Continuous excitations of the triangular-lattice quantum spin-liquid candidate $YbMgGaO_4$

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Acknowledgments

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Oak Ridge National Laboratory

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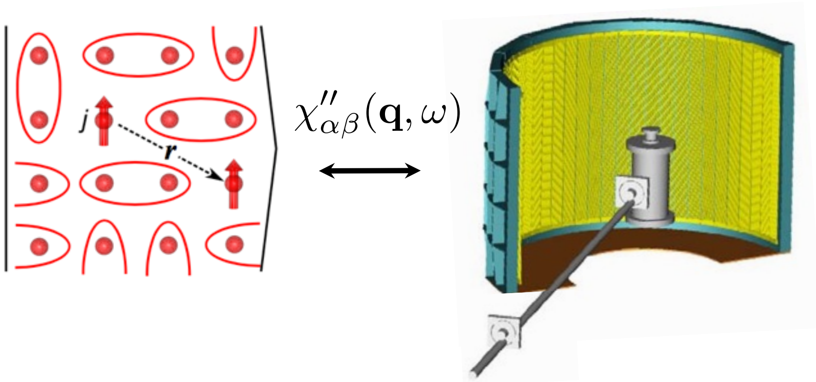
Yuan Wan

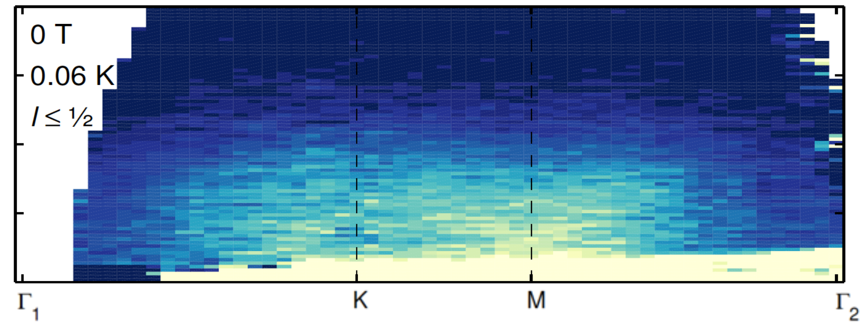
Outline

Investigation of the triangular quantum spin-liquid candidate $YbMgGaO_4$

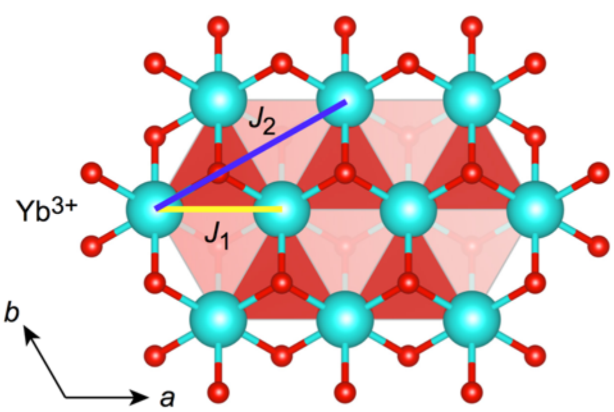
1. General context of this work



3. Inelastic neutron scattering



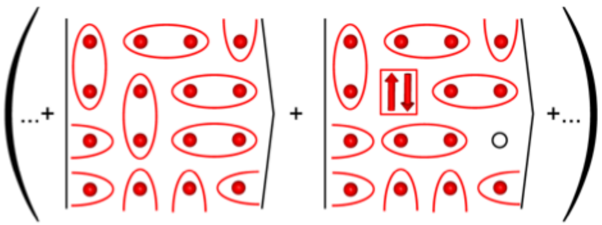
2. Structure & thermomagnetism



4. Conclusion

Spin Liquids in Mott Insulators

- Highly entangled quantum states with beautiful theoretical structure



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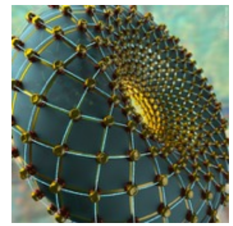

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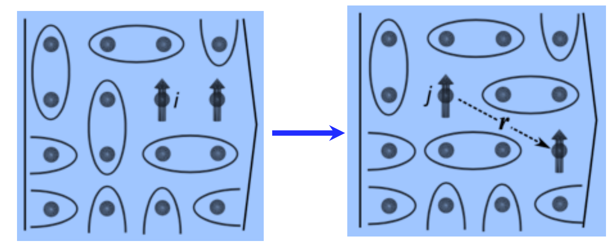


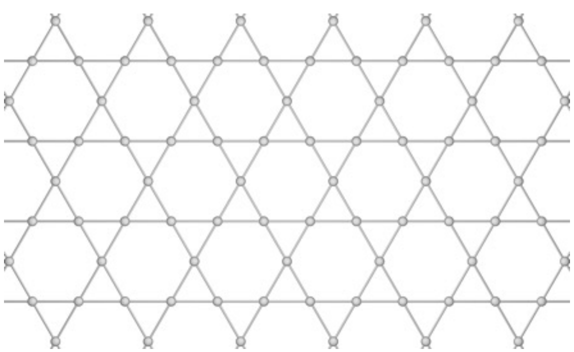
Not a product state
Intrinsically a quantum superposition

Long-range entanglement

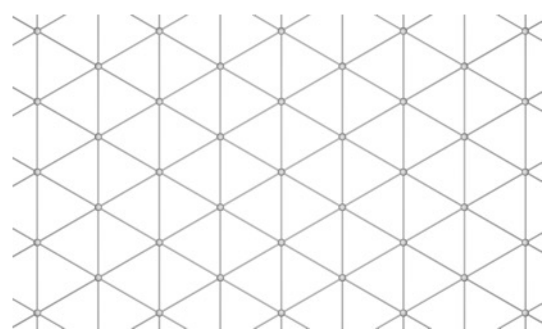
Fractional Excitations

Savary & Balents, Rep. Prog. Phys. **80**, 016502 (2017)

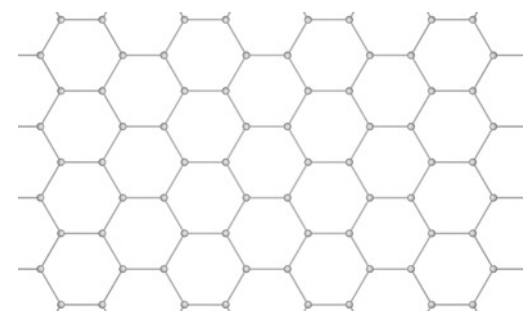
- Intense search for them in low-dimensional (2D) magnets



kagome lattice



triangular lattice



honeycomb lattice

Anderson's RVB spin-liquid or more exotic flavors of a spin-liquid remain elusive

