表題：$Tb_{2+x}Ti_{2-x}O_{7+y}$ の量子スピン液体状態

Quantum spin liquid state of $Tb_{2+x}Ti_{2-x}O_{7+y}$

H. Kadowaki[A], B. Fåk[B], J. Ollivier[B], T. J. Sato[C]
[A]*Department of Physics, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397*
[B]*Institute Laue Langevin, BP156, F-38042 Grenoble, France*
[C]*IMRAM, Tohoku University, Sendai*

Geometrically frustrated magnets have been actively studied in recent years [1]. These include classical and quantum spin systems on two-dimensional triangle [2] and kagome [3] lattices, and three-dimensional pyrochlore-lattice systems [4]. For classical systems, prototypes of which are the trianglar-lattice antiferromagnet [2] and the spin ice [5], many investigations have been performed for a few decades using a number of theoretical and experimental techniques [1]. Possibilities of quantum spin liquid (QSL) states in frustrated magnets, which date back to the theoretical proposal of the RVB state [6], are recently under hot debate. Highly-entangled many-body wave functions without magnetic long-range order (LRO), anticipated in QSL states, provide theoretically challenging problems [7]. Experimentally, finding out real QSL substances, e.g. [8], and investigating QSL states using available techniques, e.g. [9], have been attracting much interest. However, to date, nobody has found clear evidence of a QSL state, despite many trial experiments performed in the past decade [7,9].

A non-Kramers pyrochlore system $Tb_2Ti_2O_7$ (TTO) has attracted much attention since an interesting report of absence of magnetic LRO down to 0.1 K [10], which could be interpreted that TTO is a QSL candidate or quantum spin ice (QSI) [11]. On the other hand, a phase transition at $T_c \sim 0.5$ K detected by a specific heat peak suggesting a hidden LRO [12], seemed to contradict with the QSL interpretation. We resolved this contradiction by showing that ground states of TTO are highly sensitive to off-stoichiometry, i.e., x (and/or y) of $Tb_{2+x}Ti_{2-x}O_{7+y}$ [13], and that there are two ground states: an electric quadrupole ordered (QO) state ($x > x_c \sim -0.0025$) and the putative QSL state ($x < x_c$) [14] (Fig. 1 inset).

We now think that the QO state proposed in Ref. [14] or its variant likely account for the hidden LRO of TTO. On the other hand, the long-standing question of "what is the putative QSL state of TTO?" or even a simpler question of "is it really a QSL state?" are still difficult problems for present-day experimental techniques, which are not well optimized for studying QSL states. For example, in Fig. 1 we show inelastic neutron scattering (INS) spectra of TTO samples carried out at 0.1 K on IN5 and AMATERAS, which are normally known as good-energy-resolution spectrometers [16]. The energy spectrum of the QSL powder sample with $x = -0.005$ shown in Fig. 1 looks as though there are both elastic and inelastic scattering contributions. However, if this sample is in a QSL state at 0.1 K, this elastic scattering should be (at least partly) inelastic scattering with a very small energy scale [17,18].

Recently we proposed to perform INS experiments on $x = -0.005$ (QSL) and 0.005 (QO) powder samples using the extremely good energy-resolution ($\Delta E \sim 1$ μeV) spectrometers IN16B and HFBS, which have not commonly been used for studies of QSL states. Thereby, we have a good chance to observe that the seemingly elastic scattering shown in Fig. 1 is (partly) inelastic scattering, which consequently proves that the putative QSL TTO sample is definitely in a QSL state at $T = 0$. In addition, by simply comparing quasi-elastic spectra of the QSL and QO samples, distinct difference due to the two ground states can be we observed. We performed an ILL-DDT

使用施設：JRR-3M, 装置：C1-1:HER
分野：Magnetism

experiment (2018 May) of the QSL sample using IN16B, which shows interesting experimental data. However an NCNR-QAP experiment (2018 Aug.) of the QO sample using HFBS was not successful owing to large instrumental background. Now we are waiting for another experiment of the QO sample using DNA (J-PARC) to answer the questions.

