

**Metamagnetic jump in the spin- $\frac{1}{2}$  antiferromagnetic Heisenberg model on the square kagome lattice**

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The magnetization process of the spin- $\frac{1}{2}$  antiferromagnetic Heisenberg model on two-dimensional square-kagome lattice is studied theoretically. The metamagnetic jumps exist in the magnetization process at the higher edge of the  $\frac{1}{3}$  and  $\frac{2}{3}$  plateaus. The parameter-dependencies of the critical field and the magnitude of the magnetization jump at the higher edge of the  $\frac{1}{3}$  plateau are obtained by using the approximated state in the unit cell and compared with the numerical results of the exact diagonalization of 42 sites.

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The magnetization process in frustrated Heisenberg spins attracts much interest.

Kagome lattice consists of triangles and hexagons. The triangle structure makes frustration on the system. Recently, lattice with triangles, squares, and octagons, called square kagome lattice or shuriken lattice (see Fig. 1), has also been studied [1–5]. It has been reported that besides the magnetic plateaus at  $\frac{1}{3}$  and  $\frac{2}{3}$  in the magnetization process, the magnetization jump occurs at the high field edge of the  $\frac{1}{3}$  plateau [3–5]. There exists another magnetization jump between  $\frac{2}{3}$  plateau and the saturation of the magnetization, which is known to occur in kagome lattice [6,7]. Ising spins on the square kagome lattice has also been studied recently [8]. Effective Hamiltonians have been proposed to study the frustrated spin systems [2,9].

The magnetization jump, or metamagnetic jump, in anisotropic spin systems is rather easily understood as a spin flop phenomenon, which is a first-order transition between differently ordered states. In the Heisenberg antiferromagnetic spins on the square kagome lattice, on the other hand, the jump occurs in the isotropic spin systems. The magnetization jump on the square kagome lattice is also the first-order transition, but the phases are not so easily imagined as a classical spin picture. The magnetization jump is also shown to exist in the square lattice with the next-nearest-neighbor interactions ( $J_1 - J_2$  model) [10], where the first-order transition between different states occurs. Recently, another isotropic spin system (Cairo pentagon lattice [11,12]) has been discovered to have the magnetization jump. The Cairo pentagon lattice has no triangle structure but the frustration is caused by the pentagon structure. The square kagome lattice and the Cairo pentagon lattice can be constructed from the Lieb lattice, where frustration does not exist, as shown in Fig. 2. It is well known that the Lieb-lattice antiferromagnet holds the so-called Marshall-Lieb-Mattis theorem [13,14]. This theorem clarifies that this system shows the ferrimagnetic ground state. Additional interaction bonds like  $J_2$  in Fig. 2 change the behavior of the system. Other types of additional interactions were studied [15–17]. Among them, the kagome-lattice and Lieb-lattice antiferromagnets are connected by the additional interactions [15]. There is also another modulation from the kagome-lattice antiferromagnet.

In the case of the  $\sqrt{3} \times \sqrt{3}$  modulation in the kagome lattice, the magnetization jump also occurs [4,18,19]. The square kagome lattice and Cairo pentagon lattice have smaller unit cells (six spins in the unit cell) than the kagome lattice with the  $\sqrt{3} \times \sqrt{3}$  modulation (nine spins in the unit cell), so it is more appropriate to study the magnetization jump in the frustrated spin systems numerically and analytically.

Plateau and jump in the magnetization process have also been studied in the frustrated Heisenberg spin ladder [20,21] and in the anisotropic triangular antiferromagnet [10]. It is known that the triangular-lattice Heisenberg antiferromagnet shows a plateau without jumps at both edges [22–26]. Addition and removal of interactions in the triangular-lattice antiferromagnet were also studied from the viewpoints of the changing plateau behavior [27–29]. Therefore, it is worth studying how the change of interaction affects the behavior of various magnetic systems.

In this paper we study the magnetization process, especially the  $J_2/J_1$  dependencies of the critical magnetic field  $h_2$  and magnitude of the magnetization jump at  $h_2$ , in the square kagome lattice by using the approximated eigenstate and we give insights for the magnetization jump obtained numerically in this system. Rousochatzakis *et al.* [2] have introduced the effective models in the square-kagome lattice and similar lattices, i.e., the effective interactions between  $\beta$  spins around the singlet formed by four  $\alpha$  spins are obtained in the case of  $J_1 \ll J_2$ . They also gave the nearest-neighbor valence-bond description at  $J_1 \approx J_2$ , which has been studied in the kagome lattice [30,31]. Although they extensively studied the states of  $M = 0$  and the plateau boundary at  $J_2/J_1 \gg 1$ , little attention has been paid to the magnetization jump in the square-kagome lattice at  $J_2/J_1 \approx 1$ . We show that the magnetization jump at the higher edge of the  $\frac{1}{3}$  plateau can be approximated as the uniform phase of the entangled state (or the linear combination of the eigenstates) in the unit cell.

**II. SQUARE KAGOME LATTICE AND THE EXACT DIAGONALIZATION**

The square-kagome lattice is shown in Fig. 1. There are four  $\alpha$  sites and two  $\beta$  sites in the unit cell which is shown by the red square in Fig. 1. Each  $\beta$  site is shared by neighboring unit cells.

## square-kagome lattice

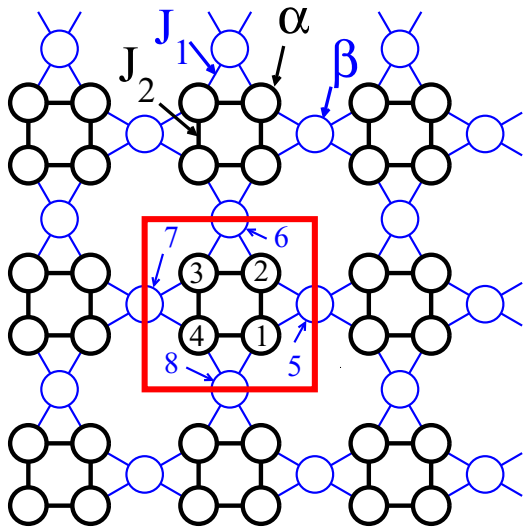


FIG. 1. Square kagome lattice. Unit cell is shown by the red square, which consists of four  $\alpha$  sites (1–4) and two  $\beta$  sites (5–8; each spin belongs to two neighboring unit cells), forming the *shuriken* structure.

