
**ORDER, DISORDER, AND PHASE TRANSITION
IN CONDENSED SYSTEM**

Decay of Multispin Multiquantum Coherent States in the NMR of a Solid

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Abstract—A model based on the Anderson adiabatic approximation, which is widely used for describing various aspects of dynamic phenomena in conventional radiospectroscopy, is proposed for describing the decay of multispin multiquantum coherent states in a solid. The coherent state relaxation function is represented by the product of two functions corresponding to spin precession in a two-component local field with a correlated and an uncorrelated component. Theoretical results of this study explain the experimental data reported in a number of publications and are in good agreement with these data.

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1. INTRODUCTION

The dynamic behavior of multispin multiquantum (MQ) coherent states formed upon exposure of the nuclear spin subsystem of a substance in the condensed matter to a certain sequence of radio frequency (rf) pulses underlies the NMR MQ spectroscopy of a solid [1].

On the one hand, MQ NMR is a powerful and often indispensable tool for studying clusters and local structures arranged, for instance, on surfaces [2], in liquid crystals [3], and nanosize cavities [4]. On the other hand, the improvement of MQ spectroscopic methods has made it possible to study experimentally the time evolution of multispin correlations by observing the coherent states formed using MQ NMR [5–10]. The formation and propagation of multiparticle correlations is of fundamental interest for statistical mechanics of irreversible processes [11]. Finally, multiparticle spin correlations may serve as a “quantum register” (see, for example, [5–9]) in quantum computations. The quantum states “prepared” in the nuclear spin system can be controlled by a sequence of rf pulses by initiating various processes (e.g., processing of quantum computations in implementation of quantum algorithms).

One of the main factors preventing the formation of a large quantum register is relaxation (decay) of the above-mentioned multispin time correlation functions (TCFs) due to the spin–spin interactions. Thus, the following two processes are in fact competing in the dynamics of coherent MQ states: (i) the formation of the above TCFs and (ii) their relaxation. If the rate of formation of coherent states is much higher than

their relaxation rate, a large quantum register can be obtained. Otherwise, it is impossible.

It can be seen from the above arguments that the relaxation of MQ states depending on time, quantum number, and the register size is one of the central (and most complicated) problems in MQ spin dynamics. These dependences were studied experimentally on protons in adamantane in [5–8] and on fluorine in CaF₂ in [9]. In these publications, the relaxation of MQ states was associated either with the secular part of the nuclear dipole–dipole interaction or with this interaction suppressed by sequences of rf pulses (see below).

The first purely phenomenological description of some results obtained in [5], which was based on the conversion of formal power expansions up to the second order in time into exponential TCFs, was given in [12]. The function describing the decay of coherent states was phenomenologically decomposed into the sum of two functions, one of which described the spin dynamics in the mean field, while the other described the spin dynamics in an uncorrelated (completely random) field. In spite of satisfactory agreement with experiment (the dependences of the coherence loss rate on quantum number M (see below) and on the number of spins in a cluster with a given value of M have been constructed), it is extremely difficult to provide a physically motivated substantiation of the decomposition of the sought TCF into the sum of (two) functions for the homogeneous system under investigation at high temperatures. At the same time, the separation of contributions to the dipole–dipole local magnetic field has been used for a long time in explaining the dynamics of the nuclear spin system of

a solid. For example, decomposition of a local field (is given in units of frequency) at a certain preferred (arbitrary) spin into two components, $H_{\text{loc}} = H_1 + H_2$ (H_1 corresponds to the nearest neighbors (of the cell) and H_2 , to more remote spins (far surroundings)) has made it possible to explain for the first time the characteristic features of the free precession signal (Fourier transform of the NMR absorption spectrum) in a solid [13]. Indeed, let H_1 and H_2 be two statistically independent contributions to a random local field with distributions functions in the form of a rectangle, $P_1(H_1)$, and a Gaussian function, $P_2(H_2)$. Then $P(H) = P_1(H_1)P_2(H_2)$, and we obtain the following expression for the free precession signal:

$$G(t) = \int_{-\infty}^{\infty} e^{i\omega t} P(H) dH = \int_{-\infty}^{\infty} e^{i\omega_1 t} P_1(H_1) dH_1 \times \int_{-\infty}^{\infty} e^{i\omega_2 t} P_2(H_2) dH_2 = \frac{\sin(bt)}{bt} \exp\left(-\frac{a^2 t^2}{2}\right),$$

which correctly explains experiment [14]. The approach described here in simplified form is based on the Anderson adiabatic approximation [14, 15]. Subsequently, this approach has been considerably developed and successfully used for solving a number of problems in spin dynamics. It can be seen that in the approach considered here, the separation of the contributions to the local field leads to the product of the corresponding correlation functions and not to their sum.

