Collective decoherence of nuclear spin clusters

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Abstract

The problem of dipole–dipole decoherence of nuclear spins is considered for strongly entangled spin clusters. We consider the pure dephasing part of the dipole–dipole interaction which can be classically interpreted as a random field fluctuating along the quantization axes. Due to the long (but finite) range nature of dipole–dipole interaction this field is expected to be partially correlated at the sites of different spins in the cluster. Consequently our results show that the dynamics of the entangled spin cluster can be described as the decoherence due to interaction with a composite bath consisting of fully correlated and uncorrelated parts. The correlated term causes the slower decay of coherence at larger times. The decoherence rate scales up as a square root of the number of spins, giving the linear scaling of the resulting error. Our theory is consistent with recent experiments reported on decoherence of correlated spin clusters.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Quantum information processing devices are expected to be an efficient tool for solving some practical problems which are exponentially hard for classical computers [1]. Their potential computational performance is achieved by exploiting the quantum evolution of many-particle systems in exponentially large Hilbert space, necessarily including evolutionary steps through entangled states. Experimental implementation of Shor’s quantum factoring algorithm in a seven spin-1/2 nuclei molecule has been demonstrated [2].

The question of whether a scalable implementation of quantum computer is possible in the near future implies therefore the question of whether one can protect the fragile entangled states from a destructive environment. The dynamics of coherence loss of entangled many-particle clusters has attracted much attention recently. Some authors have simulated the noisy
environment as a single bosonic bath embracing the whole cluster [3–6]. Classically we may think of some fluctuating random field affecting each particle in the cluster identically [7]. Such an idealized situation occurs, for example, if the characteristic wavelength of the environmental bosonic modes (for example, the characteristic phonon wavelength) is much larger than inter-particle distances (cluster size). An alternative idealized situation in which the noise sources acting on the constituents of each cluster are uncorrelated was also studied [5, 8, 9]. Here, the perturbations are considered to be statistically independent and should be described in the quantum case by individual baths related to each particle. In general, however, we cannot expect that all particles of a cluster of a finite nonvanishing size will be subject to exactly the same or completely uncorrelated fluctuations. Therefore, a realistic model of the environment should be somewhere between these two limiting cases, since both long and short range environmental modes can contribute to the interaction. Still, a quantitative account for a partially correlated environment greatly complicates the analysis [7], even for a two-particle system [10]. A specific example of a highly correlated system which is nowadays under extensive study [11, 12] is the entangled cluster of interacting nuclear spins. Until recently, experimental data on the decoherence of large clusters of highly entangled particles were also unavailable. In 2004 the coherence dynamics of groups of up to 650 entangled nuclear spins was observed for the first time [13]. Our study is motivated by this experimental breakthrough indicating the partial correlation of the environment.

In this paper we derive the dependence of decoherence rates of large spin clusters due to completely correlated and uncorrelated perturbations. The results are generalized to the system consisting of nuclear spins $I = 1/2$ experimentally studied in [13] by using a solid-state nuclear magnetic resonance (NMR) technique for powdered adamantane samples. Our results show that its dynamics resembles the decoherence due to interaction with a composite bath with a given ratio of correlated and uncorrelated terms. The dependence of the decoherence rate on the number of spins in the cluster was obtained.

This paper is organized as follows. The investigated system and experimental procedures are described in section 2. In section 3 we calculate the dynamics of the NMR signal for the cases of totally correlated/uncorrelated external perturbations and for the experimental situation when decay is caused by internal dipole–dipole interaction. Comparisons with experimental data and discussions are given in section 4. Concluding remarks are summarized in section 5.

2. System

Our study was motivated by the recent results of Krojanski and Suter [13]. In their experiments the system of nuclear spins $I = 1/2$ (protons) of the powdered adamantane sample was explored by NMR methods. Initially, a system placed in the external magnetic field $H_0$ along the $z$ axes is in thermal equilibrium. It is described by its density operator

$$\rho_{\text{eq}} = \frac{1}{2^N} \left( 1 + \frac{\gamma h H_0}{kT} \sum_j I_j^z \right),$$