The Heisenberg model on the square-kagome lattice is given by [4]

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_{\text{Zeeman}}, \quad (1)$$

where  $\mathcal{H}_1$  is the nearest-neighbor interaction between spins on the  $\alpha$  and  $\beta$  sites,

$$\mathcal{H}_1 = J_1 \sum_{(i,j), i \in \alpha, j \in \beta} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (2)$$

$\mathcal{H}_2$  is the nearest-neighbor interaction between spins on the  $\alpha$  sites,

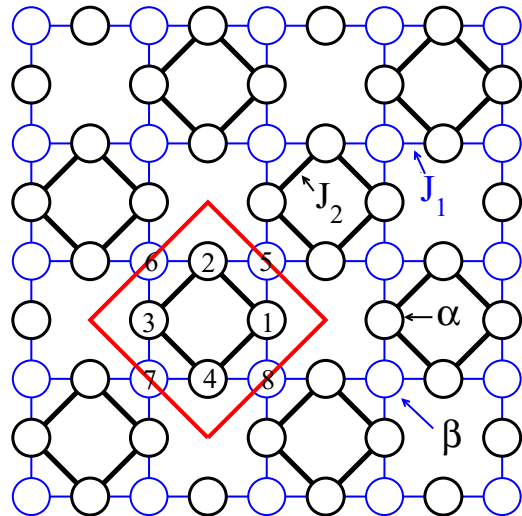
$$\mathcal{H}_2 = J_2 \sum_{(i,j), i \in \alpha, j \in \alpha} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (3)$$

and  $\mathcal{H}_{\text{Zeeman}}$  is the Zeeman energy in the magnetic field  $h$ ,

$$\mathcal{H}_{\text{Zeeman}} = -h \sum_j S_j^z. \quad (4)$$

We have reported [5] the magnetization process obtained by exact diagonalization in the square kagome lattice of  $N_s = 24, 30, 36,$  and  $42$ , where  $N_s$  is the number of spins and  $N_0 = N_s/6$  is the number of unit cell. The exact diagonalization is carried out based on the Lanczos algorithm and the Householder algorithm. The latter one is used only for the case when the dimension of the Hilbert space is small. When the dimension of the Hilbert space becomes extremely large, on the other hand, the Lanczos diagonalization is carried out using an MPI-parallelized code, which was originally developed in the study of Haldane gaps [32]. The usefulness of our program was confirmed in several large-scale parallelized calculations [5,19,33–35]. The result of the magnetization process obtained by the exact diagonalization with the parameters  $J_2/J_1 = 1.04$  and  $N_s = 42$  is shown in Fig. 3. The magnetization process of

## square-kagome lattice



## Cairo pentagon lattice

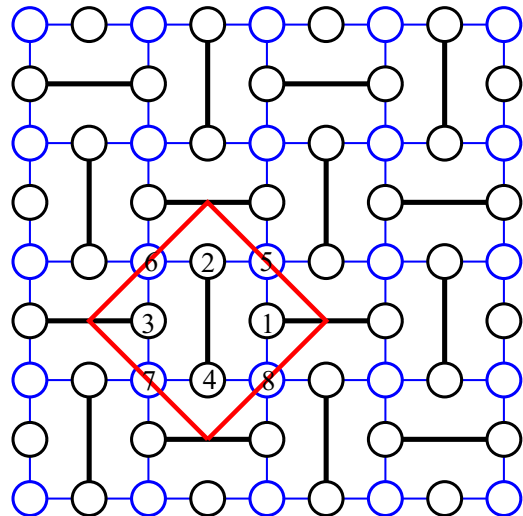


FIG. 2. Square kagome lattice, topologically same as Fig. 1 and the Cairo pentagon lattice. Unit cell is shown by the red square, which consists of four  $\alpha$  sites and two  $\beta$  sites.

the Heisenberg antiferromagnetic spins on the square kagome lattice is shown schematically in Fig. 4. There are plateaus in the magnetization process at  $M/M_s = 1/3$  and  $2/3$  when  $h_1 \leq h \leq h_2$  and  $h_3 \leq h \leq h_4$ , respectively.

The metamagnetic jump at  $h = h_2$  is determined by the Maxwell construction [5,36]

The size dependence of the jump at  $h = h_2$  is shown in Fig. 5. The size dependence of  $h_2$  is small as obtained from  $N_s = 30, 36,$  and  $42$ .

### III. MAGNETIZATION PROCESS

We define the total spin operators for  $\alpha$  spins (1–4 in Fig. 1) and  $\beta$  spins (5–8 in Fig. 1) as

$$\mathbf{S}^\alpha = \mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4, \quad (5)$$

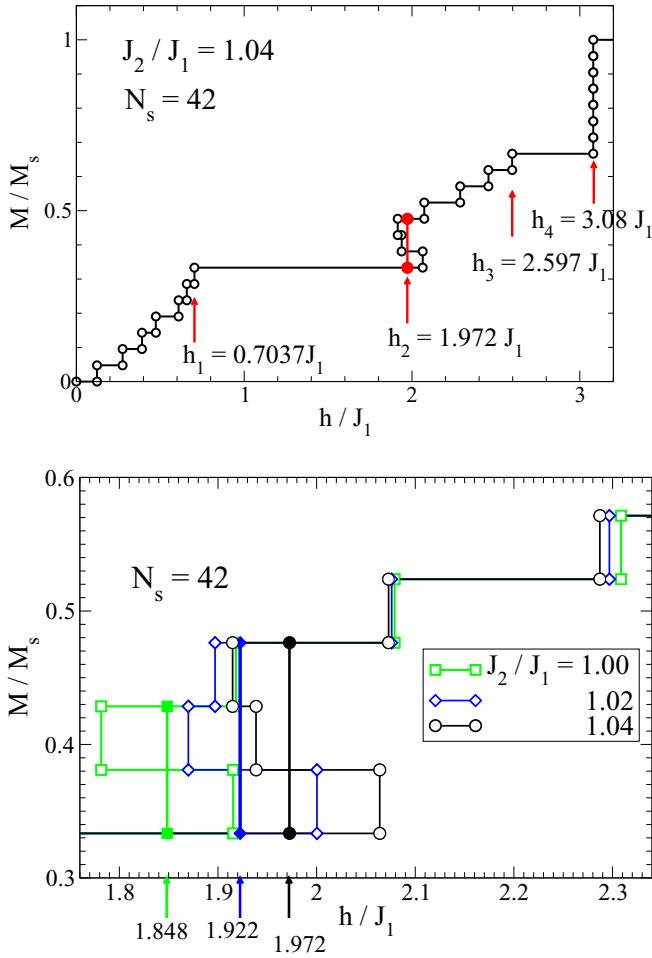


FIG. 3. Magnetization process obtained by the exact diagonalization for the system  $J_2/J_1 = 1.04$  and  $N_s = 42$  and the close-up plot of the magnetization jump for  $J_2/J_1 = 1, 1.02$ , and  $1.04$ . The results for  $J_2/J_1 = 1$  and  $1.02$  are reported in the previous papers [3–5], and the results for  $J_2/J_1 = 1.04$  are the additional data. The critical fields  $h_1, h_2, h_3$ , and  $h_4$  are indicated by the red arrows. The magnetization jumps are seen at  $h_2$  and  $h_4$

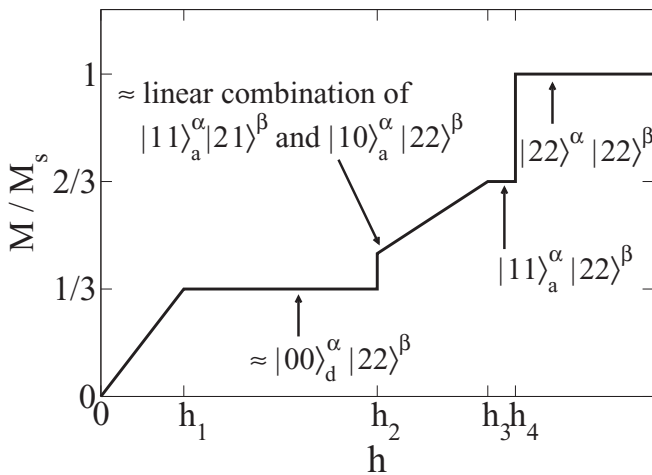


FIG. 4. Schematic figure of the magnetization process of the Heisenberg antiferromagnetic spins on square-kagome lattice. There is  $1/3$  plateau at  $h_1 \leq h \leq h_2$  and  $2/3$  plateau at  $h_3 \leq h \leq h_4$ . The magnetization jumps occur at  $h = h_2$  and  $h = h_4$

