Novel Field-induced Quantum Phase Transition of the Kagome-lattice Antiferromagnet

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The magnetization process of the $S = 1/2$ kagome-lattice quantum antiferromagnet is investigated using the numerical exact diagonalization up to 36-site clusters. Our previous finite-size scaling analysis with rhombic clusters indicated the “magnetization ramp” as a novel field-induced quantum phase transition at 1/3 the saturation magnetization. As another possible exotic behavior, we focus on the feature of the magnetization curve at 2/3 the saturation. The critical exponent analysis indicates that a different singular behavior occurs at the 2/3 magnetization.

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I. INTRODUCTION

The $S = 1/2$ kagome-lattice antiferromagnet [1] is one of the most popular frustrated quantum spin systems. Previous theoretical studies indicated that the system is disordered in the ground state [2–13]. Experimental studies to observe a novel spin liquid phase in the kagome-lattice antiferromagnet have been accelerated since discoveries of several realistic materials; herbertsmithite [14,15], volborthite [16,17] and vesignieite [18,19]. Since the quantum Monte Carlo simulation is useless for the present system, the numerical exact diagonalization is one of the best numerical method to use; another is the density matrix renormalization group (DMRG) calculation. The DMRG is often said to be powerful for the one-dimensional case, but is not so good for the two- and three-dimensional cases. Numerical diagonalization studies suggested a magnetization plateau-like behavior at 1/3 the saturation magnetization [20–24], although the classical spin systems have no plateau in the ground state [25]. In our recent numerical diagonalization study on the $S = 1/2$ kagome-lattice antiferromagnet up to $N = 36$, the calculated field derivatives of the magnetization revealed an anomalous behavior at 1/3 the saturation magnetization [26]. Namely, the field derivative is divergent on the low-field side of the critical field $H_c$, while being zero on the high-field side. This critical behavior is quite different from the conventional magnetization plateau 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.” Our recent finite-size scaling analyses, which were restricted to rhombic clusters, quantitatively revealed some characteristic features of the magnetization ramp [27,28].

The kagome-lattice antiferromagnet is also expected to exhibit another possible exotic magnetization behavior at 2/3 the saturation magnetization because some commensurate spin structure can occur. In this paper, we focus on the critical magnetization behavior at 2/3 magnetization for the system. Using the same critical exponent analysis as used for 1/3 magnetization, we investigate the field-induced phenomena at the 2/3 magnetization and reveal that a singular behavior different from the ramp may occur.

II. MODEL AND CALCULATIONS

The magnetization process of the $S = 1/2$ kagome-lattice antiferromagnet is described by the Hamiltonian

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

$$\mathcal{H}_Z = -H \sum_j S_j^z, \quad (1)$$

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where \((i,j)\) means all the nearest neighbor pairs. Throughout we use units such that \(g\mu_B = 1\). For \(N\)-site systems, the lowest energy of \(\mathcal{H}_0\) in the subspace where \(\sum_j S_j^z = M\) is denoted as \(E(N,M)\). The macroscopic magnetization \(m\) is defined by \(m \equiv M/M_s = 2M/N\), where \(M_s = N/2\) is the saturation magnetization. In our previous work \([27]\), we restricted ourselves to rhombic clusters under a periodic boundary condition to investigate the 1/3 magnetization. However, we used even-spin clusters in the present work because 2/3 the saturation magnetization does not appear in odd-spin clusters. The used system sizes of even-spin clusters were \(N = 12, 18, 24, 30\) and 36. Using a numerically exact diagonalization, we could calculate all the values of \(E(N,M)\) for these clusters.

### III. NO PLATEAU AT \(M = 2/3\)

At first, we consider whether or not a flat part of the magnetization curve due to a field-induced spin gap at \(m = 2/3\) exists for the kagome-lattice antiferromagnets. Namely, we examine whether the system has no plateau \((H_{c1} = H_{c2})\) or a finite plateau \((H_{c1} \neq H_{c2})\) at \(m = 2/3\) in the thermodynamic limit. We evaluate the length of the flat part \(H_{c2} - H_{c1}\) corresponding to the plateau width of finite-size clusters for \(N = 12, 18, 24, 30\) and 36. 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-wavelength limit. Thus, the excitation energy gap of 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 an exponentially decaying (faster than \(1/N^{1/2}\)) finite-size correction as the system size is increased. Thus, if the extrapolation by fitting the gap versus \(1/N^{1/2}\) leads to a finite gap in the thermodynamic limit, that would be strong evidence to confirm the existence of a gapped ground state. The length of the flat part \(W \equiv H_{c2} - H_{c1}\) is plotted versus \(1/N^{1/2}\) in Fig. 1. The least-squares fitting to a line leads to the following results: \(W = -0.17 \pm 0.35\). Thus the result suggests no plateau \((W = 0)\) within the errors.

