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Magnus expansion paradoxes in the study of equilibrium magnetization and entanglement in multi-pulse spin locking

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Abstract. Divergence of the Magnus expansion leads to paradoxes in the spin dynamics of solid-state NMR and in quantum informatics. This review presents results on quasi-equilibrium magnetization in a system of dipole–dipole (DD) coupled spins at times $T_2 \ll t \ll T_{1\rho}$ in multiple-pulse spin locking (T_2 is the transverse spin relaxation time and $T_{1\rho}$ is the rotating-frame spin–lattice relaxation time). It is shown how contradictions between the results obtained with the Magnus expansion and experimental data can be removed. Systems of two and three DD coupled spins in multi-pulse spin locking are considered, and the entanglement evolution is investigated using both the Magnus expansion and the exact solution. The critical temperature for an entangled state is also found.

Keywords: Floquet theorem, Floquet Hamiltonian, Magnus expansion, Magnus paradox, multi-pulse spin locking, average Hamiltonian theory, concurrence, quantum entanglement

1. Introduction

Many physical problems can be described in terms of systems of linear differential equations with periodic time-dependent coefficients [1] (in quantum mechanics, with a Hamiltonian periodically varying in time). In 1883, A M Gaston Floquet proved a remarkable theorem that asserts the existence of a periodic unitary transformation that maps a system of normal differential equations with periodic coefficients into a system of differential equations with constant coefficients [2]. A well-known example of such a procedure is the passage to a rotating reference frame (RRF) in the study of a system with dipole–dipole interactions (DDIs) in a constant magnetic field and in a circularly polarized (harmonic) magnetic field [3]. The Floquet theorem allows writing the solution of the Liouville evolution equation (in frequency units)

$$\mathrm{i}\,\frac{\mathrm{d}\rho}{\mathrm{d}t} = \left[\mathcal{H}(t), \rho(t)\right] \tag{1}$$

for the density matrix $\rho(t)$ in the form

$$\rho(t) = P(t)\exp\left(-\mathrm{i}\mathcal{H}_{\mathrm{F}}t\right)\rho_0\exp\left(\mathrm{i}\mathcal{H}_{\mathrm{F}}t\right)P^{+}(t)\,. \tag{2}$$

In (1), $\mathcal{H}(t)$ is a periodic Hamiltonian of the system [$\mathcal{H}(t+t_{\mathrm{c}}) = \mathcal{H}(t)$, where t_{c} is the period], $\mathcal{H}_{\mathrm{F}}$ is a time-independent Hamiltonian (Floquet Hamiltonian), $P(t)$ is a unitary periodic operator, $P(t+t_{\mathrm{c}}) = P(t)$, and ρ_0 is the initial density matrix.

Unfortunately, except for the above-mentioned example with an RRF, the Floquet Hamiltonian for multi-spin systems cannot be computed exactly, and approximate methods should therefore be used [4–8]. We discuss the method described in [4], known as the average Hamiltonian theory, in detail.

Let the Hamiltonian $\mathcal{H}(t)$ of a system consist of two parts,

$$\mathcal{H}(t) = \mathcal{H}_p(t) + \mathcal{H}_{\mathrm{in}}\,, \tag{3}$$

where $\mathcal{H}_{\mathrm{in}}$ describes internal interactions (for example, DDIs), and $\mathcal{H}_p(t)$ ($\mathcal{H}_p(t+t_{\mathrm{c}}) = \mathcal{H}_p(t)$) describes the effect exerted on the system by external fields (for example, by a periodic sequence of high-frequency (HF) pulses). The

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evolution of the density matrix can be conveniently represented with the aid of the evolution operator

$$\rho(t) = U(t)\rho_0 U^+(t)\,, \tag{4}$$

where the operator $U(t)$ satisfies the equation

$$\mathrm{i}\,\frac{\mathrm{d}U(t)}{\mathrm{d}t} = \mathcal{H}(t)U(t)\,, \qquad U(0) = 1\,. \tag{5}$$

To eliminate external actions from Hamiltonian (3), we pass to the interaction representation in terms of $\mathcal{H}_p(t)$. Then the evolution operator $U(t)$ can be written as

$$U(t) = U_p(t)L(t)\,, \tag{6}$$

where

$$U_p(t) = \mathrm{Texp}\left(-\mathrm{i}\int_0^t \mathcal{H}_p(t')\,\mathrm{d}t'\right). \tag{7}$$