(1)

where $N$ is the number of spins, $\gamma$ is the spin gyromagnetic ratio, $k$ is the Boltzmann constant, $T$ is the temperature and $I_j^z$ is the $z$ component of the $j$th spin operator. With the help of a special sequence of radio-frequency pulses [13] the high-order correlations between spins grow, thereby creating an ensemble of weakly coupled spin clusters as sketched in figure 1. As a result, to describe the evolution of spins in the sample it suffices to consider only the dynamics of one such cluster with a well defined number of spins $n$ [13–15]. This is called the preparation step.
Existence of high-order coherences in an $n$-spin system can be formally described by the presence of the off-diagonal elements $\rho_{ij}$ of the spin density operator in any representation whose basis states can be characterized by the total quantum magnetic numbers: $M, |i⟩ = I_z |i⟩$, $M, |j⟩ = I_z |j⟩$. Following the notation used in multiple quantum NMR experiments [16] we say that every off-diagonal density matrix element $\rho_{ij}$ represents a coherence of the order $M$ where $M = M_i - M_j$. The number of coherences (different off-diagonal elements) of the order $M$ in $n$-spin system at large $n$ is given by

$$C_{2n}^{n-M} = \frac{(2n)!}{(n-M)!(n+M)!} \propto \frac{2^n}{\sqrt{n}} \exp \left( \frac{M^2}{n} \right).$$

(2)

It is conventional to assume that after a long pulse sequence spins are prepared in the state described by the density operator $\rho(0) = U(\tau)\rho_{eq}U^\dagger(\tau)$ with all even coherences excited with equal probability [13, 14]. Here the unitary propagator $U$ governs the evolution of the system during the preparation step of duration $\tau$.

After the system has been prepared in this highly correlated state it decays under dipole–dipole interaction given by the Hamiltonian

$$H_{dd} = \sum_{j<k} d_{jk} (3I_j^z I_k^z - I^z \cdot I^z),$$

(3)

where $d_{jk} = \frac{1}{2}\hbar^2 \gamma^2 (1 - 3 \cos^2 \theta_{jk})/r_{jk}^3$ and $r_{jk}, \theta_{jk}$ are the corresponding absolute value and the angle with $z$ direction of the vector connecting $j$th and $k$th spins.

The system, evolving according to

$$\rho(t) = \exp \left( -\frac{i}{\hbar} H_{dd} t \right) \rho(0) \exp \left( \frac{i}{\hbar} H_{dd} t \right),$$

(4)

does not produce an experimentally observable signal. To analyse the effect of dipole–dipole interaction, it undergoes a conversion step by another sequence of radio-frequency pulses [14] of duration $\tau'$ described by the propagator $V$: $\rho(t + \tau') = V(\tau') \rho(t) V^\dagger(\tau')$. During this step multiple-quantum coherences are converted back to single-quantum longitudinal magnetization. After applying a resonant frequency $\pi/2$ pulse which converts the longitudinal magnetization into transverse magnetization the resulting longitudinal magnetization can be determined by measuring the free induction decay. The free induction decay amplitude right after $\pi/2$ pulse is proportional to

$$S(t) \propto \text{Tr}[I_z \rho(t + \tau')] = \text{Tr}[I_z V \rho(t) V^\dagger],$$

(5)

where $t$ is the time for which the system freely evolved under the dipole–dipole Hamiltonian between the end of the preparation step and the beginning of the conversion step. Under a special pulse sequence design [14] it is possible to achieve the condition of time reversal, $V^\dagger = U$. Using this condition, and the fact that $\rho_{eq} \propto I_z$, we obtain

$$S(t) \propto \text{Tr}[\rho(t) \rho(0)].$$

(6)

The experiment has to be repeated for a sequence of decay times $t$ to obtain the coherence decay. The overall signal can be presented as a sum of the contributions corresponding to different coherence orders $M$ [13]

$$S(t) = \sum_M S_M(t).$$

(7)

The decay times for $S_M(t)$ were also measured experimentally [13] as a function of coherence order $M$ for different cluster sizes $n$. 

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3. Theory

3.1. Decay of NMR signal due to uncorrelated/correlated external baths

First of all, consider a model when the decay of coherence occurs due to interaction with the external bath. We do not specify the bath itself and use the generic picture. In other words, in this subsection we consider the system without dipole–dipole interaction between spins. Instead we introduce some interaction with the external bath which causes the initial spin coherence to decay. In this paper we focus only on the dephasing part of this interaction. A classical analogue of such a model can be a cluster of spins in a fluctuating external magnetic field directed along the $z$ axis [7].

