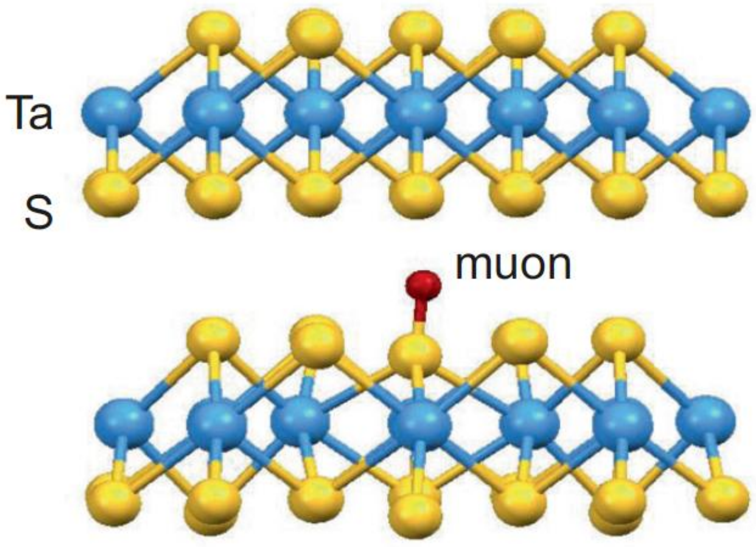
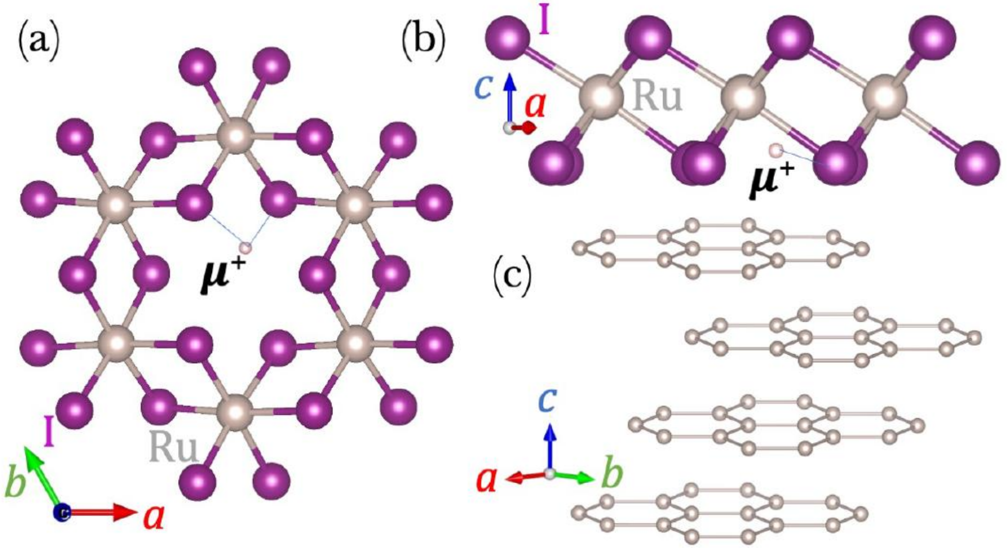
Quantum spin liquids in van der Waals materials



Ben Huddart
University of Oxford

Young Research Leaders Group Workshop 2025



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Danrui Ni

Robert Cava

Talk outline

1. Introduction to quantum spin liquids (QSLs)
2. The muon-spin spectroscopy (μSR) technique
3. μSR studies of candidate QSLs
   I. Triangular-lattice 1T-TaS_2
   II. Honeycomb-lattice α-RuI_3

Quantum spin liquids

- A quantum spin liquid (QSL) is a system of interacting localized electronic moments for which strong quantum fluctuations prevent their ordering even down to $T = 0$ K.

- Such systems are characterized by a high-degree of quantum entanglement and fractionalized excitations.

- Interactions in dimensions $d < 3$ are more effective at destabilizing magnetic order.

Geometrical frustration

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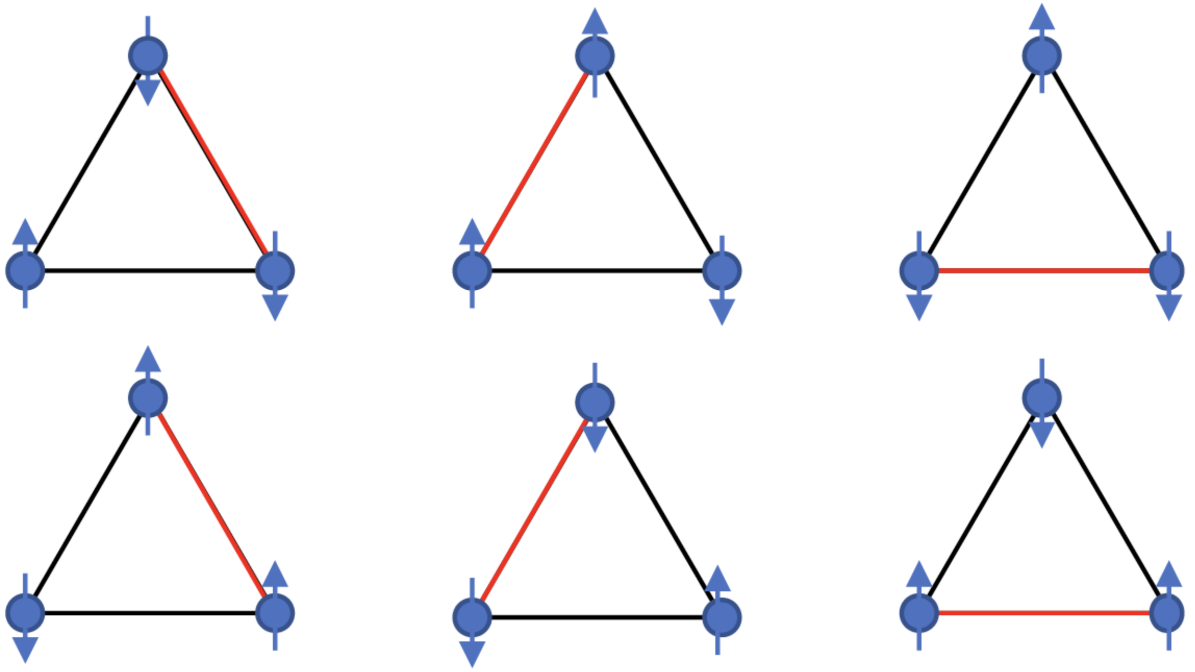
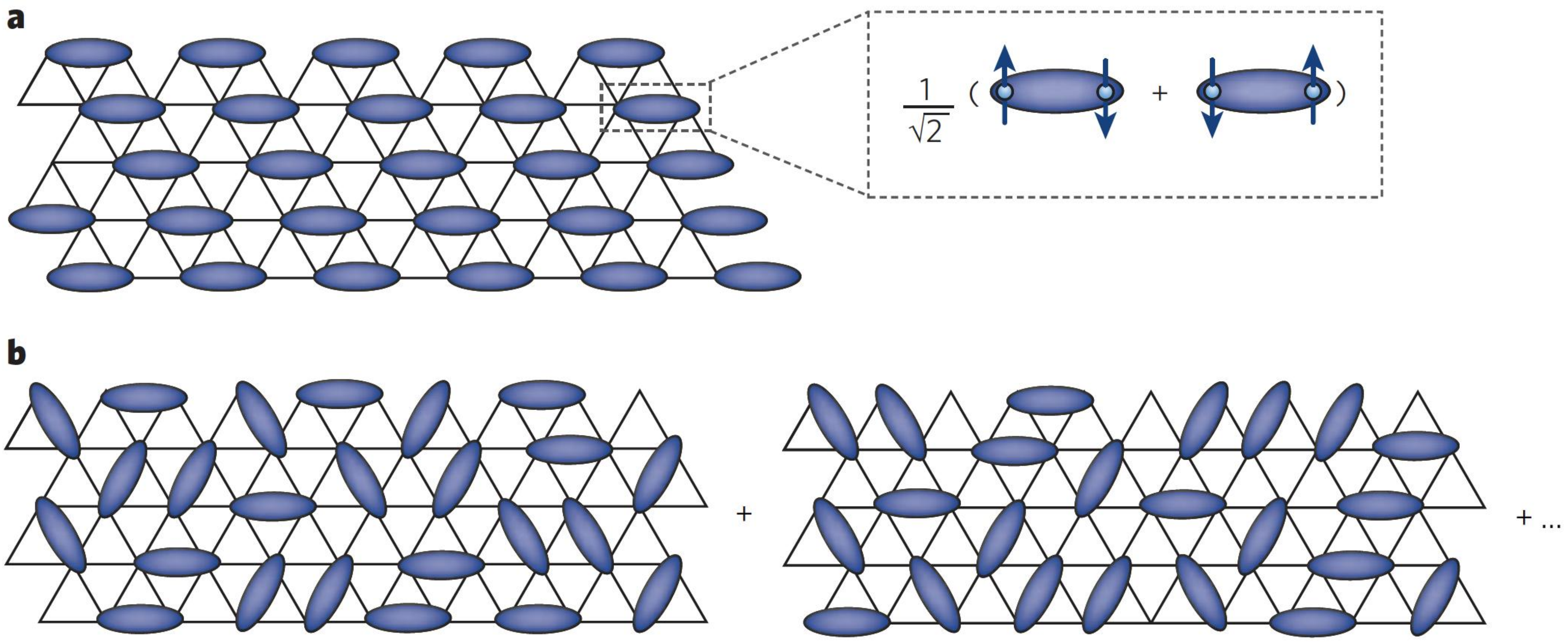
- Cannot simultaneously satisfy antiferromagnetic interactions between Ising spins on a triangle.
- Results in six degenerate ground states.



Resonating valence bonds

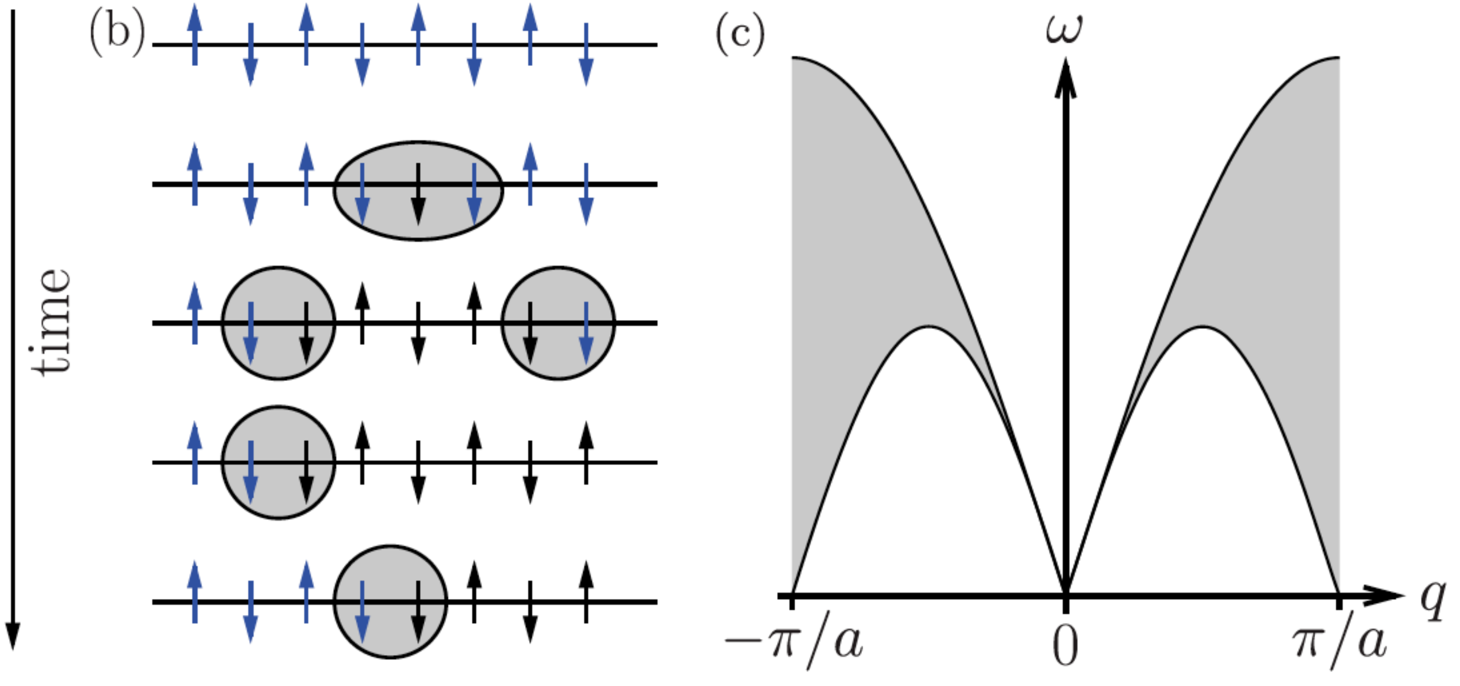


L. Balents, Nature **464**, 199 (2010)

Spinons

- Magnon (s = 1) not stable in 1D, but instead splits into two s = 1/2 spinons that can propagate by flips of pairs of spins

