

## Quantum computing of molecular magnet $Mn_{12}$

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Quantum computation in molecular magnets is studied by solving the time-dependent Schrödinger equation numerically. Following Leuenberger and Loss [Nature (London) **410**, 789 (2001)], an external alternating magnetic field is applied to populate and manipulate a superposition of single-spin states in molecular magnet  $Mn_{12}$ . The conditions to realize parallel recording and reading databases of Grover algorithms in molecular magnets are discussed in detail. It is found that an accurate time duration of the magnetic pulse as well as the discrete frequency spectrum and the amplitudes are required to design a quantum computing device.

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The quantum phase was proposed to store information in connection with a different class of computational algorithms based on the rules of quantum mechanics rather than classical physics [1]. One example is the database search problem proposed by Grover [2,3]. Different from other quantum algorithms, the superposition of quantum phases in single-particle quantum states is sufficient to implement the Grover algorithm. Grover's algorithm was successfully implemented using Rydberg atoms [4]. Recently, Leuenberger and Loss proposed that molecular magnet  $Mn_{12}$  can be used to realize the Grover algorithm by utilizing multifrequency coherent magnetic radiation [5]. In the  $S$  matrix and time-dependent perturbation theory, it is possible to populate and manipulate a superposition of single-spin states by applying a single pulse of weak alternating transverse magnetic field with a number of matching frequencies, and the state can be applied to store a multibit information. In this paper the population and manipulation of the spin excited states in an external alternating magnetic field are studied by solving the time-dependent Schrödinger equations numerically. The conditions for the magnetic pulse to implement the Grover algorithm are discussed in detail. We find that an accurate duration of magnetic pulse as well as the discrete frequencies and amplitudes are required to design a quantum computing device in the molecular magnet  $Mn_{12}$ .

Following Leuenberger and Loss, we consider a molecular magnet with spin  $S(>1/2)$  in the presence of a weak alternating transverse magnetic field. The Hamiltonian for the system reads

$$H = H_{\text{spin}} + V_{\text{low}}(t) + V_{\text{high}}(t), \quad (1)$$

where

$$H_{\text{spin}} = -AS_z^2 - BS_z^4 + g\mu_B \delta H_z S_z, \quad (2)$$

$$V_{\text{low}}(t) = g\mu_B H_0(t) \cos(\omega_0 t) S_z, \quad (3)$$

$$V_{\text{high}}(t) = \sum_{m=m_0}^{s-1} \frac{g\mu_B H_m(t)}{2} [e^{i(\omega_m t + \Phi_m)} S_+ + e^{-i(\omega_m t + \Phi_m)} S_-]. \quad (4)$$

$H_{\text{spin}}$  is a single-spin Hamiltonian for the molecular magnet  $Mn_{12}$  [6].  $|m\rangle$  are the simultaneous eigenstates of  $H_{\text{spin}}$  and the  $z$ -component spin operator  $S_z$  with the energy  $\varepsilon_m$  and the moment  $m\hbar$ .  $V_{\text{low}}$  and  $V_{\text{high}}$  are the Zeeman terms to describe the coupling between the external magnetic fields and the spin  $S$ . The magnetic field with a low frequency  $\omega_0$  is applied along the easy axis, and  $V_{\text{low}}$  supplies the necessary energy for the resonance condition. The high-frequency transverse fields are introduced to induce the transition from the initial state  $|s\rangle$  to excited states  $|m\rangle, m=m_0, \dots, s-1$ , and the frequencies  $\omega_m, m=m_0, \dots, s-2$ , mismatch the level separation by  $\omega_0$ , that is,  $\hbar\omega_m = \varepsilon_m - \varepsilon_{m+1} + \hbar\omega_0$ , and  $\hbar\omega_{s-1} = \varepsilon_{s-1} - \varepsilon_s - (s-m_0-1)\hbar\omega_0$ . The phases  $\Phi_m = \sum_{k=s-1}^{m+1} \Phi_k + \varphi_m$  ( $\varphi_m$  are the relative phases) and

$$H_m(t) = \begin{cases} H_m & \text{if } t \in \left(-\frac{T}{2}, \frac{T}{2}\right) \text{ for } m=0 \text{ and } m \geq m_0, \\ 0, & \text{otherwise.} \end{cases}$$

The time-dependent Schrödinger equation reads

$$i\hbar \frac{\partial}{\partial t} |\Psi(t)\rangle = H |\Psi(t)\rangle, \quad (5)$$

where

$$|\Psi(t)\rangle = \sum_{m=m_0}^s a_m(t) e^{-i\varepsilon_m t/\hbar} |m\rangle. \quad (6)$$

The eigenstates of  $m < m_0$  have been neglected. The low-energy states of a molecular magnet  $Mn_{12}$  behave as a single spin with  $s=10$ .