Henceforward, we use the Zeeman basis $|a⟩ = |a_1, . . . , a_n⟩$, where $a_i = ±1$ and $I^i_z|a⟩ = (a_i/2)|a⟩$. If we consider the interaction of a single spin with the bath, its evolution in the Zeeman basis is given by

$$\rho_{±1,±1}(t) = \rho_{±1,±1}(0); \quad \rho_{1,−1}(t) = \rho_{1,−1}(0)e^{−\Gamma(t)}, \quad \rho_{−1,1}(t) = \rho_{−1,1}(0)e^{−\Gamma(t)},$$

where we used the interaction representation and the explicit form of the decay function $\Gamma(t)$. The additional condition

$$\sum |c⟩|c⟩ = \text{const}$$

certainly depends on the degree of correlation of the bath at different spin sites.

3.2. Decay of NMR signal due to uncorrelated/correlated external baths

As the first example we consider the limiting case of a completely uncorrelated environment: each spin interacts with its own bath assuming there are no correlations between the baths related to different spins. In this case the matrix elements of the $n$-spin system density operator evolve according to [3]

$$\rho_{ab}(t) = \rho_{ab}(0)e^{−\Gamma_{ab}(t)},$$

where the collective decay function $\Gamma_{ab}(t)$ can be expressed in term of the single-spin decay function $\Gamma(t)$ as $\Gamma_{ab}(t) = f \Gamma(t)$ and $f = (1/2) \sum |a_i − b_j|$ is the Hamming distance between the spin states $|a⟩$ and $|b⟩$. The value of Hamming distance $f$ has the same parity as the coherence order $M$ and is within the limits $f ∈ [M, n]$. The number of configurations for given $f$ and $M$ for the system of $n$ spin-1/2 can be found as

$$2^{n−f} C^n_M C^f_f \approx \frac{2^{2n}}{\pi \sqrt{n f}} \exp \left[−\frac{(f−n/2)^2}{n/2} \right] \exp \left[−\frac{M^2}{2f} \right].$$

We can calculate the observable decay of the NMR signal $S(t)$ according to (6), (9) as

$$S(t) = \sum_{a,b} |\rho_{ab}(0)|^2 \exp[−\Gamma_{ab}(t)],$$

where we need to carry out the summation over all possible amplitudes $|\rho_{ab}|$. The signal contributions $S_M(t)$ can be evaluated by the use of the same formula (11). Although in this case one needs to take the sum over only the subset of configurations $|a⟩|b⟩ ∈ \mathbb{M}$ for which the additional condition $\sum |a_j − b_j| = 2M$ is satisfied. The situation is greatly simplified by assuming that all even coherences are initially excited with equal probability: $|\rho_{ab}(0)| = \text{const}$ if $(1/2) \sum |a_j − b_j| = 0, 2, 4, . . . ;$ while all other coherences are nonexistent ($\rho_{ab}(0) = 0$) [13, 14]. We can write

$$S_M(t) \propto \sum_{a, b \subset \mathbb{M}} \exp(−if \Gamma(t)).$$

Integrating over all $f$ with corresponding weight (10) we obtain, for $n \Gamma(t) \lesssim 1, n \gg 1$ and $M \leq n/2$,

$$S_M(t) = \exp\left(−\frac{n}{2} \Gamma(t)\right).$$
We are interested in times up to $1/e$ decay time where formula (13) is valid. Moreover, since $n\Gamma(t) \lesssim 1$ the decay function is only in the onset regime: $\Gamma(t) \lesssim 1/n \ll 1$ for $n \gg 1$. Therefore, we take only the lowest non-vanishing order of the decay function in time

$$\Gamma(t) = \alpha t^2 + O(t^3).$$

Using (13) and (14) we obtain

$$SM(t) = \exp\left(-\frac{n}{2}\alpha t^2\right).$$

We emphasize that while we used the short-time expansion for the decay function (14) we expect the formula (15) to be valid up to $1/e$ decay time for a large number of spins $n \gg 1$.