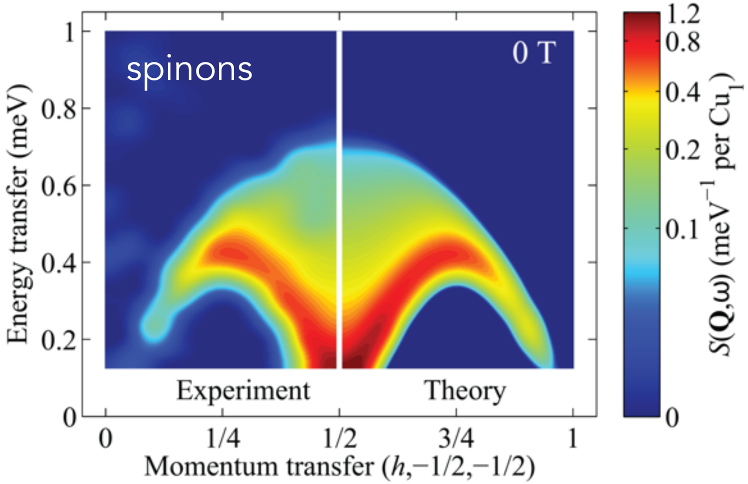
But intermediate quantum states are possible and predicted: magnon decays, plateau, etc

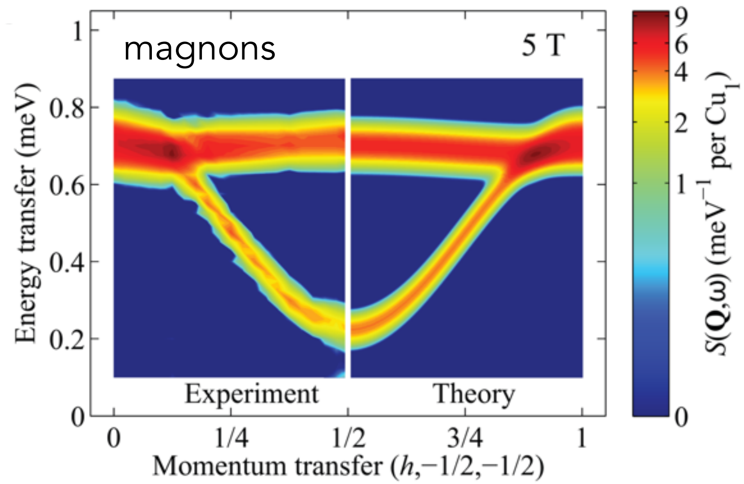
Starykh, Rep. Prog. Phys. **78**, 052502 (2015); Zhitomirsky and Chernyshev, Rev. Mod. Phys. **85**, 219 (2013)

QSL route #1: enhance quantum fluctuations

Transition-metal oxides (Cu^{2+}) on low-dimensional and/or frustrated lattices

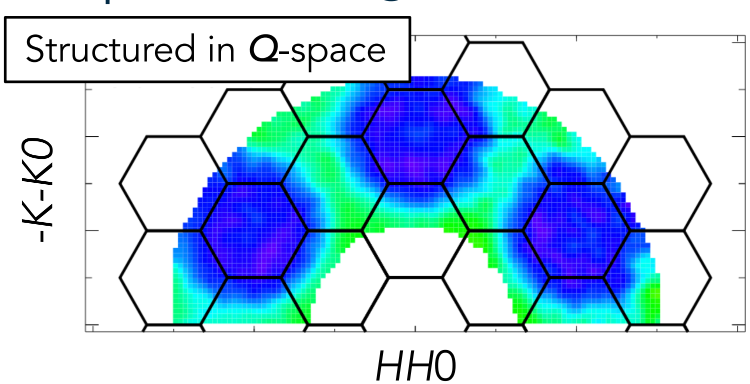
- ❑ QSLs are paradigmatic in spin-1/2 antiferromagnets in 1D

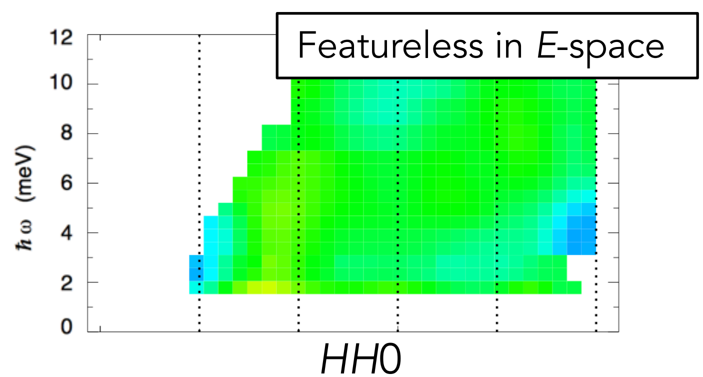




$CuSO_4.5D_2O$: Mourigal, Enderle, Rønnow *et al.*, Nat. Phys. **11**, 62-68 (2015).

- ❑ The spin-1/2 Kagome (2D) antiferromagnet remains the shiniest star





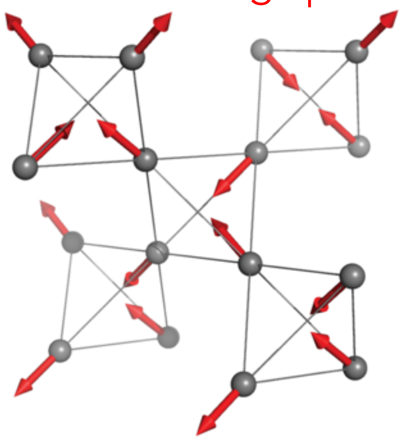
Herbertsmithite: Han *et al.*, Nature **11**, 62-68 (2012); Fu *et al.*, Science **350**, 655-658 (2016).

