Quantum magnetization plateau of an anisotropic mixed spin chain

T Sakai, T Tonegawa and K Okamoto

1Japan Atomic Energy Agency (JAEA), Spring-8, Hyogo 679-5148, and Crest JST, Japan
2Department of Mechanical Engineering, Fukui University of Technology, Fukui 910-8505, Japan
3Department of Physics, Tokyo Institute of Technology, Tokyo 152-8551, Japan
E-mail: sakai@spring8.or.jp

Abstract.

The magnetization process of the $S = 1$ and $S = 2$ spin alternating ferrimagnetic chain with the single-ion anisotropies is investigated by the numerical exact diagonalization and the density matrix renormalization group method. It is found that the mechanism of the 2/3 magnetization plateau formation due to quantum effects depends on the anisotropy constants. The obtained phase diagram of the 2/3 plateau is revealed to include three different phases.

1. Introduction

The nanowire magnet is one of the most interesting subjects in the field of low-dimensional magnets. The single chain quantum magnet [1, 2] consisting of Ni$^{2+}$ and Mn$^{3+}$ has been investigated extensively as a good candidate of useful nano-scale magnets. As a variation of the single chain magnets, recently the $S = 1$ and $S = 2$ spin alternating compound [Mn(Cl$_4$saltmen)Ni(pao)$_2$(bpy)]PF$_6$ has been synthesized. This mixed spin chain should have a ferrimagnetic ground state, which would appear as a 1/3 plateau in the magnetization curve. The ferrimagnetic feature will be discussed in another work [3]. Besides the ferrimagnetic state, the mixed spin system was predicted to exhibit another magnetization plateau at 2/3 of the saturation moment [4, 5], as a quantum effect. On the other hand, the above compound has large Ising-like single-ion anisotropies, which would possibly change magnetic properties. In the present paper, we focus on the mechanism of the 2/3 magnetization plateau in the presence of such single-ion anisotropies, using the numerical diagonalization and density matrix renormalization group methods, to obtain a phase diagram at 2/3 of the saturation magnetization.

2. Model

In order to describe the magnetic properties of the above material, we consider the $S = 1$ and $S = 2$ spin alternating Heisenberg chain with the single-ion anisotropies $D_1$ and $D_2$ for the sites of Ni$^{2+}$ and Mn$^{3+}$, respectively. The Hamiltonian is given by

$$H = \sum_j \{ s_j \cdot S_{j+1} + S_{j+1} \cdot s_{j+2} \} + \sum_j \{ D_1 (s_j^z)^2 + D_2 (S_{j+1}^z)^2 \} - H \sum_j \{ s_j^z + S_{j+1}^z \}, \quad (1)$$
The arrow (the bullet symbol) denotes an S=1/2 spin with its fixed (unfixed) projection value. Two S=1/2 spins in an ellipse form a singlet pair. At the (⟨s^z⟩, ⟨S^z⟩)=(1, 1)-type plateau two S=1/2’s in S=2 form the S^z_{tot} = 0 component of the triplet state.

where j is summed over j = 1, 3, · · · , N − 1, and N is the total number of spins in the system. sj and S_{j+1} are the S = 1 and S = 2 operators, respectively. H is the external magnetic field. We denote by M the z-component of the total spin, M = \sum_j (s^z_j + S^z_{j+1}), and also by M_s the saturation magnetization, M_s = 3/2N. We perform the numerical diagonalization and density matrix renormalization group calculations in a wide range of D_1 and D_2 to study the mechanism of the 2/3 magnetization plateau and obtain the phase diagram at M = (2/3)M_s.

3. Mechanism of the 2/3 plateau

The valence bond solid (VBS) picture [7, 8] is useful to explain the mechanism of field-induced spin gap, namely the magnetization plateau. It describes S = 1 and S = 2 as composite spins consisting of two and four S = 1/2’s, respectively. The previous work [4, 5] using the numerical diagonalization and the finite-size scaling analysis revealed that the 2/3 plateau described by the VBS shown in figure 1 (a) appears at the isotropic point (D_1, D_2)=(0,0) of the present system. It implies that the 2/3 plateau is formed by the quantum effect. However, varying D_1 and D_2 the mechanism of the 2/3 plateau is expected to be changed[6]. First, another mechanism described by figure 1 (b) should be realized at least in the limit D_2 → +∞. Second, the other mechanism accompanied with a spontaneous translational symmetry breaking described by figure 1 (c) is expected in the Ising limit, namely D_1, D_2 → −∞. Thus the three types of the 2/3 plateau can occur. In the next section we will determine phase boundaries among the three phases in the (D_1, D_2) plane, to confirm that these phases really exist.

4. Phase diagram of the 2/3 plateau

The (1/2, 3/2)-type plateau includes a singlet bond between S = 1 and S = 2 at every two unit cells, while the (1,1)-type one does not. Thus the boundary between the two plateau phases can be determined by the twisted boundary condition level spectroscopy method [9] which detects a singlet bond on the twisted coupling. We apply this method for the low-lying energy eigenvalues calculated by the numerical diagonalization of finite-size clusters up to N = 16 to obtain the boundary as solid circles in figure 2. The phase boundary between the (1/2, 3/2) and (1,1) plateaux should be Gaussian.

