Magnetization Plateaux in $S = 1$ Organic Spin Ladder BIP-TENO

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Abstract

We show a clear experimental evidence of the 1/4 plateau in the high-field magnetization measurement on the first-synthesized $S = 1$ organic spin ladder compound BIP-TENO. The theoretical mechanism of the plateau formation is also proposed, based on the frustrated interactions.

Key words: Spin ladder; spin gap; magnetization plateau; frustration

The quantization of the magnetization is one of interesting topics in the low-temperature physics. Recently the first $S = 1$ spin ladder 3,3′,5,5′-tetrakis(*N-tert*-butylaminoxyl)biphenyl, abbreviated BIP-TENO [1], was synthesized. For this organic material a plateau-like anomaly at 1/4 of the saturation magnetization was found in earlier high-field magnetization measurement [2]. If this anomaly is actually a plateau, its mechanism should be non-trivial. This is because the 1/4 plateau must occur with a two-fold degeneracy in the ground state, associated with the spontaneous breakdown of the translational symmetry, based on the necessary condition of the magnetization quantization [3]. The present high-field measurement up to 70T confirmed that it is a real plateau and successfully observed another critical field for the end of the plateau. The experimental magnetization curve at T=1.3K is shown as a solid line in Fig. 1. In the previous work [4] a possible mechanism of the 1/4 plateau formation in the $S = 1$ spin ladder, based on some frustrated exchange interactions. In this paper, we apply this mechanism for BIP-TENO and estimate the realistic coupling constants, by fitting the numerically calculated magnetization curve and temperature dependence of the susceptibility of the suitable model to the experimental results.

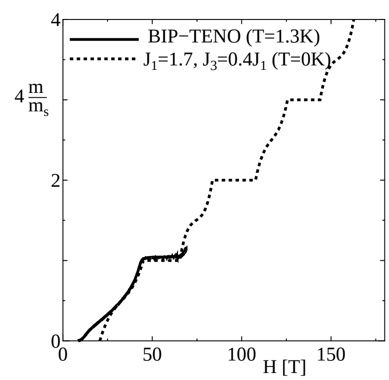

Fig. 1. Magnetization curves. Experimental result of BIP-TENO at T=1.3K (solid curve) and theoretical results based on the numerical diagonalization for finite systems of the $S = 1$ spin ladder up to $L = 8$ (dashed curve).

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In order to investigate the mechanism of the 1/4 plateau, we consider the $S = 1$ spin ladder in a magnetic field H in the presence of the 2nd- and 3rd-neighbor antiferromagnetic exchange interactions J_2 and J_3, respectively. The Hamiltonian is

$$\hat{H} = \hat{H}_0 + \hat{H}_Z \tag{1}$$

$$\begin{aligned}\hat{H}_0 = J_0 \sum_i^L \mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i} \\ + J_1 \sum_i^L (\mathbf{S}_{1,i} \cdot \mathbf{S}_{1,i+1} + \mathbf{S}_{2,i} \cdot \mathbf{S}_{2,i+1}) \\ + J_2 \sum_i^L (\mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i+1} + \mathbf{S}_{2,i} \cdot \mathbf{S}_{1,i+1}) \\ + J_3 \sum_i^L (\mathbf{S}_{1,i} \cdot \mathbf{S}_{1,i+2} + \mathbf{S}_{2,i} \cdot \mathbf{S}_{2,i+2})\end{aligned} \tag{2}$$

$$\hat{H}_Z = -H \sum_i^L (S^z_{1,i} + S^z_{2,i}), \tag{3}$$

where J_0 and J_1 are the rung and leg coupling constants, respectively. We set $J_0 = 1$ in the following analysis. The bulk magnetization is $m = M/L$, where $M \equiv \sum(S^z_{1,i} + S^z_{2,i})$. The saturation value is $m_s = 2$.

We consider the ground state at $m = 1/2$ which corresponds to the 1/4 of the saturation. According to the strong rung coupling approximation [4], we take the most important two states at each rung spin pair; the singlet and the $S^z = 1$ component of the triplet. If we introduce the pseudospin $T = 1/2$ to describe these two states, the 1/4 magnetized state of the $S = 1$ ladder (1) can be mapped onto the $m = 0$ state of the $T = 1/2$ XXZ chain including the next-nearest coupling. Based on the well-known features of the $S = 1/2$ XXZ chain, the ground state is the gapless spin fluid for $J_2 = J_3 = 0$, while the Néel (dimer) state for sufficiently large J_2 (J_3). The Néel and dimer states of the pseudospin correspond to two different mechanisms of the 1/4 plateau of the original spin ladder. The lower boundaries for the appearance of the plateau were revealed to be $J_2 \sim 0.68J_1$ and $J_3 \sim 0.31J_1$ in the strong rung limit and some quantitative phase diagrams were given in the previous work [4]. These critical values suggest that the mechanism based on the third-neighbor interaction J_3 is more realistic than J_2. The complicated crystal structure of BIP-TENO may bring about the strong third-neighbor interactions. Thus we consider the 1/4 plateau only due to J_3.

To confirm the mechanism of the plateau, we numerically calculated the magnetization curve and the temperature dependence of the susceptibility χ, using the Lanczos algorithm applied for finite systems up to $L = 8$, and fitted them to the experimental results. After several trials of the fitting, we found the most suitable parameters for BIP-TENO are $J_1 \sim 1.7$ and $J_3 \sim 0.4J_1$. The ground-state magnetization curve based on the numerical diagonalization of finite systems [6] for the parameters is shown in Fig. 1. The disagreement of H_{c1} between the calculation and the experiment can be due to the finite size effect which is generally larger for smaller magnetization. Fig. 2 shows that the χ-T curve for the same parameters, obtained by the finite-temperature Lanczos method [7] for $L = 8$, well agrees with the experimental result, in comparison with the curve for $J_3 = 0$. Thus we conclude that the most realistic parameter set is $J_1 = 1.7$ and $J_3 = 0.4J_1$.

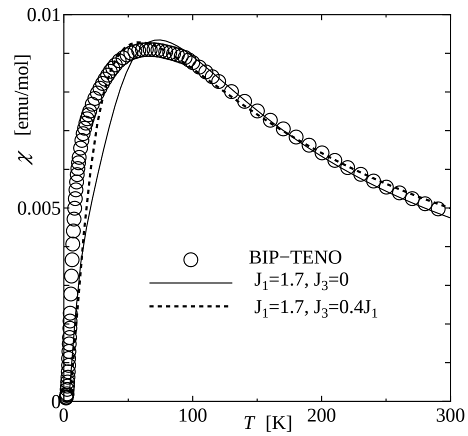

Fig. 2. Temperature dependence of the susceptibilities. The experimental result of BIP-TENO (circles) and the numerical ones calculated by the finite temperature Lanczos method for the $S = 1$ spin ladder in the cases of $J_1 = 1.7$.

In summary, we presented the result of the high-field magnetization measurement of the $S = 1$ organic spin ladder compound BIP-TENO, which indicated a clear plateau at 1/4 of the saturation magnetization. The present theoretical analyses suggest that the origin of the plateau is possibly the frustration due to the third neighbor exchange interaction.

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