



ELSEVIER

Available online at www.sciencedirect.com

SCIENCE @ DIRECT®

Journal of Magnetism and Magnetic Materials 305 (2006) 253–258

www.elsevier.com/locate/jmmm

Thermodynamic properties of mixed-spin chains in magnetic field by the transfer matrix method

H.H. Fu^{a,*}, K.L. Yao^{a,b}, Z.L. Liu^a^aDepartment of Physics and State Key Laboratory of Laser Technology, Huazhong University of Science and Technology, Wuhan 430074, China^bThe International Center of Materials Physics, Chinese Academic of Science, Shenyang 110015, China

Received 8 September 2005; received in revised form 29 November 2005

Available online 25 January 2006

Abstract

Motivated by recent experimental synthesis of molecule-based ferrimagnetics, we have investigated the thermodynamic properties of one-dimensional antiferromagnetic–ferromagnetic mixed spin- $\frac{1}{2}$ lozenge chain in external magnetic fields at low temperature, using the transfer matrix method. The magnetization and the specific heat of the spin system have been evaluated numerically from the free energy. The magnetization as a function of the magnetic field at low temperature shows step-like plateau characterized by three critical magnetic fields, which reflects the competing effect of magnetic interactions and thermal fluctuations. The double-peak structure in the curves of the specific heat is also found, which comes from the huge variation of the long-range order parameters. Our results show that the thermodynamic properties of the mixed spin- $\frac{1}{2}$ molecule-based ferrimagnetics are very analogous to the behaviors of the spin-1 and spin- $\frac{1}{2}$ mixed antiferromagnetic chain.

© 2006 Elsevier B.V. All rights reserved.

PACS: 64.40.+g; 74.25.Bt; 74.25.Ha

Keywords: Thermodynamics; Molecule-based ferrimagnetics; Transfer matrix method

1. Introduction

Molecule-based magnets and other molecular functional magnetics have received great interest in recent years [1–3]. After the discovery of the first genuinely organic ferromagnet with $T_C = 0.6$ K in 1991 [4], searching for new organic ferromagnetism exhibiting spontaneous magnetization at higher temperature is one of today's challenging targets. Ferrimagnetism has long been considered as an effective strategy to give organic materials spontaneous magnetizations by the antiferromagnetic intermolecular interactions, which would bring about antiparallel spin alignment between neighboring molecules with different magnetic moments as proposed from the theoretical works [5–7]. However, the magnetic phase transitions to a

ferrimagnetic ordered state have not been documented so far in organic molecular crystalline solids.

An experimental breakthrough work for molecular-based magnets was made by Izuoka and co-workers in 1994 [8], who synthesized a molecular complex of a ground-state triplet $S = 1$ biradical and an $S = \frac{1}{2}$ monoradical. However, they have not observed a phase transition to a bulk ferromagnetic-ordered state. It is worth noting that, recently, Hosokoshi et al. [9] adopted a structure similar to that of Izuoka et al. and claimed to succeed in making for the first time a purely organic molecule-based ferrimagnet by using a single-component strategy. They utilized a triradical including $S = 1$ and $S = \frac{1}{2}$ units within a molecule connecting the $S = 1$ and $S = \frac{1}{2}$ units by intramolecular and intermolecular antiferromagnetic interactions. This material undergoes a three-dimensional (3D) phase transition at low temperature down to 0.28 K. These works have stimulated us to study the quantum magnetic transition and the thermodynamic

*Corresponding author. Tel.: +86 278 755 7834; fax: +86 278 755 6264.
E-mail address: fuhust547@sohu.com (H.H. Fu).

properties of the organic ferrimagnets from a theoretical point of view.

The above mentioned purely organic-molecule-based ferrimagnet has been considered as a novel type of mixed-spin or hetero-spin chain theoretically [10], and studied by the Monte Carlo method [11] and transfer matrix renormalization group (TMRG) method [12]. As for low-dimensional system, the most fascinating characteristic is that they show magnetization plateaus, i.e., quantization of magnetization at low temperature near the ground state. Furthermore, when temperature is low enough, the spin correlation and the thermodynamic fluctuation are remarkable, which would develop short-range order or long-range order under certain external conditions. Meanwhile, the order parameters corresponding to these ordered states can be observed experimentally by measurement of the specific-heat. The collapse or formation of one kind of order corresponds to a peak in the specific-heat curve. Therefore, the spin correlation and thermodynamic fluctuation play an important role in the magnetic properties at low temperature for low-dimensional molecule-based ferrimagnetisms. Considering these cases, we develop the transfer matrix method by finite-size scaling, which has become increasingly important in the study of critical phenomena [13] to investigate the thermodynamic and magnetic behavior of the phenomenological ferrimagnetic biradical–monoradical alternating chain at finite temperature [14]. The remainder of this paper is organized as follows. The model and method are given in Section 2. In Section 3, the magnetization and the specific heat of spin system are calculated and discussed. Conclusions are given in Section 4.

