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Long-lived Néel states in antiferromagnetic quantum spin chains with strong uniaxial anisotropy for atomic-scale antiferromagnetic spintronics

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It has been experimentally established that magnetic adatoms on surfaces can be arranged to form antiferromagnetic quantum spin chains with strong uniaxial anisotropy and Neel states in such spin systems can be used to realize information storage. Here, we investigate eigen states, quantum spin dynamics, and life times of Neel states in short antiferromagnetic quantum spin chains with strong uniaxial anisotropy on the basis of numerical exact diagonalization method. We show rigorously that as long as the uniaxial anisotropy is very strong, the ground state and the first excitation state, being nearly degenerate, are safely separated from the other states and thus dominate the quantum dynamics of the Neel states. Through further numerical analysis, we achieve a powerful life-time expression of the Neel states for arbitrary spin and model parameters. It is interesting that for the famous Fe adatom chains on Cu_2N surface, 14 or 16 Fe adatoms are enough to obtain a practical long life-time for Neel state storage of information. These should be applicable to other similar antiferromagnetic spin systems for atomic-scale antiferromagnetic spintronics.

1. Introduction

It is inspiring that adatom-based antiferromagnets have been realized on semiconductor surfaces and used for novel magnetic information storage because their Néel states can be stabilized by strong uniaxial single-ion magnetic anisotropy[1–3]. Such nanomagnets can be fabricated adatom by adatom, and their spin anisotropy can be controlled[4–7]. Thus, one can make antiferromagnetic chains, bi-chains, nano-ribons, or nano-sheets consisting of several or tens of adatom spins with strong magnetic anisotropy and adjustable inter-spin interactions. Spin chains are of much interest because they belong to an important category of Heisenberg spin models. In fact, various one-dimensional antiferromagnetic Heisenberg models have been intensively investigated[8–15]. For $S = 1$, there exists an inetresting Haldane topological phase if there is no strong uniaxial magnetic anisotropy[16–18]. On experimental side, one usually use high spins with strong uniaxial single-ion magnetic anisotropy in adadtom spin systems[1–7]. It is known that strong uniaxial single-ion anisotropy is necessary to achieve stable Néel states. Experimentally, electrons currents injected through STM tips have been used to control the Néel states for information storage[3]. On theoretical side, some efforts have been made to understand and explore controlling the adatom-spin antiferromagnets with spin-polarized electron current[19, 20], spin current[21], and mechanical oscillator[22] and to investigate symmetry effects on spin switching of single adatoms[23]. It is believed that more significant advances and deeper insight in this field can likely lead to an atomic-scale antiferromagnetic spintronics.

Here, we investigate the intrinsic quantum dynamics and life times of Néel states in the quantum Heisenberg antiferromagnetic chain model consisting of $2N$ spins ($S \geq 1$) with strong uniaixal single-ion anisotropy. We accurately calculate eigenvalues and eigenfunctions through exact diagonalization, and rigorously show that the ground state and the first excitation can be both safely separated from the other states and well described with the two Néel states as long as the single-ion anisotropy is very strong. Then, we thereby calculate the switching rates and life times of the Néel states. Surprisingly, we achieve a unified powerful expression of the life times through fitting our accurate numerical results. More importantly, for the Fe-adatom spin antiferromagnets on Cu_2N semiconductor surface[1–3], $2N$= 14 or 16 is large enough to achieve practical life times of Néel states for information storage. More detailed results will be presented in the following.

2. Results and discussion

2.1 Spin Model and eigenstates

We start with general one-dimensional quantum Heisenberg antiferromagnetic model with strong uniaxial single-ion anisotropy,

$$\hat{H} = J \sum_{i=1}^{2N-1} \hat{\vec{S}}_i \cdot \hat{\vec{S}}_{i+1} - D \sum_{i=1}^{2N} (\hat{S}_i^z)^2, \qquad (1)$$

where the total number of the spins is $2N$, the parameter J (> 0) is the antiferromagnetic exchange constant, D (> 0) is used to characterize the single-ion magnetic anisotropy in the z axis, and $\hat{\vec{S}}_i$ is the spin operator at site i, satisfying open boundary condition. For this antiferromagnetic spin chain, there are two special states, namely Néel states, as illustrated in Fig. 1. They are important, especially when D is very large in comparison to J. Here, we do not need any inhomogeneous effective magnetic field to split the two Néel states[3, 19], but to

