Cu-NMR/NQR study of quasi-0D spin S = ½ tetramer $SeCuO_3$: ordered phase and spin excitations

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Low dimensional quantum magnetism is of great interest in theoretical and experimental physics research, owing to strong quantum fluctuations and consequently rich phase diagrams. Coupled spin clusters, called quasi-0D magnets, are of great interest because of their isolated intracluster interactions, as well as competing intercluster interactions that give rise to a 3D ordering.

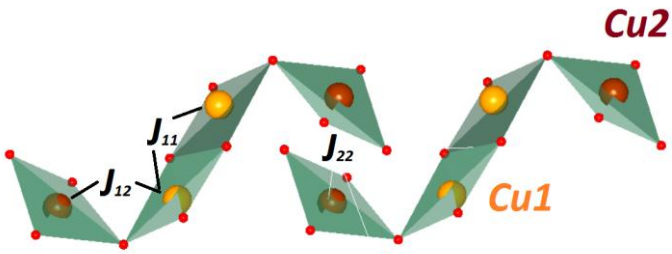

Figure 1 Tetramer structure of $SeCuO_3$ with intratetramer interactions J_{11} and J_{12} and intertetramer interaction J_{22}.

$SeCuO_3$ is monoclinic $P2_1/n$ system. Magnetism in this compound is due to a single spin on Cu $3d_{x^2-y^2}$ orbital. Two inequivalent Cu sites, Cu1 and Cu2 are connected by oxygen superexchange to other Cu sites, forming spin cluster of four spins, i.e. tetramer (Fig.1). The Heisenberg interaction parameters- J_{11} for Cu1 – Cu1, and J_{12} for Cu1 – Cu2 - have values of approx. 200 K [1]. $SeCuO_3$ orders antiferromagnetically at T_N = 8 K, which leaves a wide temperature range (8-200 K) at which low dimensional magnetism exists, making it an ideal compound for study. Multiple experimental techniques were previously used [2, 3] to determine its ordered phase and low-dimensional behaviour, but no clear picture was established. Tetramer interactions are not able to fully explain observables in disordered state, and spin structure in ordered phase is not completely clear.
NQR technique was used as a local probe of spin structure. Cu nucleus was chosen for its direct coupling to magnetic moment. By doing rotation measurements on single crystals, we show that only Cu1 site is observable by NMR technique, implying fast fluctuations of Cu2 site magnetic moments. Zero field NMR in the ordered phase, together with neutron measurements [4], provided insight into spin structure and onset of antiferromagnetic order.
Spin-lattice relaxation measurements were conducted to study spin dynamics in 3D ordered and disordered low-dimensional phase. It provided clear and direct evidence of spin ordering into gapless antiferromagnetic phase. Evidence for 1D long range correlations are present in temperature range from 8 up to 90 K. At higher temperatures activation behavior is visible, comparable to J_{11} coupling energy.
Our work showed that to completely explain the magnetism in disordered phase of $SeCuO_3$, a model beyond tetramer is needed, probably including J_{22} chain-like interaction (in accordance with our spin – lattice relaxation), and maybe existence of Dzyaloshinskii-Moriya interaction between Cu1 and Cu2 sites. Magnetic structure in ordered state was determined for Cu1 spins, studied with NMR of the corresponding nuclear spin.

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