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Spin–singlet competition responsible for magnetization plateau associated with magnetocaloric effect in a tetrameric chain

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HIGHLIGHTS

- The quantum phase transitions and magnetocaloric effect (MCE) of a tetrameric chain are investigated by Green's function theory.
- The spin–singlet competition was proposed to unpuzzle the magnetization plateau.
- The scaling behavior of inverse MCE was unveiled.

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ABSTRACT

The quantum phase transitions (QPTs) and magnetization plateau properties, together with magnetocaloric effect (MCE) of the tetrameric chain are investigated by means of Green's function theory. We reveal the uniform, dimeric and gapped (gapless) tetrameric phases, which are explicitly confirmed by the field dependence of magnetization. The spin–singlet competition is proposed to make clear the magnetization plateaus and QPTs. Simultaneously, the QPTs and quantum critical points are identified by the dips of the isoentropes, or equivalently the sharp maxima of entropy at ultra-low temperature, as well as the local minima of specific heat and the valley–peak structure of magnetic cooling rate with its sign changed. In addition, the temperature (T) and magnetic field (h) dependence of magnetic entropy change (ΔS) exhibits prominent inverse magnetocaloric effect (IMCE), implying adiabatic magnetization can generate cooling, which follows a power law dependence of h: $\Delta S \sim h^n$. The local exponent $n \approx 2$ is independent of h and T at low fields, which was observed in the antiferromagnetic materials experimentally. It is also found that the stronger the dimerization is, the larger the IMCE is, which would provide a clue to design antiferromagnetic refrigerant materials for use in magnetic cooling devices with low field controlling.

1. Introduction

In recent years, the quantum behavior of low-dimensional quantum magnets has been one of the most fascinating topics at the border of condensed matter physics, materials science and spin chemistry [1–18]. Probing the physics of magnetism has served to heighten its interest for theorist and experimentalist [3–18]. An important focus on the behavior of magnetic materials at a quantum critical point (QCP) has been evolved, reflecting critical behavior arising from quantum fluctuations, which may push a system into a different state [1,2,19]. A quantum phase transition (QPT) occurs at zero temperature upon

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tuning magnetic field or other control parameters to a critical value, which tilts the balance among the competing ground states [20]. Besides, the competition between two different ground states at a QCP results in unusual thermodynamic behaviors.

It has been shown that the magnetocaloric effect (MCE) is particularly suited to detect the quantum critical behavior, as it is more singular than specific heat close to the QCP, thus, it can be used to check the very existence of QCP [21–25]. On the one hand, at ultra-low temperature, the dips of the isoentropes, or equivalently the sharp maxima of entropy, as well as the valley–peak structure of magnetic cooling rate with its sign changed clearly demonstrated the quantum criticality. On the other hand, at finite temperature, the MCE has been studied actively for its potential application in magnetic refrigeration, which should be emphasized to understand how the magnetocaloric properties of the working substance evolve with temperature and magnetic field [26–30]. Hence, the field induced magnetic entropy change (ΔS) [26–33] is an important parameter that governs the magnetocaloric properties. In general, the application of a magnetic field causes reduction of magnetic entropy of refrigerant materials, suggesting the adiabatic demagnetization can generate cooling, which is called conventional MCE (CMCE) [26–30]. However, an inverse effect occurred in some magnetic materials has been revealed in recent studies, namely, a magnetic field causes enhancement of magnetic entropy, the phenomenon of which is termed as inverse MCE (IMCE) [24,34–40]. Such IMCE was mostly observed in antiferromagnetic and ferrimagnetic materials [35,36,40], the magnetic cooling for which can be achieved via adiabatic magnetization process. It was shown that integrating IMCE materials and CMCE ones can improve the cooling efficiency of a refrigeration device. Therefore, the field dependence of MCE that can be parametrized by critical exponent governing the transition, can not only be helpful for deeper understanding the magnetic phase transitions [19,21,35,36] and phase coexistence [37] in magnetic materials, but also provide the information on the operating field ranges in actual refrigerant cycle.

