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REVIEW

Effect of Degradation Processes Caused by a Small Perturbation on the Growth of the Average Cluster Size of Correlated Spins in Multiple Quantum NMR Spectroscopy of Solids

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Abstract

Multiple quantum (MQ) NMR spectroscopy of solids allows one to observe the growth and decay of multispin correlations. As a rule, the average size of the cluster of correlated spins is extracted from the width of the MQ spectrum. In the present article, the size distribution of such clusters is explored. To obtain the above distribution, the solutions for the amplitudes of the decomposition over complete sets of orthogonal operators for the two different models were used. By means of these models, we have taken into account the dependence of cluster degradation (the degradation of a cluster means, e.g., destruction of correlations in cluster or loss of particles in it) through two positions. The first one defines by the cluster size while the second one depends on the MQ coherence order of the cluster. It is shown that in dependence of the relation the rates of these degradation processes, the width of the MQ spectrum carries different information. If the first process is faster that the second one, then the width of the MQ spectrum is still determined by the average cluster size. When the velocity ratio becomes inverse, the width of the MQ spectrum takes on a smaller value, which is a consequence of the faster degradation of the MQ spectrum components with large orders of coherence.

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1 Introduction

The multiple quantum (MQ) NMR spectroscopy that emerged in the early 1980s has now largely defined the face of not only modern NMR, but also of "spin physics" in general [1–6]. The main reason for this lies in the wide opportunities provided by the MQ NMR for both applied and fundamental research in various fields of natural science: from chemistry, biology and medicine to the theory of quantum information and statistical mechanics of irreversible processes.

In MQ spectroscopy, initially localized quantum information is redistributed over a multiparticle system, involving (generally speaking) all particles, and is accompanied by the appearance of various, in particular, nonlocal correlations. The process of reversible redistribution of quantum information (scrambling) over multiparticle correlations is usually accompanied by irreversible (although, as a rule, partial) disturbances in the transmission process. These disturbances are called loss of coherence (or decoherence), and they can be caused by various factors. Thus, actually, two processes compete in the dynamics of MQ coherences: the development of complicated time correlation functions (TCFs) with cumbersome expressions that reflect the development of the above coherences, and their damage (or total decay) due to decoherence processes.

To study scrambling and to determine its rate, etc., four-particle out-of-time-ordered correlation (OTOC) TCFs are usually used [7–10]. These TCFs contain a lot of specific information about the most intimate processes in a multiparticle system. In addition, experimental studies of MQ NMR in multispin systems have notable advantages over other techniques of exploring OTOC multiparticle systems, such as, for example, ultracold neutral atoms [11] or trapped ions [12]. The point is that the (naturally arising) TCFs used in MQ spectroscopy belong to the class of OTOCs and can be immediately measured experimentally. These are usually four-particle TCFs that by definition contain a time-reversed stage of evolution.

Different relaxation impacts causing loss of coherence to distort the shape of the MQ spectrum; therefore, studies of these processes are very important for a corresponding method for successful measurements: e.g., the spreading of quantum information and any other substantial processes.

That is why purposeful experimental studies of the effect of disturbances causing loss of coherence (degradation or (in some sense) relaxation) in the spin system on the MQ spectra and on the growth of the average cluster size of correlated spins are carried out very intensively (see, for example, [4–6]). In addition, let us mark that earlier, it was experimentally demonstrated in [13–18] that the number of dynamically correlated spins (the number of spins in cluster) grows exponentially with time at least when degradation processes are absent. In articles [19, 20], the above fact has got a robust theoretical foundation.

As it was demonstrated by experiments [4, 5] when a small perturbation causing the clusters degradation is switched on, the width of the MQ spectrum ceases to grow with time. On this basis, the authors of [4, 5] concluded that the average cluster size of correlated spins is stabilized because of faster relaxation of

large clusters since some equilibrium cluster size does exist. In [21, 22], we proposed an alternative possibility for the stabilization of the MQ spectrum width. It was shown that the stabilization of the MQ spectrum width is a consequence of a rapid relaxation of the MQ spectrum components with large order of coherence. In the present work, the conditions for the existence of this specific mechanism will be defined below. For this purpose, we shall consider the distribution of cluster sizes [23] and take into account the dependence of the degradation rate on the cluster sizes and their coherence order [24].