As a second example, we consider the case of a completely correlated environment when the whole cluster interacts with the same external bath. The dynamics of the density matrix elements is given by [3]

$$\rho_{ab}(t) = \rho_{ab}(0) \exp(-M^2 \Gamma(t)), \tag{16}$$

and the signal $SM(t)$ decays as

$$SM(t) = \exp(-M^2 \Gamma(t)) \approx \exp(-M^2 \alpha t^2). \tag{17}$$

Formula (15) should be compared with (17). Both results show that the decay of signal $SM(t)$ can be approximated by the Gaussian function up to $1/e$ decay times for $n \gg 1$. However, the $M$ dependences for the two formulae are totally different. The decay of $SM(t)$ for an uncorrelated environment does not demonstrate any dependence on coherence order $M$, while for correlated case it strongly depends on it. Thus, we established the distinctive features of the influence of correlated/uncorrelated environments on spin cluster dynamics which can be observed experimentally by NMR methods.

3.2. Decay of the NMR signal due to internal dipole–dipole interaction

In the experiment by Krojanski and Suter [13] the decoherence is caused not by the external bath but due to internal dipole–dipole interaction between spins. However, as we show below, the resulting behaviour of the system can be interpreted with the help of results obtained in the previous subsection.

The dipole–dipole Hamiltonian (3) commutes with the Zeeman Hamiltonian

$$H_Z = -\gamma \hbar H_0 \sum_j I_j^z. \tag{18}$$

However, the complexity of the system, and especially the fact that two terms $I_j^x I_k^x$ and $I_j^y I_k^y$ do not commute, makes it impossible to find the exact (analytical or numerical) solution to the problem [14, 16, 17]. Existence of high-order coherences in the state described by the prepared density operator $\rho(0)$ also complicates the application of the traditional method of moments, which enables one to describe the decay of coherence without solving explicitly for eigenvalues and eigenstates of energy in the case of single-quantum NMR experiments [16, 18]. In our case the decay of signal is not proportional to the autocorrelation function $\text{Tr}\{I_x(t)I_x\}$, as in the case of decay of the free induction signal [17], but is given by the density operator correlator (6) where one needs to evaluate the summation of an exponentially large number of terms. In order to obtain the analytical results, we focus on the pure dephasing effect of dipole–dipole interaction, neglecting any energy exchange between spins that is described by the flip-flop term $I_j^x I_k^x + I_j^y I_k^y$ in the Hamiltonian (3). The dipole–dipole dephasing Hamiltonian has the form

$$H_{dd} = 2 \sum_{j<k} d_{jk} I_j^z I_k^z. \tag{19}$$
Because dephasing is not associated with the energy transfer mechanism it is generally the fastest source of decoherence [19, 20]. It becomes the sole process for decoherence in the limit of ‘unlike spins’ [16, 17] when spin exchange is suppressed. The consideration of only this type of interaction enables analytical calculations which are also justified by good agreement with experiment over a wide range of parameters, as will be demonstrated below.

In the Zeeman representation, the off-diagonal density matrix elements evolve according to

$$\Gamma_{ab}(t) = \frac{i}{2} \sum_{j<k} d_{jk} (a_j a_k - b_j b_k).$$

The dynamics of the normalized NMR signal (11) can be analytically expressed as

$$S(t) = 16 \prod_{j<k} \sum_{a_k, a_j, b_k} \left| \rho_{ab}(0) \right|^2 \cos^2 \left(\frac{1}{2} d_{jk} t\right).$$

The exact analytical expression (21) does not yet provide us with much information. Specifically, we intend to obtain the explicit dependence on number of spins in the cluster. For this purpose we again assume that all even coherences are initially excited with an equal probability and the size of the cluster is large $n \gg 1$ [13, 14]. After performing some algebra (the details are given in the appendix) we obtain the expression for the normalized signal

$$S_M(t) = 1 - p M_2^2 \alpha^2 / 2 - (1 - p) n \alpha^2 / 2 + O(t^4),$$

in the second order in time. Here $\alpha = M_2 / 9$ where $M_2 = (9/4) \hbar^2 \sum_j d_{jk}^2$ is the Van Vleck expression for the second moment [17] and the degree of correlation $p$ is defined as

