Short term visit to the Paul Scherrer Institute: Scientific report

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The aim of my 6 day-long visit (12-18 December 2010) to the Paul Scherrer Institute where I was hosted by A. Amato, was the investigation by muon spin rotation and relaxation (μ SR) measurements of highly frustrated magnets with a two dimensional structure.

The first studied system was the triangular antiferromagnet $FeGa_2S_4$ which consists of layers of Fe-S which are well decoupled from each other by layers of Ga-S. This system is isostructural to $NiGa_2S_4$ which was discovered as an excellent realisation of a magnet on an undistorted triangular lattice. Neutron scattering measurements evidenced the presence of short range incommensurate magnetic correlations appearing below $\simeq 18$ K, a temperature value notably higher than the position of the susceptibility peak observed around 8.5 K. μ SR measurements performed earlier on this system showed a spontaneous muon spin precession consistent with an incommensurate order. The spontaneous field at the muon was found to vanish at about 9 K. In addition, a muon spin lattice relaxation was evidenced down to the lowest investigated temperature (20 mK) with basically no thermal dependence at very low temperature. This result was in sharp contrast to Ga nuclear quadrupole resonance measurements which showed a power law dependence in the same temperature range. Concerning the high-temperature behaviour, the μ SR spectra have an exponential shape down to $\simeq 12.6$ K, with an relaxation rate monotonously increasing as the temperature is decreased. This is the classical signature of the building-up of magnetic correlations. Between 12.6 and 9 K the spectral shape is square root exponential. This response is reminiscent of the canonical spin glasses behaviour above the freezing temperature.

On its side, FeGa₂S₄ has been the subject of much fewer investigations than NiGa₂S₄. The experiments performed at PSI in December 2010 yielded the main following results. A spontaneous precession of the muon spin is observed at low temperatures up to 0.9 K. The amplitude of the signal is however quite weak and the damping of the oscillations is large. Figure 1 displays the spectrum recorded at 0.4 K as an example. The spin-lattice relaxation has a square root exponential lineshape with a finite relaxation rate which is roughly temperature independent in the range up to 0.9 K. At 2 K and above the oscillations are no longer resolved but still a relatively important fraction of the μ SR amplitude is lost. A consistent fit of the spectra has not been reached so far, but it is qualitatively clear that a magnetic transition occurs around 30 K. Above this temperature the spectra have a square root exponential relaxation lineshape at least up to 50 K with a relaxation rate which decreases as the temperature increases. A truly exponential relaxation is observed at 130 K and above. Altogether, these results are reminiscent of the ones obtained for NiGa₂S₄ with some differences: especially there is no direct evidence for correlations of incommensurate nature at low temperature in FeGa₂S₄ and the temperature region where the square root exponential lineshape is observed is much broader in FeGa₂S₄ than in NiGa₂S₄

The second part of the program dealt with another undistorted triangular antiferromagnet, namely La₂Ca₂MnO₇. This system is known to exhibit a transition to a genuinely two-dimensional antiferromagnetic order below 2.8 K. The order is of the $\sqrt{3} \times \sqrt{3}$ type. Due to the limited avalable beamtime, μ SR experiments could be performed only down to 1.8 K. While above 2.8 K, the zero-field spectra can



Figure 1: μ SR spectrum recorded in zero external field in a powder of FeGa₂S₄ at 0.4 K. The lefthand-side plot shows the short time details, while the right-hand-side one displays an extended time range. The spontaneous field at the muon site is close to 0.8 T.



Figure 2: Thermal dependence of the muon spin-lattice relaxation rate measured in an external field of 10 mT with a powder of $La_2Ca_2MnO_7$.

be satifactorily fitted to a phenomenological stretched exponential function, a sum of two components is needed to account for the sprectral shape below this temperature: the sum consists in a Gaussian and an exponential function in the relative weight ratio 2:1. The sample under investigation being a powder, one can understand this weight ratio interpreting these components in terms of the spin-spin and spin-lattice relaxation channels respectively. This results means that the spontaneous field at the muon site is zero or close to zero in the magnetic phase of La₂Ca₂MnO₇. A complete temperature scan was subsequently performed in a weak longitudinal field of 10 mT which was applied in order to decouple the muon spin from the nuclear spins of most specifically ¹³⁹La and ⁵⁵Mn. The spectra could again be fitted with the same model as in zero field. The stretched exponent clearly departed from 1 only below $\simeq 5$ K to reach a value equal to $\simeq 0.7$ near the magnetic transition. Figure 2 shows the temperature dependence of the spin-lattice relaxation rate. This rate displays a strong temperature dependence in the ordered phase, but any definitive conclusion can only be drawn when data at temperatures lower than 1.8 K are available.

As far as $FeGa_2S_4$ is concerned, the results obtained during this visit at the Paul Scherrer Institute will be published in a report about $NiGa_2S_4$ and $FeGa_2S_4$ which is in preparation. The results about $La_2Ca_2MnO_7$ require a complementary investigation below 1.8 K before their publication can be considered.

This work is performed in collaboration with A. Yaouanc and C. Marin (CEA/Grenoble), A. Amato and C. Baines (PSI), and for the part devoted to $FeGa_2S_4$ with D.E. MacLaughlin (Riverside) and S. Nakatsuji (ISSP Tokyo).