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Control and manipulation of quantum spin switching and spin correlations in [Tb_2] molecular magnet under a pulse magnetic field

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ABSTRACT

A general study of [Tb_2] molecular magnet is presented using the general spin Hamiltonian formalism. A spin-spin correlators determined for a spin wave functions in [Tb_2] are analyzed numerically and compared in details with the results obtained by means of conventional quantum mechanics. It is shown that the various expectation values of the spin operators and a study of their corresponding probability distributions allow to have a novel understanding in spin dynamics of entangled qubits in quantum [Tb_2] system. The obtained results reveal that the properties of spin-spin correlators are responsible for the entanglement of the spin qubit under a pulse magnetic field. It allows us to present some quantum circuits determined for quantum computing within SSNQ based on [Tb_2] molecule, including the **CNOT** and **SWAP** gates.

1. Introduction

The problem related with the switching rate of the atomic spins in both the modern magnetic logic and magnetic storage devices under the terahertz regime is one of the most challenging task for information processing nowadays [1,2]. The contemporary physics, applying to solve this problem, faces with serious limitations arising during technological transfer. Therefore, we have to invent new paradigms based on the quantum spin dynamics in the picosecond regime. This challenge can be met by a simulation of the quantum spin switching in a picosecond pulsed magnetic field [3,4]. Up to now, it is not a trivial task to find an accessible methods, which are able to generate the intense sub-picosecond magnetic pulses localized at the atomic scale limit. However, it was shown recently [5], that bimetallic nanorings can be treated as a potential nanoscale sources of the intense ultrashort (few tenths of Tesla) magnetic pulses. Therefore, the ability to generate strong magnetic fields localized at nanoscale is of a great interest nowadays, since it allows us to elucidate the magnetization and spin dynamics at sub-picosecond time and nanometer length scales [6]. For a **CNOT** quantum gates are using non-symmetric ligands. Ligand H_3L4 exhibits a collection of donor groups disposed in a way that could favor the aggregation of two metals in different coordination environments. Indeed, single lanthanide ions are good candidates for encoding quantum information, since they very often exhibit, as a result of their strong zero field splitting (ZFS), a very well isolated ground state doublet, which represents an effective S = 1/2, thus providing for good realizations of qubits. In addition, their spin states exhibit long decoherence (with measured relaxation times, T2, of up to 7 μs), adding a very important quality to their functionality. The reaction of H_3L4 with LnX3 (Ln^{III} = a lanthanide; X^- = Cl^- or NO_3^-) in pyridine forms dinuclear complexes with a general formula [$Ln_2X(HL4)_2(H_2L4)(py)(S)$] (S = py or H_2O) for practically the entire 4f series. A very weak coupling between both qubits is necessary for the realization of a **CNOT** quantum gate, together with a strong anisotropy of both spin carriers. The case of terbium is the most relevant to quantum computer; the high temperature $\chi_M T$ values are consistent with two uncoupled Tb^{III} ions ($^7F_6$, S = 3, L = 3, J = 6, g = 3/2) whereas the decline observed upon cooling is a consequence of the depopulation of the various m_J states, down to the Ising limit where only the two orientations $m_J = \pm 6$ of the J = 6 state of each metal are populated. These orientations correspond to the two states embodying each qubit[7]. Therefore, in present paper we explore

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the effect of a magnetic pulse with a high intensity and $T_{pulse} \gg T_{THz}$ on the switching of spin states in the magnetic molecule $[Tb_2Cl(HL4)_2(H_2L4)(py)_2]$ (hereafter referred to as $[Tb_2]$). We apply the rectangular magnetic pulse, since, as it was shown already, such a pulse leads to the switching of spin states in $[Tb_2]$ molecule [8]. The magnetic molecules (so-called molecular magnets) are the most intensively studied, so far theoretically and experimentally, objects during last decade due to their potential technological applications and a nice opportunity to investigate the fundamental properties of matter at the atomic scale limit [9]. These objects demonstrate at low temperature a slow relaxation of magnetization and give rise to magnetic hysteresis, that allows to consider a single magnetic molecule (SMM) as the smallest practical magnetic unit for memory storage or as a spin qubit for quantum computing [10,7]. Therefore, the relevance of $[Tb_2]$ molecular magnet under a magnetic field to be considered for such perspectives will be discussed in this paper. For this purpose the time-dependent spin dynamics of $[Tb_2]$ is involved within a micromagnetic model based on the general spin Hamiltonian formalism, where the spin is considered as a spatial- and time-dependent continuous function [11]. In this case the dynamics of spins obeys the Landau-Lifshitz-Gilbert (LLG) equation, involving the various energy contributions from the exchange, magnetostatic and Zeeman interactions, and magnetic anisotropy of the spin system [12,13]. It is known, that correlations are of a great importance in the study of spin systems [14]. They are directly related to the entanglement between different atomic spins, which can be employed in the field of quantum information processing [15]. Therefore, entanglement arising in the correlated quantum spin systems has become one of the most widely investigated phenomenon in quantum physics during last years [16]. Now it is well known how to create entanglement in quantum spin systems, but how it propagates inside a system is still an important fundamental question in quantum information theory. Therefore, the theoretical descriptions of the time-dependent properties of the correlated quantum spin systems are very important now days [17–19]. Recently it was claimed, that the controlled and manipulated entanglement in the quantum spin systems could be realized precisely and effectively by means of the required dynamical operations in the presence of the magnetic pulse [15,18]. Moreover, it was shown also [4] that the solid state spin-based system with a definite spin structure can be considered as a spin system of *n*-qubits (SSNQ), which demonstrates the prolonged time of the decogerence demanded for quantum information processing [20,21]. For implementation of quantum computation one can treat a quantum spin system as a SSNQ, where the couplings between the spin qubits can be controlled externally, for instance by the applied magnetic pulse or temperature. Therefore, the systematic studies of the inherent relationship between the strength of the entanglement and the peculiarities of spin structure of a SSNQ are carried out in order to find the optimal time-dependent spin structures with specific types of the controlled and engineered entanglements [17,22,23]. However, the successful implementation of quantum computation demands the physical realization not only a SSNQ, but quantum gates also [16]. It was shown recently, that molecules containing two well defined spin qubits can provide attractive prototypes for the universal controlled-NOT (**CNOT**) and **SWAP** quantum gates [10,7]. In order to realize **CNOT** both spin qubits must exhibit a mutual interaction much weaker than the energy difference between spin states within each qubit in the presence of an external magnetic field, that can be reached with a strong axially anisotropic spins. Moreover, the control and target qubits must be different, thereby they can be addressed specifically. A **SWAP** gate requires that the spins of both qubits exhibit a weak antiferromagnetic Heisenberg coupling. In this case, a system can be modified at will during a period of time, that lets the system evolves quantum mechanically and undergoes the transformation dictated by this operation [7]. In order to carry out these two operations it is necessary that spin qubits of the quantum gates have to exhibit the entangled states [24].

