ISSN 0021-3640, JETP Letters, 2017, Vol. 105, No. 8, pp. 514–518. © Pleiades Publishing, Inc., 2017.
Original Russian Text © V.E. Zobov, A.A. Lundin, 2017, published in Pis'ma v Zhurnal Eksperimental'noi i Teoreticheskoi Fiziki, 2017, Vol. 105, No. 8, pp. 499–503.

CONDENSED MATTER

On the Effect of an Inhomogeneous Magnetic Field and Many-Body Localization on the Increase in the Second Moment of Multiple-Quantum NMR with Time

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Received March 16, 2017; in final form, March 24, 2017

A change in the time dependence of the second moment of the distribution of intensities of coherences with various orders in the spectrum of multiple-quantum NMR in a solid at the inclusion of an inhomogeneous magnetic field in the effective interaction is studied. Both the secular dipole–dipole and nonspecular two-quantum interactions are considered as nucleus–nucleus interactions, which correspond to traditional experimental realizations. It is shown that, with an increase in the magnitude of the inhomogeneous field, an exponential increase in the second moment of multiple-quantum NMR with time changes to a power-law increase. The results obtained in this work indicate that this second moment, which determines the average number of dynamically correlated spins, can be used as a convenient characteristic for studying a transition to a many-body localized state.

DOI: 10.1134/S0021364017080148

A system of interacting nuclear magnetic moments in a solid is a convenient adequate model object for studying various problems of statistical physics [1]. Multiple-quantum NMR spectroscopy [2, 3] is an efficient method for studying both local properties and dynamics in these systems. For example, multiple-quantum spectroscopy was used to observe atomic clusters in solids [4], one-dimensional chains [5], an increase in the number (up to $\approx 10^4$) of dynamically correlated spins with the time [6], etc. Finally, it has been recently proposed [6–9] to apply the multiple-quantum NMR method to study the properties of a transition to a many-body localized state in inhomogeneous spin systems (i.e., in the presence of an inhomogeneous magnetic field) [10–17]. This state of matter attracts attention because of its extraordinary properties. On one hand, as in an Anderson insulator without interaction [18], diffusion is absent and equilibrium is not established in this state. On the other hand, the interaction between spins in such a state results in the dephasing of spins and, thereby, in the appearance of dynamic correlations between them at large distances. The average size of clusters of dynamically correlated spins in multiple-quantum NMR spectroscopy is characterized by the second moment of the distribution of intensities of coherences with various orders in the spectrum [19–21].

In this work, we study for the first time the effect of an inhomogeneous magnetic field on an increase in the second moment of multiple-quantum NMR with the time and demonstrate that its exponential increase changes to a power-law increase, which is apparently a manifestation of many-body localization. The proposed approach is based on analytical estimates of time-dependent spin correlation functions in the approximation of a large number of neighbors, which is accepted for NMR in solids. In contrast to our approach, most of the studies of multiparticle localization are based on the analysis of one-dimensional systems and on the numerical calculation of the dynamics of small spin clusters.

The multiple-quantum NMR method involves the measurement of the time correlation function [2, 3]

$$\Gamma_\varphi(t,\tau) = \mathrm{Tr}\{U^+(\tau)U_\varphi U(t)S_\alpha U^+(t)U_\varphi^+(\tau)S_\alpha\}/\mathrm{Tr}\{S_x^2\}, \quad (1)$$

where $U(t)$ is the operator of evolution with the Hamiltonian of the internal interaction H_d or this interaction transformed by radiofrequency pulses to a new effective Hamiltonian H_{eff}, $U_\varphi = \exp(i\varphi S_\beta)$ is the

operator of rotation by an angle of φ about the β axis, $S_\alpha = \Sigma_i S_{\alpha i}$ is the α-component of the vector operator of total spin of the nuclear system ($\alpha = x, y, z$), $S_{\alpha i}$ is the α-component of the spin operator ($S = 1/2$) at the site i, and τ stands for evolution with "reversed time." Experimental conditions $t = \tau$ will be accepted in the final formulas.

The secular part of the nuclear–nuclear dipole–dipole interaction in a strong external static magnetic field responsible for the broadening of NMR lines and, correspondingly, for the dynamics of the spin system has the form [1]

$$\begin{aligned} H_d &= \sum_{i\neq j} b_{ij} S_{zi} S_{zj} - \frac{1}{2}\sum_{i\neq j} b_{ij} S_{+i} S_{-j} \\ &= \sum_{i\neq j} b_{ij} S_{zi} S_{zj} - \frac{1}{2}\sum_{j\neq j} b_{ij} (S_{xi} S_{xj} + S_{yi} S_{yj}) \\ &= \sum_{i\neq j} \{H_{dij}^{zz} + H_{dij}^{xx} + H_{dij}^{yy}\}, \end{aligned} \tag{2}$$

where $b_{ij} = \gamma^2\hbar(1 - 3\cos^2\theta_{ij})/2r_{ij}^3$, $\mathbf{r}_{ij}$ is the vector connecting the spins i and j, θ_{ij} is the angle between the vector $\mathbf{r}_{ij}$ and the external static magnetic field, and $S_{\pm j} = S_{xj} \pm iS_{yj}$. Here and below, the energy is given in frequency units.

