



Probing internal magnetic field features of the II-IV-V₂:Mn DMS via MuSR

An in depth study exploring fundamental properties and characteristic of local magnetic features in II-IV-V₂:Mn chalcopyrite DMS systems

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BAE Systems, Adv. Systems and Tech. Nashua, NH

Spring APS Texas Section Meeting: 22-24/Mar/2012

Support:

Provided by: US DoE

Experimental Facility:

EMU Beamline at ISIS

(Rutherford Appleton Labs, Didcot, UK)



LF- μ SR

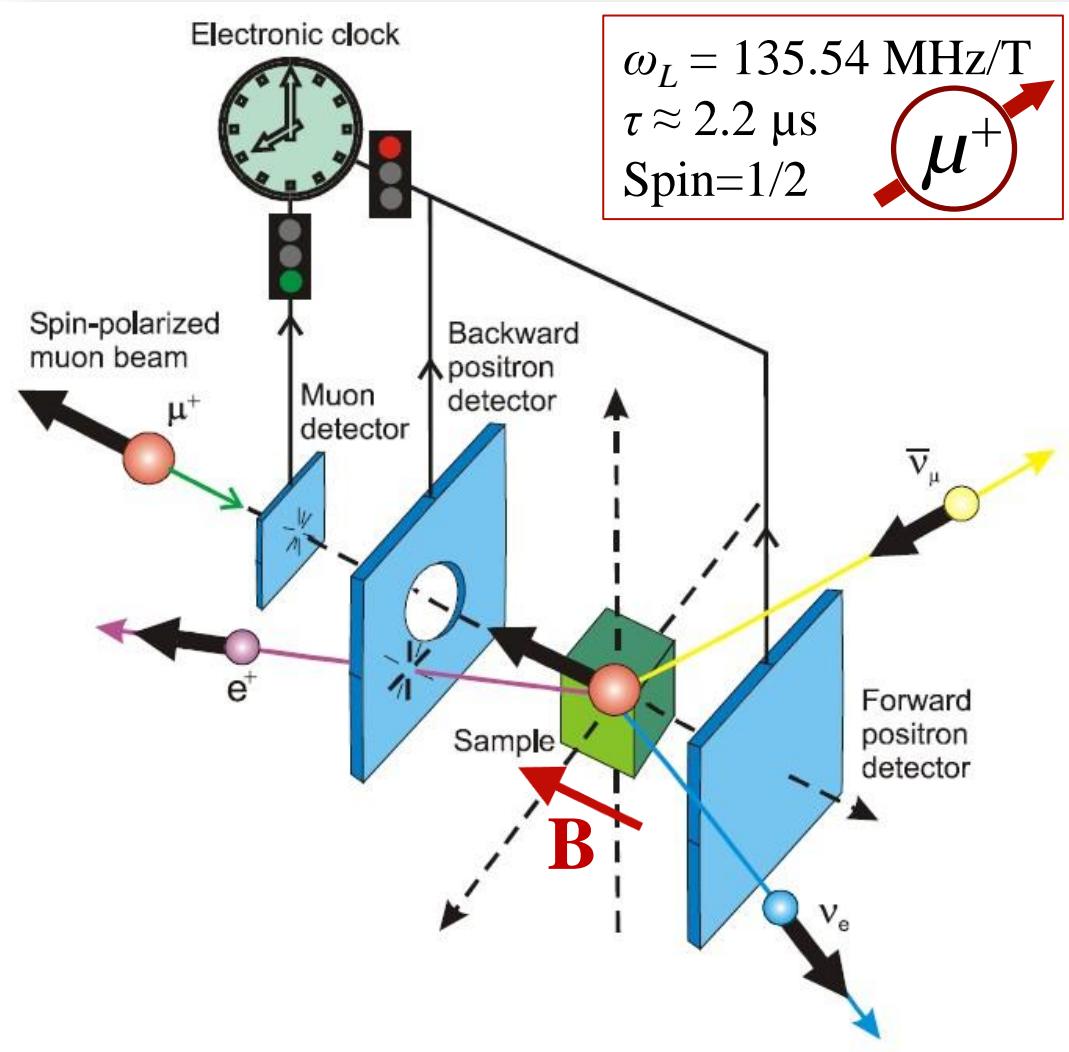
μ^+ = Local probe

Implant 100% spin polarized muons

\mathbf{B} applied \parallel to initial spin direction

Spin evolves in local environment

Evolution of polarization tracked



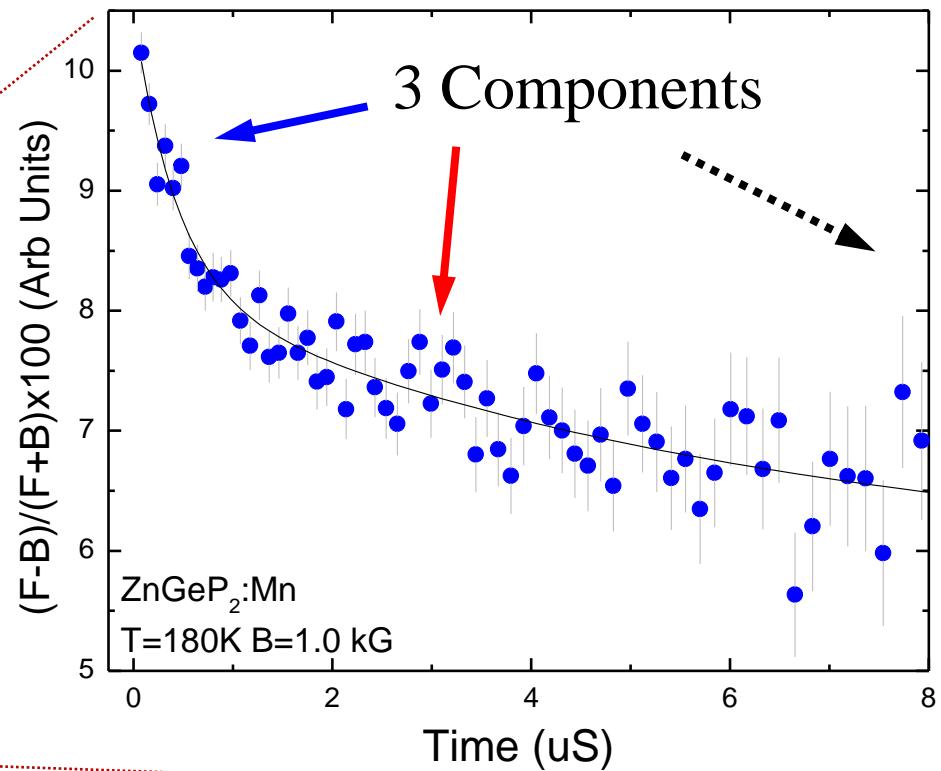
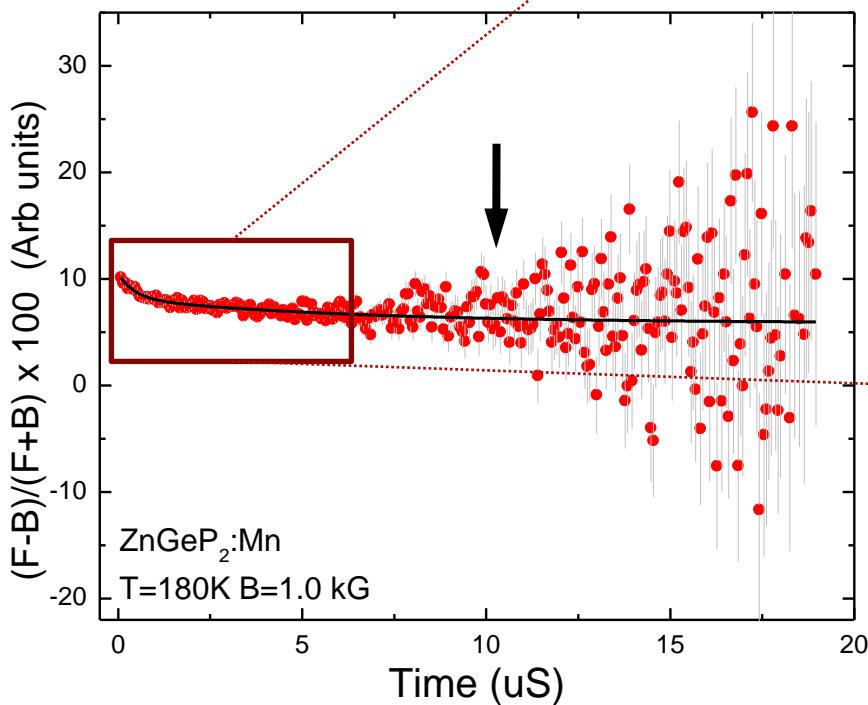