The operator $L(t)$ in (6) is defined as

$$L(t) = \mathrm{Texp}\left(-\mathrm{i}\int_0^t \widetilde{\mathcal{H}}(t')\,\mathrm{d}t'\right), \tag{8}$$

where Texp is the time-ordered exponential and

$$\widetilde{\mathcal{H}}(t) = U_p^+(t)\mathcal{H}_{\mathrm{in}}U_p(t)\,. \tag{9}$$

We now assume that the Hamiltonian $\mathcal{H}_p(t)$ is not only periodic but also satisfies the condition of cyclicity, i.e.,

$$U_p(Nt_\mathrm{c}) = \mathrm{Texp}\left(-\mathrm{i}\int_0^{Nt_\mathrm{c}} \mathcal{H}_p(t)\,\mathrm{d}t\right) = 1\,, \tag{10}$$

where N is a natural number. From formulas (6) and (9), we then obtain

$$\rho(Nt_\mathrm{c}) = L(Nt_\mathrm{c})\rho_0 L^+(Nt_\mathrm{c})\,, \tag{11}$$

$$L(Nt_\mathrm{c}) = \left[L(t_\mathrm{c})\right]^N. \tag{12}$$

Thus, in order to describe the state of the system at time instants that are multiples of the period t_c, it suffices to find the evolution of the system only over one period [4]. This conclusion is very important and useful for the investigation of multi-spin dynamics in periodic magnetic fields.

However, the problem is complicated by the presence of time-ordered exponentials (T operators) in Eqns (8) and (12), which lead to labor-consuming calculations. In 1954, Magnus suggested a transformation of the T operator into a simple exponential operator [9]. Magnus showed that

$$\begin{aligned} L(t_\mathrm{c}) &= \mathrm{Texp}\left(-\mathrm{i}\int_0^{t_\mathrm{c}} \widetilde{\mathcal{H}}(t')\,\mathrm{d}t'\right) \\ &= \exp\left[-\mathrm{i}\left(\bar{\mathcal{H}}^{(0)} + \bar{\mathcal{H}}^{(1)} + \bar{\mathcal{H}}^{(2)} + \ldots\right)t_\mathrm{c}\right], \end{aligned} \tag{13}$$

where the average Hamiltonian $\bar{\mathcal{H}}^{(0)}$ has the form

$$\bar{\mathcal{H}}^{(0)} = \frac{1}{t_\mathrm{c}}\int_0^{t_\mathrm{c}} \widetilde{\mathcal{H}}(t')\,\mathrm{d}t'\,, \tag{14}$$

and the corrections $\bar{\mathcal{H}}^{(1)}$ and $\bar{\mathcal{H}}^{(2)}$ to the average Hamiltonian are defined by the formulas

$$\bar{\mathcal{H}}^{(1)} = \frac{1}{2t_\mathrm{c}}\int_0^{t_\mathrm{c}} \mathrm{d}t_2 \int_0^{t_2} \mathrm{d}t_1 \left[\widetilde{\mathcal{H}}(t_2), \widetilde{\mathcal{H}}(t_1)\right], \tag{15}$$

$$\begin{aligned} \bar{\mathcal{H}}^{(2)} = \frac{1}{6t_\mathrm{c}}\int_0^{t_\mathrm{c}} \mathrm{d}t_3 \int_0^{t_3} \mathrm{d}t_2 \int_0^{t_2} \mathrm{d}t_1 \Big\{ &\left[\widetilde{\mathcal{H}}(t_3), \left[\widetilde{\mathcal{H}}(t_2), \widetilde{\mathcal{H}}(t_1)\right]\right] \\ &+ \left[\widetilde{\mathcal{H}}(t_1), \left[\widetilde{\mathcal{H}}(t_2), \widetilde{\mathcal{H}}(t_3)\right]\right]\Big\}, \end{aligned} \tag{16}$$

etc.

Let the order of the Hamiltonian $\widetilde{\mathcal{H}}(t)$ be equal to ω_loc. The corrections to the average Hamiltonian are by the order of magnitude proportional to powers of the small parameter $\varepsilon = \omega_\mathrm{loc}t_\mathrm{c} \ll 1$. In particular, in many cases in studying the multi-spin dynamics, we can restrict ourselves to using the average Hamiltonian. For example, the average Hamiltonian completely describes a system if the values of the Hamiltonian at different time instants commute. It is important that all the terms of series (13) are proportional to the number of spins, and this series can be used to investigate the dynamics and thermodynamics of spin systems. Expansion (13) can be used for determining the evolution of a system at the time instants that are multiple of the period of an external magnetic field. This is sufficient for describing experiments using a stroboscopic observation of magnetization.

