

Quantum magnetization plateaux of an anisotropic ferrimagnetic spin chain

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The magnetization curve of the $(S,s)=(1,1/2)$ ferrimagnetic alternating spin chain with single-ion anisotropy D is investigated with the numerical exact diagonalization of finite clusters and size-scaling analyses. The system has a plateau at $(1/3)$ of the saturation moment, which corresponds to the spontaneous magnetization for $D=0$. Varying D in the $1/3$ -magnetized ground state under the external field along the axis of D , a quantum phase transition is revealed to occur at the critical value $D/J=1.114\pm 0.001$ where the plateau vanishes. Except for the critical point, the plateau is always opening, but the mechanism is different between $D<D_c$ and $D>D_c$. The change of mechanisms is an evidence to clarify that the plateau originates from the quantization of magnetization.

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I. INTRODUCTION

The quantization of the magnetization is one of interesting phenomena in low-dimensional magnets. It is detected as a plateau in the magnetization curve. Such plateaux were actually observed in high-field measurements of several materials: the $S=1$ bond-alternating chain $[\text{Ni}_2(\text{Medpt})_2(\mu\text{-ox})(\mu\text{-N}_3)]\text{ClO}_4\cdot 0.5\text{H}_2\text{O}$ [Medpt = methyl-bis(3-aminopropyl)amine],¹⁻³ the organic $S=1$ ladder $3,3',5,5'$ -tetrakis(*N*-*tert*-butylaminoxyl)biphenyl, abbreviated BIP-TENO,⁴⁻⁸ the Shastry-Sutherland system $\text{SrCu}_2(\text{BO}_3)_2$ (Refs. 9 and 10) and the zigzag double chain NH_4CuCl_3 (Refs. 11 and 12), etc. In addition some theoretical and/or numerical analyses predicted that a magnetization plateau appears in various other systems; the polymerized chains,¹³⁻²³ the $S=3/2$ chain,²⁴⁻²⁷ the frustrated spin ladder,²⁸⁻³² several generalized spin ladders,³³⁻³⁸ distorted diamond-type spin chain,³⁹⁻⁴¹ and some layered systems.^{42,43} Using the Lieb-Schultz-Mattis theorem,⁴⁴ a general necessary condition for the presence of the plateau was derived²⁴ as

$$\bar{S} - m = \text{integer}, \quad (1)$$

where \bar{S} and m are the sum of spins over all sites and the magnetization in the unit period, respectively.

The ferrimagnetic mixed spin chains have lately attracted a lot of interest among quantum spin systems. Recent synthesizing techniques have produced a lot of such materials, for example, the bimetallic chain $\text{MM}'(\text{pbaOH})(\text{H}_2\text{O})_3\text{nH}_2\text{O}$ (Ref. 45) and the organic one $\{\text{Mn}(\text{hfac})_2\}_3(3\text{R})_2$ (Ref. 46) etc. Thus many experimental investigations have been done on such ferrimagnets, as well as theoretical ones.⁴⁷⁻⁵⁸ In the systems two different spins S and s ($S>s$) are arranged alternately in a line and coupled by the nearest-neighbor antiferromagnetic exchange interaction. They have the spontaneous magnetization $m=-s$ in the ground state. Since the lowest excitation increasing m has an energy gap, the magnetization curve has a plateau at m

$=S-s$.⁵⁴ Indeed the plateau satisfies the condition of the quantization of the magnetization (1). The plateau, however, is also realized in the classical limit where S and s are infinite with the ratio S/s fixed. Thus it is difficult to identify the plateau at $m=S-s$ as a result from the quantization of the magnetization, unlike any higher plateaux at $m=S-s+1, S-s+2, \dots, S+s-1$ which should not appear in the classical Heisenberg spin systems.^{59,60} The previous works^{61,62} to investigate the anisotropy in the exchange interaction revealed that the quantum effect stabilizes the plateau at $m=S-s$ against the XY -like anisotropy. However, it is no more than a quantitative difference between the quantum and classical systems. In this paper, to show a more definite evidence to clarify that the plateau is a result from the quantization, we investigate the single-ion anisotropy D effect. It is more realistic than the interaction anisotropy even from an experimental viewpoint. For example, the recently synthesized $(S,s)=(1,1/2)$ chain $\text{NiCu}(\text{pba})(\text{D}_2\text{O})_32\text{D}_2\text{O}$ (Ref. 63) would possibly have the anisotropy D on the Ni ion ($S=1$). Thus we study the $(1,1/2)$ system with D for $S=1$, using the numerical diagonalization and the level spectroscopy method⁶⁵⁻⁶⁸ under the twisted boundary condition.^{69,70} It will reveal that the system has a quantum phase transition with respect to D at $m=1/2$ due to the change of plateau formation mechanisms, which clarifies a full quantum nature of the plateau.

