Magnetization Plateau in the $S = 1$ Spin Ladder with Competing Interactions

Nobuhisa Okazaki$^a$,* Kiyomi Okamoto$^b$ and Tōru Sakai$^c$

$^a$Kyoto Miyama High School, Murashita, Sasari, Miyama-cho, Kitakuwata-gun, Kyoto 601-0705, Japan
$^b$Department of Physics, Tokyo Institute of Technology, Oh-okayama, Meguro-ku, Tokyo 152-8551, Japan
$^c$Tokyo Metropolitan Institute of Technology, Asahigaoka, Hino, Tokyo 191-0065, Japan

Very recently a non-trivial magnetization plateau at 1/4 of the saturation magnetization was observed in the $S = 1$ spin ladder BIP-TENO. In our previous work we proposed a possible mechanism of the plateau based on the second- and third-neighbor exchange couplings which lead to frustration. In order to confirm the realization of the mechanism, we compare the temperature dependence of the magnetic susceptibility and some critical magnetic fields obtained by the numerical calculation for the proposed model with the experimental results.

*Corresponding author:
Nobuhisa Okazaki
Email: nobuhisa@sci.himeji-tech.ac.jp
Fax: +81-771-77-0821
1 Introduction

A recent synthesized organic $S = 1$ spin ladder, 3,3',5,5'-tetrakis($N$-tert-butylaminoxyl) biphenyl, abbreviated as BIP-TENO \cite{1}, is one of interesting strongly correlated electron systems. It exhibits a field-induced spin gap which is observed as a plateau in the magnetization curve. The high-field measurement \cite{2} indicated that the plateau appears at 1/4 of the saturation moment. Such a magnetization plateau is predicted in various systems \cite{3–8}. A general condition of the quantization of the magnetization was derived from the Lieb-Schultz-Mattis (LSM) \cite{9} theorem for low-dimensional magnets \cite{10}. The necessary condition of the plateau is was described as

$$Q(S - m) = \text{integer}$$

where $Q$ is the spatial period of the ground state measured by the unit cell. $S$ and $m$ are the total spin and the magnetization per unit cell, respectively. Applying this theorem to the BIP-TENO, the 1/4 plateau is the case of $S = 2$ and $m = 1/2$. Therefore, a spontaneous breaking of the translational symmetry (maybe $Q = 2$) must occur at the plateau. In the previous work \cite{11} by the present authors two mechanisms of the 1/4 plateau of the $S = 1$ spin ladder were proposed, based on the frustrated interactions. In the next section we briefly review the mechanisms and show the phase diagrams obtained by the level spectroscopy analysis \cite{12}. The main purpose of this paper is to consider the realization of the mechanism at the 1/4 plateau of BIP-TENO, with some quantitative analyses on the critical magnetic fields and the temperature dependence of the susceptibility.

2 Mechanisms of 1/4 plateau

As an origin of the 1/4 magnetization plateau in the $S = 1$ spin ladder, we introduce the second and third-neighbor exchange interactions. The model is described by the Heisenberg Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{H}_Z$$

$$\hat{H}_0 = J_1 \sum_i^L (S_{1,i} \cdot S_{1,i+1} + S_{2,i} \cdot S_{2,i+1}) + J_\perp \sum_i^L S_{1,i} \cdot S_{2,i}$$

$$+ J_2 \sum_i^L (S_{1,i} \cdot S_{2,i+1} + S_{2,i} \cdot S_{1,i+1})$$

$$+ J_3 \sum_i^L (S_{1,i} \cdot S_{1,i+2} + S_{2,i} \cdot S_{2,i+2})$$

$$\hat{H}_Z = -H \sum_i^L (S_{1,i}^z + S_{2,i}^z)$$"
where $J_1, J_1, J_2$ and $J_3$ denote the coupling constants of the leg, rung and second- (diagonal) and third- exchange interactions, respectively (Fig. 1).