Magnus expansion (13) allows substantially simplifying the analysis of the behavior of spin systems in periodic external fields. However, the series in Eqn (13) is divergent [10] even in the case of a single-spin system in a constant magnetic field and in a circularly polarized (harmonic) magnetic field perpendicular to it [10]. Experimental studies [11, 12] of spin dynamics in the case of multi-pulse spin locking [13, 14] revealed some inconsistencies between the average Hamiltonian theory [4] and experimental data at times $T_2 < t \leqslant T_{1\rho}$ (where T_2 is the time of transverse spin relaxation and $T_{1\rho}$ is the time of spin–lattice relaxation in the RRF [3]). These inconsistencies were called the Magnus paradox in [15]. In [10], it was shown that the Magnus paradox is closely related to the convergence of expansion (13). The problem of the convergence of the Magnus series was investigated earlier in many studies [10, 16–18] and continues to be investigated at present [19].

In this article, we examine the Magnus paradox in multi-spin and few-spin problems and show how the contradictions between the average Hamiltonian theory [4] and the experimental data can be removed [11, 12] in multi-pulse spin locking. We also investigate the evolution of entanglement in a system consisting of two and three dipole-coupled spins in the case of multi-pulse spin locking. Entanglement in the two-spin system was investigated on the basis of the obtained exact solution of the two-spin problem and also in terms of the average Hamiltonian theory. In the Conclusion, we briefly list of our basic results.

2. Thermodynamic quasi-equilibrium states of systems of spins coupled by dipole–dipole interaction in multi-pulse spin locking

We examine a system of spins $s = 1/2$ coupled by a DDI in a strong external magnetic field directed along the z axis of a laboratory coordinate system. The secular part of the DDI

with respect to the field $\mathbf{H}_0$ can be expressed as

$$\mathcal{H}_{dz} = \sum_{i<j} d_{ij}\left(3I_{iz}I_{jz} - \mathbf{I}_i\,\mathbf{I}_j\right), \tag{17}$$

where $I_{i\alpha}$ ($\alpha = x, y, z$) is the projection of the angular momentum of spin i onto the axis α, $\mathbf{I}_i\,\mathbf{I}_j = I_{ix}I_{jx} + I_{iy}I_{jy} + I_{iz}I_{jz}$, and d_{ij} is the DDI constant of spins i and j.

In the case of multi-pulse spin locking, the HF pulse $P_{-y}^{90^\circ}$ at the initial instant rotates the spins about the y axis of the RRF through 90°. Then the sequence of resonance equidistant pulses P_x^{φ} rotates the spins through an angle φ about the x axis of the RRF. The arrangement of the multi-pulse sequence used in the experiment can be represented as [20, 21]

$$P_{-y}^{90^\circ} - \tau - (P_x^{\varphi} - 2\tau -)^N, \tag{18}$$

where τ and 2τ are the time intervals between subsequent pulses. It was experimentally shown in [13, 14, 22, 23] that at $\varphi = \pi/2$, sequence (18) creates a long chain of echo signals, which decay with time as $T_{2e} \sim T_2/(\omega_{loc}\tau)^4$. This time exceeds the time of decay of the free-induction signal (FIS) by several orders of magnitude.

The Hamiltonian of the system in the case of multi-pulse spin locking takes the form

$$\mathcal{H} = \omega_0 I_z - f(t)\left(I_x \cos(\omega_0 t) + I_y \sin(\omega_0 t)\right) + \mathcal{H}_{dz}, \tag{19}$$

where $\omega_0 = \gamma H_0$ is the Larmor frequency (γ is the gyromagnetic ratio), $I_\alpha = \sum_i I_{i\alpha}$ is the total projection of the angular spin moment onto the axis α ($\alpha = x, y, z$), and

$$f(t) = \varphi \sum_{k=0}^{\infty} \delta(\tau + 2k\tau - t), \tag{20}$$

is a pulsed function, where $\delta(t)$ is the Dirac delta function. The Liouville equation for the density matrix $\rho(t)$ of the system can be written in the RRF as

$$\mathrm{i}\,\frac{\mathrm{d}\rho}{\mathrm{d}t} = \left[-f(t)I_x + \mathcal{H}_{dz},\, \rho(t)\right]. \tag{21}$$

It is easy to see that

$$\mathcal{H}_{dz} = -\frac{1}{2}\,\mathcal{H}_{dx} + \mathcal{H}^{(2)} + \mathcal{H}^{(-2)}, \tag{22}$$

where $\mathcal{H}_{dx}$ is obtained from $\mathcal{H}_{dz}$ (17) by replacing the index z with the index x, and

$$\mathcal{H}^{(2)} = \frac{3}{4}\sum_{i<j} d_{ij} I_i^+ I_j^+, \qquad \mathcal{H}^{(-2)} = \frac{3}{4}\sum_{i<j} d_{ij} I_i^- I_j^-. \tag{23}$$

