# Probing Local Features in Dilute Magnetic Semiconducting $ZnGeP_2$:Mn via $\mu^+SR$


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## Project Focus

•DMS systems gaining importance as prospects in spin-based electronics

•Mechanism responsible for connecting local magnetic features to bulk magnetic properties – not yet understood in DMS systems

## MuSR & $\mu^+$ as Local Probe [1]

•Muon Spin Relaxation utilizes unique sensitivity of 100% spin polarized and positively charged muons to probe local magnetic and electronic environment

•Local B-field environment for $\mu$:

$$\mathbf{B}_{loc} = \langle \mathbf{B}_{loc} \rangle + \delta \mathbf{B}_{loc} = \mathbf{B}_{ext} + \mathbf{B}_{dip} + \mathbf{B}_{hyp} + \mathbf{B}_{fermi} + \delta \mathbf{B}_{loc}$$

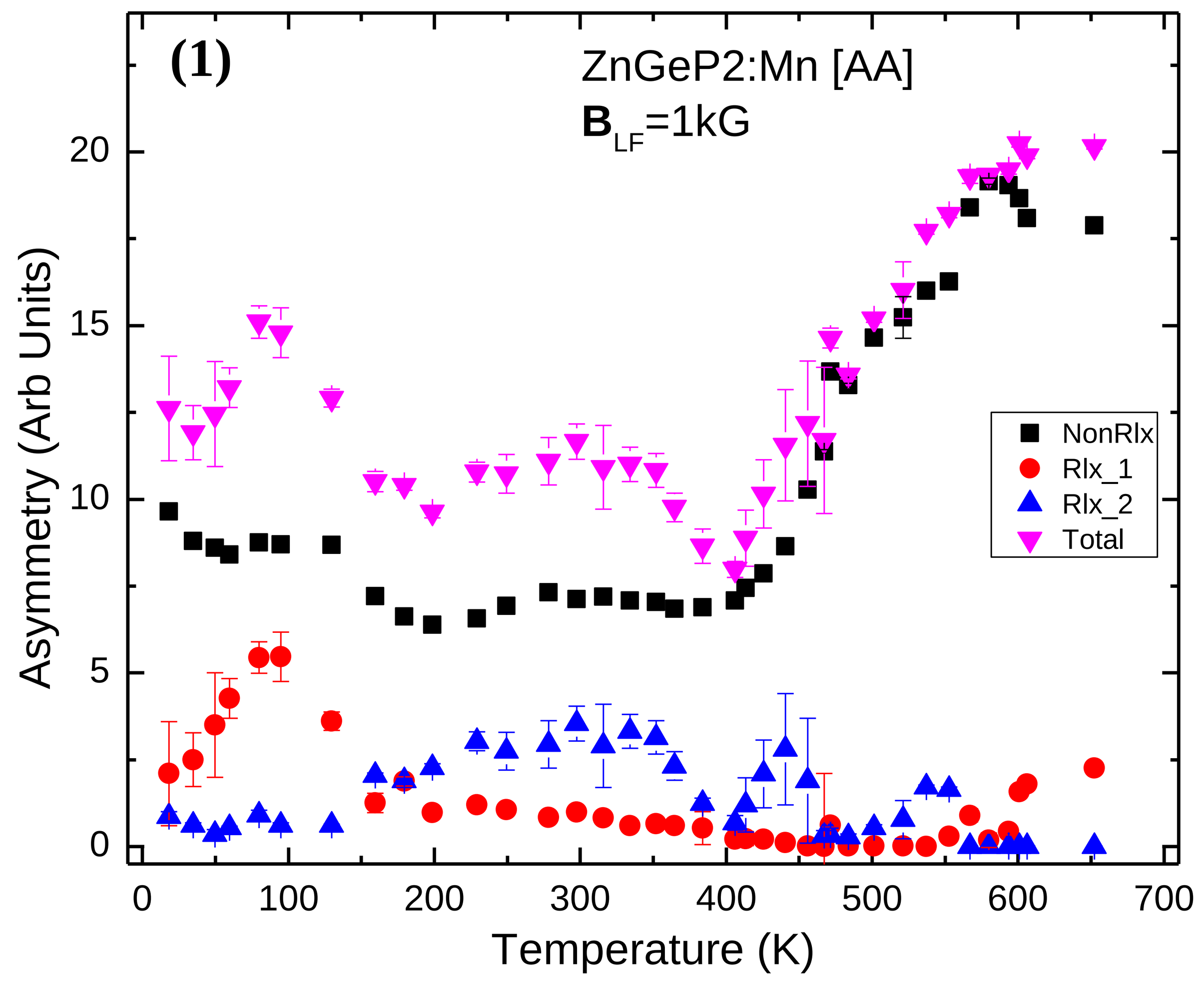
$B_{ext}$ =Applied external field

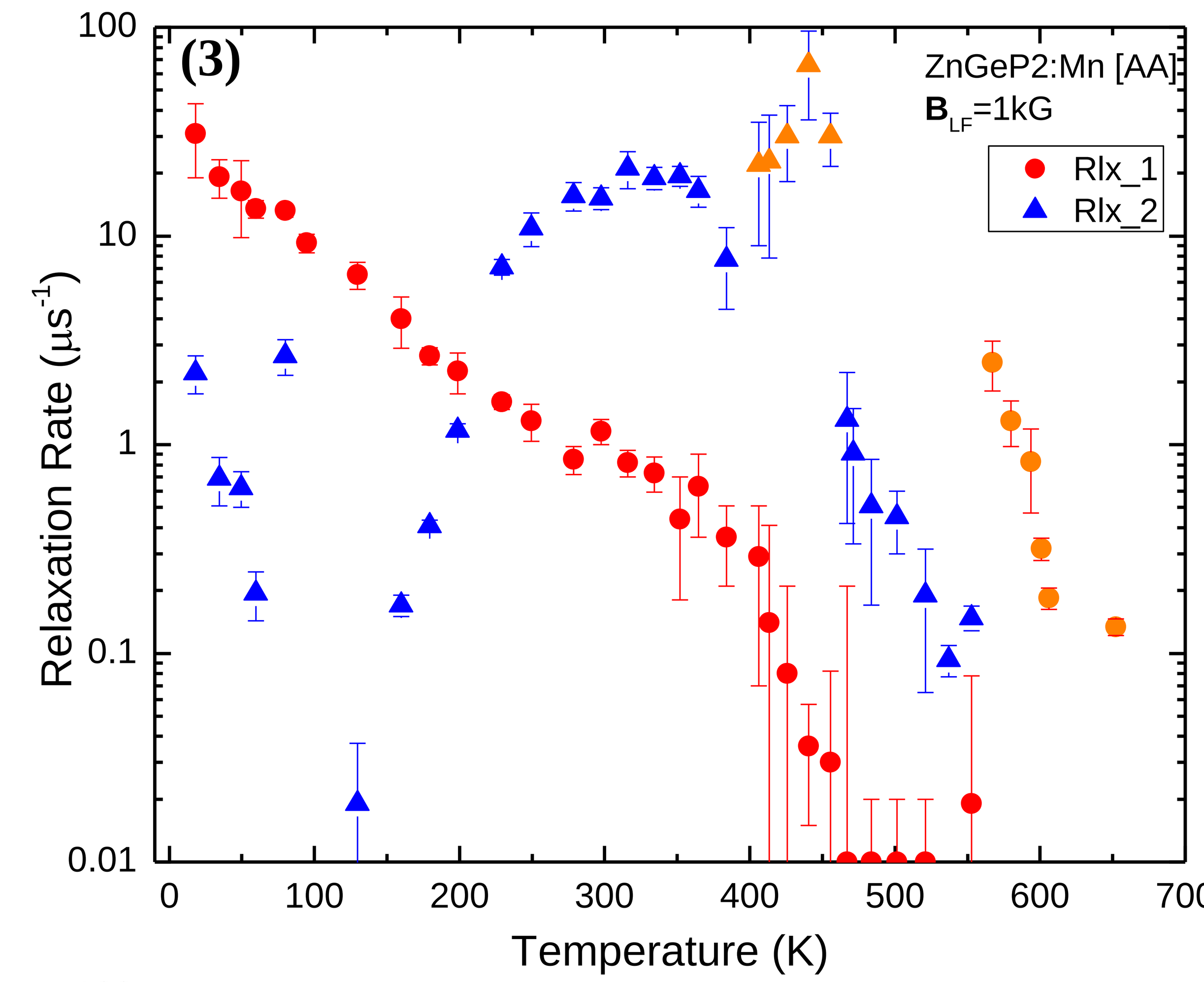
$B_{dip}$ = dipolar field
→ Sum of localized moments over entire crystal
→ Including site to site differences

