Many-Spin Entanglement in Multiple Quantum NMR with a Dipolar Ordered Initial State

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Abstract—Many-spin entanglement is investigated in a gas of spin-carrying molecules (atoms) in a nanopore under NMR conditions with a dipolar ordered initial state. To estimate the number of entangled spins, the second moment of the distribution of the intensities of multiple quantum (MQ) NMR coherences is used, which provides a lower bound for the quantum Fisher information. Many-spin entanglement is investigated at different temperatures and different numbers of spins.

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1. INTRODUCTION

Entanglement [1] is an important concept in quantum mechanics. In particular, it is responsible for the superiority of quantum computers over their classical counterparts. The quantum superiority [2] recently demonstrated on a programmable superconducting processor is also related to the concept of entanglement, which is absent in classical physics. Among the numerous methods for studying entanglement, we focused on multiple quantum (MQ) NMR in solids [3], which is widely used to study entanglement in binary systems [4–7].

It also turned out that MQ NMR spectroscopy [3] makes it possible to extract information on many-spin entanglement [8] by using the quantum Fisher information [9, 10], which is a key concept in quantum information theory. The quantum Fisher information describes the rate of change of quantum states determined by the density matrix under variation of the observable responsible for the MQ coherence in MQ NMR spectroscopy [3]. In MQ NMR spectroscopy, this observable is determined by the operator of the total projection of the spin angular momentum onto the direction of a strong external magnetic field. Using the quantum Fisher information [11], one can obtain important information about many-spin entanglement for the analysis of the MQ NMR spectrum, since there is a relationship between the second moment of the spectrum [12] and the quantum Fisher information [8, 13]. In addition, the second moment of the MQ NMR spectrum determines the lower bound of the quantum Fisher information [8]. This means that MQ NMR spectroscopy is an effective method for solving quantum information problems.

Many-spin entanglement was studied in [13] for a nonspherical nanopore filled with a gas of spin-carrying molecules in a strong external magnetic field [14, 15]. The thermodynamic equilibrium initial state of the system was determined by the one-spin Zeeman interaction with the external magnetic field [16]. However, many-spin entanglement can be investigated when the same system is initially prepared in a dipolar ordered state [17] using either the method of adiabatic demagnetization in a rotating reference frame (RRF) [17, 18] or a two-pulse Broekaert–Jeener sequence [17, 19]. The MQ NMR dynamics with such an initial state was simulated both for small spin systems [16, 20] and for a system consisting of 200–600 spin-carrying molecules (atoms) filling a nanopore [21]. However, the approaches developed for these studies are restricted to the high temperature case and cannot be applied to studying many-spin entanglement.

In the present work, we consider the intermediate case of low Zeeman and high dipolar temperatures. Magnetic ordering [22] is beyond the scope of this article. Notice that the two-pulse Broekaert–Jeener experiment [19] was developed for the high-temperature case. We show theoretically that the experiment of [19] can also be carried out in the intermediate temperature case, which we consider here. It was shown [21] that, in the MQ NMR experiment with a dipolar ordered initial state, MQ NMR coherences appear faster than in the MQ NMR experiment with an initial thermodynamic equilibrium state in a strong external magnetic field. This fact is important for the study of many-spin entanglement, since it involves the calculation of the second moment of the distribution of MQ
NMR coherences. This fact is also important for studying the propagation of many-spin correlations [14, 23–25] and their localization [26, 27]. In this case, an important role is played by out-of-time ordered correlations, which are associated with the distribution of MQ NMR coherences.

In this article, we investigate many-spin entanglement with the use of the MQ NMR spectrum of spin-carrying atoms (molecules) in a nanopore when the system is prepared in a dipolar ordered state. In Section 2, we describe the theory of MQ NMR dynamics at low Zeeman temperature and high dipolar temperature. An analytical solution for the MQ NMR dynamics of a three-spin system, obtained at the same temperatures, is described in Section 3. The second moment of the MQ NMR spectrum is considered in Section 4 as a measure of many-spin entanglement. In Section 5, we investigate the dependence of many-spin correlations on the dipolar temperature and the number of spins in the system. A brief summary of the results obtained is given in Section 6. The Appendix shows that the two-pulse Broekaert–Jeener sequence results obtained is given in Section 6. The Appendix shows that the two-pulse Broekaert–Jeener sequence can be used in the case when the Zeeman temperature is low and the dipolar temperature is high.