The article is structured as follows. Section 2 contains some general formulas for MQ spectroscopy. Section 3 considers the ideal case without degradation processes. In Sect. 4, we estimate the average cluster size in the presence of degradation, and in Sect. 5, we study the change in the MQ spectrum under these conditions. Finally, Sect. 6 provides a discussion.

2 Hamiltonians and Basic Relations for Correlation Functions

It is well known [25] that the broadening of NMR lines in nonmetallic diamagnetic solids is mainly attributed to the secular part of internuclear dipole–dipole interactions, which completely determines the dynamics of a nuclear spin system:

$$H_d = H_d^{zz} = \sum_{i \neq j} b_{ij} S_{zi} S_{zj} - (1/2) \sum_{i \neq j} b_{ij} S_{+i} S_{-j}, \tag{1}$$

where $b_{ij} = (1/2)\gamma^2 (1 - 3\cos^2 \theta_{ij})/ r^3_{ij}$, r_{ij} is a vector connecting spins i and j, θ_{ij} is the angle between the vector r_{ij} and an external static magnetic field, γ is the gyromagnetic ratio, and $S_{\alpha i}$ is the α -component ($\alpha = x, y, z, +, -$) of the vector operator of spin at site i. Henceforth, we express energy in frequency units. When using pulse methods in the NMR of solids, the basic Hamiltonian (1) is usually transformed by spin alchemy (various sequences of rf pulses) into other Hamiltonians that are of interest for the researcher [26]. For example, in the conventional MQ NMR spectroscopy, the original Hamiltonian is transformed into a two-spin/two-quantum effective Hamiltonian [1–3]:

$$H_{DQ} = (-1/4) \sum_{i \neq j} b_{ij} (S_{+i} S_{+j} + S_{-i} S_{-j}), \tag{2}$$

which is nonsecular with respect to the external static magnetic field. The equilibrium high temperature density matrix in a strong static magnetic field H_0 is represented as [25]

$$\rho_{eq} \propto 1 + \frac{\gamma \hbar H_0}{kT} \sum_{j=1}^{N} S_{zj}.$$

Here, k is the Boltzmann constant, T is temperature, and N is the total number of spins in a sample. Under the effect of Hamiltonian (2) during the so-called preparation period of length t, the original magnetization is transformed into various rather

complicated TCFs that depend on the product of a various number of spin operators K, called clusters. Therefore, this matrix is transformed into a nonequilibrium density matrix, which is conveniently represented as a sum ρ_M of off-diagonal elements with a certain difference of magnetic quantum numbers M, called MQ coherences (M is the order of a coherence, which simultaneously numbers the position of the coherence in the MQ spectrum),

$$\rho_M(t) = \sum_{K=M}^{K=N} \sum_{\{i\}} \sum_{q} g_{KMq\{i\}}(t)|KMq\{i\}>. \tag{3}$$

In Eq. (3), $|KMq\{i\}\rangle$ is a basis operator in which K one spin operators form a product that relates Zeeman states differing by M units and $\{i\}$ is the numbers of sites of the crystalline lattice occupied by a given cluster. Thus, $\{i\}$ is in fact a multiindex. The index q enumerates different basis states with identical values of K and M. The summation over $\{i\}$ implies the summation over both the set of clusters and the set of spins in each cluster. The expression under the summation sign depends only on differences of coordinates; i.e., the dependence on one of the coordinates is missing. Setting this coordinate to be arbitrary, we find that, with respect to other coordinates, the expression under summation decays quite rapidly. Thus, a cluster is a group of spins for which the expression under summation is not negligible for every $S_{zj}(t)$ included in the $\varrho_{\text{eq}}(0)$.

The arising coherences are marked by a phase shift φ [1, 3]. The phase shift is proportional to $M\varphi$, where M is an integer. Therefore K-spin correlations are additionally distinguished at least by the number of quanta ($M \leq K$ for $S = 1/2$) [1–3].

To study the possible loss of coherence in a correlated cluster, its effect on the MQ spectra, and to investigate the growth of the average cluster size with correlated spins under fairly realistic conditions, experimenters usually use a small perturbation introduced into the system in a controlled manner. Therefore, e.g., in articles [4–6] during the preparatory period, to the effects of the double quantum Hamiltonian that produces MQ coherences (Hamiltonian (2)), the Hamiltonian Σ produced perturbations, usually added, with a little coefficient. This coefficient could be changed in experiments.