II. CLASSICAL SYSTEM

The ferrimagnetic $(S,s)=(1,1/2)$ mixed spin chain with the single-ion anisotropy under an external magnetic field H is described by the Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{ex}}, \quad (2)$$

$$\mathcal{H}_0 = \sum_{j=1}^N \{S_j \cdot s_j + s_j \cdot S_{j+1} + D(S_j^z)^2\}, \quad (3)$$

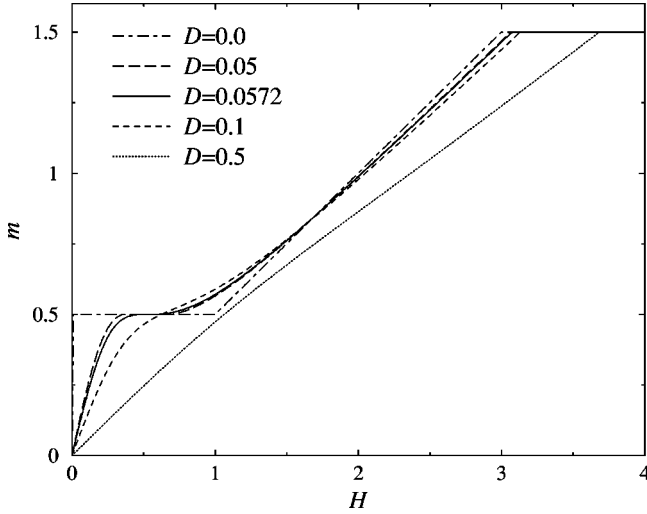


FIG. 1. Magnetization curves of the classical $(S,s)=(1,1/2)$ chain for various D . The plateau appears at $m=1/2$ only for $D < D_c=0.0572$.

$$\mathcal{H}_{\text{ex}} = -H \sum_{j=1}^N (S_j^z + s_j^z). \quad (4)$$

Throughout this paper, we consider only the case when the external field is along the symmetry axis of D . We note that $D(s_j^z)^2$ gives a constant $D/4$. In order to clarify the quantum nature of the plateau later, we show the properties of the classical spin system with the same Hamiltonian where S and s are the classical vectors with the amplitudes 1 and $1/2$, respectively. The magnetization process at $T=0$ can be solved with the standard variation of the Hamiltonian with respect to the two vectors S and s . In the isotropic case of the classical system the ground-state magnetization curve begins at the spontaneous magnetization $m=S-s=1/2$ and has a plateau there, shown as a dot-dashed line in Fig. 1. The complete Néel order along H ($S_j^z=1, s_j^z=-1/2$) is realized in the plateau state, while the canted Néel order (in the xy plane) occurs for $1/2 < m < 3/2$. Thus the plateau should originate from the classical Néel order. Since the Néel order is oriented in the xy plane for $D > 0$, the magnetization curve starts from $m=0$. The plateau at $m=1/2$ still appears for small positive D , as a long dashed line ($D=0.05$) in Fig. 1, because the Néel order along H can be realized there. With increasing D , however, the plateau disappears at the critical value $D_c=0.0572$ and it does not appear any more for $D > D_c$, shown in Fig. 1. The break down of the plateau due to the easy plane anisotropy $D > 0$ is qualitatively the same as the case of the XY -like coupling anisotropy.^{61,62} In the quantum system discussed in the following sections, however, the plateau will be revealed to appear again for larger D due to the quantum effect, in contrast to the coupling anisotropy.