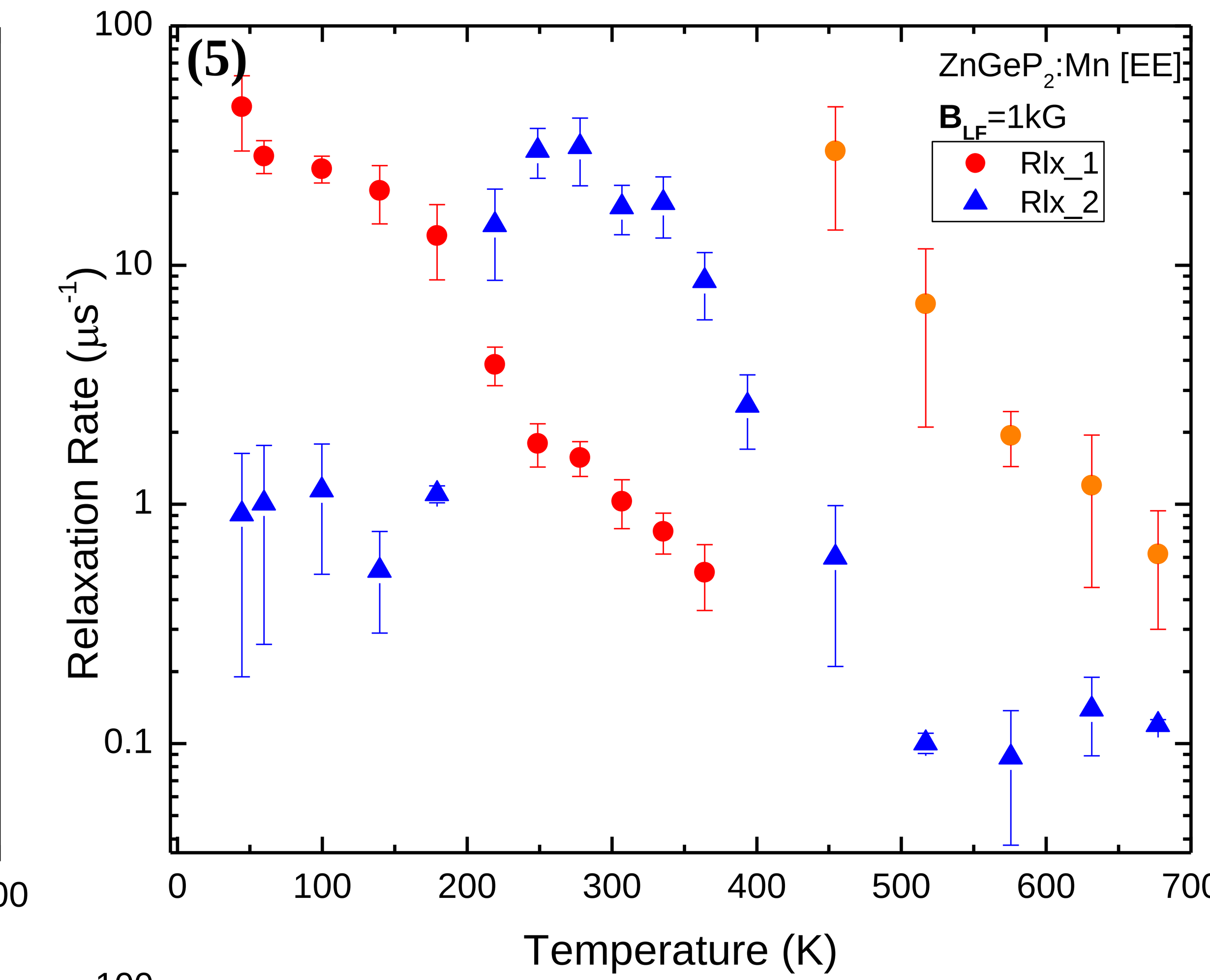
$B_{hyp}$= Field from HF interaction
→ Short range magnetic interaction between $\mu^+$ and local electronic moments (cf: wavefunction overlap)