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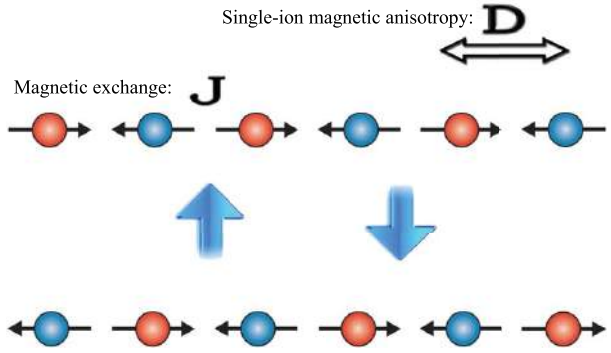

FIG. 1. An illustration of the two Néel states of the spin chain. An adatom spin is presented with a ball with an arrow. Parameter D is the uniaxial anisotropy of each spin, and J the coupling constant between the nearest spins.

experimentally prepare a specific Néel state, one can use an STM tip to inject a spin-polarized electron current on the first adatom spin[3]. If being applied to similar spin rings with uniaxial anisotropy, such as antiferromagnetic molecule wheels[24], the Hamiltonian (1) needs some modification to make the spin operators satisfy periodic boundary condition.

Using $\hat{S}_i^{\pm} = \hat{S}_i^x \pm i\hat{S}_i^y$, we have $\hat{\vec{S}}_i \cdot \hat{\vec{S}}_{i+1} = \hat{S}_i^z \cdot \hat{S}_{i+1}^z + \frac{1}{2}(\hat{S}_i^+ \cdot \hat{S}_{i+1}^- + \hat{S}_i^- \cdot \hat{S}_{i+1}^+)$. The ideal Néel states ($|N_1\rangle$ and $|N_2\rangle$) are certainly not the eigenstates of Hamiltonian (1) due to the transverse part including the raising and lowing operators $\hat{S}_i^{\pm}$, but the strong single-ion anisotropy D in the z axis makes the spin tend to orient in the z axis. Consequently, there are large (D/J dependent) weight of the two Néel states in the ground state and low excitations, which implies that the two Néel states for large D/J can be stable enough to be used for information storage[3]. Using exact diagonalization method[25] to the Hamiltonian (1), we can obtain the spin eigenvalues and eigenfunctions. For convenience, we shall use J as our unit in the following, which means that anisotropy parameter D and energy E can be scaled in terms of J.

In Fig. 2 we present the energy eigenvalues depending on N (1 through 5) and D/J (10, 3, and 1) for $S = 1$ and 3/2. For each of the cases, the ground state G and the first excitation E_1 are both separated from the other states. The trend is that the separation increases with D/J and S. The corresponding energy gaps between the ground states and the first excitation ones are summarized in Table I. It is clear in the table that the gap decreases with N, S, and D/J. The weights of the Néel states in the ground state (G) and the first excitation (E_1) as functions of N are presented in Table II for the two spin values and the three D/J ones. It can be seen that the Néel weights increase with D/J, but decrease with N. Except the special case of $N = 1$, the Néel weights increase with S, too. Because we are interested in the cases with strong uniaxial anisotropy, the ground state and low excitations are far from the regime of the Haldane state[16–18].

It is easy to prove that the total spin z-component $\hat{S}^z = \sum_i \hat{S}_i^z$ is conserved because it is commutable with the Hamiltonian (1). All the energy eigenstates can be classified in terms of the eigenvalue S^z of $\hat{S}^z$. Generally speaking, for a finite antiferromagnetic chain with $2N$ spins, the ground state is a spin single state. When D/J is very large, the ground state can be approximately constructed with a superposition of the two Néel states. For general D/J, we can always construct the following two eigenstates from the Néel states.

