Sequential magnetic ordering of kagome-plane and interlayer Cu moments in clinoatacamite revealed by site-selective NMR

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Frustrated quantum magnets provide a fertile ground for unconventional magnetic ground states arising from competing exchange interactions and reduced dimensionality. The naturally occurring mineral clinoatacamite, a polymorph of the parent compound of the kagome spin-liquid material herbertsmithite, is an important model system in this context. Its magnetic lattice consists of Cu^{2+} (S=1/2) ions coupled through oxygen-mediated superexchange interactions, giving rise to strong magnetic frustration. Previous μSR and neutron diffraction studies reported two magnetic transitions near 18 K and 6.5 K, but their microscopic origin has remained unresolved [1]. Here, we investigate clinoatacamite by site-selective ^{1}H and ^{35}Cl nuclear magnetic resonance. The NMR spectra reveal magnetic transitions at both temperatures, while the site-dependent spin-lattice relaxation rates provide direct evidence that the Cu2/Cu3 kagome planes develop long-range magnetic order below 18 K, whereas the interlayer Cu1 moments remain dynamically fluctuating until they order below 6 K. These results establish a microscopic picture of sequential magnetic ordering in clinoatacamite and reconcile the previously reported experimental observations.

References:
[1] L. Stödter, C. Kastner, H. O. Jeschke, et al., arXiv:2601.21024