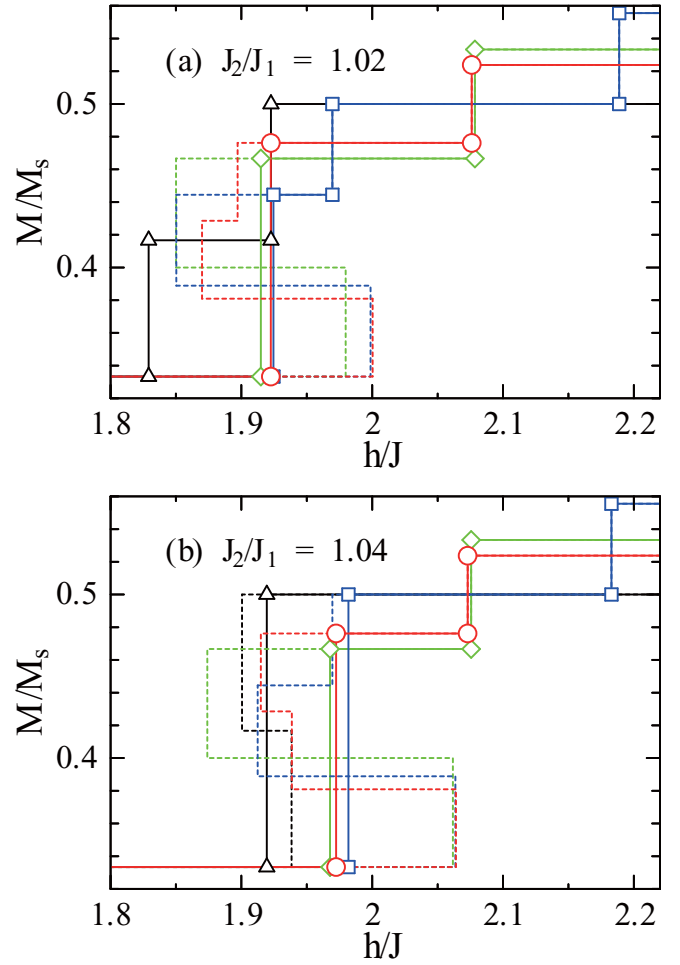


FIG. 5. Close-up plot of the magnetization process near  $1/3$  plateau on square-kagome lattice with (a)  $J_2/J_1 = 1.02$  and (b)  $J_2/J_1 = 1.04$  obtained by the exact diagonalization. The results of  $J_2/J_1 = 1.02$  were already reported in the previous paper [5], and these of  $J_2/J_1 = 1.04$  are added to study the  $J_2/J_1$  dependence on  $h_2$ . Black triangles, green diamonds, blue squares, and red circles are obtained in the systems of  $N_s = 24, 30, 36$ , and  $42$ , respectively. The broken lines represent the results before the Maxwell construction is carried out [5]. Magnetization jumps are seen in all cases except for the case of  $J_2/J_1 = 1.02$  and  $N_s = 24$ . Note that the critical value  $h_2$  depends very little on the size  $N_s$  at  $J_2/J_1 = 1.02$  and  $1.04$  except for  $N_s = 24$ .

and

$$\mathbf{S}^\beta = \mathbf{S}_5 + \mathbf{S}_6 + \mathbf{S}_7 + \mathbf{S}_8, \quad (6)$$

respectively. If the system preserves the translational symmetry,  $\mathbf{S}_5$  and  $\mathbf{S}_6$  should be the same as  $\mathbf{S}_7$  and  $\mathbf{S}_8$ , respectively. Since we are interested in the ground states in the magnetic field and the excited states from the plateau states, the translational symmetry may be broken in general. Since  $\beta$  spins belong to two unit cells simultaneously, the total spin in the unit cell is given by

$$\mathbf{S} = \mathbf{S}^\alpha + \frac{1}{2}\mathbf{S}^\beta. \quad (7)$$

When  $J_1 = 0$  we can obtain the eigenstates of the Hamiltonian ( $\mathcal{H}_2 + \mathcal{H}_{Zeeman}$ ) as shown in Fig. 6 (see Appendix A).

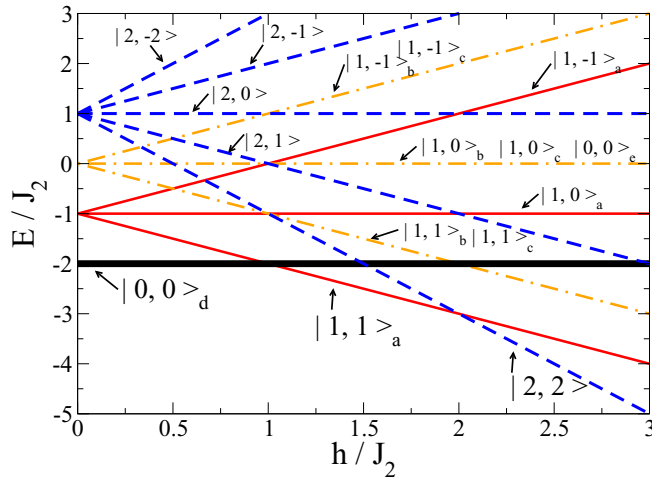


FIG. 6. Eigenvalues of  $\mathcal{H}_2 + \mathcal{H}_{Zeeman}$  of four  $\alpha$  spins as a function of external magnetic field.

We study the  $1/3$  plateau state and the magnetization jump at the higher edge of the magnetic field ( $h = h_2$ ) by using the approximate states of the entangled state in the unit cell. We also discuss the jump between  $2/3$  plateau and the saturated state at  $h = h_4$ , which is obtained exactly.

### A. $1/3$ plateau state at $h_1 < h < h_2$

When  $J_2 = 0$ , the square kagome lattice is the same as the Lieb lattice as seen in Fig. 2. The Lieb lattice has no frustration. The ground state of the Lieb lattice with classical spins at  $h = 0$  is the ferrimagnetic state, i.e., all  $\alpha$  spins are up and all  $\beta$  spins are down, resulting in the magnetization of  $1/3$  of the saturation value. Even if the spins are quantum spins with  $S = 1/2$ , the ferrimagnetic state with the  $1/3$  magnetization survives, although the amplitudes of the local spins are reduced by the quantum effects.

In the other limit of  $J_1 = 0$  at  $h = 0$ , four  $\alpha$  spins form the spin singlet state and the  $\beta$  spins are arbitrary.

When  $0 < J_1 \ll J_2$ , the effective interactions between  $\beta$  spins have been studied by Roussochatzakis *et al.* [2] using degenerate perturbation theory. They have shown that the ground state at  $J_1 \ll J_2$  and  $h = 0$  can be approximated by the singlet state of four  $\alpha$  spins and the crossed-dimer valence bond crystal state of  $\beta$  spins, resulting in the plateau at  $M = 0$  due to a finite spin gap. When  $J_1 \approx J_2$ , the ground states at  $h = 0$  are different from the ground state at  $J_1 \ll J_2$  and  $h = 0$  and are not definitely determined [2].