$$\begin{cases} |+\rangle = c_1(|N_1\rangle + |N_2\rangle + O_1|\mathcal{O}_+\rangle + \cdots) \\ |-\rangle = c_2(|N_1\rangle - |N_2\rangle + O_2|\mathcal{O}_-\rangle + \cdots), \end{cases} \quad (2)$$

where $|\mathcal{O}_{\pm}\rangle$ is defined as $\sum_{i=1}^{2N-1}(\hat{S}_i^+ \cdot \hat{S}_{i+1}^- + \hat{S}_i^- \cdot \hat{S}_{i+1}^+)(|N_1\rangle \pm |N_2\rangle)$, and c_1, c_2, O_1, and O_2 are coefficients to be determined. Actually, our exact diagonalization results show that when $2NS$ is even, the ground state G is $|+\rangle$ and the first excitation E_1 is $|-\rangle$; and when $2NS$ is odd, we have G=$|-\rangle$ and E_1=$|+\rangle$. This is in accordance with the theoretical results obtained by spin coherent state path integral[26, 27]. The higher excitation states with E_i ($i \geq 2$) can be constructed in the similar way.

2.2 Quantum dynamics of Néel states

We shall mainly focus on the subspace of the states with $S^z = 0$ because the ground state and the low excitation states including the Néel states belong to this subspace. For convenience, we shall use $|g\rangle$ and $|e_i\rangle$ ($i \geq 1$) to denote all the eigenstates in the $S^z = 0$ subspace. Because this subspace is closed under the Hamiltonian (1), the time evolution of the two Néel states can be expanded as

$$|\tilde{N}_a(t)\rangle = f_0^a e^{iE_0 t/\hbar}|g\rangle + \sum_{j\geq 1} f_j^a e^{iE_j t/\hbar}|e_j\rangle, \quad (3)$$

where E_j and f_j^a ($j \geq 0$, a =1,2) are the eigenvalues and expansion coefficients of the j-th eigenstates. Here, of course, we have $|\tilde{N}_a(0)\rangle = |N_a\rangle$, $|g\rangle$=G, and $|e_1\rangle$=E_1. Then, the weight of $|N_a\rangle$ in $|\tilde{N}_a(t)\rangle$ can be expressed as

$$\chi_a^2(t) = |f_0^a + \sum_{j\geq 1} f_j^a e^{i\Delta E_j t/\hbar}|^2, \quad (4)$$

where $\Delta E_j = E_j - E_0$. The total Néel weight of $|N_1\rangle$ and $|N_2\rangle$ in $|\tilde{N}_1(t)\rangle$ can be defined as $W_N(t) = \chi_1^2(t) + \chi_2^2(t)$. $W_N(t)$ reflects how well the Néel states describe the quantum antiferromagnetic chain. The two-state approximation results in a simplified expansion of $|\tilde{N}_a(t)\rangle$, such as

$$|\tilde{N}_1(t)\rangle \propto \cos(\frac{\Delta E_1}{2\hbar}t)|N_1\rangle + \sin(\frac{\Delta E_1}{2\hbar}t)|N_2\rangle. \quad (5)$$

We present $\chi_1^2(t)$ and $W_N(t)$ in Fig. 3 for D/J=10 , 3, and 1. For $\chi_1^2(t)$, the two-state approximation is also presented for comparison. It is clear that $\chi_1^2(t)$ is a