2. Theoretical aspects

To study the properties of a molecular magnet, we apply in the present work the theoretical approach based on the irreducible tensor operator (ITO) technique [25]. In the framework of this approach a molecular magnet with an arbitrary topology and an arbitrary number of the magnetic sites (or states), *N*, can be ascribed by the local spins $S_1, S_2, \ldots, S_N$, which can have the different values in general. In this case, one can use the following successive scheme to present a spin coupling in the system:

$$S_1 + S_2 = S^{(2)}, S^{(2)} + S_3 = S^{(3)}, \ldots, S^{(N-1)} + S_N = S, \quad (1)$$

where *S* represents the complete set of the intermediate spin quantum numbers $S^{(k)}$ with $k = 1,2,\ldots, N-1$. The eigenstates $|SM\rangle_i$ of the general spin Hamiltonian (GSH) $\widehat{H}_{spin}$ of the system are given by the linear combinations of the basis states $|S^{(\mu)}M^{(\mu)}\rangle$:

$$|SM\rangle_i = \sum_{\mu=1}^{N} c_{i\mu} |S^{(\mu)}M^{(\mu)}\rangle, \quad (2)$$

where $M = -S, \ldots, S$ and the coefficients $c_{i\mu}$ can be evaluated by diagonalization of the $\widehat{H}_{spin}$. The main physical interactions contributing to this spin Hamiltonian were considered in previous studies [26,27]. Based on these works we can study the spin dynamics of the system using the LLG equation derived from the quantum theory with the GSH. For instance, to study the spin structure of $[Tb_2]$ molecule we apply the GSH of the following form:

$$\widehat{H}_{spin} = \widehat{H}_{ex} + \widehat{H}_{an} + \widehat{H}_{ZEE} + \widehat{H}_{pulse}(t), \quad (3)$$

where $\hat{H}_{ex}$ represents the isotropic exchange interaction in the Heisenberg-Dirac Hamiltonian, H_{an} represents the anisotropic exchange interaction due to the axial single-ion anisotropy, H_{ZEE} represents the interaction between the spin system and the external magnetic field, and $\widehat{H}_{pulse}(t)$ is an external magnetic pulse. Each term of the $\widehat{H}_{spin}$ can be presented as a combination of the irreducible tensor operators [25,26]. For instance, all isotropic terms of the GSH are described by the rank-0 tensor operators which have the non zero matrix elements only for the eigenstates with the same total spin quantum number *S* (ΔS, $\Delta M = 0$). In this case, the representative matrices can be decomposed into the blocks depending only on the value of *S* and *M*. While all anisotropic terms of the GSH are described by the rank-2 tensor operators related to the non zero matrix elements between eigenstates with $\Delta S = 0, \pm 1, \pm 2$. Their matrices cannot be decomposed into the blocks depending only on the total spin quantum number *S* due to the *S*-mixing between spin states with different *S*. We apply the ITO's technique to the GSH within the MAGPACK code, which has been developed to study efficiently the magnetic properties of a various nanoscale magnets [25].

To study the magnetic properties of $[Tb_2]$ molecule, we'll treat each Tb atom as a single atomic spin interacting with its own surrounding. Such a treatment is a suitable approximation [7]. Besides that, we can also restrict ourselves considering the GSH in the framework of the mean field theory, as a simple but very useful approach [28]. In this case such a "mean field" spin Hamiltonian, which describes the interaction of the spin $\widehat{\mathbf{S}}$ with the external magnetic field, is determined by its flux $\mathbf{H}_{eff}$ and can be written as:

$$\widehat{H}_{spin} = -\gamma \mathbf{H}_{eff}\widehat{\mathbf{S}}, \tag{4}$$

where γ is the gyromagnetic ratio coming from the relation between magnetic moment and spin. The effective magnetic field $\mathbf{H}_{eff}$ describes the external magnetic field (H_z directed along arbitrary Z axis), the anisotropy ($\mathbf{H}_{an}$) and exchange ($\mathbf{H}_{ex}$) fields, and the external magnetic pulse ($\mathbf{H}_{pulse}(t)$) field. The applied "mean field" [29] approximation leads to $\mathbf{H}_{eff} \Rightarrow \mathbf{H}_{eff}^{mean}$ with replacement $\widehat{\mathbf{S}} \Rightarrow \mathbf{M} = \gamma\langle\widehat{\mathbf{S}}\rangle$. It takes into account an average effect of the neighbors but neglects the correlation effects between the spin and its neighbors. Since each Tb atom is considered as a single atomic spin, such approach is quite suitable for [Tb_2] molecule. Thereon, using the Eq. (4), we obtain the following equation for the dynamics of spin [13]:

$$\frac{\partial\langle\widehat{\mathbf{S}}\rangle}{\partial t} = \frac{1}{1+\lambda^2}\langle\widehat{\mathbf{S}}\rangle \times \mathbf{H}_{eff}^{mean} - \frac{\lambda}{1+\lambda^2}\langle\widehat{\mathbf{S}}\rangle \times (\langle\widehat{\mathbf{S}}\rangle \times \mathbf{H}_{eff}^{mean}), \tag{5}$$