In this study, we solve the formulated problem of relaxation of MQ states using a modification of the above-mentioned popular Anderson model. In Section 2, the nuclear system under investigation and the method for observing MQ states are described. The theory of the decay of coherent MQ states under the action of the secular part of the dipole–dipole interaction is expounded in Section 3. In Section 4, the decay of MQ states is considered in the case when the nuclear spin system evolves during the relaxation time under the action of not the secular dipole–dipole interaction, but a considerably suppressed interaction obtained from the secular part of the dipole–dipole interaction by exposing the nuclear spin system to a specially constructed sequence of rf pulses. The results are discussed and compared with experimental data in Section 5. Finally, the formal derivation of one most essential formulas of this article is given in the Appendix.

2. DYNAMICS OF COHERENT MQ STATES IN A SOLID

It is well known [14] that the main reason for NMR line broadening in nonmetallic diamagnetic solids is the secular part of nuclear dipole–dipole interactions,

which completely determines the dynamics of the nuclear spin system:

$$H_d = \sum_{i \neq j} b_{ij} S_{zi} S_{zj} - \frac{1}{2} \sum_{i \neq j} b_{ij} S_{+i} S_{-j} = H_{zz} + H_{ff} \quad (1)$$

$$= \sum_{i \neq j} (3/2) b_{ij} S_{zi} S_{zj} - \frac{1}{2} \sum_{i \neq j} b_{ij} \vec{S}_i \vec{S}_j = H_{zz} + H_{ex},$$

where $b_{ij} = (1/2) \hbar^2 (1 - 3\cos^2 \theta_{ij}) / r_{ij}^3$, \mathbf{r}_{ij} is the vector connecting spins i and j , θ_{ij} is the angle formed by vector \mathbf{r}_{ij} with a constant external magnetic field, γ is the gyromagnetic ratio, and $S_{\alpha i}$ is the α component ($\alpha = z, +, -$) of the vector spin operator at site i . Here and below, the energy is expressed in the frequency units.

Usually, when impulse methods are used in NMR of solids, base Hamiltonian (1) is transformed with the help of “spin alchemy” (various sequences of rf pulses) into other Hamiltonians that are of interest for researchers [16]. For example, in traditional NMR MQ spectroscopy, the initial Hamiltonian is transformed into the effective Hamiltonian [17, 18]

$$H_{\text{eff}} = -\frac{1}{2} \sum_{i \neq j} b_{ij} (S_{+i} S_{+j} + S_{-i} S_{-j}), \quad (2)$$

which is nonsecular relative to external magnetization. Under the action of this Hamiltonian, the initial magnetization is “pumped” into various TCFs of a complex structure (depending on the product of a various number Q of spin operators that will be referred to as clusters) over the so-called preparatory period of duration τ . In other words, the equilibrium low-temperature density matrix in a strong constant magnetic field H_0 , which has the form [14]

$$\rho_{\text{eq}} = 1 + \frac{\hbar H_0}{kT} \sum_{j=1}^N S_{zj},$$

where k is the Boltzmann constant, T is the temperature, and N is the total number of spins in the sample, is transformed into a nonequilibrium density matrix, which can be conveniently represented as the sum of nondiagonal elements ρ_M with a certain difference of magnetic quantum numbers M called coherent MQ states (M is the degree of coherence):

$$\rho(t) = \exp(-iH_{\text{eff}} t) \rho_{\text{eq}} \exp(iH_{\text{eff}} t) = \sum_M \rho_M(t), \quad (3)$$

$$\rho_M(t) = \sum_{Q=M}^N \sum_{\{i\}} \sum_p g_{QM_p\{i\}}(\tau) |Q, M, p, \{i\}\rangle;$$

here $|Q, M, p, \{i\}\rangle$ is the basis operator in which Q one-spin operators form the product connecting the Zeeman states differing by M unities and $\{i\}$ are the number of crystal lattice sites occupied by the given cluster. Thus, $\{i\}$ is actually a multi-index. Summation over $\{i\}$ implies summation over a set of clusters as well as over

the set of spins in each cluster. The expression being summed depends only on the difference of the coordinates defined by it (i.e., there is no dependence on one of the coordinates). Having defined this coordinate arbitrarily, we find that the expression summed over other coordinates decays quite rapidly. Here, we use the term cluster for the group of spins for which the expression to be summed is not negligibly small. Index p labels different basis states with identical values of Q and M . The coherent states formed during the preparation period are tagged with the help of phase shift proportional to time [17, 18]. The emerging phase shift is proportional to M , where M is an integer. Thus, Q -spin correlations differ in the number of quanta as well ($M \sim Q$) [1, 17–19]. These coherent states subsequently relax over time t under the action of secular dipole–dipole Hamiltonian (1) (or the same Hamiltonian averaged over the pulsed sequence [16]).