- Gives rise to a continuum of excitations

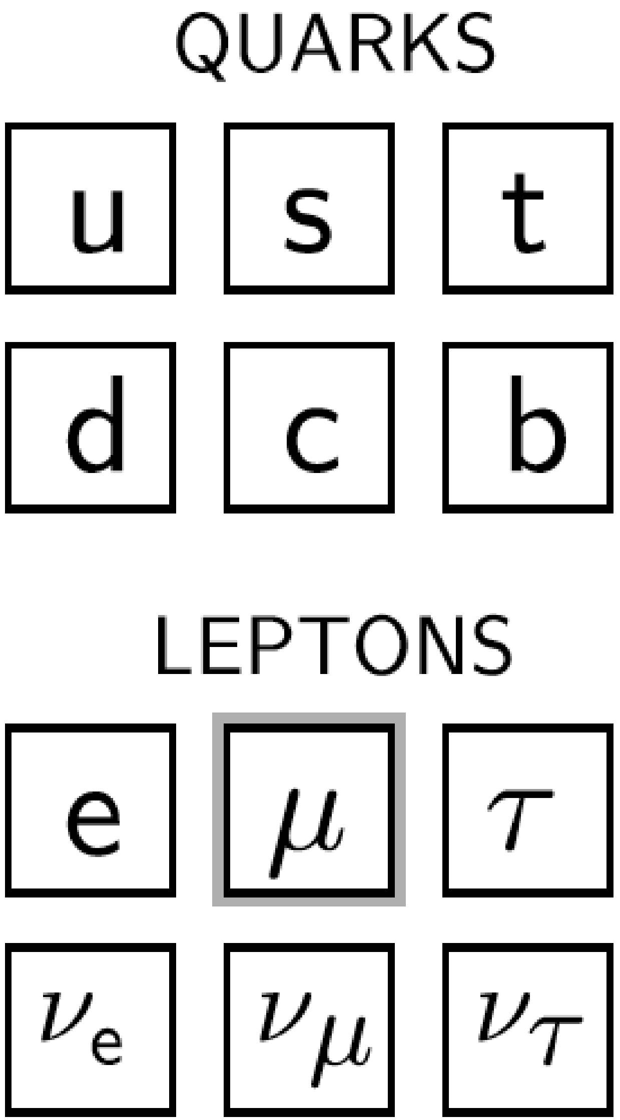


T. Lancaster, Contemp. Phys. **64**, 127 (2023)

Muon-spin spectroscopy

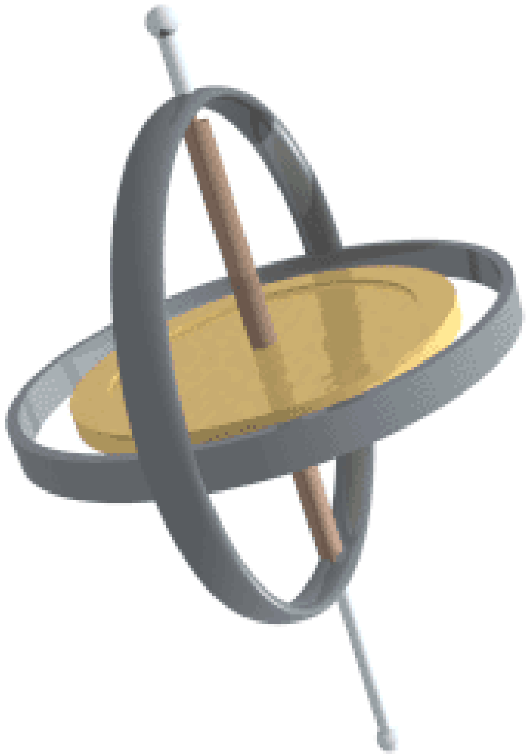
What is a muon?

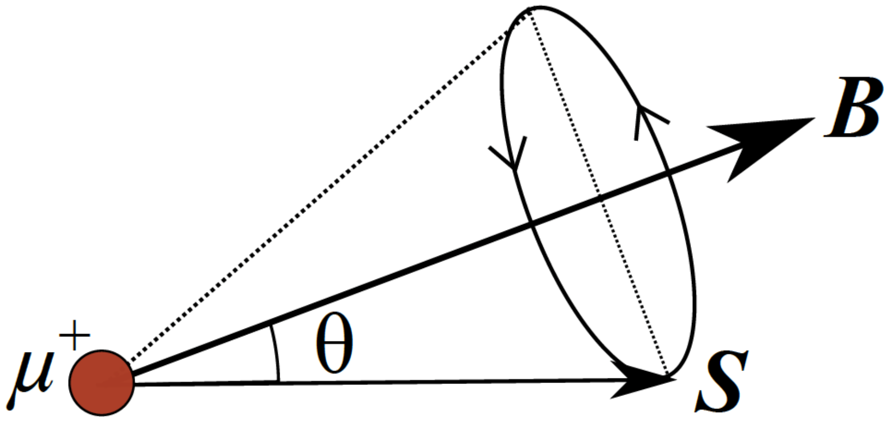
- First discovered as constituent of cosmic rays
- Spin-1/2 lepton
- Unstable, average lifetime τ = 2.2 μs
- In μ^+SR we use the positive muon (antimuon)



S. J. Blundell, *Contemporary Physics*, 40, 175 (1999)

Spin precession





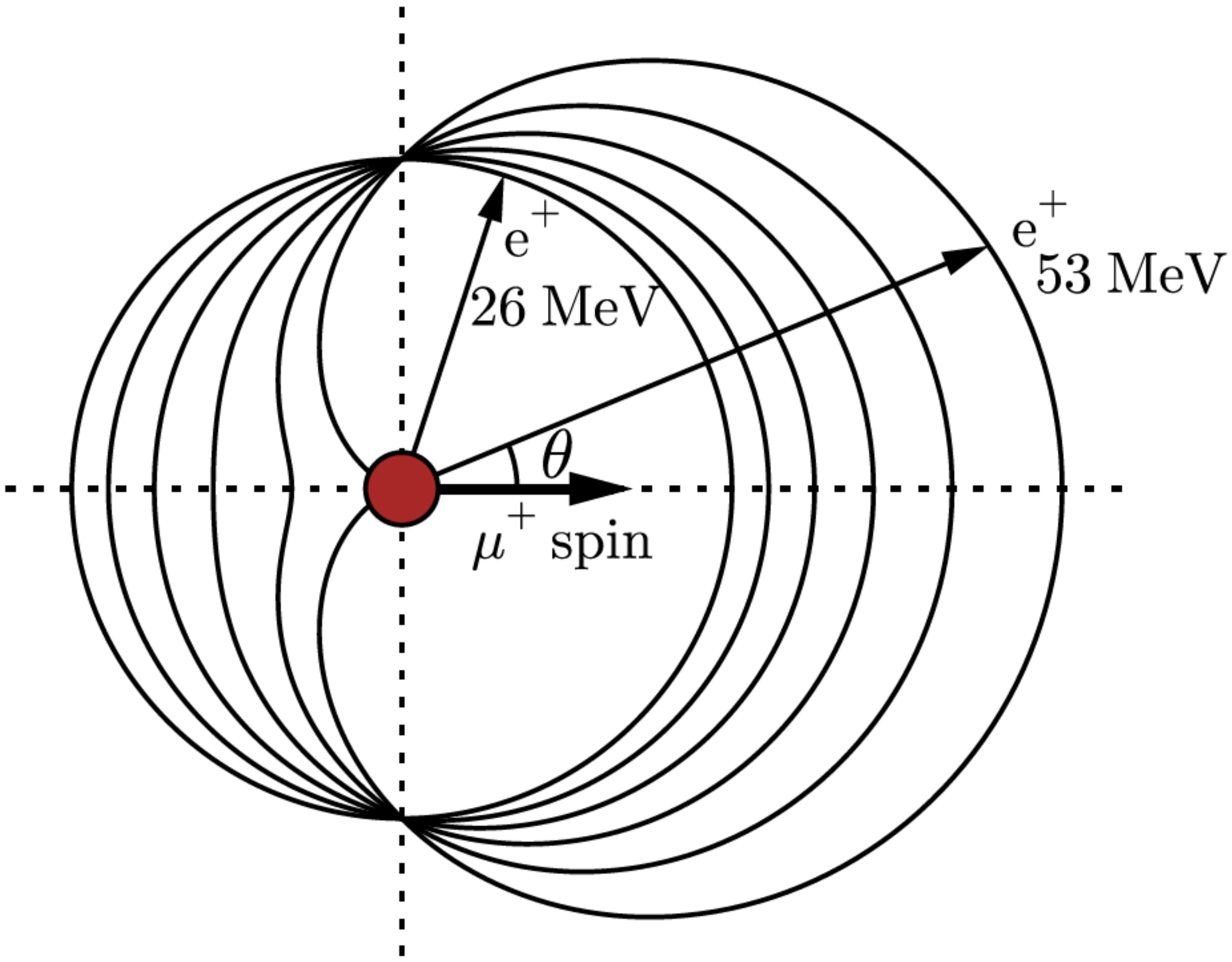
$$\frac{d\langle \boldsymbol{S} \rangle}{dt} = \gamma_\mu \langle \boldsymbol{S} \rangle \times \boldsymbol{B}$$