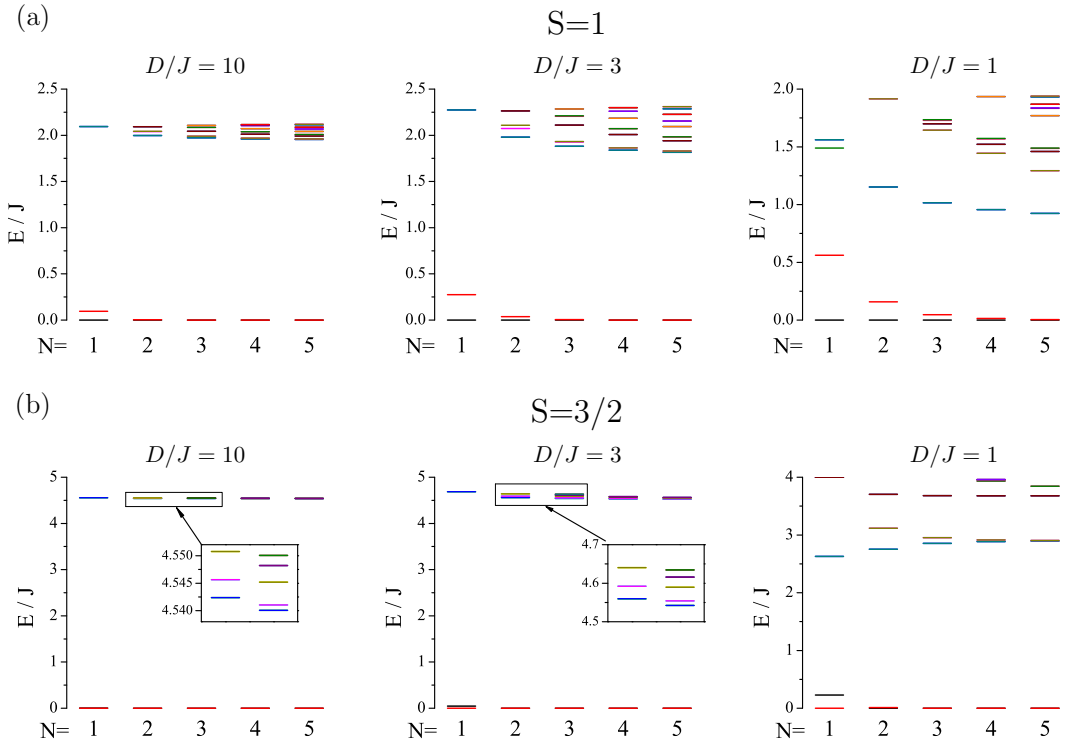

FIG. 2. The eigen energies (E/J) of Hamiltonian (1) for $S = 1$ (a) and $S = 3/2$ (b), with $D/J = 10$, 3, and 1.

TABLE I. The N-dependent energy gap ΔE between the ground state G and the first excitation E_1 for two spin values (1 and 3/2) and three D/J (1, 3, and 10).

		$N=1$	$N=2$	$N=3$	$N=4$	$N=5$
$S=1$	$D/J=1$	0.561	0.158	4.76×10^{-2}	1.43×10^{-2}	4.32×10^{-3}
	$D/J=3$	0.275	3.66×10^{-2}	4.95×10^{-3}	6.68×10^{-4}	9.03×10^{-5}
	$D/J=10$	9.48×10^{-2}	4.41×10^{-3}	2.05×10^{-4}	9.56×10^{-6}	4.46×10^{-7}
$S=3/2$	$D/J=1$	0.228	1.24×10^{-2}	6.64×10^{-4}	3.56×10^{-5}	1.90×10^{-6}
	$D/J=3$	4.53×10^{-2}	4.44×10^{-4}	4.34×10^{-6}	4.25×10^{-8}	4.15×10^{-10}
	$D/J=10$	5.09×10^{-3}	5.69×10^{-6}	6.36×10^{-9}	7.96×10^{-12}	8.81×10^{-15}

periodic function of t and $W_N(t)$ is almost a constant except a narrowly oscillating noise due to the higher states. For small D/J such as 1, the maximal value of $\chi_1^2(t)$ is approximately 0.8 and the Néel weight $W_N(t)$ is less than 0.9, but for large D/J such as 10, $\chi_1^2(t)$ can be well described with $\cos^2(t/2T)$ and the Néel weight becomes larger than 0.99. Here, the time period is equivalent to $P = 2\pi T$, and $1/T$ reflects the switching rate (or frequency) between the two Néel states. It is surprising that for this case of $S = 2$ and $N = 2$, T increases by five orders of magnitudes when D/J changes from 1 to 10.

2.3 Life times of Néel states

Because $\chi_1^2(t)$ is a well-defined periodic function of t, the quantity T, the time spent by a switching circle between the two Néel states, can be used to characterize the life times of the Néel states. In the case of two-spin chains ($N = 1$) with $S \leq 3$, we can calculate eigenstates and $|\tilde{N}_a(t)\rangle$ exactly. For $S = 1$, we obtain $\Delta E_1 = J[\sqrt{4(D/J)^2 + 4D/J + 9} - 2D - 1]/2$, and T can be expressed as $(2D/J + 1)/2$ when D/J is large. For higher spins, we can achieve $T \propto (2D/J + 1)^{2S-1}$ for both integer and half odd integer spins by using a usual perturbation method. Generally speaking, we can also use exact diagonalization method to calculate ΔE and T for arbitrary S and N. In Fig. 4 we present the calculated T as functions of D/J for $N = 1$, with S taking nine values from 1 to 5. In Fig. 5 we present our accurate calculated T curves for N=1 through 4 and S=1, 3/2, 2, and 5/2.

