},$$

$$e^{tA_2} = \begin{pmatrix} 1 - (b/\Omega)^2(1 - \cos \tau \Omega) & (b/\Omega) \sin \tau \Omega & (ab/\Omega^2)(1 - \cos \tau \Omega) \\ -b/\Omega \sin \tau \Omega & \cos \tau \Omega & (a/\Omega) \sin \tau \Omega \\ (ab/\Omega^2)(1 - \cos \tau \Omega) & -a/(\Omega^2) \sin \tau \Omega & 1 - a/(\Omega^2)^2(1 - \cos \tau \Omega) \end{pmatrix}.$$  \hfill (B5)

where $a = 2\pi \xi^2 F_0$, $b = 2\pi \hbar \rho(1 + \xi)(1 + \xi^2 + \xi^6)F_R$, and $\Omega = (a^2 + b^2)^{1/2}$.