$$P(t) = \cos^2\theta + \sin^2\theta \cos(\gamma_\mu B t)$$

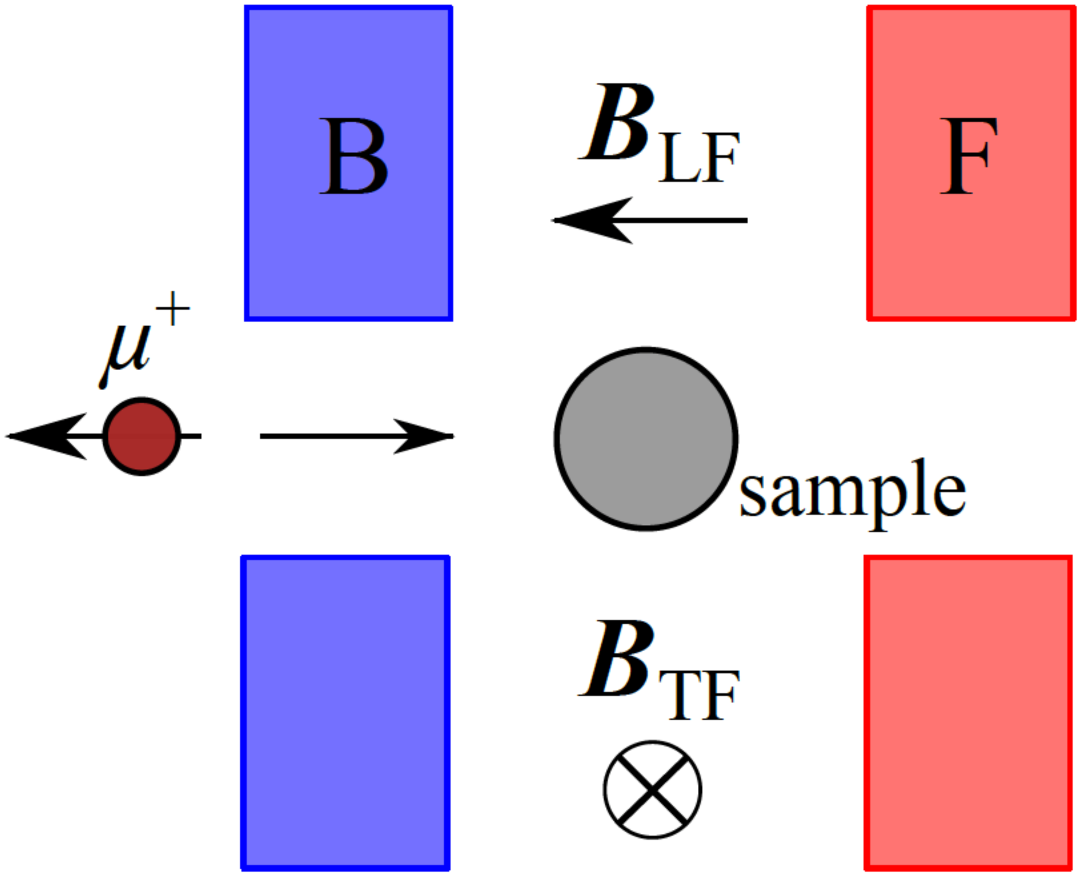
μSR is not a scattering technique

$$\mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu$$

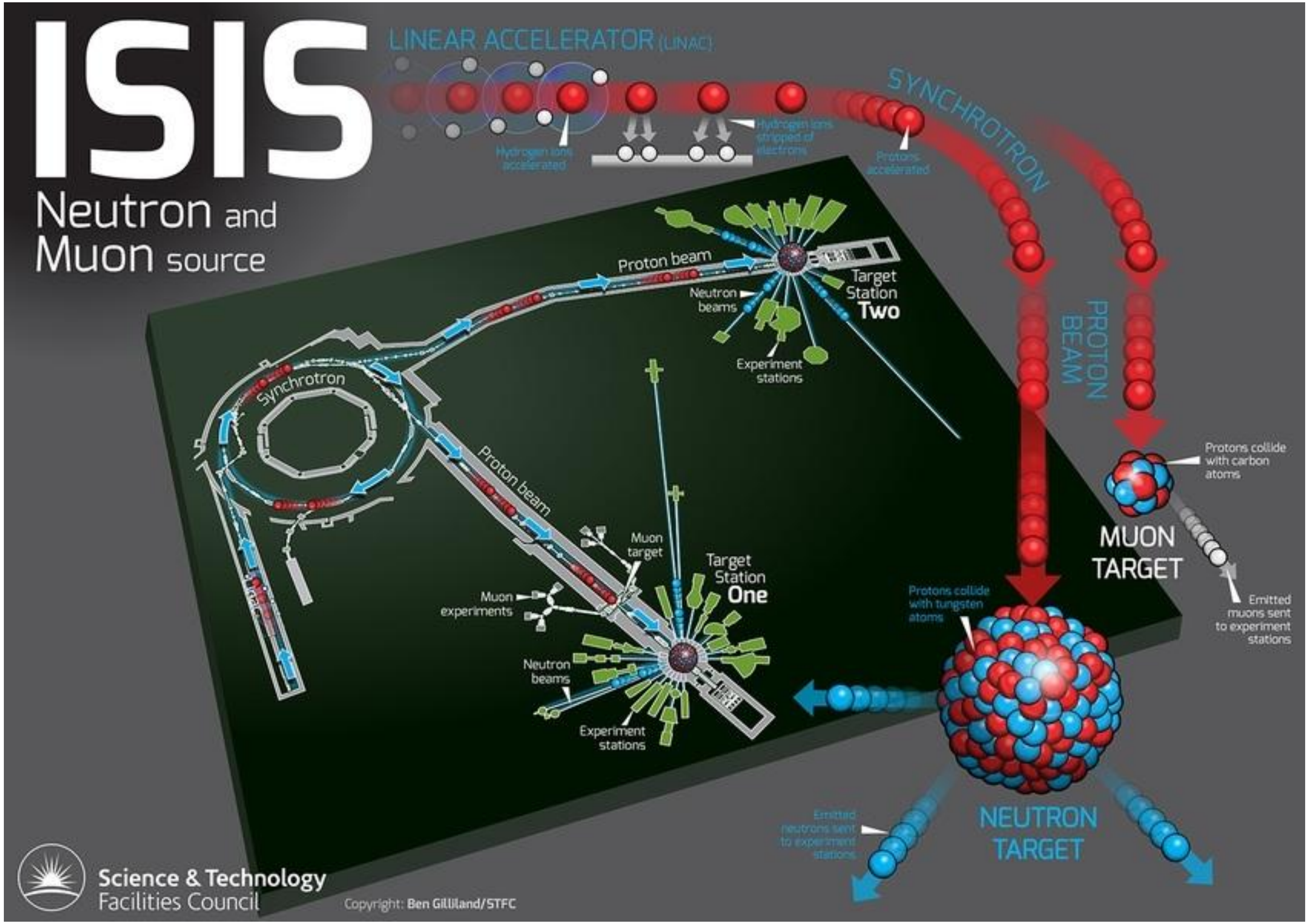
τ = 2.2 μs



Experimental setup



$$A(t) = \frac{N_{\mathrm{B}}(t) - N_{\mathrm{F}}(t)}{N_{\mathrm{B}}(t) + N_{\mathrm{F}}(t)}$$


ISIS
Neutron and Muon source
LINEAR ACCELERATOR (LINAC)
SYNCHROTRON
Hydrogen ions accelerated
Hydrogen ions stripped of electrons
Protons accelerated
PROTON BEAM
Proton beam
Target Station Two
Neutron beams
Experiment stations
Synchrotron
Proton beam
Muon target
Target Station One
Muon experiments
Neutron beams
Experiment stations
Protons collide with carbon atoms
MUON TARGET
Emitted muons sent to experiment stations
Protons collide with tungsten atoms
Emitted neutrons sent to experiment stations
NEUTRON TARGET
Science & Technology Facilities Council
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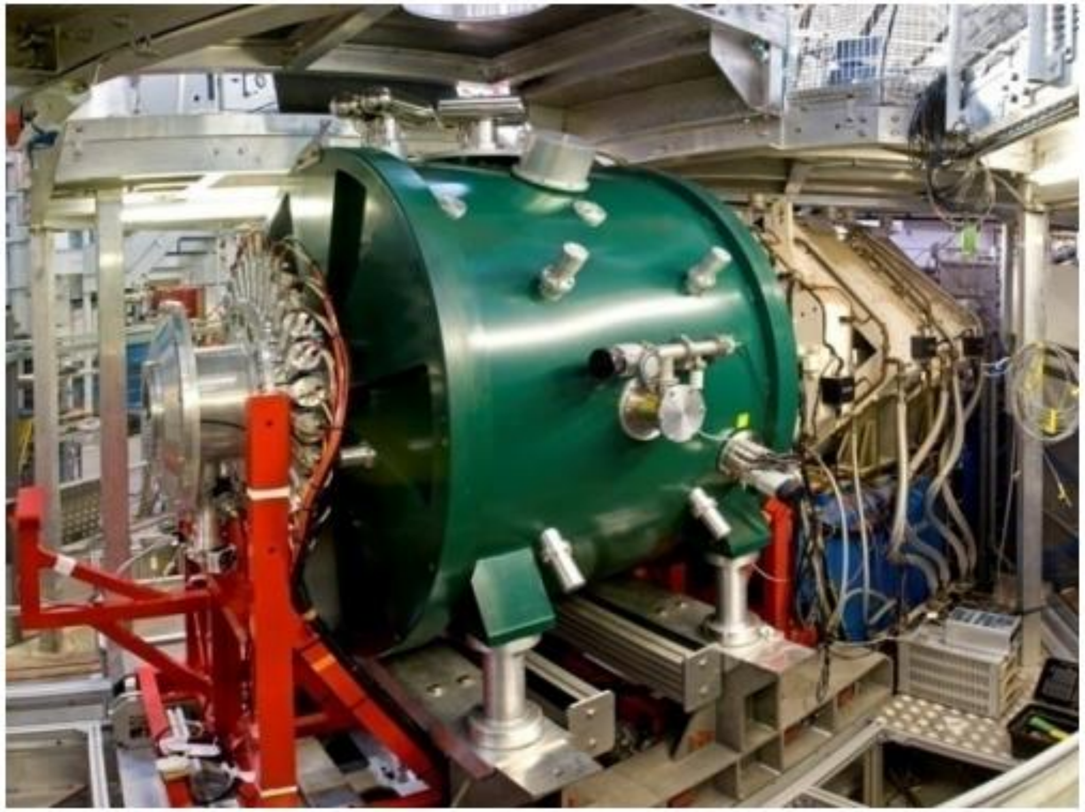
$$\pi^+ \to \mu^+ + \nu_\mu$$

μSR spectrometers

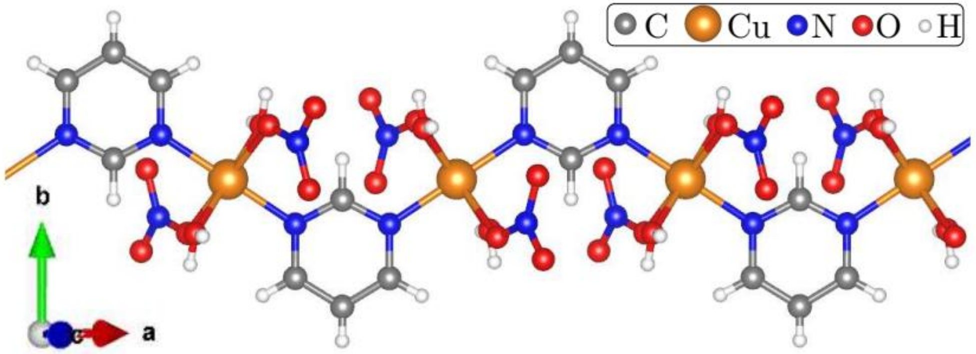


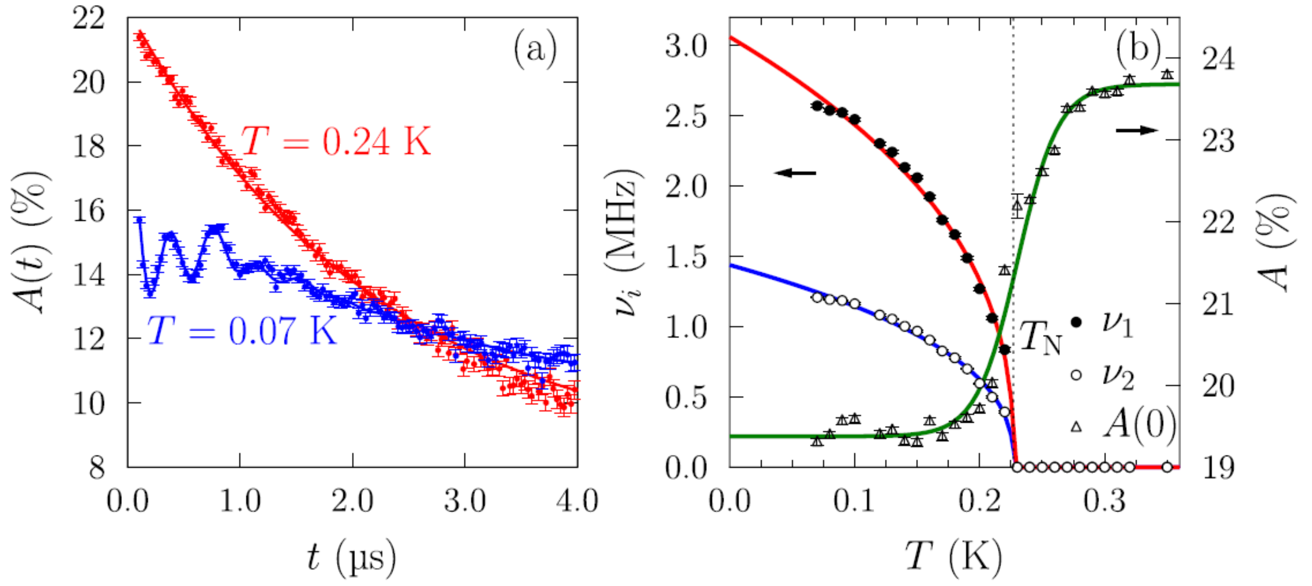
HiFi (ISIS)

GPS (PSI)

Magnetism

- Muons precess at a frequency $\omega = \gamma_\mu B$
- Fields as small as ≈ 0.04 mT are measurable
- Muons are a *local* probe