III. QUANTUM MECHANISMS OF PLATEAU

In the quantum spin system described by the Hamiltonian (2) it would be efficient to introduce the composite spin picture, where $S=1$ is considered as the triplet state of two $1/2$

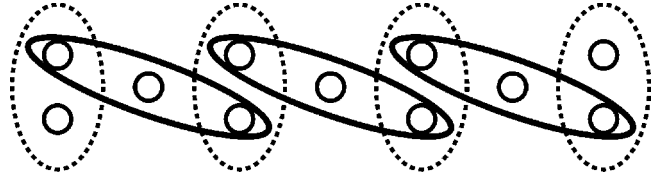


FIG. 2. Schematic picture of the Haldane mechanism of the plateau at $m=1/2$. Each circle represents a $1/2$ spin. A solid ellipse is a trimer and a dotted ellipse means symmetrized two spins.

spins.⁷¹ Using this picture we clarify the two different mechanisms of the plateau at $m=1/2$; the Haldane and large- D mechanisms for small and large D , respectively. The names of these two mechanisms originate from the Haldane and large- D phases of the $S=1$ antiferromagnetic chain where the critical point was revealed to be $D \sim 1$.⁷²⁻⁷⁴ Similar mechanisms were also proposed for the $1/3$ plateau of the $S=3/2$ chain.^{26,27}

A. Haldane Plateau

For smaller D each $S=1$ site can be in any state of $S^z = -1, 0$, and $+1$. Thus the two $1/2$ spins representing $S=1$ have only to be symmetrized. In the plateau state at $m=1/2$ ($1/3$ of the saturation magnetization), the antiferromagnetic trimer state, which is schematically shown in Fig. 2, is expected to be realized. In Fig. 2 a solid ellipse represents the trimer

$$|\uparrow\rangle = \frac{1}{\sqrt{6}}(|\uparrow\uparrow\downarrow\rangle - 2|\uparrow\downarrow\uparrow\rangle + |\downarrow\uparrow\uparrow\rangle), \quad (5)$$

and a dotted one corresponds to an $S=1$ site where the two $1/2$ spins should be symmetrized. When the plateau is based on the effective mechanism in Fig. 2, we call it the Haldane plateau. In the ideal state where this picture is exactly realized, the expectation values of the z component are $\frac{1}{3}, -\frac{1}{6}$, and $\frac{1}{3}$ for the three spins in each trimer, respectively. Thus the original system is expected to have $\langle S^z \rangle = \frac{2}{3}$ and $\langle s^z \rangle = -\frac{1}{6}$ in the ideal Haldane state. It suggests that the Haldane state has the Néel order along H , but the amplitude is smaller than the classical system ($\langle S^z \rangle = 1$ and $\langle s^z \rangle = -\frac{1}{2}$) because of the quantum spin reduction. We note that the $D = -\infty$ case is not the ideal Haldane case. In this case, classical values $\langle S^z \rangle = 1$ and $\langle s^z \rangle = -1/2$ will be realized.

B. Large- D plateau

Each $S=1$ site tends to have $S^z=0$ for larger positive D . In the composite spin picture $S^z=0$ corresponds to one of the triplet states

$$\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle). \quad (6)$$

Thus the large- D mechanism of the $1/3$ plateau ($m=1/2$) is presented schematically in Fig. 3, where a rectangle represents the triplet state (6) at $S=1$ site and each $s=1/2$ site has $s^z=1/2$. In the ideal large- D state the expectation values

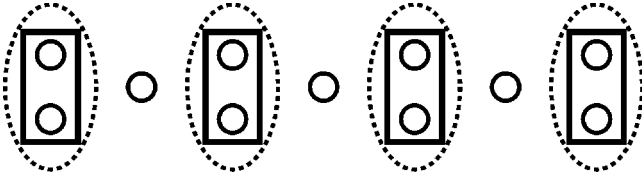


FIG. 3. Schematic picture of the large- D mechanism of the plateau at $m=1/2$. Each circle represents a $1/2$ spin. A solid rectangle is one of the triplet (6).

should be $\langle S^z \rangle = 0$ and $\langle s^z \rangle = +1/2$. Obviously no Néel order is realized along z axis in the large- D phase. The large- D plateau results from the quantization of $S=1$. Thus it is never realized in the classical spin system.