It is very interesting that all these (D/J)-T curves can be satisfactorily fitted with one simple function,

$$T = A\frac{\hbar}{J}(2\frac{D}{J} + 1)^{N(2S-1)} \qquad (6)$$

where A is a constant depending on S and N only. It

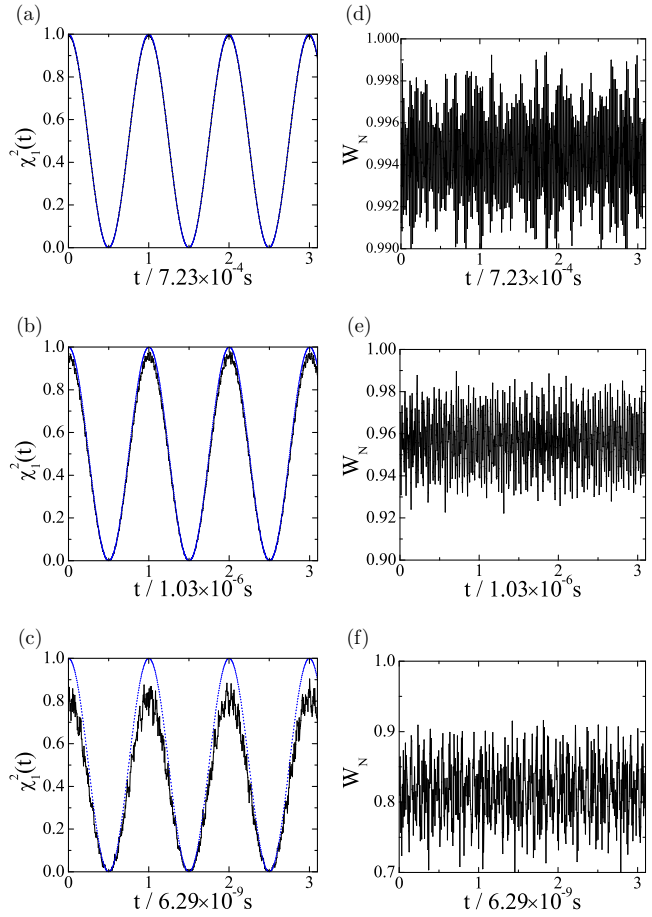

FIG. 3. The time dependence of $\chi_1^2(t)$ (black lines in the left column) and $W_N(t)$ (the right column) for $S = 2$, $2N = 4$, and $D/J = 10$ (a,d), 3 (b,e), and 1 (c,f), respectively. The two-state approximated results of $\chi_1^2(t)$ (blue dash lines in the left column) are also presented for comparison.

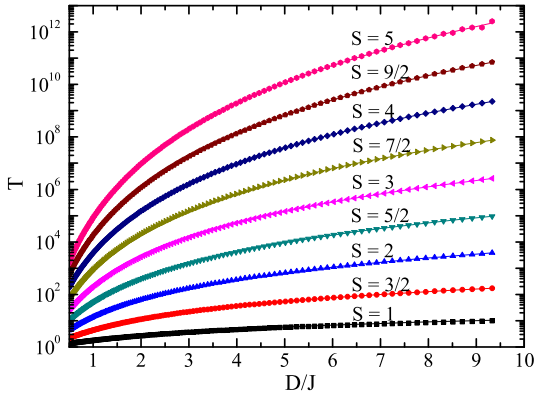

FIG. 4. The life times T (in unit of $\hbar/J$) of the two-spin chains as functions of D/J for nine S values, fitted with Eq. (5).

TABLE II. The N-dependent Néel weights in the ground state (G) and the first excitation (E_1) for different S and D/J.

