Magnetization Process of the Spin-S Kagome-Lattice Heisenberg Antiferromagnet

Hiroki Nakano and Tōru Sakai

1 Graduate School of Material Science, University of Hyogo, Kamigori, Hyogo 678-1297, Japan
2 Japan Atomic Energy Agency, SPring-8, Sayo, Hyogo 679-5148, Japan

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The magnetization process of the spin-S Heisenberg antiferromagnet on the kagome lattice is studied by the numerical-diagonalization method. Our numerical-diagonalization data for small finite-size clusters with S = 1, 3/2, 2, and 5/2 suggest that a magnetization plateau appears at one-third of the height of the saturation in the magnetization process irrespective of S. We discuss the S dependences of the edge fields and the width of the plateau in comparison with recent results obtained by real-space perturbation theory.

Frustrated spin systems have attracted much attention from many condensed-matter physicists. One of the fascinating systems among them is the kagome-lattice Heisenberg antiferromagnet. Unfortunately, our understanding of this system is still far from complete in spite of many experimental and theoretical studies. In the S = 1/2 system, in particular, discoveries of several realistic materials such as herbertsmithite,\textsuperscript{1,2} volborthite,\textsuperscript{3,4} and vesignieite\textsuperscript{5,6} have accelerated theoretical studies.\textsuperscript{7–29} However, there remain some unresolved issues; one of them is the spin-gap problem of whether the spin excitation above the singlet ground state is gapped or gapless.

On the other hand, fewer studies on spin-S systems have been carried out. As candidate S = 1 kagome-lattice systems, m-MPYM-BF\textsubscript{4}\textsuperscript{30,31} NaV\textsubscript{3}(OH)\textsubscript{6}(SO\textsubscript{4})\textsubscript{2}\textsuperscript{32} [C\textsubscript{6}N\textsubscript{2}H\textsubscript{8}]\textsubscript{2}[Ni\textsubscript{3}F\textsubscript{6}(SO\textsubscript{4})\textsubscript{2}]\textsubscript{2}\textsuperscript{33} and KV\textsubscript{3}Ge\textsubscript{2}O\textsubscript{4}\textsuperscript{34} are known. Theoretical studies\textsuperscript{9,35–39} for the S = 1 case are also limited. Studies on the S > 1 cases have only started recently; candidate kagome-lattice systems of Cs\textsubscript{2}Mn\textsubscript{3}LiF\textsubscript{12}\textsuperscript{40} for S = 2 and NaBa\textsubscript{2}Mn\textsubscript{3}F\textsubscript{11}\textsuperscript{41} for S = 5/2 have been reported, together with theoretical studies\textsuperscript{42–44} as well as an analysis based on the semiclassical limit.\textsuperscript{13}

Under these circumstances, then, we are faced with a question: do any systematic behaviors exist in the spin-S kagome-lattice Heisenberg antiferromagnet under magnetic fields? The purpose of this letter is to extract such systematic behavior of the magnetization processes of this model for various S by numerical-diagonalization calculations that are unbiased against approximations. With the same motivation, Zhitomirsky recently investigated the frustrated Heisenberg model under magnetic fields by real-space perturbation theory taking into account fluctuations around a classical configuration.\textsuperscript{44} He found that in the magnetization process of the kagome-lattice antiferromagnet, the so-called \textit{udd} state is stable at one-third of the height of the saturation, at which a magnetization plateau appears irrespective of the value of S. He also derived an expression for the 1/S expansion for both the edge fields of this height. The comparison between the present numerical-diagonalization results and the results from real-space perturbation theory should contribute to our understanding of the frustration effect in the kagome-lattice antiferromagnet.

The Hamiltonian that we study in this research is given by $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{Zeeman}}$, where

$$\mathcal{H}_0 = \sum_{\langle i,j \rangle} J \mathbf{S}_i \cdot \mathbf{S}_j,$$

and

$$\mathcal{H}_{\text{Zeeman}} = -H \sum_j S_j^z.$$

Here, $\mathbf{S}_i$ denotes the spin operator at site $i$, where the sites are the vertices of the kagome lattice. The spin operator satisfies $\mathbf{S}_i^2 = S(S + 1)$. The sum of $\mathcal{H}_0$ runs over all the nearest-neighbor pairs in the kagome lattice. Energies are measured in units of $J$; hereafter, we set $J = 1$. The number of spin sites is denoted by $N$, where $N/3$ is an integer. We impose the periodic boundary condition for clusters with site $N$.