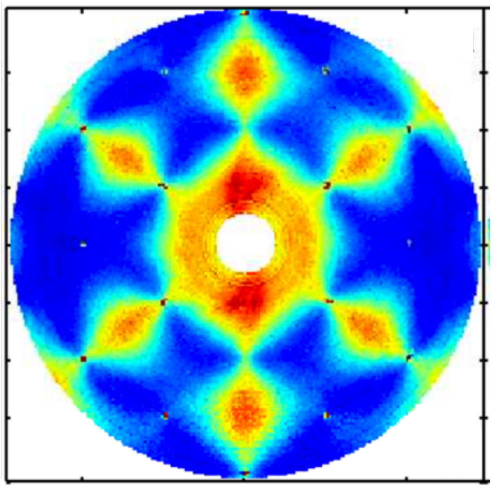
QSL route #2: utilize spin-orbit coupling

Rare-earth pyrochlore oxides (3*f* shell) or 4*d*/5*d* shells on the honeycomb lattice

- ❑ Classical and quantum spin-ice

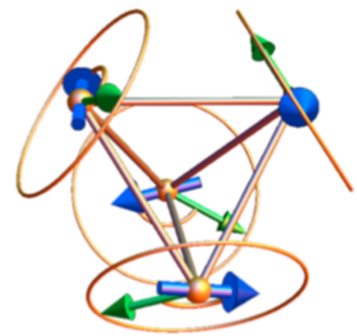
Ising spins

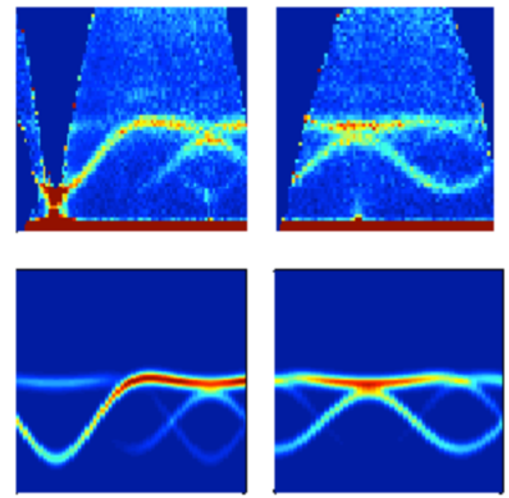




$Ho_2Ti_2O_7$: Fennell *et al.*, Science, 326, 415 (2009).

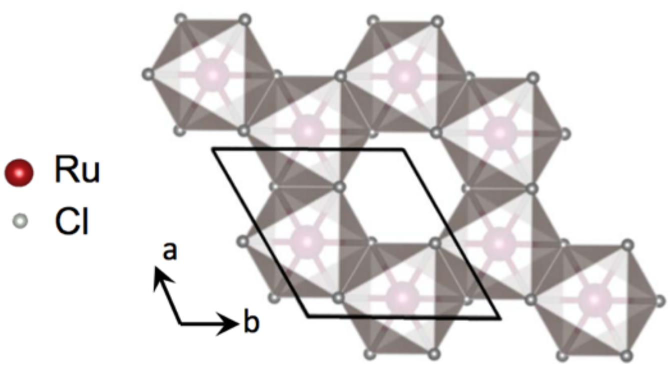
Anisotropic exchange

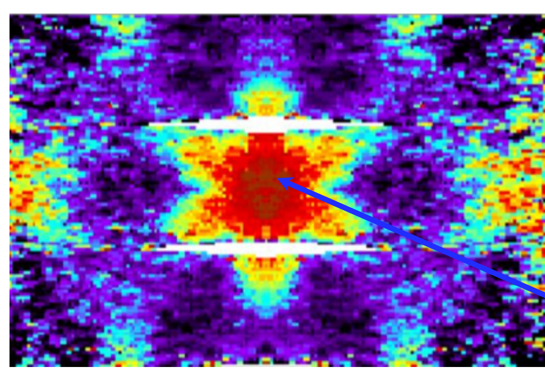




$Yb_2Ti_2O_7$: Ross *et al.*, Phys. Rev. X 1, 021002 (2011)

- ❑ Approximate "Kitaev materials" from bond-directional exchange



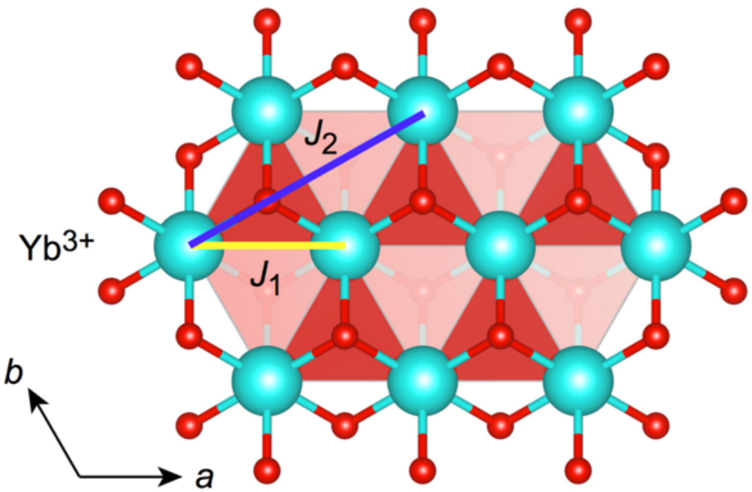


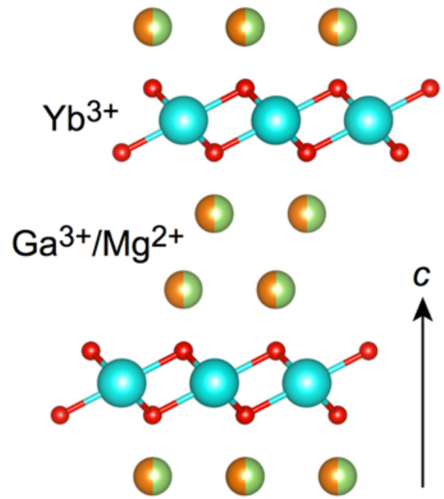
$RuCl_3$: Plumb et al.,
Phys. Rev. B **90**, 041112(R) (2014)
$RuCl_3$: Banerjee *et al.*
Nat. Mater. **15**, 733–740 (2016)
+ arXiV:1609.00103 (2016)

Majorana pairs scattering?