The (1,2,-1,2)-type plateau phase, where the spontaneous double periodicity appears, continues to the Ising limit. Thus the quantum phase transition towards the phase is expected to belong to the Ising universality class. In such a case the phenomenological renormalization [10] is a good method to determine the phase boundary. To estimate the critical value D_{2c} for fixed D_1 by this method, we should numerically solve the N dependent fixed point D_{2c}(N) defined by the form,

\[ (N + 4)\Delta_{N+4}(D_1, D_{2c}) = N\Delta_N(D_1, D_{2c}), \]  

and extrapolate it to the thermodynamic limit N → ∞. The D_2 dependence of the scaled gap N\Delta for D_1 = −1 is shown in figure 3. It suggests that the fixed point (the cross point in figure

Figure 1. Schematic representations for (a) the (⟨s^z⟩, ⟨S^z⟩)=(1/2, 3/2)-type, (b) the (⟨s^z⟩, ⟨S^z⟩)=(1, 1)-type and (c) the (⟨s^z⟩, ⟨S^z⟩)=(1, 2, −1, 2)-type 2/3-plateau state.
Figure 2. The $\frac{2}{3}$-plateau phase diagram on the $D_2$ versus $D_1$ plane. Solid circles are the phase boundary between the $(1/2,3/2)$ and (1,1)-type plateau phases, obtained by the twisted boundary condition level spectroscopy. Solid squares are the phase boundary between the (1,1) and (1,2,-1,2)-type plateau phases, obtained by the phenomenological renormalization.

Figure 3. Scaled gap for $D_1 = -1$ is plotted versus $D_2$ for $N = 8$, 12 and 16. The cross point between $(N + 4)\Delta_{N+4}$ and $N\Delta_N$ gives the $N$ dependent fixed point $D_{2c}(N)$. Since the $N$ dependence is quite small, we use $D_{2c}(N)$ obtained from the largest system size ($N = 12$) as the critical point $D_{2c}$.

3) seems almost independent of $N$. Thus we determine $D_{2c}$ from the largest-$N$ result up to $N = 16$. The estimated $D_{2c}$ for various values of $D_1$ is plotted as the phase boundary of the (1,2,-1,2)-type plateau phase in figure 2 (solid squares). It indicates that this plateau phase adjoins only the (1,1)-type one.

Within the present analyses of the phase boundary another possible 2/3 plateau mechanism (0,2) is not found, although it is expected to appear in the large-$D_1$ region. If it appears, it would adjoin the (1/2,3/2)-type plateau phase. In order to confirm the mechanism of the 2/3 plateau, we calculate the expectation values of $s^z$ and $S^z$ at each site using the density matrix renormalization group method applied for 84-spin system under open boundary condition. The obtained result for $D_1 = 5$ and $D_2 = 2$ is shown in figure 4. It clearly shows that the plateau is still (1/2,3/2)-type even in such a large-$D_1$ region. This result is also a direct evidence to confirm that the (1/2,3/2)-type 2/3 plateau really exists and it is quite stable against large $D_1$. 
Figure 4. Expectation values of $s^z$ and $S^z$ at each site for $D_1 = 5$ and $D_2 = 2$ calculated by the density matrix renormalization group method applied for $N = 84$ under the open boundary condition. It justifies that even in large-$D_1$ region the $2/3$ plateau is $(1/2,3/2)$-type, not (0,2)-type. A small deviation from $(1/2,3/2)$ is supposed to be a quantum effect.

5. Summary
The $2/3$ magnetization plateau of the $S = 1$ and $S = 2$ spin alternating chain is investigated in the presence of single-ion anisotropies, using the numerical diagonalization and the density matrix renormalization group method. Three types of plateaux are found on the $D_1$-$D_2$ plane, as shown in Fig.2. For the single chain magnet consisting of Ni$^{2+}$ and Mn$^{3+}$, $D_2$ was estimated by the ESR measurement [11] as $\sim -0.3$, but $D_1$ was difficult to be determined because its effect was too small on ESR. Since $D_1$ is expected to be smaller than $D_2$, the material is likely to lie in the $(1/2,3/2)$-type plateau phase. It would be interesting to measure the magnetization process of this material in high magnetic field.

Acknowledgment
We wish to thank T. Hikihara by whom the DMRG program employed in a part of this study is coded. This work has been partly supported by Grants-in-Aid for Scientific Research (B) (No. 17340100) and Scientific Research (C) (No. 16540329, No. 18540340) from the Ministry of Education, Culture, Sports, Science and Technology of Japan. We further thank the Supercomputer Center, Institute for Solid State Physics, University of Tokyo, the Information Synergy Center, Tohoku University, and the Computer Room, Yukawa Institute for Theoretical Physics, Kyoto University for computational facilities.

References
[1] Clerac R, Miyasaka H, Yamashita M and Coulon C 2002 J. Am. Cem. Soc. 124 12837
[2] Miyasaka H, Cléric R, Mizushima K, Sugiura K, Yamashita M, Wernsdorfer W and Coulon C 2003 Inorg. Chem. 42 8203
[3] Tonegawa T, Sakai T, Okamoto K and Kaburagi M 2006 this Proceedings
[4] Yamamoto S and Sakai T 2000 Phys. Rev. B 62 3795
[5] Sakai T and Yamamoto S 2000 J. Phys.: Condens. Matter 12 9787
[6] Sakai T and Okamoto K 2002 Phys. Rev. B 65 214403
[7] Affleck I, Kennedy T, Lieb E H and Tasaki H 1987 Phys. Rev. Lett. 59 799
[8] Affleck I, Kennedy T, Lieb E H and Tasaki H 1988 Commun. Math. Phys. 115 477
[9] Kitazawa A 1997 J. Phys. A: Math. Gen. 30 L285
[10] Nightingale M P 1976 Physica A 83 561
[11] Oshima Y, Nojiri H, Asakura K, Sakai T, Yamashita M and Miyasaka H 2006 Phys. Rev. B to appear.