Scientific report for ESF HFM Grant:

NMR/NQR Study of the Kagomé Compounds $ZnCu_3(OH)_6Cl_2$ and $Pr_3Ga_5SiO_{14}$

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Purpose of the visit

Two novel families with kagomé geometry, which show no magnetic instability down to at least 50 mK, have been highlighted recently. The atacamite family $Zn_xCu_{4-x}(OH)_6Cl_2$, with herberthsmitithe (x = 1) as the first structurally perfect realization of the spin-1/2 kagomé lattice [1], and the family of rear-earth based langasites $RE_3Ga_5SiO_{14}$. For the latter, the Nd case has been extensively studied since Nd-langasite represents a realization of an anisotropic large-spin kagomé lattice [2]. It has been proposed that the second member of the langasite family, Pr-langasite, similarly exhibits a spin-liquid ground state in zero magnetic field, while the magnetic field itself induces short-range ordering of Pr^{3+} magnetic moments [3,4]. The zero-field results, primarily based on neutron diffraction and heat capacity measurements, appeared to be in a contradiction with μSR results that we have obtained for the same compound. Namely, our preliminary studies pointed at a glassy-like state with weak static internal magnetic fields emerging already at elevated temperatures. The main purpose of my visit was therefore to perform an in-depth analysis of our μSR data of the Pr-langasite and to do additional NQR measurement, as another zero-field local-probe technique to complement the μSR results. Second, ^{35}Cl NMR investigation of partially oriented herbertsmithite powders was planned, in order to further inspect the role of magnetic anisotropy [5] and impurities [6] in this material.

Description of the work carried out during the visit and of the main results obtained

The majority of my time while visiting the host institution was devoted to the Pr-langasite. Based on our μSR data we determined the nature of the ground state of this material, which enabled us to accurately simulate μSR relaxation curves. Our comprehensive μSR analysis gave information about static and dynamic properties of this state. Our results gave first clear indications of weak internal static magnetic fields setting in below approximately 40 K. Their size at muon (oxygen) sites is limited to only few ten Gauss, making this an original state. The magnetic order however does not appear as a classical long-ranged Néel order, but rather as glassy-like short-ranged order. It is important to note that this spin-glass behavior applies to bulk of the sample. We were also able to follow the temperature evolution of a dynamical mechanism of muon relaxation. This relaxation is intimately related to the dynamics of the internal magnetic field at muon sites originating from electronic spins. Surprisingly, the peak

in the muon relaxation was observed at significantly lower temperatures than the temperature of the onset of the static internal fields.

In order to complement the above results we performed another zero-field local-probe investigation on the same material, namely a nuclear quadrupolar resonance study. Static NQR spectra were detected above and below the transition temperature. Similarly as in the case of the Nd-langasite these revealed large distributions of local environments. The large width of the spectra effectively masked their shape transformation below the spin-glass-like transition temperature. However, the corresponding changes in spin dynamics were accurately followed through the temperature dependence of NQR spin-lattice relaxation time. Our NQR spin-lattice relaxation confirmed the shifted peak of the spin-lattice relaxation rate with respect to the onset temperature of static internal magnetic fields.

The initially planned thorough analysis of the ^{35}Cl NMR data of partially ordered powder of herbertsmithite and further measurements were left to be performed later.

Future collaboration with host institution and projected publications/articles resulting or to result from the grant

We plan to pursue our ongoing collaboration in future on few selected examples of highly frustrated magnets. First we plan to complete our study of another member of the langasite family, Sm-langasite, and continue with a couple of new copper-based compounds with frustrated geometry. For the latter, we will perform μSR at ISIS and PSI, NMR will be done in France and ESR in Slovenia. The work performed during my visit will results in a publication in near future.

References:

[1] M. P. Shores *et al.*, J. Am. Chem. Soc. **127**, 13462 (2005).
[2] A. Zorko *et al.*, Phys. Rev. Lett. **100**, 147201 (2008).
[3] H. D. Zhou *et al.*, arXiv: cond-mat 0808.2819.
[4] L. L. Lumata *et al.*, arXiv: cond-mat 0811.3367.
[5] A. Zorko *et al.*, Phys. Rev. Lett. **101**, 026405 (2008).
[6] A. Olariu *et al.*, Phys. Rev. Lett. **100**, 087202 (2008); M. A. de Vries *et al.*, Phys. Rev. Lett. **100**, 157205 (2008).

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