Spin Dynamics of the A-Site Ordered Breathing Pyrochlore Compounds $LiGaCr_4O_8$ and $LiInCr_4O_8$

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Introduction and Experimental

Spinel oxides AB_2O_4 have proven to be a rich reservoir for emergent phenomena such as spin liquids, spin ice, and magnetic monopoles [1]. This is mainly owed to the octahedrally coordinated B atoms, which form the pyrochlore lattice [1]. The particularly interesting example is the A-site ordered spinel compounds, $LiGaCr_4O_8$ and $LiInCr_4O_8$ [2]. The key ingredient of magnetism is a breathing pyrochlore lattice with two different nearest-neighbor exchange coupling constants J and J'. This provides a possibility of tuning magnetic phases by controlling the breathing parameter, $B_f = J'/J$, which measures the degree of frustration. $LiGaCr_4O_8$ and $LiInCr_4O_8$ have the largely different value of $B_f \sim 0.6$ and 0.1, respectively. The former shows the physics of a uniform pyrochlore lattice while the latter is close to an isolated tetrahedron. Their magnetic properties are expected to differ markedly from each other. However, there are no systematic, comparative investigations.

Experimental

High frequency ESR experiments were performed at the EMR Facility at 328.8 GHz in a broadband transmission setup with a sweepable 15/17 T superconducting magnet.

Results and Discussion

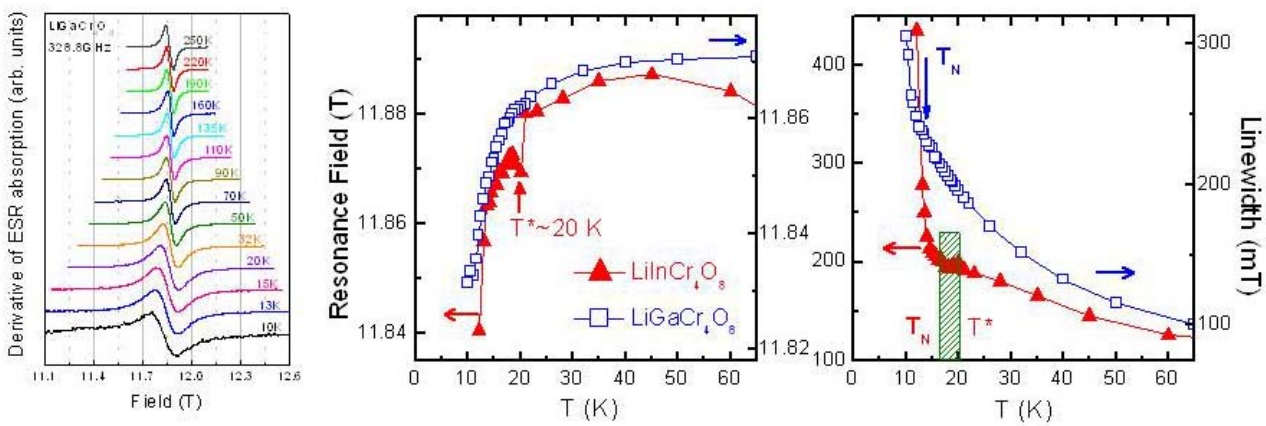

Fig. 1 (Left panel) Temperature dependence of ESR spectra at 328.8 GHz. (Middle and right panels) Temperature dependence of resonance field, H_{res} and peak-to-peak linewidth, ΔH_{pp}.

In Fig. 1 we summarize the temperature dependence of ESR spectra and their parameters. The ESR linewidth, ΔH_{pp}(T) of $LiGaCr_4O_8$ exhibits a critical-like line broadening for $T > T_N = 13.8$ K and then continues to increase upon cooling below T_N and finally wipes out at a few degrees below T_N, which is unexpected for conventional antiferromagets. The resonance field, H_{res}(T) undergoes a shift to lower fields upon cooling. This behavior may be related to persisting spin fluctuations inherent to the frustrated pyrochlore lattice. In contrast, ΔH_{pp}(T) of $LiInCr_4O_8$ shows a weak temperature dependence upon approaching $T_N = 15.9$ K and then exponential-like increase for temperatures below T_N. This indicates the presence of spin singlet correlations at elevated temperatures, which turn to a long-range ordered one due to a structural distortion at T_N. The abrupt drop of H_{res} at $T^* \sim 20$ K gives a hint for a hidden transition, possibly to another (collective) singlet state. Our comparative ESR study reveals the quantum phase transition tuned by changing the breathing parameter.

Acknowledgements

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References

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[2] Okamoto, Y., *et al*., Phys. Rev. Lett., **110**, 097203 (2013).