IV. QUANTUM CRITICAL POINT

The existence of the two different mechanisms of the $1/3$ plateau suggests that there is a quantum phase transition between them with respect to the parameter D in the ground state at $m=1/2$. A useful order parameter to investigate the phase transition is the spin excitation gap at $m=1/2$,

$$\Delta \equiv E(M+1) + E(M-1) - 2E(M). \quad (7)$$

$E(M)$ is the lowest energy level in the subspace where the eigenvalue of $\sum_j (S_j^z + s_j^z)$ is M and $m=1/2$ corresponds to $M=N/2$ for the N -unit system. Δ is also the length of the plateau. Based on the analogy with the $S=1$ chain, it is expected that the critical point D_c between the Haldane and large- D phases is a Gaussian fixed point. Thus the gap would vanish just at D_c and open in both phases. The scaled gap $N\Delta$ calculated for several finite systems under the periodic boundary condition using the numerical diagonalization is plotted versus D in Fig. 4. It indicates that there exists a critical point at $D \sim 1.1$ where the scaled gap $N\Delta$ is independent of N , namely the system is gapless ($\Delta \sim 1/N$). It also suggests that the gap is opening for both sides of D_c , as

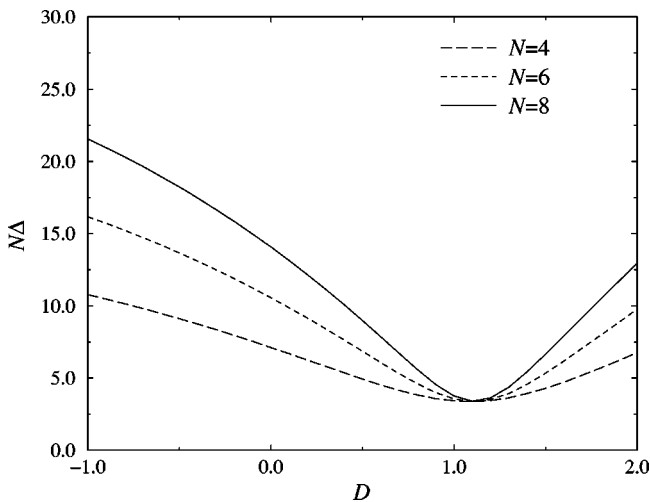


FIG. 4. Scaled gap $N\Delta$ plotted versus D . It suggests that the critical point is around $D=1.1$.

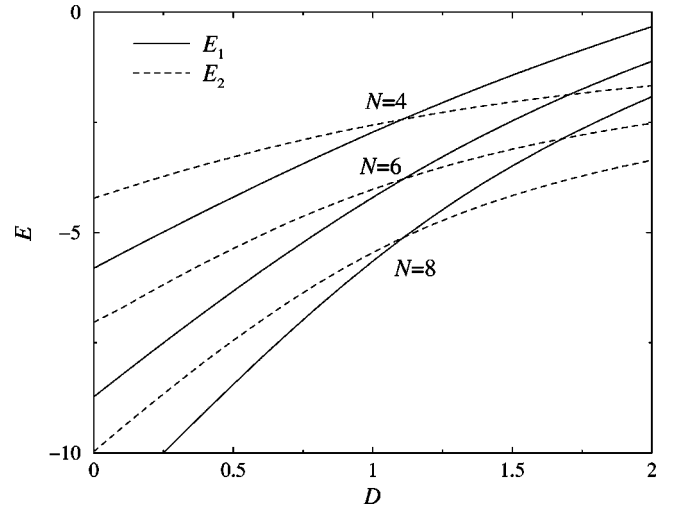


FIG. 5. Lowest two energy levels under the twisted boundary condition. The crossing point is the size-dependent critical point.

expected. In general, the fixed point $D_{c,N,N+2}$ of the phenomenological renormalization equation⁶⁴

$$N\Delta_N(D) = (N+2)\Delta_{N+2}(D') \quad (8)$$

depends on the system size N for small clusters. However, the behavior of the scaled gap in Fig. 4 suggests that the fixed point is almost independent of N , that is, $D_{c,4,6} \sim D_{c,6,8}$. It implies that the higher-order size correction [$o(1/N)$] of the gap Δ is negligible even for $N=4, 6$, and 8 in the present system. Thus some systematic size-scaling analyses would lead to a precise estimation of the critical point in the thermodynamic limit, even based only on such small cluster calculations.