The Grover scheme adapts to describe the quantum computational read-in and decoding of the quantum data register

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$\mathbf{a}=(a_s, a_{s-1}, \dots, a_{m_0})$ . As a simple example, we discuss the case of  $m_0=9$ . At the first stage of the so-called ‘‘read-in,’’ the initial values of  $\{a_m^{(0)}:m=10,9\}$  are set to be  $\{a_{10}^{(0)}=1, a_9^{(0)}=0\}$ . After irradiating the molecular magnet  $\text{Mn}_{12}$  with a coherent magnetic pulse with the relative phase  $\varphi_9^{(1)}$  and the duration  $T$ , one has the quantum data register  $\mathbf{a}=(a_{10}^{(1)}, a_9^{(1)})$  with

$$a_{10}^{(1)} = \cos\left(\frac{g\mu_B H_9}{2\hbar} \langle 9|S_-|10\rangle T\right), \quad (7)$$

$$a_9^{(1)} = -ie^{-i\varphi_9^{(1)}} \sin\left(\frac{g\mu_B H_9}{2\hbar} \langle 9|S_-|10\rangle T\right). \quad (8)$$

At the second stage of the so-called ‘‘decoding,’’ the solutions of Eqs. (7) and (8) become the initial values of  $\{a_m\}$  in Eq. (5), and the relative phase is set to be  $\varphi_9^{(2)}=0$ . At the end of decoding, one obtains

$$a_9^{(2)} = \begin{cases} -i \sin\left(2\frac{g\mu_B H_9}{2\hbar} \langle 9|S_-|10\rangle T\right) & \text{for } \varphi_9^{(1)}=0 \\ 0 & \text{for } \varphi_9^{(1)}=\pi. \end{cases} \quad (9)$$

If the condition of  $g\mu_B H_9 \langle 9|S_-|10\rangle T/2\hbar \ll 1$  is satisfied, the result in Eq. (9) is in good agreement with Ref. [5]. For a longer duration time,  $T \gg (g\mu_B H_9 \langle 9|S_-|10\rangle/2\hbar)^{-1}$ , the condition fails to implement the Grover algorithm.

For  $m_0 < 9$ , it is difficult to solve Eq. (5) analytically. However, as it is a single-particle problem with a finite number of states, it is possible for us to solve the differential equations [Eq. (5)] numerically instead of evaluating the  $S$ -matrix perturbatively as Leuenberger and Loss did. The results of the explicit numerical calculation are more reliable than other approximate results. For an illustration, we focus on the case of  $m_0=5$ . In this case six states can be used for quantum computation. The parameters used here are the same as the ones used in Ref. [5]:  $\omega_0=5 \times 10^7 \text{ s}^{-1}$ ,  $T=10^{-7} \text{ s}$ ,  $H_0=H_9=2 \text{ G}$ ,  $H_8/H_0=-0.04$ ,  $H_7/H_0=-0.25$ ,  $H_6/H_0=-0.61$ , and  $H_5/H_0=-1.12$ . For the molecular magnet  $\text{Mn}_{12}$ ,  $A$  and  $B$  in Eq. (2) are  $0.56 \text{ K}$  and  $1.11 \times 10^{-3} \text{ K}$ , respectively [6]. According to Ref. [5], the relative phases are  $\varphi_9^{(1)}=\varphi_8^{(1)}=\varphi_7^{(1)}=0$  and  $\varphi_6^{(1)}=\varphi_5^{(1)}=\pi$  for encoding the number  $13_{10}=1101_2$ . Now, we solve six first-order differential equations for  $\{a_m(t)\}$  in Eq. (6). The initial values of the  $\{a_m\}$  are set to be  $a_{10}^{(0)}=1$  and  $a_m^{(0)}=0$  for  $m \neq 10$ . After a duration  $T$ , we have  $\mathbf{a}^{(1)}=(a_{10}^{(1)}, a_9^{(1)}, a_8^{(1)}, a_7^{(1)}, a_6^{(1)}, a_5^{(1)})$ . At the second stage for decoding the numbers  $\mathbf{a}^{(1)}$  become the initial values of Eq. (5) and the relative phases should be set by  $\varphi_9^{(2)}=\varphi_7^{(2)}=\varphi_5^{(2)}=0$  and  $\varphi_8^{(2)}=\varphi_6^{(2)}=\pi$ . Once again, the six first-order differential equations with the new initial conditions are solved numerically. At the end of decoding, we obtain  $\mathbf{a}^{(2)}=(a_{10}^{(2)}, a_9^{(2)}, a_8^{(2)}, a_7^{(2)}, a_6^{(2)}, a_5^{(2)})$ . If the Grover algorithm is implemented successfully, according to Leuenberger and Loss, one should have  $|a_{10}^{(2)}| \approx 1, |a_9^{(2)}| \approx 2|a_9^{(1)}| \approx 2\eta$ ,  $|a_8^{(2)}|$