2. THEORY OF MQ NMR DYNAMICS IN A NANOPORE AT LOW ZEEMAN AND HIGH DIPOLAR TEMPERATURES

The MQ NMR dynamics in a nanopore is determined by the Hamiltonian [13, 15]

\[ H_{MQ} = -\frac{D}{4}(I^+)^2 + (I^-)^2, \]

where

\[ I^± = \sum_{j=1}^{N} I_j^±, \]

(1)

(2)

\(N\) is the number of spins in a nanopore, \(I_j^±\) are raising and lowering operators of spin \(j\), and \(D\) is the dipole–dipole interaction (DDI) constant averaged over the fast molecular diffusion of spin-carrying atoms (molecules) in the nanopore. We emphasize that the DDI constant \(D\) is the same for all pairs of interacting spins in the nanopore [13, 15]. The density matrix \(\rho(t)\) in the preparation period of the MQ NMR experiment [3] can be obtained from the Liouville evolution equation [17, 22]

\[ i\hbar \frac{d\rho(t)}{dt} = [H_{MQ}, \rho(t)] \]

(3)

with the initial thermodynamic equilibrium density matrix

\[ \rho(0) = \rho_{eq} = \frac{1}{Z} \exp \left( \frac{\hbar \omega_0}{k_B} \alpha_z I_z + \frac{\hbar}{k_B} \beta_d H_{dc} \right), \]

(4)

where

\[ Z = \text{Tr} \left\{ \exp \left( \frac{\hbar \omega_0}{k_B} \alpha_z I_z + \frac{\hbar}{k_B} \beta_d H_{dc} \right) \right\} \]

is the partition function, \(k_B\) is the Boltzmann constant, \(\omega_0\) is the Larmor frequency, \(I_z\) is the operator of the projection of the total spin angular momentum onto the \(z\) axis, which is directed along a strong external magnetic field, \(H_{dc}\) is the secular part of the DDI Hamiltonian in a strong external magnetic field, and \(\alpha_z\) and \(\beta_d\) are the inverse Zeeman and dipole temperatures. We consider the case when the Zeeman temperature is low (\(\hbar \omega_0 \alpha_z / k_B \gg 1\)) and the dipolar temperature is high (\(\hbar \beta_d / k_B \ll 1\)). We assume that \(\omega_0 = 2\pi \cdot 500 \times 10^6 \text{s}^{-1}\) and \(D = 2\pi \times 10^4 \text{s}^{-1}\). In the Appendix, we prove that the two-pulse Broekaert–Jeener sequence [17, 19] results in a dipolar ordered state even at a low Zeeman temperature. Another method for creating a dipolar ordered state of the system is adiabatic demagnetization [17, 18]. Using these methods, we can obtain a system in the thermodynamic equilibrium state with the density matrix

\[ \rho_i = \frac{1}{Z_i} \exp \left( \frac{\hbar \beta_d H_{dc}}{k_B} \right) = \frac{1}{Z_i} \left( 1 + \frac{\hbar \beta_d}{k_B} H_{dc} \right), \]

(5)

where the partition function is

\[ Z_i = \text{Tr} \left\{ \exp \left( \frac{\hbar \beta_d H_{dc}}{k_B} \right) \right\} \approx 2^N. \]

(6)

We will investigate the MQ NMR dynamics in a nanopore on the basis of Eq. (3) with the initial state (5). It is also important that the Hamiltonian \(H_{dc}\) is partially averaged by fast molecular diffusion in the nanopore, and the averaged Hamiltonian can be expressed as [21, 28]

\[ H_{dc} = \frac{D}{2} (3I_z^2 - I^2), \]

(7)

where \(I\) is the square of the spin angular momentum.