In the traditional multiple-quantum NMR scheme [2, 3], the irradiation of the sample by a multipulse sequence usually transforms the Hamiltonian (2) to the so-called two-spin/two-quantum Hamiltonian

$$\begin{aligned} H_{DQ} &= -\frac{1}{2}\sum_{i\neq j} b_{ij} (S_{xi} S_{xj} - S_{yi} S_{yj}) \\ &= -\frac{1}{4}\sum_{i\neq j} b_{ij} (S_{+i} S_{+j} + S_{-i} S_{-j}) \end{aligned} \tag{3}$$

and time correlation functions (1) determined by the directions $\alpha = \beta = z$ are chosen. Other variants are also possible. In particular, directions $\alpha = z$ and $\beta = x$ were chosen in [22] and directions $\alpha = x$ and $\beta = x$ were chosen in [23]. The last choice made it possible to measure an increase in correlations in the process of development of the free induction decay, when the evolution of $U(t)$ is determined by the dipole–dipole interaction (2).

The authors of [9] proposed and used a multipulse sequence with 16-pulse cycle, which provided the Hamiltonian

$$H_{\text{eff}} = uH_{DQ} + vH_d + g\sum_i h_i S_{zi} \tag{4}$$

with arbitrarily related parameters u, v, and g and, naturally, with the inversion of their signs at the stage of evolution with reversed time.

The multiple-quantum NMR spectrum is obtained after the Fourier transform of the time correlation function $\Gamma_\varphi(t, \tau)$ (1) with respect to the variable φ. The second moment of the multiple-quantum spectrum [19–21] is given by the formula

$$\begin{aligned} \langle n^2(t)\rangle &= -d^2\Gamma_\varphi(t,t)/d\varphi^2 \,|_{\varphi=0} \\ &= -\mathrm{Tr}\{[S_\beta, S_\alpha(t)]^2\}/\mathrm{Tr}\{S_x^2\}, \end{aligned} \tag{5}$$

where $S_\alpha(t) = U(t)S_\alpha U^+(t)$. In [20], we transformed Eq. (5) at $\beta = x$ to the formula more convenient for calculations:

$$\begin{aligned} &\langle n^2(t,\tau)\rangle \\ &= 2\sum_{i,j,f,q} \mathrm{Tr}\left\{S_{xj} S_{xf} \left\{S_{\alpha i}(t) - \overline{S_{\alpha i}(t)}^{(f)}\right\} S_{\alpha q}(\tau)\right\}/\mathrm{Tr}\{S_x^2\}. \end{aligned} \tag{6}$$

The symbol $\overline{S_{\alpha j}(t)}^{(f)}$ means that the spin f is reversed in the Hamiltonian appearing in the time evolution operator; i.e., this Hamiltonian now has the form $4S_{xf}HS_{xf}$, where H_{dfj}^{zz} and H_{dfj}^{yy} are replaced by $-H_{dfj}^{zz}$ and $-H_{dfj}^{yy}$, respectively.

It is remarkable that the terms with $i = f$ in Eq. (6) change sign at $\alpha = z$. The sum of these terms ensures the initial condition $\langle n^2(0)\rangle = 1$ in Eq. (5) at $\alpha = z$ and $\beta = x$. We will neglect this contribution when considering large clusters with $\langle n^2(t)\rangle \gg 1$. We calculated the quantity $\langle n^2(t)\rangle$ in [20] for homogeneous spin systems with a large number of neighbors surrounding each nucleus in the lattice. The nucleus–nucleus interaction was described in [20] by Hamiltonian (4) with the parameters $u = 1$ and $v = g = 0$, and the transition to the parameters $u = 1 - p$, $v = p \ll 1$, and $g = 0$ was examined in [24].

To analyze the effect of inhomogeneous magnetic fields h_i in Eq. (4) on the second moment of the multiple-quantum spectrum, we consider the case $g \neq 0$ with the normal distribution law with the variance W^2 for inhomogeneous magnetic fields:

$$P(gh_i) = \frac{1}{\sqrt{2\pi W^2}}\exp\left(-\frac{g^2h_i^2}{2W^2}\right). \tag{7}$$

First, we take the Hamiltonian (4) with the parameters $u = 0$ and $v = 1$ and choose the axes $\alpha = x$ and $\beta = x$. In this case, to estimate the time dependence $\langle n^2(t)\rangle$ in [20], we summed chains of flip-flop spin pairs $b_{kj}S_{+k}S_{-j}$, which are coupled by the zz interaction, connecting the initial spin i and the "reversed" spin f in Eq. (6).