2. Theoretical model and numerical method

Compared with transition-metal-based molecular ferrimagnets [15], organic-molecule-based magnetism is characterized by the following features. In organic-molecule-based magnetic materials, spin density is distributed over many atomic sites in an open-shell molecule, and hence, the intermolecular spin–spin interaction has a multicentered or multicontact nature. Furthermore, in some cases, intramolecular interactions in stable organic molecules are in the

same order of magnitude as the intermolecular interactions in crystalline solid states. In these cases, the magnetic degree of freedom for $S > \frac{1}{2}$ molecules and the spatial symmetry of intermolecular interaction should be taken into account when considering the magnetic properties of molecular assemblages containing $S > \frac{1}{2}$ molecules. The phenomenological spin Hamiltonian with Ising-like interactions in the presence of an applied static magnetic field H , is given by

$$H = \sum_{i=1}^N \left[J_1 \left(S_{i,b1}^z \cdot S_{i,m}^z + S_{i-1,m}^z S_{i,b1}^z \right) + J_2 \left(S_{i,b2}^z \cdot S_{i,m}^z + S_{i-1,m}^z S_{i,b2}^z \right) + J_3 S_{i,b1}^z \cdot S_{i,b2}^z - H \left(S_{i,m}^z + S_{i,b1}^z + S_{i,b2}^z \right) \right], \quad (1)$$

where $S_{i,b1}^z$, $S_{i,b2}^z$ and $S_{i,m}^z$ denote the spin- $\frac{1}{2}$ operators and N is the number of unit cells. Hamiltonian (1) is illustrated in Fig. 1. $S_{i,b1}^z$ and $S_{i,b2}^z$ are coupled by the intramolecular ferromagnetic exchange interaction, $J_3 < 0$, to give a molecular site of biradical or the two-spin site. The biradical spins are coupled with a neighboring monoradical spin of $S_{i,m}^z = 1/2$ by the antiferromagnetic exchange interactions $J_1 > 0$ and $J_2 > 0$. This is the simplest model Hamiltonian for molecular ferrimagnets possessing the above features. The last term in Eq. (1) represents the Zeeman energy of the spins. To confront the experimental findings on the molecule-based ferrimagnet, we set $J_1 = 0.1$, $J_2 = 0.2$ and $J_3 = -1.0$ in our calculations. It should be mentioned that the effects of the various parameter ratios of J_1 , J_2 and J_3 on the thermodynamic properties have already been reported in our earlier publication [14], to which the interested reader is referred for more studied details. In this paper, we focus on the effects of external magnetic fields on the thermodynamic properties of the ferrimagnetic mixed-spin system.

The quantum transfer matrix is widely used to study the one-dimensional spin system at finite temperature numerically. In above model, for any site we define a spin variable $S(r, i) = \pm 1/2$, $r = (m, b1, b2)$, with the periodic boundary condition as

$$S_{N,m}^z = S_{1,m}^z, \quad S_{N,b1}^z = S_{1,b1}^z \quad \text{and} \quad S_{N,b2}^z = S_{1,b2}^z. \quad (2)$$

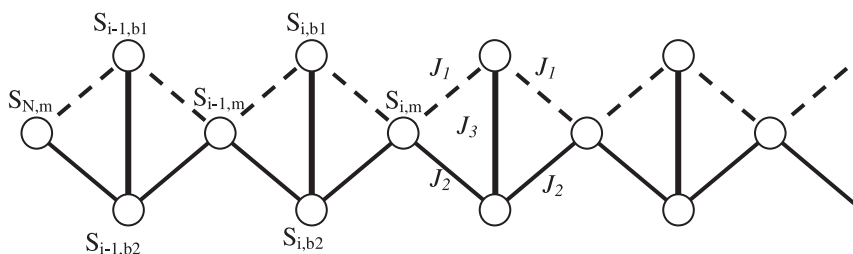


Fig. 1. The multicentered ferromagnetic chain as a model for organic molecule-based ferrimagnets. The biradical molecules with two unpaired electron spins interact with adjacent monoradicals.

Download English Version:

<https://daneshyari.com/en/article/1805061>

Download Persian Version:

<https://daneshyari.com/article/1805061>

[Daneshyari.com](https://daneshyari.com)