As we have shown previously [5], the  $1/3$  plateau state at  $h_1 \leq h \leq h_2$  for  $J_2/J_1 \lesssim 0.96$  is different from the states for  $J_2/J_1 \gtrsim 0.96$ . When  $J_2/J_1 \lesssim 0.96$ , the  $1/3$  plateau state is the ferrimagnetic state, similar to that in the Lieb lattice. When  $J_2/J_1 \gtrsim 0.96$ , the plateau state can be approximated by the similar state at  $J_1 = 0$ , i.e., the spin singlet state is formed by four  $\alpha$  spins and all  $\beta$  spins align up. The latter approximation is justified numerically for  $J_2/J_1 \gtrsim 1$ . The exact diagonalization studies [4,5] show that in the region of magnetic field  $0 < h < h_1$ ,  $\langle S_i^z \rangle$  ( $i \in \beta$ ) is obtained to be nearly proportional to  $h$ , while  $\langle S_i^z \rangle$  ( $i \in \alpha$ ) is almost zero. We approximate the  $1/3$  plateau state as the direct product state of  $|00\rangle_d^\alpha$  for four  $\alpha$  spins and

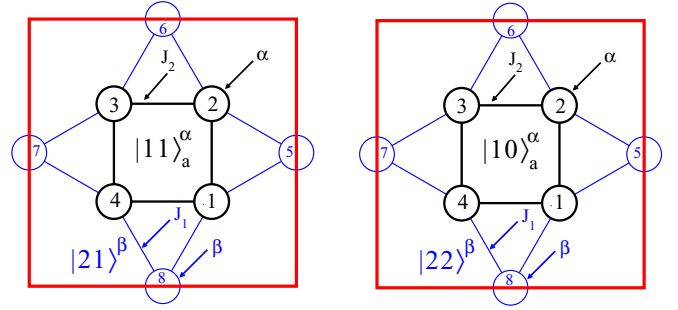


FIG. 7. Approximate state at  $h \gtrsim h_2$  is an entangled state of the two states of  $|11\rangle_a^\alpha |21\rangle^\beta$  and  $|10\rangle_a^\alpha |22\rangle^\beta$ .

all up states for  $\beta$  spins, i.e.,

$$|0,0\rangle_d^\alpha |2,2\rangle^\beta, \quad (8)$$

which can be justified if  $J_2/J_1 \gg 1$ . Although the condition  $J_2/J_1 \gg 1$  is not fulfilled in the present case, we treat  $\mathcal{H}_1$  as a perturbation. The magnetization of this state is

$$\frac{M}{M_s} = \frac{1}{3}. \quad (9)$$

The energy of this state is approximated as

$$E_h^{(h_1 \leq h \leq h_2)} \approx N_0(-2J_2 - h). \quad (10)$$

### B. Magnetization jump at $h = h_2$

When  $h$  is larger than  $h_2$ ,  $\alpha$  spins no longer stay a singlet state  $|0,0\rangle_d^\alpha$ . In order to increase the magnetization from  $1/3$  of the saturation, four  $\alpha$  spins should become one of the spin-triplet states, which may be  $|1,1\rangle_a^\alpha$  state, because this state has the lowest energy when  $J_1 = 0$  and  $J_2 < h < 2J_2$  (see Fig. 6). The  $z$  component of the  $\beta$  spins surrounding the  $\alpha$  spins may decrease the  $z$  component  $S_z^\beta$  by changing from  $|2,2\rangle^\beta$  to  $|2,1\rangle^\beta$ , as shown in the right figure in Fig. 7. However, this state is not the eigenstate of  $\mathcal{H}_1$  as shown in Appendix D. We approximate the eigenstate just above the magnetic field  $h_2$  as a linear combination of  $|11\rangle_a^\alpha |21\rangle^\beta$  and  $|10\rangle_a^\alpha |22\rangle^\beta$  (See Fig. 8).

The state at the field  $h$  just above the higher edge of the  $1/3$  plateau  $h_2$  ( $h = h_2 + 0$ ) is studied in Appendix D and the

$$\begin{aligned} |1,1\rangle_a^\alpha &= \frac{1}{2} \left( \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} - \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} + \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} - \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} \right) \\ |1,0\rangle_a^\alpha &= \frac{-1}{\sqrt{2}} \left( \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} - \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} \right) \\ |0,0\rangle_d^\alpha &= \frac{1}{\sqrt{12}} \left( \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} + \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} + \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} + \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} \right. \\ &\quad \left. - 2 \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} - 2 \begin{array}{c} \text{O} \quad \text{O} \\ \text{O} \quad \text{O} \end{array} \right) \end{aligned}$$

FIG. 8. Some eigenstates of  $\mathcal{H}_2 + \mathcal{H}_{Zeeman}$ . Open circles are up spins and filled circles are down spins at sites 1, 2, 3, and 4.

energy is approximately given by

$$E_h^{(h=h_2+0)} \approx N_0 \left( \frac{1}{4} J_1 - J_2 - \frac{5}{4} h - \frac{1}{4} \sqrt{h^2 - 2J_1 h + 9J_1^2} \right). \quad (11)$$

On the other hand the energy at the  $1/3$  plateau is approximated by Eq. (10). The upper edge of the  $1/3$  plateau is obtained by

$$E_h^{(h_1 \leq h \leq h_2)} = E_h^{(h=h_2+0)}. \quad (12)$$

Then we obtain

$$h_2 = \frac{(J_1 + J_2)(-J_1 + 2J_2)}{J_2} = 2J_1 \frac{(1 + \frac{\delta}{2})(1 + 2\delta)}{1 + \delta} \quad (13)$$

$$\approx 2J_1 \left( 1 + \frac{3}{2} \delta \right), \quad (14)$$

where

$$\delta = \frac{J_2 - J_1}{J_1}, \quad (15)$$

and we have assumed

$$0 \leq \delta \ll 1. \quad (16)$$

Although the absolute value of  $h_2$  given in Eq. (13) is a little bit deviated from the value obtained by the exact diagonalization [ $h_2 = 2$  in Eq. (13), while  $h_2 = 1.848$  is obtained by the exact diagonalization at  $J_2 = J_1$ ], the  $(J_2 - J_1)/J_1$  dependence of  $h_2$  is in good agreement between Eq. (13) and the exact diagonalization, as shown in Fig. 9. We will discuss the interaction between the excitations in the next section.

At the magnetic field just above  $h_2$ , the eigenstate is approximated as

$$|\Psi\rangle \approx \frac{-\sqrt{2}J_2}{\sqrt{J_1^2 + 2J_2^2}} |11\rangle_a^\alpha |21\rangle^\beta + \frac{J_1}{\sqrt{J_1^2 + 2J_2^2}} |10\rangle_a^\alpha |22\rangle^\beta. \quad (17)$$

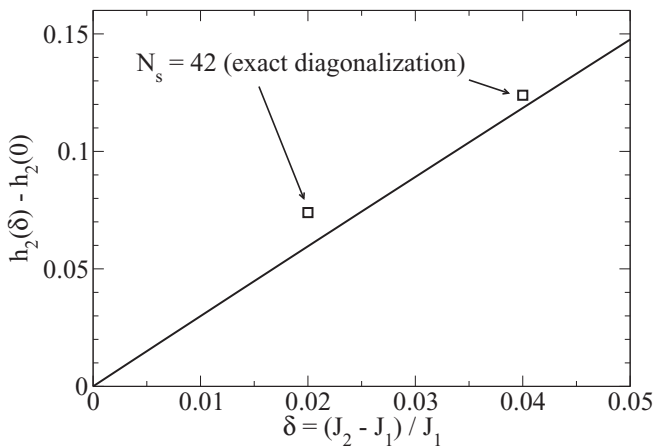


FIG. 9. The critical field  $h_2$  vs  $(J_2 - J_1)/J_1$  given in Eq. (13) (solid line) and the numerical results of the exact diagonalization in the system  $N_s = 42$  (squares).