### IV. CRITICAL EXPONENTS

In order to characterize a possible unconventional magnetization behavior at \(m = 2/3\) for the kagome-lattice antiferromagnet in a quantitative way, we introduce the critical exponent \(\delta\). It is defined by the form

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

and it is an important index to specify the universality class of the field-induced quantum phase transition. Previous theoretical works indicated \(\delta = 2\) for some typical one-dimensional gapped systems \([29,30]\) while \(\delta = 1\) for two-dimensional systems \([31]\). In order to clarify the anomalous critical behavior at \(m = 2/3\) for the kagome-lattice antiferromagnet, we estimate \(\delta\) by using finite-size scaling developed in a previous work \([32]\). Although the method was proposed for one-dimensional systems, it can be easily generalized to two dimensions. We assume the asymptotic form of the size dependence of the energy as

\[
\frac{1}{N} E(N,M) \sim \epsilon(m) + C(m) \frac{1}{N^\delta} \quad (N \to \infty),
\]

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 the highest magnetic field, corresponding to \(m = 2/3\) in the thermodynamic limit are defined as \(H_{c1}\) and \(H_{c2}\), respectively, and have the form

\[
E(N, \frac{N}{3}, \frac{N}{3} + 1) = E(N, \frac{N}{3}) - H_{c2} \quad (N \to \infty).
\]

In order to consider the critical magnetization behaviors for \(m < 2/3\) and \(m > 2/3\) independently, we define the critical exponents \(\delta_-\) and \(\delta_+\) with the forms

\[
m - \frac{2}{3} \sim (H - H_{c2})^{1/\delta_-}.
\]

\[
\frac{2}{3} - m \sim (H_{c1} - H)^{1/\delta_+}.
\]

If we define the quantities \(f_+(N)\) and \(f_-(N)\) with the forms

\[
f_+(N) \equiv [E(N, \frac{N}{3} + 2) + E(N, \frac{N}{3}) - 2E(N, \frac{N}{3} - 1)] \quad (8)
\]

\[
f_-(N) \equiv [E(N, \frac{N}{3} + 2) + E(N, \frac{N}{3}) - 2E(N, \frac{N}{3} + 1)] \quad (9)
\]
these asymptotic forms are expected to be
\[ f_-(N) \sim \frac{1}{N^{\delta_-}} + O\left(\frac{1}{N^{\theta+1}}\right) \quad (N \to \infty), \quad (10) \]
\[ f_+(N) \sim \frac{1}{N^{\delta_+}} + O\left(\frac{1}{N^{\theta+1}}\right) \quad (N \to \infty), \quad (11) \]
as long as we assume the form in Eq.(4). Thus, the exponents \( \delta_- \) and \( \delta_+ \) can be estimated from the slopes of the \( \ln f_-\ln N \) and \( \ln f_+\ln N \) plots, respectively, under the conditions \( \theta > \delta_- - 1 \) and \( \theta > \delta_+ - 1 \). Plots of \( \ln f_- \) and \( \ln f_+ \) versus \( \ln N \) for the kagome lattice antiferromagnets are shown in Fig. 2 and 3, respectively. In the case when \( f_- \) or \( f_+ \) is negative because of some accidental finite-size effects, we neglect it when estimating the critical exponent. (Since these negative values of \( \ln f_- \) and \( \ln f_+ \) possibly result from an instability just below \( m = 2/3 \), we cannot exclude the possibility of a magnetization jump there.) Applying the standard least-squares fitting of lines to useful points for \( N = 12, 18, 24, 30 \) and 36, we estimate the exponents as \( \delta_- = 2.5 \pm 1.3 \) and \( \delta_+ = 2.8 \pm 1.9 \). These results suggest that \( \delta_- \) and \( \delta_+ \) are larger than 1, which means that the field derivative \( dm/dH \) is divergent on both the higher and the lower field sides of \( m = 2/3 \), different from \( m = 1/3 \) where \( dm/dH = 0 \) on the higher field side. Because the present size-scaling analysis indicated \( H_{c1} = H_{c2} \), a singular behavior should occur at a single critical field \( H_c \) \( (= H_{c1} = H_{c2}) \). This would be a possible novel field induced quantum phase transition, although the mechanism is still an open question.

In order to show a global behavior of the magnetization, we present the magnetization curves calculated for \( N = 30 \) and 36 in Fig. 4. Dotted and solid curves are the magnetization curves for \( N = 30 \) and 36, respectively. In addition, \( m = 1/3, 2/3 \) and 1 are shown as long dashed lines. Actually, a jump appears just below \( m = 2/3 \) for \( N = 36 \) in Fig. 4.

\[ Fig. 2. \ln(f_-) \text{ is plotted versus } \ln(N). \]

\[ Fig. 3. \ln(f_+) \text{ is plotted versus } \ln(N). \]

\[ Fig. 4. \text{Magnetization curves calculated by using the exact numerical diagonalization for } N = 30 \text{ and 36. Dotted and solid curves correspond to } N = 30 \text{ and 36, respectively. Long dashed lines indicate } m = 1/3, 2/3 \text{ and 1.} \]

V. CONCLUSIONS

In summary, we have investigated critical magnetization behaviors at \( m = 2/3 \) for the \( S = 1/2 \) kagome lattice quantum antiferromagnet, using the numerical exact diagonalization of general clusters up to \( N = 36 \). The system is revealed to exhibit unconventional critical properties; \( \delta_- \) and \( \delta_+ \) are larger than 1 and no magnetization plateau \( (H_c = H_{c1} = H_{c2}) \), namely the field derivative \( \chi \) is divergent at a single critical field \( H_c \). The conclusion suggests that a new quantum phase transition possibly occurs at \( m = 2/3 \) of the kagome lattice antiferromagnet.

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