Rare-earth triangular-lattice in $YbMgGaO_4$

- Layered compound discovered in 2015 (Q. Zhang, Renmin Univ., China)





Space-group: *R-3m*

Perfect triangular lattice

No DM interaction

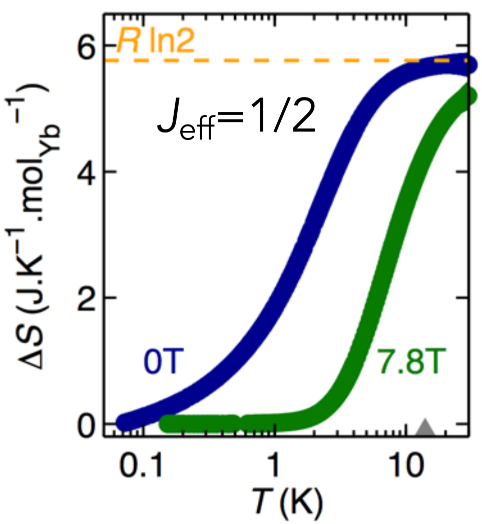
Yb^{3+} ($4f^{13}$) $J = 7/2$

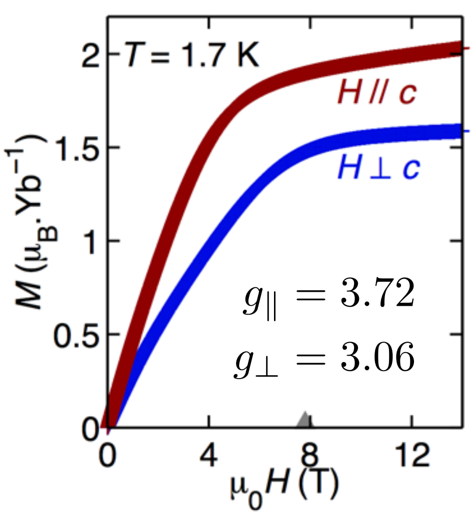
Can be made as large crystal

Ga^{3+}/Mg^{2+} randomness

Y. Li *et al*, Scientific Reports **5**, 16419 (2015); Y. Li *et al*. Phys. Rev. Lett. **115**, 167203 (2015).

- Thermo-magnetic properties point at a J_{eff} = 1/2 XXZ antiferromagnet





Small saturation field H_s<8T

High-*T* magnetic susceptibility

$$\theta_W^{\perp} = -4.8K$$

$$\theta_W^{\parallel} = -3.2K$$

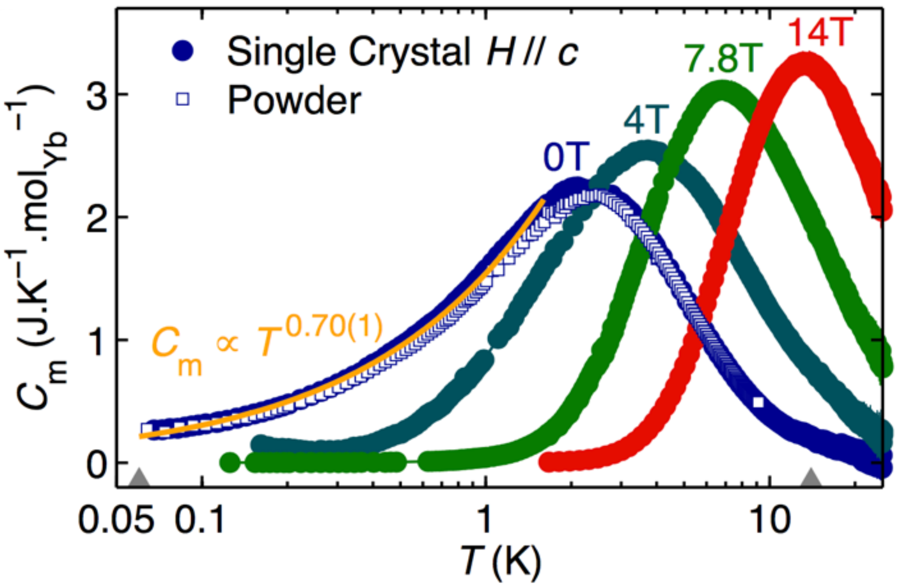
Convex *M(H)* even for $T < \theta_W$

Slope of *M(H)* above H_s: Van-Vleck?

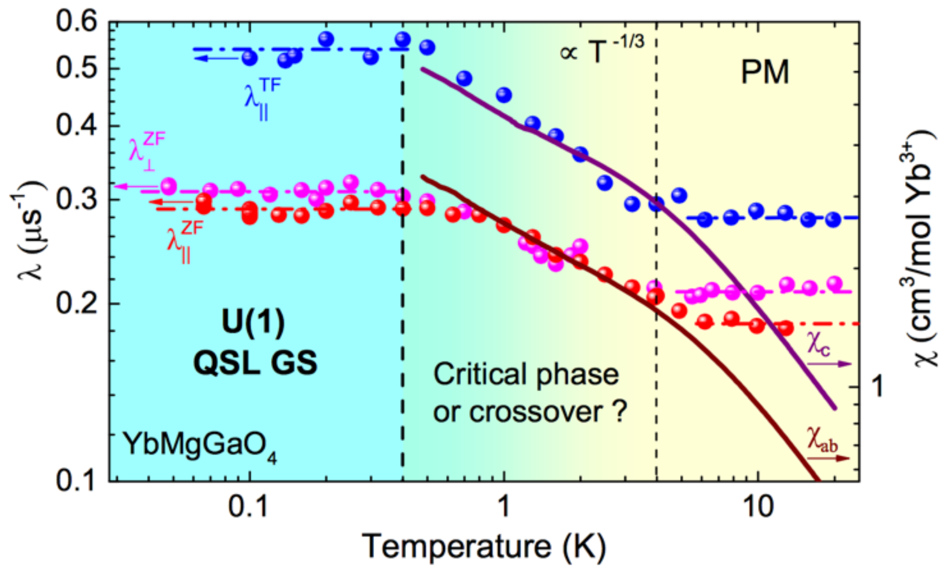
Quantum Spin Liquid Phenomenology

- No-sign of long-range magnetic order in C_p and μSR

See also: Y. Xu *et al*, Phys. Rev. Lett. **117**, 267202 (2016)   Y. Li *et al*, Phys. Rev. Lett. **117**, 097201 (2016)

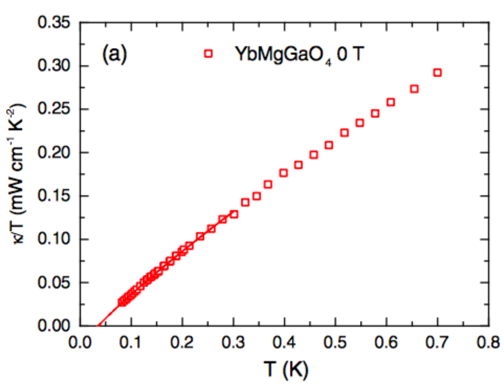


Power-law specific heat below ~2K



Dynamics slow-down but no freezing

- To be contrasted with recent thermal conductivity results



No detectable magnetic contribution to κ

Strong phonon-impurity scattering

Y. Xu *et al*, Phys. Rev. Lett. **117**, 267202 (2016)

Spin-space anisotropy and crystal-electric field

❑ Evidence for weak "off-diagonal" exchange anisotropy terms

Yb^{3+} local symmetry allows 4 terms in n.n. exchange matrix: $\underbrace{J^{zz}, J^{\pm}}_{\text{XXZ}}, \underbrace{J^{\pm\pm}, J^{z\pm}}_{\text{pseudo-dipolar}}$