LF- μ SR on ZnGeP₂:Mn: T=180 with B_{LF}=1.0 kG

Asymmetry: ‘corrected’ average of counts

$$A(t) = \frac{N_F(t) - \alpha N_B(t)}{N_F(t) + \alpha N_B(t)}$$

Data: Shown



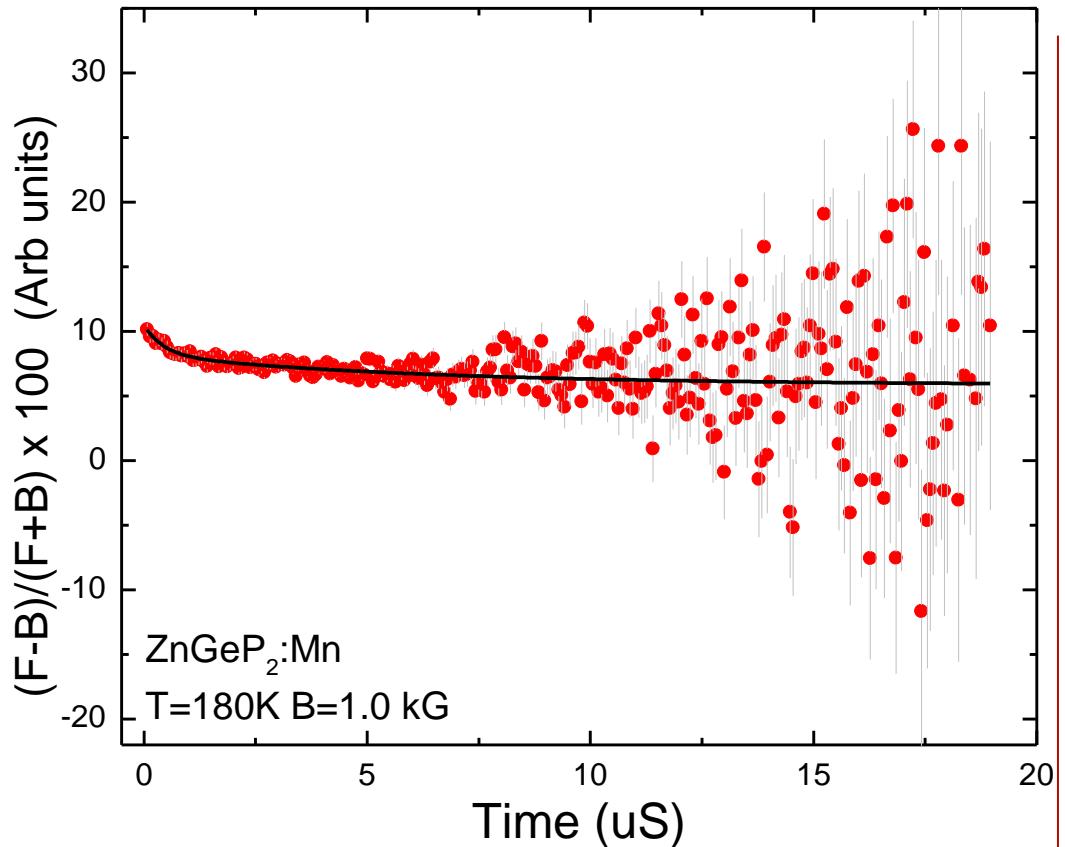
Faster: $1 \rightarrow 25+ \text{ us}^{-1}$

Slower: $\sim 0.01 \rightarrow 1.0 \text{ us}^{-1}$

Non-Rlx: $\lambda < 0.01 \text{ us}^{-1}$



Polarization Function



$$P(t) = A_1 e^{-t/T_1} + A_2 e^{-t/T_2} + A_3$$

Fit Parameters: A_i, T_j

$$\frac{1}{T_j} = \frac{2\Delta_i^2}{\nu_j}$$

For dilute alloys:

$$\rho(\Delta_i) = \sqrt{\frac{2}{\pi}} \frac{a}{\Delta_i^2} \exp\left(-\frac{a^2}{2\Delta_i^2}\right)$$

$$\Delta = \gamma_\mu B_i$$

$$P_z^L(t) = \int_0^\infty P_z^G(t) \rho(\Delta) d\Delta = \dots$$

ie: A. Schenk, *Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics* (Adam Hilger Ltd, Bristol, 1985).



B-Field at Mu+ Site

Localized magnetic density contributing to \mathbf{B}_{loc} via:

- 1) classical dipole interaction
- 2) RKKY [metals] or via hyperfine field [insulators]

$$\begin{aligned}\mathbf{B}_{\text{loc}} &= \langle \mathbf{B}_{\text{loc}} \rangle + \delta \mathbf{B}_{\text{loc}} \\ &= \mathbf{B}_{\text{ext}} + \mathbf{B}_{\text{dip}} + \mathbf{B}_{\text{hyp}} + \mathbf{B}_{\text{fermi}} + \delta \mathbf{B}_{\text{loc}}\end{aligned}$$

v

\mathbf{B}_{ext} = Applied External Field

$$\mathbf{B}_{\text{dip}} = \frac{\mu_0}{4\pi} \sum_{i=1}^N (-g_i \mu_B) \left[-\frac{\mathbf{J}_i}{r_i^3} + \frac{3(\mathbf{J}_i \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} \right] + \frac{\mu_0}{4\pi} \sum_{i=1}^N (-\gamma_i \hbar) \left[-\frac{\mathbf{I}_i}{r_i^3} + \frac{3(\mathbf{I}_i \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} \right]$$

= dipolar field -- sum of contributions from localized magnetic moments over whole crystal

\mathbf{B}_{hyp} = field from HF interaction; $\mathbf{B}_{\text{hyp}} = \frac{\mu_0}{4\pi} \sum_{i=1}^{N'} \frac{(-g_i \mu_B) H_{\mathbf{r}_i}}{v_c} \mathbf{J}_i$
short range magnetic interaction between μ^+ and local magnetic moments near μ^+ site

$\mathbf{B}_{\text{Fermi}}$ = Fermi magnetic field operators, i.e.:

Fermi contact interaction [magnetic interaction of μ^+ & e^- spins for s & p e^- metals]

RKKY – indirect exchange between μ^+ and unpaired e^- via conduction e^- [d & f materials]

Transferred hyperfine field [μ^+ & e^- wavefunction overlap in insulators]