We note that the quantization axis in the left-hand side of (22) is the z axis, whereas in the right-hand side, it is the x axis. Using the transformation

$$\rho(t) = L(t)\bar{\rho}L^+(t), \tag{24}$$

where

$$L(t) = \exp\left[\mathrm{i}\left(\int_0^t f(t')\,\mathrm{d}t' - \omega_e t\right)I_x\right], \tag{25}$$

we obtain the following equation for the density matrix $\bar{\rho}(t)$:

$$\mathrm{i}\,\frac{\mathrm{d}\bar{\rho}(t)}{\mathrm{d}t} = \left[\omega_e I_x - \frac{1}{2}\,\mathcal{H}_{dx} + \Phi(t)\mathcal{H}^{(2)} + \Phi^*(t)\mathcal{H}^{(-2)},\, \bar{\rho}(t)\right]. \tag{26}$$

In Eqn (26), $\omega_e = \varphi/(2\tau)$ is the effective field and $\Phi(t)$ is a periodic function with the mean $(1/\varphi)\sin\varphi$. The average Hamiltonian $\bar{\mathcal{H}}$ can be represented (up to terms of the order of $\varepsilon\omega_{loc}$, where $\varepsilon = 2\tau\omega_{loc}$, $\omega_{loc} \sim T_2^{-1}$) in the form

$$\bar{\mathcal{H}} = \omega_e I_x - \frac{1}{2}\,\mathcal{H}_{dx} + \frac{\sin\varphi}{\varphi}\left(\mathcal{H}^{(2)} + \mathcal{H}^{(-2)}\right). \tag{27}$$

In addition to average Hamiltonian (27), the total Hamiltonian contains small rapidly oscillating terms, which can be disregarded at times $t \leqslant T_2$. By the instant $t \sim T_2$, the spin system reaches a quasi-equilibrium state described by the density matrix ρ_{eq}, which in the high-temperature approximation [24, 25] can be represented as

$$\rho_{eq} = Z^{-1}(1 - \beta\bar{\mathcal{H}}), \qquad Z = \mathrm{Tr}\,(1), \tag{28}$$

where β is the inverse temperature. Taking into account that the initial density matrix ρ_0 in the multi-pulse spin locking [see (18)] has the form

$$\rho_0 = Z^{-1}(1 - \beta_0\omega_0 I_x), \qquad Z = \mathrm{Tr}\,(1), \tag{29}$$

where β_0 is the initial inverse temperature, we use the energy conservation law

$$\mathrm{Tr}\,\{\rho_0\bar{\mathcal{H}}\} = \mathrm{Tr}\,\{\rho_{eq}\bar{\mathcal{H}}\} \tag{30}$$

to derive the relation

$$\frac{\beta}{\beta_0} = \omega_e\omega_0\left[\omega_e^2 + \left(\frac{1}{4} + \frac{3}{4}\,\frac{\sin^2\varphi}{\varphi^2}\right)\omega_{loc}^2\right]^{-1}, \tag{31}$$

where $\omega_{loc}^2 = \mathrm{Tr}\,(\mathcal{H}_{dx}^2)/\mathrm{Tr}\,(I_x^2)$, and it follows from (22) that

$$\frac{\mathrm{Tr}\left(\mathcal{H}^{(2)}\mathcal{H}^{(-2)}\right)}{\mathrm{Tr}\,(I_x^2)} = \frac{3}{8}\,\omega_{loc}^2. \tag{32}$$

Letting M_0 and M_{eq} denote the initial and quasi-equilibrium magnetizations, we use Eqns (28), (30), and (31) to obtain the following result:

$$\frac{M_{eq}}{M_0} = \frac{\mathrm{Tr}\,(\rho_{eq}I_x)}{\mathrm{Tr}\,(\rho_0 I_x)} = \omega_e^2\left[\omega_e^2 + \left(\frac{1}{4} + \frac{3}{4}\,\frac{\sin^2\varphi}{\varphi^2}\right)\omega_{loc}^2\right]^{-1}. \tag{33}$$

The theoretical result in (33) agrees well with the experimental data in [11] at the times $t \sim T_2$.