By the end of the period of free evolution, a new pulsed sequence is applied to the system, which reverses the sign of effective Hamiltonian (2) and thus performs “time inversion” [20, 21], owing to which the order is again pumped to the observable (single-quantum longitudinal magnetization). The amplitude of the partial (for the given value of M) magnetization can be measured with the help of a $\pi/2$ pulse rotating the magnetization to the plane perpendicular to the external magnetic field. To determine the relaxation rate, the experiment is repeated multiply for different values of t .

It should be emphasized that the MQ states can be observed only under certain conditions, as a consequence of which all contributions to the coherence of the given order appear after the restoration (mixing) period with the same phase [17]. In view of fundamental importance of the above circumstance, we consider this aspect of MQ spectroscopy in detail.

The amplitude and phase of partial magnetization are completely determined by the previous history of evolution of the spin system. For example, if the evolution of the system at the preparatory stage occurs under the action of Hamiltonian (2) (the corresponding propagator is $U(\tau) = \exp(-iH_{\text{eff}}\tau)$), while the evolution during mixing period t occurs under the action of a certain (generally different) Hamiltonian H'_{eff} (the corresponding propagator is $V(t) = \exp(-iH'_{\text{eff}}t)$), the total magnetization amplitude is described by the following expression [17]:

$$\begin{aligned} \rho_0(\tau, t, \varphi) &= \text{Tr}\{S_z(\tau, t, \varphi)\} \\ &= \text{Tr}\{S_z V^\dagger(t) \exp[-iH_d t] U^\dagger(\tau) S_z U(\tau) \\ &\quad \times \exp[iH_d t] V(t)\}. \end{aligned}$$

We calculate the trace of this expression in the basis of eigenfunctions of secular dipole–dipole Hamilto-

nian (1), denoting them by $|i\rangle$ and $|j\rangle$. Writing complex matrix elements in the form

$$\begin{aligned} P_{ij}(\tau) &= \langle i | U^\dagger(\tau) S_z U(\tau) | j \rangle, \\ Q_{ij}(t) &= \langle i | V^\dagger(t) S_z V(t) | j \rangle, \end{aligned}$$

we obtain

$$\rho_0(\tau, t, \varphi) = \sum_{i,j} P_{ij}(\tau) Q_{ji}(t) \exp[-i(\epsilon_i - \epsilon_j)t].$$

Here, ϵ_i and ϵ_j are the eigenvalues (in units of frequency) of Hamiltonian (1). It follows from the above arguments that if the Hamiltonians controlling the evolution of the spin system at the preparatory stage and over the mixing period are different, the MQ states formed during the preparatory stage will only be additionally transformed during mixing. If, however, the operator controlling the evolution of the system during mixing period is constructed so that $V^\dagger(t) = U(\tau)$ or differs from $U(\tau)$ only in phase factor φ , i.e.,

$$V^\dagger = \exp(-iS_z)\varphi \exp(iS_z)$$

(it is precisely this situation that takes place upon time inversion), the observed signal acquires the form of a Fourier series in the coherent state of various orders [17]:

$$\rho_0(\tau, y, \varphi) = \sum_n \sum_{i,j} |P_{ij}|^2 \exp(in\varphi) \exp[-i(\epsilon_i - \epsilon_j)t].$$

Thus, for the above evolution of the events, each partial coherent state includes the contribution from all coherent states of the given order, differing from the neighboring state only in phase by $\pm \varphi$.

Proceeding from the simplest statistical model [17, 18], it is assumed in experiments that the distribution of coherent states of different orders in the multiquantum spectrum is Gaussian:

$$g_M(\varphi) = \exp[-M^2/N(\varphi)].$$

Dispersion $N(\varphi)/2$ of the distribution in this model is determined by the number $N(\varphi)$ of spins between which dynamic correlation sets in over preparation time τ due to interaction (2). This number, which is known as the number of correlated spins or the effective size of a cluster, increases with preparation time τ . In Section 5, while describing the experiment, we will denote this quantity by K .

3. RELAXATION OF MQ STATES UNDER THE ACTION OF THE SECULAR PART OF THE NUCLEAR DIPOLE–DIPOLE INTERACTION

We consider an individual spin cluster from the sum in formula (3), which contains Q spins, is located in set $\{i\}$ of crystal lattice sites with a fixed geometry, and has degree of coherence M . During free evolution under the action of interaction (1), the operator component

of the density matrix generally changes. These changes can also be accompanied by variations of parameters M , Q , p , and $\{i\}$. If, however, the cluster is large, we can disregard the changes in number M (Hamiltonian (1) commutes with the Zeeman Hamiltonian), in dipole energy, and in the remaining numbers for not very long time periods. Therefore, it is expedient to use in further analysis the Anderson model [14, 15] (adiabatic approximation), in accordance with which we will assume that only the phase of the operator component of the density matrix changes:

$$(M, Q, p, l, \{i\} | t) = |M, Q, p, l, \{i\} \times \exp \left[-i \int_0^t dt_1 (M, Q, p, l, \{i\} | t_1) \right]. \quad (4)$$

