Quantum critical phenomena in magnetization process of the Kagome and triangular lattice antiferromagnets

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Abstract. Using the numerical exact diagonalization of finite-size clusters up to 39 spins, we investigate the magnetization process of the Kagome lattice antiferromagnet, as well as the triangular lattice antiferromagnet. A finite-size scaling analysis at 1/3 of the saturation magnetization indicates that an unconventional quantum phase transition, so called a magnetization ramp, occurs for the Kagome lattice antiferromagnet. In contrast, the triangular lattice antiferromagnet is revealed to exhibit a conventional 1/3 magnetization plateau. These conclusions are based on the estimated critical exponent \( \delta \) defined as \( |m - m_c| \sim |H - H_c|^{1/\delta} \). The exponents at the lower and higher field sides of \( m = 1/3 \) \( \delta_- \) and \( \delta_+ \) are estimated as \( \delta_- = 1.9 \pm 1.0 \) and \( \delta_+ = 0.5 \pm 0.2 \) for the Kagome lattice, while \( \delta_- = 1.0 \pm 0.2 \) and \( \delta_+ = 0.8 \pm 0.2 \) for the triangular lattice.

1. Introduction

The \( S = 1/2 \) Kagome\textsuperscript{[1]} and triangular lattice antiferromagnets are ones of the most popular frustrated quantum spin systems. The previous theoretical studies indicated that the former system is disordered in the ground state \([5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16]\), while the latter has the 120\degree long-range order\([2, 3, 4]\). Experimental studies to observe a novel spin liquid phase have been accelerated since discoveries of several realistic materials; the herbertsmithite\([18, 19]\), the volborthite\([20, 21]\) and the vesignieite\([22]\) for the Kagome lattice, the organic compound \( \kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3 \) for the triangular lattice\([17]\). Since the quantum Monte Carlo simulation and the DMRG calculation are useless for these systems, the numerical exact diagonalization is one of the best numerical method for them. The numerical diagonalization studies suggested that both systems have the 1/3 magnetization plateau \([23, 24, 25, 26]\), although the classical spin systems have no plateau on both lattices in the ground state\([28, 29]\). In our recent numerical diagonalization study on the \( S = 1/2 \) Kagome lattice antiferromagnet up to \( N = 36 \), the calculated field derivatives revealed an anomalous behavior at 1/3 of the saturation magnetization\([30]\). Namely, the field derivative is diversing at the low-field side of the critical field \( H_c \), while almost zero at the high-field side. This critical behavior is quite different from conventional magnetization plateaux in two-dimensional systems where the field derivative is finite at both sides of \( H_c \). To distinguish such an anomalous property at the 1/3 magnetization of the Kagome lattice from conventional plateaux, we called it a “magnetization ramp”. However,
its mechanism is still an open problem. In this paper, to clarify such an unconventional behavior around the 1/3 magnetization of the $S = 1/2$ Kagome lattice antiferromagnet, comparing with the triangular one, we applied the numerical diagonalization for both systems up to $N = 39$ which is the largest cluster at present. In addition we estimated the critical exponent $\delta$ by the finite-size scaling proposed by the previous work[32], to investigate the quantum critical behavior more quantitatively.

2. Model and calculation
The magnetization processes of the $S = 1/2$ kagome and triangular lattice antiferromagnets are described by the Hamiltonian

$$
\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_Z,
$$

$$
\mathcal{H}_0 = \sum_{\langle i,j \rangle} S_i \cdot S_j,
$$

$$
\mathcal{H}_Z = -H \sum_j^N S_j^z,
$$

where $\langle i, j \rangle$ means all the nearest neighbor pairs on each lattice. Throughout we use the unit such that $g\mu_B = 1$. For $N$-site systems, the lowest energy of $\mathcal{H}_0$ in the subspace where $\sum_j S_j^z = M$ (the macroscopic magnetization is $m = M/N$) is denoted as $E(N, M)$. We restrict us to the rhombic cluster under the periodic boundary condition to keep the 120$^{\circ}$ rotational symmetry for a systematic finite-size scaling. Using the numerical exact diagonalization, we can calculate all the values of $E(N, M)$ available for the rhombic clusters with $N = 12, 21, 27, 36$ and 39, to obtain the ground state magnetization curves. In our previous work[30] the analysis of the field derivative calculated as the form

$$
\chi^{-1} = \frac{E(N, M + 1) - 2E(N, M) + E(N, M - 1)}{1/M_s},
$$

revealed an exotic behavior of the Kagome lattice antiferromagnet at $m = 1/3$, called a magnetization ramp, using the result up to $N = 36$. This phenomenon is characterized by a difference between the lower and higher field sides of $m = 1/3$; $\chi$ is diverging at the lower side like a plateau in one-dimensional systems, while is very small (possibly zero) at the higher one. The magnetization and the field derivative curves of the Kagome lattice antiferromagnet for $N = 39$ also supported such a ramp-like behavior[31] at least qualitatively.