$$p = \frac{1}{n} \left(\sum_j d_{jk}\right)^2 / \sum_j d_{jk}^2,$$

so that $0 \leq p \leq 1$. Formula (22) is valid only at short time scales $n \alpha t^2 \ll 1$, while we are also interested in much larger times up to $n \alpha t^2 \sim 1$. However, expansion of the signal $S_M(t)$ in higher orders in time becomes exceedingly difficult. Therefore, to continue (22) to the longer times we use the analogy with the investigated limiting cases (15) and (17). Formula (22) contains two terms proportional to $M^2$ and $n/2$ which can be regarded as contributions from correlated and uncorrelated perturbations to spin dynamics, respectively. In fact, the interaction described by Hamiltonian (19) can be semiclassically interpreted as the perturbing magnetic
field at the site of each spin (parallel or antiparallel to the strong external magnetic field) produced by all other spins in a cluster. The consequent spread of Larmor frequencies for different spins in the cluster causes destructive interference, or dephasing, observable by the decay of the NMR signal. The limit of totally correlated perturbation \( p = 1 \) corresponds to the case \( d_{jk} \equiv \text{const} \) leading to the same perturbing field for each spin in the cluster. In contrast, the case of absolutely random coefficients \( \langle d_{jk} \rangle = 0 \) gives \( p = 0 \) and fully uncorrelated dynamics. The realistic situation is expected to be in between these two limiting cases. Thus, we write (22) as

\[
S_M(t) = p \exp(-M^2 \alpha t^2) + (1 - p) \exp\left(-\frac{n}{2} \alpha t^2\right),
\]

which is mathematically exact in up to the second order in time but continued to the longer times \( n \alpha t^2 \sim 1 \). The total magnetic resonance signal from the cluster \( S(t) \) can be obtained by summation over all contributions from different coherence orders \( S_M(t) \) according to formulae (7) and (24)

\[
S(t) = \frac{p}{\sqrt{n \alpha t^2 + 1}} + (1 - p) \exp\left(-\frac{n}{2} \alpha t^2\right).
\]

In order to understand whether the obtained formulae (24) and (25) adequately describe the real experimental situation we should check them with experimental data. A comparison of the presented theory and experiment is given in the next section.

4. Comparison of theory with experiment, and discussion

Recent experiments [13] allowed us to estimate the degree of correlation parameter for spin clusters in adamantane samples. In figure 2 we show curves of the decay rates of various
coherence orders for different cluster sizes fitted to experimental points. The decoherence rate was defined as the inverse of $1/e$ decay time and was evaluated by solving the algebraic equation $S_M(t) = 1/e$ where $S_M(t)$ is given by formula (24). The degree of correlation $p$ and the Van Vleck second moment $M_2$ were extracted with the use of MATLAB software by weighted least squares fitting to experimental data for every cluster size $n$. We minimized $\sum_i (f(x_i) - f_i)^2/\Delta_i^2$, where $x_i$ and $f_i$ are experimental points, $f(x_i)$ are corresponding theoretical solutions and $\Delta_i$ are experimental errors denoted by vertical bars in figure 2. Obtained values of $p$ and $M_2$ are given in table 1. As follows from the formula, the definition of second moment [18] is determined by geometrical configurations and does not depend on cluster size $n$. Its moderate fluctuations around an average value ($M_2 = (1.60 \pm 0.05) \times 10^9$ s$^{-2}$) can be attributed to experimental errors and corrections at small $n$. Obtained values for the second moment are comparable but not identical with the previous theoretical estimates and experimental measured values $M_2 \approx 2.6 \times 10^9$ s$^{-2}$ for powdered solid adamantane [21, 22]. The difference can be explained either by the crude nature of the chosen model and the neglect of flip-flop terms or by discrepancy in the adamantane samples used in different experiments. The question could be resolved by additional measurement of the second moment $M_2$ for the given sample.

Taking the average value of $M_2 = 1.6 \times 10^9$ s$^{-2}$ and values for $p$ from table 1 it is possible to predict the temporal dependence of the total NMR signal from the highly correlated spin cluster (25) which was measured independently [13] for different cluster sizes. The results shown in figure 3 are in good agreement with experiment. As can be seen from
Figure 4. Examples of temporal dependence of the signal from a highly correlated spin cluster with size $n = 116$ and three values of degree of correlation $p$ for perturbation: $p = 0$ (dashed line, uncorrelated perturbation), 0.33 (solid line, partial correlation corresponding to experimental situation), 1 (dotted line, correlated perturbation). The inset shows the decoherence rate as a function of coherence order $M$.