Here, l is the number of operators S_{zk} in operator $|M, Q, p, l, \{i\}$. If we confine our analysis to not very long time intervals, we can assume that the longitudinal component of the local field, which mainly determines phase relaxation, is static [22]. Then we obtain the following expression for a cluster containing n operators S_{+k} and m operators S_{-k} ($M = n - m$):

$$(M, Q, p, l, \{i\} | t) \quad \{i\}_{0M}(t) = \prod_j \exp \left[-i \sum_{\substack{k=1 \\ k \in \{i\}}}^n b_{kj} - \sum_{\substack{q=1 \\ q \in \{i\}}}^m b_{qj} \right] 2S_{zj} t. \quad (5)$$

Indices “ k ” and “ q ” on the right-hand side of this expression denote only the spins of the cluster under investigation, while index “ j ” denotes the spins that do not belong to the selected cluster.

The local dipole magnetic field acting on each spin of the cluster splits into two components: a certain mean value, which is approximately the same for all spins of the cluster (i.e., independent of the spin number in the cluster) and is produced, in the case of dense clusters, by the spins located at distances much larger than the cluster size, and the component depending on the spin number in the cluster. Naturally, the most probable value of each of these fields is zero due to the high-temperature approximation used here. We can now write relation (5) in the form

$$\{i\}_{0M}(t) = \prod_{j,j'} \exp(-ih_{\{i\}j}^0 M 2S_{zj} t) \times \exp \left[-i \sum_{k=1}^n b_{kj} - \sum_{q=1}^m b_{qj'} \right] 2S_{zj} t. \quad (6)$$

Here, we assume that

$$h_{\{i\}j}^0 = \frac{1}{m+n} \sum_{k=1}^n b_{kj} + \sum_{q=1}^m b_{qj} \quad (7)$$

is the mean field produced by “remote” spins, which is the same for each spin k and q in the cluster under

investigation. Indices j and j' denote the spins contributing to the field component independent of the spin number in the cluster and depending on it, respectively.

The main contribution to each product in Eq. (6) comes from different spins so that their contributions to phase relaxation are independent. Averaging over operators $\{S_{zj}\}$ and $\{S_{zj'}\}$ (i.e., calculating the trace), we obtain

$$\{i\}_{0M}(t) = \prod_j \cos(Mh_{\{i\}j}^0 t) \times \prod_{kj'} \cos(b_{kj'} t) \prod_{qj'} \cos(b_{qj'} t). \quad (8)$$

As usual, in the case of a large number of approximately equivalent neighbors surrounding each spin in the lattice and the cluster as a whole (generally, the number of neighbors $Z \rightarrow \infty$), we replace the product of cosines in formula (8) by Gaussian functions [14, 23]. This gives

$$\{i\}_{0M}(t) \approx \exp(-a_{\{i\}}^2 M^2 t^2) \exp[-(m+n) B_{\{i\}}^2 t^2 / 2],$$

where

$$2a_{\{i\}}^2 = \sum_j b_{ij}^2, \quad B_{\{i\}}^2 = \sum_{j'} b_{ij'}^2.$$

A more formal derivation of this relation without using visual images of close and remote spins is given in the Appendix.

Generally speaking, dealing with long time intervals, we should also take into account the time dependence of the z components of cluster spins associated with flip-flop processes (the contributions of these processes to auto-correlation functions of spins were considered in [23]), which would lead to additional decay as compared to that defined by formula (8). Therefore, we substitute $Q = m + n + l$ for $m + n$ in what follows. In addition, coefficients $B_{\{i\}}^2$ and $2a_{\{i\}}^2$ must substantially depend, for example, on the geometry of cluster $\{i\}$. A dense cluster has a larger number of equivalent distant neighbors, while each spin in a loose (dendrite) cluster contains a large number of approximately equivalent neighbors from the number of spins that do not belong to the cluster. Thus, to derive the final expression, we must multiply quantity $\{i\}_{0M}(t)$ by the amplitude of the MQ state and average over the arrangement of spins in cluster $\{i\}$ in the lattice. As in the familiar case of averaging over the orientations of crystallite in a powder, we must average the exponential itself rather than its exponent. In addition, we must carry out averaging over the number Q of spins in the cluster. Since we are not in a position to adequately take into account these (as well as other)

fine details, we write the final expression for the partial magnetization in the form

$${}_{0M}(t) = \exp(-a^2 M^2 t^2) \exp(-Kb^2 t^2/2), \quad (9)$$

where K , a^2 , and b^2 will henceforth be assumed to be certain constants that can be determined from experimental data.

