Epitaxial $Mn_5Ge_3C_x$ films - effect of carbon doping on magnetic anisotropy studied by means of ^{55}Mn NMR

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Objectives

Mn_5Ge_3(001)/Ge(111) epitaxial films: a potential source of polarized carriers directly into Ge.
Interesting features: high spin polarization (42%), easy fabrication, high Curie temperature (T_C = 296 K), and full compatibility with Si and Ge based technology

Doping with carbon :

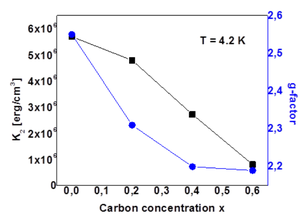
- **Increased Curie temperature from 296 K up to 430 K**
- **Decreased uniaxial magnetocrystalline anisotropy, evidenced by FMR**

Kittel's equations:

$$\left(\frac{\omega}{\gamma}\right)^2 = H_{\parallel}\left(H_{\parallel} + 4\pi M - \frac{2K_u}{M}\right)$$

$$\frac{\omega}{\gamma} = H_{\perp} - 4\pi M + \frac{2K_u}{M}$$

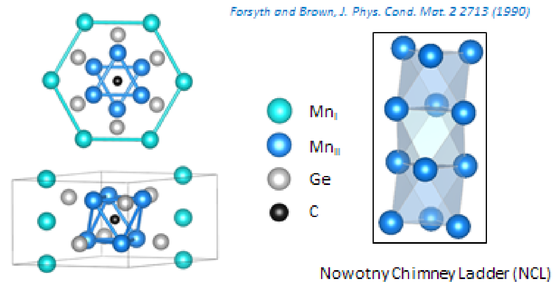
$$\gamma = \frac{g\mu_B}{\hbar}$$



Mn_5Ge_3 – crystal structure

- Nowotny phase, hexagonal structure – space group $P6_3/mcm$
- Mn positions: Mn_I (4d) : μ_I = 1.94 μ_B and Mn_{II} (6g) : μ_{II} = 3.34 μ_B

Forsyth and Brown, J. Phys. Cond. Mat. 2 2713 (1990)



Nowotny Chimney Ladder (NCL)

$Mn_5Ge_3C_x$: Carbon enters into 2(b) voids in the center of Mn_{II} octahedron in a highly ordered manner, maximum uptake: x=0.5 ➡ every second 2(b) void occupied

R. Kalvig et al., Phys. Rev. B 105 094405 (2022)

Aim of this study: to understand the origin of magnetocrystalline anisotropy in the entire concentration range

Methods

Experimental tool: ^{55}Mn NMR

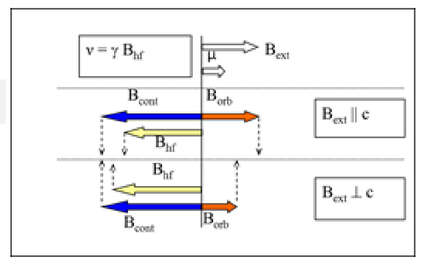
$$\nu_{NMR} = \gamma B_{eff} \approx B_{hf} \approx B_{hf}^{cF} + B_{hf}^{orb} = -A_{cF}\mu^{S}_{loc} + A_{orb}\mu^{L}_{loc}$$

isotropic anisotropic

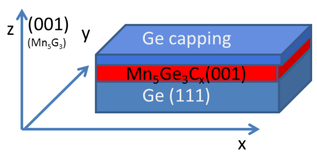
From calculations of hyperfine fields in Mn_5Ge_3 ➡ A_{cF}= 12 T/μ_B A_{orb}= 27 T/μ_B

S. Picozzi, A. Continenza, and A. J. Freeman, Phys. Rev. B 70, 235205 (2004)

^{55}Mn: $\gamma = 10{,}553\frac{MHz}{T}$



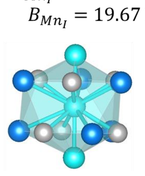
Results: ^{55}Mn NMR in the undoped Mn_5Ge_3 film (300 nm)

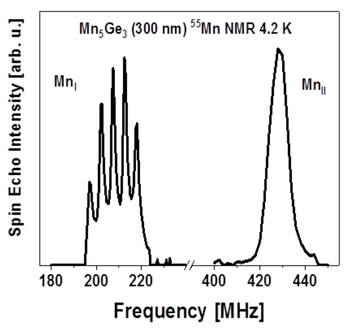


Zero-Field ^{55}Mn NMR

Mn_I in (4d):