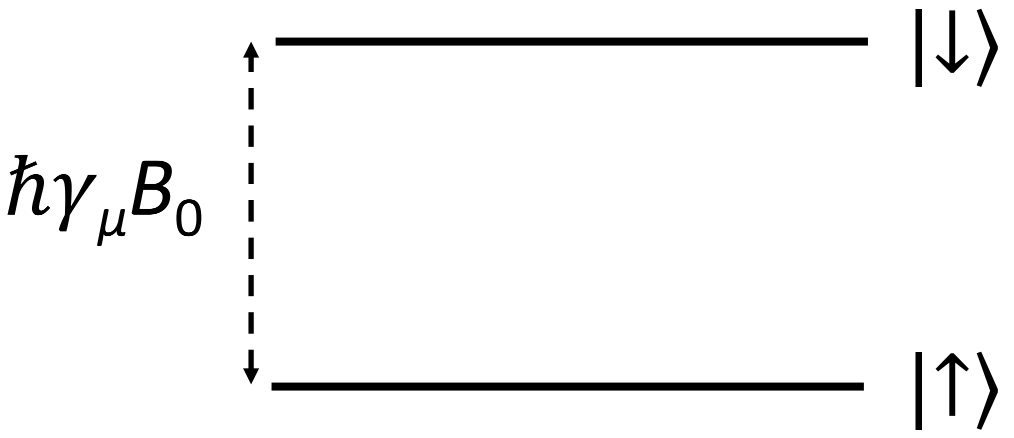




B. M. Huddart *et al.*, Phys. Rev. B **103**, L060405 (2021)

Magnetisation dynamics

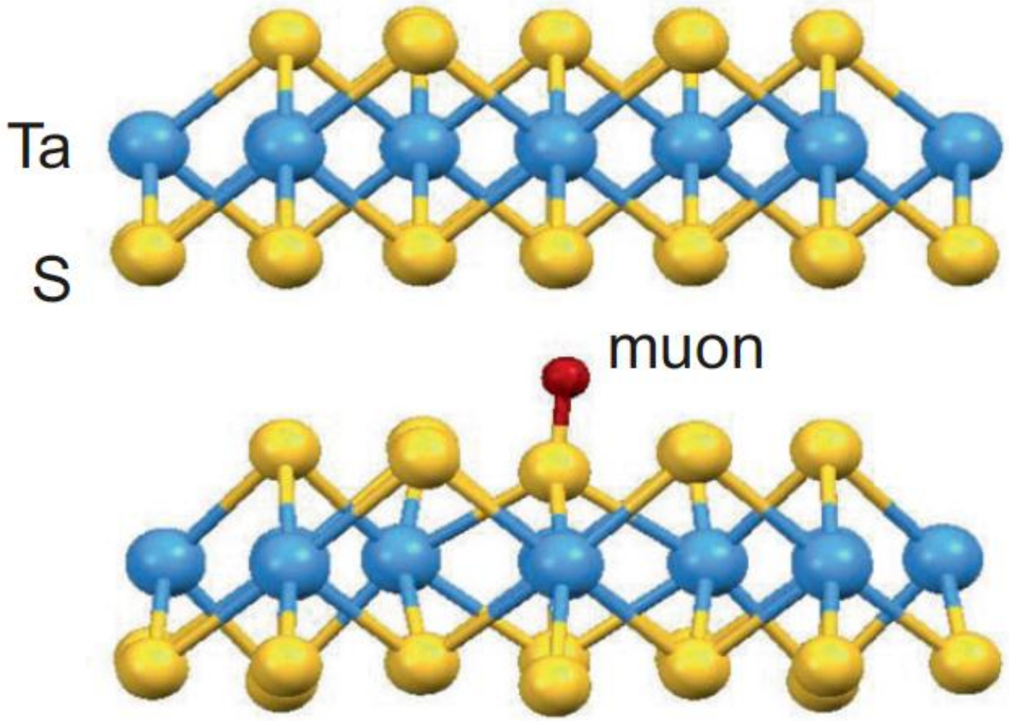
- A magnetic field applied parallel to the muon spin creates an energy gap between the spin states
- Fluctuating magnetic fields can cause transitions between up and down states
- Leads to a relaxation of the polarisation $P_z(t) \propto (n_\uparrow - n_\downarrow)$



$$P_z(t) = P_z(0)e^{-\lambda t}$$

1T-TaS_2

A candidate triangular-lattice QSL

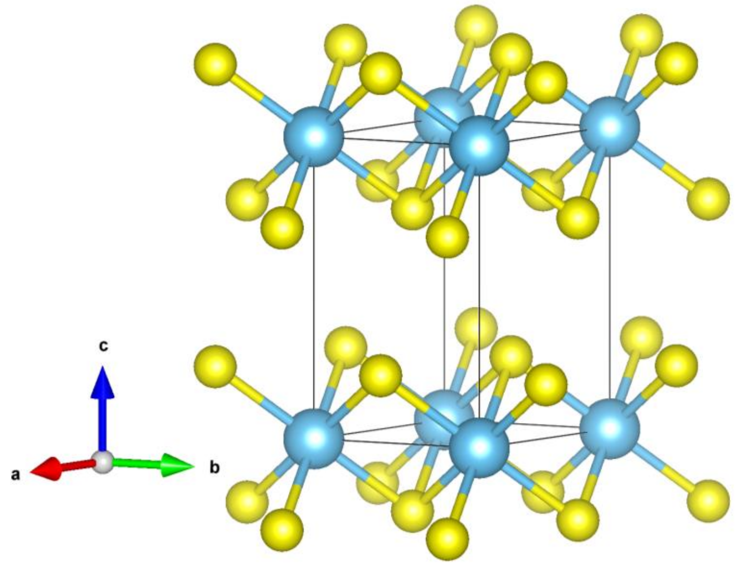


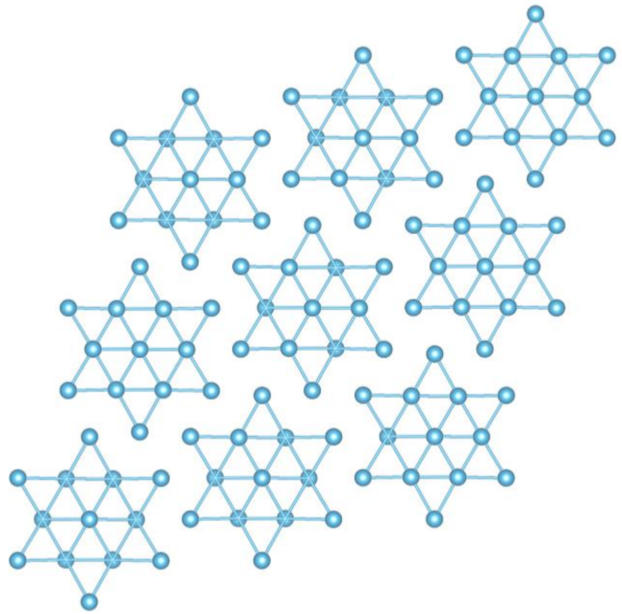
S. Mañas-Valero *et al.*, npj Quantum Mater. 6, 69 (2021)

1T-TaS_2

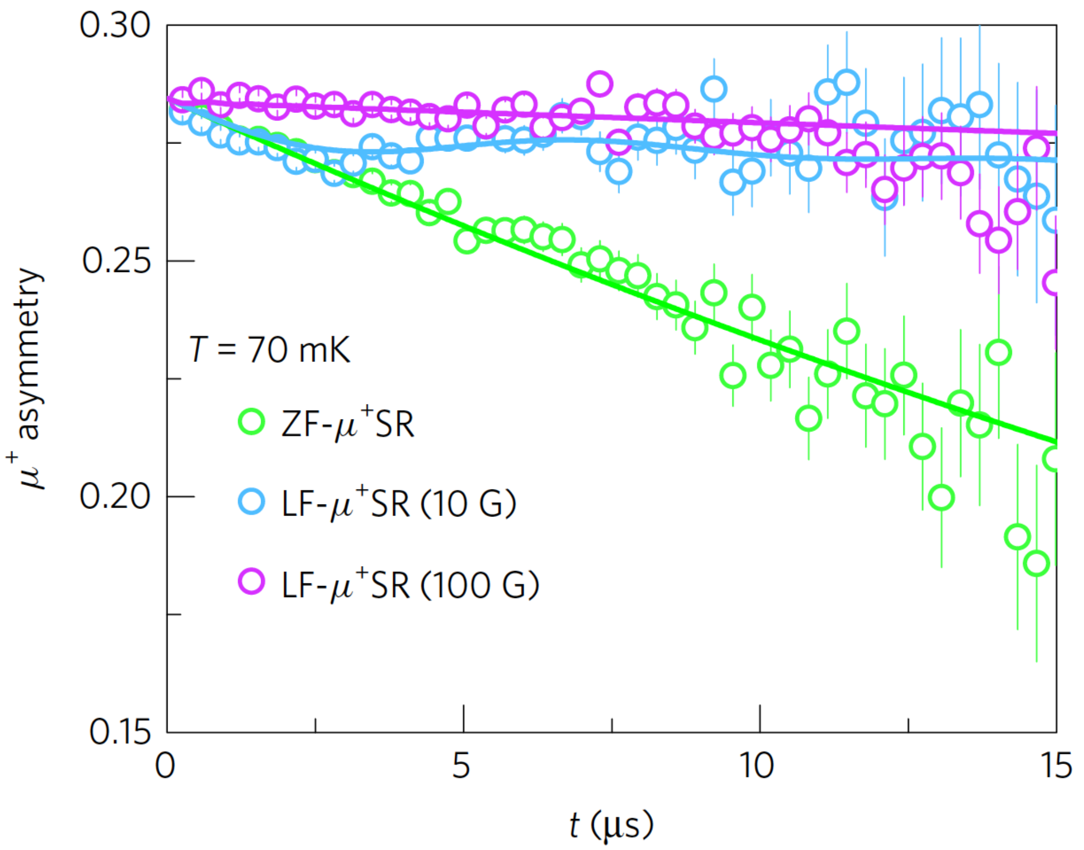
- Layered compound that undergoes a series of charge-density-wave (CDW) instabilities.

- For commensurate CDW (C-CDW) phase below 200 K, Ta atoms form a triangular lattice of Star-of-David clusters, with one unpaired spin-1/2 per cluster.





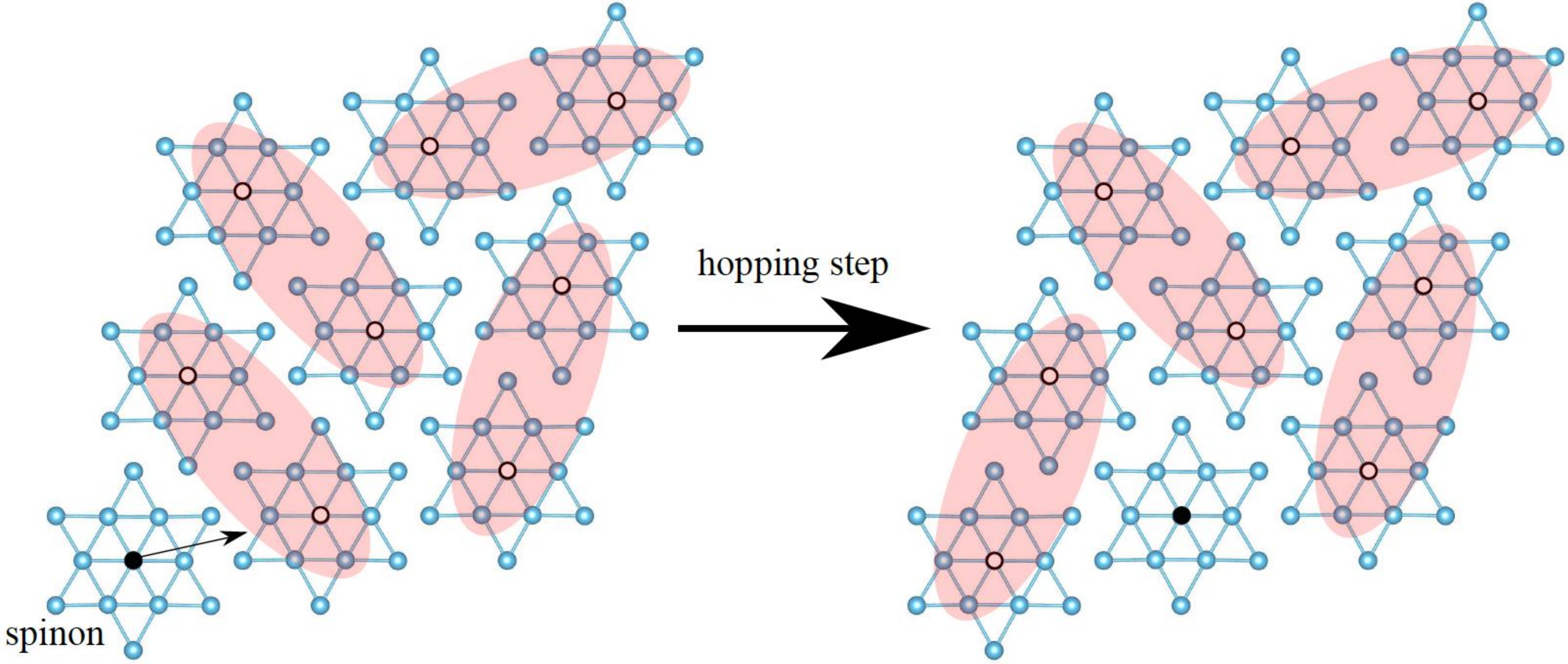
Absence of magnetic order



M. Klanjšek *et al.* Nat. Phys. **13**, 1130 (2017)

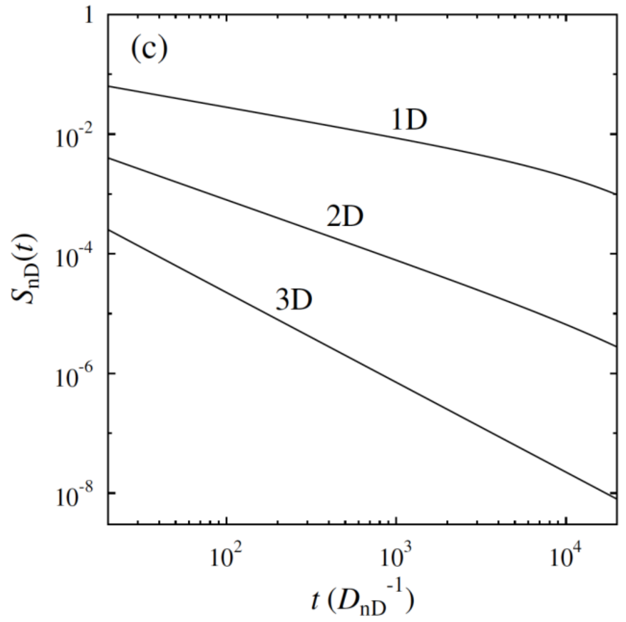
Here we instead focus on the **dynamics**