Using formula (9), we can derive the expression for the decay rate (the quantity reciprocal to the time of attenuation by a factor of e):

$$\frac{1}{bt_e} = \sqrt{\frac{K}{2} \left(1 + \frac{2a^2 M^2}{Kb^2}\right)}. \quad (10)$$

To obtain the total signal of transverse magnetization, the partial components from relation (9) must be summed. Replacing the summation by integration, which can be done for not very long time intervals ($a^2 t^2 + 1/K \ll 6$), and following statistical theory [18], we obtain

$$\begin{aligned} {}_0(t) &= \int_0^\infty dM {}_{0M}(t) \frac{2}{\sqrt{K}} \exp\left(-\frac{M^2}{K}\right) \\ &= \frac{1}{\sqrt{a^2 K t^2 + 1}} \exp\left(-\frac{Kb^2 t^2}{2}\right). \end{aligned} \quad (11)$$

4. RELAXATION OF COHERENT STATES UNDER THE ACTION OF THE EFFECTIVE HAMILTONIAN

If a sequence of pulses averaging the dipole Hamiltonian [16] is applied to the system on the time interval of free evolution, relations (4) and (5) generally remain valid, although the relation between functional $\{ \}$ from formula (4) and the dipole constants is significantly complicated. For example, for the MREV-8 sequence, the dipole contribution from Hamiltonian (1) is transformed into the Hamiltonian [16]

$$\bar{H}_d^{(2)} = \frac{2}{18} [(H_{dx} - H_{dy}), [H_{dy}, H_{dx}]]. \quad (12)$$

Here, ρ is the time interval separating pulses,

$$H_{dx} = \exp\left(-\frac{i S_y}{2}\right) H_d \exp\left(\frac{i S_y}{2}\right), \quad (13)$$

$$H_{dy} = \exp\left(-\frac{i S_x}{2}\right) H_d \exp\left(\frac{i S_x}{2}\right),$$

and S_x and S_y are the x and y components of the total spin of the system. It should be noted that along with principal Hamiltonian (12), various cross contributions to the Hamiltonian, which are associated with imperfection of the pulsed action on the system, are also preserved [16].

As in the previous section, we can decompose the local field acting on a spin of the cluster into two components, one of which (associated, for example, with

Results of regressive analysis of the dependence of the coherence loss rate on "quantization" parameter M and number K of correlated spins based on formula (10)

K (τ , ms)	$\frac{Kb^2}{2}$, ms ⁻²	a^2 , ms ⁻²
26 (240)	1490.7	46
41 (300)	2117.4	63
71 (360)	3088	85
116 (420)	4821	109
189 (480)	7658	122
309 (540)	12398	133
477 (600)	17582	190
650 (660)	23145	205

remote spins) is almost the same for all spins of the cluster, while the other (associated, for example, with the nearest neighbors) has different values for different spins. If the number of equivalent spins constituting each group is large, the contribution to relaxation corresponding to short time intervals is decisive. This contribution is described [16] by the second moment of the Gaussian one-spin TCF (the second moment of this function is associated with the second moment of the conventional NMR spectrum) due to dipole-dipole interaction (1):

$$\bar{M}_2(\text{MREV-8}) = M_2, \quad = 0.065 M_2^2 \rho^4. \quad (14)$$

Thus, expression (9) for the partial magnetization turns out to be valid, the only difference being that parameters a and b are reduced in accordance with scaling factor ($a_p^2 = a^2$, $b_p^2 = b^2$). In addition, ratio a^2/b^2 may change because interaction (12) is generally not a paired but a multiparticle interaction.

5. DISCUSSION OF RESULTS AND COMPARISON WITH EXPERIMENTAL DATA

To compare the above theory with the experimental data reported in [5–7] and to explain the results obtained in these publications, the points on the experimental curves describing the dependence of the decay rate of coherent states on MQ coherence order M for different average numbers of involved spins (different cluster sizes K) (see Fig. 5 from [5]) were scanned and digitized. Approximation of these curves by formula (10) using nonlinear regressive analysis has made it possible to determine constants a and b appearing in formula (10). The results are compiled in the table. It should be noted that since the experimental error for large values of M sharply increased (see Fig. 5 from [5]), we used only nine points in regressive analysis (the central point and four points on each side) for each curve.

