We study the dynamical correlations between two coupled spins depending on time and the value of the spin quantum numbers. In the high-temperature approximation, we obtain analytic expressions for the mutual information and the quantum and classical parts of correlations. We consider both orthogonal and nonorthogonal measurements in the basis of spin coherent states. We show that at small times, the quantum part of correlations becomes much less than the classical part as the spin quantum numbers increase, while the situation is quite different at times equal to half the quantum period.

Keywords: spin dynamics, high-temperature approximation, mutual information, generalized measurement, spin coherent state, quantum correlation

1. Introduction

Up to now, numerous experiments have been performed using the nuclear magnetic resonance (NMR) method to demonstrate implementations of quantum algorithms on nuclear spins at high temperatures (see [1]). It is well known that there is no quantum entanglement, but the dynamical correlations arising in quantum calculations have both quantum and classical parts [2], [3]. Moreover, these high-temperature quantum correlations can ensure rather fast implementations of some quantum algorithms [1]–[7]. No explanations for the mechanisms of such an acceleration have yet been obtained although there are numerous works analyzing the relation between quantum and classical correlations (see, e.g., [7]).

Instead of arbitrary states of quantum systems for which no general solution describing the relation between the quantum and classical correlation parts has yet been obtained [7], we consider correlations typical of NMR [8]. Polarization is very small for nuclear spins in a strong static magnetic field at room temperature $T$, $\beta = \hbar \omega_0 / kT \approx 10^{-5} \ll 1$ ($\omega_0$ is the Larmor frequency), and the equilibrium density matrix used to describe the NMR is hence taken in the form

$$\hat{\rho}_{eq} = \frac{1 + \beta \hat{S}_z}{Z},$$

where $Z$ is the partition function, $\hat{S}_\alpha = \sum_j \hat{S}_{j\alpha}$, $\hat{S}_{j\alpha}$ is the $\alpha$-component of the $j$th spin operator, $\alpha = x, y, z$, and the magnetic field is directed along the $z$ axis. To observe the NMR signal, a radio-frequency (RF) magnetic field pulse is applied to the system, thus rotating the spins through the angle $90^\circ$ about the axis $y$ of the rotating reference frame (RRF),

$$\hat{\rho}(0) = \hat{Y} \hat{\rho}_{eq} \hat{Y}^{-1} = \frac{1 + \beta \hat{S}_x}{Z}.$$
This initial density matrix varies in time as

\[ \hat{\rho}(t) = \hat{U}(t)\hat{\rho}(0)\hat{U}^{-1}(t) = \frac{1 + \beta\hat{U}(t)\hat{S}_Z\hat{U}^{-1}(t)}{Z} \]

where \( \hat{U}(t) = e^{-i\hat{H}t/\hbar} \). In the course of time, polarization of separate spins in the field (the initial ordering of Zeeman subsystem (1)) becomes the correlation between the spin and its local field created by the dipole–dipole interactions. At small times, this correlation is described by a term of the form

\[ -\frac{i\beta t}{\hbar Z} [\hat{H}_d, \hat{S}_z] = -\frac{i\beta t}{Z} \sum_{i<j} a_{ij} [(3\hat{S}_{iz}\hat{S}_{jz} - \hat{S}_i \cdot \hat{S}_j), \hat{S}_z] = \frac{3\beta t}{Z} \sum_{i\neq j} a_{ij}\hat{S}_{iz}\hat{S}_{jy} \]

in expression (2). This correlation has been well studied for NMR because it is responsible for the solid echo [9] arising when the system is exposed to the action of a second RF pulse of rotation through 90° about the RRF \( x \) axis. If the second RF pulse rotates the spins by 45°, then the abovementioned correlation becomes the ordering of the spin–spin subsystem, which ensures a nonzero value of the average energy of dipole–dipole interactions [10]. But such correlations (3) have not yet been separated into the quantum and classical parts.

Here, we obtain such a separation for a model system. We take a system of two large spins \( S_1 \) and \( S_2 \) with a dipole–dipole interaction between the projections on the \( z \) axis given by the Hamiltonian

\[ \hat{H}_{SS} = \frac{J}{S_2} \hat{S}_{1z}\hat{S}_{2z}. \]

To study the limit as \( S_1, S_2 \rightarrow \infty \), we take the coupling constant divided by the spin value \( S_2 \geq S_1 \) following [11], [12]. Such a model allows tracing the transition from the case of two spins \( S_2 = S_1 = 1/2 \), where the dynamical correlations are equally separated into the quantum and classical parts [13], to the case of two classical magnetic momenta, where only one part remains. The large spin in nuclear systems is formed, for example, in the case of intermolecular interaction of fast rotating symmetric molecules of adamantane in a solid matrix. The fast rotation equates the dipole coupling constants between the 16 nuclei of \( ^1H \) in one molecule and the nucleus of \( ^13C \) in another molecule [14]. An even greater number of nuclei with equal coupling constants can be obtained for moving molecules in nanovoids [15].