Practical Material: II–IV–V₂:Mn

ZnGeP₂:Mn

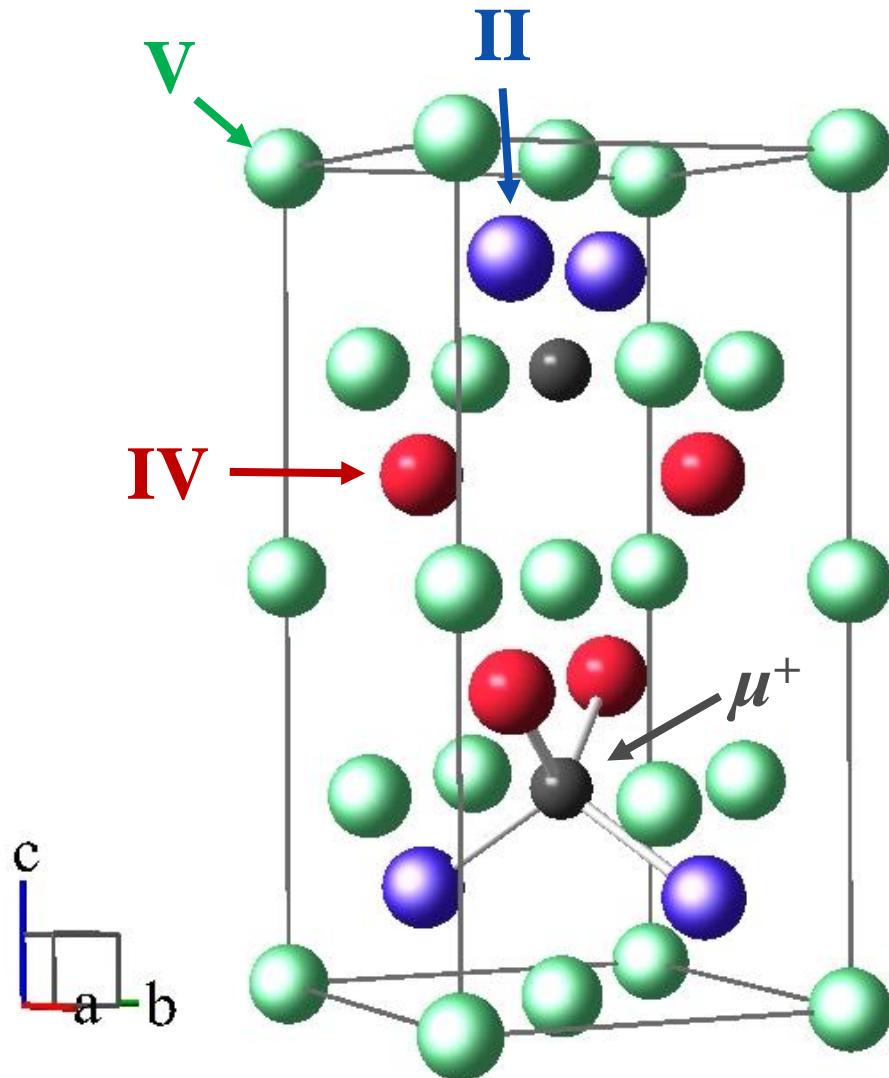
- $E_g \approx 2.0$ eV & FM order above RT
($T_c = 310\text{K}$ to 350K)
- Prime choice for *spin*-based devices

CdGeAs₂:Mn

- $E_g \approx .67$ eV & FM order above RT
($T_c = 355\text{K}$)

Mn²⁺ Substitution:

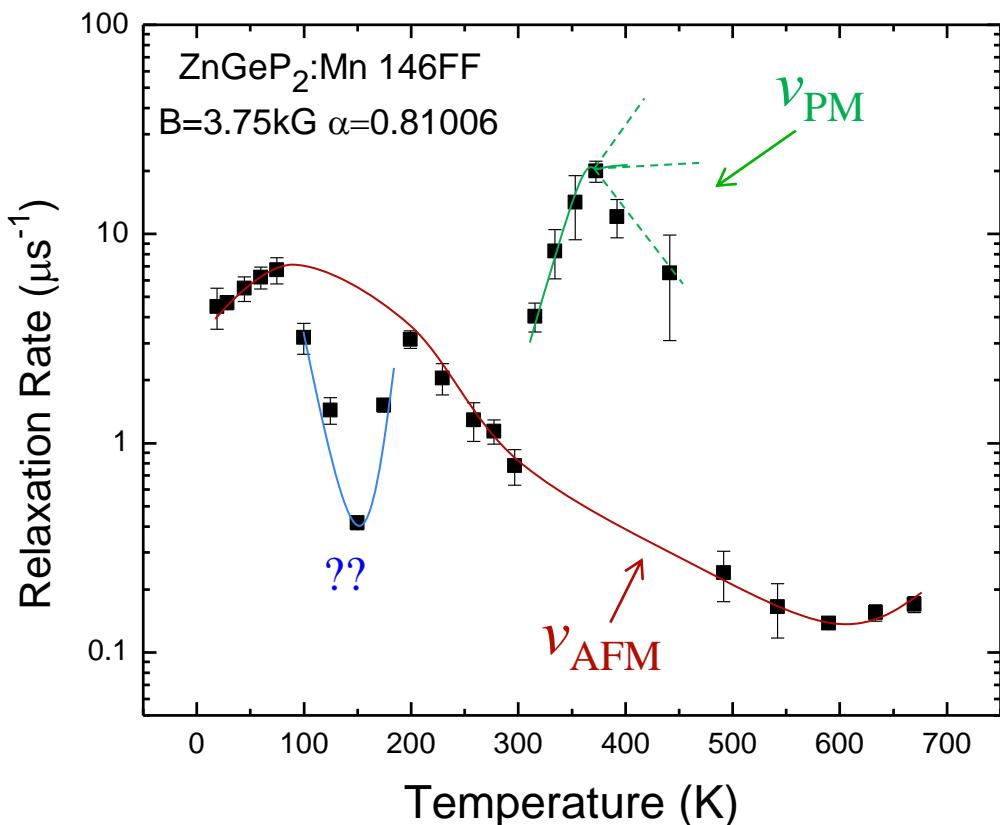
- Isovalent – Group-II
- Double Acceptor – Group-IV
- Light conc. of Mn²⁺ on IV
- Heavy conc. of Mn²⁺ on II
→ AFM if Mn only on II
- With Mn sub on IV sites, provides extra carriers, mix results in FM order