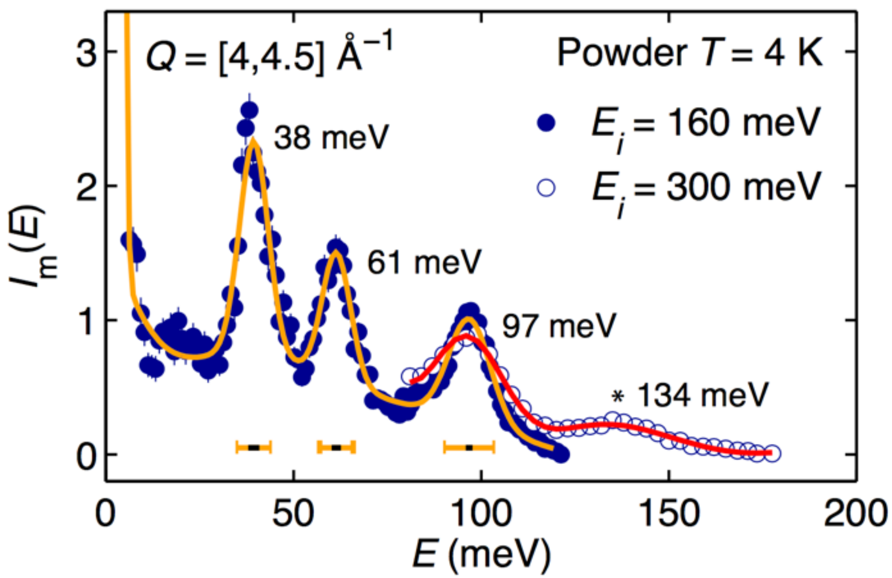
$$+2J_{\pm\pm}\left(S_1^{x_0}S_2^{x_0} - S_1^{y_0}S_2^{y_0}\right) + J_{z\pm}\left(S_1^{z_0}S_2^{y_0} + S_1^{y_0}S_2^{z_0}\right)$$

Y.-D. Li *et al.*, Phys. Rev. B **94**, 035107 (2016) + arXiv:1608.06445 (2016), 1612.03447 (2016) & 1703.01876 (2017).
Z. Zhu *et al.*, arXiv:1703.0297 (2017)

Electron spin resonance at *T* = 10 K implies that $J^{z\pm}$ is vanishingly small

Y. Li *et al.* Phys. Rev. Lett. **115**, 167203 (2015)

❑ Evidence that disorder plays a role from width of CEF levels



Expect 4 Kramers doublet by splitting *J*=7/2

Ground-state doublet well-separated from three excited doublets

High-energy modes are very broad

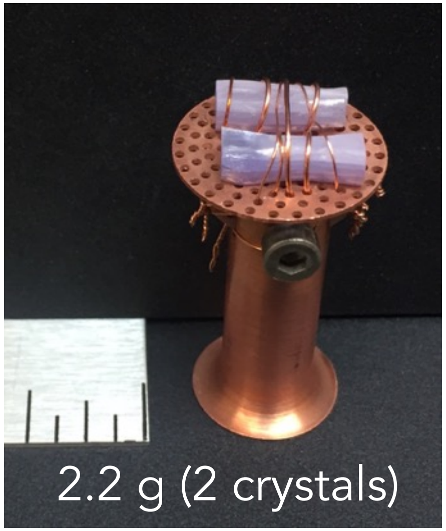
An additional mode is observed

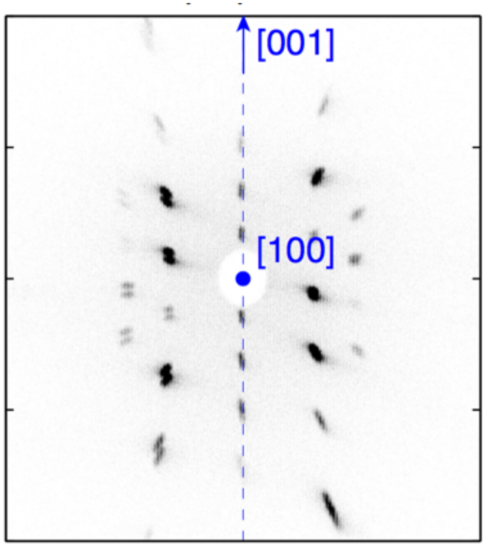
Detailed study concludes distribution of *g*

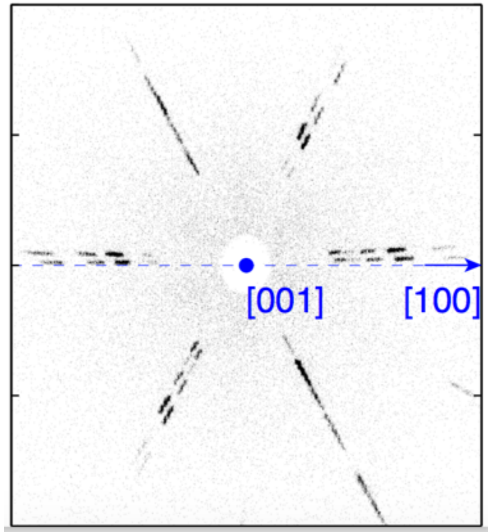
Y. Li *et al.*, arXiv:1702.01981 (2017), to appear in PRL

High quality single-crystals can be obtained

- Floating zone growth (Zhiling Dun and Haidong Zhou)