It seems important to study the spins \( S > 1/2 \) not only in the NMR theory but also in the theory of quantum computers because quantum computations can be performed on qudits (quantum systems with \( d \) levels, \( d = 2S + 1 \) for spins). The use of qudits promises several advantages (e.g., the same dimension of the computational basis can be obtained on them for fewer spins \( 2^n \leftrightarrow d^n \) [16]–[18], but such systems have not yet been studied sufficiently well. There are many publications where measures of quantum correlations such as the quantum and geometric discords for two qudits (\( S = 1/2 \) for two spins) [7], [13], [19] or in the cases reducible to them [20]–[22] were calculated. These measures were calculated for several states with two spins \( S = 1 \) [23] and symmetric states with large \( d \) [24]. Their values were estimated for \( 2 \times n \) systems [3], [5], [25], [26], and the geometric discord was also estimated for \( m \times n \) systems [26]–[28].

The dynamics of correlations of two coupled large spins has not yet been investigated. We here consider it in the high-temperature approximation. We calculate the time-evolution of the density-matrix mutual information and the mutual information of the classical distribution function over directions, which were obtained by generalized measurements (POVM\(^1\) measurements) [7], [29] with a basis of spin coherent states (SCS) [22], [30], [31]. We study how the relation between the quantum and classical correlations depends on the spin value and compare the results obtained by orthogonal and nonorthogonal measurements.

\(^1\)Positive operator-valued measure.
2. Mutual information of two spins

We consider a system of two spins $S_1$ and $S_2$ with spin–spin coupling (4). For each spin, we use a basis of states with a certain value of the projection on the axis $z$, i.e., $|m⟩$, where $m$ takes $2S+1$ values,

$$-S, \ldots, -S+1, \ldots, S-1, S.$$  

(5)

For two spins, the basis has the form of the direct product $|m_1⟩ ⊗ |m_2⟩$.

We consider the system in equilibrium in a strong magnetic field exceeding spin–spin coupling (4) with density matrix (1). The time-evolution of matrix (2) can be written explicitly as

$$\hat{ρ}(t) = e^{-i\hat{H}_{SS}/\hbar} \hat{ρ}(0)e^{i\hat{H}_{SS}/\hbar} =$$

$$= \frac{1}{Z} \left[ 1 + \frac{β}{2} (\hat{S}_1 e^{-i\tau \hat{S}_2 z} + \hat{S}_1 e^{i\tau \hat{S}_2 z} + \hat{S}_2 e^{-i\tau \hat{S}_1 z} + \hat{S}_2 e^{i\tau \hat{S}_1 z}) \right] \frac{1}{Z} [1 + β\Delta \hat{ρ}(t)],$$  

(6)

where $\hat{S}_j = \hat{S}_{jx} ± i\hat{S}_{jy}$, $τ = tJ/S_2$ is the dimensionless time, $Z = d_1d_2$, and $d_j = 2S_j + 1$.

The correlation between spins is measured by the mutual information [7], [29]

$$I(\hat{ρ}) = S_N(\hat{ρ}_1) + S_N(\hat{ρ}_2) - S_N(\hat{ρ}),$$  

(7)

where $S_N(\hat{ρ}) = -\text{Tr} \{ \hat{ρ} \log_2 \hat{ρ} \}$ is the von Neumann entropy and $\hat{ρ}_1 = \text{Tr}_2 \hat{ρ}$ and $\hat{ρ}_2 = \text{Tr}_1 \hat{ρ}$ are the respective reduced density matrices after the trace of matrix (6) is calculated over the states of the second or the first spin. We calculate the trace and obtain

$$\hat{ρ}_1 = \frac{1 + βS_1 g_2(t)}{d_1}, \quad \hat{ρ}_2 = \frac{1 + βS_2 g_1(t)}{d_2},$$  

(8)

$$g_j(t) = \frac{1}{d_j} \sum_{m=-S_j}^{S_j} e^{±im\tau} = \frac{\sin(d_j \tau/2)}{d_j \sin(\tau/2)},$$  

(9)

We calculate the von Neumann entropy in the lowest order in $β$ [2], [3], [8],

$$S_N(\hat{ρ}) = -\text{Tr}(\hat{ρ} \log_2 \hat{ρ}) \approx \log_2 Z - \frac{β^2}{2Z \log 2} \text{Tr}(\Delta \hat{ρ})^2,$$  

(10)

and obtain

$$I(\hat{ρ}) = S_1(S_1 + 1)b[1 - g_2(t)] + S_2(S_2 + 1)b[1 - g_1(t)],$$  

(11)

where $b = β^2/6\log 2$.

Figure 1 illustrates the time dependence of the results obtained for the mutual information of two spins (11) for different values of the spin quantum number $S_1 = S_2 = S$ and the classical momenta (see formula (A.5) in the appendix). The time is taken in dimensionless units $τS = tJ$. The results are given in $2bS^2$ units (i.e., the ratios $I(\hat{ρ})/2bS^2$ are given). In such units, the curves of the classical mutual information dependence $J_c(P_c)/2bS^2$ given by (A.5) coincide for different $S$. There are no correlations at $t = 0$. We obtain

$$I(\hat{ρ}) \approx \frac{2}{3}bS_1(S_1 + 1)S_2(S_2 + 1)τ^2 = \frac{2}{3}bS_1(S_1 + 1)(1 + \frac{1}{S_2})(Jt)^2,$$  

(12)

$$J_c(P_c) \approx \frac{2}{3}b(S_1)^2(S_2)^2τ^2 = \frac{2}{3}b(S_1)^2(Jt)^2.$$  

(13)
Quantum mutual information (12) exceeds classical mutual information (13) because its squared total momentum as a result of quantum fluctuations is equal to $S_j(S_j + 1)$ rather than to $S_j^2$.