where λ is the damping constant. The effective magnetic field $\mathbf{H}_{eff}^{mean}$ can be found as a variation of the free magnetic energy with magnetization $\mathbf{M}$:

$$\mathbf{H}_{eff}^{mean}(\mathbf{M}, t) = -\frac{\delta F}{\delta \mathbf{M}},$$

where F is the free energy of the magnetic system:

$$F = -Nk_BT \ln Z(H_z)$$

with the partition function

$$Z(H_z) = \sum_{M,\mu} \exp[-\epsilon_\mu(M)/k_BT] \sum_{M} \exp[-g_e M H_z/k_BT], \tag{6}$$

where k_B and g_e are the Boltzmann constant and the Landé factor, respectively. Here, we have the energy levels $\epsilon_\mu(M)$, which are the eigenvalues of the $\widehat{H}_{spin}$, and, thus, can be derived from diagonalization procedure. Index μ runs over the energy levels with a given total spin projection $\mathbf{M}$. Once the energy levels of the spin Hamiltonian are obtained, one can evaluate a different thermodynamic properties of the magnetic system such as the magnetization, the magnetic susceptibility, and the magnetic specific heat. Since further in our study of [Tb_2] molecule its anisotropic part of the spin Hamiltonian will be treated only as a scalar, the magnetic properties of the system do not depend on the direction of the external magnetic field. Thereby, we can consider the external magnetic field H_z directed along arbitrary Z axis, that is chosen as a spin quantization direction of the molecule. In this case we can write the energies of the system as $\epsilon_\mu(M_s) + g_e M H_z$. Using this expression, one can evaluate the magnetization $\mathbf{M}$ via a standard thermodynamic definition:

$$\mathbf{M} = \frac{\partial F(M, H_z)}{\partial \mathbf{H}} = NkT\frac{\partial \ln Z}{\partial \mathbf{H}}. \tag{7}$$

Thereby, the effective magnetic field $\mathbf{H}_{eff}^{mean}$ can be derived from the free energy functional as:

$$\mathbf{H}_{eff}^{mean} = -\frac{\delta(F(M, H_z) + F(t))}{\delta \mathbf{M}} =$$

$$= -\frac{\partial F(M, H_z)}{\partial \mathbf{M}} + H_{pulse}^{x}(t). \tag{8}$$

Thus, we have derived, by means of Eqs. (5) and (8), a general form of the time-dependent spin equation for a system of the spins, precessing in an effective magnetic field $\mathbf{H}_{eff}^{mean}$, with interactions specified directly in the magnetic molecule. By solving Eq. (5), we can find the time-dependent expectations values $\langle\widehat{S}^{x,y,z}\rangle(t)$, which describe the spin dynamics of the system. We use them further to define the spin–spin correlation functions $\mathcal{C}_{ij}$ for the entangled ground state $|SM_0^{(ij)}\rangle = a_i|SM\rangle_i + b_j|SM\rangle_j$ of the system [30]:

$$\mathcal{C}_{ij}^{xx}(t) = \langle SM_0^{(ij)}|\widehat{S}_i^x(t)\widehat{S}_j^x(t)|SM_0^{(ij)}\rangle = \langle S_i^x\rangle(t)\langle S_j^x\rangle(t), \tag{9}$$

$$\mathcal{C}_{ij}^{yy}(t) = \langle SM_0^{(ij)}|\widehat{S}_i^y(t)\widehat{S}_j^y(t)|SM_0^{(ij)}\rangle = \langle S_i^y\rangle(t)\langle S_j^y\rangle(t), \tag{10}$$

$$\mathcal{C}_{ij}^{zz}(t) = \langle SM_0^{(ij)}|\widehat{S}_i^z(t)\widehat{S}_j^z(t)|SM_0^{(ij)}\rangle = \langle S_i^z\rangle(t)\langle S_j^z\rangle(t), \tag{11}$$

$$\mathcal{C}_{ij}^{xy}(t) = \langle SM_0^{(ij)}|\widehat{S}_i^x(t)\widehat{S}_j^y(t)|SM_0^{(ij)}\rangle = \langle S_i^x\rangle(t)\langle S_j^y\rangle(t), \tag{12}$$

$$\mathcal{C}_{ij}^{xz}(t) = \langle SM_0^{(ij)}|\widehat{S}_i^x(t)\widehat{S}_j^z(t)|SM_0^{(ij)}\rangle = \langle S_i^x\rangle(t)\langle S_j^z\rangle(t), \tag{13}$$

$$\mathcal{C}_{ij}^{yz}(t) = \langle SM_0^{(ij)}|\widehat{S}_i^y(t)\widehat{S}_j^z(t)|SM_0^{(ij)}\rangle = \langle S_i^y\rangle(t)\langle S_j^z\rangle(t). \tag{14}$$

The ground state is in the subspace $\mathcal{A}$ for which $M^{(\mu)} = 0$ for all μ. The entanglement entropy between a subspace $\mathcal{A}$ and the rest of the system $\mathcal{R}$ is given by:

$$S_{\mathcal{A}} = -\mathrm{Tr}(\rho_{\mathcal{A}}\log_2\rho_{\mathcal{A}}), \tag{15}$$

where $\rho_{\mathcal{A}}$ is the reduced density matrix of the subspace $\mathcal{A}$ obtained by tracing out over all those parts of the Hilbert space not associated with $\mathcal{A}$. We consider the subspace $\mathcal{A}_{ij} \equiv \{i,j\}$ consisting of all spin pairs (not only neighboring) i and j of the ground states $|S^{(\mu)}M^{(\mu)} = 0\rangle$ [14]. For the rest of the system $\mathcal{R}$ is given by $\mathcal{R}_{kl} \equiv \{k,l\}$. The matrix elements of the reduced density matrix, needed to calculate the entanglement, can be written in terms of the spin–spin correlation functions $\mathcal{C}_{ij}^{\alpha\beta}(t)$ (see Eqs. (9)–(14):

$$\rho_{\mathcal{A}}^{(ij)}(t) = \sum_{\alpha,\beta\in x,y,z}\langle SM_0^{(ij)}|\widehat{S}_i^{\alpha}(t)\widehat{S}_j^{\beta}(t)|SM_0^{(ij)}\rangle\rho_{\mu\nu}^{(ij)}, \tag{16}$$

where

$$\rho_{\mu\nu}^{(ij)} = |SM_0^{(ij)}\rangle_\mu \otimes |SM_0^{(ij)}\rangle_\nu.$$