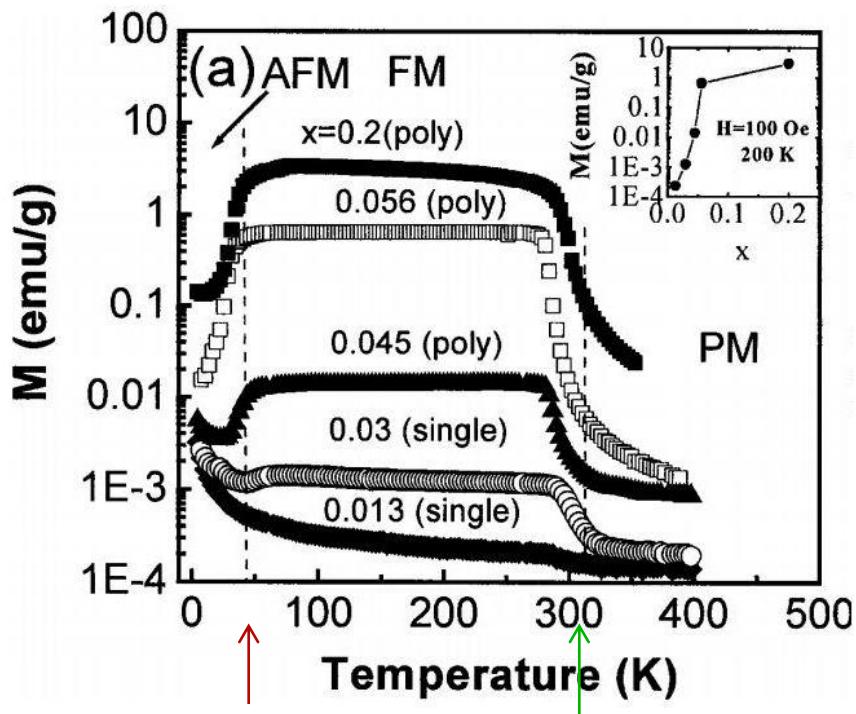
ZnGeP₂:Mn – Preliminary results

High quality, single crystal, P-type samples, doping <5% Mn → Dilute regime



NOTE: Lines *only* guide for the eyes

P.W. Mengyan, et al., Unpublished data, ISIS, Dec 2012

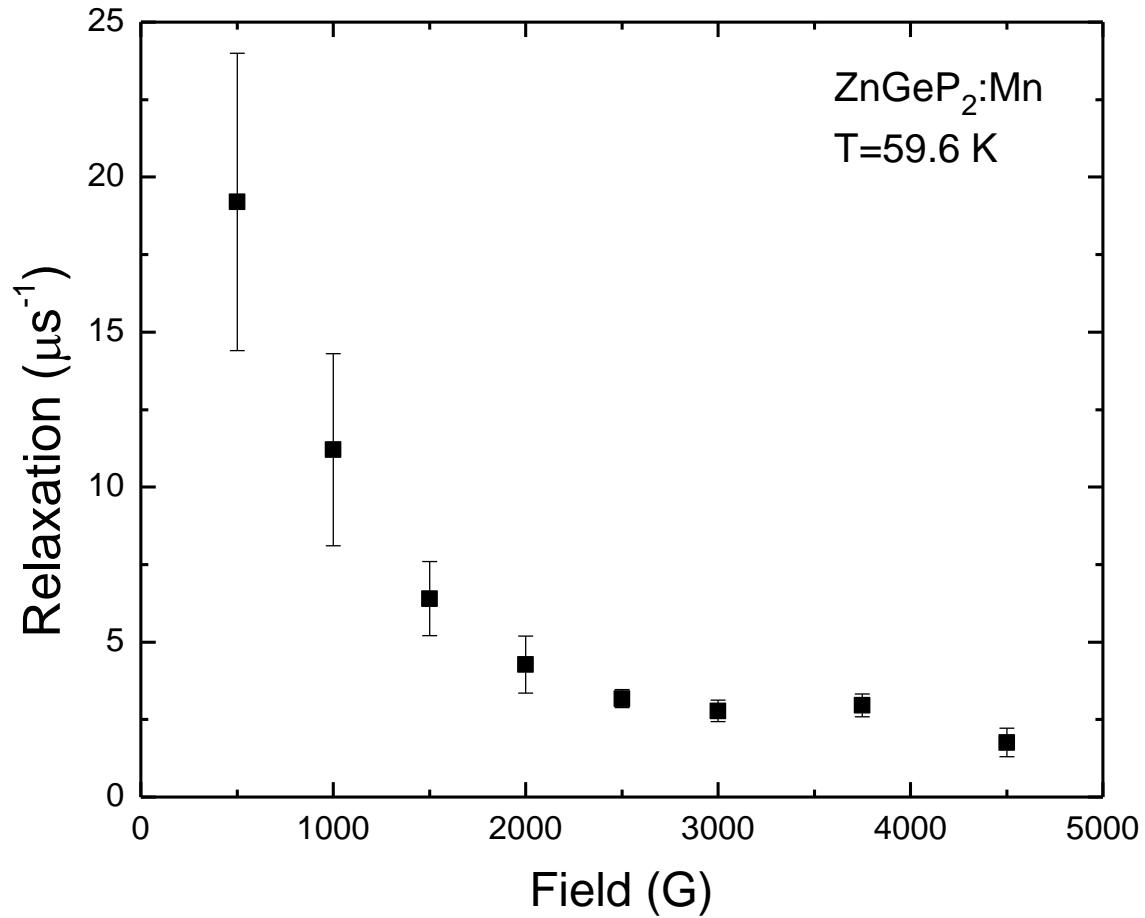


S. Cho, et al., PRL 88 (2002) 257203

P.W. Mengyan, et al. APS (2012)



ZnGeP₂:Mn – Preliminary results





Future Work

Additional data fitting & theoretical modeling

- Develop model designed specifically for these DMS systems
- Reanalyze data with new model, extract fluctuation rates, energies, etc

These measurements supplementary to additional work to characterize internal field properties and characteristics, ie:

- distribution
- structure
- dynamics
- how magnetism carried/transferred throughout DMS



Thank you

Questions, Comments, etc?

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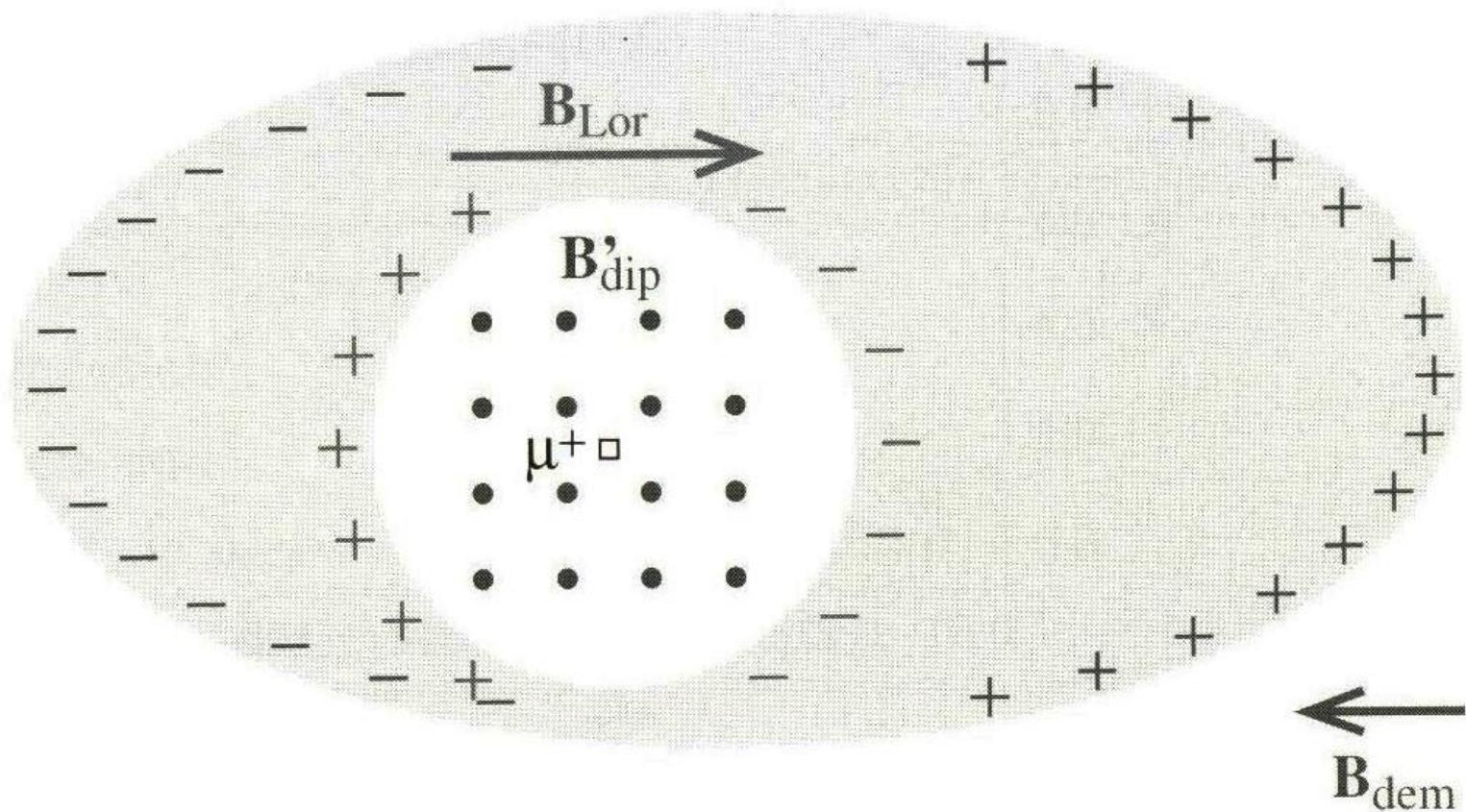
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B-Field at Mu+ Site

\mathbf{B}_{dip}

$$\mathbf{B}_{\text{loc}} = \mathbf{B}_{\text{ext}} + (\mathbf{B}'_{\text{dip}} + \mathbf{B}_{\text{Lor}} + \mathbf{B}_{\text{dem}} + \delta\mathbf{B}_{\text{dip}}) + \mathbf{B}_{\text{hyp}}$$