At large times, the classical mutual information $J_c(P_c)$ given by (A.5) tends to its limit value. The quantum mutual information $I(\hat{\rho})$ given by (11) behaves differently: it first enters a plateau and then decreases to zero at $\tau = 2\pi$. The periodic time variation with the period $T = 2\pi S/J$ arises because the energy levels are discrete (such a behavior of simple quantum systems is well known; see, e.g., [11]–[13], [21]). For $t = T$, the difference between the phase increment of different energy levels (6) is an integer multiple of $2\pi$. Hence, $g^2_j(T) = 1$ (see (9)) and $I(\hat{\rho}) = 0$. In the case of classical magnetic momenta, their projections on the magnetic field and their energies vary continuously. Hence, $g_{cJ}(t) \to 0$ as $t \to \infty$ (see (A.4)), and the curve $J_c(P_c)$ given by (A.5) enters a plateau. For $S = 3/2$, the mutual information periodicity can be seen in Fig. 1 ($I(\hat{\rho}) = 0$ for $tJ = 3\pi$). For $S = 9/2$, the mutual information disappears beyond the displayed region for $tJ = 9\pi$. As $S \to \infty$, the period becomes infinite, and the functions $I(\hat{\rho})/2bS^2$ given by (11) and $J_c(P_c)/2bS^2$ given by (A.5) coincide at finite times.

3. Orthogonal measurements

Mutual information (7) is used to measure the total correlations, which are sums of the classical and quantum correlations. The classical correlations can be calculated by the measurement described in [7]. To perform a von Neumann measurement, we must project the state $\hat{\rho}(t)$ on a complete basis of orthogonal wave functions $|\Psi_m\rangle$ using a complete system of projection operators,

$$\hat{\Pi}_m = |\Psi_m\rangle\langle\Psi_m|, \quad \sum_m \hat{\Pi}_m = 1. \quad (14)$$

In the case of a system with $S_1 = 1/2$, the complete set of mutually orthogonal projection operators consists
of two projection operators of general form,

$$\hat{\Pi}_{1\pm} = \frac{1}{2}[1 \pm (n_x \hat{\sigma}_1 + n_y \hat{\sigma}_1 + n_z \hat{\sigma}_1)],$$

where $n_\alpha$ are the direction cosines, $\hat{\sigma}_\alpha$ is the Pauli matrix, and $\alpha = x, y, z$. In the case of generalized POVM measurements, which we consider in the next section, the functions $|\Psi_m\rangle$ in operators (14) can be nonorthogonal, and these operators are then already not projection operators strictly speaking [29].

Density matrix (6) after projection for the first spin becomes

$$\hat{\Pi}_1(\hat{\rho}) = \frac{1}{Z}[1 + \beta \hat{\Pi}_1(\Delta \hat{\rho}(t))],$$

where

$$\hat{\Pi}_1(\Delta \hat{\rho}(t)) = \sum_m (\hat{\Pi}_{1m} \otimes \hat{E}_2) \Delta \hat{\rho}(t)(\hat{\Pi}_{1m} \otimes \hat{E}_2)$$

and $\hat{E}_2$ is the unit matrix. In the lowest order with respect to the inverse temperature, mutual information (7) becomes

$$I(\hat{\Pi}_1(\hat{\rho})) = S_N(\hat{\Pi}_1(\hat{\rho}_1)) + S_N(\hat{\Pi}_1(\hat{\rho}_2)) - S_N(\hat{\Pi}_1(\hat{\rho})) =$$

$$= \frac{\beta^2}{2 \log 2} \left\{ \frac{1}{Z} \text{Tr}(\hat{\Pi}_1(\Delta \hat{\rho}))^2 - \frac{1}{d_1} \text{Tr}_1(\hat{\Pi}_1(\Delta \hat{\rho}_1))^2 - \frac{1}{d_2} \text{Tr}_2(\Delta \hat{\rho}_2)^2 \right\}. \quad (17)$$

The value of this measure of the classical correlation depends on the choice of basis (14). It was proposed [7] to consider all bases and to take the maximum value of correlation (17) as the universal measure. Such a program can be implemented only in several simple cases, for example, in the case of a two-level system, which we consider below.

If we subtract classical part (17) from all correlations (7), then we obtain the quantum part of the correlations

$$Q = I(\hat{\rho}) - I(\hat{\Pi}_1(\hat{\rho})) = \frac{\beta^2}{2 \log 2} \left\{ \frac{1}{Z} \text{Tr}(\Delta \hat{\rho})^2 - \frac{1}{Z} \text{Tr}(\hat{\Pi}_1(\Delta \hat{\rho}))^2 - \frac{1}{d_1} \text{Tr}_1(\Delta \hat{\rho}_1)^2 + \frac{1}{d_1} \text{Tr}_1(\hat{\Pi}_1(\Delta \hat{\rho}_1))^2 \right\}. \quad (18)$$

We minimize this quantity over the measurement bases and obtain the entropy measure of quantum correlations, i.e., the quantum discord [7]. Measure (18) without optimization was called a measurement-dependent discord [7].

