Magnetic Fluctuations in ZnGeP₂:Mn

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Project Focus

•Dilute Magnetic Semiconducting (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 & μ^+ 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 μ :

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



 \boldsymbol{B}_{ext} = Applied external field

 $\boldsymbol{B}_{dip} = \text{dipolar field}$

 \rightarrow Sum of localized moments over entire crystal

 \rightarrow Including site to site differences

 \boldsymbol{B}_{hvp} = Field from HF interaction

 \rightarrow Short range magnetic interaction between μ^+ and local electronic moments (cf: wavefunction overlap)

 \boldsymbol{B}_{fermi} = Fermi contact interaction

 \rightarrow Mag. interaction of $\mu^+\&e^-$ spins for $s\&p e^-$ metals

 \rightarrow RKKY – indirect exchange between μ^+ and unpaired e⁻ via conduction e⁻ [*d*&*f* materials] \rightarrow Transferred hyperfine field [μ^+ & e⁻ wavefunction overlap in insulators]

 δB_{loc} = Contribution from fluctuation in neighboring magnetic moments $\rightarrow v$

Material Properties: ZnGeP₂:Mn

- $E_g \approx 1.83$ eV to 2.0eV (decreases as Mn conc. increases) [2]
- FM order above RT ($T_c \approx 310$ K 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
- Mn²⁺ 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

AFM Fluctuations (•)[5]:

the AFM order produced by group II substitution only

• Powder XRD results [3]

support 2nd ordering transition but lacks evidence to conclusively demonstrate if small inclusions of MnP dominate magnetic features

• NMR [4]

Suggests 90+% Mn atoms in MnP impurity phase with nm sized clusters for 8% to 15% Mn; no additional information for samples with Mn concentration < 8%

The Experiment

• LF muon spin relaxation measurements:

- EMU & HiFi spectrometers on surface muon channel at ISIS in Didcot, UK ullet
- Helios spectrometer on M20 surface muon channel at TRIUMF in Vancouver, Canada
- 4 different ZnGeP₂:Mn samples, varying Mn concentration
- Temperature scans: ~2 K to 700 K at $\mathbf{B}_{LF} = \{0.1, 0.375, 0.7, 1.5\}$ T
- B-field scans at various temperatures
- Time dependent muon spin polarization, P(t), fit with up to 3 Lorentzian relaxing and 1 non-relaxing component

Samples

- BAE Systems provided 6 high quality, p-type ZnGeP₂:Mn
- •All samples cut from the same single crystal boule from starting melt of 1.6% Mn grown by horizontal gradient freeze technique

- $\frac{1}{T_1} \sim \frac{2\Delta_i^2}{v} |T_l^{-1}| = \mu^+ \text{Rlx rate; } v = \text{Spin fluctuation rate}$
 - $\Delta_i = \gamma_u \delta \mathbf{B}_i = (\mu \text{ gyromagnetic ratio})(\text{RMS value of fluctuating field})$

Short Range FM Correlations (■):

• Further analysis and modeling required to positively identify and further characterize short range correlations

FM Transition Precursors (△):

- Further analysis and modeling required to positively identify and further characterize fluctuations above 350 K
- Preliminary analysis of spin precession data hint at SP at T > 300 K
- SQUID Measurements at 350 K show field induced FM that collapses to 0 at zero applied field

Future Work and Open Questions

- Overall: Improve understanding of magnetism within DMS systems
- Additional data analysis: improve separation of relaxation rates in regions containing more than 2 components; ie. 300K to 500K
- Modeling of fluctuations in DMS systems for AFM, FM, SP
- Link between dilute local moments and bulk magnetism?
- How is magnetism distributed throughout sample?, ie: (1) MnP impurity phase with clustering throughout? (2) Distributed relatively uniformly throughout?

[5] Uemura, Phys Rev B. **31** (1985) 546; Moriya, Prog. Theor. Phys, **16** (1956) 23





[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