Accordingly in the articles [4, 5], the authors used Hamiltonian

$$H_p = (1-p)H_{DQ} + p\Sigma. \tag{4}$$

The parameter p authors could change in experiments. The dipole–dipole interaction was chosen as a perturbation in [4], and the dipole–dipole and Zeeman interactions were chosen in [5]. On the contrary, in [6], the unknown perturbation Σ remained unchanged, but the coefficient δ (the multiplier written before the dipole–dipole interaction) could change in the course of experiments by experimenters:

$$H_{\delta,F(B)} = \pm\delta H_d^{yy} + \Sigma. \tag{5}$$

In Eq. (5) H_d^{yy} is the secular part of the dipole–dipole Hamiltonian (1) but secular with respect to the quantization axis Y, not Z. As follows from the results of

work [1–3] for both of the above options, TCFs (which are one of the possible types "OTOC"-correlators) those have been measured in MQ experiments and discussing in the present article can be written in a unified way:

$$\Gamma_{\varphi}(t,\tau) = Tr\{U_2^{+}(\tau)U_{\varphi}U_1(t)S_zU_1^{+}(t)U_{\varphi}^{+}U_2(\tau)S_z\}/Tr\{S_z^2\}, \tag{6}$$

where $U_{1\ (2)}\ (t)$ is the usual unitary evolution operator with the Hamiltonian $H_{1\ (2)}$ taken from Eq. (4) (or Eq. (5)). In formula (6), which contains the aforementioned evolution operators, index "1" denotes the direct evolution in time, and index "2" denotes the inverse ones. We introduced the notation τ for time-reversed evolution. Under experimental conditions $\tau = t$, $\Gamma_{\varphi}(t) = \Gamma_{\varphi}(t,t)$. $U_{\varphi} = \exp\ (i\varphi S_z)$ is the operator of rotation through an angle φ around the z axis. As we mentioned above, rotation through an angle φ marks and then allows one to distinguish TCFs corresponding to coherences of various orders M, which is determined by the difference in magnetic quantum numbers [1]. The full MQ NMR spectrum can be obtained by means of the Fourier transform of the TCF (6) with respect to the variable φ.

3 Dynamics of MQ Coherences in a Solid

Let us begin the analysis of the TCF (6) with the ideal case when there is no perturbation and complete time reversibility takes place. It means that $H_1 = H_2$. The form of the Hamiltonian will not be specified yet. We assume that the evolution of the system in time can be described on the basis of orthogonal operators $|j\rangle$ and the corresponding amplitudes $A_j(t)$ [19, 23, 27–32]:

$$S_z(t) = \sum_{j=0}^{\infty} A_j(t)|j\rangle; \quad A_j(t) = \frac{\langle j|S_z(t)\rangle}{\langle j|j\rangle}. \tag{7}$$

Each subsequent basis operator is obtained from the previous one after calculating the commutator with the Hamiltonian in accordance with the recurrent equation:

$$|0\rangle = S_z, \quad |1\rangle = i\big[H,|0\rangle\big], \quad |j+1\rangle = i\big[H,|j\rangle\big] + v_{j-1}^2|j-1\rangle \quad (\text{at } j \geq 1), \quad v_j^2 = \langle j+1|j+1\rangle/\langle j|j\rangle. \tag{8}$$

In the above expressions, the angle brackets denote the calculation of the trace.

The contributions to the orthogonal operator numbered j in formula (8) can obviously contain the maximum $j+1$ spin operators and, accordingly, $j+1$ lattice summation indices [19]. Indeed, the transition from operator $|j-1\rangle$ to operator $|j\rangle$ is accompanied by a commutation with the Hamiltonian of the two-spin interaction (H_d^{yy} or H_{DQ}), which is each time accompanied by the addition of at most one spin operator (lattice index). Of course, the operator also contains contributions with a smaller number of spins (lattice indices). Therefore, if the number $j \geq K$, this vector can also contribute to the K-spin cluster. However, these contributions can be neglected due to their smallness. The point is that at small times they grow with time in proportion to t^j. At large times ($t > T_2 = 1/M_2^{1/2} \propto 1/Z^{1/2}$, where Z is the number of approximately equivalent nearest neighbors), the functions $A_j\ (t)$ decay rapidly.

Therefore, as the number increases, their amplitude decreases as $A_j(t) \sim Z^{-j/2}$. Thus, (7) can be regarded as a certain (somewhat diffuse) expansion into clusters with different numbers of spins [23].

