Helical Ordering of Spin Trimers Found in a Distorted Kagome Lattice

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ABSTRACT

In magnetic materials with geometrically frustrated interactions in triangular, kagome, or pyrochlore lattice systems, various kinds of nontrivial orderings are observed; e.g., non-collinear and incommensurate spin structures, as well as successive transitions through partially disordered states. Most of these orderings are difficult to predict because in general a large number of spin structures are degenerate. Recently, we have found a novel ordering phenomenon realized in a distorted kagome lattice of S = 7/2 4f-spins of Gd ions in $Gd_3Ru_4Al_{12}$, which effectively can be considered to be a triangular lattice of S = 21/2 spin trimers. First, it is very rare that quantum mechanical multimerization is realized in localized and metallic f-electron systems. Second, it is significant that a spontaneous chiral symmetry breaking was found in the process of sinusoidal to helical successive phase transitions.

INTRODUCTION

Cooperative phenomenon among interacting electrons gives rise to a diversity of spontaneous ordering structures. Even in simple structures of ferromagnetic and antiferromagnetic orderings, where electron spins align in parallel and antiparallel ways, respectively, the details of the magnetic exchange interactions are widely varied. If the magnetic ions are located on a triangular or kagome lattice, and if the exchange interaction is antiferromagnetic, one cannot find a simple structure to satisfy the interactions consistently. In such cases, the system usually ends up with a so-called 120˚ structure as a result of compromises being made. In the crystal structure of $Gd_3Ru_4Al_{12}$ shown in Fig. 1, with a distorted kagome, or breathing kagome lattice, one may simply speculate that a 120˚ spin structure should be realized.

SPIN TRIMER FORMATION

In $Gd_3Ru_4Al_{12}$, Nakamura *et al.* recently pointed out that the Gd3+ spins of S = 7/2 on the nearest neighbor triangle form a “spin trimer” state with S = 21/2 [1]. They showed that the anomalous temperature dependence of the magnetic susceptibility and specific heat can be well explained by a spin trimer model H = $J(S_1 \bullet S_2 + S_1 \bullet S_2 + S_1 \bullet S_2)$ with a ferromagnetic exchange constant of J=13.5 K. Although such quantum spin states are frequently observed in insulating d-electron systems with small spin moments, they are rarely observed in mostly metallic f-electron systems with relatively large angular moments. Only one exception is a spin dimer formation in $YbAl_3C_3$ [2]. The spin trimer formation in $Gd_3Ru_4Al_{12}$ may be the first known case in f-electron systems.

HELICAL ORDERING OF SPIN TRIMERS

Another interesting property found in $Gd_3Ru_4Al_{12}$ is the successive magnetic phase transitions at 18.5 K and 17.5 K. Since the binding energy of the spin trimer is higher than 100 K, these phase transitions at low temperatures are considered to be the orderings among well developed spin trimers. To investigate the ordered spin structure, we have utilized a resonant X-ray diffraction method using synchrotron radiation at the Photon Factory, High Energy Accelerator Research Organization, in Tsukuba, Japan. Element and orbital selectivity by using X-ray energies near the absorption edge of the target element, effective usage of a polarized incident beam and polarization analysis, high Q-resolution, applicability of tiny samples, and applicability to the neutron absorbing elements like Gd, are the advantages of resonant X-ray diffraction over neutron diffraction, which is a typical method used to investigate magnetic structures.

Through careful measurements and data analysis, especially from the results of the polarization analysis, we concluded that the transition at 18.5 K is an ordering of spin trimers from a paramagnetic to sinusoidal structure, and that the transition at 17.5 K is a transition from a sinusoidal to helical structure; these transitions are shown in Fig. 1.

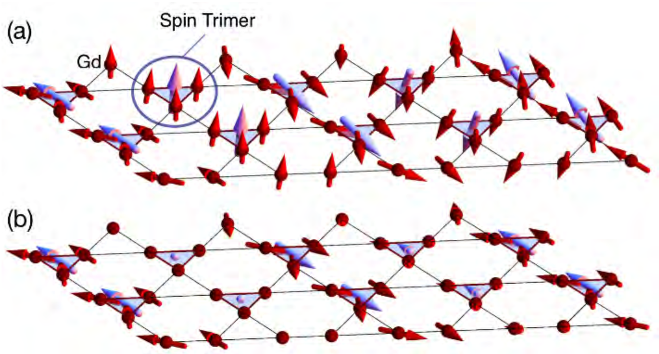

Fig.1: (a) Helical trimer spin structure below 17.5 K, propagating along the a*-axis. The total spins of each trimer is represented by the bigger arrows at the center of colored triangles. (b) Sinusoidal trimer spin structure between 17.5 K and 18.5 K.

An important problem of this sinusoidal structure is that there remain magnetic sites with small or even vanishing ordered moments. This means that unreleased magnetic entropy or degeneracy remains, which must be lifted at lower temperatures. The sinusoidal structure just below the Néel order reflects the weak anisotropy in the c-plane and the preferable propagation vector of (0.27, 0, 0) for the magnetic exchange interaction via the conduction electrons. However, it is not preferable to maintain this collinear structure down to lower temperatures due to the thermodynamic reason of magnetic entropy.

The spin system of $Gd_3Ru_4Al_{12}$ chooses to become helical below 17.5 K by inducing the c-axis spin component, i.e., by transforming the structure into a non-collinear form. In other words, the chiral degeneracy in the sinusoidal structure is lifted spontaneously by the transition to the helical structure, which allows all the Gd spins to fully develop.

Chirality plays an important role in a wide range of fields in nature, from biology, chemistry, and particle physics to materials science. Recently, in magnetic materials with a chiral crystal structure, the emergence of nontrivial chiral objects such as skyrmions and chiral soliton lattices have been attracting wide interest both for applications and basic science. Although the crystal structure of $Gd_3Ru_4Al_{12}$ is not chiral, the present discovery of the spontaneous breaking of chiral symmetry is expected to stimulate further research and deeper understanding of chiral magnets [4].

Acknowledgements: This work was supported by JSPS KAKENHI Grant number 18K187370A. The synchrotron experiments were performed under the approval of the Photon Factory Program Advisory Committee (No. 2018G039).

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Reference 3 was published on 28 January 2019 of the online version of the *Journal of the Physical Society of Japan* as an Editor's Choice article.

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