Collapse of Ferrimagnetism in Two-Dimensional Heisenberg Antiferromagnet due to Frustration

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We study ferrimagnetism in the ground state of the antiferromagnetic Heisenberg model on the spatially anisotropic kagome lattice, in which ferrimagnetism of the conventional Lieb-Mattis type appears in the region of weak frustration whereas the ground state is nonmagnetic in the isotropic case. Numerical diagonalizations of small finite-size clusters are carried out to examine the spontaneous magnetization. We find that the spontaneous magnetization changes continuously in the intermediate region between conventional ferrimagnetism and the nonmagnetic phase. Local magnetization of the intermediate state shows strong dependence on the site position, which suggests non-Lieb-Mattis ferrimagnetism.

KEYWORDS: antiferromagnetic Heisenberg spin model, ferrimagnetism, frustration, numerical-diagonalization method, Lanczos method

Ferrimagnetism has been studied extensively as an important phenomenon that has both ferromagnetic nature and antiferromagnetic nature at the same time. One of the fundamental keys to understanding ferrimagnetism is the Marshall-Lieb-Mattis (MLM) theorem. This theorem clarifies some of the magnetic properties in the ground state of a system when the system has a bipartite lattice structure and when a spin on one sublattice interacts antiferromagnetically with a spin on the other sublattice. Under the condition that the sum of the spin amplitudes of spins in each sublattice is different between the two sublattices, one finds that the ground state of such a system exhibits ferrimagnetism. In this ferrimagnetic ground state, spontaneous magnetization is realized and its magnitude is a simple fraction of the saturated magnetization. We hereafter call ferrimagnetism of this type the Lieb-Mattis (LM) type.

Some studies in recent years, on the other hand, reported cases when the magnitude of the spontaneous magnetization of the ferrimagnetism is not a simple fraction of the saturated magnetization. The ferrimagnetic ground state of this type is a nontrivial quantum state whose behavior is difficult to explain well only within the classical picture. Ferrimagnetism of this type was first predicted in ref. 10 using the quantum rotor model. The mechanism of this ferrimagnetism has not been understood sufficiently up to now. Hereafter, we call this case the non-Lieb-Mattis (NLM) type. Note that in the cases when NLM ferrimagnetism is present, the structure of the lattices is limited to being one-dimensional. Recall that the above conditions of the MLM theorem do not include the spatial dimension of the system; the MLM theorem holds irrespective of the spatial dimensionality. We are then faced with a question: can NLM ferrimagnetism be realized when the spatial dimension is more than one?

The purpose of this letter is to answer the above question concerning the existence of NLM ferrimagnetism in higher dimensions. In this letter, we consider a case when we introduce a frustrating interaction into a two-dimensional lattice whose interactions satisfy the conditions of the MLM theorem. When the frustrating interaction is small, ferrimagnetism of the LM type survives; however, the ferrimagnetism is destroyed with the increase in the frustrating interaction and the system finally becomes nonmagnetic due to the considerably large frustrating interaction. We examine the behavior of the collapse of the ferrimagnetism and the existence of an intermediate region between the LM ferrimagnetic and nonmagnetic phases by means of the numerical-diagonalization method applied to finite-size clusters. Our study of the two-dimensional system successfully clarifies the existence of the intermediate phase and captures a feature of NLM ferrimagnetism.

First, we explain the model Hamiltonian examined in this letter. The Hamiltonian is given by

\[ \mathcal{H} = \sum_{i \in A, j \in B} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i \in A, j \in B'} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i \in B, j \in B'} J_2 \mathbf{S}_i \cdot \mathbf{S}_j, \]  

where \( \mathbf{S}_i \) denotes an \( S = 1/2 \) spin operator at site \( i \). Sublattices A, B, and B' and the network of antiferromagnetic interactions \( J_1 \) and \( J_2 \) are depicted in Fig. 1. Here, we consider the case of isotropic interactions. The system size is denoted by \( N_s \); the saturation magnetization is \( M_{\text{sat}} = N_s / 2 \). Energies are measured in units of \( J_1 \); thus, we take \( J_1 = 1 \) hereafter. We examine the properties of this model in the range of \( 0 < J_2 / J_1 \leq 1 \).