3. Critical exponent
In order to characterize the unconventional magnetization behavior at $m = 1/3$ of the Kagome lattice antiferromagnet in a quantitative way, we introduce the critical exponent $\delta$. It is defined by the form

$$
|m - m_c| \sim |H - H_c|^{1/\delta},
$$

which is an important index to specify the universality class of the field induced quantum phase transition. The previous theoretical works indicated $\delta = 2$ for some typical one-dimensional gapped systems[33, 34], while $\delta = 1$ for two-dimensional systems[35]. In order to clarify the anomalous critical behavior at $m = 1/3$ of the Kagome lattice antiferromagnet, we estimate $\delta$ by the finite-size scaling developed by the previous work[32], and compare it with the triangular lattice antiferromagnet. Although the method was proposed for one-dimensional systems, it can
be easily generalized for two dimensions. We assume the asymptotic form of the size dependence
of the energy as
\[ \frac{1}{N} E(N, M) \sim \epsilon(m) + C(m) \frac{1}{N^\theta} \quad (N \to \infty), \]
(4)
where \( \epsilon(m) \) is the bulk energy and the second term describes the leading size correction. We
also assume that \( C(m) \) is an analytic function of \( m \). The lowest and highest magnetic
field corresponding to \( m = 1/3 \) in the thermodynamic limit are defined as \( H_{c1} \) and \( H_{c2} \), respectively,
as the form
\[ E(N, \frac{N}{3}) - E(N, \frac{N}{3} - 1) \to H_{c1} \quad (N \to \infty), \]
(5)
\[ E(N, \frac{N}{3} + 1) - E(N, \frac{N}{3}) \to H_{c2} \quad (N \to \infty). \]
(6)
In order to consider the critical magnetization behaviors for \( m < 1/3 \) and \( m > 1/3 \)
independently, we define the critical exponents \( \delta_- \) and \( \delta_+ \) by the forms
\[ m - \frac{1}{3} \sim (H - H_{c2})^{1/\delta_+}, \]
(7)
\[ \frac{1}{3} - m \sim (H_{c1} - H)^{1/\delta_-}. \]
(8)
If we define the quantities \( f_+(N) \) and \( f_-(N) \) by the forms
\[ f_\pm(N) \equiv \pm[E(N, \frac{N}{3} \pm 2) + E(N, \frac{N}{3}) - 2E(N, \frac{N}{3} \pm 1)], \]
(9)
the asymptotic forms of them are expected to be
\[ f_\pm(N) \sim \frac{1}{N^{\delta_\pm}} + O\left(\frac{1}{N^{\theta+1}}\right) \quad (N \to \infty), \]
(10)
as far as we assume the form (5). Thus the exponents \( \delta_- \) and \( \delta_+ \) can be estimated from the
slope of the \( \log f_\pm - \log N \) plot, respectively, under the condition \( \theta > \delta_\pm - 1 \). In order to avoid
an oscillation of the finite-size correction due to the cluster shape dependence, we just use the
rhombic clusters under the periodic boundary condition with \( N = 12, 21, 27, 36, \) and \( 39 \). The
plots of \( \log f_\pm \) versus \( \log N \) for the triangular and Kagome lattice antiferromagnets are shown in
Figures. 1 and 2, respectively. Figure 1 suggests that the calculated points are well fitted to a
line for each of \( f_- \) and \( f_+ \) in the case of the triangular lattice. Thus applying the standard least
square fitting to lines (Dashed and long-dashed lines are used to obtain \( \delta_+ \) and \( \delta_- \), respectively.)
for \( N = 12, 21, 27, 36, 39, \delta_- \) and \( \delta_+ \) are estimated as follows:
\[ \delta_- = 1.0 \pm 0.2, \quad \delta_+ = 0.8 \pm 0.2, \]
for the triangular lattice. The errors are estimated from the deviation of points from the fitted
lines. It would be reasonable to conclude \( \delta_- = \delta_+ = 1 \) at \( m = 1/3 \) of the triangular lattice
antiferromagnet, as expected for conventional magnetization plateau in two dimensions. On the
other hand, Figure 2 indicates quite different feature of the Kagome lattice antiferromagnet.
The same least square fitting yields the following estimates:
\[ \delta_- = 1.9 \pm 1.0, \quad \delta_+ = 0.5 \pm 0.2, \]
Figure 1. \( \log(f) \) is plotted versus \( \log(N) \) for the triangular lattice antiferromagnet. Solid circles and squares are useful to estimate the critical exponents \( \delta^+ \) and \( \delta^- \), respectively.