One of the most precise methods to determine the Gaussian fixed point of the one-dimensional quantum systems is the level spectroscopy^{65–68} under the twisted boundary condition.^{69,70} The twisted boundary condition means that the sign of the coupling constant for the XY component is switched in the exchange interaction at the boundary. According to the method, the critical point is determined as a crossing point of the lowest two energy levels under twisted boundary conditions. The two levels E_1 and E_2 are plotted versus D for the finite system with $N=4, 6$ and 8 in Fig. 5. The size dependence of the crossing point is so small that a precise D_c is expected to be obtained. Since the effect of some irrelevant fields⁶⁶ should yield the size correction proportional to $1/N^2$, we extrapolate the size-dependent D_c to the thermodynamic limit using the plot versus $1/N^2$ in Fig. 6. The result is $D_c = 1.114 \pm 0.001$. The good agreement of the size correction with $1/N^2$ in Fig. 6 justifies the accuracy of the present estimation. In fact, the plateau-nonplateau phase boundary of some spin ladder systems determined by the level spectroscopy even with small cluster calculations well agreed with the exact result in some ideal limits.^{6,31,38} The above scaled gap analysis also supports the existence of the quantum critical point in the present system.

In order to confirm the realization of the two mechanisms of the plateau discussed in the preceding section, the expect-

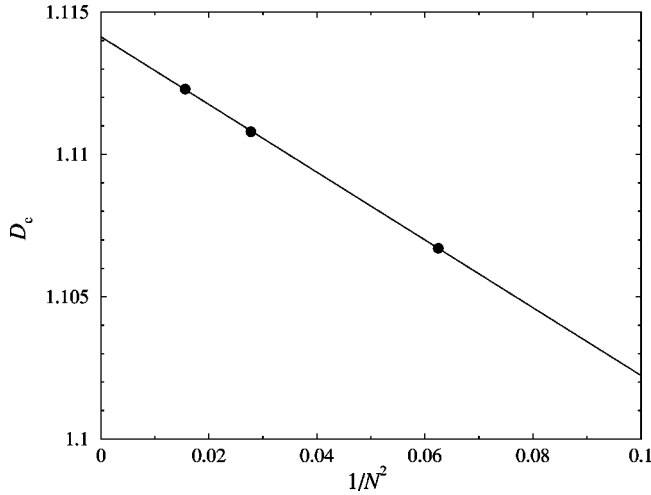


FIG. 6. Estimation of the critical point D_c in the thermodynamic limit based on fitting a line $\sim 1/N^2$.

tation values of the z component $\langle S^z \rangle$ and $\langle s^z \rangle$ for finite systems are plotted versus D in Fig. 7. It indicates that the sign of $\langle s^z \rangle$ switched around the critical point. It means that the Néel order along z axis exists only in the Haldane phase. Thus it is also consistent with the schematic pictures of the two plateau mechanisms.

V. MAGNETIZATION CURVES

Finally, we present the ground-state magnetization curve of the quantum systems described by the Hamiltonian (2) for several values of D , using some size-scaling techniques⁷⁵ applied for the numerical energy levels of finite systems up to $N=10$. The conformal field theory in one-dimensional quantum systems^{76–78} applied to the present model predicted that the in gapless phases the size dependence of the energy gap have the asymptotic forms