In formula (6), we transfer the operators of inverse evolution from the first operator S_z to the second. As a result, it acquires a direct time dependence $S_z(t)$. Substituting series (7) into the transformed formula (6), we obtain

$$\Gamma_{\varphi}(t) = \mathrm{Tr}\{U_{\varphi} \sum_{j=0}^{\infty} A_j(t)|j\rangle\, U_{\varphi}^{+} \sum_{j'=0}^{\infty} A_{j'}(t)|j'\rangle\}/Tr\{S_z^2\}. \tag{9}$$

Taking into account what has been said in the previous paragraph and the fact that the rotation operator U_{φ} does not change the number of spin operators in the vector, with a large number of equivalent nearest neighbors in expression (9), we will leave only the terms with $j=j'$. Then, we get

$$\Gamma_{\varphi}(t) = \sum_{j=0}^{\infty} \Gamma_{\varphi,j}(t). \tag{10}$$

Here, we have

$$\Gamma_{\varphi,j}(t) = \mathrm{Tr}\{A_j^2(t) U_{\varphi}|j\rangle\, U_{\varphi}^{+}|j\rangle\}/Tr\{S_z^2\}. \tag{11}$$

For $\varphi=0$, we find

$$P(K,t) = \Gamma_{0,K-1}(t) = A_{K-1}^2(t)\langle K-1 \mid K-1\rangle/Tr(S_z^2). \tag{12}$$

In addition, in this case, the condition is fulfilled: $\Gamma_{\varphi=0}(t) = \sum_{K=1}^{\infty} \Gamma_{\varphi=0,K-1}(t) = \sum_{K=1}^{\infty} P(K,t) = 1$. The new designation in formula (12) was introduced to emphasize that $P(K,t)$ is actually the distribution over the number of clusters with $K=j+1$ spins.

For $\varphi \neq 0$, by representing in formula (11), the orthogonal operator $|j\rangle = \sum_M |j\rangle_M$ as a sum of elements with a certain order of coherence, we find

$$\Gamma_{\varphi,K}(t) = \Gamma_{0,K}(t) \sum_M \exp(i\varphi M) g_{KM}, \tag{13}$$

where

$$g_{KM} = \langle K-1 \mid K-1\rangle_M / \langle K-1 \mid K-1\rangle. \tag{14}$$

In Eq. (13), we took into account that the contribution to the trace from the terms whose coherence of the second operator differs from "- M" is zero.

Let us note that the g_{KM} function actually represents a form of the MQ spectrum.

A simple formula sufficiently adequately describing the shape of the MQ spectrum at least for fairly short periods of time for the first time was obtained in [3] on the base of the statistical model despite the fact that the model did not have a good reason. Therefore, according to ref. [3], we have

$$g_{KM} = \begin{cases} \frac{1}{N_g}\begin{pmatrix} 2K \\ K-M \end{pmatrix}, & M \neq 0 \\ \frac{1}{2N_g}\left[\begin{pmatrix} 2K \\ K \end{pmatrix} - 2^K\right], & M = 0 \end{cases}, \tag{15}$$

where $N_g = 4^K - \frac{1}{2}\left[\begin{pmatrix} 2K \\ K \end{pmatrix} + 2^K\right]$ is the normalization factor, $\begin{pmatrix} n \\ m \end{pmatrix}$ is binomial coefficient. For large clusters, the form of the MQ spectrum (15) is replaced by the Gaussian function

$$g_{KM} = \frac{1}{\sqrt{\pi K}} \exp\left(-\frac{M^2}{K}\right). \tag{16}$$

These relations allow calculating time dependences of the average cluster size (average number of spins in it):

$$\overline{K} = \sum_{K=1}^{\infty} K\, P(K,t). \tag{17}$$

This sort of dependences, observed experimentally in solids, represent either a power-law function of time or an exponential function of time. As we have shown in the work [19], these two types of dependence can be conveyed using two models with the following examples of amplitudes:

1. Gaussian functions [19, 28]

$$A_0(t) = \exp(-t^2/2), \quad A_j(t) = \frac{t^j}{j!}\exp(-t^2/2), \quad P(K,t) = \frac{t^{2(K-1)}}{(K-1)!}\exp(-t^2). \tag{18}$$