The formula (25) allows us to analyse the influence of the degree of correlation on spin dynamics. Figure 4 shows the decay of the NMR signal for spin clusters of intermediate size $n = 116$ and three representative examples of degree of correlation $p$: $p = 0$ (uncorrelated dynamics), $p = 0.33$ (partially correlated dynamics corresponding to the experimental situation) and $p = 1$ (correlated dynamics). One can see that initially all three curves decay equally. However, at later times the signal from the spin cluster subject to correlated perturbation exhibits slower decay compared to that for uncorrelated perturbation. That result comes from the behaviour of decoherence rate as function of coherence order $M$. As can be seen from the inset of figure 4, for uncorrelated perturbation all coherence orders decay with the same, comparatively high, rate $(n/\alpha)^{1/2}$. By contrast, the decay rate for correlated spin dynamics increases linearly with absolute value of $M$ as $|M|^{1/2}$. For the most probable configurations, which according to (2) are those with $M \approx 0$, the decay rate for correlated perturbation is actually less than that for uncorrelated perturbation. The fact that the correlated environment is acting more delicately on specific groups of states is not surprising. In particular,
quantum computing error avoidance schemes based on decoherence-free subspaces [4, 23] are based on this property.

For implementation of large-scale quantum computation, the scaling of decoherence rate with number of qubits is important. From the expression (25) it transpires that the decoherence rate of a spin cluster defined as inverse 1/decay time always increases as \( \propto \sqrt{n} \) with number of spins \( n \), although the corresponding factor depends on degree of correlation \( p \). The square root of \( n \) scaling was indeed experimentally discovered recently by Krojanski and Suter [13].

For quantum information processing applications it is also important to evaluate the error of a quantum computer, represented by a cluster of highly correlated spins, induced by dipole–dipole interaction between spins. The error is defined as the deviation of the NMR signal from its initial value due to decoherence processes during the time required for elementary gate operation \( t_g \): \( \delta_n = 1 - S(t_g) \). In order to provide successful implementation of quantum error correction schemes, one needs to maintain this error below the small threshold guaranteeing fault-tolerance operation of these procedures [1]. Taking the smallness of the parameter \( \delta_n \) into account, one can use (25) to obtain

\[
\delta_n \propto n t^2. \tag{26}
\]

This shows that if the error is small it scales linearly with number of spins independently of the degree of correlation. The linear scaling of error agrees with theoretical results for bosonic models of the environment [6, 9], and suggests that the worst case scenario of ‘superdecoherence’ [3] is not realized for this particular system.

5. Summary

In summary, we have presented a theory of coherence decay of entangled spin clusters states due to internal dipole–dipole interactions. Its dynamics resembles the decoherence due to interaction with a composite bath consisting of fully correlated and uncorrelated parts. The perturbation due to correlated terms leads to the slower decay of coherence at larger times. The decoherence rate scales up as a square root of the number of spins giving a linear scaling of the resulting error. The results obtained can be useful in the analysis of decoherence effects in spin-based quantum computers.

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Appendix

The signal contributions \( S_M(t) \) can be evaluated by use of the formula (11)

\[
S_M(t) = \sum_{a,b \in M} |\rho_{ab}(0)|^2 \exp[-\Gamma_{ab}(t)]. \tag{A.1}
\]

Here domain \( \mathcal{M} \) denote all configurations \( |a\rangle \langle b| = |a_1 \ldots a_n\rangle \langle b_1 \ldots b_n| \) related to the certain coherence order \( M \):

\[
\sum_j (a_j - b_j) = 2M. \tag{A.2}
\]
For every configuration $|a⟩⟨b|$ we can divide the total set of $n$ spins into two subsets $E$ and $N$,

$$a_i = b_i, \quad \forall i \in E; \quad a_j = -b_j, \quad \forall j \in N. \quad (A.3)$$