To simplify the calculations, it was proposed to replace the above entropy measure with a geometric measure of quantum correlations, i.e., the geometric discord [7], for which in the high-temperature approximation we obtain

$$D_G = \min_{\Pi_1} \text{Tr}(\hat{\rho} - \hat{\Pi}_1(\hat{\rho}))^2 = \frac{\beta^2}{Z^2} \min_{\Pi_1} \{\text{Tr}(\Delta \hat{\rho})^2 - \text{Tr}(\hat{\Pi}_1(\Delta \hat{\rho}))^2\}. \quad (19)$$

This relation is based on the property $\text{Tr}(\hat{\Pi}(\Delta \hat{\rho}))^2 = \text{Tr}(\hat{\Pi}(\Delta \hat{\rho})\Delta \hat{\rho})$. Although this measure has certain drawbacks noted in [28], [32], [33], it is currently often used to estimate quantum correlations [3], [19]–[28]. It was proposed in [28] to improve this measure by changing it to

$$\bar{D}_G = \frac{D_G}{\text{Tr}(\hat{\rho})^2}. \quad (20)$$
In the high-temperature approximation, this transformation is related to multiplying (19) by \( Z \).

We calculate these measures for two-spin system (4) in state (6) for \( S_1 = 1/2 \) and \( S_2 \geq 1/2 \). We apply projection operators (15), perform several calculations, and obtain

\[
\frac{\text{Tr}(\hat{\Pi}_1(\Delta \hat{\rho}))}{Z} = \frac{S_2(S_2 + 1)}{3} \left( \cos^2 \frac{\tau}{2} + n_z^2 \sin^2 \frac{\tau}{2} \right) + \frac{n_x^2}{8} (1 + g_2(2t)) + \frac{n_y^2}{8} (1 - g_2(2t)),
\]

\[
\frac{\text{I}(\hat{\Pi}_1(\hat{\rho}))}{3b} = \frac{S_2(S_2 + 1)}{3} n_z^2 \sin^2 \frac{\tau}{2} + \frac{n_x^2}{8} (1 + g_2(2t)) + \frac{n_y^2}{8} (1 - g_2(2t)) - \frac{n_z^2}{4} g_2^2(t).
\]

The maximum value of quantity (22) is attained for the direction cosines \( n_x = n_y = 0, n_z = 1 \) at any times. This implies the respective formulas for the classical correlations and the quantum discords

\[
C_N = \max_{\hat{\Pi}_1} I(\hat{\Pi}_1(\hat{\rho})) = S_2(S_2 + 1)b \sin^2 \frac{\tau}{2}, \quad D_N = I(\hat{\rho}) - C_N = \frac{3}{4} b[1 - g_2^2(t)].
\]

Quantity (21) attains its maximum for \( n_x = 1 \) at small times \( \tau < \tau_c \) and for \( n_z = 1 \) at large times \( \tau > \tau_c \). Here, \( \tau_c \) is the solution of the equation

\[
\frac{S_2(S_2 + 1)}{3} \sin^2 \frac{\tau}{2} = \frac{1 + g_2(2t)}{8}.
\]

We obtain \( \tau_c = \pi/2 \) for \( d_2 = 2 \), \( \tau_c = 0.574 \) for \( d_2 = 3 \), and the estimate \( \tau_c \approx \sqrt{3}/\sqrt{2S_2(S_2 + 1)} \approx \sqrt{6}/d_2 \) for \( d_2 \gg 1 \). Finally, the geometric discord has the form

\[
D_G = \begin{cases} \beta^2 \left[ \frac{S_2(S_2 + 1)}{3} \sin^2 \frac{\tau}{2} + \frac{1 - g_2(2t)}{8} \right], & \tau < \tau_c, \\ \frac{\beta^2}{4Z}, & \tau > \tau_c. \end{cases}
\]

The time behavior of the two measures is similar. The correlations increase and enter a plateau. The differences are in their approach to the plateau and in the plateau height. This height is independent of \( d_2 \) in the case of the entropy measure, while it decreases as \( 1/d_2 \) from \( \hat{\rho}^2 \) in definition (19) compared with \( \hat{\rho} \log_2 \hat{\rho} \) in definition (18) in the case of the geometric measure because of the additional factor \( 1/Z \). Such differences were noted in [3], [28] for the qubit \( (d_1 = 2) \) related to a multiqubit system \( (d_2 = 2^n) \) by the scheme of deterministic quantum calculation with one qubit (DQC1). They can be removed by transformation (20).

