Effect of silicon doping on low temperature magnetic interactions in epitaxial films of $Mn_5(Ge_{1-x}Si_x)_3$: zero-field ^{55}Mn NMR study

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Mn_5Ge_3 and Mn_5Si_3 are two isostructural compounds crystallizing in the hexagonal $D8_8$ structure (space-group $P6_3/mcm$) that can be mutually alloyed over the entire concentration range. The two end compositions of the $Mn_5(Ge_{1-x}Si_x)_3$ series exhibit very different magnetic behavior: Mn_5Ge_3 is a metallic ferromagnet with a Curie temperature of 296 K whereas Mn_5Si_3 reveals a complex antiferromagnetic order at low temperatures: a chiral spin structure below 65 K and a collinear spin arrangement between 65 K and 100 K. Their unit cell contains two formula units, with the manganese atoms in two Wyckoff crystallographic positions: 4*(d)* and 6*(g)* (here denoted as Mn_I and Mn_{II} sites, respectively). It was shown that the $Mn_5(Ge_{1-x}Si_x)_3$ films can be grown epitaxially on Ge(111) substrates. With the aim to understand local magnetic properties in each of the two manganese sites as a function of Si content we have undertaken a zero-field ^{55}Mn NMR study in a series of the $Mn_5(Ge_{1-x}Si_x)_3$ epitaxial films with silicon concentration ($0 \le x \le 0.5$).

^{55}Mn NMR experiment probes the local hyperfine field: in the pristine Mn_5Ge_3 compound at 4.2 K the manganese atoms located at the 4*(d)* positions give the resonance signal at 207 MHz (central frequency of the quadrupolar structure) corresponding to the local hyperfine field of 19,6 T, whereas those in the 6*(g)* sites experience hyperfine field of 40,8 T (resonance frequency 430 MHz).

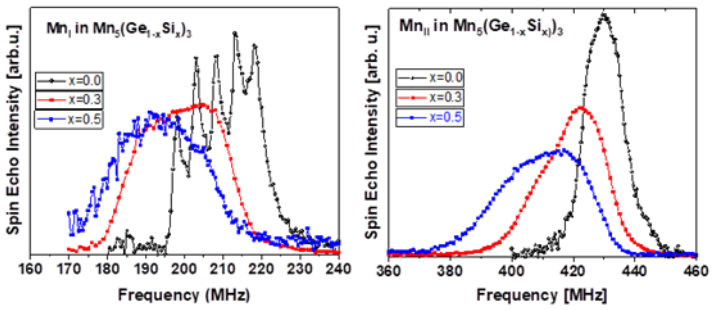

Fig.1 ^{55}Mn NMR spectra recorded at 4.2 K from the $Mn_5(Ge_{1-x}Si_x)_3$ epitaxial films (60 nm thick) as a function of silicon content a) Mn_I environments b) Mn_{II} environments.

Upon silicon substitution two effects are readily visible (Fig 1): i) the NMR spectra shift towards lower frequencies evidencing the drop of the average hyperfine field on both manganese sites and ii) the NMR spectrum from the Mn_{II} sites develops a broad low frequency profile revealing the presence of a Mn_{II} population with the reduced magnetic moment. The thorough analysis of NMR data shows that substitution of Ge atoms by Si significantly modifies the exchange interactions involving the 6*(g)* manganese sites, leading to a drop of the magnetic moment of those Mn_{II} atoms that have Si neighbour instead of Ge. The effect on the Mn_I sites is secondary and consists in a smaller contribution of the transferred hyperfine field due to the reduced magnetic moment of the Mn_{II} neighbors and a loss of the quadrupolar structure due to the inhomogeneity brought by Ge/Si replacement.