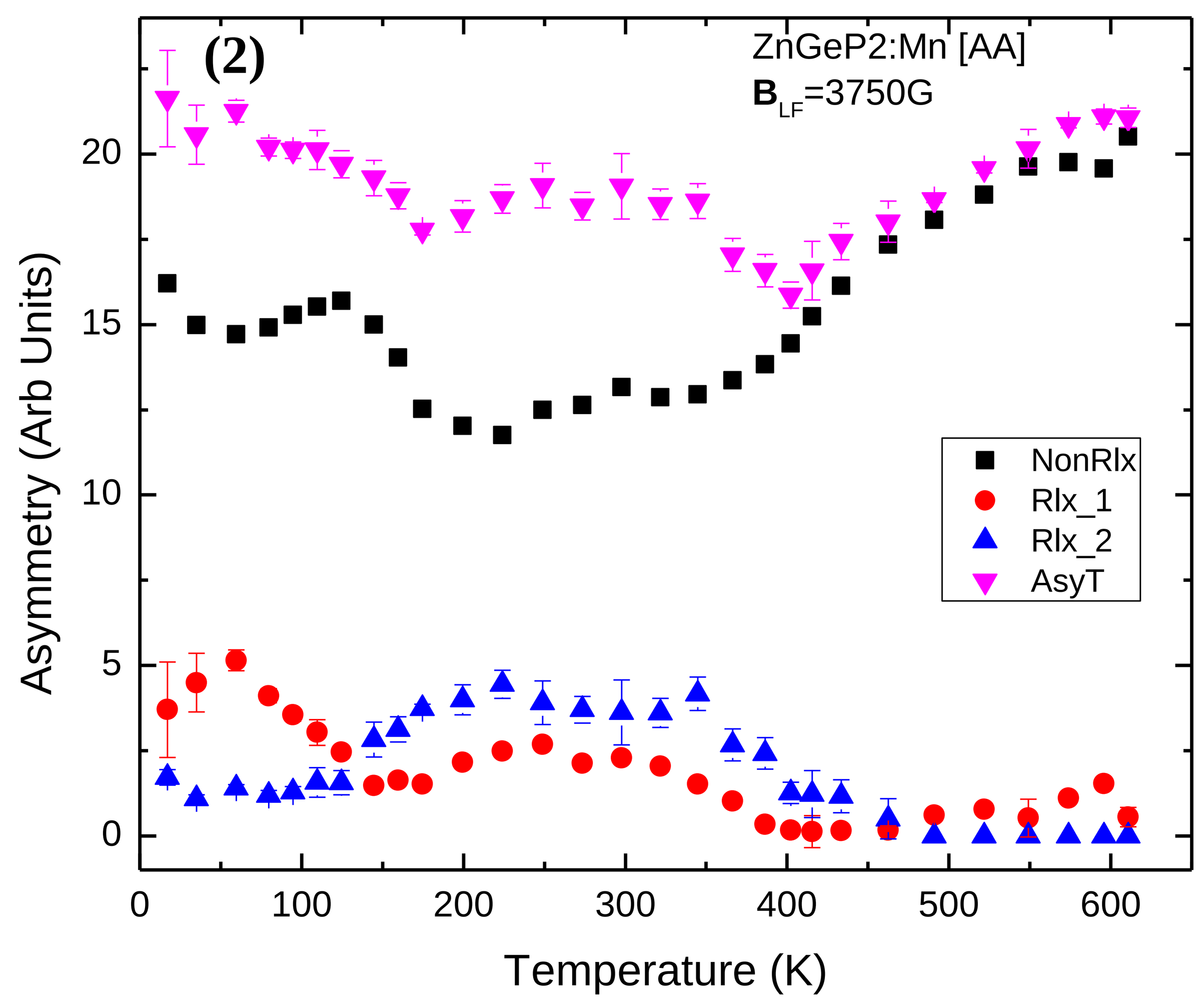
$B_{fermi}$ =Fermi contact interaction
→ Mag. interaction of $\mu^+$&e⁻ spins for *s&p* e⁻ metals
→ RKKY – indirect exchange between $\mu^+$ and unpaired e⁻ via conduction e⁻ [*d&f* materials]
→ Transferred hyperfine field [$\mu^+$ & e⁻ wavefunction overlap in insulators]