$\approx 0, |a_7^{(2)}| \approx 2|a_7^{(1)}| \approx 2\eta, |a_6^{(2)}| \approx 2|a_6^{(1)}| \approx 2\eta, |a_5^{(2)}| \approx 0$  and  $\eta \ll 1$ . Instead, our numerical calculation gives

$$|a_{10}^{(1)}| \approx 0.95, |a_9^{(1)}| \approx 0.31, |a_8^{(1)}| \approx 0.04,$$

$$|a_7^{(1)}| \approx 0.01, |a_6^{(1)}| \approx 0.02, |a_5^{(1)}| \approx 0.01$$

and

$$|a_{10}^{(2)}| \approx 0.82, |a_9^{(2)}| \approx 0.59, |a_8^{(2)}| \approx 0.08,$$

$$|a_7^{(2)}| \approx 0.06, |a_6^{(2)}| \approx 0.01, |a_5^{(2)}| \approx 0.01.$$

The values of  $\{\mathbf{a}^{(1)}, \mathbf{a}^{(2)}\}$  are complex numbers (not real) and we only present their absolute values here. Therefore, the numerical solution shows that the encoding number is  $0001_2$ , but not  $1101_2$  as expected by Leuenberger and Loss at the specific values of the applied field and  $T$ . However,  $\{\mathbf{a}^{(1)}, \mathbf{a}^{(2)}\}$  depend on  $T$ , and one may be still able to reach to the encoding number  $1101_2$  by choosing a specific duration of pulse.

To clarify the feasibility of the Leuenberger and Loss scheme, we study a simple, but nontrivial, case of  $m_0=8$  as a representative example. The key approximation in the method of Leuenberger and Loss is that  $S_{m,s}^{(j)}=0$  for  $j < n$  ( $=s-m_0$ ). The transitions from  $|k\rangle$  to  $|m\rangle$ , i.e.,  $S_{m,k}^{(j)}$  ( $m < k < s$ ) and all higher-order amplitudes  $S_{m,s}^{(j)}$  are neglected. In the case of  $m_0=8$ , the first-order perturbation is required to be zero:  $S_{9,10}^{(1)}=S_{8,9}^{(1)}=0$ , or at least  $|S_{9,10}^{(1)}| \ll |S_{9,10}^{(2)}| (=|S_{8,10}^{(2)}|)$  and  $|S_{8,9}^{(1)}| \ll |S_{9,10}^{(2)}| (=|S_{8,10}^{(2)}|)$ . In fact, the quantum amplitudes of the transitions by the perturbation  $S$ -matrix formula are given by

$$S_{9,10}^{(2)} = \frac{2\pi}{i} \left(\frac{g\mu_B}{2\hbar}\right)^2 \frac{H_0 H_9 e^{i\Phi_9}}{\omega_0} \langle 9|S_-|10\rangle \frac{T}{2\pi}, \quad (10)$$