Suppose that a function \(G(t, \phi)\) describes a signal, averaged over the equilibrium density matrix, after the periods of preparation, evolution, and mixing in the MQ NMR experiment [3]. It can be expressed as [13]

\[ G(t, \phi) = \text{Tr} \{ \exp(iH_{MQ}^0 t) \exp(i\phi I_z) \} \times \exp(-iH_{MQ}^0 t) \rho_i \exp(iH_{MQ}^0 t) \times \exp(-i\phi I_z) \rho_i \exp(i\phi I_z) \}

(8)

= \text{Tr} \{ \exp(i\phi I_z) \rho(t) \exp(-i\phi I_z) \} \rho(t), \]

where

\[ \rho(t) = \exp(-iH_{MQ}^0 t) \rho_i \exp(iH_{MQ}^0 t) \]

(9)
is a solution of Eq. (3) with the initial condition (5). The density matrix $\rho(\tau)$ is expanded in a series as

$$\rho(\tau) = \sum_n \rho_n(\tau),$$  \hspace{1cm} (10)

where $\rho_n(\tau)$ is the contribution to $\rho(\tau)$ from the $n$th-order MQ-coherence [29]. Then the function $G(\tau, \phi)$ in Eq. (8) can be rewritten as

$$G(\tau, \phi) = \sum_n e^{i\phi n} \text{Tr}[\rho_n(\tau)\rho_{-n}(\tau)],$$  \hspace{1cm} (11)

where we take into account that

$$[I_z, \rho_n(\tau)] = n\rho_n(\tau).$$  \hspace{1cm} (12)

For further calculations, it is necessary to introduce the normalized intensities $J_n(\tau) (n = 0, \pm 2, \pm 4, \ldots)$ of MQ NMR coherences:

$$J_n(\tau) = \frac{\text{Tr}[\rho_n(\tau)\rho_{-n}(\tau)]}{\text{Tr}[\rho^2_1(\tau)]}. $$  \hspace{1cm} (13)

Using Eqs. (9) and (10), we can check that

$$\sum_n J_n(\tau) = \frac{\sum_n \text{Tr}[\rho_n(\tau)\rho_n(\tau)]}{\text{Tr}[\rho^2_1(\tau)]} = \frac{\text{Tr}[\rho^2(\tau)]}{\text{Tr}[\rho^2_1(\tau)]} = 1. $$  \hspace{1cm} (14)

From Eq. (14) we can conclude that the sum of the MQ NMR coherences is preserved during the preparation period of the MQ NMR experiment [3].

The basis consisting of the eigenstates of the operator $I_z$ (called a multiplicative basis) is widely used for numerical calculations of MQ NMR dynamics [30]. Due to the rapid expansion of the Hilbert space with an increase in the number of spins, such calculations are possible only for systems with a small number of spins. This approach is not suitable for investigating many-spin entanglement. Since the Hamiltonian $H_{MQ}$ of Eq. (1) consists of two blocks for two possible values of the spin angular momentum ($F = S(S + 1)$, $S = 3/2, 1/2$). These blocks and the corresponding eigenvalues and eigenstates are given in [13]. The density matrix of the system also consists of two blocks, $\rho^{3/2}(\tau)$ and $\rho^{1/2}(\tau)$ and

$$\rho^{3/2}(0) = \frac{1}{Z} \begin{pmatrix} e^{3b/2} & 0 & 0 & 0 \\ 0 & e^{-3b/2} & 0 & 0 \\ 0 & 0 & e^{-3b/2} & 0 \\ 0 & 0 & 0 & e^{3b/2} \end{pmatrix},$$  \hspace{1cm} (15)

$$\rho^{1/2}(0) = \frac{1}{Z} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}. $$  \hspace{1cm} (15)

where $b = \hbar D/k_B T$ and $T$ is temperature. By simple calculations, we can obtain the density matrices $\rho^{3/2}(\tau)$ and $\rho^{1/2}(\tau)$, which allow us to find the intensities of the MQ NMR coherences.

In the systems under consideration, only the zeroth-order and plus/minus second-order MQ NMR coherences appear. The intensities of these coherences are

$$J_0(\tau) = 1 - \frac{1}{2} \sin^2(\sqrt{3}D\tau),$$  \hspace{1cm} (16)

$$J_{\pm2}(\tau) = \frac{1}{4} \sin^2\left(\frac{3b}{2}\sqrt{3}D\tau\right).$$  \hspace{1cm} (16)

According to expressions (16), the sum of the intensities of the MQ coherences is equal to unity according to Eq. (14). The dependences of the calculated intensities $J_n(\tau) (n = 0, \pm 2)$ are shown in Fig. 1 as a function of the evolution time.
4. THE SECOND MOMENT OF THE MQ NMR SPECTRUM AS A MEASURE OF MANY-SPIN ENTANGLEMENT

