Short visit 3639 – «HFM» ESF Activity - Final report

I was awarded a short visit grant (Nb 3639) to perform muSR experiment at the ISIS large scale facility in UK. The purpose of this beam time was to explore with muSR the ground state of new Cu2+ S=1/2 kagome compounds, isomorphic to the now-celebrated Herbertsmithite, namely CdCu3(OH)6Cl2 and MgCu3(OH)6Cl2. Due to the different ionic radii as compared to Zn, we expected (1) that changes of the unit cell parameters would modify significantly the antiferromagnetic coupling and the Dzyaloshinski-Moriya interaction which are likely to drive the nature of the ground state, a question that is still deeply debated (2) to reduce the tendency to intersite mixing. Further, we proposed to draw the phase diagram of the related families of compounds AxCu4-x(OH)6Cl2, A=Cd or Mg. The comparison of the Zn, Cd and Mg based atacamite should help to characterize the weak 3D couplings at play and which stabilize 3D order when x is reduced below c.a. 0.5.

Experiments were performed at the MuSR spectrometer. A. D. Hillier was our local contact. We focused our beam time to the study of the Mg compounds which turned out to show a rather slow muon relaxation rate at base temperature which is uniquely suited to the ISIS pulsed muon source. The Cd counterparts are best suited to continuous muon sources available for instance at the PSI facility (CH). Our chemist co-workers managed to synthesized good quality powder materials only in a limited range of composition around x=1, so that we postponed the study of the whole magnetic phase diagram to next beam times. The x=1 composition corresponds ideally to the case where the kagome planes are fully magnetically decoupled from each others, i.e. the best case where to expect a spin liquid behavior. Most experiments were performed in the sub-kelvin temperature range (50mK – 4K) in a dilution refrigerator. We could study in details four different samples spanning the Mg concentration rate range [0.85-1] and obtained by two different synthesis routes - hydrothermal and Redox synthesis – which likely impact the local structure of the material.

Our main finding is that none of the materials we studied showed a magnetic transition down to the base temperature (50mK) of the experiment. muSR being highly sensitive to minute internal magnetic fields, this is a robust statement. The Mg analogue of Herbertsmithite is thus the second material to qualify for a spin liquid behavior after Herbertsmithite. Despite the absence of a transition to a magnetically frozen state, we could detect a slight increase of the relaxation rate below 1K quite similar to the one found in Herbertsmithite and that was tentatively attributed to magnetic defects. Interestingly this low T relaxation varies significantly in between the four studied compounds. Further analysis in relation with accurate refinements of the structure of these samples will help understanding in depth their properties.