Long-range ordered superlattice of carbon within $Mn_5Ge_3C_{0.5}$ epitaxial films

R. Kalvig,[1] E. Jędryka,[1] M. Wójcik,[1] M.Petit,[2] and L. Michez[2]

[1] *Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Warszawa, Poland*

[2] *Aix Marseille Univ, CNRS, CINAM, Marseille, France*

Mn_5Ge_3 is a metallic ferromagnet with the Curie temperature of 296 K, that can be increased up to 450 K by the addition of carbon [1]. It crystallizes in the hexagonal D_{88} structure with Mn atoms occupying two magnetically and structurally inequivalent positions: 4d (Mn_I) and 6g (Mn_{II}). Recent ^{55}Mn NMR studies performed on a pristine Mn_5Ge_3 evidenced the unquenched orbital moment of Mn in both sites, giving rise to a high out-of-plane anisotropy [2]. The anisotropy of the orbital moment was found to be strongly reduced by the presence of carbon, which enters interstitially occupying the 2(b) octahedral voids and strongly reduces the magnetic moment of the Mn_{II} atoms located in the corners of a host octahedron [3]. We now present the ^{55}Mn NMR study performed on a series of epitaxial 30 nm thick $Mn_5Ge_3C_x$ films in the entire concentration range (nominal carbon content varying between $0 < x < 0.85$). Up to x=0.5 the Mn_{II} atoms are found in two distinctly different magnetic states, labelled as Mn_{II} (no carbon nearest neighbor) and Mn_{II_C} (one carbon nn). The latter site gives rise to a satellite NMR line shifted down by 82 MHz - with increasing carbon concentration the signal intensity is systematically transferred from Mn_{II} to Mn_{II_C}. The NMR spectra recorded from films with a nominal carbon content above x=0.5 are identical: the original Mn_{II} NMR signal disappears, but no second satellite line is observed, even though there are two available 2(b) positions around the Mn_{II} atoms. These observations imply that the spatial distribution of carbon is not statistical, tending to a selective occupancy of every second available 2(b) site in the Mn_5Ge_3 lattice along the c-direction and setting the limit for the uptake of carbon in the Mn_5Ge_3 lattice to x=0.5. Considering that the 2(b) voids are located in the same atomic plane as the Mn_I atoms, in case of their random occupancy one would expect a distribution of different Mn_I environments. However, the observed evolution of the Mn_I NMR line as a function of carbon concentration reveals that the carbon-filled voids are ordered also in-plane, and form a highly ordered superlattice within the hexagonal D_{88} structure. This postulate is confirmed by the reported stabilization of the Mn_5Ge_3 crystal lattice in presence of carbon. A self-organizing tendency of carbon is well documented in different carbon- and graphen-containing nanostructures, but to our knowledge this is the first evidence of carbon long range order within an alien crystal lattice. This observation may encourage efforts to grow other highly ordered carbon-doped systems in view of engineering their magnetic properties.

References:

[1] M. Gajdzik, C. Sürgers, M. T. Kelemen. H. Löhneysen, J. Magn. Magn. Mat. **221**, 248 (2000)

[2] R. Kalvig, E. Jedryka, M. Wojcik, G. Allodi, R. De Renzi, M. Petit, L. Michez, Phys. Rev. B **97**, 174428 (2018)

[3] R. Kalvig, E.Jedryka, M. Wojcik, M. Petit, L. Michez, Phys. Rev. B **101**, 094401 (2020)