Especially, one-dimensional quantum spin chains with competing interactions are known to present exotic physical properties [5–13,16]. One of the most intriguing features is the magnetization plateau [5–7,13,16]. It is worth noting that, Oshikawa, Yamanaka and Affleck [41] who extended the Lieb–Schultz–Mattis [42] theorem, proposed a necessary condition $n(S-m)=$ integer for the appearance of magnetization plateau in spin chains, where n is the period of ground state, S is the magnitude of spin, and m is the magnetization per site. It is well-known that, no magnetization plateau appears until saturation in the spin-1/2 uniform antiferromagnetic chain [9,10], the ground state of which can be described by a Tomonaga–Luttinger liquid (TLL) with fermionic spin-1/2 spinon excitations [43]. Hence, the competition between magnetic interactions has attracted much attention and resulted in unconventional quantum phases. For instance, neglecting the weak interchain coupling, the malachite $Cu_2(OH)_2CO_3$ can be modeled as a bond-alternating (dimeric) spin chain [12], in which a zero magnetization plateau with a large energy gap was observed. The trimer chain (J_1–J_2–J_2) compound $Cu_3(P_2O_6OD)_2$ exhibited a 1/3 plateau with gapless low-lying excitations [13]. Owing to the lack of experimental realization, more complicated quantum spin systems with multiple exchange interactions have not been sufficiently studied. Very recently, an organic tetrameric chain (J_1–J_2–J_3–J_2) compound $\beta-2$, 6–Cl_2–V has been synthesized [16], wherein the zero and 1/2 plateaus were unveiled, and low field low temperature TLL was also revealed. In addition, at low field, the enhancement of entropy was observed, implying that IMCE may appear. Thus, the study of the quantum criticality and MCE of low-dimensional spin systems, particularly the exactly soluble, is of great importance for understanding the magnetic and thermodynamic properties of these materials. To the best of our knowledge, the one-dimensional *XY* model which was introduced by Lieb et al. [42], continues to provide new information on the quantum behavior of magnetic systems. It is still the best one to feature exactly magnetic phase transitions. It was clearly demonstrated by de Lima et al. [5–7], who presented rather rich quantum critical behaviors using the anisotropic and isotropic *XY* model. Theoretically, the spin-1/2 *XY* chain can provide an excellent ground for rigorous study of various properties of quantum magnetic systems. Performing Jordan–Wigner transformation [42,44], the model can be mapped onto a system of noninteraction spinless fermions, in which the quantum correlation and thermodynamic quantities can be calculated exactly beyond the rough random phase approximation (RPA) [45] by Green's function theory.

In this paper, we will consider the isotropic *XY* model in a magnetic field on the tetrameric chain, which was addressed by Yamaguchi et al. [16], who restricted their analysis to the study of thermodynamic properties experimentally. The aim of this work is to present the magnetization plateau nature, the quantum criticality and MCE of the tetrameric chain. The remainder of the paper is organized as follows. In the forthcoming section, we present the model Hamiltonian and give an outline of Green's function method. In Section 3, the thermal Drude weight, the magnetization, the magnetic cooling rate, the entropy and the magnetic entropy change are calculated; the quantum phase diagram and scaling behavior will be explored; and a comparison with the experimental results will be made. Finally, we draw a conclusion in Section 4.

2. Model Hamiltonian and Method

We take into account the tetrameric chain illustrated in Fig. 1(b) with each molecule ($M_1(M_1')$ and $M_2(M_2')$) carrying $S=1/2$, as a one-dimensional isotropic *XY* model with the superlattice consisting of four effective spin-1/2 sublattices, the Hamiltonian of which in an external magnetic field can be written as

$$H=\sum_l\left[\frac{J_1}{2}S_{1,l}^+S_{2,l}^-+\frac{J_2}{2}\left(S_{2,l}^+S_{3,l}^-+S_{4,l}^+S_{1,l+1}^-\right)+\frac{J_3}{2}S_{3,l}^+S_{4,l}^-+H.c\right]-g\mu_BB\sum_l\sum_{i=1}^4S_{i,l}^z, \quad (1)$$