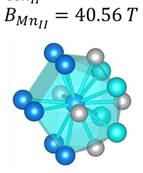
$\mu_{Mn_I} = 1.94\mu_B$
$\nu_{Mn_I} = 207.5\ MHz$
$B_{Mn_I} = 19.67\ T$





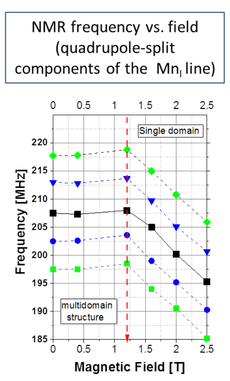
Mn_{II} in (6g):

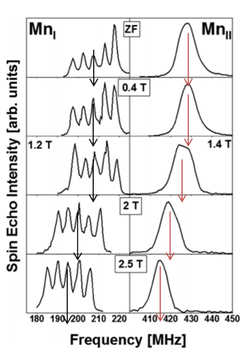
$\mu_{Mn_{II}} = 3.34\mu_B$
$\nu_{Mn_{II}} = 428\ MHz$
$B_{Mn_{II}} = 40.56\ T$



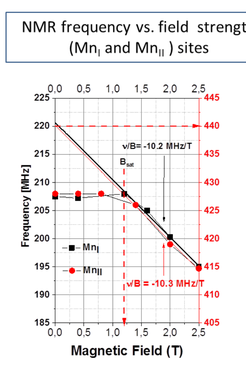
External magnetic field perpendicular to the film

NMR frequency vs. field (quadrupole-split components of the Mn_I line)





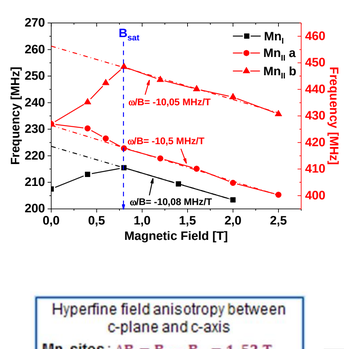
NMR frequency vs. field strength (Mn_I and Mn_{II}) sites

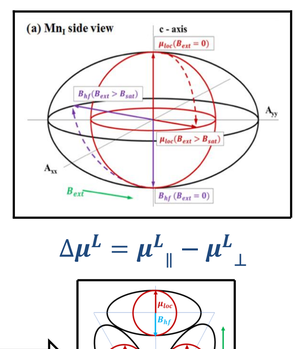


$$\Delta\nu \sim (3cos^2\theta - 1)$$
$$\theta = const.$$

In demagnetized film NMR signal originates from magnetization oriented along the easy axis (c-direction)

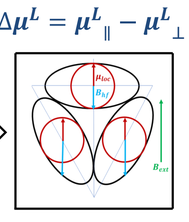
External magnetic field in-plane of the film



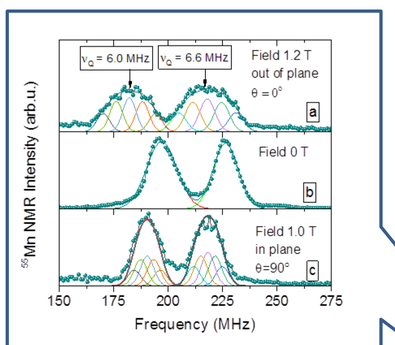


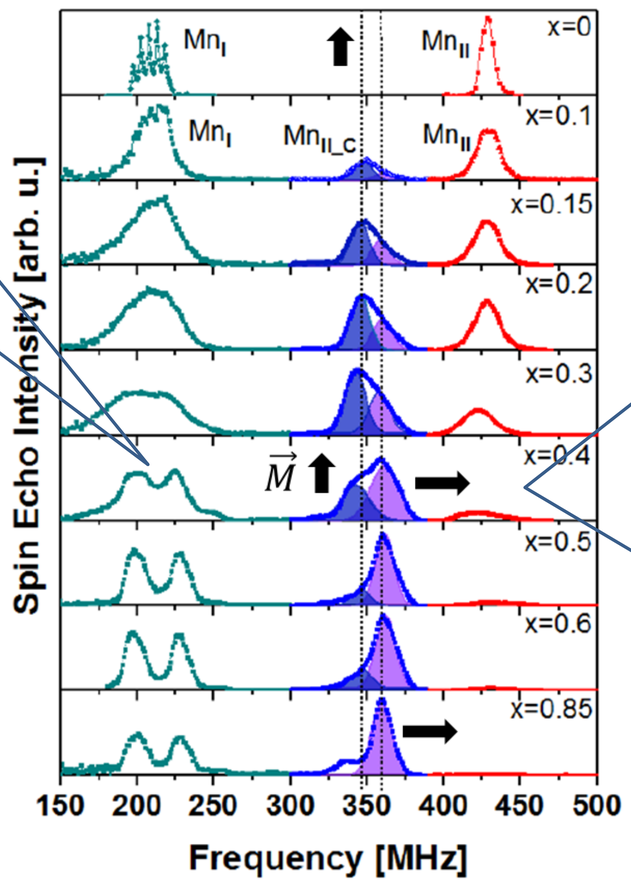
$$\Delta\mu^L = \mu^L_{\parallel} - \mu^L_{\perp}$$