The autocorrelation function of the transverse components of the spins at a large number of neighbors was approximated by a Gaussian with the second

moment $B^2 = \sum_j b_{ij}^2$ caused by the zz interactions. Inhomogeneous fields lead to an additional relaxation of the autocorrelation function of transverse spin components, which is manifested in an increase in the second moment of the Gaussian describing them

$$\Gamma_d(t) = \exp\{-(B^2 + W^2)t^2/2\}. \quad (8)$$

After such a change, by formulas derived in [20], the desired estimate is obtained in the form

$$\langle n^2(t)\rangle = (2/3)(B/x)^{2/3}\{e^{xt} - 2e^{-xt/2}\cos(\sqrt{3}xt/2 - \pi/3)\} + (1/6)\{e^{xt} + 2e^{-xt/2}\cos(\sqrt{3}xt/2) - 3\}, \quad (9)$$

where $x = \left(\frac{2\sqrt{\pi}B^4}{\sqrt{B^2 + W^2}}\right)^{1/3}$. At $xt \gg 1$, Eq. (9) gives an exponential increase with the time:

$$\langle n^2(t)\rangle \sim \exp(xt). \quad (10)$$

If $xt \ll 1$, which is the case at a large inhomogeneity $B^2/W^2 \ll 1$, the second term in Eq. (9) vanishes and the first term gives a power law

$$\langle n^2(t)\rangle \cong B^2t^2. \quad (11)$$

According to [20], such a result implies that only the zz interactions are retained in Eq. (2) and the flip–flop contribution to the Hamiltonian is neglected because it is suppressed by inhomogeneous fields. Expression (11) is the first term of the expansion in a power series of time for the result obtained in [20]:

$$\langle n^2(t)\rangle = \sum_f \sin^2 b_{if}t. \quad (12)$$

At large times, when the interaction between nearby spins satisfies the condition $|b_{ij}|t \gg 1$, the dependence is transformed to

$$\langle n^2(t)\rangle \cong 4\pi^2\gamma^2\hbar t/(9\Omega\sqrt{3}), \quad (13)$$

where Ω is the volume per site of the lattice. Here, we used the results of calculation of the sum in Eq. (12) after its change to the integral over the space obtained in [25–27] at the calculations of the form of the spectrum of magnetically diluted of spin systems. Dependence (13) is presented for three-dimensional lattices. For systems of a lower dimension d, using estimates from [27], we obtain the dependence $\langle n^2(t)\rangle \sim t^{d/3}$.

A similar problem was solved in [9–13, 16] when estimating an increase in quantum correlation, i.e., entanglement between the spin i and its environment in the state of many-body localization. Following the cited works, at large times for each of the nearby spins for which $|b_{ij}|t > \pi/2$, we take an oscillation-averaged contribution of 1/2 to the sum (12). Further, if the zz interaction in Eq. (2) is taken with the constant $b_{ij} = J_0\exp(-|\mathbf{r}_{ij}|/\xi)$, the sum will be determined by the number of spins inside a sphere with the radius $|\mathbf{r}_{ij}| = \xi\ln(2t/\pi J_0)$. The contribution from other spins is small because of the fast weakening of the interaction with distance. The resulting estimate is $\langle n^2(t)\rangle \sim \ln^d(2t/\pi J_0)$. Finally, we note that a similar estimate for the dipole–dipole interaction with a power-law dependence on the distance provides a correct time dependence $\langle n^2(t)\rangle \sim t^{d/3}$, but with an underestimated coefficient as compared to the results in [27] and Eq. (13).

We now consider the Hamiltonian (4) with the parameters $u = 1$ and $v = 0$ at the directions of the axes $\alpha = z$ and $\beta = x$ in Eq. (6). In this case, the chain between the initial spin and reversed spin f in Eq. (6) is constructed by alternating the xx and yy interactions. Following [20], to estimate an increase in $\langle n^2(t,\tau)\rangle$, we estimate the contributions from two segments of evolution in Eq. (1) independently, i.e., under the assumption that

$$\langle n^2(t)\rangle \sim G(t)G(\tau), \quad (14)$$

where $G(t)$ (and $G(\tau)$) is defined in the form of a series of convolutions of the autocorrelation functions, which are approximated by the Gaussian

$$\Gamma_e(t) = \exp\{-M_{2e}t^2/2\}, \quad (15)$$

where the average value of the second moment $M_{2c} = 5B^2/16 + 3W^2/4$ is obtained according to the rules from [20]. The Laplace image of the series $G(t)$ has the form