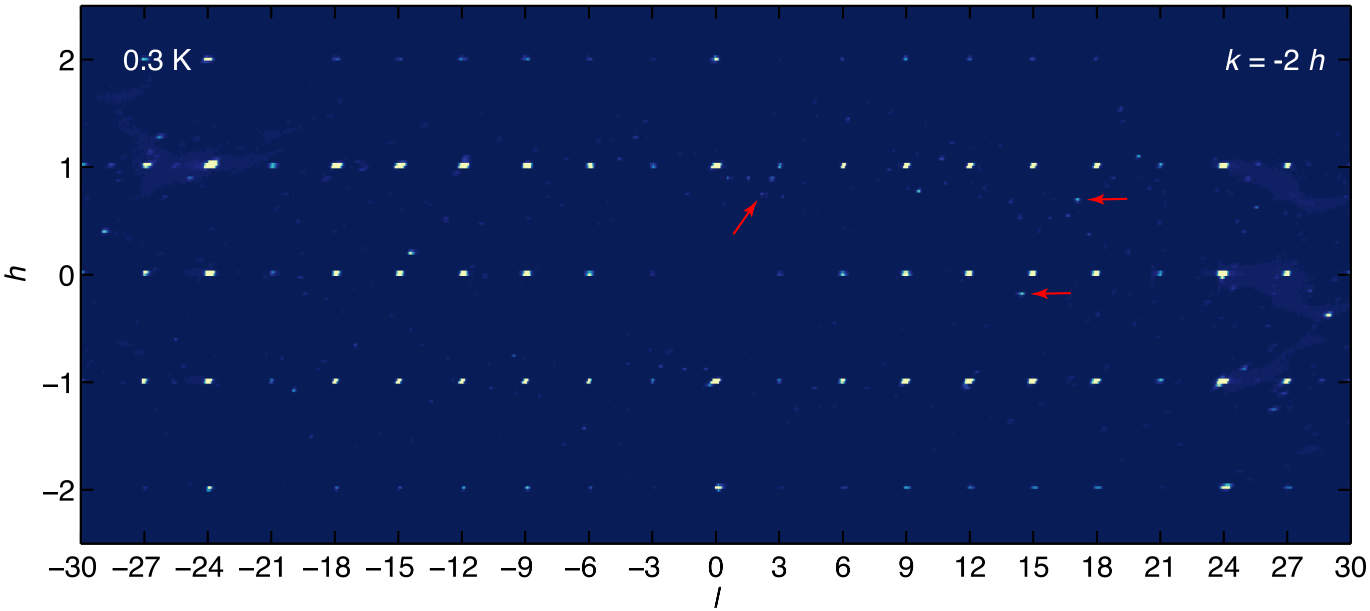






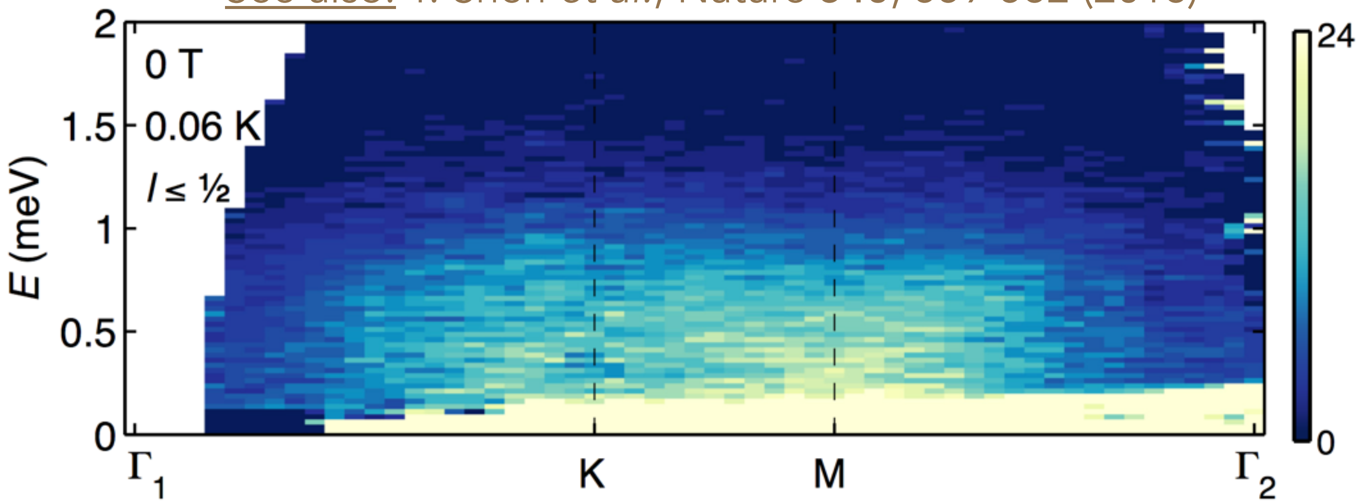
Aligned with ~1.5 degrees

Predominantly a single grain



Neutron scattering in zero-field

<u>See also:</u> Y. Shen *et al.*, Nature **540**, 559-562 (2016)