By the use of definitions (A.3) the decay function $\Gamma_{ab}(t)$ can be simplified as

$$\Gamma_{ab}(t) = i \frac{t}{4} \sum_{j,k} d_{jk}(a_ja_k - b_jb_k) + i \frac{t}{4} \sum_{j \in N, k \in E} d_{jk}(a_ja_k - b_jb_k) + i \frac{t}{2} \sum_{j \in E, k \in N} d_{jk}(a_ja_k - b_jb_k) = it \sum_{j \in E, k \in N} d_{jk}a_ja_k. \quad (A.4)$$

Assuming that all even coherences are initially excited with equal probability, namely $|\rho_{ab}(0)| = \text{const}$ for all $a, b \in \mathbb{N}$ (and for all other even order coherences), we obtain the following formula for the decay of $S_M(t)$ according to (A.1) and (A.4)

$$S_M(t) \propto \sum_{a, b \subset \mathbb{N}} \exp \left[ -it \sum_{j \in E, k \in N} d_{jk}a_ja_k \right]. \quad (A.5)$$

We can redistribute the summation in (A.5) in the following way

$$\sum_{a, b \subset \mathbb{N}} = \sum_{E, N} \sum_{a_j, a_k \in \mathbb{N}}. \quad (A.6)$$

Here the first sum in the left part of the equation is over all possible choices of subsets $E, N$ in the set of $n$ spins and second sum is over all possible values of $a_j, a_k$ for $i \in E, k \in N$. It is easy to see that values $a_j$ for $j \in E$ can take any values $a_j = \pm 1$ since they do not contribute to (A.2) and, therefore, do not change coherence order $M$. We still have the condition for the values $a_k, k \in N$:

$$\sum_{k \in N} a_k = M. \quad (A.7)$$

Thus, we can evaluate the summation over $a_j$ for $j \in E$ first and obtain

$$S_M(t) \propto \sum_{E, N} \sum_{a_j, a_k \in \mathbb{N}} \prod_{j \in E} \cos \left( t \sum_{k \in N} d_{jk}a_k \right) \prod_{j \in E} \left( 1 - \frac{t^2}{2} \sum_{j \in E} \sum_{k \in N} d_{jk}a_k \right)^2 + O(t^4). \quad (A.8)$$

Now we consider the term $\sum_{j \in E} (\sum_{k \in N} d_{jk}a_k)^2$ to express it in terms of parameters of the material. We write

$$d_{jk} = \bar{d}_j + \delta_{jk}, \quad (A.9)$$

where average coupling constant is defined as

$$\bar{d}_j = f^{-1} \sum_{k \in N} d_{jk}. \quad (A.10)$$

Here $f$ is Hamming distance between $|a⟩$ and $|b⟩$ or the number of spins in subset $N$. Note, that $\sum_{k \in N} \delta_{jk} = 0$. By use of (A.7) and (A.9) we obtain

$$\left( \sum_{k \in N} d_{jk}a_k \right)^2 = (\bar{d}_j)^2 M^2 + \sum_{k \in N} \delta_{jk}^2, \quad (A.11)$$
where we neglected cross-terms $\sum_{k \in N} \delta_{jk} a_k$ whose contribution is negligible for the large cluster sizes. For $n \gg 1$ we also approximate the summation over subsets by the summation over total cluster with correction to the number of terms in the sum: $\sum_i = (n/(n-f)) \sum_{j \in \mathbb{E}} = (n/f) \sum_{j \in \mathbb{N}}$. These assumptions should lead to the asymptotically correct value of $S_M(t)$ for $n \to \infty$. We then evaluate the signal decay as

$$S_M(t) \propto \sum_{E,N} \left[ 1 - \frac{t^2}{2} \frac{n - f}{n} (p M^2 + (1 - p)(n/f) \sum_i d_{ik}^2) \right] + O(t^4), \quad (A.12)$$

where the parameter $p$ is defined as

$$p = \frac{1}{n} \left( \sum_j d_{jk}^2 \right)^2 / \sum_j d_{jk}^2. \quad (A.13)$$

After integration over all possible $f$ we deduce the closed, analytical form for the signal, exact up to second order in time,

$$S_M(t) = 1 - \frac{t^2}{2} \frac{M_2}{9} \left( p M^2 + \frac{(1 - p) n}{2} \right) + O(t^4), \quad (A.14)$$

Here $M_2 = (9/4)\hbar^{-2} \sum_j d_{jk}^2$ is Van Vleck expression for the second moment [17].

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