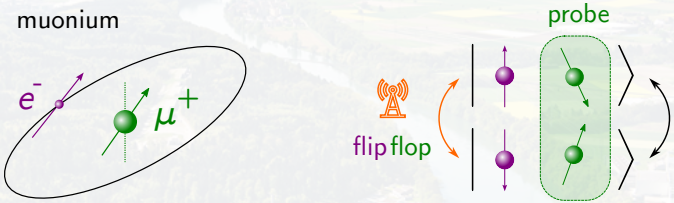
Multi-Quantum Spectroscopy of Anisotropic Muonium



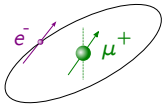
Andrin Doll
Thomas Hicken, Thomas Prokscha, Zaher Salman

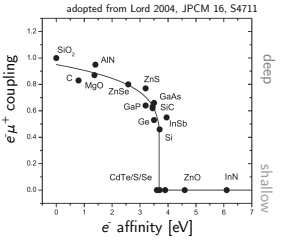
PSI

Main Idea

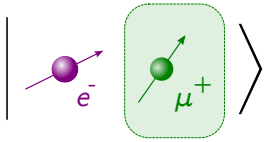
1. Microwave excitation in μSR
   - Electron gyromagnetic ratio γ_e
   - ➥ Muonium transitions

muonium





2. Relation to previous work
   1. μSR with GHz microwaves
      - Muonium spectroscopy
        Patterson, Rev. Mod. Phys, **1988**
        Nishimura *et al.*, Phys. Rev. A, **2021**
   2. μSR with MHz radio-frequencies
      - RF muon rotation
        Giblin *et al*, Nucl. Instrum. Meth. Phys. Res., **2014**
      - DEMUR, Decoupling, Delayed formation
        Patterson, Rev. Mod. Phys, **1988**
        Lord *et al*, J. Phys. Condens. Matter, **2004**



probe

Main Idea

1. Microwave excitation in μSR
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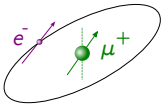
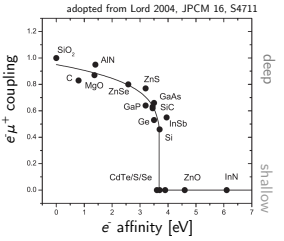
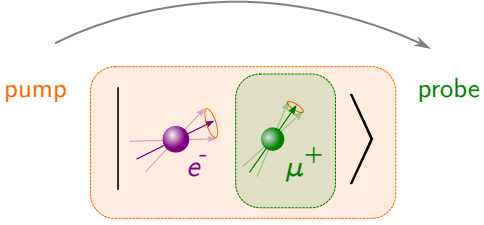
muonium





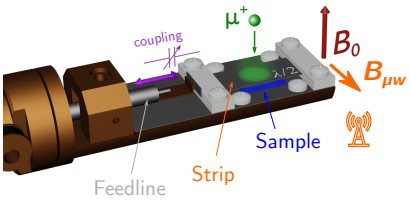
mechanisms .... and why bother?

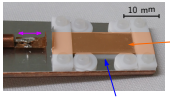


Realization in practice

probehead

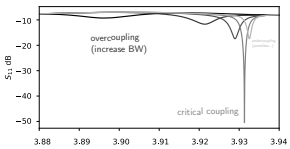
λ/2: 3.8 GHz (28 mm)



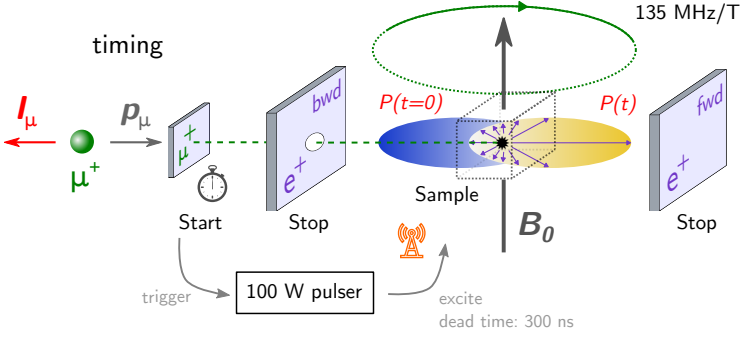


coupling

example data at 80 K (at low T, up to 30 G of uwave field)