In the case of a two-spin system with arbitrary spins, we project state (6) on eigenstates (5) of the operator \( S_{1z} \) with the projection operators

\[
\hat{\Pi}_{S_{1m}} = \vert m \rangle \langle m \vert.
\]

For the classical correlations and the measurement-dependent discord, we then obtain the respective expressions

\[
C_N^{(S_1)} = I(\hat{\Pi}_{S_1}(\hat{\rho})) = S_2(S_2 + 1)b[1 - g_1^2(t)], \quad Q_N^{(S_1)} = S_1(S_1 + 1)b[1 - g_2^2(t)].
\]

4. Nonorthogonal measurements

According to result (26) obtained in the preceding section, we have \( C_N^{(S_1)} = Q_N^{(S_1)} \) at small times (this relation holds at all times for \( S_1 = S_2 \)). Therefore, for an orthogonal measurement with a chosen basis, we show that the quantum correlations can be preserved for large spins \( S_2 \to \infty \). The fact that the quantum
properties are preserved in this limit, which manifests itself in a violation of the Bell inequality, was noted for a singlet state of two spins in [34], [35]. For systems in other states, the transition from quantum spin to the classical momentum as $S \to \infty$ is expected [11], [31], [36]. We specify to which case our state (6) belongs. For this, we change the measurement basis.

It is assumed that the SCS (Bloch states) are closest to the state of classical momenta [30]:

$$|\theta, \varphi \rangle = R(\theta, \varphi)|S\rangle = \sum_{m=-S}^{S} \left( \frac{2S}{S+m} \right)^{1/2} \left( \cos \frac{\theta}{2} \right)^{S+m} \left( e^{i\varphi} \sin \frac{\theta}{2} \right)^{S-m} |m\rangle,$$

(27)

where $\theta$ and $\varphi$ are the polar and azimuthal angles on the unit sphere (Bloch sphere). These states are obtained from the ground state $|S\rangle$ by the rotation operator $R(\theta, \varphi)$ and are a superposition of states (5) with different projections $m$. In state (27), the average spin projections

$$\langle \theta, \varphi | \hat{S}_z \rangle = S \cos \theta, \quad \langle \theta, \varphi | \hat{S}_z \rangle = S \sin \theta \cos \varphi,$$

(28)

are the same as in the case of classical momentum (A.1) considered in the appendix. The completeness condition

$$\frac{2S+1}{4\pi} \int \langle \theta, \varphi \rangle \langle \theta, \varphi | \sin \theta d\theta d\varphi = 1$$

is satisfied for the SCS basis, but this basis is not orthogonal:

$$|\langle \theta, \varphi | \theta', \varphi' \rangle|^2 = \cos^{4S} \frac{\Theta}{2}, \quad \cos \Theta = \cos \theta \cos \theta' + \sin \theta \sin \theta' \cos(\varphi - \varphi').$$

We take the SCS system as the measurement basis in (14), perform the POVM measurement, which reduces to multiplying by the SCS and calculating the trace, and obtain the classical density function for the probability distribution of the angle values

$$(4\pi)^2 P_{12}(\theta_1, \varphi_1, \theta_2, \varphi_2; t) = d_1 d_2 \text{Tr}[|\theta_1, \varphi_1 \rangle \langle \theta_1, \varphi_1 | \otimes |\theta_2, \varphi_2 \rangle \langle \theta_2, \varphi_2 | \hat{\rho}(t)] =$$

$$= d_1 d_2 \langle \theta_1, \varphi_1, \theta_2, \varphi_2 | \hat{\rho}(t) |\theta_1, \varphi_1, \theta_2, \varphi_2 \rangle.$$

(29)

We use the SCS properties

$$\langle \theta_j, \varphi_j | \hat{S}_{j\pm} | \theta_j, \varphi_j \rangle = S_{j\pm} = S_j \sin \theta_j e^{\pm i\varphi_j},$$

$$\langle \theta_j, \varphi_j | e^{\pm i\tau \hat{S}_j} | \theta_j, \varphi_j \rangle = \left( \cos \frac{\tau}{2} \mp i \cos \theta_j \sin \frac{\tau}{2} \right)^{2S_j} \equiv \xi_{j\pm}(t)$$

to obtain function (29):

$$P_{12}(\theta_1, \varphi_1, \theta_2, \varphi_2; t) = \frac{1}{(4\pi)^2} \left\{ 1 + \frac{\beta}{2} \left[ S_{1+} \xi_{2+}(t) + S_{1-} \xi_{2-}(t) + S_{2+} \xi_{1+}(t) + S_{2-} \xi_{1-}(t) \right] \right\}.$$

(30)

The further calculations repeat the calculations for the classical momenta in the appendix. Integrating over the angles, we obtain the reduced distributions

$$P_1(\theta_1, \varphi_1; t) = \frac{1}{4\pi} \{ 1 + \beta S_{1g_2}(t) \}, \quad P_2(\theta_2, \varphi_2; t) = \frac{1}{4\pi} \{ 1 + \beta S_{2g_1}(t) \}$$

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and mutual information (A.3) in the high-temperature approximation

\[ J_{BB}(P_{12}) = (S_1)^2 b[f_2(t) - g_2^2(t)] + (S_2)^2 b[f_1(t) - g_1^2(t)], \]  

(31)

where

\[ f_j(t) = \frac{1}{2} \int_{-1}^{+1} \xi_j(t) \xi_j^{(t)} d \cos \theta_j = \sum_{n=0}^{2S_j} \binom{2S_j}{n} \left( \cos \frac{\tau}{2} \right)^{4S_j - 2n} \left( \sin \frac{\tau}{2} \right)^{2n} \frac{1}{2n + 1} = \]