Suppose $\{|\mathcal{A}_{ij}\rangle\}$ and $\{|\mathcal{R}_{kl}\rangle\}$ are the orthonormal basis states of the many-body Hilbert space of the subsystems $\mathcal{A}$ and $\mathcal{R}$. A general quantum spin state of the composite system can be described by a wave function [31]:

$$|\Psi\rangle = \sum_{ij,kl} C_{ij,kl}|\mathcal{A}_{ij}\rangle|\mathcal{R}_{kl}\rangle. \tag{17}$$

Here, a rectangular matrix $\mathbf{C}$ can be presented always in the form $\mathbf{UDV}^\dagger$, where $\mathbf{U}$ is unitary, $\mathbf{D}$ is diagonal and the rows of $\mathbf{V}$ are orthonormal. It is known already as the singular-value decomposition (SVD) and is similar to the principal-axis transformation of a symmetric square matrix [16]. Using this decomposition in Eq. (17) and forming a new basis by combining the $|\mathcal{A}_{ij}\rangle$ with $\mathbf{U}$ and the $|\mathcal{R}_{kl}\rangle$ with $\mathbf{V}^\dagger$, one can obtain the Schmidt decomposition [16]:

$$|\Psi\rangle = \sum_{k=1}^{Rank}\sigma_k|\Phi_k^{\mathcal{A}}\rangle|\Phi_k^{\mathcal{R}}\rangle, \tag{18}$$

which represents the total wave function $|\Psi\rangle$ of the system as a single sum of products of the orthonormal functions. Here the *Rank* number of the terms is limited by the smallest one of the two Hilbert spaces and the weight factors σ_k are the matrix elements of the diagonal matrix $\mathbf{D}$. If $|\Psi\rangle$ is normalized, their absolute magnitudes squared sum to one. The entanglement properties of a system are performed with the set of σ_k:

$$S_{\mathcal{A}}(t) = -\sum_{k=1}^{Rank}\tau_k(t)\log_2\tau_k(t), \tag{19}$$

where

$$\tau_k(t) = \langle S_i^{\alpha}\rangle(t)\langle S_j^{\beta}\rangle(t)\sigma_k^2. \quad (20)$$

3. Results and discussion

Nowadays the superconductors, trapped ions, nuclear magnetic resonance in organic liquids, quantum dots, molecular magnets are considered as the most promising candidates for generating a switch over a spin states, that is necessary condition needed to realize a spin qubits. Therefore, a certain new magnetic molecules, such as the complex Tb^{+3}- $[Tb_2Cl(HL4)_2(H_2L4)(py)_2]$, are treated now for perspective applications in the quantum computers [7]. The calculated (within the GSH model) spin levels of $[Tb_2]$ molecule are shown in Fig. 1. They are grouped according to the spin moment M. The obtained energy spectrum demonstrates several peculiar features such as the presence of the levels belonging to $M = 0$ (the ground state and low-lying excited states) and $M = \pm(1 \div 6)$ (the high-lying excited states) separated by a small gap, and the sets of the excited levels ($\geqslant$80 meV) well separated from the low-lying ones. Thereby, such a spin system demonstrates the ability to generate a spin switching over states. However, there are several key points, or counterparts related to successful realization of the quantum computing, such as the direct interaction of a spin qubit with the external media that eventually leads to the loss of quantum information, and a spin switching achieved only in the presence of the external magnetic field. Up to now no single-molecular system could be found in the literature, where the prime element involved in quantum-information processing is shielded within a spin qubit from the external media, and a spin qubit is realized thereto in the absence of the external magnetic field. Therefore, in this work we consider a possibility to switch the spin states in the system by pulses of a magnetic field of the different amplitude and duration. Recently, we have studied and discussed the spin dynamics of the Co- octaethylporphyrin (CoOEP) molecule in the low spin (LS) and high spin (HS) states induced by the applied magnetic pulse of 36.8T [27]. We have shown that in case of the HS state a spin switching of the CoOEP molecule is characterized by a long lifetime and depends on the magnitude and duration of the applied field. The applied external field reverted the system from the LS state to its ground state via a spin switching, whereas the system in the HS state remained in the excited state for a long time. Here, in contrast to work [27], we study the spin dynamics of $[Tb_2]$ magnetic molecule under the magnetic pulse with the duration of time about hundreds picoseconds. In Fig. 2 we present the result of our calculations, which demonstrate switching of a spin $\langle S^z\rangle$from one state to another caused by the magnetic pulse with amplitude of 27T and duration of 173 ps. It is well seen, that switching between two distinguishable spin states (here as spin-up $|\Uparrow\rangle$ and spin-down $|\Downarrow\rangle$) occurs practically at the same time when a magnetic pulse is emerging. Moreover the effect of spin switching remains steady even the applied magnetic field is switched off after 173 ps. Therefore, in order to control a spin switching over a spin states in n-qubit system based on $[Tb_2]$ molecule, we propose to use a scheme, which involves two magnetic pulses operating consecutively. We show in Fig. 3 the result of our calculations obtained for two applied magnetic pulses. We find in this case that both magnetic pulses applied consecutively to $[Tb_2]$ molecule lead in turn to a spin switching $\langle S^z\rangle$ between two distinguishable states. It means that one can control a spin states in the system by using both switch on and off of the magnetic pulses.