Corrections (13) to time-independent Hamiltonian (27) are proportional to powers of the small parameter $\varepsilon = 2\tau\omega_{loc} \ll 1$ and are small by themselves. Therefore, it can be assumed that Hamiltonian (27) approximately determines the Floquet Hamiltonian [see (2)]. By extending the Redfield hypothesis on the spin temperature in an RRF [26], it can be assumed that thermodynamic equilibrium determined by the Floquet Hamiltonian $\mathcal{H}_F$ eventually sets in. The equilibrium density matrix in the high-temperature approximation [24, 25] has the form

$$\rho_{eq} = Z^{-1}(1 - \beta_{eq}\mathcal{H}_F), \qquad Z = \mathrm{Tr}\,(1), \tag{34}$$

where β_{eq} is the inverse spin temperature in equilibrium. If average Hamiltonian (27) gives an approximate expression for the Floquet Hamiltonian $\mathcal{H}_F$, we can conclude that the equilibrium magnetization M_{eq} defined as

$$M_{eq} = \mathrm{Tr}\,\{\rho_{eq}I_n\} \tag{35}$$

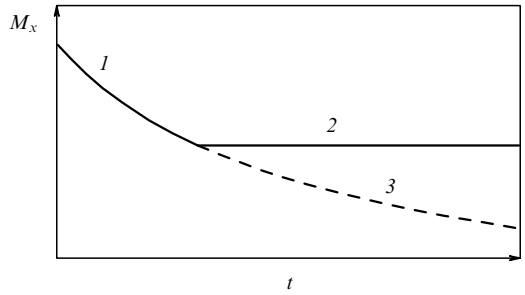

Figure 1. Time dependence of the magnetization M_x (schematic): (*1*) variation of M_x in the time interval preceding the establishment of equilibrium (34); (*2*) equilibrium magnetization (35); and (*3*) variation of the magnetization according to experimental data [12].

is nonzero. (In (35), **n** is the quantization axis for the Hamiltonian $\mathcal{H}_F$.) However, the experimental data in [12] show that at times $T_2 \ll t \ll T_{1\rho}$, the quasi-equilibrium polarization decreases to zero. A theoretical analysis based on Provotorov's equations [27] leads to the same result. The evolution of the magnetization in accordance with Eqns (34) and (35) and the corresponding experimental data [12] are presented schematically in Fig. 1. The theoretical results based on the description of the spin dynamics in a periodic magnetic field and on thermodynamic considerations are inconsistent with the experimental data. This result was first obtained in our study [21]; it was called the Magnus paradox by Abragam and Goldman [15]. This name of the paradox is connected with the approximate method of the definition of the Floquet Hamiltonian via expansion (13).

We now discuss the reasons for the appearance of the Magnus paradox. First of all, we note that the above evolution of the spin system in a periodic magnetic field can be described in terms of different time-independent Hamiltonians. Indeed, we assume that a time-independent Hamiltonian $\bar{\mathcal{H}}$ has been found using Eqn (13). Then the evolution of the system over a single period is determined by

$$L(t_c) = \exp(-i\bar{\mathcal{H}}t_c). \tag{36}$$

For an arbitrary direction **m** ($|\mathbf{m}| = 1$) in the spin space for spins 1/2, we have $\exp(-4\pi i N\mathbf{m}\hat{S}) = 1$, where N is an integer. From Eqns (6)–(9), we find

$$\exp(-4\pi i N\mathbf{m}\hat{S})\exp(-i\bar{\mathcal{H}}t_c) = \exp\left[-i\left(\frac{4\pi N}{t_c}(\mathbf{m}\hat{S}) + \tilde{\tilde{\mathcal{H}}}\right)t_c\right], \tag{37}$$

where the Hamiltonian $\tilde{\tilde{\mathcal{H}}}$ satisfies the equation

$$\mathrm{T}\exp\left[-i\int_0^{t_c}\exp\left(\frac{4i\pi N}{t_c}\,\mathbf{m}\hat{S}t\right)\tilde{\tilde{\mathcal{H}}}\exp\left(-\frac{4i\pi N}{t_c}\,\mathbf{m}\hat{S}t\right)dt\right] = \exp(-i\bar{\mathcal{H}}t_c). \tag{38}$$

Another reason for the appearance of the Magnus paradox is related to the divergence of the Magnus expansion [10]. It can be assumed that the Magnus series that was used to obtain average Hamiltonian (27) diverges at times $T_2 \ll t \ll T_{1\rho}$. To simplify the subsequent analysis, we also assume that $\varphi \approx 2\pi/n$, where n is an even number, and $|\varphi - 2\pi/n| \lesssim \varepsilon$. Under these conditions, the heating of the system at $t \gg T_2$ is mainly determined by the n-spin resonance process, where the simultaneous flip of n dipole-coupled spins leads to the absorption of energy, which is determined by some harmonic of the periodic external field. The excess (lack) of energy in the case of such a flip of n spins is compensated by a transfer of part of the energy into the dipole reservoir [25]. The probability of this process is expressed as [20]

$$W \sim \varepsilon^n \omega_{loc} \exp\left[-\frac{(2\pi - n\varphi)^2}{6\varepsilon^2}\right]. \tag{39}$$

It follows from Eqn (39) that the above n-spin resonance process can be neglected at times $t \lesssim T_2$, and Hamiltonian (27) at these times gives a suitable approximation to the Floquet Hamiltonian.