(32)

The time-evolution of the mutual information \( J_{BB}(P_{12}) \) given by (31), which is obtained by POVM measurements with the SCS basis, is shown in Fig. 1. At small times \( \tau = tJ/S \ll 1 \), we obtain \( J_{BB}(P_{12}) \approx J_c(P_c) \), where \( J_c(P_c) \) is determined by formula (13). At large times, the curve of the dependence \( J_{BB}(P_{12}) \) walks away downwards. This effect arises because of the packet spreading from states with different projections \( S_1, S_2 \) to the SCS (with different values of the phase factor \( e^{i\pi S_1} \)). Nevertheless, the packets are gathered together at \( t = T \) because time dependence (6) is periodic.

If we now subtract the obtained classical part of correlations (31) from the total correlation \( I(\hat{\rho}) \) given by (11), then the difference is the quantum part of the correlations,

\[ Q_{BB} = I(\hat{\rho}) - J_{BB}(P_{12}) = \]

(33)

\[ = (S_1)^2 b[1 - f_2(t)] + (S_2)^2 b[1 - f_1(t)] + S_1 b[1 - g_2^2(t)] + S_2 b[1 - g_1^2(t)]. \]

In the case under study, the measurements were performed symmetrically for both spins. Another measure of classical correlations can be obtained by performing the POVM measurement only for one of the spins, for example, for the second spin. In this case, instead of (31), we obtain

\[ J_{B2} = S_1(S_1 + 1)b[f_2(t) - g_2^2(t)] + (S_2)^2 b[1 - g_1^2(t)], \]

(34)

which implies the quantum part

\[ Q_{B2} = I(\hat{\rho}) - J_{B2} = S_1(S_1 + 1)b[1 - f_2(t)] + S_2 b[1 - g_1^2(t)]. \]

(35)

We note that a measure called the Gaussian quantum discord was previously introduced in systems with a continuous spectrum obtainable by the POVM measurement with a basis of field (boson) coherent states [37], [38].

5. Discussion

The time dependence of the fractions of quantum correlations obtained by different measurement methods is shown in Fig. 2 as the ratios \( Q_{BB}/I(\hat{\rho}) \), \( Q_{B2}/I(\hat{\rho}) \), \( Q_{B2}^{(S_1)}/I(\hat{\rho}) \), and \( D_{BB}/I(\hat{\rho}) \) for different values of \( S_1 \) and \( S_2 \). At small times \( \tau = tJ/S_2 \ll 1 \), we use formulas (33), (35), and (12) to obtain the results of POVM measurements with the SCS basis

\[ \frac{Q_{BB}}{I(\hat{\rho})} \approx \frac{S_1 + S_2 + 1}{(S_1 + 1)(S_2 + 1)}, \quad \frac{Q_{B2}}{I(\hat{\rho})} \approx \frac{1}{S_2 + 1}. \]

(36)
Fig. 2. Time evolution of the quantum part of the correlations for different spin pairs indicated by the numbers on the curves: (1) $S_1 = S_2 = 3/2$, (2) $S_1 = S_2 = 9/2$, (3) $S_1 = 1/2$ and $S_2 = 9/2$, and (4) $S_1 = 9/2$ and $S_2 = 1/2$. The results of orthogonal measurements for the first spin, $D_N/I(\tilde{\rho})$ and $Q_N^{(S_1)}/I(\tilde{\rho})$, are shown by solid lines. The results of nonorthogonal measurements for measurements for both spins, $Q_{BB}/I(\tilde{\rho})$, are shown by dotted lines and for measurements for the second spin only, $Q_{B2}/I(\tilde{\rho})$, are shown by dashed lines.

which is consistent with the data illustrated in Fig. 2. It follows from results (23) and (26) obtained after projection on orthogonal basis (25) that the classical and quantum contributions coincide at small times: $C_N(S_1) = Q_N^{(S_1)} = I(\tilde{\rho})/2$. For $S_1 = S_2$, this relation remains the same at all times. The coincidence of the contributions was previously proved in the special case $S_1 = S_2 = 1/2$ in [13].

To reveal the causes of the differences between the two measuring methods, we consider density matrix (6) at small times,

$$\rho(t) \sim \frac{1}{Z} \left[ 1 + \beta (\tilde{S}_{1x} + \tilde{S}_{2x}) + \frac{\beta J}{S_2} (\tilde{S}_{1y}\tilde{S}_{2x} + \tilde{S}_{2y}\tilde{S}_{1x}) \right]. \quad (37)$$

After orthogonal measurement (16) with projection operators (25), the contribution of the term $\tilde{S}_{1y}\tilde{S}_{2x}$ is zero, and the classical part of the correlations remaining after the measurement is hence equal to half the total correlations for any spin value. After the POVM measurement of state (37) for the second (large) spin, both terms contribute to the classical part of correlations (34) (or to (31) after POVM measurements of state (37) for both spins). Only the quantum fluctuations are lost in calculations of the average squared operator of the spin component, i.e., we have $S_j^2/3$ instead of $S_j(S_j + 1)/3$. As $S_j \to \infty$, these differences, which arise because of fluctuations, disappear. We can thus see that the fraction of classical correlations increases as the spin increases and the fraction of quantum correlations respectively decreases and tends to zero in the limit as $S_j \to \infty$.