Further, we investigate the dynamics of the spin–spin correlations and entanglement entropy in SSNQ based on $[Tb_2]$ molecule, where spins are coupled via the GSH and affected by the time-dependent magnetic pulse. For these purposes the time-evolution

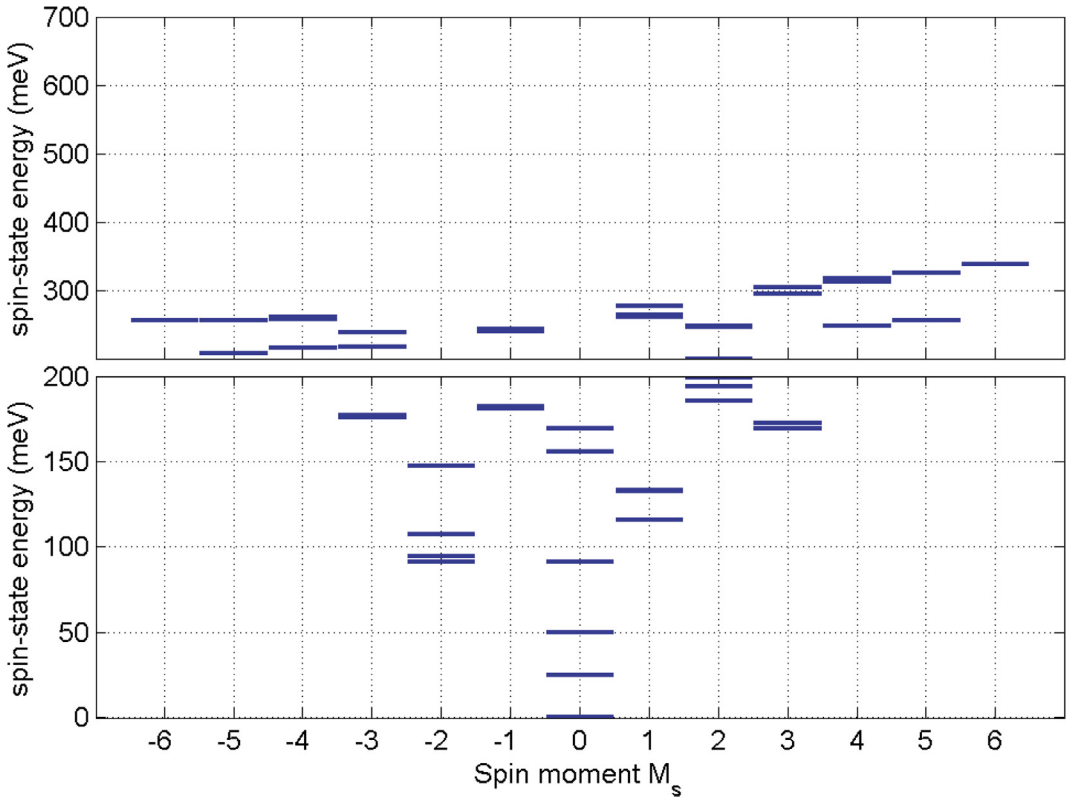

Fig. 1. (Color online) Spin structure of $[Tb_2]$ magnetic molecule calculated using the parameterization reported in [7].

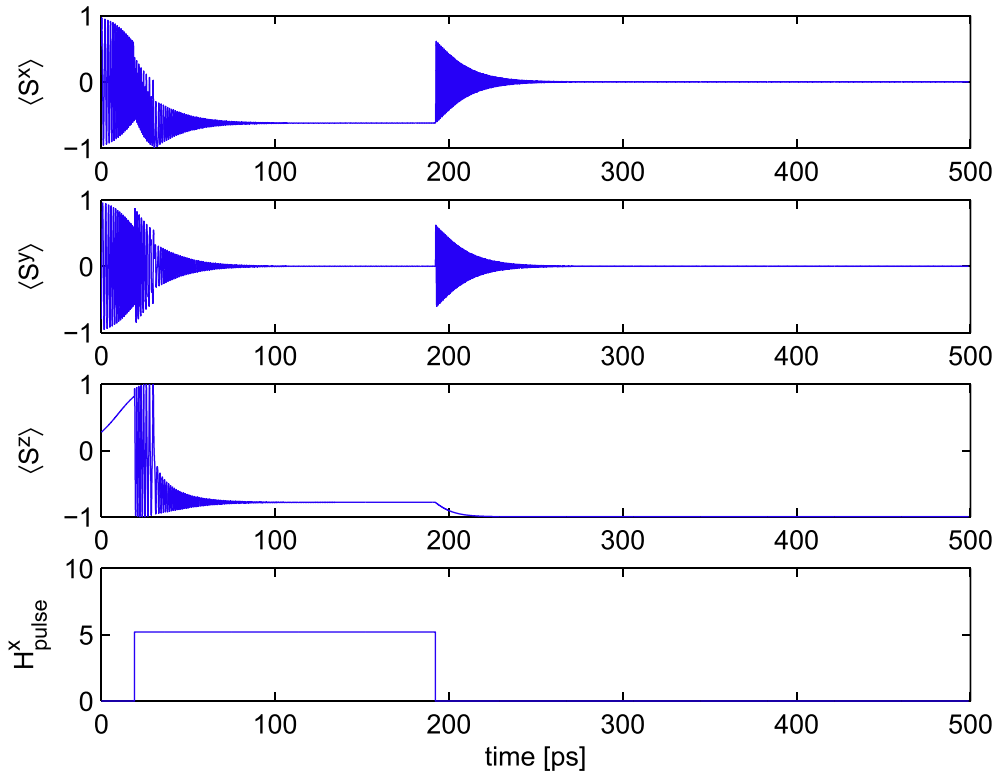

Fig. 2. (Color online) The evolution of the time-dependent quantum mechanical expectations $\langle S^{x,y,z}\rangle(t)$ of [Tb_2] molecule under a magnetic pulse H_{pulse} = 27T.