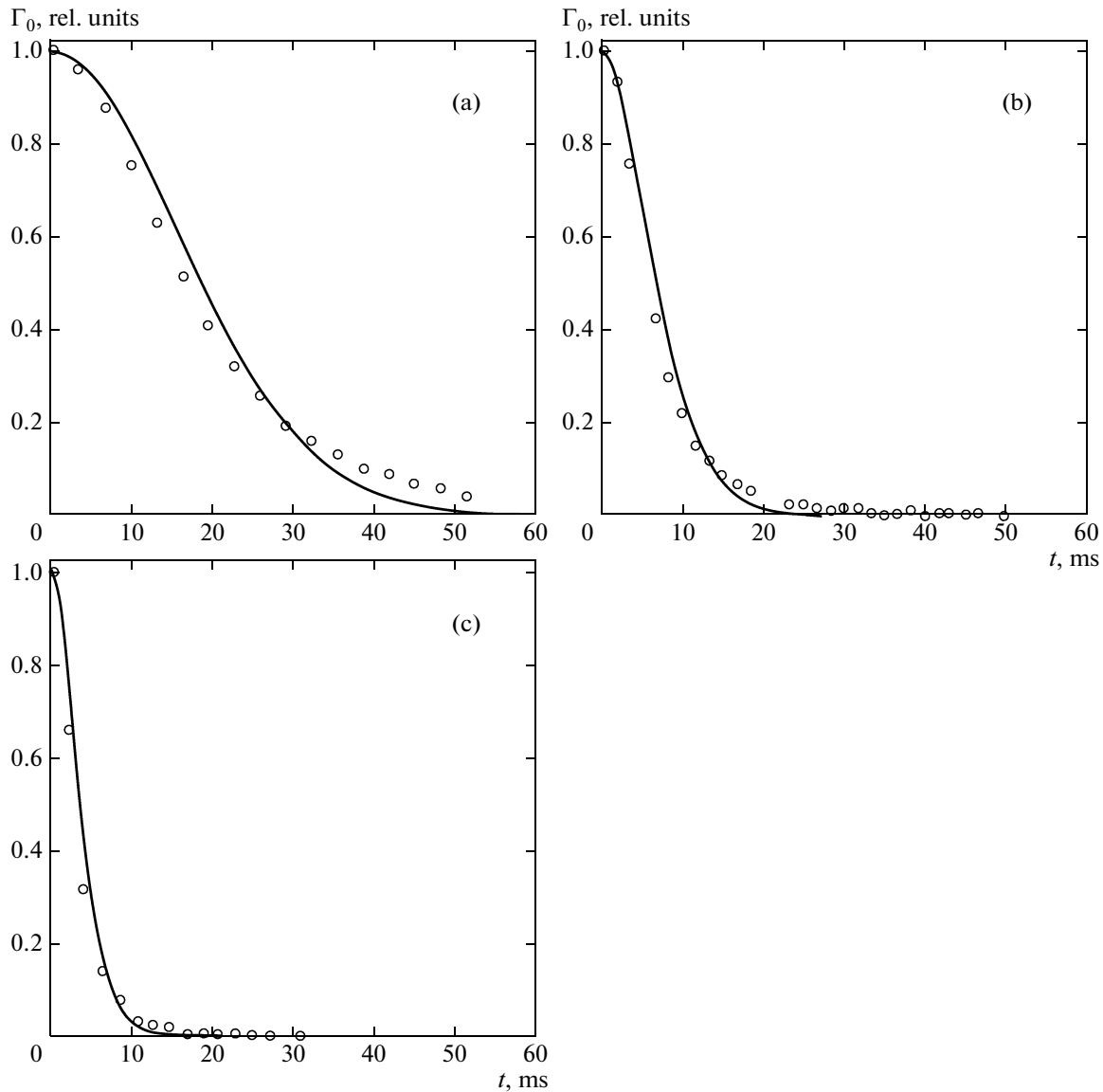


Fig. 1. Decay of multispin coherent clusters for different numbers of correlated spins: $K = 24$ (a), 309 (b), and 650 (c). Circles correspond to experiment, and solid curves are calculated by formula (11) with parameters from the table.

The values of a and b obtained in this way have made it possible to satisfactorily describe, using relation (11), the experimental time dependences of the decay rate of coherent states for different sizes K of the cluster [5]. Figure 1 show examples of comparison of experimental and theoretical results. A relatively slight discrepancy between theory and experiment for large time intervals can be explained by the following factors.

The approximation of time-independent local magnetic fields (formula (5)) gives good results only for short time intervals, for which TCF (9) decreases by a factor of e , while the variation of the contributions from individual spins (cofactors in formula (8)) on this time interval is K times smaller. If, however, we are interested in the decay of coherence states over long

time intervals, we must take into account the time dependence of frequency in expression (4). In the Anderson model for a large number Z of neighbors, the field is described by a random Gaussian process $\tilde{h}(t)$; i.e., $\langle \tilde{h}(t) \rangle = \langle \tilde{h}_0(t) \rangle$. In this case, expression (9) for the conventional dipole–dipole interaction [14] is replaced by the following expression:

$$\begin{aligned}
 {}_0M(t) &= \exp[-2a^2 M^2 {}_d(t)] \\
 &\times \exp[-Kb^2 {}_d(t)],
 \end{aligned}
 \tag{15}$$

where

$${}_d(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \langle \tilde{h}(t_1) \tilde{h}(t_2) \rangle.$$