But the n-spin resonance process cannot be ignored at $t \sim W^{-1} \gg T_2$. On the other hand, such a resonance process cannot be taken into account by adding the higher-order terms in (13) to (27) because of the divergence of the Magnus expansion.

To obtain an approximate time-independent Hamiltonian describing the dynamics of the system at times $t \sim W^{-1}$, we pass to a coordinate system rotating with the frequency $\pi/(n\tau)$ about the x axis of the RRF used above and perform the averaging according to the preceding scheme. Then the average Hamiltonian $\mathcal{H}_0'$ takes the form

$$\mathcal{H}_0' = \left(\frac{\varphi}{2\tau} - \frac{\pi}{n\tau}\right)I_x - \frac{1}{2}\,\mathcal{H}_{dx}. \tag{40}$$

By the time $t \sim W^{-1}$, the spin system reaches a quasi-equilibrium state,

$$\rho_{qe}' = \frac{1}{Z}(1 - \alpha_{qe}'\mathcal{H}_0'), \qquad Z = \mathrm{Tr}(1), \tag{41}$$

where α_{qe}' is the corresponding inverse spin temperature. It can be easily verified that quasi-equilibrium state (41) leads to a smaller quasi-equilibrium magnetization than quasi-equilibrium state (28). Therefore, the slow decay of the magnetization at times $T_{1\rho} \geqslant t \geqslant T_2$, which is easily explained in terms of Provotorov's equations [27], can also be described using a Floquet Hamiltonian. In this approach, the decay of the magnetization is explained by a rearrangement of the quasi-equilibrium states. Notably, quasi-equilibrium state (28) in the system at times $t \approx T_2$ in the case under consideration is rearranged at $t \approx W^{-1}$ into quasi-equilibrium state (41).

3. Entanglement in a system of two (three) spins connected by dipole–dipole interaction in multi-pulse spin locking

We come across another example of the Magnus paradox when studying the problem of entanglement [28] in a system consisting of two (three) spins coupled by DDIs in the case of multi-pulse spin locking [13, 14]. Instead of pulse sequence (18), we here consider the sequence

$$P_{-y}^{90^\circ} - \tau - (P_x^{90^\circ} - a\tau - P_x^{90^\circ} - 2\tau)^N, \tag{42}$$

where a is a free parameter. In the standard basis $|00\rangle$, $|01\rangle$, $|10\rangle$, $|11\rangle$ that is used for solving problems of quantum informatics in two-spin systems [28], the matrix representation of Hamiltonian (17) has the form

$$\mathcal{H}_{dz} = \frac{d_{12}}{2}\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & -1 & 0 \\ 0 & -1 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \tag{43}$$

After a preparatory pulse applied at the instant $t = 0$, the density matrix ρ_0 of the system at an arbitrary temperature acquires the form

$$\rho_0 = \frac{1}{Z}\exp(\beta I_x), \qquad \beta = \frac{\hbar\omega_0}{k_B T}, \qquad Z = 4\cosh^2\frac{\beta}{2}. \quad (44)$$

Under the action of pulse sequence (42) on the system, just as in [13, 14], anisotropic DDIs are described by the Hamiltonians $\mathcal{H}_{dz}$ and $\mathcal{H}_{dy}$, with

$$\mathcal{H}_{dy} = d_{12}(3I_{1y}I_{2y} - \mathbf{I}_1\mathbf{I}_2). \quad (45)$$

As a result, the operator of the evolution of the system over a single period of the pulse sequence $U((a+2)\tau)$ acquires the form

$$U\big((a+2)\tau\big) = \exp(-i\tau\mathcal{H}_{dz})\exp(-ai\tau\mathcal{H}_{dy})\exp(-i\tau\mathcal{H}_{dz}), \quad (46)$$

and the density matrix of the system $\rho((a+2)\tau)$ after the evolution can be represented as

$$\rho\big((a+2)\tau\big) = U\big((a+2)\tau\big)\rho_0 U^+\big((a+2)\tau\big). \quad (47)$$