correlation functions $C_{ij}^{\alpha\beta}(t)$ (see Eqs. (9)–(14) have been calculated and used to study the dynamics of entanglement entropy in SSNQ of quantum [Tb_2] system. In Fig. 3 we plot the evolution of the entanglement entropy for n-qubit [Tb_2] system (with n = 49 of a spin states) using the following parameters in our calculations: S = 3; H_{ZEE} = 3.1T; H_{pulse}=27T; J_{ex} = −0.0014 meV; the axial ZFS parameter D = −1.5 meV, reported recently for [Tb_2] magnetic molecule [7]. As shown in Fig. 3 the entanglement exhibits oscillatory behavior at the beginning of time evolution from $t = 0$ up to $t = 85$ ps, when the pulsed magnetic field is emerging in the system. Stabilization comes up at the end of the lifetime of the first magnetic pulse. During the time interval between two consecutive magnetic pulses the entanglement starts to oscillate again within a short interval of time about 40 ps and reaches its maximum value. After that, the emergence of the second pulse causes the strong oscillations with a very short duration of time about 10 ps and leads the system to its maximum entanglement immediately. Thus, the obtained results reveal that the consecutively applied magnetic pulses with a certain duration and amplitude enable us to achieve the maximum entanglement in SSNQ, which does not change during a lifetime of an emerging pulse.

To define the entanglement of a SSNQ in the ground state, we determine the eigenvalues and eigenfunction of 49×49 matrix and ascribe the ground state through $|SM_0\rangle \equiv |SM = 0\rangle$: $\{|00\rangle, |10\rangle, |20\rangle, |30\rangle, |40\rangle, |50\rangle, |60\rangle\}$. By applying the Hadamard transform [24], we find two-qubit transformation $\left\{|SM_0^{(ij)}\rangle\right\}$. Based on this result and on realization of two-qubit **CNOT** gate reported for [Tb_2] earlier [10], we obtain the entangled states for quantum [Tb_2] system:

$$\mathbf{CNOT}: |\mathbf{SM_0^{(ij)}}\rangle \mapsto \left\{|S0\rangle_i, |S0\rangle_i \oplus |S0\rangle_j\right\}$$

This procedure can be extended to other controlled unitary operations like **CU** operation defined as [32]:

$$\mathbf{CU}: |S0\rangle_i\langle S0|_j \otimes I_s + |S0\rangle_i\langle S0|_j \otimes U_s^{(ij)}$$

The above unitary operation performs in general the transformation U_s over a set of qubits s (in the case of two qubits s = 2) if the state of the controlled qubit i is $|1\rangle$, and does not act otherwise. The transformation

$$U_s^{(ij)} = \exp(-i\widehat{H}_{spin}t) = \exp(i\gamma\mathbf{H}_{eff}\widehat{\mathbf{S}}_{ij}t)$$

is the operational representation of **CU** gate for a SSNQ. Based on that, we can perform here the class of the quantum computers that allows one to evaluate the ground state quantum algorithms in a register of a SSNQ with the physical spin–spin correlation functions of the form $C_{ij}^{\alpha\beta}(t)$. In the case of [Tb_2] qubit one can obtain the correlation functions with $\widehat{W}_s = \widehat{U}_s^{\dagger}\widehat{U}_s$ and $\widehat{U}_s$ unitary operators acting on s [33]. For this purpose [Tb_2] qubit is initialized at first in the state $|SM_0^{(ij)}\rangle = a_i|SM_0\rangle_i + b_j|SM_0\rangle_j$ by applying the unitary Hadamard gate (**H**) to the state $|SM_0\rangle_i$. Second, one makes it interact with the qubit system, initially in a certain pure state $|SM_0\rangle_i$, through two controlled unitary operations $\widehat{U}_s$, associated to the U_s operations respectively. After that, the final state of the quantum register $|\Psi_f\rangle$ can be ascribed by

$$\begin{aligned}|\Psi_f\rangle &= \widehat{U}_s\widehat{U}_s|SM_0^{(ij)}\rangle|SM_0\rangle \\ &= a_i|SM_0\rangle_i \otimes U_s|SM_0\rangle + b_j|SM_0\rangle_j \otimes U_s|SM_0\rangle. \end{aligned} \quad (21)$$

Thus,

$$U_s^{(ij)} = U_s^{(i)}U_s^{(j)} = \exp(i\gamma\mathbf{H}_{eff}\widehat{\mathbf{S}}_i t)\exp(i\gamma\mathbf{H}_{eff}\widehat{\mathbf{S}}_j t). \quad (22)$$

In Fig. 4 we present the obtained results of the time evolution of U_s calculated for different values of t. An effective field $\mathbf{H}_{eff}$ is proportional to the spin operator $\langle\widehat{\mathbf{S}}\rangle$. It means qualitatively, that the $\mathbf{H}_{eff}$ behaves in the same manner as the $\langle\widehat{\mathbf{S}}\rangle$ does, and it is well seen from the Fig. 5. Thus, dealing with a magnetic pulse of a certain