For short time intervals $t \ll t_c$ (t_c is the correlation time), the TCFs from Eq. (15) remain Gaussian functions of time, while for $t > t_c$, these functions become exponential:

$$\rho_M(t) = \exp(-2a^2 M^2 t) \exp(-Kb^2 t). \quad (16)$$

Further, the values of a and b determined by us make it possible to plot the dependence reflecting the increase in the coherence loss rate (reciprocal time of decay of function $\rho_0(t)$ by a factor of e) from relation (11) on the number K of correlated spins. The equation for the sought time t_e ,

$$t_e^2 K b^2 + \ln(K a^2 t_e^2 + 1) = 2, \quad (17)$$

was solved numerically. Figure 2 shows the resultant dependence on the log–log scale. The straight line is plotted using the least squares method. The equation for this straight line has the form

$$y = 0.528x + 0.886. \quad (18)$$

The values of $y = \log t_e$ calculated with the help of Eq. (18) are in good agreement with the experimental results [5] (see Fig. 2) both in the value and in the form of the dependence on $x = \log K$. Thus, the approach described here has made it possible to connect the results of two different measurements of relaxation rate: for coherent states with a definite number M and the total rate. It should be noted that an analogous root dependence was also observed in CaF_2 [9].

Finally, if averaging pulse sequences exist during free evolution period, time dependence $\rho(t)$ can be described analogously. However, since Hamiltonian (12) represents the four-particle interaction, the time dependence generated by it contains the powers of the random quantity up to the third:

$$\rho_{\text{eff}}^{-(2)}(t) = \rho_{\text{eff},0}^{-(2)} [C_1^{-1}(t) + C_2^{-2}(t) + C_3^{-3}(t)]. \quad (19)$$

A modification of the Anderson model taking into account the quadratic contribution was considered in [24] as applied to narrowing of the NMR line by a strong rf field under magic angle conditions. However, it is only important for our analysis that, according to [24], exponential decay (16) in the case of a quadratic Gaussian process is observed sooner than in the case of TCF (15), which corresponds to experimental results (see right group of the decay curves in Fig. 3 in [6]).

It was noted in the previous section that the action of a narrowing pulse sequence on the system during its free evolution only reduces the relaxation rate of functions $\rho_0(t)$ in accordance with scaling factor K^{-1} , which can amount to many tens (see formula (14)). However, the shape these functions for time intervals over which the corresponding TCF decrease by a factor of e is Gaussian. Thus, the above theory also provides an explanation for another important experimental result from [6]. In that study, the authors analyzed among other things the dependence of the relaxation rates of coherent states (coherence loss) on number K of spins in the cluster for purely dipole–dipole interaction (1)

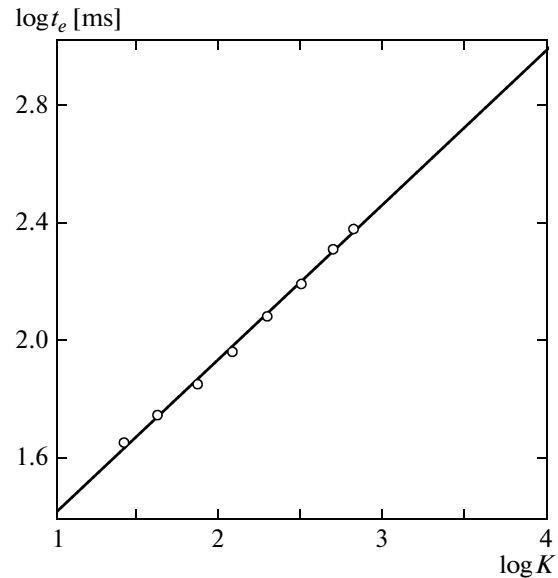


Fig. 2. Dependence of the decay rate of multispin coherent states on the number of correlated spins. Circles correspond to the result of solution of Eq. (17) with parameters from the table; the solid straight line is plotted by the least squares method (formula (18)).

during free evolution as well as in the case when narrowing pulse sequences leading to an effective Hamiltonian analogous to that defined by formula (14) are applied to the sample. It can be seen from Fig. 4 from [6] that these dependences of the relaxation rate for the dipole evolution as well as under the action of a narrowing sequences of pulses on the spin system are completely proportional and differ only in the decay rate by approximately a factor of 50.

Thus, the resultant dependence for the coherence loss rate, which is close to the \sqrt{K} dependence, confirms that the form of the corresponding time dependences is close to Gaussian, which in turn indicates the independence of the dynamics of individual spins in a large correlated cluster over the free evolution period (a large number of independent contributions to the dynamics). In [12], the experimentally observed above-mentioned dependence of the coherence loss rate on the number K of correlated spins was explained assuming that each spin in the cluster relaxes independently from other spins and that the relaxation function of each spin in the cluster is Gaussian. It should be emphasized, however, that for large values of K , the specific form of the relaxation dependence for each spin is in fact immaterial (see above). What is important is the parity of the corresponding function of time. The rest will be “taken care of” by the central limiting theorem. It is for this reason that we observe the root dependences of relaxation rates not only in dipole evolution, but also under the action of narrowing sequences of pulses on the spin system.