\mathbf{B}_{ext} = Applied External Field

$$\mathbf{B}_{\text{dip}} = \frac{\mu_0}{4\pi} \sum_{i=1}^N (-g_i \mu_B) \left[-\frac{\mathbf{J}_i}{r_i^3} + \frac{3(\mathbf{J}_i \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} \right] + \frac{\mu_0}{4\pi} \sum_{i=1}^N (-\gamma_i \hbar) \left[-\frac{\mathbf{I}_i}{r_i^3} + \frac{3(\mathbf{I}_i \cdot \mathbf{r}_i)\mathbf{r}_i}{r_i^5} \right]$$

= dipole field expressed as sum of contributions from magnetic moments over whole crystal

\mathbf{B}'_{dip} = Sum of fields from within *Lorentz sphere* centered at μ^+ site
sphere taken to be large enough to allow \mathbf{B}_{dip} sum to converge

\mathbf{B}_{Lor} = Field from charges on surface of Lorentz sphere, ie: $\mathbf{B}_{\text{Lor}} = \frac{\mu_0}{3} \mathbf{M}_{\text{Lor}}$
 \mathbf{M}_{Lor} = vector sum magnetic moments inside sphere per unit volume

\mathbf{B}_{dem} = Field from charges on the surface of sample, ie: $\mathbf{B}_{\text{dem}} = -\mu_0 \mathbf{N} \mathbf{M}_{\text{meas}}$
 \mathbf{N} = Demagnetization tensor ; \mathbf{M}_{meas} = Bulk magnetization of sample

$\delta\mathbf{B}_{\text{dip}}$ = Fluctuating component of dipolar field contribution

\mathbf{B}_{hyp} = field from HF interaction; $\mathbf{B}_{\text{hyp}} = \frac{\mu_0}{4\pi} \sum_{i=1}^{N'} \frac{(-g_i \mu_B) H_{\mathbf{r}_i}}{\nu_c} \mathbf{J}_i$
short range magnetic interaction between μ^+ and local magnetic moments near μ^+ site



Experimentally Accessible Analog to Hydrogen

	Muon	Proton
Mass (m_p)	$0.1126 \approx 1/9$	1
Spin	$1/2$	$1/2$
Gyro. Ratio, γ ($s^{-1} T^{-1}$)	8.51607×10^8 $\approx 3.2 \times \gamma_P$	2.67520×10^8
Lifetime, τ (μs)	2.19709	Stable
	Muonium	Hydrogen
Red. e^- mass (m_e)	0.995187	0.999456
G. S. Radius (\AA)	0.531736	0.529465
G. S. Energy (eV)	-13.5403	-13.5984

Muonium ($\text{Mu} \equiv \mu^+ e^-$)

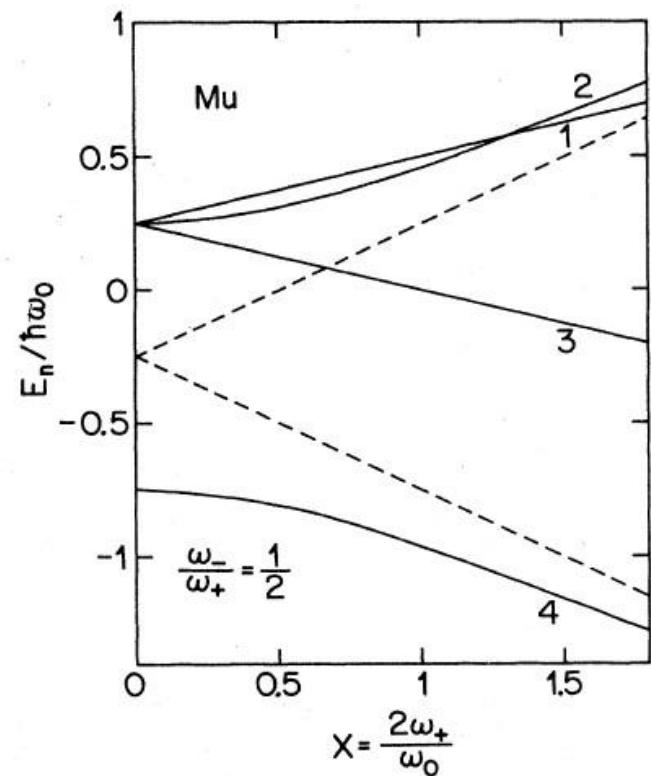
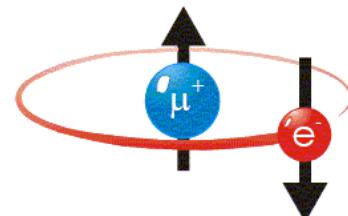


FIG. 1. The hyperfine energy-level (Breit-Rabi) diagram for isotropic 1s-Mu as a function of the dimensionless magnetic field $x = B(g_\mu\mu_\mu - g_e\mu_B)/(\hbar A)$. A fictitious value for the quantity ω_-/ω_+ has been used for clarity; its true value is 0.9904. The dashed lines are the high-field asymptotes for levels 2 and 4.



Why μ^+ ?

	Muon	Proton
Mass (m_p)	$0.1126 \approx 1/9$	1
Spin	$1/2$	$1/2$
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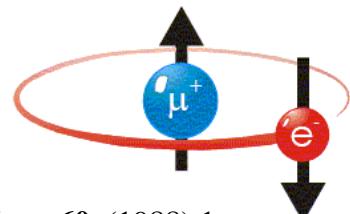
$$\mu^+ \sim p^+$$

$$m_\mu \approx 1/9 m_p$$
$$m_\mu \approx 207 m_e; S=1/2$$

Local probe

$$\text{Mu}^0 = \mu^+ + e^-$$

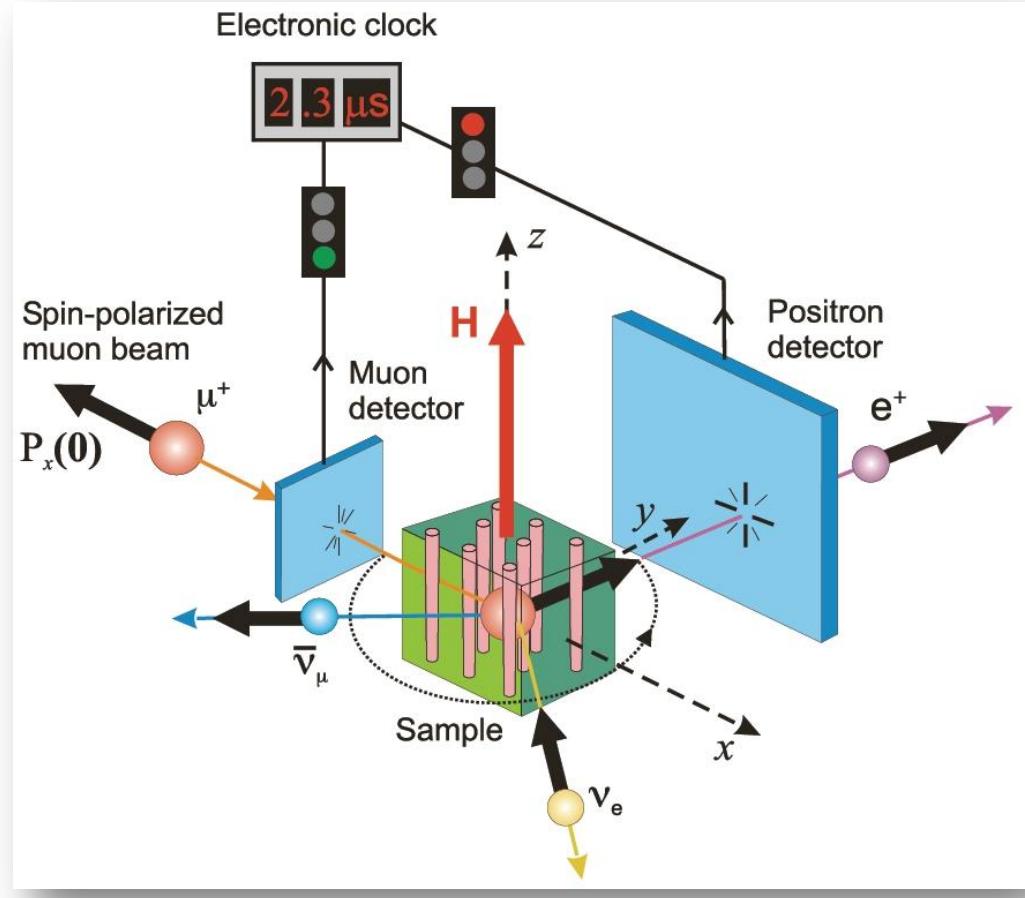
Muonium ($\text{Mu} \equiv \mu^+ e^-$)