$$S_{8,10}^{(2)} = \frac{2\pi}{i} \left(\frac{g\mu_B}{2\hbar}\right)^2 \frac{H_8 H_9 e^{i\Phi_8} e^{i\Phi_9}}{\omega_0} \langle 8|S_-|9\rangle \langle 9|S_-|10\rangle \frac{T}{2\pi}, \quad (11)$$

$$S_{9,10}^{(1)} = \frac{1}{i} \frac{g\mu_B H_9 e^{i\Phi_9}}{2\hbar} \langle 9|S_-|10\rangle \frac{\sin(\omega_0 T/2)}{\omega_0/2}, \quad (12)$$

$$S_{8,9}^{(1)} = \frac{1}{i} \frac{g\mu_B H_8 e^{i\Phi_8}}{2\hbar} \langle 8|S_-|9\rangle \frac{\sin(\omega_0 T/2)}{\omega_0/2}, \quad (13)$$

where  $\Phi_9=\varphi_9$  and  $\Phi_8=\Phi_9+\varphi_8$  ( $\varphi_9$  and  $\varphi_8$  are the relative phases). The first two terms  $\{S_{9,10}^{(2)}, S_{8,10}^{(2)}\}$  result from the second-order perturbation contributions with the energy conservation. From the condition  $|S_{9,10}^{(2)}| \approx |S_{8,10}^{(2)}|$ , one can deduce the field amplitude  $H_8/H_0=0.16$ . However, the lower-order terms  $S_{9,10}^{(1)}$  and  $S_{8,9}^{(1)}$ , in general, are not exactly equal to zero even if they do not satisfy the energy conservation. They can be zero only if  $\omega_0 T=2\pi l$ , where  $l$  is an integer. Due to these  $S_{9,10}^{(1)}$  and  $S_{8,9}^{(1)}$  terms, the amplitudes of  $a_8(t)$  and  $a_9(t)$  in the state  $|\Psi(t)\rangle$  oscillate periodically with  $T$  as shown in Fig. 1.

Now, let us assume the parameters:  $T=1.0 \times 10^{-9} \text{ s}$ ,  $H_0=200 \text{ G}$ ,  $H_9=20 \text{ G}$ , and  $\omega_0=4\pi \times 10^9 \text{ s}^{-1}$ . This gives

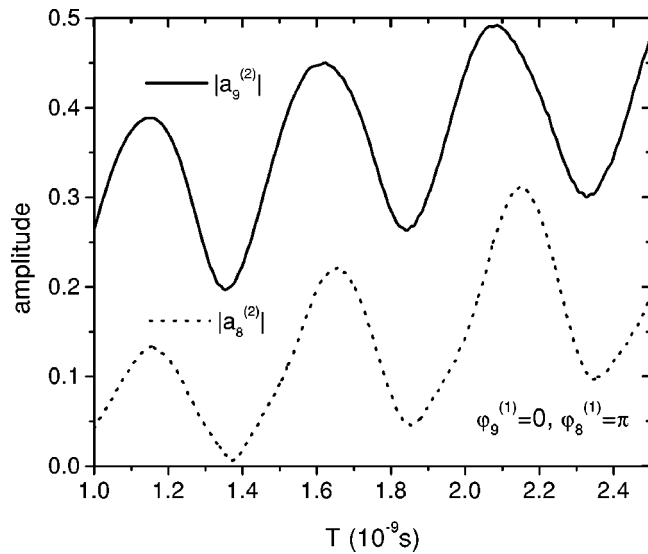


FIG. 1. The amplitudes  $|a_9^{(2)}|$  and  $|a_8^{(2)}|$  for  $m_0=8$  oscillate with the pulse duration  $T$  in the unit  $10^{-9}$  s (for  $\varphi_9^{(1)}=0$  and  $\varphi_8^{(1)}=\pi$ ). The parameters for the magnetic pulse are  $H_0=200$  G,  $H_9=20$  G,  $H_8=0.16H_0$ , and  $\omega_0=4\pi\times 10^9$  s $^{-1}$ .