We calculate the lowest energy of $\mathcal{H}_0$ in the subspace belonging to $\sum_j S_j^z = M$ by numerical diagonalizations based on the Lanczos algorithm and/or the Householder algorithm. The energy is denoted by $E(N_s, M)$, where $M$ takes an integer or a half odd integer value up to the saturation value $M_s = SN_s$. We often use the normalized magnetization $m = M/M_s$. Part of the Lanczos diagonalizations were carried out using the MPI-parallelized code, which was originally developed in the study of Haldane gaps.\textsuperscript{45} The usefulness of our program was previously confirmed in large-scale parallelized calculations.\textsuperscript{19,27,46}

The magnetization process for a finite-size system is obtained by considering the magnetization increase from $M$ to $M + 1$ in the field

$$H = E(N_s, M + 1) - E(N_s, M),$$

under the condition that the lowest-energy state with magnetization $M$ and that with magnetization $M + 1$ become the ground state in specific magnetic fields.

First, let us present our results of the magnetization processes for $S = 1, 3/2, 2$, and $5/2$; results are shown in Fig. 1. The maximum sizes of the clusters treated in
Fig. 1. (Color) Magnetization processes for $S = 1, 3/2, 2, \text{ and } 5/2$ in (a), (b), (c), and (d), in which the maximum sizes are $N_s = 24, 18, 15, \text{ and } 12$, respectively. Black circles, red triangles, blue squares, green pentagons, light-blue inverted triangles, and violet diamonds linked by lines of the same color denote the cases of $N_s = 9, 12, 15, 18, 21, \text{ and } 24$, respectively.

This study are $N_s = 24, 18, 15, \text{ and } 12$ for $S = 1, 3/2, 2, \text{ and } 5/2$, respectively. The cluster shapes we calculated are the same as those in Ref. 9. Note here that, the shapes for $N_s = 9, 12, \text{ and } 21$ are rhombic and that the others are nonrhombic. In Fig. 1(a), the entire range of the cases up to $N_s = 18$ and part of the range in the case of $N_s = 21$ were already reported in Ref. 9 and the rest of the range in the case of $N_s = 21$ and the results for $N_s = 24$ are additionally presented in the present study. For the cases of $S > 1$, there are no reports on numerical-diagonalization calculations of the magnetization processes to the best of our knowledge.

The most noteworthy behavior is observed at one-third of the height of the saturation, where behavior similar to a magnetization plateau appears irrespective of the value of $S$. A detailed discussion concerning the edges and the width of this height will be given later. The next characteristic behavior is a jump near the saturation. Note here that all the states within the jump are numerically degenerate at the saturation field $H_s (= 6JS)$. A similar jump is known to occur in several cases.$^{11,47−49}$ Regarding the existence or the absence of the degeneracy, this behavior is different from the magnetization jump observed in square-kagome-lattice and Cairo-pentagon-lattice antiferromagnets, and so on.$^{50−53}$ Although this behavior occurs irrespective of the value of $S$, the skip of $m$ at the jump gradually decreases as $S$ is increased. Around $m = (9S − 2)/(9S)$ near the jump for $S = 1, 3/2, 2, \text{ and } 5/2$, a region where the gradient of the magnetization process seems small may exist, although the behavior is very faint. To clarify the existence of the magnetization plateau at this height, future studies of larger systems are required. Note here that this height does not correspond to the $m = 7/9$ plateau near the jump in the $S = 1/2$ case in Ref. 49 but that it corresponds to $m = 5/9$ for $S = 1/2$, at which the existence of another plateau has been pointed out for the $S = 1/2$ kagome-lattice antiferromagnet.$^{23,24}$ There appears an overhanging behavior at $m = 17/21$ only for $N_s = 21$ in the $S = 1$ case; this may be an artifact owing to a finite-size effect because it is very small and because it does not appear for $N_s = 24$.

Hereafter, we focus our attention on the behavior at $m = 1/3$.