[1] C. Lacroix et al. Introduction to Frustrated Magnetism (Springer, Berlin, Heidelberg, 2011).
[2] G. H. Wannier, Phys. Rev. 79, 357 (1950).
[3] I. Syôzi, Prog. Theor. Phys. 6, 306 (1951).
[4] J. S. Gardner et al. Rev. Mod. Phys. 82, 53 (2010).
[5] S. T. Bramwell and M. J. P. Gingras, Science 294, 1495 (2001).
[6] P. W. Anderson, Mater. Res. Bull. 8, 153 (1973).
[7] L. Savary and L. Balents, Rep. Prog. Phys. 80, 016502 (2017).
[8] K. Hirakawa, H. Kadowaki, K. Ubukoshi, J. Phys. Soc. Jpn. 54, 3526 (1985).
[9] B. Fåk et al. Phys. Rev. B 95, 060402 (2017); Y. Shen et al. Nature 540, 559 (2016); Z. Zhu et al. Phys. Rev. Lett. 119, 157201 (2017).
[10] J. S. Gardner et al. Phys. Rev. Lett. 82, 1012 (1999).
[11] H. R. Molavian et al. Phys. Rev. Lett. 98, 157204 (2007).
[12] N. Hamaguchi et al. Phys. Rev. B 69, 132413 (2004).
[13] T. Taniguchi, H. Kadowaki, B. Fåk, J. Ollivier et al. Phys. Rev. B 87, 060408(R) (2013).
[14] H. Takatsu, S. Kittaka, A. Kasahara, Y. Kono, T. Sakakibara, Y. Kato, S. Onoda, B. Fåk, J. Ollivier, J.W. Lynn, T. Taniguchi, M. Wakita, H. Kadowaki, Phys. Rev. Lett. 116, 217201 (2016).
[15] M. Wakita, T. Taniguchi, H. Edamoto, H. Takatsu, H. Kadowaki, J. Phys. CS 683, 012023 (2016).
[16] H. Kadowaki, M. Wakita, B. Fåk, J. Ollivier, S. Ohira-Kawamura, K. Nakajima, H. Takatsu, M. Tamai, J. Phys. Soc. Jpn. 87, 064704 (2018); H. Kadowaki, M. Wakita, B Fåk, J. Ollivier, S. Ohira-Kawamura, K. Nakajima, J. W. Lynn, Phys. Rev. B 99, 014406 (2019).
[17] M. Hermele et al., Phys. Rev. B 69, 064404 (2004).
[18] C-J. Huang et al. Phys. Rev. Lett. 120, 167202 (2018); Y. Kato and S. Onoda, Phys. Rev. Lett. 115, 077202 (2015).

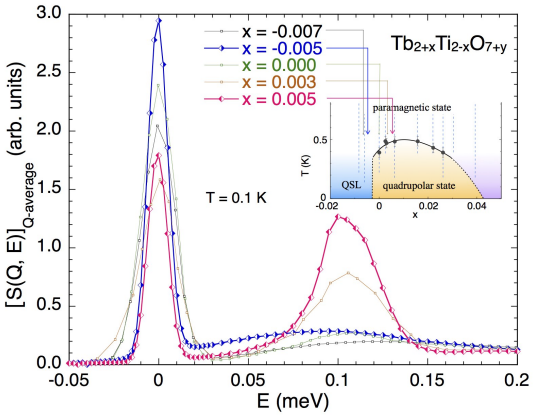

Fig.1 Energy spectra S(Q,E) averaged in a wide Q-range taken using crystal samples (x = -0.007, 0.000, 0.003) and powder samples (x = -0.005, 0.005) [16]. Inset shows the x-T phase diagram of Ref. [15].

Fig. 1.