$|S_{8,10}^{(2)}|\approx 0.11$ . Following Leuenberger and Loss, we set  $\varphi_9^{(2)}=0$  and  $\varphi_8^{(2)}=0$ . Then, the recording number  $00_2$  corresponds to  $\{\varphi_9^{(1)}=\pi, \varphi_8^{(1)}=\pi\}$ ,  $01_2$  to  $\{\varphi_9^{(1)}=0, \varphi_8^{(1)}=\pi\}$ ,  $10_2$  to  $\{\varphi_9^{(1)}=\pi, \varphi_8^{(1)}=0\}$ , and  $11_2$  to  $\{\varphi_9^{(1)}=0, \varphi_8^{(1)}=0\}$ . The results by our numerical calculation are shown in the Table I (where  $m=9,8$  represent the binary digits  $2^0$  and  $2^1$ , respectively). Using the above parameters, the Grover algorithm can be implemented as suggested by Leuenberger and Loss. However, the values  $\{|a_{10}^{(2)}|, |a_9^{(2)}|, |a_8^{(2)}|\}$  are not constants and will be changed with the duration of the magnetic pulse. In a certain range,  $|S_{9,10}^{(1)}|$  or  $|S_{8,9}^{(1)}|$  will have a magnitude close to  $|S_{m,s}^{(n)}|$ , which can deform the recording, or even more seriously destroy the recording completely. In Fig. 1 we plot the amplitudes of  $|a_9^{(2)}|$  and  $|a_8^{(2)}|$  with varying  $T$  (for  $\varphi_9^{(1)}=0$  and  $\varphi_8^{(1)}=\pi$ ), which corresponds to  $01_2$ . The amplitudes  $|a_9^{(2)}|\sim 0.26$  and  $|a_8^{(2)}|\sim 0.04$  for  $T=10^{-9}$  s. The value 0.26 can be regarded as single-digit number 1 and 0.04 as 0. The values  $|a_9^{(2)}|$  and  $|a_8^{(2)}|$  are varied with  $T$ . The first maximum of  $|a_8^{(2)}|\sim 0.107$  at  $T\sim 1.16\times 10^{-9}$  s, the second maximum  $|a_8^{(2)}|\sim 0.22$  at  $T\sim 1.66\times 10^{-9}$  s, which becomes larger than the first minimum of  $|a_9^{(2)}|\sim 0.19$  at  $T\sim 1.37\times 10^{-9}$  s. Therefore, we conclude that an accurate duration

TABLE I. The numerical results for the case of  $m_0=8$ .

Decoded number	$ a_{10}^{(1)} $	$ a_9^{(1)} $	$ a_8^{(1)} $	$ a_{10}^{(2)} $	$ a_9^{(2)} $	$ a_8^{(2)} $
$00_2$	0.99	0.13	0.10	1.00	0.07	0.01
$01_2$	0.99	0.13	0.10	0.96	0.26	0.04
$10_2$	0.98	0.14	0.10	0.98	0.04	0.20
$11_2$	0.98	0.14	0.10	0.94	0.28	0.19

of pulse should be chosen for a stable and clear quantum recording.

The above discussion and conclusion can be generalized straightforwardly to the cases of any  $m_0$ . The more quantum states are used to store information, the higher precision is required to design the magnetic pulses. It is clear that the lower-order perturbation series,  $j<n$ , of the  $S$  matrix do not vanish for a general  $T$  even when the relative transitions do not satisfy energy conservation. Therefore, to implement the encoding and decoding in the molecular magnets by applying an external alternating magnetic field, one should be very careful to design the appropriate  $T$  as well as the matching frequencies and amplitudes of the field.

In conclusion, we studied the quantum computing scheme proposed by Leuenberger and Loss by solving the time-dependent schrödinger equation instead of the perturbation theory. Our calculation shows that some aspects of the scheme should be improved. We point out that the energy conservation used in the perturbation theory is not valid since the duration of pulse in the problem is very short such that the contributions from the processes with no energy conservation becomes non-negligible. It follows that the multibit codes are  $T$  dependent, not as expected in the perturbation theory. Thus,  $T$  control is one of the important steps in future applications. More detailed calculations show that the multibit codes in the model of Leuenberger and Loss are stable to a reasonable extent with respect to some variation of  $T$  as well as frequency  $\omega_0$  and magnitude  $H_0$  of magnetic field. Leuenberger and Loss' scheme may have the potential, in future, to implement Grover's algorithm. There will be some scope for further study to improve the stability of codes recording and decoding.

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