Spinon diffusion

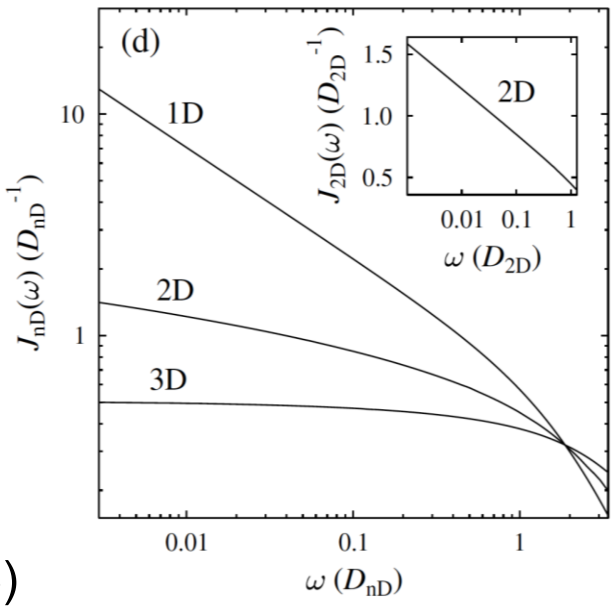


Studying spin diffusion with LF μSR

- The spin autocorrelation function for diffusion in *n* dimensions has the form
  $$S_{\mathrm{nD}}(t) = [\exp(-2D_{\mathrm{nD}}t)I_0(2D_{\mathrm{nD}}t)]^n,$$
  where I_0 is a zeroth order Bessel function.



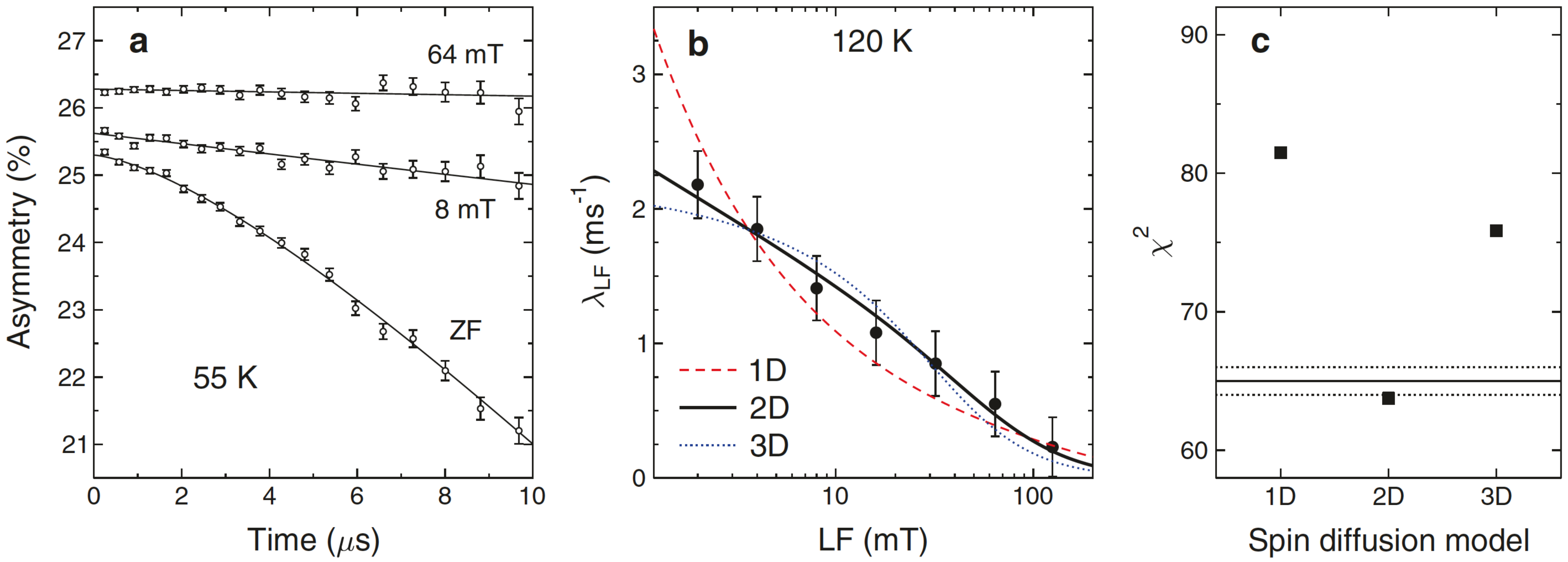
- The relaxation rate $\lambda(B) \propto J_{\mathrm{nD}}(\omega)$
  - spectral density $J_{\mathrm{nD}}(\omega) = \mathrm{FT}[S_{\mathrm{nD}}(t)]$
  - probe frequency $\omega = \gamma_e B$



F L Pratt *et al*. J. Phys. Conf. Ser. **2462**, 012038 (2023)

Spinon diffusion in 1T-TaS_2

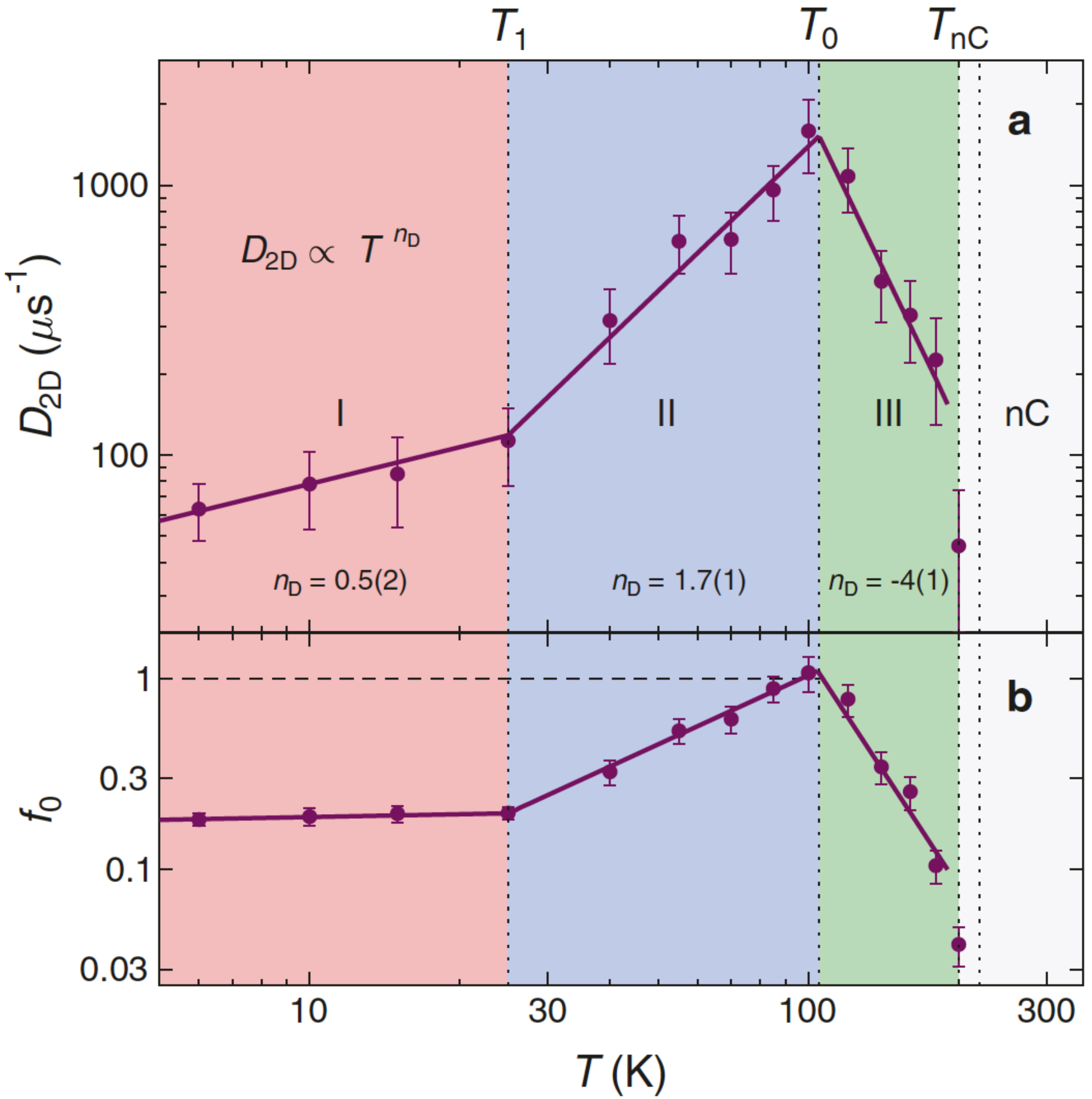
$$A(t) = A_0 \exp(-\lambda_{\mathrm{LF}} t)$$



S. Mañas-Valero *et al.*, npj Quantum Mater. 6, 69 (2021)

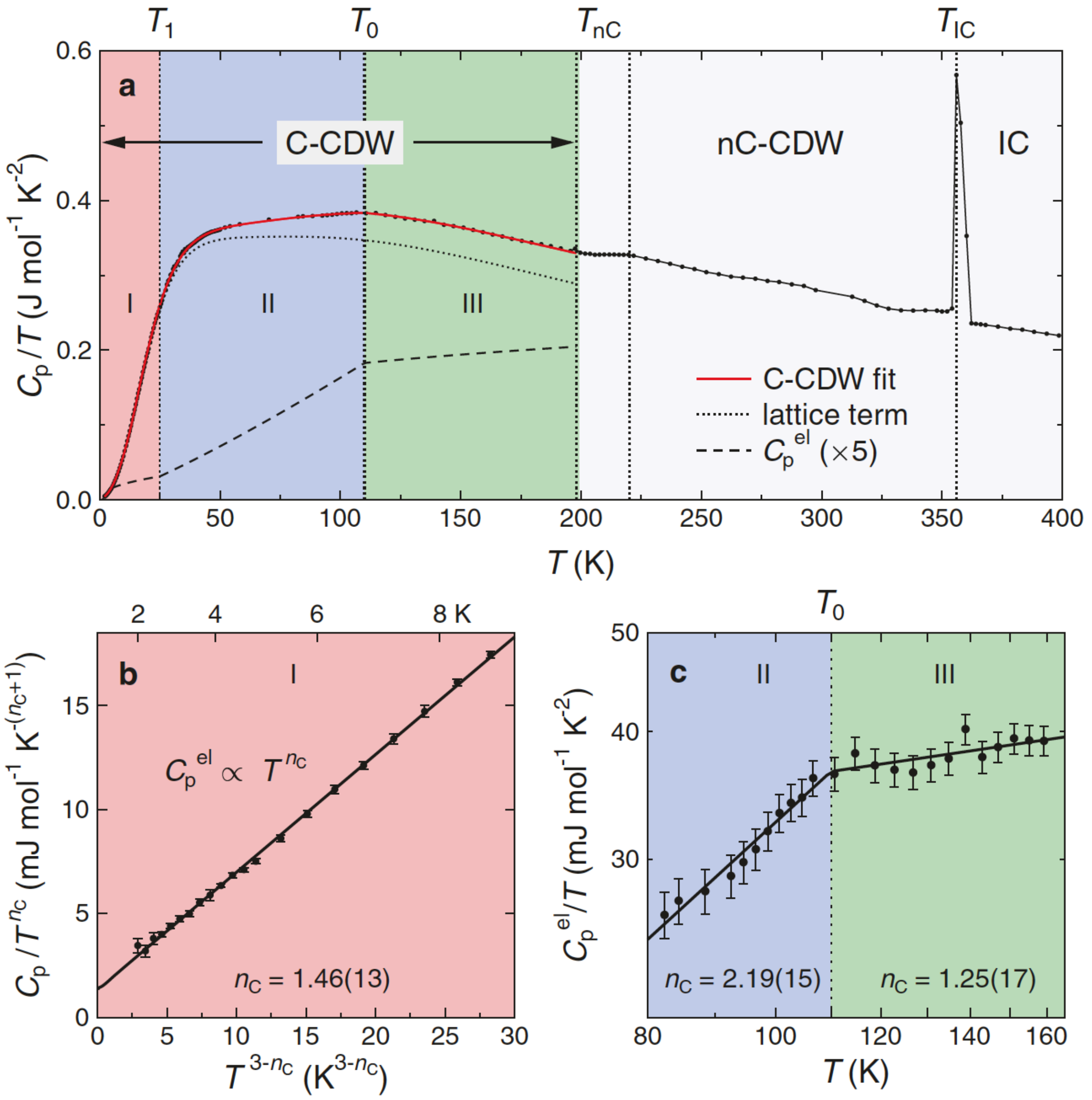
Diffusion rate

- Obtain $D_{2\mathrm{D}}$ from fits to $\lambda(B)$ at each temperature.
- $D_{2\mathrm{D}} \propto T^{n_\mathrm{D}}$
- Find three distinct regions in temperature.
- n_D can be related to predictions of QSL models.