Expression (8) for the MQ NMR signal \( G(\tau, \phi) \) can be expanded in a series in terms of the phase increment of the pulses:

\[
G(\tau, \phi) = \text{Tr}[\rho(\tau) \exp(i\phi I_z) \rho(\tau) \exp(-i\phi I_z)] = \text{Tr}[\rho^2(\tau)] - \phi^2 \text{Tr}[\rho^2(\tau) I_z^2 - (\rho(\tau) I_z)^2] + O(\phi^3).
\]  

One can prove \([31]\) that the quantum Fisher information \( F_Q(\rho, I_z) \) \([32]\) satisfies the inequality

\[
F_Q(\rho, I_z) \geq 4\text{Tr}[\rho^2 I_z^2 - (\rho I_z)^2].
\]  

At the same time, it is easy to check that the expression \(2\text{Tr}[\rho^2(\tau) I_z^2 - (\rho(\tau) I_z)^2] \) is equal to the second moment \( M_2 \) of the intensity distribution of MQ NMR coherences \([12]\),

\[
M_2 = \sum_n n^2 J_n(\tau),
\]  

where \( J_n(\tau) (n = 0, \pm 2, \pm 4, \ldots) \) is defined by Eq. (13). Thus, the second moment of the distribution of MQ NMR intensities provides a lower bound for the quantum Fisher information \( F_Q(\rho, I_z) \). It was also shown in \([9, 10]\) that if

\[
F_Q(\rho, I_z) > nk^2 + (N - nk)^2,
\]  

where \( n \) is the integer part of \( N/k \), then the system with the density matrix \( \rho(\tau) \) contains \( k + 1 \) entangled spins \([33–35]\). The results of the numerical analysis of many-spin entanglement in a system of spin-carrying molecules (atoms) initially prepared in a dipolar ordered state are presented in the next section.

5. NUMERICAL ANALYSIS OF MANY-SPIN ENTANGLEMENT AT DIFFERENT TEMPERATURES AND DIFFERENT NUMBER OF SPINS IN THE SYSTEM

The model of spin-carrying molecules (atoms) in a nanopore in a dipolar ordered state expands the possibilities of studying many-spin entanglement compared with the related model of \([13]\), in which the system was initially in thermodynamic equilibrium in a strong external magnetic field. The model of \([13]\) is inapplicable for studying the time evolution of the system, because the distribution of MQ NMR coherences quickly becomes stationary \([15]\). In this model, many-spin entanglement changes with temperature in a very narrow temperature range. For example, all spins are entangled in a system consisting of 201 spins already at a temperature of \( T = 6.856 \times 10^{-3} \text{K} \) \([13]\).

The time dependence of the quantum Fisher information in a system consisting of 101 spins is shown in Fig. 2 for different temperatures. Figure 2a shows that, at a temperature of \( T = 6 \times 10^{-4} \text{K} \), only pairwise entanglement exists. At a temperature of \( T = 3.2 \times 10^{-4} \text{K} \), a strip appears in Fig. 2b in which inequality (20) can be satisfied for \( 19 \leq k \leq 46 \). Thus, there is many-spin entanglement in spin clusters consisting of 20–47 spins at a temperature of \( 3.2 \times 10^{-4} \text{K} \). When temperature decreases, the width of the strip in which many-spin entanglement exists increases. At a temperature of \( T = 1.6 \times 10^{-4} \text{K} \) (Fig. 2c), clusters of 19–87 entangled spins appear, and, at a temperature of \( T = 4.8 \times 10^{-5} \text{K} \) (Fig. 2d), 11–92 entangled spins are observed.

The temperature dependence of the maximum number of entangled spins during evolution \( 0 \leq D_T \leq 3 \) for different numbers of spins in the nanopore is shown in Fig. 3. The maximum number of entangled spins \( n_{\text{max}} \) decreases with increasing temperature. The maximum number of entangled spins increases as the number of spins in the nanopore increases, because the system in the nanopore becomes denser.

6. CONCLUSIONS

We have investigated many-spin entanglement in a system of spin-carrying molecules (atoms) filling nonspherical nanopores under the conditions of an MQ NMR spectroscopy experiment. The spins were initially in a dipolar ordered state. We have found the dependence of many-spin entanglement on the temperature and the number of spins in the nanopore.