Note that in the case of \( J_2 = 0 \), sublattices B and B' are combined into a single sublattice; the system satisfies the above conditions of the MLM theorem. Thus, ferrimagnetism of the LM type is exactly realized in this case. In the case of \( J_2 = J_1 \), on the other hand, the lattice of the system is reduced to the kagome lattice. The ground state of the system on the kagome lattice...
without a magnetic field is known to be singlet from numerical-diagonalization studies,\textsuperscript{12–15} which indicates that the ground state is nonmagnetic. One thus finds that LM ferrimagnetism collapses between $J_2 = 0$ and $J_2 = J_1$. Consequently, we survey the region between the two cases.

Next, we discuss the method we use here, which is numerical diagonalization based on the Lanczos algorithm. It is known that this method is nonbiased beyond any approximations and reliable for many-body problems such as the present model. A disadvantage of this method is that the available system sizes are limited to being small because the dimension of the matrix grows exponentially with respect to the system size. To treat systems that are as large as possible, we have developed parallelization in our numerical calculations using the OpenMP and MPI techniques, either separately or in a hybrid way.\textsuperscript{16}

In this letter, we treat the finite-size clusters depicted in Fig. 2 when the system sizes are $N_s = 12$, $N_s = 24$, $N_s = 27$, and $N_s = 30$ under the periodic boundary condition and $N_s = 33$ under the open boundary condition. Note that each of these clusters forms a regular square although clusters (b) and (d) are tilted. The next larger size under the condition that a regular square is formed is $N_s = 48$, which is too large to handle using the present method, even when one uses modern supercomputers.

Before our numerical-diagonalization results for the finite-size clusters are presented, let us consider the directions of the spins in the ground state within the classical picture. We here examine the spin directions of classical vectors with length $S$ depicted in Fig. 3. One obtains the energy of the spin state with angle $\theta$ to be $E/J_1 = -2N_s/3S^2[2\cos(\pi - \theta) + (J_2/J_1)\cos(2\theta)]$. This expression of the energy indicates that for $J_2/J_1 \leq 1/2$, the state of $\theta = 0$, namely, ferrimagnetism of the LM type, is realized. Thus, the normalized magnetization of this state is $M/M_{\text{sat}} = 1/3$. One finds, on the other hand, that for $J_2/J_1 > 1/2$, the lowest-energy state is realized for nonzero $\theta$ when $J_1/J_2 = 2\cos(\theta)$ is satisfied. The normalized magnetization of this state is $M/M_{\text{sat}} = (J_2/J_1 - 1)/3$. When $J_2/J_1$ becomes unity, the magnetization finally vanishes. This classical argument will be compared with our finite-size results obtained from numerical diagonalizations.

Now, let us present our numerical results for the quantum case. First, we show our data for the lowest energy in each subspace of $S_z^{\text{tot}}$, which reveal the magnetization of the systems. Figure 4 depicts our results for the system with $N_s = 30$ depicted in Fig. 2(d). Note that $M_{\text{sat}} = 15$ in this case. For $J_2/J_1 = 0.5$, the energies from $S_z^{\text{tot}} = 0$ to $S_z^{\text{tot}} = 5$ are numerically identical, which means that $M/M_{\text{sat}}$ becomes $1/3$ and that ferrimagnetism of the LM type is realized. For $J_2/J_1 = 1$, the energy for $S_z^{\text{tot}} = 0$ is lower than the other energies for larger $S_z^{\text{tot}}$. The ground state of this case is nonmagnetic. For $J_2/J_1 = 0.6$, the energies from $S_z^{\text{tot}} = 0$ to $S_z^{\text{tot}} = 2$ are the same; thus, we find that the spontaneous magnetization is $M = 2$, which is smaller than the value for ferrimagnetism of the LM type. One finds that a state with intermediate magnetization appears between LM-type ferrimagnetism and the nonmagnetic state, at least according to the finite-size calculations.

Next, we examine the region of such an intermediate
condition is close to that for the $N_s = 24$ case under the periodic boundary condition. This is consistent with the fact that there are 21 sites in the inner part of cluster (c) of $N_s = 33$ under the open boundary condition. Our present results for both boundary conditions imply that the presence of the intermediate phase is irrespective of the boundary conditions.