Mu^0 ~light isotope H
ie: Early history of H impurities



TF- μ SR



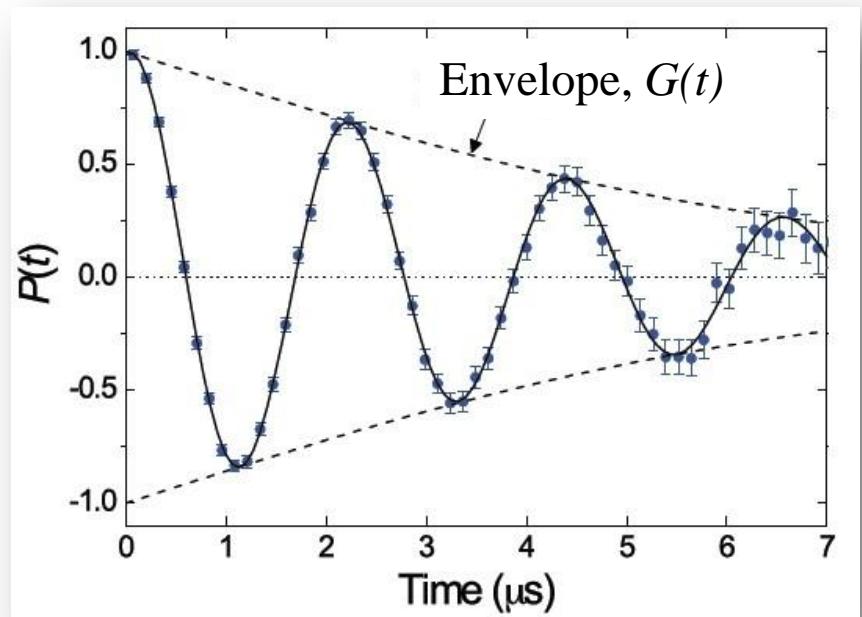
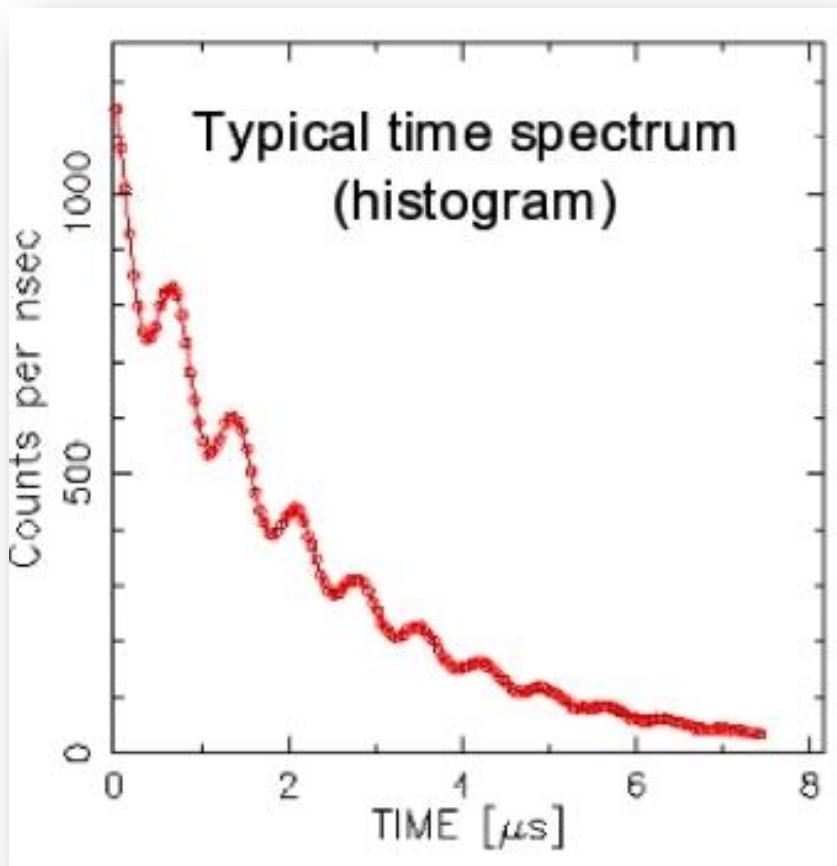
Field applied \perp to initial spin polarization
→ μ^+ spin precession about applied field at:

$$v_{\mu+} = \gamma_\mu \times |\mathbf{B}| \quad |\gamma_\mu| = 135.54 \text{ MHz/T}$$

$Mu^0 = \mu^+ + e^-$
→ spin-orbit coupling
→ affects local field of μ^+
→ diff prec. Freq for:
 $|\uparrow_\mu\rangle + |\uparrow_e\rangle$ & $|\uparrow_\mu\rangle + |\downarrow_e\rangle$



TF- μ SR: Sample signal from relaxing μ^+



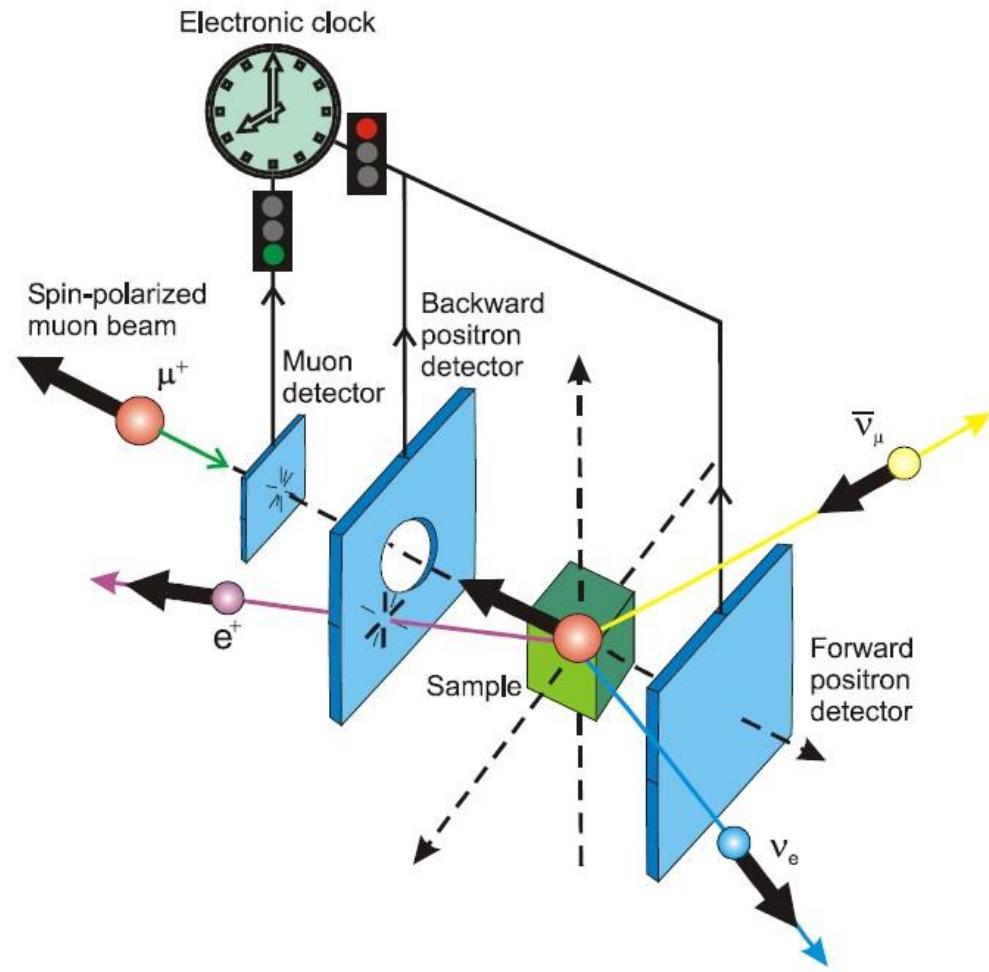
$$P(t) = G(t) \cos\left(\gamma_{\mu^+} |\mathbf{B}_{\mu^+}| + \delta\right)$$