For the Kagome lattice antiferromagnet. Exponent \( \delta_- \) has a large error because the line fitting is not good. It does not converge with respect to the system size well, but seems to still increasing with \( N \). The same line fitting to the points for \( N = 27, 36 \) and \( 39 \) yields the estimation \( \delta_- = 4.6 \pm 0.3 \). Thus we can just conclude \( \delta_- \geq 2 \) at most. It means that the diversing behavior of the field derivative at \( H_{c1} \) is stronger than one-dimensional systems. It is led to two possibilities. One is a jump (a first-order transition) in the magnetization curve. A magnetization jump which also appears near the saturation was proved[36]. The other is an anomalous continuous transition. A similar phenomenon was reported in the metal-insulator transition of the Hubbard chain with next-nearest neighbor hopping[?]. In comparison with \( \delta_- \), \( \delta^+ \) is more conclusive, because the fitting error is much smaller. According to the above result of the line fitting, we conclude \( \delta^+ \) is smaller than unity. Thus the field derivative \( \chi \) should be zero at the higher field side of \( H_{c2} \). It also justifies a property of the magnetization ramp.

4. Ramp versus plateau
We consider whether a flat part of the magnetization curve at \( m = 1/3 \) exists or not for the triangular and Kagome lattice antiferromagnets. Namely, we examine whether each system has no plateau \( (H_{c1} = H_{c2}) \) or a finite plateau \( (H_{c1} \neq H_{c2}) \) at \( m = 1/3 \) in the thermodynamic limit. We evaluate the length of the flat part \( H_{c2} - H_{c1} \) corresponding to the plateau width of the finite-size clusters with \( N = 12, 21, 27, 36 \) and \( 39 \) for both systems. If the system has a gapless excitation like a spin wave from some ordered states, the low-lying energy spectrum is expected to be proportional to the wave vector \( k \) in the long wave length limit. Thus the excitation energy gap of the finite-size systems should have the asymptotic form \( \sim 1/N^{1/2} \) in two-dimensional gapless systems. On the other hand, in gapped systems the gap is expected to converge to the thermodynamic limit with exponentially decaying (faster than \( 1/N^{1/2} \)) finite-size correction, as the system size increases. Thus if the extrapolation by fitting the gap versus \( 1/N^{1/2} \) leads to a finite gap in the thermodynamic limit, it would be a strong evidence to confirm the gapped ground state. The length of a flat part \( H_{c2} - H_{c1} \) is plotted versus \( 1/N^{1/2} \) in Figure 3, where open triangles and solid circles are for the triangular and Kagome lattice antiferromagnets,
Figure 2. log(f) is plotted versus log(N) for the Kagome lattice antiferromagnet. Solid circles and squares are useful to estimate the critical exponents $\delta^+$ and $\delta^-$, respectively.

Figure 3. Plateau width $H_{c2} - H_{c1}$ is plotted versus $1/N^{1/2}$. Open triangles and solid squares are for the triangular and Kagome lattice antiferromagnets, respectively. Fitted lines are used for the extrapolation to the thermodynamic limit.

respectively. The least square fitting to a line leads to the following results: $H_{c2} - H_{c1} = 0.3 \pm 0.2$ for the triangular lattice and $H_{c2} - H_{c1} = -0.3 \pm 0.5$ for the Kagome lattice. Obviously we can conclude that the triangular lattice antiferromagnet has the 1/3 magnetization plateau. In contrast, the result for the Kagome lattice suggests that it possibly has a single critical field $H_c = H_{c1} = H_{c2}$. Thus we conclude that the Kagome lattice antiferromagnet exhibits a 1/3 magnetization ramp as a single quantum phase transition at $H_c$.

In the recent magnetization measurement[20] on a candidate of the Kagome lattice antiferromagnet Volborthite several step-like behaviors were observed, but it has not reached $m = 1/3$ yet. The same measurement is still going on to observe an anomaly at $m = 1/3$, which
is expected to be about 60T. It would be interesting to detect some unconventional features.

5. Summary
We have investigated critical magnetization behaviors at $m = 1/3$ for the $S = 1/2$ triangular and Kagome lattice quantum antiferromagnets, using the numerical exact diagonalization of rhombic clusters up to $N = 39$. The triangular lattice is revealed to have the critical exponents $\delta_\perp = \delta_\parallel = 1$ and a finite plateau, which are consistent with a conventional magnetization plateau in two-dimensional systems. On the other hand, the Kagome lattice is revealed to exhibit unconventional critical properties; $\delta_\perp < 1 < \delta_\parallel$, namely the field derivative $\chi$ is diverging at the lower field side, while zero at the higher one of a single critical field $H_c = H_{c1} = H_{c2}$. The conclusion supports the magnetization ramp behavior at $m = 1/3$ of the Kagome lattice antiferromagnet.

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