The magnetization at  $h = h_2 + 0$  is

$$\begin{aligned} \langle m \rangle &= \frac{1}{J_1^2 + 2J_2^2} \left( \frac{1}{3} J_1^2 + J_2^2 \right) = \frac{4 \left( 1 + \frac{3}{2} \delta + \frac{3}{4} \delta^2 \right)}{9 \left( 1 + \frac{4}{3} \delta + \frac{2}{3} \delta^2 \right)} \\ &\approx \frac{4}{9} \left( 1 + \frac{1}{6} \delta \right). \end{aligned} \quad (18)$$

If the interaction between the excitations from the state at the  $1/3$  state were repulsive, the Bose-Einstein condensation of magnons would happen, which has been shown to be realized in several materials [37–41]. In that case the magnetization would increase continuously when the magnetic field is increased from  $h_2$ . However, as we will show numerically in the next section, the interaction between the excitations from the state at the  $1/3$  state is attractive. Then the excitations occur on every unit cell in the ground state at  $h = h_2 + 0$ . In this case the magnetization jumps from  $1/3$  to the value given in Eq. (18).

### C. $2/3$ plateau at $h_3 < h < h_4$ and the jump at $h = h_4$

We study the  $2/3$  magnetization plateau and jump at  $h = h_4$  in this subsection in order to make clear the mechanism of the jump in this system. All spins align to the  $z$  direction at  $h > h_4$ . This state is written as the direct product of the  $S^\alpha = 2, S_z^\alpha = 2$  state of four  $\alpha$  spins ( $|2, 2\rangle^\alpha$ ) and the  $S^\beta = 2, S_z^\beta = 2$  state of four  $\beta$  spins ( $|2, 2\rangle^\beta$ ). We write the state at  $h > h_4$  as

$$|2, 2\rangle^\alpha |2, 2\rangle^\beta. \quad (19)$$

The magnetization per unit cell is  $M_s/N_0 = 3$  ( $M/M^s = 1$ ) and the energy per unit cell is obtained as

$$\frac{E_h^{(h \geq h_4)}}{N_0} = (J_2 + 2J_1 - 3h). \quad (20)$$

When we decrease the magnetic field below  $h_4$ , the magnetization jumps from the fully polarized state ( $M/M_s = 1$ ) to the  $2/3$  plateau. This jump can be understood as follows. In this  $2/3$  plateau the spins at the  $\beta$  sites are aligned to the  $z$  direction, while the four  $\alpha$  spins form the spin triplet  $|1, 1\rangle_a^\alpha$ , since  $|1, 1\rangle_a^\alpha$  is the lowest state within  $S = 1, S_z = 1$  states for four  $\alpha$  spins (see Fig. 6). Note that both  $|2, 2\rangle^\alpha |2, 2\rangle^\beta$  and  $|1, 1\rangle_a^\alpha |2, 2\rangle^\beta$  are eigenstates of the Hamiltonian with the energy given as Eq. (20) and

$$\frac{E_h^{(h_3 \leq h \leq h_4)}}{N_0} = (-J_2 + J_1 - 2h), \quad (21)$$

respectively. In both states the shared  $\beta$  spins are all ups. Therefore, any spatially mixed states of  $|2, 2\rangle^\alpha |2, 2\rangle^\beta$  and  $|1, 1\rangle_a^\alpha |2, 2\rangle^\beta$  are also the eigenstates. If the fraction of  $p$  ( $0 \leq p \leq 1$ ) of the unit cells is the state  $|2, 2\rangle^\alpha |2, 2\rangle^\beta$  and  $(1 - p)$  of the unit cells is the state  $|1, 1\rangle_a^\alpha |2, 2\rangle^\beta$ , the energy is

$$\frac{E}{N_0} = p \frac{E_h^{(h \geq h_4)}}{N_0} + (1 - p) \frac{E_h^{(h_3 \leq h \leq h_4)}}{N_0}. \quad (22)$$

The lowest energy is obtained by  $p = 0$  for  $h < h_4$  and by  $p = 1$  for  $h > h_4$ , where the critical value  $h_4$  is obtained by the equation

$$E_{h_4}^{(h \geq h_4)} = E_{h_4}^{(h_3 \leq h \leq h_4)}. \quad (23)$$

We obtain

$$h_4 = J_1 + 2J_2. \quad (24)$$

In this subsection no approximation is used. A similar situation has been studied for the magnetization jump to the saturated magnetization in kagome lattice [6,7].

#### IV. INTERACTION BETWEEN EXCITATIONS

In this section we consider the interaction between excitations. We take  $x = M/M_s$ , where  $M$  is the magnetization and  $M_s$  is the saturation value of the magnetization,  $M_s = N_s/2$  ( $N_s$  is the number of sites). We define the energy  $E(x)$  as the lowest energy at  $h = 0$  (the eigenvalue of  $\mathcal{H}_1 + \mathcal{H}_2$ ) among the eigenstates having the same magnetization  $x$ . In the finite system,  $x$  can have the discrete values

$$x = \frac{2n}{N_s}, \quad (25)$$

where

$$n = 0, \pm 1, \pm 2, \dots, \pm \frac{N_s}{2}. \quad (26)$$

We have assumed that  $N_s$  is an even number. We define the lowest energy per site at  $h = 0$  among the states with the magnetization  $x$ ,

$$\epsilon(x) = \frac{E(x)}{N_s}. \quad (27)$$

In Fig. 10 we plot  $\epsilon(x)$  as a function of  $x$  obtained by the exact diagonalization with  $J_2/J_1 = 1.04$  and  $N_s = 36$ . The magnetization process is calculated as

$$\begin{aligned} h(x) &= E(x) - E\left(x - \frac{2}{N_s}\right) \\ &= N_s \left[ \epsilon(x) - \epsilon\left(x - \frac{2}{N_s}\right) \right]. \end{aligned} \quad (28)$$

In the limit of  $N_s \rightarrow \infty$ , it becomes

$$h(x) = 2 \frac{d\epsilon(x)}{dx}. \quad (29)$$

In the magnetization process, we plot the magnetization  $x$  as a function of  $h$ , as shown in Fig. 3.