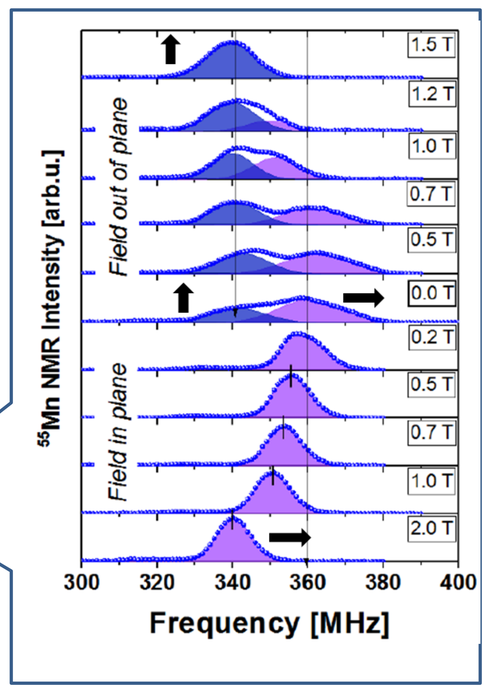
Hyperfine field anisotropy between c-plane and c-axis
Mn_I sites : $\Delta B = B_I - B_{\perp} = 1.52$ T
Mn_{II} sites : ΔB = up to 4.08 T
modulated in-plane

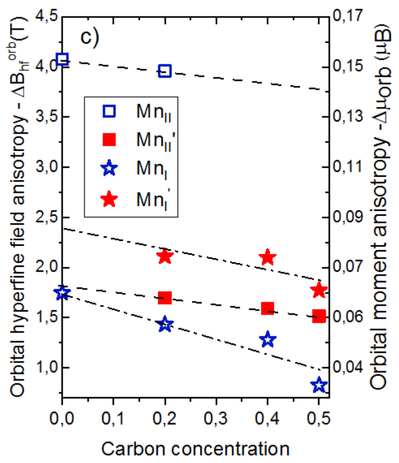


Results: ^{55}Mn NMR in $Mn_5Ge_3C_x$ films ($t = 30$ nm; $0 \leq x \leq 0.85$)









Mn_{II} atoms with a carbon neighbour
new spectrum component at 344 MHz due to co-existing signals from in-plane and out-of-plane orientation

$x \geq 0.5$: **No changes in the NMR spectrum**

Conclusions

- Strong anisotropy of ^{55}Mn hyperfine fields between the hexagonal c-axis and the c-plane in Mn_I and Mn_{II} lattice sites within pristine Mn_5Ge_3. Mn_{II} site: an additional modulation of hyperfine field in the c-plane.
- Isotropic Fermi contact term constitutes the main component of hyperfine fields, but a significant anisotropic contribution due to the anisotropy of orbital moment μ_L is evidenced by the NMR data.
- Carbon dopant occupies selectively interstitial 2*b* voids, locally changing the anisotropy of orbital contribution to the hyperfine field on those Mn_{II} atoms that form a host octahedron. The modulation of anisotropy within the c-plane is lifted. Mn_I sites, more distant from the dopant location are less affected.
- A linear drop of hyperfine field anisotropy upon carbon doping corresponds to the change of bulk uniaxial magnetocrystalline anisotropy evidenced by FMR, and reflects the increasing number of Mn_{II} atoms affected by their carbon neighbors.
- The above observations confirm that the magnetocrystalline anisotropy observed in this system has a single ion origin and can be linked to the observed anisotropic orbital moment of manganese.