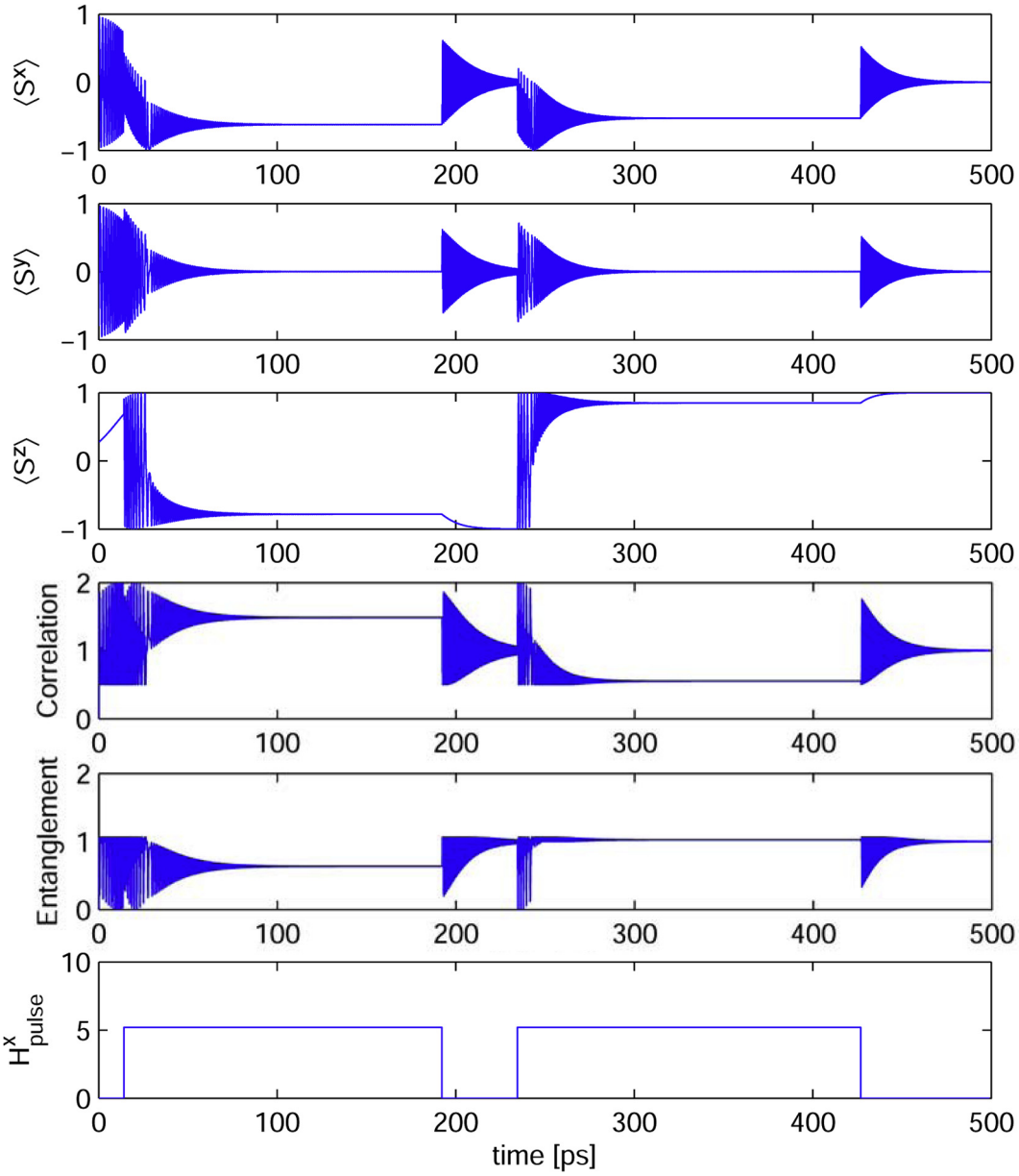

Fig. 3. (Color online) The evolution of the time-dependent quantum mechanical expectations $\langle S^{x,y,z}\rangle(t)$, correlation function and entanglement entropy of [Tb_2] molecule under two magnetic pulses H_{pulse} = 27T.

duration and amplitude, we can create a quantum register and operate with its help in the quantum algorithm, creating in the certain time the **CU**, **CNOT**, **SWAP** gates by means of an effective field $\mathbf{H}_{eff}$ and the spin operator $\langle\widehat{\mathbf{S}}\rangle$ in quantum [Tb_2] system. For instance, as it is seen from Fig. 4, during the action of the first magnetic pulse U_s creates the quantum **CNOT** gate:$|10\rangle \mapsto |11\rangle$, and the quantum **SWAP** gate is available in an interval of time between two magnetic pulses, whereas by the action of the second magnetic pulse the quantum **CNOT** gate:$|10\rangle \mapsto |10\rangle$ is realized. It is worth to note, that in quantum computing one is faced with a necessity to have a deal with the physical interactions in the Hamiltonian. Actually, these interactions not necessarily would have to lead to a quantum mechanical evolution of the system, which can be interpreted as **CNOT** gate. Nevertheless, there is always possible to find within Hamiltonians available for various physical systems the two-qubit gates from which, in conjunction with one-qubit gates, the **CNOT** gates can be realized [34]. For instance, in Fig. 4 we demonstrate one of an explicit construction for how to realize the **CNOT** gates from the two-body interactions $\widetilde{U}_s^{yzz} \equiv U_s^y U_s^z U_s^z$ that are available in our quantum [Tb_2] system. However, the key problem is how many **CNOT** and one-qubit gates are necessary and sufficient in order to implement any two-qubit gate. To solve this problem, one can employ the following criterion, that makes it is possible to determine the number of **CNOT** gates needed to realize a two-qubit gate with a help of one-qubit operations: U_s can be realized using two **CNOT** gates if and only if $\mathrm{Tr}(U_s)$ is real [24,35,36]. In our case $\mathrm{Tr}(U_s) = 2\cos(\gamma \mathbf{H}_{eff}\widehat{\mathbf{S}}_{ij}t)$ is real, and the relevant solution for [Tb_2] system is illustrated in Fig. 4, where $Re(U_s^z)$ is shown in the form of two **CNOT** gates.