Next, let us examine the system-size dependence of the edges at $m = 1/3$, i.e., the lower-field edge $H_{c1}$ and the higher-field edge $H_{c2}$; results are shown in Fig. 2. In all cases of $S$, the size dependences of $H_{c1}$ and $H_{c2}$ are not large. Recall here that in the case of $S = 1/2$, the discontinuous size dependence between $N_s = 18$ and 21 is known to occur in $H_{c2}$; the decrease is about 20%. In the case of $S = 1$, on the other hand, the decrease between $N_s = 18$ and 21 is about 6%, which is much smaller than that in the case of $S = 1/2$. It is therefore reasonable to use the values of $H_{c1}$ and $H_{c2}$ of the largest cluster for each $S$ as substitutes for those.
of the infinite-size system without extrapolation when we carry out a detailed analysis. Note also that the small size dependences of $H_{c1}$ and $H_{c2}$ are related to the properties of the $m = 1/3$ states. After the $m = 1/3$ states were studied in an analysis based on the semiclassical limit,\(13\) it was pointed out that the $m = 1/3$ states reveal a nine-site structure in the unit cells of the spin states from the analysis of an effective Hamiltonian obtained by perturbation theory from the Ising limit,\(42\) although Refs. 10 and 42 did not clarify the existence of the plateau or present estimates of $H_{c1}$ and $H_{c2}$. If the states form the nine-site structure, the states are energetically stable when $N_s/9$ is an integer; on the other hand, the energies are higher when $N_s/9$ is not an integer than when $N_s/9$ is an integer. This argument suggests that for finite-size $H_{c1}$ and $H_{c2}$, $H_{c1}$ ($H_{c2}$) becomes lower (higher) only when $N_s/9$ is an integer. However, such behavior is not observed in Fig. 2. Thus, the present results do not support the nine-site structure in the $m = 1/3$ states. The same situation was pointed out in Ref. 27 for the $S = 1/2$ Kagome-lattice antiferromagnet. For future studies, numerical data for $H_{c1}$ and $H_{c2}$ are shown in Table I together with the singlet ground-state energy $E(N_s, 0)$.

Next, let us examine the $S$ dependence of $H_{c1}$ and $H_{c2}$. We plotted $H_{c1}$ and $H_{c2}$ for the largest cluster as a function of $1/S$; the result is shown in Fig. 3 together with $H_{c1}$ and $H_{c2}$ for the $N_s = 42$ cluster in the $S = 1/2$ case reported in Ref. 27 as the largest-cluster result for $S = 1/2$. We also draw the curves of the expressions

$$\frac{H_{c1}}{JS} = 2 - \frac{1}{8S} - \frac{1}{4S^2},$$

(4)

$$\frac{H_{c2}}{JS} = 2 + \frac{3}{8S} + \frac{1}{4S^2},$$

(5)

derived in Ref. 44. One observes that the dependence of $H_{c2}$ changes between $S = 1$ and $S = 3/2$ and that the numerically obtained $H_{c2}$ for $S \geq 3/2$ approaches a value of 2 with increasing $S$, which is the value for an infinite $S$, namely, the classical case. The agreement of $\lim_{S \to \infty} H_{c2}$ with the classical value suggests that the substitution explained in the above is reasonable. Note here that the $1/S$ dependence of the numerically obtained $H_{c2}$ is convex upward. This convex dependence is clearly different from Eq. (5). On the other hand, $H_{c1}$ shows an almost linear dependence on $1/S$; the line seems to approach a value that is slightly smaller than 2, which is also the value for the classical case. The reason for the difference of this value from the classical value is unclear at present. The dependence of $H_{c1}$ may change above $S = 5/2$ if we assume a continuous dependence toward the classical limit. Thus, the $1/S$ dependence must be concave upward when $S$ is large. This concave dependence is clearly different from Eq. (4). Note additionally that the almost smooth $S$-dependences of our results in Fig. 3 do not support the existence of the nine-site structure in the $m = 1/3$ state pointed out in Ref. 42.

