Probing the phase diagram of the frustrated quantum spin magnet $LiCuSbO_4$ with high-field ESR spectroscopy

The complex oxide $LiCuSbO_4$ has been recently put forward as a possible realization of the Heisenberg quantum spin-1/2 chain magnet which exhibits the so-called spin liquid state. During my research visit at the Institute for Materials Research we have undertaken a detailed study of this compound by high magnetic field ESR spectroscopy. The experimental data reveal interesting signatures of the conjectured magnetic field induced spin phase which possibly arises in this system at low temperatures close to the spin saturation field of 12 T.

Transition metal oxides provide indispensable playground for studies of fundamental models of quantum magnetism. Recently the $LiCuSbO_4$ (LCSO) was suggested as a material where a quantum spin-liquid state is possibly realized [1]. In LCSO the Cu^{2+} spins-1/2 are arranged in chains (Fig. 1) which are frustrated due to competing exchange interactions. Thermodynamics and neutron scattering data reported in Ref. [1] reveal short-range incommensurate spin correlations below $T \sim 8$ K. However, no long-range magnetic order was found down to 100 mK. The analysis of the dispersion of magnetic excitations has revealed the ferromagnetic nearest neighbor magnetic coupling $J_1 = -75$ K and a substantial antiferromagnetic next nearest neighbor coupling $J_2 = 34$ K. The ratio $|J_2/J_1| = 0.45$ exceeds the critical value of 0.25 that separates a collinear spin arrangement in the Heisenberg spin-1/2 chain for $|J_2/J_1| < 0.25$ from an incommensurate spin spiral configuration for $|J_2/J_1| > 0.25$. Interestingly, specific heat measurements suggest the occurrence of a magnetic field induced phase at $T < 2$ K close to the saturation field $B_s = 12$ T. Recent theories predict that close to B_s exotic spin-nematic (multipolar) order can occur in frustrated spin chains (see, e.g., [2]).

Using the excellent pulse magnetic field high-frequency electron spin resonance (ESR) facility at the IMR Tohoku we have studied a polycrystalline sample of LCSO in a broad range of excitation frequencies in the sub-THz domain, in magnetic fields up to 20 T and temperatures down to 500 mK.

At $T > 50$ K the ESR spectrum of LCSO at all studied frequencies exhibits a single line with the g-factor $g = 2.18$ typical for Cu^{2+}. At fields $B < 7$ T the signal remains single line and practically unchanged even at low temperatures (Fig. 2, left). In particular, no signatures of magnetic order could be revealed. However, a peculiar effect has been observed in the high field domain $B \geq 9$ T. There, below $T \sim 20$ K, the ESR spectrum develops a structure (Fig. 2, right). Besides the main peak P2, the low-field (P1) and the high-field (P3) satellites begin to develop. Their distance from the central peak P2 seems to be independent of the field strength but their intensity grows with field and with decreasing the temperature.

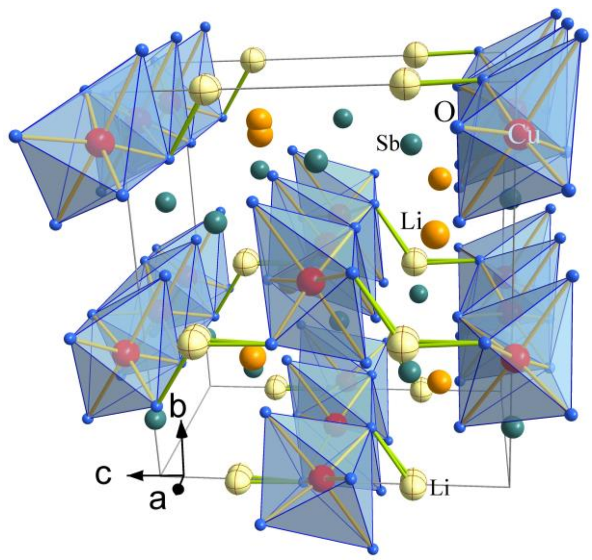

Fig. 1 Crystallographic structure of $LiCuSbO_4$. Cu^{2+} ions (red) bonded to O^{2-} ligands (blue) form edge-sharing CuO_6 chains along the a-axis and provide a realization of the one-dimensional Heisenberg spin-1/2 quantum magnet.

The structure in the ESR spectrum of LCSO is most likely not due to the g-factor anisotropy because it is absent at high temperatures despite the narrowing of the line at high T. It can be tentatively attributed to the growth of the short-range spin correlations in the low temperature regime. It is tempting to conclude that a possible reason for a peculiar shape of the ESR spectrum could be the development of a staggered field in the short-range ordered state. It is remarkable that the satellite peaks are seen only at high fields where the distinct

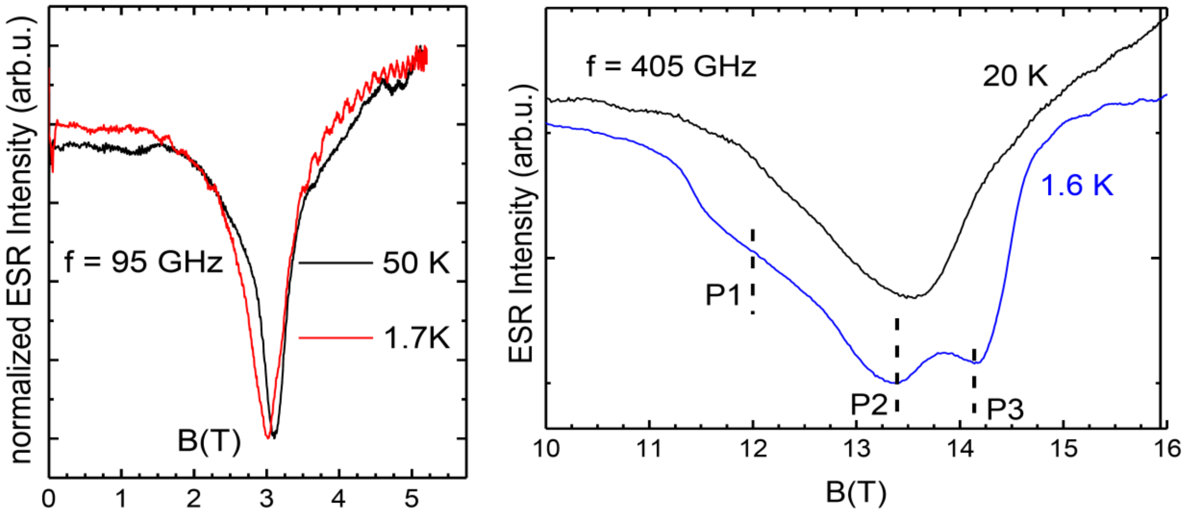

Fig. 2 Representative ESR spectra of $LiCuSbO_4$ at a frequency f = 95 GHz, and a resonant field B_{res} = 3 T (left), and at f = 405 GHz, B_{res} = 13.4 T (right). The spectrum remains single line down to low temperature at fields B_{res} < 7 T, but develops a structure (peaks P1, P2 and P3) at T < 20 K at higher fields

high-field phase has been identified near the saturation field B_s [1]. Motivated by this conjecture we have started a theoretical analysis of the possible spin structure of such high-field phase in LCSO. This work is currently in progress.

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References

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Vladislav Kataev (Leibniz Institute for Solid State and Materials Research IFW Dresden)
E-mail: v.kataev@ifw-dresden.de
http://www.ifw-dresden.de/about-us/people/dr-vladislav-kataev/