$$L_G(p) = \frac{\omega_c w(p)}{1 - \omega_c w(p)}, \quad (16)$$

where $\omega_c = B/2^{1/2}$ is the average interaction per vertex and $w(p)$ is the Laplace image of the Gaussian (15). The behavior of the function $G(t)$ at large times is determined by the minimum zero of the denominator in Eq. (16):

$$G(t) \approx \exp(p_{\min}t). \quad (17)$$

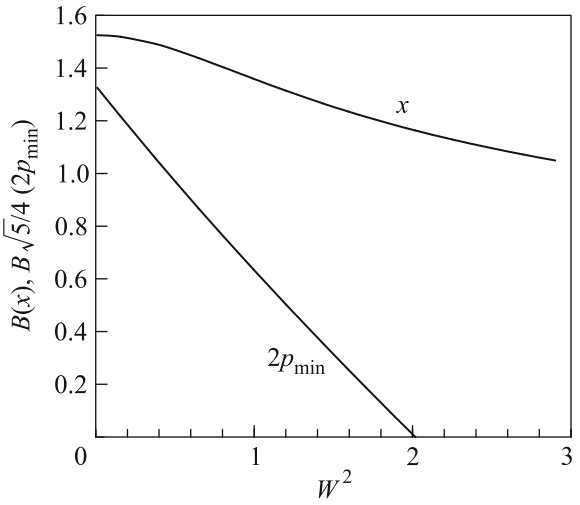
To calculate this zero, we use the equation

$$\delta\exp(s^2)\mathrm{erfc}(s) = 1, \quad (18)$$

where erfc(s) is the is the complementary error function and

$$\delta = B\sqrt{\pi/4M_{2e}}, s = p/\sqrt{2M_{2e}}. \quad (19)$$

At $W = 0$, we found $s_{\min} = p_{\min}/\sqrt{2M_{2e}(W = 0)} = 0.47$, where $M_{2e}(W = 0) = 5B^2/16$. The parameters $s_{\min}$ and $p_{\min}$ will decrease with an increase in the rms inhomogeneity of the field W, as is shown in the fig-



Exponents x and $2p_{\min}$ of the exponential growth of $\langle n^2(t)\rangle$ with time in frequency units of B and $B\sqrt{5}/4$ versus the variance W^2 of the inhomogeneous magnetic field.

ure, and vanish at $\delta = 1$, which corresponds to the value

$$W^2/M_{2e}(W = 0) = \pi 16/15 - 4/3 = 2.018. \quad (20)$$

Thus, an increase in the inhomogeneous field will result in a decrease in the exponent $2p_{\min}$ of the exponential increase in the second moment of the multiple-quantum spectrum (14), which determines the average size of a cluster of dynamically correlated spins. The exponential growth will cease at the critical value (20). With a further increase in W, an increase changes to a decrease. This means that the included contribution from chains to $\langle n^2(t)\rangle$ is no longer leading and another approximation should be used.

Indeed, in the preceding example with the dipole–dipole interaction in the presence of a large inhomogeneous field, a transition occurs to a power-law increase specified by Eq. (11) or (13), which is caused by the zz interaction. A particular role of such interactions in the state of many-body localization was specially emphasized in [10–17]. In the general case, the authors of the cited works proposed to change to new variables, effective spins τ_{zi}, which diagonalize the Hamiltonian. However, the cost of this transition is the appearance of multispin interactions. The zz interaction is absent in the initial two-spin/two-quantum Hamiltonian (3), but it appears after the change to new variables. Such a transition can be performed by, e.g., approximate canonical transformations [14, 16]. As a result, one can obtain the effective three-spin interaction

$$H_{\text{eff}} = \sum_{i,j,k} J_{ijk}\tau_{zi}\tau_{zj}\tau_{zk},$$

where the constants are small, $J_{ijk} \sim B^4/W^3$. This interaction leads to a power-law increase in $\langle n^2(t)\rangle$ in the form of Eq. (11) with a small coefficient of about B^{10}/W^8. It is noteworthy that this coefficient at another choice of the axes $\alpha = x$ and $\beta = x$ increases by a factor of W^2/B^2. The reason is that the time dependence in Eq. (6) at $\alpha = z$ is determined by the correction term after the canonical transformation S_z.

To summarize, according to the reported calculations, the analysis of the second moment of the multiple-quantum NMR spectrum allows studying the effect of the inhomogeneous magnetic field on the growth rate and size of the average cluster of dynamically correlated spins. Thus, the characteristics of the transition to the many-body localized state can be studied. The average correlation length was previously proposed in [9] as a measure of localization, but to theoretically calculate it, it is necessary to calculate the entire multiple-quantum NMR spectrum, which is hardly possible. The fundamental advantage of the second moment of multiple-quantum NMR is that, to determine it, it is sufficient to calculate the time correlation function given by Eq. (5) or (6).

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Translated by R. Tyapaev