LF- μ SR

B applied \parallel to μ^+ spin pol.
See time evolution of P(t)
along original direction

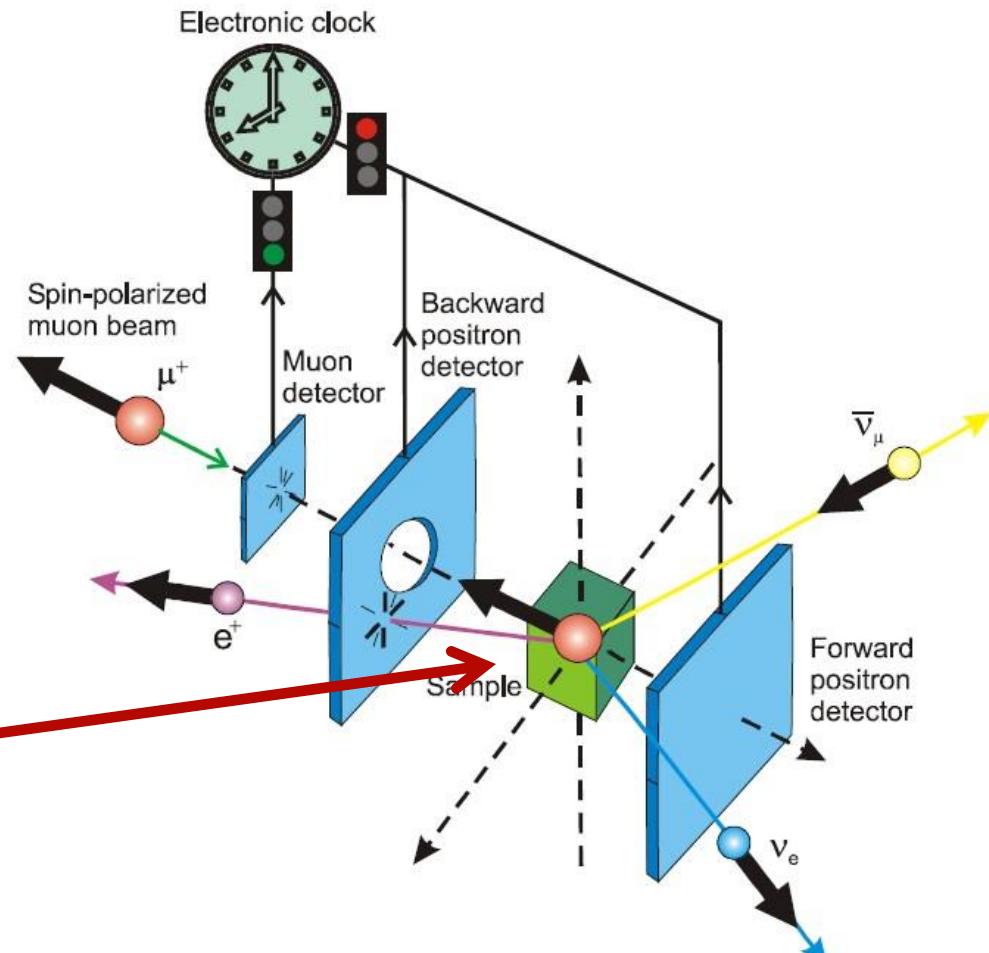
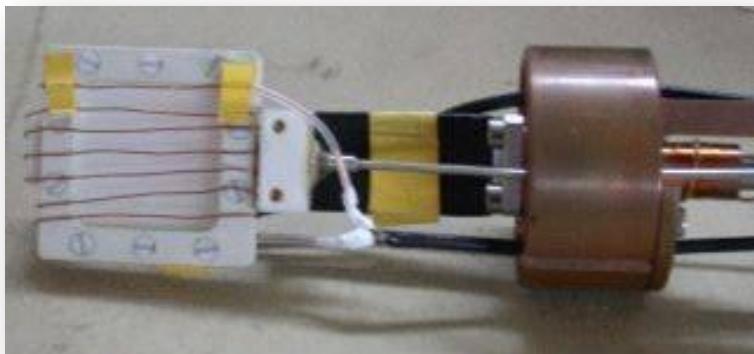
=> Change in Spin P(t) from:
1) local environment (nearby
nuclear moments)
2) muonium motion
(e^- spin-flip w/ each site
change, transferring back to
 μ^+ contributing to $\Delta P(t)$)





RF- μ SR

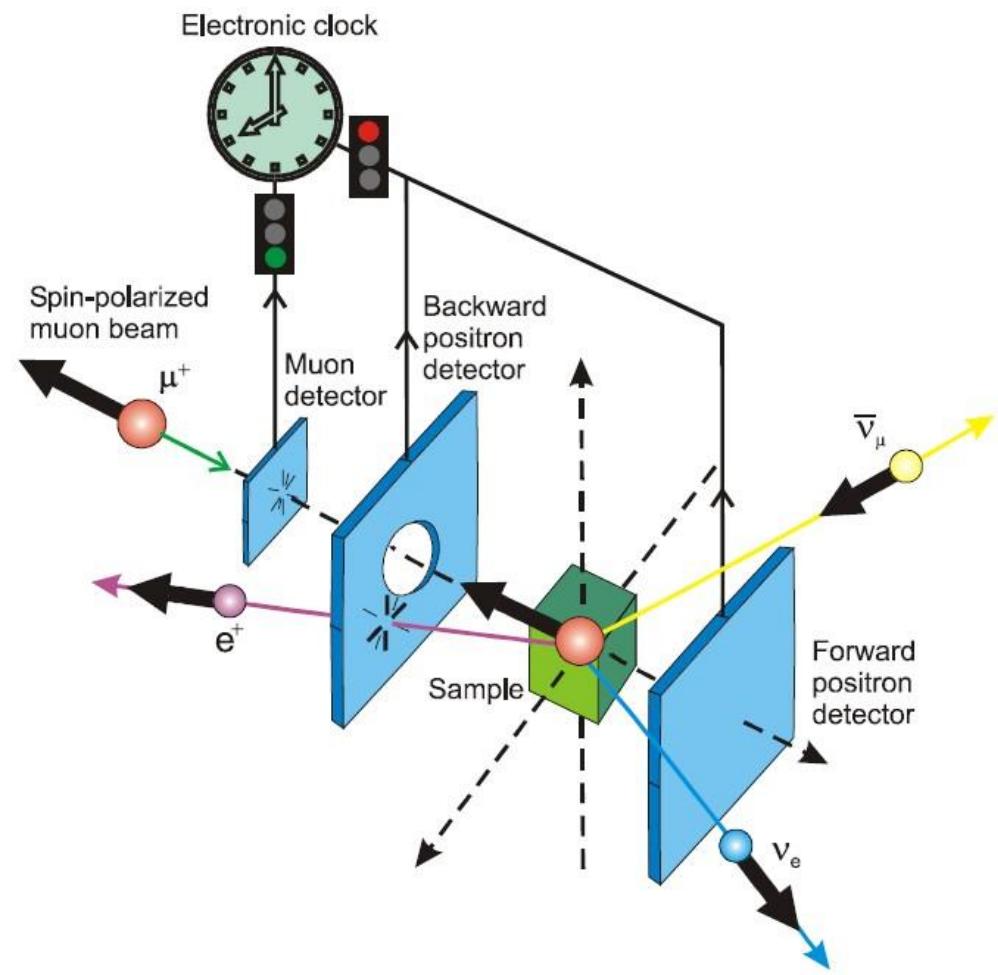
Start with LF setup
Oscillating field applied
to drive transitions
between Zeeman level(s)