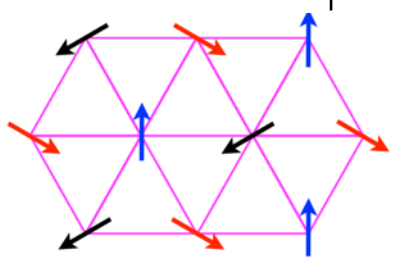
Broad magnetic signal

Bandwidth ~1.5 meV

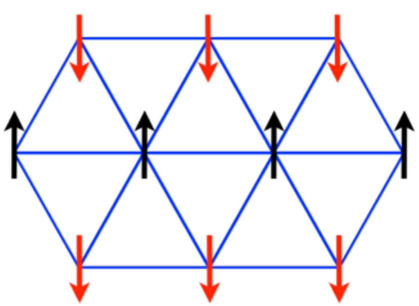
Almost featureless

More intense at the *M*-point of the BZ

120° structure = K-point



Stripe structure = M-point

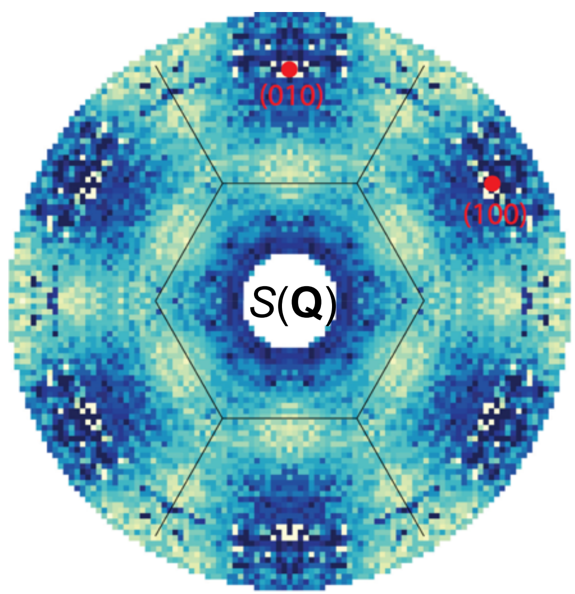


Structured in **Q**-space

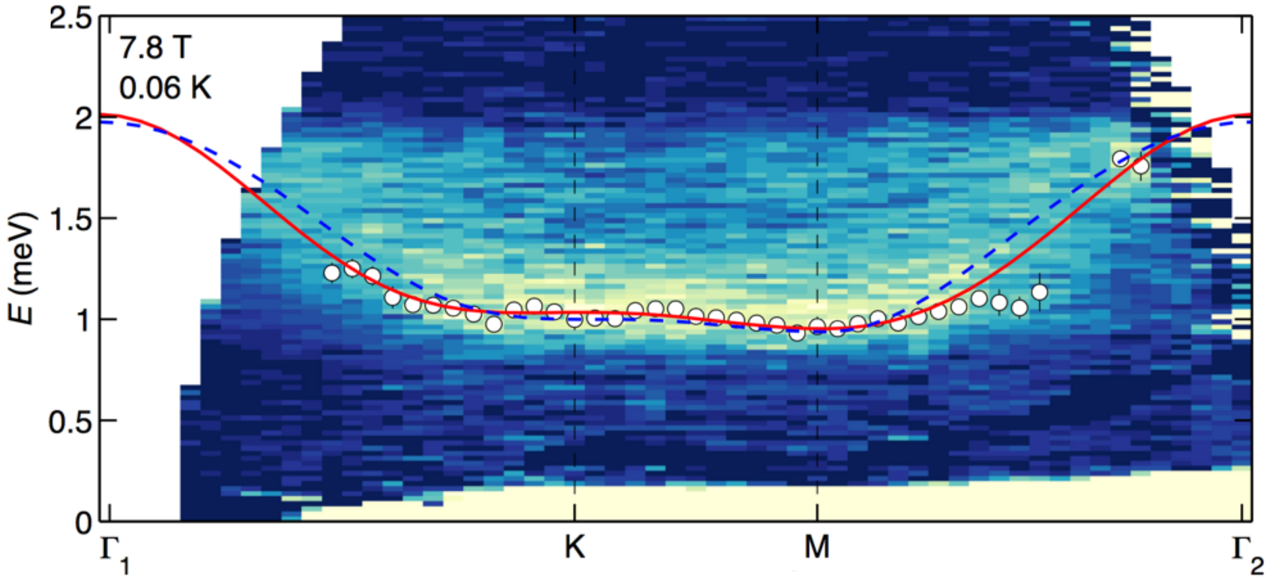
Maximum ~15% of the spectral weight is elastic

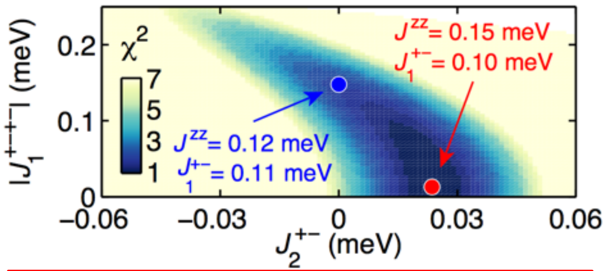
Diffuse scattering peaks at the *M*-point of the Brillouin zone!

[0,1.6] meV



Neutron scattering above the saturation field





$J_1^{zz} + J_2^{zz} = 0.15\text{meV}$

$J_1^{\pm} = 0.109 \text{ meV}, J_2^{\pm} = 0.22 J_1^{\pm}$

Best fit: includes J_2

- ❑ Modeling the dominant "mode-like" feature

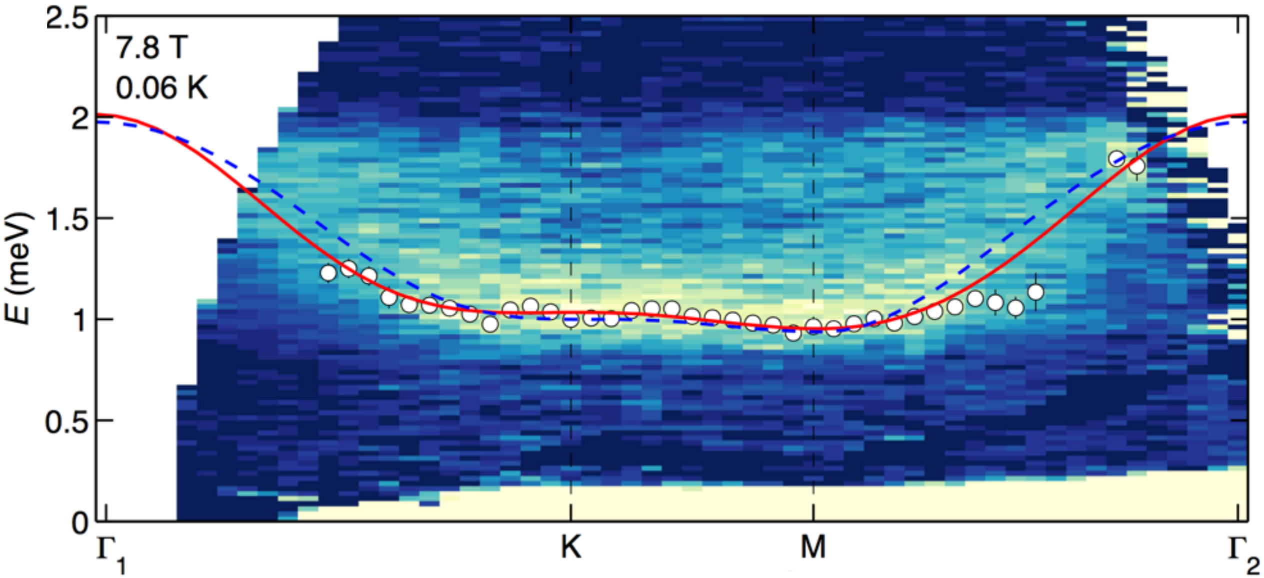
Yb^{3+} local symmetry allows 4 terms in exchange matrix: $\overbrace{J^{zz}, J^{\pm}}^{\text{XXZ}}, \underbrace{J^{\pm\pm}, J^{z\pm}}_{\text{pseudo-dipolar}}$