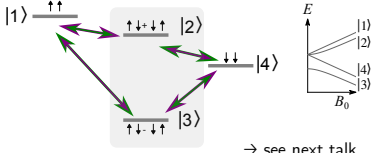
timing



see also Doll *et al.*, PRR 7, 033059, 2025

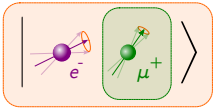
Detectable microwave transitions

strong coupling

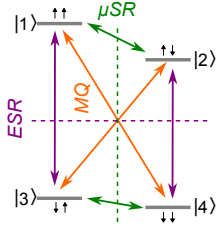


→ see next talk

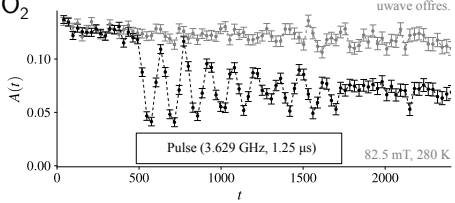
pump probe



weak coupling + anisotropy

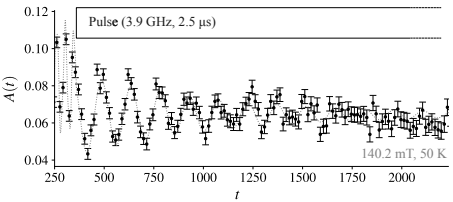


SiO_2



$t_\pi \approx 70$ ns

Si

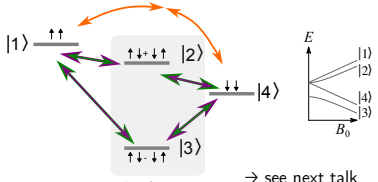


see also Doll *et al.*, PRR 7, 033059, 2025

Detectable microwave transitions

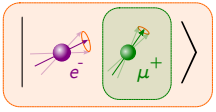
strong coupling

DQ RF:
S. R. Kreitzman, Hyperfine Interact. 65, 1055 (1991)

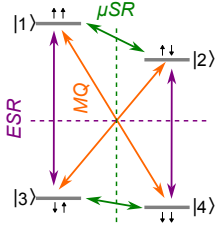


→ see next talk

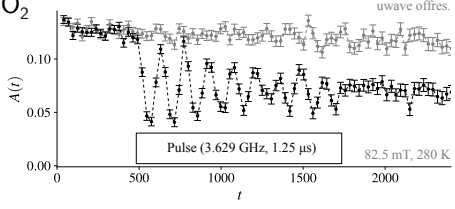
pump probe



weak coupling + anisotropy



SiO_2



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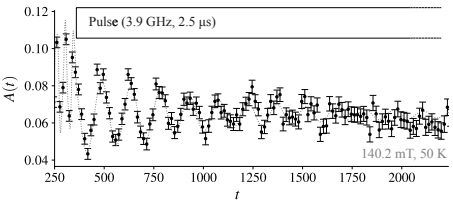


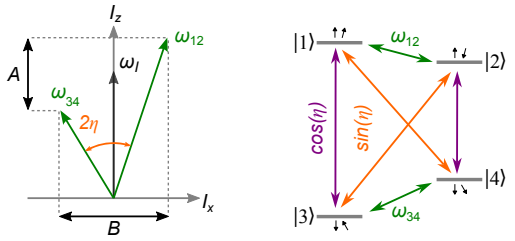
see also Doll *et al.*, PRR 7, 033059, 2025

State mixing by dipolar coupling

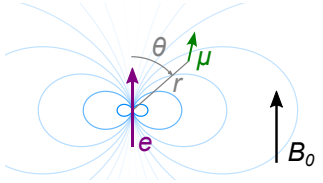
$$\hat{\mathcal{H}}_0 = \underbrace{\omega_S \cdot \hat{S}_z + \omega_I \cdot \hat{I}_z + A \cdot \hat{S}_z \hat{I}_z}_{\text{diagonal}} + \underbrace{B \cdot \hat{S}_z \hat{I}_x}_{\text{mixing}}$$

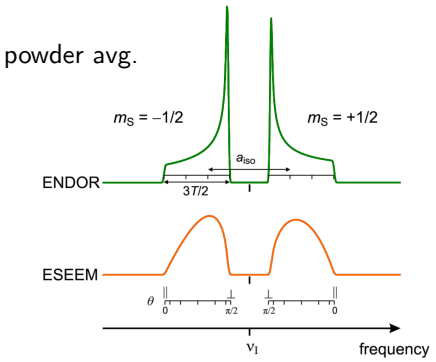
$S = 1/2$ and high-field limit
$I = 1/2$ (not in high-field limit)

many more routes to state mixing
for $S > 1/2$ or $I > 1/2$
see e.g. Schweiger/Jeschke book



$$A = T \cdot (3\cos^2(\theta) - 1) + a_{\text{iso}}$$
$$B = 3T \sin(\theta)\cos(\theta) \quad \rightarrow \eta \neq 0$$

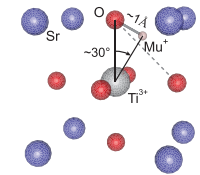


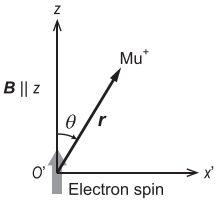


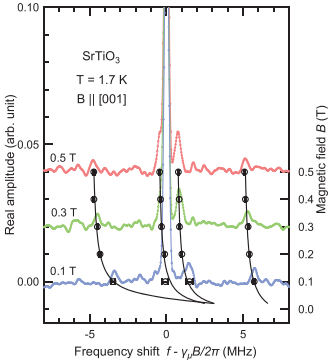
MQ-μSR on $SrTiO_3$

crystal: TF

Salman et al., PRL 113, 156801 (2014)
Ito *et al.*, APL 115, 192103 (2019)