The conclusion that the maximum contribution of the classical correlations can be obtained by POVM measurements is not new and is consistent with the general theory [7]. But in the case $S_1 = S_2 = 1/2$, which is considered more frequently than the others, the passage from von Neumann orthogonal measurements to POVM measurements gives a refinement that is insignificant in value [7]. The difference between the results for the classical mutual information obtained by the two measurement methods is greater in the system of a qubit and a qutrit [39]. We obtained a similar result for the system of two large spins in the state with
density matrix (37). At least for this state, the estimation methods based on orthogonal measurements can distort the actual relation between the quantum and classical correlations.

We now analyze the evolution of quantum correlations at large times. At $t = T$, the correlations disappear in the quantum system because $I(\hat{\rho}) = 0$. For $t \approx T/2$, the correlation is maximum, as can be seen in the graphs of mutual information in Fig. 1. Moreover, the fractions of quantum and classical correlations depend on the measurement method (see Fig. 2). If $S_1 \neq S_2$, then results (23) and (26) obtained after projection on the orthogonal basis (25) give a periodic time dependence of the fractions of classical and quantum correlations. These fractions are equal to each other at small times but attain different values for $\sin^2(\tau/2) = 1$:

$$
\frac{C_N^{(31)}}{I(\hat{\rho})} \approx \frac{S_2(S_2 + 1)}{S_1(S_1 + 1) + S_2(S_2 + 1)}, \quad \frac{Q_N^{(31)}}{I(\hat{\rho})} \approx \frac{S_1(S_1 + 1)}{S_1(S_1 + 1) + S_2(S_2 + 1)}.
$$

Such dependences are shown in Fig. 2 for $S_1 = 1/2$, $S_2 = 9/2$ and $S_1 = 9/2$, $S_2 = 1/2$. According to (11), (23), and (26), the ratio $Q_N^{(31)}/I(\hat{\rho})$ either decreases (for $S_1 < S_2$) or increases (for $S_1 > S_2$) with time (in the latter case, we replace $S_2$ with $S_1$ as the parameters $J$ and $\tau$ are scaled). On the other hand, after POVM measurements, relations (31) and (34) with $\sin^2(\tau/2) = 1$ (only one term $f_j(t) = 1/(4S_1 + 1)$ remains in series (32)) imply

$$
\frac{J_{B2}}{I(\hat{\rho})} \approx \frac{S_1(S_1 + 1)/(4S_2 + 1) + S_2^2}{S_1(S_1 + 1) + S_2(S_2 + 1)}, \quad \frac{J_{BB}}{I(\hat{\rho})} \approx \frac{S_1^2/(4S_2 + 1) + S_2^2/(4S_1 + 1)}{S_1(S_1 + 1) + S_2(S_2 + 1)}.
$$

From (33) and (35), we further derive the estimates for $S_1 = S_2 = S \gg 1$:

$$
\frac{Q_{BB}}{I(\hat{\rho})} \approx 1 - \frac{1}{4S} \quad \text{and} \quad \frac{Q_{B2}}{I(\hat{\rho})} \approx \frac{1}{2} \left( 1 + \frac{3}{4S} \right).
$$

Based on Fig. 2 and the above formulas, we conclude that for $t \sim T/2$, the orthogonal measurement gives a value of the fraction of quantum correlations less than that obtained by the POVM measurement with the SCS basis, i.e., the obtained relation between the results is opposite to that in the case of small times. To explain this effect, we write the density matrix for $t = T/2$ ($\tau = \pi$):

$$
\hat{\rho} \left( \frac{T}{2} \right) = \frac{1}{Z} \left\{ 1 + \frac{\beta}{2} [\hat{S}_{1z}(-1)^{m_2} + \hat{S}_{2z}(-1)^{m_1}] + \hat{S}_{1z}(-1)^{m_2} + \hat{S}_{2z}(-1)^{m_1} \right\}.
$$

If the value of spin projections varies by one, then the function $e^{i\pi S_{1z}}$ changes its sign. The corresponding terms in (41) are $\beta[\hat{S}_{1z}(-1)^{m_2} + \hat{S}_{2z}(-1)^{m_1}]$ for integer-valued spins and $\beta[\hat{S}_{1z}(-1)^{m_2-1/2} + \hat{S}_{2z}(-1)^{m_1-1/2}]$ for half-integer spins. In the case of orthogonal measurements with projection operators (25) for one of the spins (the first), the contribution is given by one term. In contrast to the case of small times, the value of the contribution of this term is variable in this case. For example, for $S_1 < S_2$, we only have the “small field of the small spin rotates the large spin” contribution, while the “large spin rotates the small spin” contribution is lost. We obtain expressions (38) for the fractions of classical and quantum correlations. The result of projection (29) of state (41) on the SCS in the POVM measurement is more complicated: SCS (27) is formed by the sum of states (5) with different spin projections on the $z$ axis, which in (29) and (30) give a sign-alternating series with the sums

$$
\xi_{j\pm} \left( \frac{T}{2} \right) = [\cos \theta_j]^2 S_j.
$$
As a result, after POVM measurements, we obtain results (39) that are less in value than result (38) obtained by orthogonal measurements.