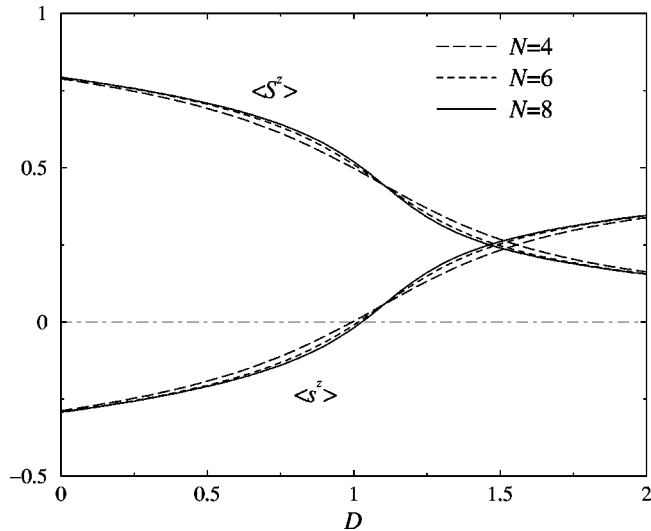


FIG. 7. Sublattice magnetizations $\langle S^z \rangle$ and $\langle s^z \rangle$ plotted versus D . It suggests that the sign of $\langle s^z \rangle$ is changed at the critical point.

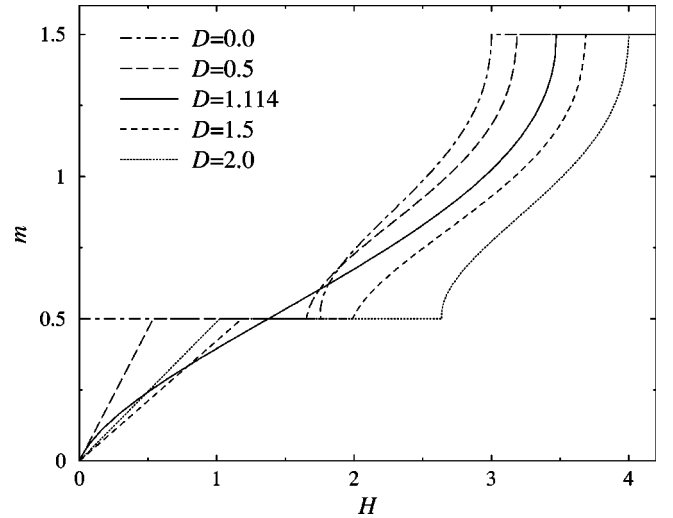


FIG. 8. Magnetization curves of the quantum system for several values of D . The plateau at $m=1/2$ is caused by the Haldane mechanism for $D=0$ and 0.5 , while the large- D one for $D=1.5$ and 2.0 . The curve has no plateau just at the critical value $D=1.114$.

$$E(M+1) - E(M) \sim H(m) + \pi v_s \eta \frac{1}{N}, \quad (9)$$

$$E(M) - E(M-1) \sim H(m) - \pi v_s \eta \frac{1}{N}, \quad (10)$$

where N and M vary with $m=M/N$ fixed. Thus the forms are useful to estimate the magnetic field H for several values of m , which can be obtained from all the combinations of N and M for available finite systems. The method works except for the plateau. On the other hand, at the massive pointlike plateaux the Shanks transformation,⁷⁹ which is a technique to accelerate the convergence of sequences, is useful. The general form of the transformation for a sequence a_n is

$$a'_n = \frac{a_{n-1}a_{n+1} - a_n^2}{a_{n-1} + a_{n+1} - 2a_n}. \quad (11)$$

We show the result of the magnetization curve for several values of D in Fig. 8, where only the polynomial curves suitably fitted to the obtained points based on the above method. Figure 8 shows that with increasing D up to the critical point $D=D_c=1.114$ the plateau at $m=1/2$ is shrinking, while it is opening again for $D>D_c$, in contrast to the classical system. The plateau is due to the Haldane mechanism for $D<D_c$ while the large- D mechanism for $D>D_c$. This change of mechanisms should be a clear evidence of the quantum nature in the plateau formation.

VI. SUMMARY

The magnetization process of the $(S,s)=(1,1/2)$ ferromagnetic mixed spin chain with the single-ion anisotropy D was investigated using the numerical exact diagonalization and some size-scaling analyses. It revealed that the mechanism of the plateau at $m=1/2$ changes from the Haldane to large- D one at the Gaussian quantum critical point $D_c=1.114 \pm 0.001$. It justifies that the plateau at $m=1/2$ originates from the quantization of the magnetization, although a similar plateau also appears in the classical system.

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