			$N=1$	$N=2$	$N=3$	$N=4$	$N=5$
$S=1$	$D/J=1$	G	0.864	0.750	0.693	0.634	0.575
		E_1	1.000	0.854	0.735	0.650	0.580
	$D/J=3$	G	0.964	0.935	0.916	0.892	0.867
		E_1	1.000	0.958	0.921	0.893	0.868
	$D/J=10$	G	0.996	0.991	0.988	0.985	0.980
		E_1	1.000	0.994	0.989	0.985	0.980
$S=3/2$	$D/J=1$	G	0.900	0.875	0.822	0.770	0.720
		E_1	0.968	0.882	0.823	0.770	0.720
	$D/J=3$	G	0.985	0.946	0.957	0.941	0.926
		E_1	0.991	0.973	0.957	0.941	0.926
	$D/J=10$	G	0.999	0.995	0.994	0.992	0.991
		E_1	0.999	0.996	0.994	0.992	0.991

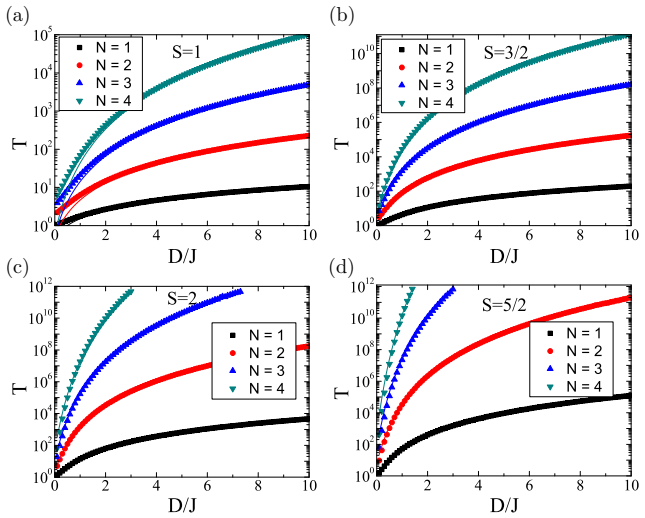

FIG. 5. The life times T (in unit of $\hbar/J$) of the $2N$ chains as function of D/J for $S = 1$ (a), $S = 3/2$ (b), $S = 2$ (c), and (d) $S = 5/2$, fitted with Eq. (5).

is surprising that, as we show in Fig. 6, A can be well fitted with $A = ba^N$, and furthermore the parameters a and b can be well fitted with $a = 0.2427 \times 4.1545^S$ and $b = 0.5007 \times S^{-2.0713}$. The fitted data of A, a, and b are summarized in Table III. Consequently, we obtain a unified expression for T as functions of D, J, N, and S. It can be used to extrapolate T with given D and J for higher S and larger N. It should be pointed out that although T increases with increasing D or decreasing J, too small J will be harmful to stability against thermal fluctuations. Although T increases exponentially with N increasing, one cannot use too long spin chains for practical information storage because the Néel weight will decrease with N increasing. Therefore, for a practically useful system, one should keep a balance between a large T and a good stability of the Néel states.

TABLE III. Fitted results of A, a, and b for the antiferromagnetic spin-S chains including $2N$ spins.

Spin	A				a	b
	N=1	N=2	N=3	N=4		
S=1	0.506	0.519	0.531	0.542	1.023	0.4952
S=3/2	0.445	0.909	1.839	3.725	2.029	0.2202
S=2	0.500	2.039	8.527	36.47	4.178	0.1183
S=5/2	0.640	5.245	47.49	371.9	8.415	0.07596
S=3	0.894	15.74	289.2	4871	17.68	0.05080