We performed POVM measurements with the SCS basis for both one and two spins. The fraction of quantum correlations is less in the first case. On the other hand, in the orthogonal measurements of state (37) for one or two spins, we obtain the same values of the classical and hence of the quantum correlations. Indeed, if after the orthogonal measurement for the first spin, we perform the orthogonal projection of the second spin on the eigenstates of the operator $\hat{S}_{2y}$, then there are no additional losses of the correlations. Such an observation about the coincidence of the results of measurements for one or two qubits was made in [39]. How to measure is the question, and no final answer to this question has yet been obtained [7], [39], [40]. For instance, Luo [40] believes that the fraction of the semiclassical correlation that includes a part of the quantum correlations is obtained when one spin is measured.

6. Conclusion

Correlations (3) in our model, observed by the NMR method, have form (37) at small times. For two spins $S_1 = S_2 = 1/2$, these correlations are equally divided into the quantum and classical parts, according to the results of orthogonal measurements. As the spin increases, the fraction of classical correlations increases, and the fraction of quantum correlations correspondingly decreases and tends to zero in the limit as $S \to \infty$. To obtain such a result, we passed from orthogonal measurements to POVM measurements with the SCS basis. As time increases, the linear time dependence changed into a dependence in the form of the sum of exponentials with different periods. After the time period $T = 2\pi S/J$ equal to the greatest common divisor of these periods for $S_1 = S_2 = S$, the correlations again become zero. At times close to half the period, we observe the greatest degree of correlation between the two spins, which is characterized by the maximum of mutual information. In this region, the relative fraction of quantum correlations increases compared with the region of small times. Because the phase factors of neighboring states with projections that differ by one have different values at such times, the POVM measurement with the SCS basis becomes too rough as a measuring tool. The point is that in the SCS, the spread of projections $m$ is of the order of $\sqrt{S} \gg 1$ [31]. The orthogonal measurements in the case of projection on separate states with certain values of $m$ permit obtaining a greater amount of classical correlations. Nevertheless, the quantum correlations form a noticeable part equal to half all the correlations for $S_1 = S_2$. For $S_1 < S_2$, their fraction decreases by a factor of $(S_1/S_2)^2$ if the measurements are performed for the first spin.

Appendix: Mutual information of two classical momenta

We consider a system of two classical magnetic momenta. We describe their states by polar and azimuthal angles $\theta$ and $\varphi$ on the unit sphere (Bloch sphere) in terms of which the projections of the classical momentum $\vec{S}$ on the coordinate axes can be expressed as

$$S_z = S \cos \theta, \quad S_y = S \sin \theta \sin \varphi, \quad S_x = S \sin \theta \cos \varphi. \quad (A.1)$$

Instead of the density matrix $\hat{\rho}(t)$ given by (6), the states of the ensemble are now described by the density function of the probability distribution of the angle values on the Bloch sphere:

$$P_c(\theta_1, \varphi_1, \theta_2, \varphi_2; t) = \frac{1}{(4\pi)^2} \left\{ 1 + \frac{\beta}{2} [S_{1+}e^{-i\tau S_{2z}} + S_{1-}e^{i\tau S_{2z}} + S_{2+}e^{-i\tau S_{1z}} + S_{2-}e^{i\tau S_{1z}}] \right\}, \quad (A.2)$$

where

$$S_{jz} = S_j \cos \theta_j, \quad S_{j\pm} = S_j \sin \theta_j e^{\pm i\varphi_j}.$$
The classical mutual information is

\[ J_c(P_c) = S_{Sh}(P_{c1}) + S_{Sh}(P_{c2}) - S_{Sh}(P_c), \]  

(A.3)

where

\[ S_{Sh}(P_c) = -\int\int P_c(\theta_1, \varphi_1, \theta_2, \varphi_2; t) \log_2 P_c(\theta_1, \varphi_1, \theta_2, \varphi_2; t) \sin \theta_2 d\theta_2 d\varphi_2 \sin \theta_1 d\theta_1 d\varphi_1 \]

is the Shannon entropy. Instead of taking the trace, we must calculate the integral over the Bloch sphere. The reduced distribution density for the first momentum (the calculations for the second momentum are similar) is given by

\[ P_{c1}(\theta_1, \varphi_1; t) = \int P_c(\theta_1, \varphi_1, \theta_2, \varphi_2; t) \sin \theta_2 d\theta_2 d\varphi_2 = \frac{1}{4\pi} \{ 1 + \beta S_{1x} g_{c2}(t) \}, \]

(A.4)

\[ g_{c_j}(t) = \frac{1}{2} \int_{-1}^{+1} e^{\pm i\tau S_j \cos \theta_j} d\cos \theta_j = \frac{\sin \tau S_j}{\tau S_j}. \]

We obtain the expression for classical mutual information (A.3) in the high-temperature approximation:

\[ J_c(P_c) = (S_1)^2 b[1 - g_{c2}(t)] + (S_2)^2 b[1 - g_{c1}(t)]. \]

(A.5)

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