Picture: J. Lord, RF- μ SR and Pulsed Techniques,
<http://www.isis.stfc.ac.uk/groups/muons/muon-training-school>



ZF- μ SR



No net B applied
See time evolution of P(t)
in natural environment

=> Change in Spin P(t)
from:
1) local environment
(nearby nuclear moments)
2) μ^+ motion



Practical Material: II–IV–V₂:Mn

ZnGeP₂:Mn

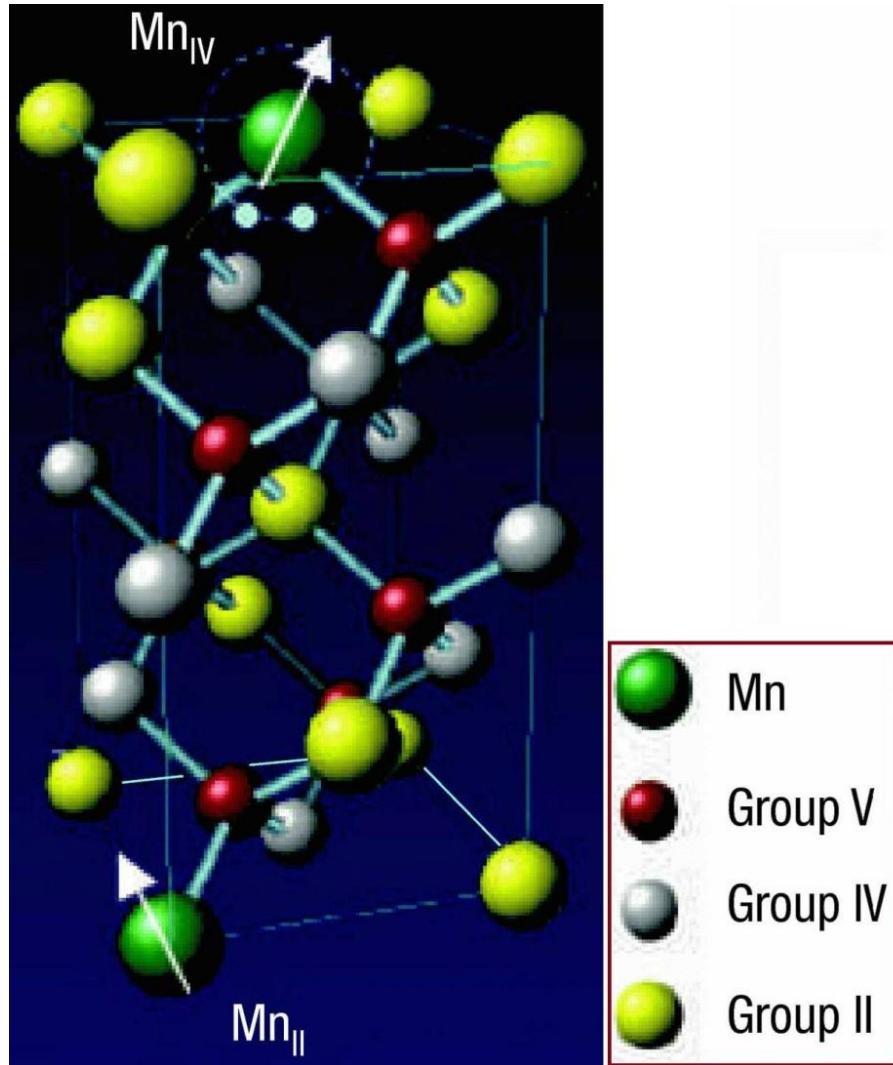
- $E_g \approx 2.0$ eV & FM order above RT ($T_c = 310K$ to $350K$)
→ Prime choice for *spin*-based devices

CdGeAs₂:Mn

- $E_g \approx .67$ eV & FM order above RT ($T_c=355K$)

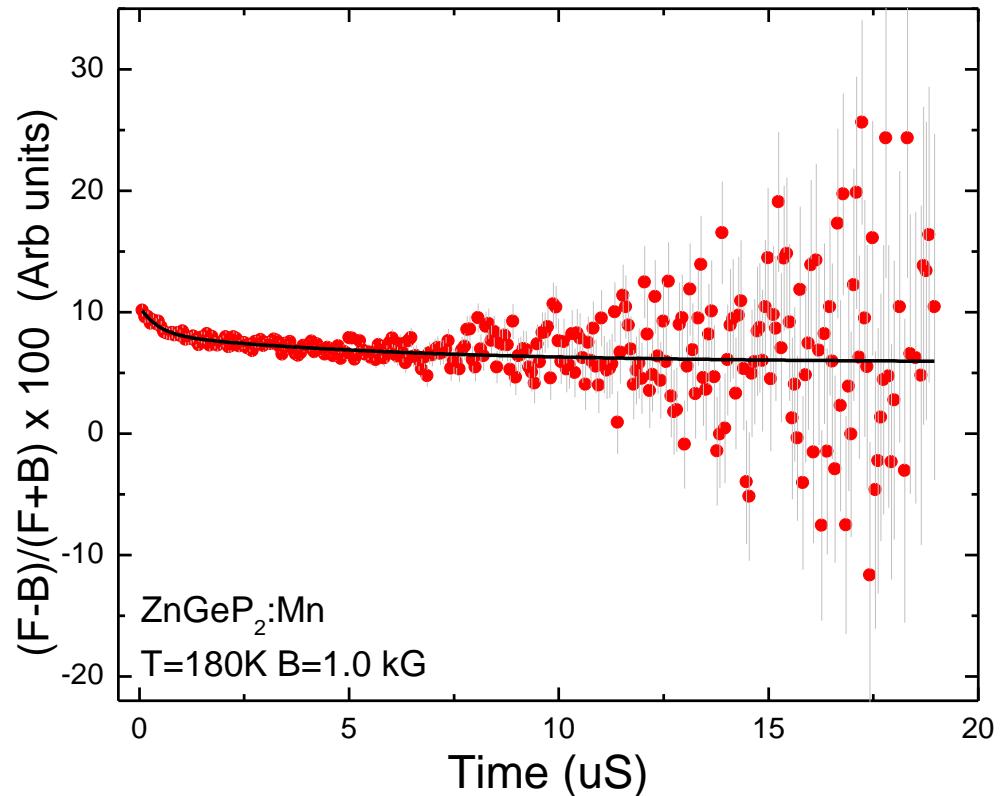
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- Isovalent – Group-II
- Double Acceptor – Group-IV
- Light conc. of Mn²⁺ on IV
Heavy conc. of Mn²⁺ on II
→ AFM if Mn only on II
- With Mn sub on IV sites, provides extra carriers, mix results in FM order





Polarization Function



$$P_z^G(t) = e^{-t/T_1} + e^{-t/T_2} + C$$

$$P_z(t) \approx e^{\left(\frac{-t}{T_1}\right)^\beta} \left| \frac{1}{T_1} = \frac{2\Delta_i^2}{\nu} \right.$$

$$\rho(\Delta_i) = \sqrt{\frac{2}{\pi}} \frac{a}{\Delta_i^2} \exp\left(\frac{-a^2}{2\Delta_i^2}\right) \left| \Delta_i = \gamma_\mu B_i \right.$$

$$P_z^L(t) = \int_0^\infty P_z^G(t) \rho(\Delta) d\Delta = \dots = \exp\left[-\left(\frac{4a^2 t}{\nu}\right)^{1/2}\right]$$