Finally, let us analyze the width of the height at

| $S$ | $N_s$ | $H_{c1}/J$ | $H_{c2}/J$ | $E(N_s, 0)/(N_s JS^2)$ |
|-----|-------|-------------|-------------|---------------------|
| 1   | 24    | 1.7502      | 2.7430      | -1.4266894          |
| 3/2 | 18    | 2.7016      | 3.9827      | -1.2895265          |
| 2   | 15    | 3.6697      | 5.0085      | -1.2259126          |
| 5/2 | 12    | 4.5944      | 6.0308      | -1.1835511          |

Table 1. Edge fields for the $m = 1/3$ height in the magnetization process of the spin-$S$ Kagome-lattice Heisenberg antiferromagnet for the largest cluster treated in the present study. We also present the energy per site of the singlet ground state.$^{44}$

Fig. 2. System-size dependence of (a) $H_{c2}$ and (b) $H_{c1}$. Circles, triangles, squares, and diamonds denote the cases of $S = 1$, 3/2, 2, and 5/2, respectively.

Fig. 3. $S$-dependence of $H_{c1}$ and $H_{c2}$ of the largest-size clusters. Circles and squares represent the results of $H_{c1}$ and $H_{c2}$ for the largest-size clusters, respectively. Equations (4) and (5) are drawn as dashed and broken curves, respectively.
with reliable numerical estimates for spin-$S$ also different from Eq. (6). Note here that the present dependence of the Haldane gap. This feature is in which we also draw the line Fig. 4. Width $\Delta$ for the largest-size clusters plotted as a function of $1/S$. The dashed line denotes Eq. (6).

$m = 1/3$ in the magnetization process, where the width is given by $\Delta = H_{c2} - H_{c1}$; the result is shown in Fig. 4, in which we also draw the line

$$\frac{\Delta}{J} = \frac{1}{2} + \frac{1}{2S}, \quad (6)$$

obtained from Eqs. (4) and (5). The most striking feature is that the numerically obtained $\Delta$ increases as $S$ is increased in spite of the fact that $\Delta$ is regarded as an energy gap in the magnetic field. Regarding the $S$ dependence of the energy gap, it is well known that the Haldane gap of the integer-$S$ Heisenberg chain shows exponential decay with respect to $S$. The $S$-dependence of Haldane’s expression can be compared with reliable numerical estimates for spin-$S$ Haldane gaps. The present increase in $\Delta$ is contradictory to the dependence of the Haldane gap. This feature is also different from Eq. (6). Note here that the present observation of $\Delta$ in units of $J$ approaching a nonzero limit does not contradict the simple expectation that the plateau width will vanish in the classical limit because the plateau should be measured in comparison with $H_s$, which is linear in $S$. The nonzero limit of $\Delta$ in units of $J$ is different from $1/2$ in Eq. (6). One cannot, unfortunately, deny the possibility that $\Delta$ is overestimated in the present numerical-diagonalization study owing to the finite-size effect. This is a possible reason for the disagreement. Thus, studies tackling calculations of larger clusters should be carried out in future. Another possible reason for the disagreement is that the perturbation treatment around the classical configuration may be too rough to properly capture the essential quantum effect in the $m = 1/3$ state of the kagome-lattice antiferromagnet. The roughness may be reduced by additionally taking into account the effect from the spin waves; an examination along such a direction should be carried out in a future study.

In summary, we have investigated the magnetization process of the general spin-$S$ Heisenberg antiferromagnet on the kagome lattice by the numerical-diagonalization method. We have found that a magnetization plateau appears at one-third of the height of the saturation even in the cases of large $S$. Our analysis of the edge fields and the width of the plateau suggests that the numerical-diagonalization results disagree with the equations obtained by real-space perturbation theory. The present study, based on an unbiased and non-perturbative theoretical method, presents significant information concerning general spin-$S$ antiferromagnets with frustrations, which will contribute to future studies. In the case of $S = 1/2$, the $m = 1/3$ state of several frustrated systems shows an interesting phase transition between the ferrimagnetic state and another state accompanied by a novel spin-flop phenomenon. Future studies taking this distortion into account could help us to more clearly establish the relationship between the nine-site structure pointed out in Ref. 42 and unbiased numerical data. When the kagome lattice is spatially anisotropic, a non-Lieb-Mattis-type ferrimagnetic ground state of the $S = 1/2$ system exists near the isotropic point. It should be examined in future what happens to these nontrivial phenomena when $S \geq 1$. Further study on general spin-$S$ systems would greatly contribute to our understanding of the frustration effect in quantum spin systems.

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