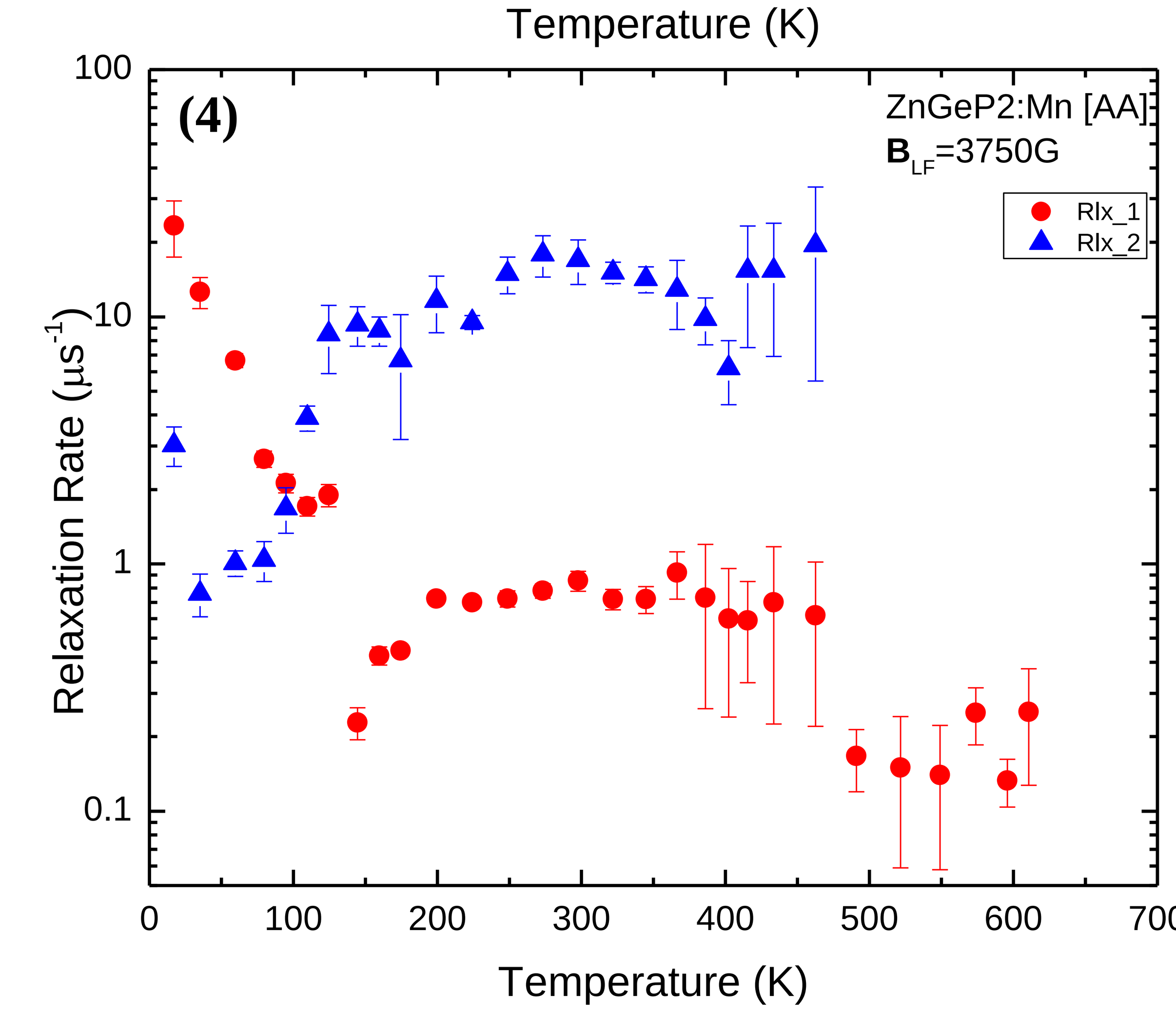
$\delta B_{loc}$= Contribution from fluctuation in neighboring magnetic moments → $\nu$