S. Mañas-Valero *et al.*, npj Quantum Mater. 6, 69 (2021)

Heat capacity



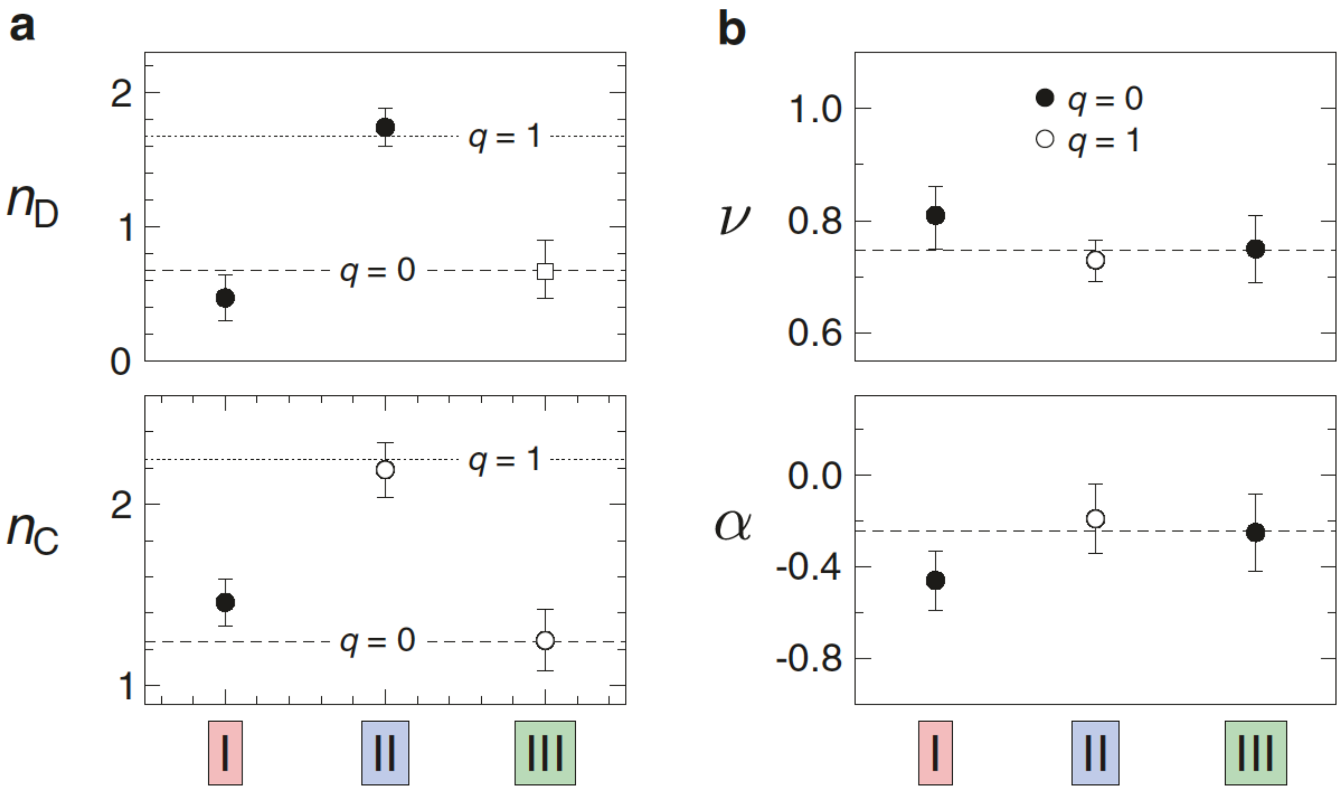
- In region I phonon contribution given by a simple Debye T^3 term.
- In regions II and III require anisotropic Debye term for acoustic phonons and two Einstein terms for optical phonons.
- Electronic contribution to the heat capacity $C_p^{el} \propto T^{n_C}$

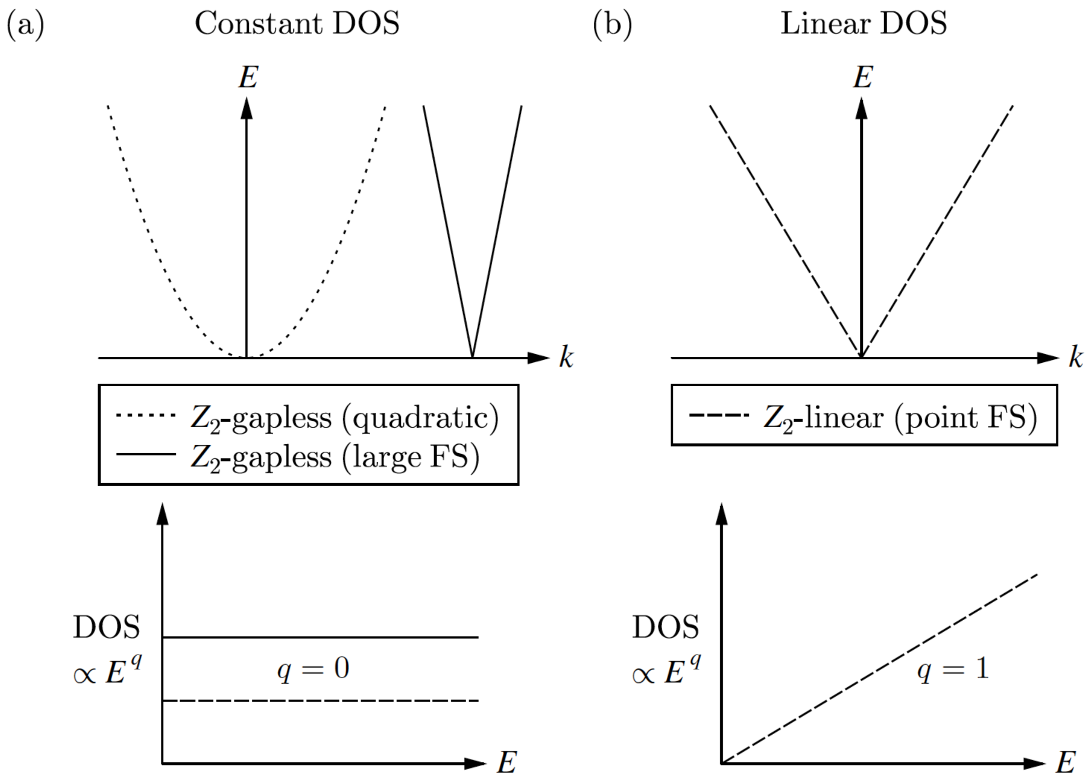
S. Mañas-Valero *et al.*, npj Quantum Mater. 6, 69 (2021)

Critical parameters

$$n_{\mathrm{D}} = \frac{2}{\nu} - 2 + q$$

$$n_{\mathrm{C}} = 1 - \alpha + q$$

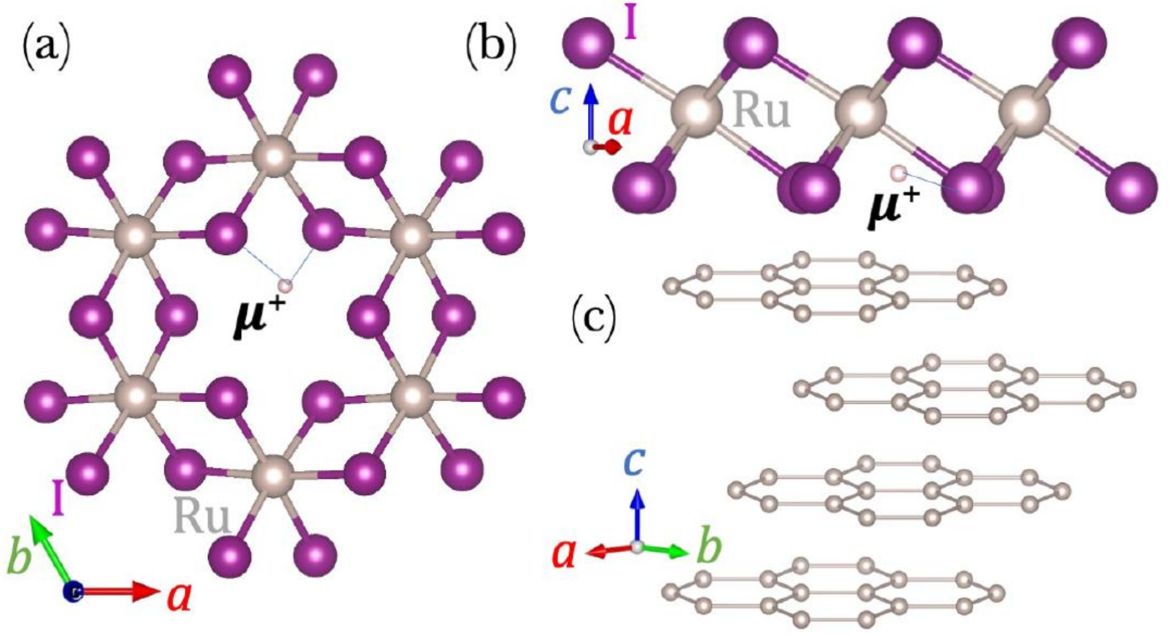




S. Mañas-Valero *et al.*, npj Quantum Mater. 6, 69 (2021)

α-RuI_3

A candidate Kitaev QSL



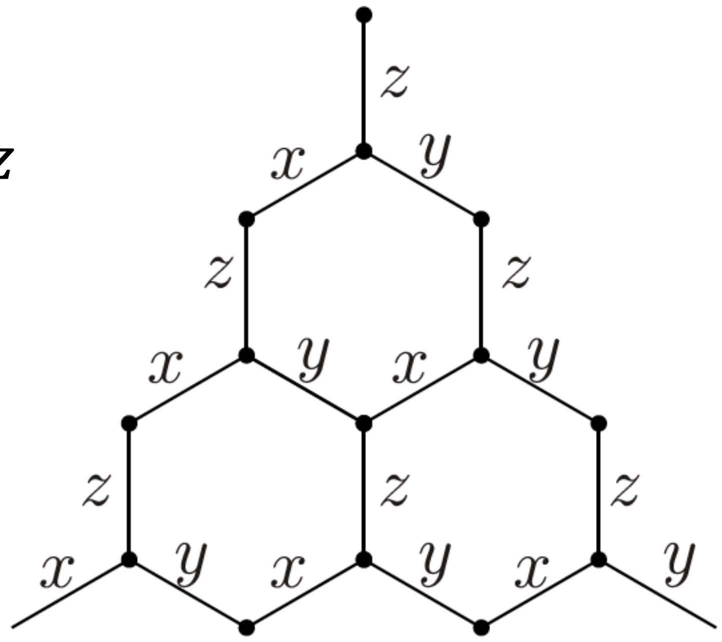
H. C. H. Wu *et al.* arXiv:2406.04065 [cond-mat.str-el]

The Kitaev model

- The Kitaev model is an exactly solvable model with a QSL ground state.

$$\hat{H} = -J_x \sum_x \hat{\sigma}_i^x \hat{\sigma}_j^x - J_y \sum_y \hat{\sigma}_i^y \hat{\sigma}_j^y - J_z \sum_z \hat{\sigma}_i^z \hat{\sigma}_j^z$$

- This is an example of exchange frustration, rather than geometrical frustration.