In the region  $0 < x \leq 1/3$  ( $0 < h \leq h_1$ ), the graph of  $\epsilon(x)$  is downward convex (blue lines). This downward convex curvature means the repulsive interaction between the magnonlike excitations from the totally singlet state at  $h = 0$ . The plateau at  $x = 1/3$  corresponds to the kink of  $\epsilon(x)$  at  $x = 1/3$ . Above  $x = 1/3$  the graph of  $\epsilon(x)$  is upward convex (red lines) as shown in Fig. 10. Although the difference between the red lines connecting the nearest circles and the black broken line connecting the third-nearest circles is very small, it is much larger than the numerical errors of the exact diagonalization (relative errors should be less than  $10^{-10}$  for example). The downward convex curvature means that the attractive interaction works between the excitation from the plateau state. Thus the entangled states studied approximately in the previous section will be created in all unit cells, resulting in the finite jump in the magnetization. The straight line of  $\epsilon(x)$

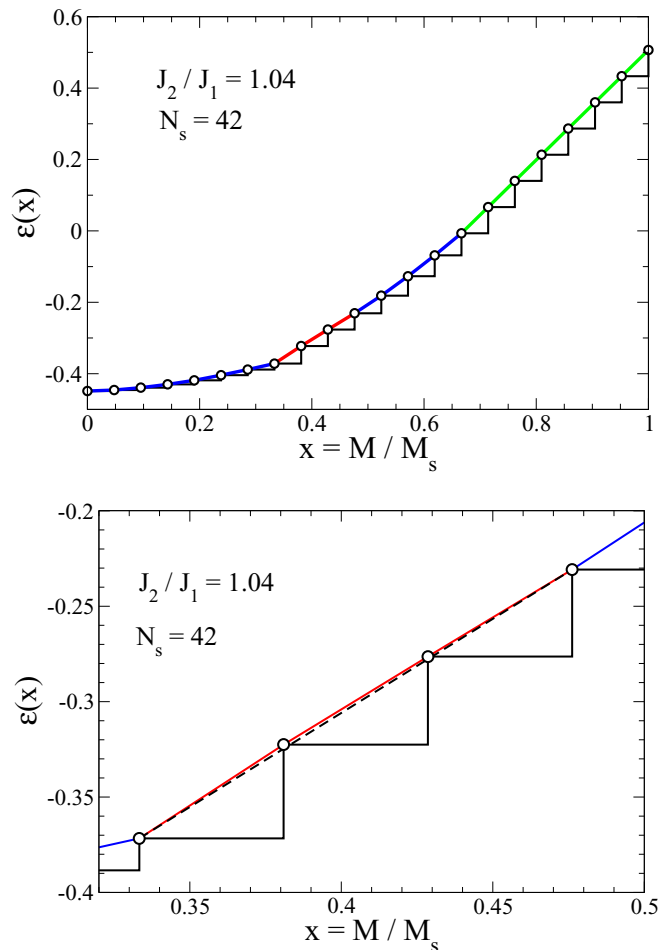


FIG. 10. Energy per site [ $\epsilon(x)$ ] as a function of  $x = M/M_s = n/21$  obtained by the exact diagonalization ( $N_s = 42$ ). At  $0 < x \leq 1/3$ ,  $\epsilon(x)$  is downward convex as shown by the blue line. A kink is seen at  $x = 1/3$ . At  $1/3 \leq x \leq 10/21$  the curvature is upward convex (the red line) and a little higher than the black broken line connecting the third-nearest circles at  $x = 1/3 = 7/21$  and  $x = 10/21$ . At  $10/21 \leq x \leq 2/3$ ,  $\epsilon(x)$  is again downward convex (the blue line). At  $x = 2/3$  there is a kink again, and all circles at  $2/3 \leq x \leq 1$  are on the straight green line.

at  $2/3 \leq x \leq 1$  is consistent with no interaction between the excitation from the  $2/3$  state or from the fully saturated state.

Finally, we would like to comment on the experimental situation. Although a good candidate material for the present system depicted in Fig. 1 was reported [42] and the numerical study was done [43], the material has a further additional distortion. Owing to this addition, the behavior of this material is different from the present result [43]. Even though there is such a difference, good candidate materials will be found in the near future.

#### V. CONCLUSION

In this paper we study the magnetization process of the Heisenberg antiferromagnet on the square kagome lattice by using the approximated wave function. We take the approximation that the ground state just above the higher edge of the  $1/3$  plateau is the entangled state of the  $S = 1$  triplet states of

the  $\alpha$  spins on the square and the  $S = 2$  quintet state of the  $\beta$  spins,  $|10\rangle_a^\alpha |22\rangle^\beta$  and  $|11\rangle_a^\alpha |21\rangle^\beta$ . Since the  $\beta$  spins are shared by neighboring unit cells, the magnetization of the entangled states depends on the coefficient of two states. In spite of the crude approximation taken in this paper, it gives the reasonable  $J_2/J_1$  dependence of the value of the critical field  $h_2$  and the magnitude of the magnetization jump, which are obtained by the exact diagonalization study. The approximation is justified when  $J_2/J_1 \gg 1$ . The reason it seems to work well even when  $J_2/J_1 \gtrsim 1$  would be the frustration, which reduces the effective coupling ( $J_1$ ) between the spins forming triangles with respect to the coupling ( $J_2$ ) in the spins forming squares without frustrations.

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### APPENDIX A: EIGENSTATES OF $\mathcal{H}_2 + \mathcal{H}_{\text{Zeeman}}$

The eigenstates of  $\mathcal{H}_2$  for four  $\alpha$  spins on the corner of the square is written as the linear combination of  $|\sigma_1, \sigma_2, \sigma_3, \sigma_4\rangle$ , where  $\sigma_j = \uparrow$  or  $\downarrow$ . The eigenstates are also written as  $|S, S_z\rangle^\alpha$ , where  $S$  is the total spin for four spins on  $\alpha$  sites and  $S_z$  is the  $z$  component of total spin. The same notations are used for the four  $\beta$  spins (5, 6, 7, and 8).

There are one  $S = 2$  quintet, three  $S = 1$  triplets and two  $S = 0$  singlets. The  $S = 2$  states are given as

$$|2, 2\rangle^\alpha = |\uparrow\uparrow\uparrow\uparrow\rangle, \quad (\text{A1})$$

$$|2, 1\rangle^\alpha = \frac{1}{2}(|\downarrow\uparrow\uparrow\uparrow\rangle + |\uparrow\downarrow\uparrow\uparrow\rangle + |\uparrow\uparrow\downarrow\uparrow\rangle + |\uparrow\uparrow\uparrow\downarrow\rangle), \quad (\text{A2})$$

$$|2, 0\rangle^\alpha = \frac{1}{\sqrt{6}}(|\uparrow\uparrow\downarrow\downarrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle + |\downarrow\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle), \quad (\text{A3})$$

$$|2, -1\rangle^\alpha = \frac{1}{2}(|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\uparrow\downarrow\downarrow\rangle + |\downarrow\downarrow\uparrow\downarrow\rangle + |\downarrow\downarrow\downarrow\uparrow\rangle), \quad (\text{A4})$$

$$|2, -2\rangle^\alpha = |\downarrow\downarrow\downarrow\downarrow\rangle. \quad (\text{A5})$$

