

MAR14-2013-004226

Abstract for an Invited Paper
for the MAR14 Meeting of
the American Physical Society

Breathing Pyrochlore Lattice Realized in the A-Site Ordered Spinel Oxides LiGaCr₄O₈ and LiInCr₄O₈

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A Cr spinel oxide ACr₂O₄ with a nonmagnetic A²⁺ ion at the tetrahedral site provides an interesting playground for studying magnetic frustration in a pyrochlore lattice made of Cr³⁺ ions with an $S = 3/2$ spin. We found a novel type of frustrated lattice called “breathing” pyrochlore lattice, which is made of Cr³⁺ ions in two A-site ordered spinel oxides, LiGaCr₄O₈ and LiInCr₄O₈ [1]. Because of the large size mismatch between Li⁺ and Ga³⁺/In³⁺ ions, they alternately occupy the tetrahedral sites so as to form a Zinc Blende lattice. This transforms the conventional pyrochlore lattice into an alternating array of small and large tetrahedra, while keeping their shapes regular. LiGaCr₄O₈, with a lesser degree of alternation, shows similar magnetic properties to the conventional Cr spinel oxides such as ZnCr₂O₄. In contrast, LiInCr₄O₈ shows a spin-gap behavior in its magnetic susceptibility caused by a large alternation of magnetic interaction in the more breathing pyrochlore lattice. This suggests that LiInCr₄O₈ exists in a proximity to an exotic singlet ground state based on a tetramer singlet formed in the smaller tetrahedron, although it finally goes to a magnetically ordered state below 13 K, which may be triggered by a structural transition. We will also present NMR and neutron scattering measurements carried out to elucidate the nature of these compounds, and our recent results on solid solutions between the two compounds.

The work has been done in collaboration with T. Nakazono, Z. Hiroi, Y. Tanaka, M. Yoshida, M. Takigawa, T. Masuda (ISSP, Univ. of Tokyo), G. J. Nilsen, H. Mutka, T. Hansen (ILL), and J. P. Attfield (Univ. of Edinburgh).

[1] Y. Okamoto, G. J. Nilsen, J. P. Attfield, and Z. Hiroi, Phys. Rev. Lett. 110, 097203 (2013).