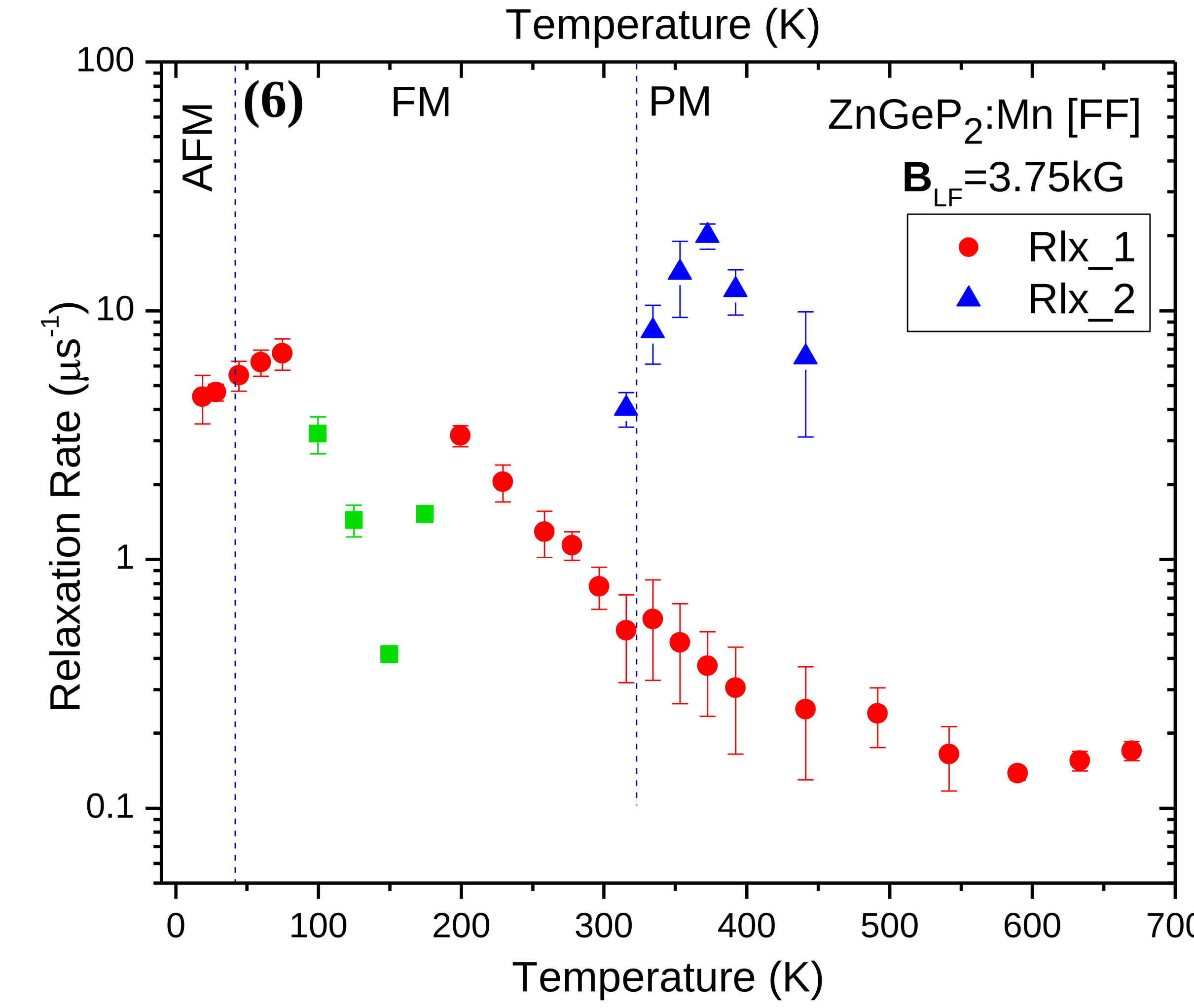




**Fig 1:**
•T-dep of asymmetry at $\mathbf{B}_{LF}$=1kG
•Lowest Mn concentration
•Dips in total amplitude correlate to regions in which we do not detect full signal due to limitations of the experimental setup
ie: $\mu^+$ relaxes too fast to resolve

**Fig 2:**
•T-dep of asymmetry at $\mathbf{B}_{LF}$ = 3.75kG
•Lowest Mn concentration
•Higher field provides additional alignment ∥ to B → increase in Asy

**Fig 3:**
•T-dep rlx rates at $\mathbf{B}_{LF}$ = 1kG
•Lowest Mn concentration
•*Rlx_1* (●): AFM fluctuations
•*Rlx_2* (▲):
T < 100K – Sharp rise, unknown
100K < T < 575K – short range FM correlations
•(● & ▲): T > 400K – fluctuations related to spin polaron formation

**Fig 4:**
•T-dep rlx rates at $\mathbf{B}_{LF}$ = 3.75kG
•Lowest Mn concentration
•(● & ▲) ibid **Fig 3**

**Fig 5:**
•T-dep rlx rates at $\mathbf{B}_{LF}$ = 1kG
•Mid range Mn concentration
•(●, ▲, ● & ▲) ibid **Fig 3**

**Fig 6:**
•T-dep rlx rates at $\mathbf{B}_{LF}$ = 3.75kG
•Highest Mn concentration
•(● & ▲) ibid **Fig 3**
•(■) Unknown – consistent feature across all samples

## $ZnGeP_2$:Mn

• $E_g \approx$ 1.83eV to 2.0eV (decreases as Mn conc. increases) [2]
• FM order above RT ($T_c \approx$ 310K to 350K) [2]
• AFM below 47K for Mn > 5% [2]
• PM/AFM below 47K mixed state for Mn < 5% [2]
• Prime candidate for spin-based electronics
  (1) Semiconducting properties
  (2) FM and AFM characteristics
• $Mn^{2+}$ substitution:
  (1) Group II: Isovalent (high concentration of $Mn^{2+}$)
  (2) Group IV: Double Acceptor (light concentration of $Mn^{2+}$)
  (3) Result of (1) and (2) [ie hole abundance] is strong FM coupling instead of the AFM order produced by group II substitution only
• Powder XRD results [3]
  (1) support 2nd ordering transition
  (2) lacks evidence to conclusively demonstrate if small inclusions of MnP dominate magnetic features
• NMR [4]
  (1) suggests 90+% of Mn atoms in MnP impurity phase with nm sized clusters for 8% to 15% Mn
  (2) No additional information for samples with Mn concentration < 8%