We write the three  $S = 1$  triplets as  $|1, S_z\rangle_a$ ,  $|1, S_z\rangle_b$ , and  $|1, S_z\rangle_c$ , which are given by

$$|1, 1\rangle_a^\alpha = \frac{1}{2}(|\downarrow\uparrow\uparrow\uparrow\rangle - |\uparrow\downarrow\uparrow\uparrow\rangle + |\uparrow\uparrow\downarrow\uparrow\rangle - |\uparrow\uparrow\uparrow\downarrow\rangle), \quad (\text{A6})$$

$$|1, 0\rangle_a^\alpha = \frac{1}{\sqrt{2}}(-|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\uparrow\downarrow\downarrow\rangle), \quad (\text{A7})$$

$$|1, -1\rangle_a^\alpha = \frac{1}{2}(-|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\uparrow\downarrow\downarrow\rangle - |\downarrow\downarrow\uparrow\downarrow\rangle + |\downarrow\downarrow\downarrow\uparrow\rangle), \quad (\text{A8})$$

$$|1, 1\rangle_b^\alpha = \frac{1}{\sqrt{2}}(|\downarrow\uparrow\uparrow\uparrow\rangle - |\uparrow\uparrow\downarrow\uparrow\rangle), \quad (\text{A9})$$

$$|1, 0\rangle_b^\alpha = \frac{1}{2}(-|\uparrow\uparrow\downarrow\downarrow\rangle - |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle + |\downarrow\uparrow\uparrow\downarrow\rangle), \quad (\text{A10})$$

$$|1, -1\rangle_b^\alpha = \frac{1}{\sqrt{2}}(-|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\uparrow\downarrow\downarrow\rangle), \quad (\text{A11})$$

$$|1, 1\rangle_c^\alpha = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\uparrow\uparrow\rangle - |\uparrow\uparrow\uparrow\downarrow\rangle), \quad (\text{A12})$$

$$|1, 0\rangle_c^\alpha = \frac{1}{2}(-|\uparrow\uparrow\downarrow\downarrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle - |\downarrow\uparrow\uparrow\downarrow\rangle), \quad (\text{A13})$$

$$|1, -1\rangle_c^\alpha = \frac{1}{\sqrt{2}}(-|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\downarrow\downarrow\uparrow\rangle). \quad (\text{A14})$$

We write two  $S = 0$  singlets as  $|0, 0\rangle_d$  and  $|0, 0\rangle_e$ , which are given by

$$|0, 0\rangle_d^\alpha = \frac{1}{\sqrt{12}}(|\uparrow\uparrow\downarrow\downarrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle + |\downarrow\uparrow\uparrow\downarrow\rangle - 2|\uparrow\downarrow\uparrow\downarrow\rangle - 2|\downarrow\uparrow\downarrow\uparrow\rangle), \quad (\text{A15})$$

$$|0, 0\rangle_e^\alpha = \frac{1}{2}(|\uparrow\uparrow\downarrow\downarrow\rangle - |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle - |\downarrow\uparrow\uparrow\downarrow\rangle). \quad (\text{A16})$$

The  $S = 2$  states are the eigenstates of  $\mathcal{H}_2$  with the eigenvalue  $J_2$ ,

$$\mathcal{H}_2|2, S_z\rangle^\alpha = J_2 N_0 |2, S_z\rangle^\alpha, \quad (\text{A17})$$

where  $S_z = 2, 1, 0, -1$ , or  $-2$ . One of the  $S = 1$  states ( $|1, S_z\rangle_a^\alpha$ ) has the eigenvalue  $-J_2$ , and the other two  $S = 1$  states ( $|1, S_z\rangle_b^\alpha$  and  $|1, S_z\rangle_c^\alpha$ ) have eigenvalue 0,

$$\mathcal{H}_2|1, S_z\rangle_a^\alpha = -J_2 N_0 |1, S_z\rangle_a^\alpha, \quad (\text{A18})$$

$$\mathcal{H}_2|1, S_z\rangle_b^\alpha = 0, \quad (\text{A19})$$

$$\mathcal{H}_2|1, S_z\rangle_c^\alpha = 0, \quad (\text{A20})$$

where  $S_z = \pm 1$  or 0.

Two  $S = 0$  eigenstates ( $|0,0\rangle_d^\alpha$  and  $|0,0\rangle_e^\alpha$ ) have eigenvalues  $-2J_2$  and  $0$ , respectively,

$$\mathcal{H}_2|0,0\rangle_d^\alpha = -2J_2N_0|0,0\rangle_d^\alpha, \quad (\text{A21})$$

$$\mathcal{H}_2|0,0\rangle_e^\alpha = 0. \quad (\text{A22})$$

In Fig. 6 the eigenvalues of  $\mathcal{H}_2 + \mathcal{H}_{\text{Zeeman}}$  are plotted as a function of the external magnetic field  $h$ . The ground state of  $\mathcal{H}_2 + \mathcal{H}_{\text{Zeeman}}$  is  $|0,0\rangle_d^\alpha$ ,  $|1,1\rangle_a^\alpha$ , and  $|2,2\rangle^\alpha$  for  $0 < h < J_2$ ,  $J_2 < h < 2J_2$ , and  $h > 2J_2$ , respectively.

### APPENDIX B: 2/3 PLATEAU AND $h_4$

In this appendix we calculate the critical magnetic field  $h_4$  at which the state changes from the 2/3-plateau state to the state of all spins up. The 2/3-plateau state is the state in which four  $\alpha$  spins (sites 1–4 in Fig. 1) form the  $|1,1\rangle_a^\alpha$  state and all  $\beta$  spins are up state  $(|2,2\rangle)^\beta$ . We consider sites 1–8 in Fig. 1. Then we can write  $\mathcal{H}_1$  as

$$\mathcal{H}_1 = J_1 \sum_{(i,j):i=1\sim 4,j=5\sim 8} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (\text{B1})$$

For example, we consider the term containing  $S_5$  in Eq. (B1),

$$J_1 \left[ \frac{1}{2} [S_5^-(S_1^+ + S_2^+) + S_5^+(S_1^- + S_2^-)] + S_5^z(S_1^z + S_2^z) \right]. \quad (\text{B2})$$

Since

$$(S_1^+ + S_2^+)|1,1\rangle_a^\alpha = 0, \quad (\text{B3})$$

and

$$S_5^+|2,2\rangle^\beta = 0, \quad (\text{B4})$$

we can show that the state of the direct product of  $|1,1\rangle_a^\alpha$  and all up states of spins 5–8 ( $|2,2\rangle^\beta$ ) are the eigenstates of  $\mathcal{H}_1$  and  $\mathcal{H}$ . We obtain the energy of this state as

$$E_h^{(h_3 \leq h \leq h_4)} = N_0(J_1 - J_2 - 2h), \quad (\text{B5})$$

where  $N_0$  is the number of the unit cell.

At  $h > h_4$  all spins are aligned to the  $z$  direction and the energy is

$$E_h^{(h \geq h_4)} = N_0(2J_1 + J_2 - 3h). \quad (\text{B6})$$

The magnetization jump from  $M/M_s = 2/3$  to 1 occurs at  $h = h_4$ , at which

$$E_h^{(h_3 \leq h \leq h_4)} = E_h^{(h \geq h_4)}. \quad (\text{B7})$$

We obtain

$$h_4 = J_1 + 2J_2. \quad (\text{B8})$$

### APPENDIX C: $0 \leq h < h_1$ AND $h_1 < h < h_2$

When  $h = 0$ , the true ground state should be the total singlet state of all spins, and the ground state at small  $h$  might be a complicated state. We do not address the ground state at small  $h$  in detail in this paper. However, as shown by numerical study [4,5], in the region of magnetic field  $0 < h < h_1$ ,  $\langle S_i^z \rangle$  is nearly proportional to  $h$  for  $i \in \beta$ , while it is almost zero for  $i \in \alpha$ . We may take a simplified picture that the state for the  $\alpha$  spins is approximated as the singlet  $|00\rangle_d^\alpha$ . This approximation is justified if  $J_1 \ll J_2$ , since  $|00\rangle_d^\alpha$  is the ground state for

$\mathcal{H}_2 + \mathcal{H}_{\text{Zeeman}}$  for  $h < J_2$ , as shown in Fig. 6. Although the condition  $J_1 \ll J_2$  is not fulfilled in the present case, we treat  $\mathcal{H}_1$  as a perturbation. In the region  $0 < h < h_1$  the system is considered in the state that the  $\alpha$  spins make  $|00\rangle_d^\alpha$  and the locally excited  $\beta$  spin from the singlet state extend over the system forming a spin-wave-like state with the repulsive interaction between excitations. If there were no interactions between the excitations as in the case at  $h = h_4$  discussed in Appendix B, or if there were attractive interaction between the excitations, the magnetization jump would occur.