An important characteristic of NLM ferrimagnetism is that the local magnetization in sublattice exhibits long-distance periodicity, which is absent in LM-type ferrimagnetism. Note that one cannot detect this periodicity in the cases under the periodic boundary condition. We thus examine the local magnetization in the intermediate phase for the case under the periodic boundary condition; the results for $N_s = 33$ are depicted in Fig. 6. For $J_2/J_1 = 0.5$ with LM-type ferrimagnetism, the local magnetization shows weak dependence on the position of sites, although $\langle S_1^z \rangle$ at edge sites 1 and 6 is slightly larger than those at interior sites, where the site numbers are illustrated in the inset of Fig. 6. This small difference originates from the edge effect due to the open boundary condition. For $J_2/J_1 = 0.5$, the edge effect does not seem to affect $\langle S_1^z \rangle$ at internal sites. For $J_2/J_1 = 0.53$ and 0.57, on the other hand, $\langle S_1^z \rangle$ at edge sites 1 and 6 becomes very small. The behavior of this appearance of the edge effect is different from the case of $J_2/J_1 = 0.5$. For $J_2/J_1 = 0.53$ and 0.57, one finds a strength dependence of $\langle S_1^z \rangle$ on the position of the site from site 2 to site 5. For $J_2/J_1 = 0.53$, $\langle S_1^z \rangle$ at sites next to the edges seems to be affected by the edge sites. It is unclear whether or not the case of $J_2/J_1 = 0.53$ corresponds to NLM type ferrimagnetism at present. For $J_2/J_1 = 0.57$, on the other hand, the strong dependence on the site position suggests the existence of origins that are different from the edge effect.

The system size $N_s = 33$ is sufficiently small for long-distance periodicity to be observed clearly. Although the present results are not decisive evidence of the periodicity, our finding of the large change in $\langle S_1^z \rangle$ is considered as possible evidence. In order to obtain decisive evidence,
calculations on systems of larger sizes are required, which are unfortunately difficult at the present time. Instead of the present two-dimensional kagome case, we are now examining a quasi-one-dimensional system on a kagome stripe lattice. Both systems partly share the same lattice structure. The system on the kagome stripe lattice reveals the clear appearance of NLM ferrimagnetism in the intermediate region. Results will be published elsewhere.

The phenomenon of ground-state magnetization changing continuously with respect to a continuous parameter in a model Hamiltonian has been reported in other cases. Tonegawa and co-workers reported such a phenomenon in spin systems with anisotropic interactions. It is unclear at present whether or not the states of this continuously changing magnetization in the anisotropic case show long-distance periodicity because the behavior of local magnetization has not been investigated yet. Since this phenomenon disappears in the isotropic case when the quantum effect is stronger than that in the anisotropic case, this phenomenon is considered to arise from the anisotropy. From this point of view, the origin of this phenomenon seems to be different from that of intermediate ferrimagnetism in the isotropic case studied here. Another reported phenomenon is partial ferromagnetism in the Hubbard model when the system is hole-doped near the half-filled Mott insulator. The origin of this phenomenon has been clarified to be the formation of spin polarons around doped holes. The mechanism of these two cases is different from the present case of NLM ferrimagnetism.

Finally, we briefly discuss possible future experiments. For volborthite, eq. (1) was proposed as a model Hamiltonian from the argument of its crystal structure, although NLM ferrimagnetism has not yet been observed in this material. A theoretical study on the spatial anisotropy of this material indicated that the deviation of the anisotropy from the isotropic kagome point is not particularly large. This is consistent with our present result because the nonmagnetic ground state is realized around the region of weak anisotropy as shown in Fig. 5. In order to observe NLM ferrimagnetism experimentally, it is necessary to realize a case with larger anisotropy. The measurement of volborthite under high pressure in the direction of the a-axis or discoveries of new materials might lead to such an observation.

In summary, we have clearly shown the existence of a ground state of non-Lieb-Mattis type ferrimagnetism in a two-dimensional lattice that lies between the well-known Lieb-Mattis type ferrimagnetic phase and the nonmagnetic phase including the kagome-lattice system. The nontrivial ferrimagnetism we have found in the intermediate phase occurs as a consequence of magnetic frustration. Our present result indicates that non-Lieb-Mattis ferrimagnetism is a general phenomenon irrespective of the spatial dimensionality.

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