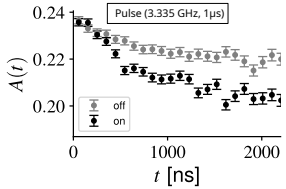


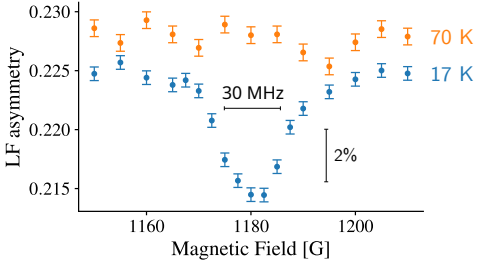


$T = 15$ MHz (dip coup)

Figures adapted from Ito *et al.*

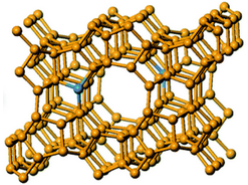
powder: MQ

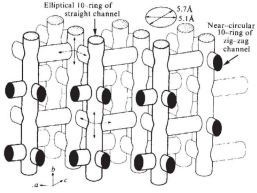




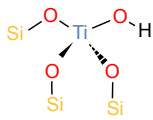
Titanium silicalite-1

Zeolite





Active site

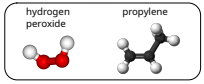


$g_\perp \approx 1.93$

$g_\parallel \approx 1.99$

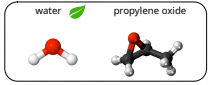
Morra et al., Chem. Eur. J. 20 (2014)

Input

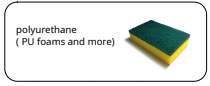


TS-1

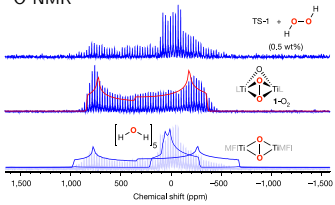
Output



Product



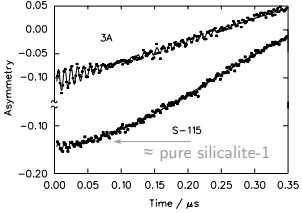
^{17}O NMR



Gordon *et al.*, Nature 586, 708-713 (2020)

→ active site is dinuclear !

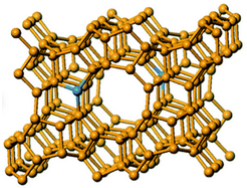
Zeolite TF μSR

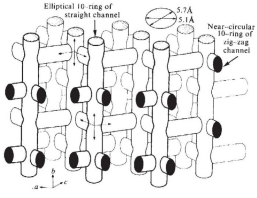


Arsenau et al., Phys. B: Condens. Matter 326, 2003

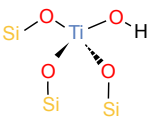
MQ-μSR on TS-1

Zeolite





Active site

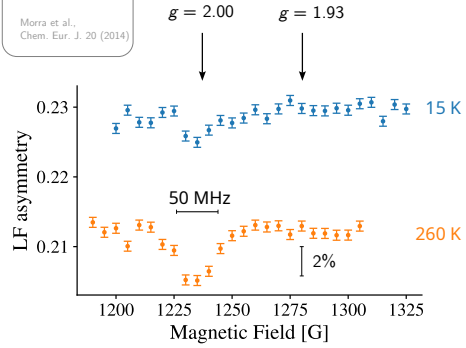


Sealed flypast cell

$g_\perp \approx 1.93$

$g_\parallel \approx 1.99$

Morra et al., Chem. Eur. J. 20 (2014)



→ electron probably on Si

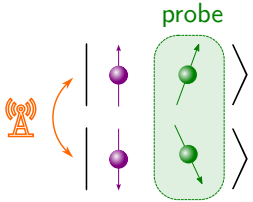
→ upcoming experiments on silicalite-1 to clarify role of Ti

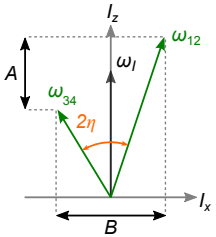
Summary and Outlook

- MQ-μSR
  - ➥ Selective probe of anisotropic muonium
- Further work
  1. How small η are detectable
  2. How to extract powder spectra

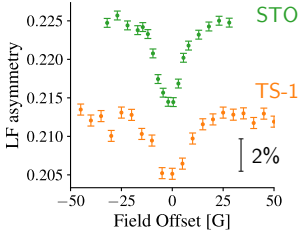
  → Complementarity to ALC
  3. Increase sensitivity !

  → Time integral
  On/Off contrast
  HIMB rates

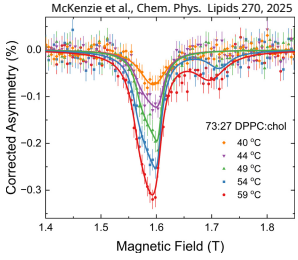




MQ-μSR



ALC



Acknowledgment

- Zaher Salman
- Thomas Hicken
- Thomas Prokscha

Swiss muon source (SμS)

PSI

Thank you for your attention!