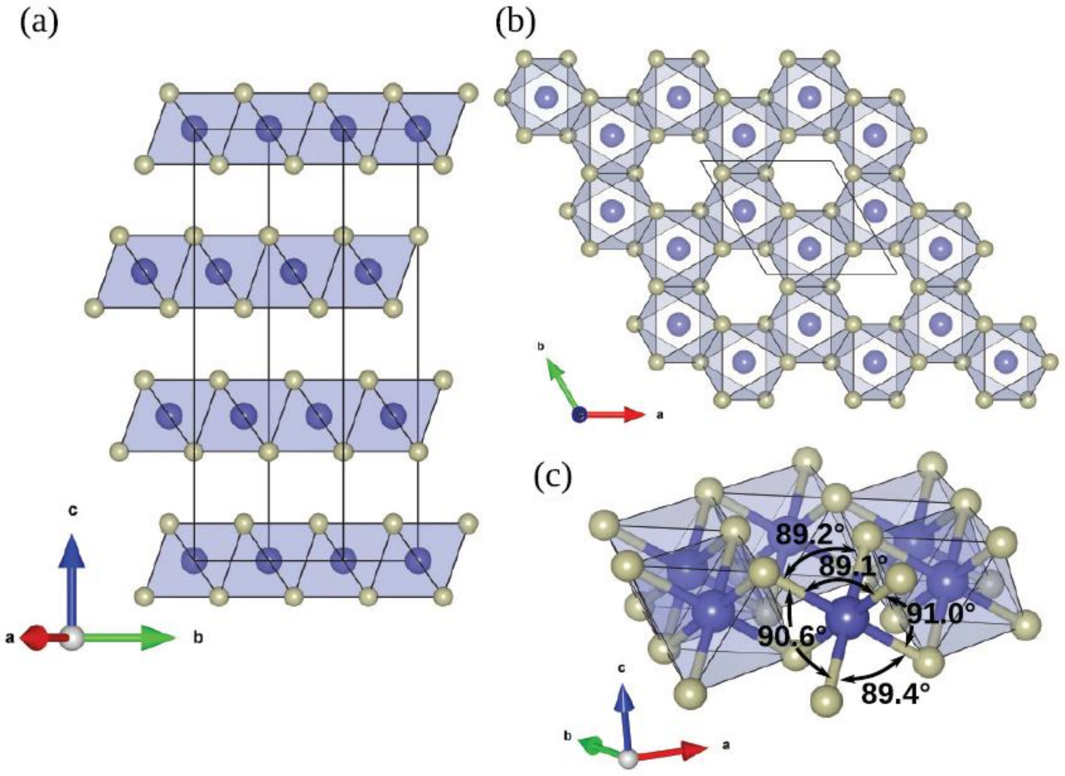


A. Kitaev, Ann. Phys. (N. Y.) **321**, 127 (2006)
T. Lancaster, Contemp. Phys. **64**, 127 (2023)

Candidate Kitaev QSLs

- Materials with *S*=1/2 spins on a honeycomb lattice and significant spin-orbit-coupling are good candidates for hosting Kitaev QSLs

- In α-$RuCl_3$, indirect hopping through Cl gives rise to a Kitaev interaction.

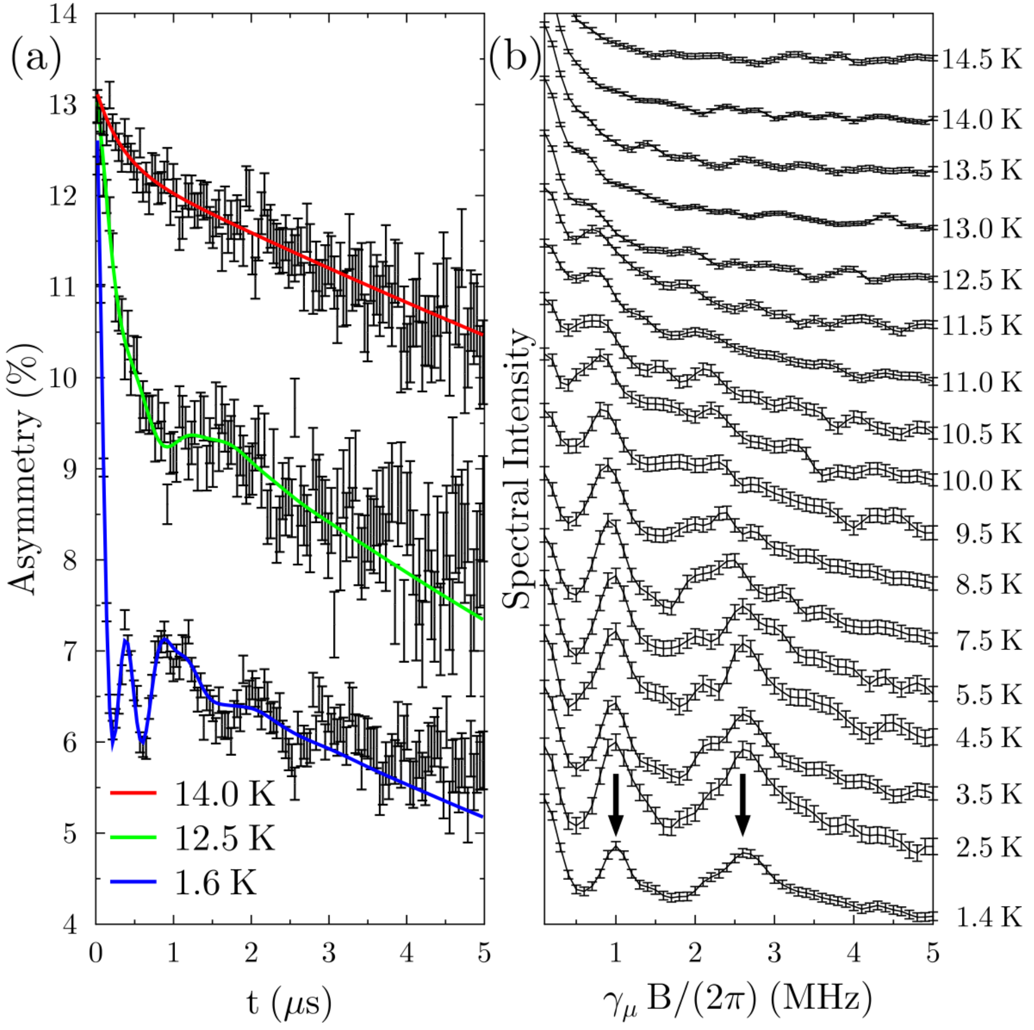


K. W. Plumb et al. Phys. Rev. B **90**, 041112(R) (2014)

Candidate Kitaev QSLs: α-$RuCl_3$

- α-$RuCl_3$ orders magnetically into a zigzag antiferromagnetic state at around 7 to 14 K in zero-field.

- The ordering temperature is sample-dependent and possibly depends on stacking faults.

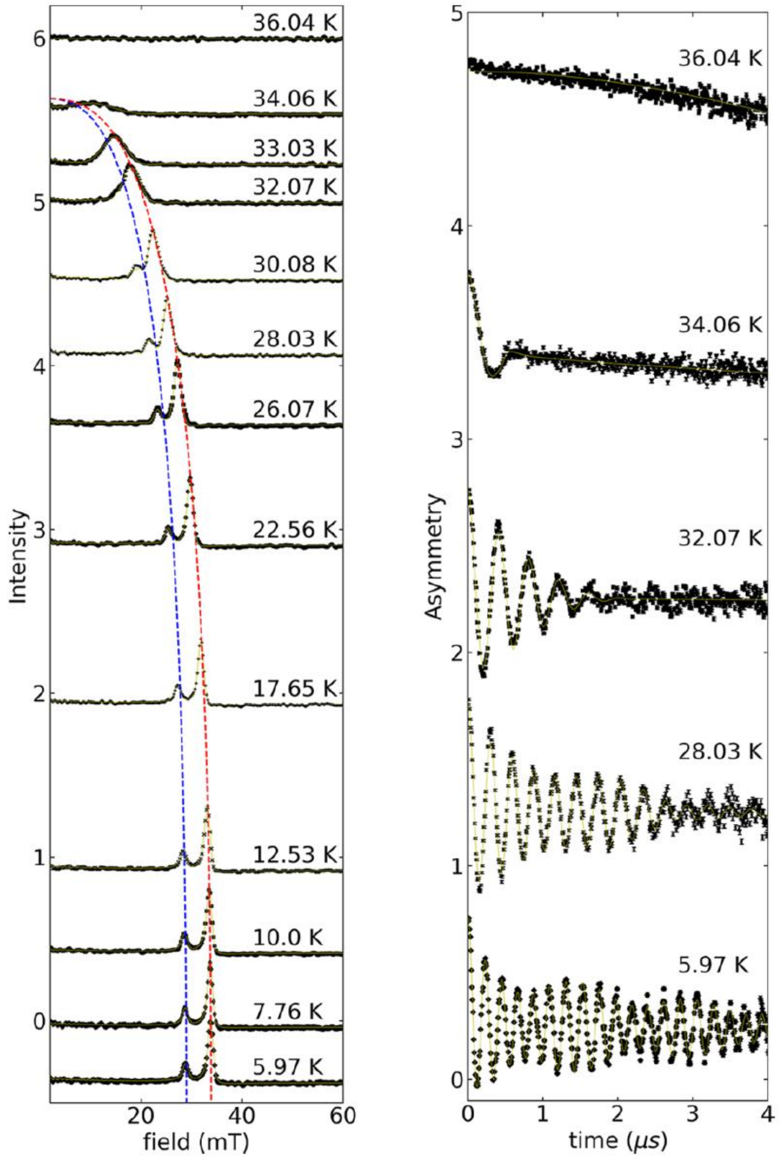


F. Lang *et al.* Phys. Rev. B **94**, 020407(R) (2016).

Candidate Kitaev QSLs: α-$RuBr_3$

- α-$RuBr_3$ was found also to possess a zigzag antiferromagnetic ground state with a higher transition temperature at 34 K.

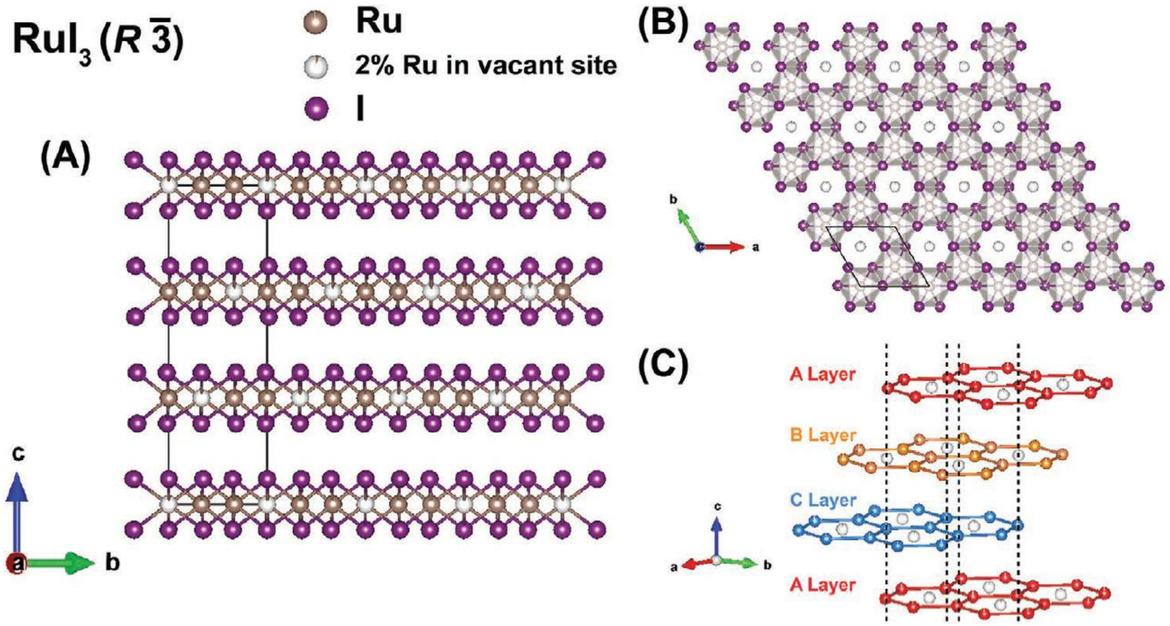
- The magnetic ordering in both compounds has been attributed to non-Kitaev interactions in the Hamiltonian.



T. Weinhold *et al.*, Phys. Rev. B **109**, 014440 (2024)

α-RuI_3

- The isostructural iodide α-RuI_3 has only recently been synthesized and has shown promising potential for being a candidate QSL.
- The material is synthesised at moderately elevated pressures and is stable under ambient conditions.
- No long-range magnetic order down to 0.35 K.