2. Inverse hyperbolic cosine functions [19, 31]

$$A_0(t) = \frac{1}{ch^2(t/\sqrt{2})}, \quad A_j(t) = \frac{1}{ch^2(t/\sqrt{2})}\frac{th^j(t/\sqrt{2})}{j!}, \quad P(K,t) = \frac{(th^2(t/\sqrt{2}))^{K-1}K}{ch^4(t/\sqrt{2})}. \tag{19}$$

In Eqs. (18) and (19), time is scaled in units of the second moment of the corresponding function $A_o(t)$. Summing up the series (17), we find

1. $$\overline{K} = 1 + t^2. \tag{20}$$

2. $$\overline{K} = 1 + 2\mathrm{sh}^2(t/\sqrt{2}). \tag{21}$$

4 Degradation of Correlated States due to Relaxation Processes

In the presence of a perturbation, the expressions for the orthogonal operators $|j\rangle$ and the corresponding amplitudes $A_j(t)$ will change. Assuming that the perturbation is small, we take its effect into account phenomenologically by adding the relaxation function as a damping factor to $P(K, t)$ and preserving the unperturbed values for $|j\rangle$ and $A_j(t)$. Later in the text, we will introduce the notation T for the duration of the preparatory period, keeping the notation t for the current time within the interval [0, T]. The damped factor will be defined as well as in [21, 22, 24], in the form of the product of two factors

$$\Gamma_{KM}(T) = \exp(-KB^2 t_T^2/2)\exp(-A^2 M^2 t_T^2) = F_K(T)F_M(T). \tag{22}$$

Here,

$$t_T^2 = \left\langle (T-t)^2 \right\rangle = \int_0^T (T-t)^2 R(t)\mathrm{d}t \tag{23}$$

is average over the instant of time of appearance of coherence t in the interval [0, T], characterized by the probability density R (t):

$$R(t) = (\mathrm{d}K(t)/\mathrm{d}t)/K(T),$$

where $K(t)$ is a time-dependent number of spins in the cluster. We neglect the contribution from the initial value $K(0)$. In formula (22), $A^2 = p^2 A_\Sigma^2, B^2 = p^2 B_\Sigma^2$, where A_Σ^2 and B_Σ^2 are some constants specifying the effect of the perturbation on the cluster spins. In this case, the parameter B^2 characterizes the uncorrelated contribution to the local field at each of the spins, independent of the contributions to the other spins. The parameter A^2, on the other hand, characterizes the mean field that acts in a correlated manner on all spins of the cluster. Note that the values of the constants A and B and their ratio can vary within wide limits, since it depends on the type of perturbation and the properties of the spin system. We will consider them as phenomenological parameters.

In previous works [21, 22, 24], we calculated (22) for one middle cluster. Now the calculation should be performed for each term of sum (17) according to the number of spins in the cluster at time T. In this case, the growth parameters will be expressed in terms of K and T using the following formulas:

For power growth with exponent 2 (quadratic growth):

$$K(T) = M_2^{(K)} T^2, \quad M_2^{(K)} = T^2/K. \tag{24}$$

For exponential growth:

$$K(T) = exp(a_K T), \quad a_K = (1/T)\ln K. \tag{25}$$

After performing simple calculations, we find:

For quadratic growth (24):

$$\left\langle (T-t)^2 \right\rangle = T^2/6. \tag{26}$$

For exponential growth (25):

$$\left\langle (T-t)^2 \right\rangle = 2/a_K^2 - 2T/(a_K K(T)) - T^2/K(T). \tag{27}$$

Let us estimate the change in the average cluster size due to damping. Substitute the damping function $F_K(T) = exp(-K(T)B^2 t_T^2/2)$ into series (17):

$$\overline{K}(T) = \sum_{K=1}^{\infty} K\, F_K(T) P(K,T)/N(T). \tag{28}$$

Here, we have

$$N(T) = \sum_{K=1}^{\infty} F_K(T) P(K,T), \tag{29}$$

$N(T)$ is normalizing factor equal to the Loschmidt echo amplitude.

For the case of Gaussian amplitudes (18) with a quadratic growth of the cluster size (24) and (26), we find

$$N(T) = r^2 \exp\{-T^2(1-r^2)\}, \quad \overline{K}(T) = 1 + T^2 r^2, \tag{30}$$

where $r^2 = exp(-B^2T^2/12)$.