**Acknowledgement:**
Research is supported by the U.S. Department of Energy (Grant DE-SC0001769).


## Samples

• BAE Systems provided high quality, p-type $ZnGeP_2$:Mn

•All samples cut from the same single crystal boule from starting melt of 1.6% Mn

• AA → lowest Mn content; FF → Highest Mn content

## The Experiment

• LF muon spin relaxation measurements performed using the EMU MuSR spectrometer on a surface muon channel at ISIS in Didcot, UK

• 4 different $ZnGeP_2$:Mn samples, varying Mn concentration

• Temperature scans at $\mathbf{B}_{LF}$=1kG and $\mathbf{B}_{LF}$=3.75kG

• B-field scans at various temperatures

• P(t) fit with two Lorentzian relaxing components and one non-relaxing component


**References**
[1] A. Schenk, *Muon Spin Rotation Spectroscopy: Principles and Applications […]* (Adam Hilger Ltd, Bristol, 1985).
[2] Cho, et al., Phys Rev Lett. **88** (2002) 257203
[3] Aitken, et al., *Chem Mater* **19** (2007) 5272-5278
[4] Hwang, et al *Appl Phys Lett* **83** (2003) 1809-1811
[5] Uemura, Phys Rev B. **31** (1985) 546; Moriya, Prog. Theor. Phys, **16** (1956) 23


## Observed Features

**AFM Fluctuations (●, *Rlx_1*)[5]:**

• $\frac{1}{T_1} \sim \frac{2\Delta_i^2}{\nu}$ | $T^{1}_{1} = \mu^+$ Rlx rate; $\nu$=Spin fluctuation rate
$\Delta_i = \gamma_\mu \mathbf{B}_i$=($\mu$ gyromagnetic ratio)(RMS value fluctuating field)

**Short Range FM Correlations (▲, *Rlx_2*):**

• Additional measurements and modeling required to positively identify and further characterize short range correlations

**Fluctuations related to Spin Polaron (● & ▲):**

• Additional measurements and modeling required to positively identify and further characterize fluctuations above 400K

• $CdGeAs_2$:Mn(3%)
[very similar properties to $ZnGeP_2$:Mn, $T_c$ > 300K]
Spin precession results indicate SP above 300K

## Future Work and Open Questions

• Overall goal: Further characterize magnetic properties and further the understanding of magnetism within DMS systems
• Additional analysis and modeling to achieve better separation of relaxation rates in regions that clearly have more than 2 relaxing components; ie. 300K to 500K region in the 1kG measurements of sample 'AA' (Fig 3)
• Higher field LF measurements to slow fluctuations enough to *actually* be able to measure and follow fluctuations through transition regions
• Muon spin precession measurements to characterize local magnetic fields and features; ie:
  (1) Identify $\mu^+$, $Mu^0$ and $Mu^0$-like states
  (2) Check for well defined internal fields in FM regime
  (3) Investigate spin polaron formation and properties
• Modeling of fluctuations in DMS systems for AFM, FM, SP
• This work is start of the large scale project of studying the local magnetic features in DMS II-IV-$V_2$ and II-VI systems
• Link between local magnetic moments and bulk magnetism?
• How is magnetism distributed throughout sample?, ie:
  (1) MnP impurity phase with clustering throughout?
  (2) Distributed relatively uniformly throughout?
  (3) Something else entirely?