The studies performed allow us to conclude that MQ NMR spectroscopy is a subtle and useful method for studying various problems of quantum informatics. In particular, it is a very effective method for studying quantum entanglement.
Two-Pulse Broekaert–Jeener Experiment at Low Zeeman Temperature and High Dipolar Temperature

Initially, the system is in a state of thermodynamic equilibrium in a strong external magnetic field with the density matrix

\[ \sigma_i = \frac{\exp(\beta z \omega_0 I_z)}{Z_i}, \quad Z_i = \text{Tr}(\exp(\beta_z \omega_0 I_z)), \]  

(A.1)

where \( \beta_z \) is proportional to the inverse temperature of the lattice. After the first resonant \( x \)-pulse, we obtain

\[ \sigma'(0) = \exp\left(\frac{i \pi}{2} I_x\right) \sigma_i \exp\left(-i \frac{\pi}{2} I_x\right) = \frac{\exp(\beta_z \omega_0 I_x)}{Z_i}, \]  

(A.2)

where \( I_\alpha \) is the operator of the projection of the total spin angular momentum onto the axis \( \alpha = x, y, z \). Then the system freely evolves over time \( \tau \), and after that a second resonant \( y \)-pulse is applied, which rotates the spins through angle \( \theta \) around the \( y \)-axis of the RRF. As a result, we find that

\[ \]
After time $T_2$ ($T_2$ is the spin relaxation time [17]), the system reaches a state of thermodynamic equilibrium,
\[ \sigma_f = \frac{\exp(\alpha_Z \omega_h I_z + \beta_d H_{dc})}{Z_f}, \quad (A.4) \]
where $\alpha_Z$ and $\beta_d$ are the inverse Zeeman and dipolar temperatures. It is obvious that the system has a single equilibrium state, and the temperatures $\alpha_Z$ and $\beta_d$ in the equilibrium state are determined from the conservation laws:
\[ \text{Tr}[I_\sigma(\tau)] = \text{Tr}[I_\sigma(\tau)] = \text{Tr}[I_\sigma(\tau)], \quad (A.5) \]
\[ \text{Tr}[H_{dc}\sigma(\tau)] = \text{Tr}[H_{dc}\sigma(\tau)]. \quad (A.6) \]

We can rewrite $\text{Tr}[I_\sigma(\tau)]$ as
\[ \text{Tr}[I_\sigma(\tau)] = \frac{1}{Z_i} \text{Tr}[\exp(-i\Theta I_y)\exp(-iH_{dc}.\tau)] \]
\[ \times \exp(-iH_{dc}.\tau) \exp(\beta_d \omega_h I_y) \exp(iH_{dc}.\tau) \]
\[ = \frac{1}{Z_i} \text{Tr}[(I_z \cos \theta - I_x \sin \theta) \exp(-iH_{dc}.\tau)] \]
\[ \times \exp(\beta_d \omega_h I_y) \exp(iH_{dc}.\tau)] \]
\[ = \frac{1}{Z_i} \text{Tr}[\exp(-i\pi I_y)(I_z \cos \theta - I_x \sin \theta)] \]
\[ \times \exp(-iH_{dc}.\tau) \exp(\beta_d \omega_h I_y) \exp(iH_{dc}.\tau) \exp(i\pi I_y)] \]
\[ = -\frac{1}{Z_i} \text{Tr}[(I_z \cos \theta - I_x \sin \theta) \exp(-iH_{dc}.\tau)] \]
\[ \times \exp(\beta_d \omega_h I_y) \exp(iH_{dc}.\tau)] = 0. \quad (A.7) \]

In expression (A.7), we took into account that $[\exp(-i\pi I_y), H_{dc}] = 0$. Since we consider the case of a high dipolar temperature, Eq. (A.5) can be rewritten as
\[ 0 = \frac{1}{Z_f} \text{Tr}[I_z \exp(\alpha_Z \omega_h I_z)] \]
\[ + \frac{\beta}{Z_f} \text{Tr}[I_z \exp(\alpha_Z \omega_h I_z)H_{dc}]. \quad (A.8) \]

Notice that $\text{Tr}[I_z] = \text{Tr}[I_z H_{dc}] = 0$. In such a case, $\alpha_Z = 0$ satisfies Eq. (A.5). Thus, in this case we obtain a dipolar ordered state.

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Fig. 3. The maximum number of entangled spins, averaged over the evolution time ($0 \leq D \tau \leq 3$) as a function of temperature for (a) $N = 51$, (b) $N = 75$ (b), and (c) $N = 101$. 
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