Hereafter we put $J_\perp=1$. $\hat{H}_Z$ is the Zeeman term where $H$ denotes the magnetic field along the $z$-axis and the eigenvalue $M$ of the conserved quantity $\sum_i(S^z_{1,i} + S^z_{2,i})$ is a good quantum number. The macroscopic magnetization is represented by $m = M/L$. In this definition the $1/4$ of the saturation magnetization corresponds to $m = 1/2$. In order to explain the mechanism of the plateau at $m = 1/2$, we use the degenerate perturbation theory around the strong rung coupling limit $J_1, J_2, J_3 \ll 1$ [13, 14]. For the two spins at each rung, we take only two dominant states; the singlet $\Psi_{0,0} \equiv |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$ and the triplet $\Psi_{1,1} \equiv |\uparrow\uparrow\rangle$. We introduce a pseudo spin $T$ for each rung coupling and map the two original states singlet $\Psi_{0,0}$ and triplet $\Psi_{1,1}$ of the $S$ picture to the $|\downarrow\rangle$ and $|\uparrow\rangle$ states of $T$, respectively. Effective Hamiltonian in pseudo spin can be written as follows:

$$\hat{H}_{\text{eff}} = \frac{8(J_1 - J_2)}{3} \sum_i (T^x_i \cdot T^x_{i+1} + T^y_i \cdot T^y_{i+1}) + \frac{J_1 + J_2}{2} \sum_i (T^z_i \cdot T^z_{i+1}) + \frac{8J_3}{3} \sum_i (T^x_i \cdot T^x_{i+2} + T^y_i \cdot T^y_{i+2}) + \frac{J_3}{2} \sum_i (T^z_i \cdot T^z_{i+2}).$$

This is the Hamiltonian of the $T = 1/2$ XXZ chain with the second-neighbor interaction. The magnetization $m = 1/2$ of the original system corresponds to $m = 0$ in the pseudo-spin system. Referring the well-known features of the $S = 1/2$ frustrated XXZ chain [12], sufficiently large $J_2$ and $J_3$ lead to the Néel order and dimerization of the pseudo spins, respectively. They correspond to the field-induced spin gap at $m = 1/2$, that is the $1/4$ plateau in the original system. The boundary between the spin-fluid and plateau phases is of the Kosterlitz-Thouless (KT) type [15]. Therefore, the pseudo-
spin picture gives two different mechanisms of the plateau; Néel order and dimerization, denoted as plateaux A and B, respectively.

The KT phase boundary can be determined precisely, using the recently developed level spectroscopy [12] applied to the low-lying energy levels of the finite chains obtained by the numerical diagonalization. We show only thus-obtained phase diagrams in the $J_2$-$J_1$ ($J_3 = 0$) and $J_3$-$J_1$ ($J_2 = 0$) planes in Figs. 2 and 3, respectively. The two gapless phases in Fig. 2 correspond to two different ordered states in the classical limit, depending on whether the rung or diagonal interactions are dominant [16]. Note that there is an upper bound of $J_1 (\sim 0.7)$ for the plateau A, while no bound for the plateau B.

![Figure 2: Phase diagram on the $J_2$-$J_1$ plane at $m = 1/2$](image)

![Figure 3: Phase diagram on the $J_3$-$J_1$ plane at $m = 1/2$.](image)
3 Comparison with Experiment

Based on the obtained phase diagrams, we discuss the realistic mechanism of the 1/4 plateau of BIP-TENO. The ratio $J_1/J_\perp$ of BIP-TENO was estimated as $J_1/J_\perp \sim 1.2$, fitting the observed temperature dependence of the susceptibility $\chi$ to the numerical calculation for the $S = 1$ simple spin ladder [1]. Fig. 2 suggests that there is no chance of the Néel plateau for $J_1/J_\perp \sim 1.2$. In addition the required value of the $J_2/J_1$ for the plateau is about 0.69 even for $J_1/J_\perp < 0.7$. $J_1/J_\perp \sim 0.69$ is too large for the realization. Thus the Néel mechanism due to $J_2$ should be discarded.

Next we consider the possibility of the dimer plateau due to $J_3$. According to Fig. 3, the plateau would appear if $J_3/J_1 > 0.39$ for $J_1/J_\perp \sim 1.2$. $J_3/J_1 \sim 0.39$ is not so far from the realization, because the lattice spacing along the leg is much smaller than the rung in the crystal structure of BIP-TENO [1]. Thus we examine the dimer mechanism due to $J_3$ more quantitatively.