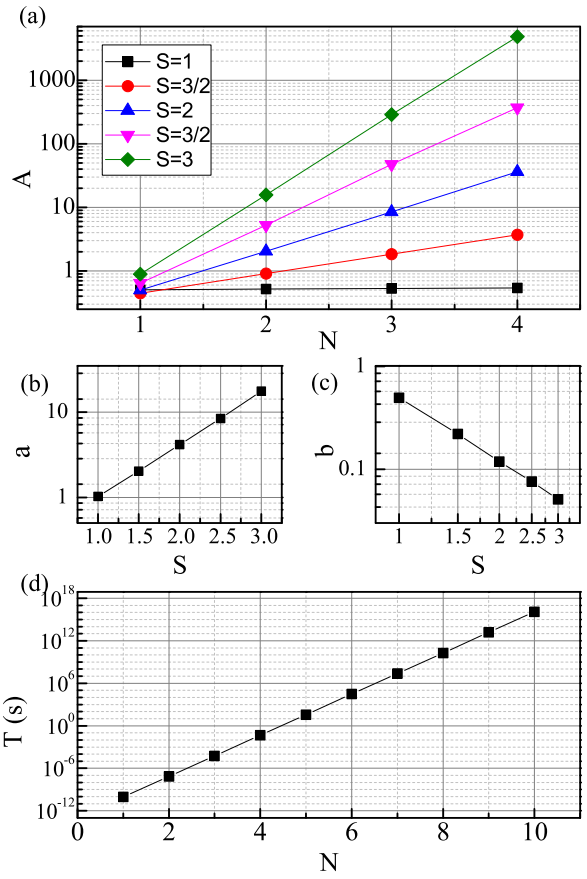

FIG. 6. (a) Fitting of the numeric results of A depending on N for different S; (b) fitting of a depending on S; (c) fitting of b depending on S; and (d) Plot of the life time T (in second) depending on N defined in Eq. (6) for the Fe adatom spin chains on Cu_2N surface.

2.4 Long life times in real adatom spin chains

For the short antiferromagnetic chains of Fe adatom spins on Cu_2N surface, experimental result reveals that $S = 2$, $J = 0.7$ meV, and $D = 1.87$ meV [5]. In this case, we have $D/J = 2.67$, belonging to the regime of strong uniaxial anisotropy. As a result, we obtain a simple formula of T (in second) depending on N,

$$T = 1069^N \times 1.120 \times 10^{-13}. \quad (7)$$

We plot it in Fig. 6(d). The expression (6) implies that the switching rate $(1/T)$ will be decreased approximately by 1000 times when we add two more Fe-adatom spins to the chain, which is consistent with the low-temperature limit (with external thermal effects being frozen) of the experimental results[3].

This implies that the life times T can be very long, reaching 1.9 days, 5.7 years, and 6057 years when N is equivalent to 6, 7, and 8, respectively. This theoretical trend is also consistent with the experimental result of the Fe-adatom spin system[3]. These results show that for such antiferromagnetic chains, 14 or 16 spins (for $2N$) should be enough to achieve stable Néel states for practical information storage.

On the other hand, for practical usage, we need to consider other factors affecting the life times of Néel states. First, we consider possible transverse single-ion anisotropy E and transverse magnetic field B_x which appear as additional $\sum_i \{E[(\hat{S}_i^x)^2 - (\hat{S}_i^y)^2] + \gamma B_x \hat{S}_i^x\}$ in the Hamiltonian. Our calculations reveal that as long as D/J is not less than 1, there is little change in T even when E/J and $\gamma B_x/J$ reach to 0.2. Then, we investigate effect of spin exchange anisotropy on T, showing that the effect is very small for $D/J > 1$. Therefore, our T results are robust and technically sound.

3. Conclusions

In summary, we have investigated the intrinsic quantum dynamics and life times of Néel states in the quantum Heisenberg antiferromagnetic chains with strong uniaixal single-ion anisotropy. For typical values of spin, chain length, and magnetic anisotropy, we have used exact diagonalization method to accurately calculate eigenvalues and eigenfunctions, and shown rigorously that the ground state and the first excitation are both safely separated from the other states and can be well described with the two Néel states as long as both D/J and S are large enough and N is not too large. Through investigating accurate time evolution of the Néel states, we have determined their switching rates and life times. Surprisingly, we have achieved a unified powerful expression of the life times for arbitrary values of N, S, D, and J. Furthermore, we show that for the Fe-adatom spin antiferromagnets on Cu_2N semiconductor surface[1–3], $2N$= 14 or 16 is large enough to achieve practically long life times for the Néel states. These theoretical results should be useful to help realize the Néel state storage of information and atomic-scale antiferromagnetic spintronics.

Acknowledgments

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