6. CONCLUSIONS

The above analysis shows that a slight modification of the Anderson model, which is widely used in radio spectroscopy, makes it possible to satisfactorily describe qualitatively and quantitatively the physical phenomena observed during relaxation of a quantum register. Thus, in spite of the drawbacks of the Anderson model associated with a certain “phenomenology” of the description, this model can be useful for studying relaxation phenomena in coherent spin systems.

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APPENDIX

Performing averaging over operators $\{S_{z_j}\}$ in expression (5) and calculating the trace, we obtain

$$\langle \{i\} \rangle_{0M}(t) = \prod_j \cos \left[\sum_{\substack{k=1 \\ k \in \{i\}}}^n b_{kj} - \sum_{\substack{q=1 \\ q \in \{i\}}}^m b_{qj} \right] t. \quad (\text{A.1})$$

In calculating the coherence damping, we must average expression (A.1) over all possible versions n and m of arrangement of operators S_{+k} and S_{-k} , respectively, in a cluster for $M = n - m$. To perform averaging, we introduce parameter μ_i assuming values ± 1 in accordance with the sign of the operator $S_{\pm i}$ located at site i ,

$$\langle \{i\} \rangle_{0M}(t) = \left\langle \prod_j \cos \left[t \sum_{i=1}^K \mu_i b_{ij} \right] \right\rangle_{\{\mu_i\}}, \quad (\text{A.2})$$

and carry out averaging over random quantities μ_i satisfying the condition $\sum_{i=1}^K \mu_i = M$, where $K = m + n$.

As usual, for a large number of approximately equivalent neighbors surrounding each spin in the lattice (and in the cluster as a whole; generally, the number of neighbors $Z \rightarrow \infty$), we replace the products of cosines in formula (A.2) by Gaussian functions [14, 15, 22]. The decay of a Gaussian function is determined by a second-order correlation function, which is equal to the second-order term in expansion (A.2):

$$\begin{aligned} & \left\langle \sum_j \frac{t^2}{2} \sum_{i=1}^K \mu_i b_{ij} \right\rangle_{\{\mu_i\}}^2 \\ &= \frac{t^2}{2} \sum_j \sum_{i=1}^K b_{ij}^2 \mu_i^2 \end{aligned} \quad (\text{A.3})$$

$$+ \frac{t^2}{2} \sum_j \sum_{q=1}^K \sum_{\substack{i=1 \\ i \neq q}}^K b_{qj} b_{ij} \mu_i \mu_q.$$

Substituting $\mu_i^2 = 1$ and $\mu_i \mu_q = (M^2 - K')/(K')^2$ into formula (A.3), we obtain the following expression for the Gaussian function:

$$\begin{aligned} \langle \{i\} \rangle_{0M}(t) &= \exp \left[-\frac{t^2}{2} \sum_j \sum_{i=1}^K b_{ij}^2 - \frac{t^2}{2} (M^2 - K') \right. \\ &\quad \left. \times \sum_j \frac{1}{K'} \sum_{i=1}^K b_{ij}^2 \right]. \end{aligned} \quad (\text{A.4})$$

Time expansion (A.3) with allowance for (A.4) can be written in the form analogous to that proposed in Appendix of [12]:

$$\begin{aligned} & \frac{t^2}{2} K' B_{\{i\}}^2 + \frac{t^2}{2} (M^2 - K') B_{\{i\}}^2 P_{\{i\}} \\ &= \frac{t^2}{2} K' (1 - P_{\{i\}}) B_{\{i\}}^2 + \frac{t^2}{2} M^2 P_{\{i\}} B_{\{i\}}^2, \end{aligned} \quad (\text{A.5})$$

where

$$\begin{aligned} B_{\{i\}}^2 &= \frac{1}{K'} \sum_j \sum_{i=1}^K b_{ij}^2, \\ B_{\{i\}}^2 P_{\{i\}} &= \sum_j \frac{1}{K'} \sum_{i=1}^K b_{ij}^2. \end{aligned}$$

Further, using an expression similar to (A.5), the sum of correlations functions with preexponential parameter P is written in [12], while here we obtain product (A.4) with parameter P in the exponent. Other differences are associated with the fact that in [12] the cluster is assumed to be dense and summation is hence performed only inside the cluster (indices k and j in formula (A.13) from Appendix in [12]), while we do not proffer beforehand any assumptions concerning the cluster topology. The cluster can also be loose (of dendrite type); in this case, index j corresponds to the surroundings and index i to the cluster itself. In contrast to the expression from [12], expression (A.5) derived here should be averaged over the arrangement of spins of cluster $\{i\}$ over the lattice.

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