![Figure 4: The ratio of the critical magnetic field ($B_2/B_1$).](image)

Using the numerical diagonalization of finite clusters up to $L = 8$ (16 spins) and the size scaling technique on the model (2), we estimate the ratio of the critical magnetic field $B_1$ that spin gap disappears and $B_2$ that plateau begin to appear. The results for $J_3/J_1 = 0.4$ and 0.5 are plotted versus $J_1$ in Fig. 4, gathered with the experimental result of BIP-TENO. It suggests that the calculated $B_2/B_1$ is closer to that of BIP-TENO for $J_1/J_\perp \sim 1.7$, rather than for $J_1/J_\perp \sim 1.2$. However the experimental estimation $J_1/J_\perp \sim 1.2$ is not so conclusive, because the fitted curve was not obtained by the numerical diagonalization of the $S = 1$ ladder, but by some mean field approximation for the rung interaction. Thus it would be important to fit the direct numerical calculation for the $S = 1$ ladder including $J_3$ to
observed $\chi$ for $J_1/J_\perp = 1.7$. We performed the finite-temperature Lanczos method to calculate the temperature dependence of $\chi$ for the system $L = 8$. The results for $J_1/J_\perp = 1.7$ and various values of $J_3/J_1$ are shown in Fig. 5, together with the experimental result of BIP-TENO. The numerical curve well agrees with the measured one for $J_3/J_1=0.4$ or 0.5. Therefore, the dimer plateau is expected to realize, if $J_1/J_\perp \sim 1.7$ and $J_3/J_1 \sim 0.4$ or 0.5 are satisfied in BIP-TENO. A little difference in $B_2/B_1$ between the model calculation and the experiment is possibly due to the finite size effect, because we used only the values for $L = 4$ and 8 to estimate it. Indeed the ratio $B_2/B_1$ of the finite systems is revealed to increase with increasing $L$ toward the experimental result.

![Graph showing temperature dependence of susceptibility $\chi$.](image)

Figure 5: The temperature dependence of the susceptibility $\chi$.

4 summary

We proposed two mechanisms of the 1/4 magnetization plateau in the $S = 1$ frustrated spin ladder. They are described by the Néel order and dimerization of the pseudo-spin system. Comparing the ratio of the two critical fields $B_2/B_1$ and the temperature dependence of $\chi$ between the model calculation and the experiment, we conclude that the dimer mechanism due to the third neighbor interaction is more suitable for BIP-TENO.

Acknowledgements

We wish to thank Professor Tsuneaki Goto for sending some experimental data of BIP-TENO.
References

[1] K. Katoh, Y. Hosokoshi, K. Inoue and T. Goto J. Phys. Soc. Jpn. 69 (2000) 1008.
[2] T. Goto, M. I. Bartashevich, Y. Hosokoshi, K. Kato and K. Inoue: Physica B 294 & 295 (2001) 43.
[3] D. C. Cabra, A. Honecker and P. Pujol: Phys. Rev. Lett. 79 (1997) 5126.
[4] D. C. Cabra and M. D. Grynberg: Phys. Rev. Lett. 82 (1999) 1768.
[5] K. Totsuka: Phys. Rev. B57 (1998) 3435.
[6] T. Tonegawa, T. Nishida and M. Kaburagi: Physica. B 246 & 247 (1998) 368.
[7] T. Tonegawa, K. Okamoto and M. Kaburagi: Physica B 294 & 295 (2001) 39.
[8] T. Sakai and M. Takahashi: Phys. Rev. B 57 (1998) R3201.
[9] E. Lieb, T. D. Schultz and D. C. Mattis: Ann. Phys. (N. Y.) 16 (1961) 407.
[10] M. Oshikawa, M. Yamanaka, and I. Affleck: Phys. Rev. Lett. 78 (1997) 1984.
[11] K. Okamoto, N. Okazaki and T. Sakai: J. Phys. Soc. Jpn. 70 (2001) 636.
[12] K. Nomura and K. Okamoto: J. Phys. A: Math. Gen. 27 (1994) 5773.
[13] N. Okazaki, K. Okamoto and T. Sakai: J. Phys. Soc. Jpn. 69 (2000) 2419.
[14] F. Mila: Eur. Phys. J. B6 (1998) 201.
[15] J. M. Kosterlitz and D. J. Thouless: J.Phys.C 6 (1973) 1181.
[16] T. Sakai and N. Okazaki: J. Appl. Phys. 87 (2000) 5893.
[17] J. Jaklic and P. Prelovsek, Phys. Rev. Lett. 74 (1995) 3411.