In a similar way, the density matrix $\rho((a+2)M\tau)$ after M periods of pulse sequence (42) can be expressed as

$$\rho\big((a+2)M\tau\big) = U\big((a+2)M\tau\big)\rho_0 U^+\big((a+2)M\tau\big). \quad (48)$$

After simple calculations, we obtain

$$\exp(-i\tau\mathcal{H}_{dz}) = \begin{pmatrix} e^{-it/2} & 0 & 0 & 0 \\ 0 & \dfrac{1+e^{it}}{2} & \dfrac{e^{it}-1}{2} & 0 \\ 0 & \dfrac{e^{it}-1}{2} & \dfrac{1+e^{it}}{2} & 0 \\ 0 & 0 & 0 & e^{-it/2} \end{pmatrix},$$

$$\exp(-ia\tau\mathcal{H}_{dy}) = \frac{1}{2}\begin{pmatrix} e^{-iat/2}+e^{iat} & 0 & 0 & e^{iat}-e^{-iat/2} \\ 0 & 1+e^{-iat/2} & e^{-iat/2}-1 & 0 \\ 0 & e^{-iat/2}-1 & 1+e^{-iat/2} & 0 \\ e^{iat}-e^{-iat/2} & 0 & 0 & e^{-iat/2}+e^{iat} \end{pmatrix}. \quad (49)$$

In matrices (49), $t = \tau d_{12}$ is the dimensionless time. Using (49), we obtain a matrix representation of the evolution operator $U((a+2)\tau)$ in the form

$$U\big((a+2)\tau\big) = \frac{1}{2} \times \begin{pmatrix} e^{-i\frac{a+2}{2}t}+e^{i(a-1)t} & 0 & 0 & -e^{-i\frac{a+2}{2}t}+e^{i(a-1)t} \\ 0 & 1+e^{-i\frac{a-4}{2}t} & e^{-i\frac{a-4}{2}t}-1 & 0 \\ 0 & e^{-i\frac{a-4}{2}t}-1 & 1+e^{-i\frac{a-4}{2}t} & 0 \\ -e^{-i\frac{a+2}{2}t}+e^{i(a-1)t} & 0 & 0 & e^{-i\frac{a+2}{2}t}+e^{i(a-1)t} \end{pmatrix}. \quad (50)$$

Matrix (50) is central-symmetric (CS) ($u_{ij} = u_{5-i,5-j}$, $i, j = 1, 2, 3, 4$) [29] and, using the orthogonal transformation

$$G = \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 0 & 0 & 1 \\ 0 & 1 & 1 & 0 \\ 0 & 1 & -1 & 0 \\ 1 & 0 & 0 & -1 \end{pmatrix} \quad (51)$$

it can be decomposed into two blocks with 2×2 dimensions. As a result, we obtain the following analytic expression for the matrix representation of $\rho((a+2)M\tau)$ after M periods of pulse sequence (42):

$$\rho\big((a+2)M\tau\big) = U\big((a+2)M\tau\big)\rho_0 U^+\big((a+2)M\tau\big) = \frac{1}{8\cosh^2(\beta/2)} \times \begin{pmatrix} 2\cosh^2\frac{\beta}{2} & \sinh\beta\, e^{i\frac{3a-6}{2}Mt} & \sinh\beta\, e^{i\frac{3a-6}{2}Mt} & 2\sinh^2\frac{\beta}{2} \\ \sinh\beta\, e^{-i\frac{3a-6}{2}Mt} & 2\cosh^2\frac{\beta}{2} & 2\sinh^2\frac{\beta}{2} & \sinh\beta\, e^{-i\frac{3a-6}{2}Mt} \\ \sinh\beta\, e^{-i\frac{3a-6}{2}Mt} & 2\sinh^2\frac{\beta}{2} & 2\cosh^2\frac{\beta}{2} & \sinh\beta\, e^{-i\frac{3a-6}{2}Mt} \\ 2\sinh^2\frac{\beta}{2} & \sinh\beta\, e^{i\frac{3a-6}{2}Mt} & \sinh\beta\, e^{i\frac{3a-6}{2}Mt} & 2\cosh^2\frac{\beta}{2} \end{pmatrix}. \quad (52)$$