In the 1/3-plateau region ( $h_1 < h < h_2$ ), the ground state is approximated by the direct product of  $|00\rangle_d^\alpha$  for four  $\alpha$  spins and all  $\beta$  spins are aligned up, i.e.,  $|2,2\rangle^\beta$ . The energy of this state is approximated as

$$E_h^{(h_1 \leq h \leq h_2)} \approx N_0(-2J_2 - h). \quad (\text{C1})$$

### APPENDIX D: $h \gtrsim h_2$

In this appendix we show the matrix elements of  $\mathcal{H}_1$  between the eigenstates of  $\mathcal{H}_2$  at the magnetic field just above  $h_2$ .

Using the definition of  $|1S_z\rangle_a^\alpha$ ,  $|1S_z\rangle_b^\beta$ , and  $|1S_z\rangle_c^\alpha$ , we obtain

$$\begin{aligned} \mathcal{H}_1|11\rangle_a^\alpha = J_1 & \left[ \frac{1}{2\sqrt{2}}|10\rangle_a^\alpha S_a^{+\beta} + \frac{1}{4}|10\rangle_b^\alpha S_b^{+\beta} \right. \\ & + \frac{1}{4}|10\rangle_c^\alpha S_c^{+\beta} + \frac{1}{2}|11\rangle_a^\alpha S_a^{z\beta} + \frac{1}{2\sqrt{2}}|11\rangle_b^\alpha S_b^{z\beta} \\ & \left. + \frac{1}{2\sqrt{2}}|11\rangle_c^\alpha S_c^{z\beta} \right], \quad (\text{D1}) \end{aligned}$$

$$\begin{aligned} \mathcal{H}_1|10\rangle_a^\alpha = J_1 & \left[ \frac{1}{2\sqrt{2}}|11\rangle_a^\alpha S_a^{-\beta} + \frac{1}{4}|11\rangle_b^\alpha S_b^{-\beta} + \frac{1}{4}|11\rangle_c^\alpha S_c^{-\beta} \right. \\ & + \frac{1}{2}|1-1\rangle_a^\alpha S_a^{+\beta} + \frac{1}{2\sqrt{2}}|1-1\rangle_b^\alpha S_b^{+\beta} \\ & \left. + \frac{1}{2\sqrt{2}}|1-1\rangle_c^\alpha S_c^{+\beta} \right], \quad (\text{D2}) \end{aligned}$$

$$\begin{aligned} \mathcal{H}_1|1-1\rangle_a^\alpha = J_1 & \left[ \frac{1}{2\sqrt{2}}|10\rangle_a^\alpha S_a^{-\beta} + \frac{1}{4}|10\rangle_b^\alpha S_b^{-\beta} \right. \\ & + \frac{1}{4}|10\rangle_c^\alpha S_c^{-\beta} + \frac{1}{2}|1-1\rangle_a^\alpha S_a^{z\beta} \\ & \left. + \frac{1}{2\sqrt{2}}|1-1\rangle_b^\alpha S_b^{z\beta} + \frac{1}{2\sqrt{2}}|1-1\rangle_c^\alpha S_c^{z\beta} \right], \quad (\text{D3}) \end{aligned}$$

where  $\mathbf{S}_a^\beta$ ,  $\mathbf{S}_b^\beta$ , and  $\mathbf{S}_c^\beta$  are the spin operators for the  $\beta$  spins defined by

$$\mathbf{S}_a^\beta = \mathbf{S}^\beta = \mathbf{S}_5 + \mathbf{S}_6 + \mathbf{S}_7 + \mathbf{S}_8, \quad (\text{D4})$$

$$\mathbf{S}_b^\beta = -\mathbf{S}_5 + \mathbf{S}_6 + \mathbf{S}_7 - \mathbf{S}_8, \quad (\text{D5})$$

and

$$\mathbf{S}_c^\beta = \mathbf{S}_5 + \mathbf{S}_6 - \mathbf{S}_7 - \mathbf{S}_8. \quad (\text{D6})$$

The  $z$  component, the raising operator, and the lowering operator of  $\mathbf{S}_a^\beta$ ,  $\mathbf{S}_b^\beta$ , and  $\mathbf{S}_c^\beta$ , are defined as usual, for example,

$$S_a^{\pm\beta} = (S_5^x + S_6^x + S_7^x + S_8^x) \pm i(S_5^y + S_6^y + S_7^y + S_8^y). \quad (\text{D7})$$



Since  $|1S_z)_b^\alpha$ ,  $|1S_z)_c^\alpha$ , and  $|1-1)_a^\alpha$  have higher energy than  $|11)_a^\alpha$  and  $|10)_a^\alpha$ , we restrict ourselves in the subspace in  $|11)_a^\alpha$  and  $|10)_a^\alpha$  and neglect other states. Then  $\mathcal{H}_1$  is approximated in the basis of  $|11)_a^\alpha|21)^\beta$  and  $|10)_a^\alpha|22)^\beta$  as

$$\mathcal{H}_1 \approx \frac{1}{2} J_1 \begin{pmatrix} S_a^{z\beta} & \frac{1}{\sqrt{2}} S_a^{+\beta} \\ \frac{1}{\sqrt{2}} S_a^{-\beta} & 0 \end{pmatrix}. \quad (\text{D8})$$

Since

$${}^\beta \langle 21|S_a^{z\beta}|21)^\beta = 1, \quad (\text{D9})$$

and

$${}^\beta \langle 22|S_a^{+\beta}|21)^\beta = {}^\beta \langle 21|S_a^{-\beta}|22)^\beta = 2, \quad (\text{D10})$$

we obtain

$$\mathcal{H}_1 \approx J_1 \begin{pmatrix} \frac{1}{2} & \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} & 0 \end{pmatrix}. \quad (\text{D11})$$

In this subspace ( $|10)_a^\alpha|22)^\beta$  and  $|11)_a^\alpha|21)^\beta$ ) the Hamiltonian is approximated as

$$\mathcal{H}^{(h=h_2+0)} \approx \begin{pmatrix} \frac{1}{2} J_1 - J_2 - \frac{3}{2} h & \frac{1}{\sqrt{2}} J_1 \\ \frac{1}{\sqrt{2}} J_1 & -J_2 - h \end{pmatrix}. \quad (\text{D12})$$

The eigenvalues of Eq. (D12) are

$$E_h^{(h=h_2+0)} = N_0 \left( \frac{1}{4} J_1 - J_2 - \frac{5}{4} h \pm \frac{1}{4} \sqrt{h^2 - 2J_1 h + 9J_1^2} \right). \quad (\text{D13})$$

We take the minus sign for the square root, since the state with lower energy is realized.

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