4. Conclusions

In the present paper a general study of [Tb_2] molecular magnet is carried out using the general spin Hamiltonian formalism. We present for this molecule the results of the theoretical study of

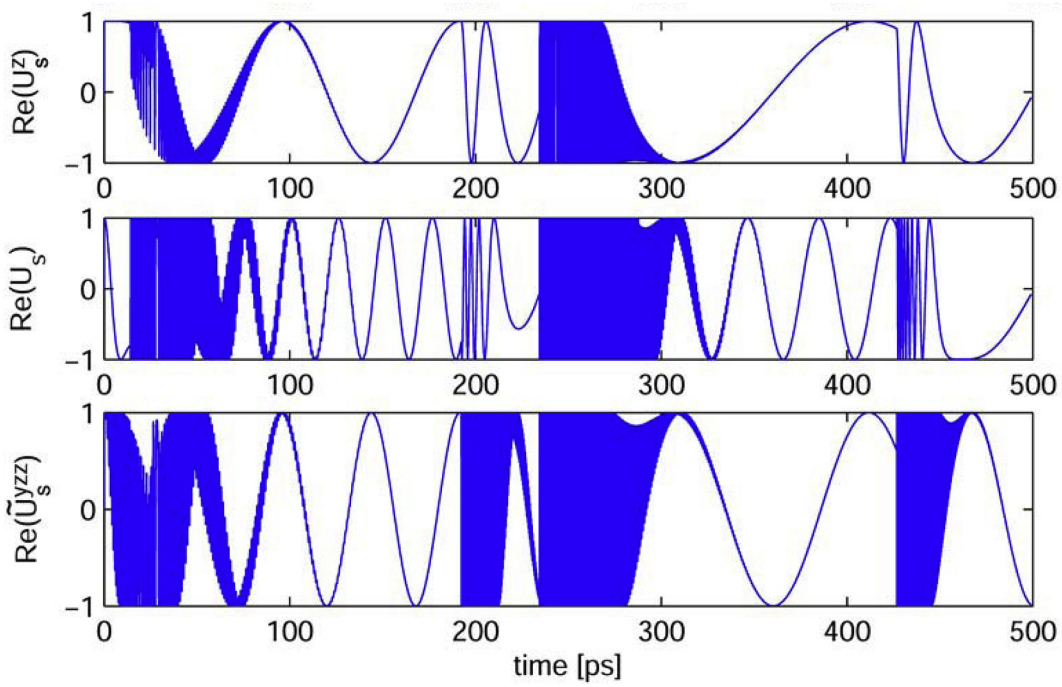

Fig. 4. (Color online) The time evolution behavior of the real parts (Re) of U_s^z, U_s, and $\tilde{U}_s^{yzz}$ in [Tb_2] molecule under two magnetic pulses H_{pulse} = 27T.

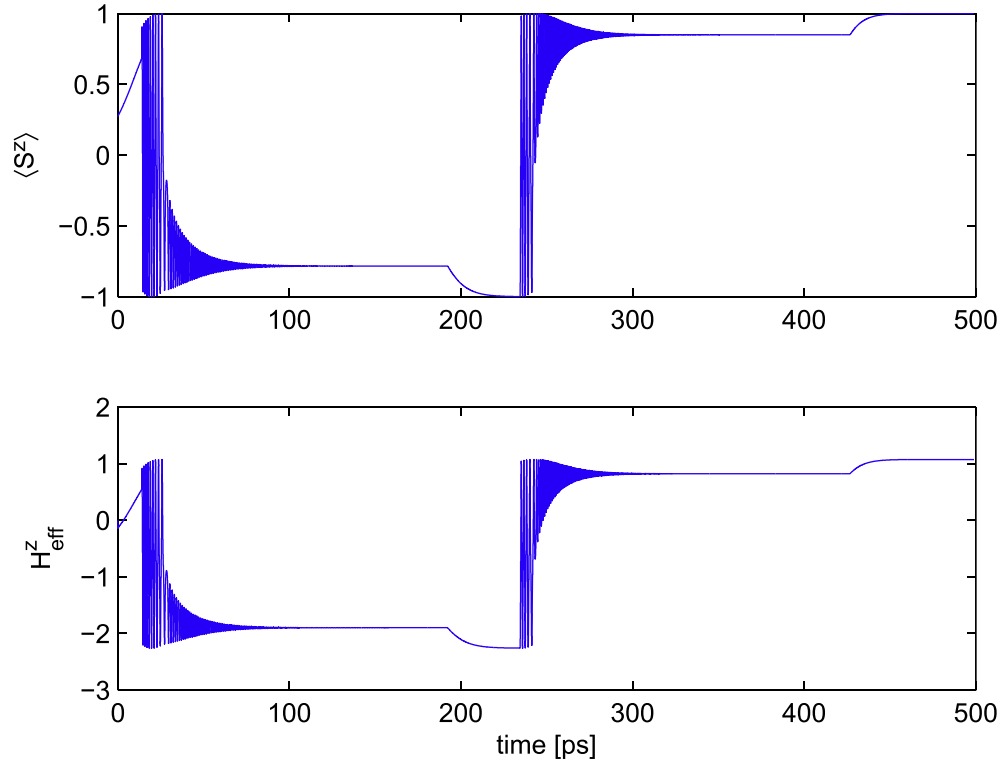

Fig. 5. (Color online) The time evolution behavior of the quantum mechanical expectation $\langle S^z \rangle(t)$ and effective magnetic field H_{eff} in [Tb_2] molecule under two magnetic pulses H_{pulse} = 27T.

the spin behavior affected by the magnetic pulse with the duration of 200 ps and amplitude of 27T. Based on the LLG equation the spin dynamics of [Tb_2] molecule is described. We obtain also a general form of the time-dependent spin equation for a system of spins precessing in an effective magnetic field with specified interactions. In order to control a spin switching of n-qubit [Tb_2] system, we propose a scheme, which involves two magnetic pulses acting consecutively on the magnetic molecule. It is shown, that one can control a spin states in the system by switching on and off of the magnetic pulses. The spin–spin correlators of a spin wave functions in [Tb_2] molecule are analyzed numerically and compared in details with the results obtained by standard quantum mechanics.

We demonstrate that, besides the various expectation values of the spin operators, a study of their corresponding probability distributions allows to have a novel understanding in spin dynamics of entangled qubit. It is shown also via conventional quantum mechanics that the properties of spin–spin correlators are responsible for various entanglement in spin qubit under a pulse magnetic field. We propose with the general spin Hamiltonian some compact schemes for implementation of quantum computing in SSNQ based on [Tb_2] molecule, including the **CNOT** and **SWAP** gates.

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