The density matrix (52) is also CS. Entanglement in the system exists if the 'concurrence' (as defined by Wootters [30]) is positive. To calculate the concurrence, the matrix $\tilde\rho((a+2)M\tau) = (\sigma_y\otimes\sigma_y)\rho^*(\sigma_y\otimes\sigma_y)$ must be found, where σ_y is the Pauli matrix. The concurrence is determined in [30] via the square roots of the eigenvalues of the product of matrices $\rho((a+2)M\tau)\tilde\rho((a+2)M\tau)$. Taking into account that the product of CS matrices again gives a CS matrix, the following formulas can be obtained [29] for the square roots of its eigenvalues:

$$\lambda_1 = \frac{1}{2}\left\{\frac{1}{2}\sqrt{\left(1+\tanh^2\frac{\beta}{2}\right)^2 - 4\tanh^2\frac{\beta}{2}\cos^2\left(\frac{3a-6}{2}Mt\right)} + \tanh\frac{\beta}{2}\left|\sin\left(\frac{3a-6}{2}Mt\right)\right|\right\}, \quad (53)$$

$$\lambda_2 = \frac{1}{2}\left\{\frac{1}{2}\sqrt{\left(1+\tanh^2\frac{\beta}{2}\right)^2 - 4\tanh^2\frac{\beta}{2}\cos^2\left(\frac{3a-6}{2}Mt\right)} - \tanh\frac{\beta}{2}\left|\sin\left(\frac{3a-6}{2}Mt\right)\right|\right\}, \quad (54)$$

$$\lambda_3 = \lambda_4 = \frac{1-\tanh^2(\beta/2)}{4}. \quad (55)$$

Using formulas (53)–(55), we express the concurrence [30]

$$C = \max\{0,\ 2\lambda_{\max} - \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4\}, \quad (56)$$

where $\lambda_{\max} = \max\{\lambda_1, \lambda_2, \lambda_3, \lambda_4\}$, as

$$C = \max\left\{0,\ \tanh\frac{\beta}{2}\left|\sin\left(\frac{3a-6}{2}Mt\right)\right| - \frac{1-\tanh^2(\beta/2)}{2}\right\}. \quad (57)$$

The entanglement in the problem under consideration arises at the critical temperature

$$T_{cr} = \frac{\hbar\omega_0}{k_B\left|\ln(\sqrt{2}-1)\right|}, \quad (58)$$

and emerges at time instants

$$t = \frac{\pi + 2\pi n}{3|a-2|}\,, \tag{59}$$

where n is a nonnegative number. We note that the entanglement in the system is absent at the initial instant $t = 0$.

In the foregoing, the analysis of the entanglement has been carried out by exact methods, without any approximations. However, this problem can also be solved using the method described in Section 2, based on an approximate calculation of the Floquet Hamiltonian (average Hamiltonian). The average Hamiltonian for this system at $2\pi/(a+2)\tau \gg \omega_{\rm loc}$ has the form

$$\bar{\mathcal{H}} = \frac{2}{2+a}\,\mathcal{H}_{dz} + \frac{a}{2+a}\,\mathcal{H}_{dy}\,. \tag{60}$$

For $N = 2$, the Hamiltonians $\mathcal{H}_{dz}$ and $\mathcal{H}_{dy}$ commute; therefore, all corrections (15) and (16) to Hamiltonian (60) vanish. Hence, the calculations that use average Hamiltonian (60) (the Floquet Hamiltonian) give the same results as those based on the above exact solution.

At $N > 2$, however, the Hamiltonians $\mathcal{H}_{dz}$ and $\mathcal{H}_{dy}$, no longer commute in general and correction terms (15) and (16) to average Hamiltonian (60) do not vanish. Unfortunately, at $N > 2$, the entanglement cannot be studied by analytic methods. Our numerical investigation of the entanglement of the two-spin and single-spin subsystems in a three-spin system $(N = 3)$ shows that the results obtained without the use of average Hamiltonian (60) differ from the results obtained with its use. In particular, in the latter case, entangled states in the evolution of the system arise at times that differ from those obtained without the use of average Hamiltonian (6). These results can be considered to be a manifestation of the same Magnus paradox that was discussed in Section 2.

4. Conclusion

The Magnus expansion [9] for the Floquet Hamiltonian allows efficiently investigating problems of spin dynamics in periodic magnetic fields. Among many important results obtained with the use of the Magnus expansion [9], we especially note the creation of methods of high-resolution nuclear magnetic resonance (NMR) in solids [31, 32], the results of time-reversal experiments [33], and the development of the multi-quantum NMR method [34, 35]. Recent studies [36, 37] indicate the relation of the concepts considered in this paper to the physics of topological materials.

At the same time, the divergence of the Magnus expansion [10] and the ambiguity of the Floquet Hamiltonian lead to paradoxes, which should be taken into account in the theoretical analysis of the dynamics of spin systems in periodic magnetic fields. In this paper, we have revealed the physical factors responsible for these paradoxes. They are related to multi-spin resonance processes of the absorption of the energy of external magnetic fields. These processes lead to rearrangements of quasi-equilibrium states in the course of the system evolution.

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