For the case of the inverse hyperbolic cosine (19) with exponential growth of the cluster size (25) and (27), we take into account that for small B^2 and large T, the main contribution to (28) comes from large K, therefore, we take the decay function in the form

$$F_K(T) \cong \exp(-K(T)B^2/a^2). \tag{31}$$

In Eq. (31), $a^2 = a_{\overline{K}}^2 = 2$ is the parameter of unperturbed growth (21). Therefore, we get

$$N(T) = (1 - \mathrm{th}^2(T/\sqrt{2})\exp(-B^2/a^2))^{-2}\mathrm{ch}^{-4}(T/\sqrt{2}), \tag{32}$$

$$\overline{K}(T) = 1 + 2\frac{\mathrm{sh}^2(T/\sqrt{2})\exp(-B^2/a^2)}{1 + \mathrm{sh}^2(T/\sqrt{2})(1 - \exp(-B^2/a^2))}. \tag{33}$$

As it follows from the obtained expressions, in the presence of decay processes, monotonic growth $\overline{K}(T)$ slows down at large times T. With the exponential growth described by formula (33), a plateau is reached:

$$\overline{K}(T) = 1 + 2\frac{\exp(-B^2/a^2)}{1 - \exp(-B^2/a^2)},$$

whereas with the quadratic growth described by formula (30), a decrease in $\overline{K}(T)$ was observed.

5 The MQ Spectrum Shape

In the experiment, the average size of a cluster is determined from the MQ spectrum, by comparing to the Gaussian function or via finding the order of coherence M, at which the spectrum falls off "e" times. Taking into account the decay processes, the shape of the MQ spectrum will be set by the series

$$G_M(T) = \sum_{K=|M|}^{\infty} g_{KM} F_M(T) F_K(T) P(K,T)/N_1(T), \tag{34}$$

where we have

$$N_1(T) = \sum_{M} \sum_{K=|M|}^{\infty} g_{KM} F_M(T) F_K(T) P(K,T),$$

$N_1(t)$ is the new normalizing factor.

To analytically estimate the shape of the spectrum given by the relation (34), it is necessary to make some approximations. Let us assume the form-factor g_{KM} in the Gaussian form (16). We take out the factor $F_M(T)$, which is weakly dependent on K, outside the sign of the sum, and as the sum itself is the average of Gaussian functions, we replace it by the Gaussian function with the mean defined above $\overline{K}(T)$ (17). Therefore, we get

$$G_M(T) \approx \frac{1}{N_2(T)\sqrt{\pi \overline{K}(T)}} \exp\left(-\frac{M^2}{\overline{K}(T)}\right) exp\left\{-M^2 A^2 \left\langle (T-t)^2 \right\rangle\right\}, \tag{35}$$

with

$$N_2(T) \approx \frac{1}{\sqrt{\pi \overline{K}(T)}} \int_{-\infty}^{\infty} \exp\left(-\frac{M^2}{\overline{K}(T)}\right) exp\{-M^2 A^2 \left\langle (T-t)^2 \right\rangle\} dM$$
$$= \frac{1}{\sqrt{1 + \overline{K}(T) A^2 \langle (T-t)^2 \rangle}}.$$

Hence, comparing the sum of exponents in (35) with the Gaussian function with one effective cluster size, we obtain for that exponent

$$\frac{1}{K_{eff}} = \frac{1}{\overline{K}(T)} + A^2 \left\langle (T-t)^2 \right\rangle. \tag{36}$$

A more accurate summation of the Gaussian functions in (34), which we carried out in [23] without considering decay of correlations, showed that the resulting shape of the spectrum remains Gaussian at small M, and the wings are described by a simple exponential function:$G_M(T) \sim \exp\left(-\frac{|M|}{\sqrt{\overline{K}_e}}\right)$, here $\overline{K}_e$ differs from $\overline{K}$ by a numerical factor. The exponential shape of the spectrum was observed experimentally [4]. In the same paper, it was argued that relaxation is also described by an exponential function of M: $F_M(T) \sim exp\{-|M|A_e f(T)\}$. If we take $A_e = A$ and $f(T) = \sqrt{\langle (T-t)^2 \rangle}$, then the result for K_{eff} differs from (36) by a numerical coefficient, but gives the same qualitative dependence on the parameters. Therefore, for approximate estimates, let us return to formula (36). Thus, for both cases considered above, we obtain the following results:

1. With the quadratic increase in the number of spins in the cluster (see (26) and (30)), we have:

$$\frac{1}{K_{eff}} = \frac{1}{\overline{K}(T)} + \frac{A^2T^2}{6}. \tag{37}$$

2. With the exponential increase in the number of spins in the cluster (31), taking the average at large T in (27) as $\left\langle (T-t)^2 \right\rangle \cong 2/a^2$,

$$\frac{1}{K_{\text{eff}}} = \frac{1}{\overline{K}(T)} + \frac{2A^2}{a^2}. \tag{38}$$

6 Discussion

We begin our discussion with the case of the exponential increase in the number of spins in a cluster (38). If the preparation time T is not too long, so that

$$exp(aT) < 4a^2/B^2 \text{ and } exp(aT) < a^2/A^2, \tag{39}$$

then the effective size of a cluster is equal to its size in the absence of perturbations:

$$K_{\text{eff}} \cong \frac{1}{2} exp(aT) = \frac{1}{2} exp\left(\sqrt{2}T\right). \tag{40}$$

At large T, if conditions (39) are violated, the growth slows down. Let $A = 0$, then for $T \to \infty$

$$K_{\text{eff}} = \frac{2a^2}{B^2} = \frac{4}{B^2}. \tag{41}$$

Let $B = 0$, then for $T \to \infty$

$$K_{\text{eff}} = \frac{a^2}{2A^2} = \frac{1}{A^2}. \tag{42}$$

In both cases, the experiment will demonstrate the cessation of cluster growth. However, the physical reasons are different. In the case described by Eq. (41), due to the faster decay of the contribution from large clusters in series (28) and (34), the average cluster size is determined by smaller clusters, the size of which depends on the value of the damage parameter B.

On the contrary, in case (42), there is no faster damage of large clusters, but there is a faster decay of clusters with a large M. Because of this, the shape of the MQ spectrum stabilizes, and it leads to stabilization of the apparent size of the cluster extracted from it [21, 22]. In this case, the growth of the average cluster size with increasing T can continue. Which of the cases, and in what proportion, is realized in the spin system depends on the ratio of parameters A and B.

For the experimental estimation of these parameters, one can use the results of [13], where the relaxation under the action of dipole–dipole interaction was measured depending on the time interval t_d between the preparatory period and mixing period. As we have shown earlier [24], formula (22) at $t_d = t_T$ well describes results of the experiment [13]. Therefore, as follows from [13], the parameter A^2 exceeds B^2 by more than two times.

Note that the assumption about the possible stabilization of the average cluster size due to relaxation processes was proposed in [4, 5]. In [4], the average value of the stable cluster size (K_0) was estimated from the relaxation of the components of the MQ spectrum, which depends on M; i.e., through the stabilization of the MQ spectrum. In light of the above, the conclusion that this value really characterizes the real size of the cluster needs additional justification.

Let us now discuss the situation when the number of spins in a cluster grows in accordance with the quadratic law (37). If the preparatory period T is not too long, the effective cluster size is equal to its size in the absence of perturbations:

$$K_{\text{eff}} \cong T^2. \tag{43}$$

At large T, the growth slows down, passes through a maximum and falls off. Let $A = 0$, then for $T \to \infty$

$$K_{\text{eff}} = T^2 \exp\left(-\frac{B^2T^2}{12}\right). \tag{44}$$

If $B = 0$, then for $T \to \infty$, one gets

$$K_{\text{eff}} = \frac{6}{A^2T^2}. \tag{45}$$

The difference between the dependencies for $T \to \infty$ in the considering situation and in the previous case is due to weaker, power-law growth of the cluster size, which does not compensate for the action of exponential relaxation processes. As in the previous case, the result (44) shows a decrease in the average size of real clusters, while the result reflected by Eq. (45) is associated with

narrowing of the MQ spectrum due to the *M*-dependent relaxation of the spectral components.

In conclusion, the studies performed in the present work for model systems have shown the sensitivity of the MQ spectrum width on the ratio of the rates of relaxation processes caused by two different mechanisms. The first mechanism is the consequence of the fact that the components of the local field act independently on each spin in the cluster exists. The second mechanism is the result of the presence of a component of the field that acts in a correlated manner on all the spins of the cluster. Therefore, to extract reliable information from MQ spectra, it is necessary, on the one hand, to control the proportion of these processes, and on the other hand, to further develop the theory of the shapes of MQ spectra. The latter is all the more important because for conventional three-dimensional systems, computer modeling is very difficult for obvious reasons while it largely advanced [33] for one-dimensional systems.

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