D.Ni et al., Adv. Mater. **34**, 2106831 (2022)

α-RuI_3: absence of magnetic order

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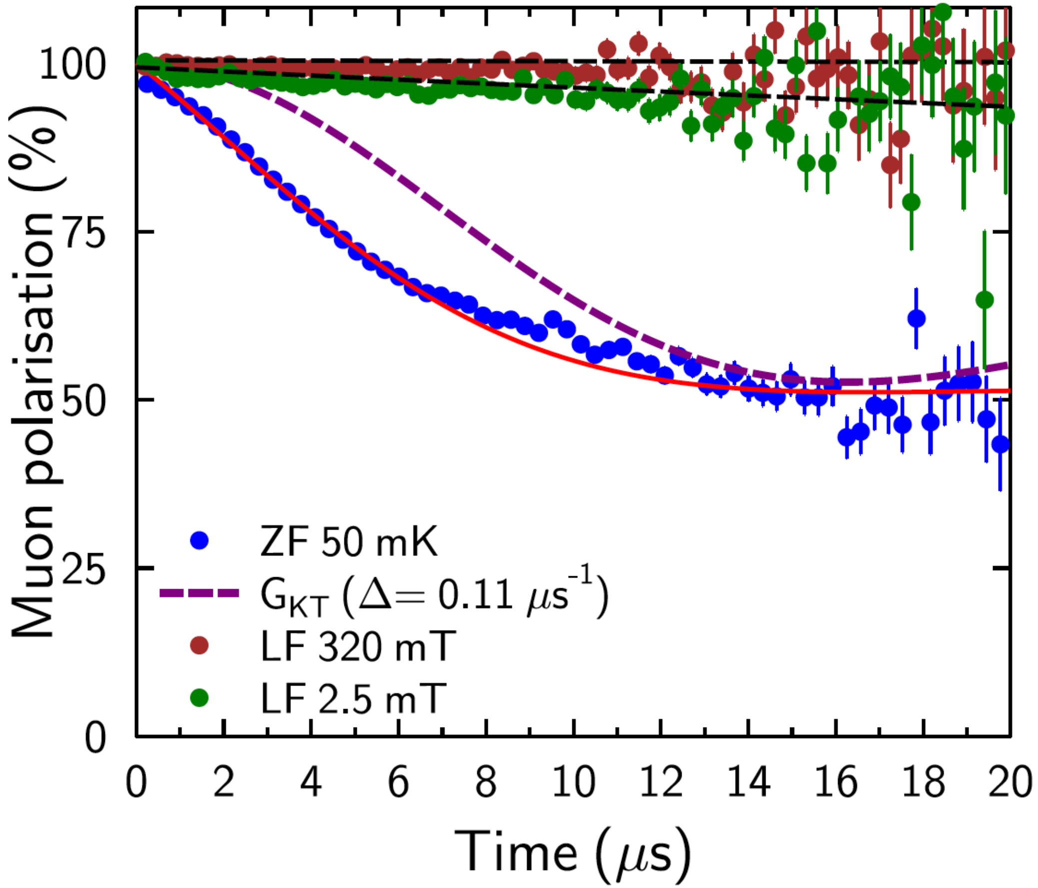
- No oscillations in the muon polarization down to 50 mK

  → Suggests no LRO.

- Nuclear broadening at muon site insufficient to explain this relaxation, must be additional relaxation processes occurring.

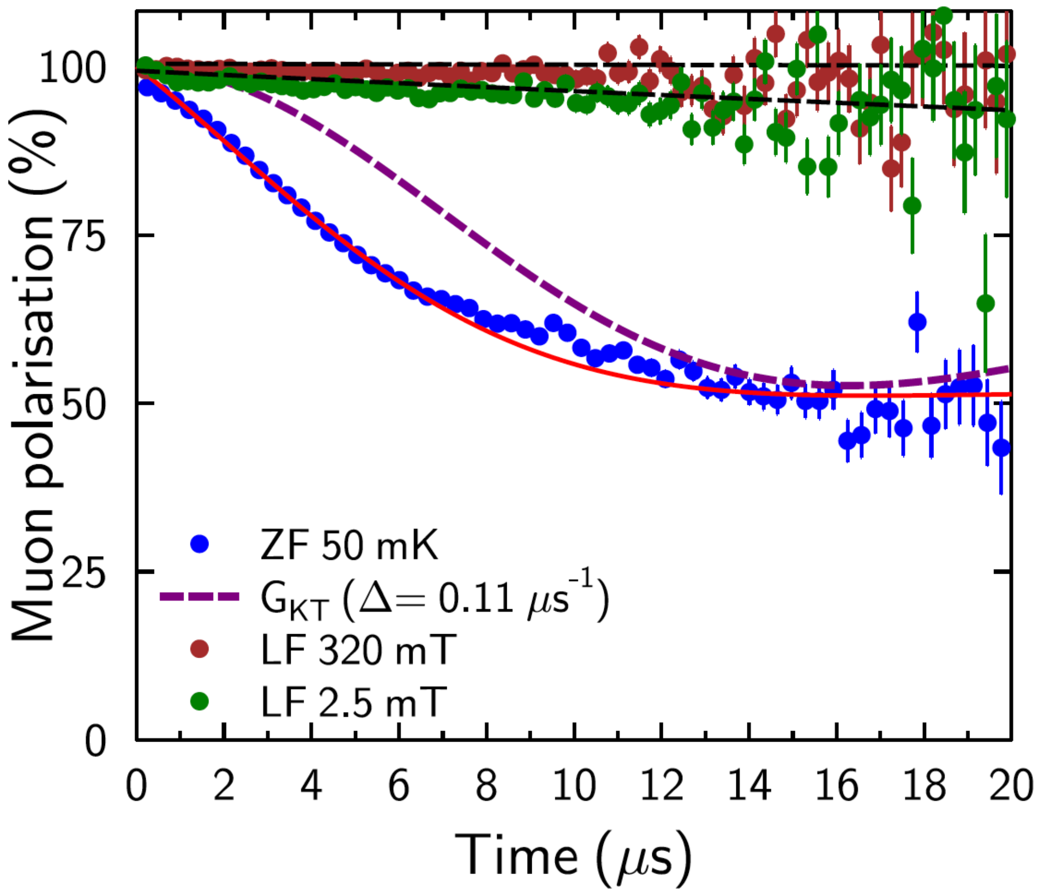


H. C. H. Wu *et al.* arXiv:2406.04065 [cond-mat.str-el]

α-RuI_3: longitudinal field μSR

- Applying LF quenches contribution of the nuclear moments, but some exponential relaxation remains, which is due to the fluctuating Ru^{3+} moments.

- The dynamical relaxation weakens with increasing longitudinal field.

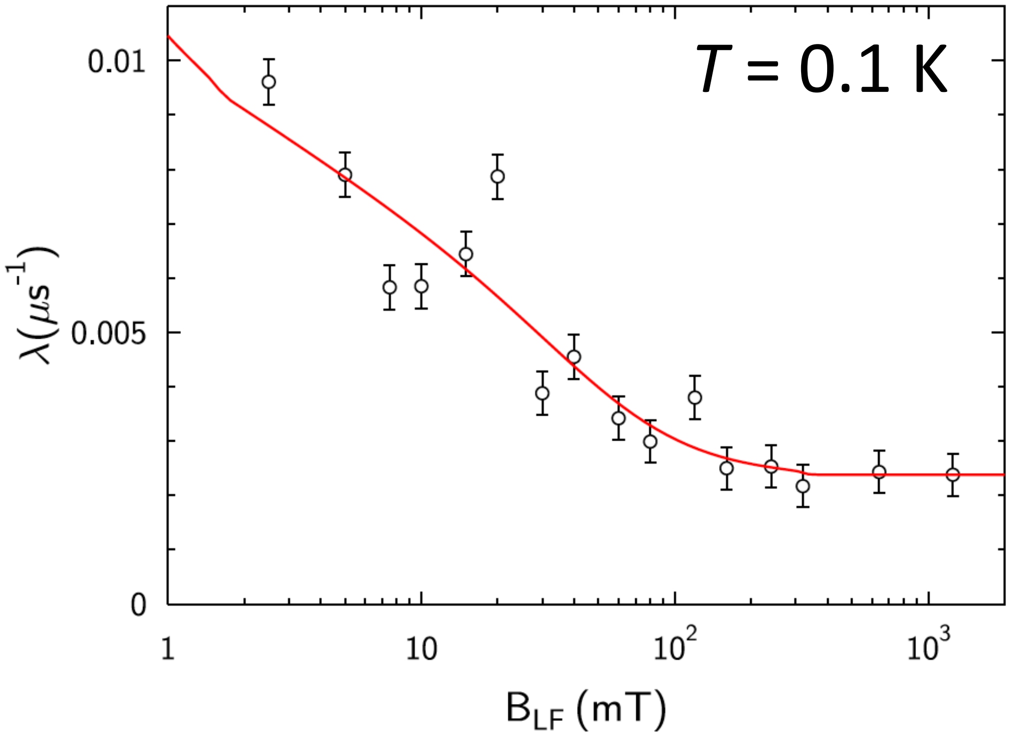


H. C. H. Wu *et al.* arXiv:2406.04065 [cond-mat.str-el]

α-RuI_3: field-dependent relaxation rate

$$\lambda(B_{\mathrm{LF}}) = \lambda_{2\mathrm{D}}(B_{\mathrm{LF}}) + \lambda_0$$

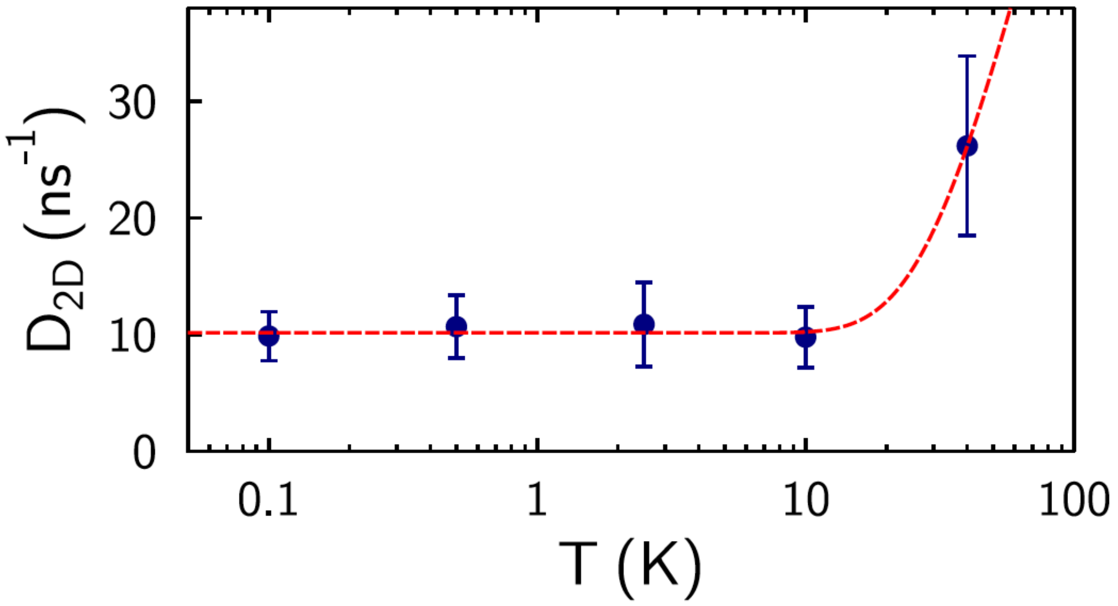
- Field-dependent term due to excitations diffusing in 2D
- 0D field-independent term due to localized excitations
- $\lambda_{2\mathrm{D}}$ obtained from the Fourier transform of the autocorrelation function for 2D spin diffusion



H. C. H. Wu *et al.* arXiv:2406.04065 [cond-mat.str-el]

α-RuI_3: spinon diffusion

- Diffusion rate approximately constant up to 10 K, but then rises rapidly at 40 K.
- Diffusion coefficient $D = \frac{1}{2} v l = D_{2D} l^2$ so that $D_{2D} = \frac{v}{2l}$



- Rising diffusion rate suggests falling mean free path l.

- High-T: Ru^{3+} moments correlated only to nearest neighbours (classical regime) → $l = a$
- Low-T: QSL state with long-range entanglement between spin pairs → $l = \xi$

Summary

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- The reduced dimensionality of van der Waals magnets makes them promising candidate for realizing a quantum spin liquid.
- Muons are highly sensitive probes of magnetic fields and are therefore an excellent tool for confirming the absence of magnetic order.
- Muons are also sensitive to dynamics associated with spinon diffusion through the field-dependence of the LF relaxation rate.
- Following the diffusion rate as a function of temperature allows comparison to predictions of various QSL models and a measure of the quantum entanglement length.