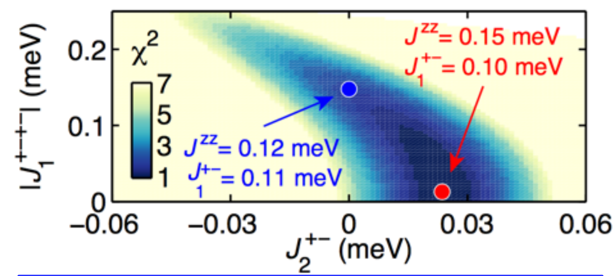
Phenomenological J_2 XXZ exchange, $J_1^{z\pm} \approx 0$ from ESR

$$\mathcal{H} = \sum_{\langle i,j \rangle} \left[J_1^{zz} S_i^z S_j^z + J_1^{\pm} \left(S_i^+ S_j^- + S_i^- S_j^+ \right) + J_1^{\pm\pm} \left(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^- \right) \right]$$
$$+ \sum_{\langle\langle i,j \rangle\rangle} \left[J_2^{zz} S_i^z S_j^z + J_2^{\pm} \left(S_i^+ S_j^- + S_i^- S_j^+ \right) \right] - \mu_0 \mu_B \sum_i \left[g_\perp \left(H^x S_i^x + H^y S_i^y \right) + g_\parallel H^z S_i^z \right]$$

Y. D. Li *et al.*, Phys. Rev. B **94**, 035107 (2016); arXiv:1608.06445 (2016).

Neutron scattering above the saturation field





$J_1^{zz} = 0.12$ meV, $J_1^{\pm} = 0.11$ meV

$|J_1^{\pm\pm}| = 0.15$ meV

Alternative Fit

- Modeling the dominant "mode-like" feature

Yb^{3+} local symmetry allows 4 terms in exchange matrix: $\overbrace{J^{zz}, J^{\pm}}^{\text{XXZ}}, \underbrace{J^{\pm\pm}, J^{z\pm}}_{\text{pseudo-dipolar}}$

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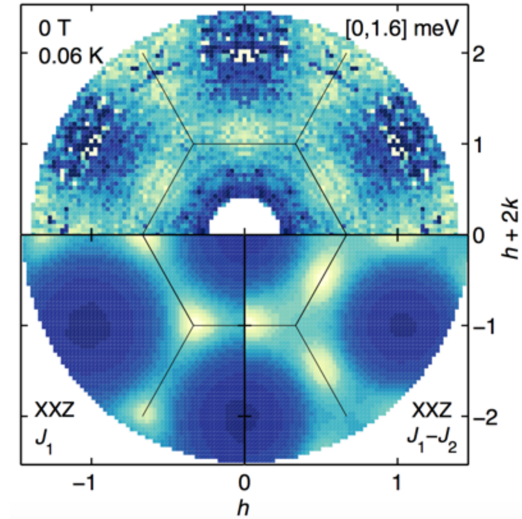
Y. D. Li *et al.*, Phys. Rev. B **94**, 035107 (2016); arXiv:1608.06445 (2016).

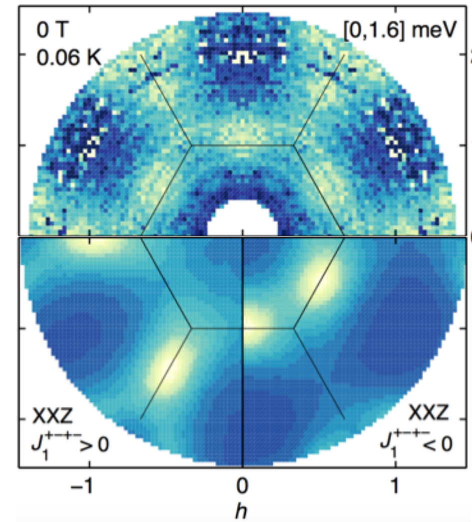
Additional pieces of the puzzle

- ❑ Strong or weak pseudo-dipolar exchange?

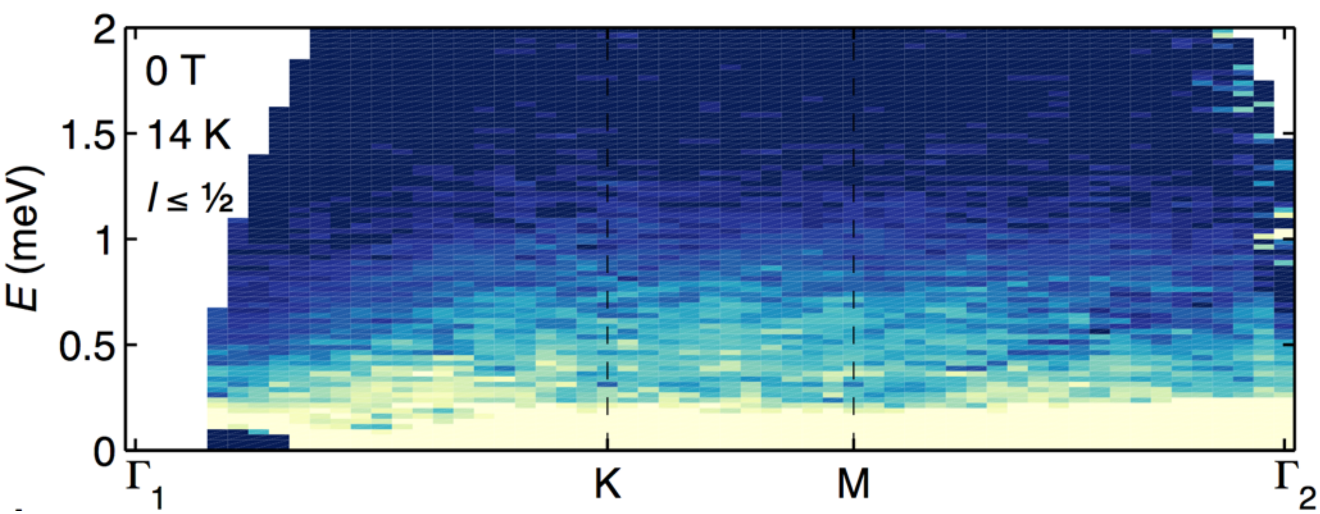
Performed classical Monte-Carlo simulations for both sets of exchange interactions, $T \sim 1.3$K

Strong $J_1^{\pm\pm}$ produces unobserved modulation in the diffuse scattering pattern





- ❑ Temperature dependence is very unusual



Continuum persists at $T > 3\theta_W$

Spectral weight expected to concentrate at Q~0

Conclusions and future work $YbMgGaO_4$

- We believe we found the nominal Hamiltonian for this new material:

  A J_{eff}=1/2 XXZ triangular-lattice antiferromagnet (moderate planar anisotropy) with surprisingly $J_2 \approx 0.2\ J_1$ and likely role of charged inter-plane disorder

- Theoretical interpretation remains an open question

  Jun Zhao and Gang Chen's work in Nature: U(1) QSL with spinon Fermi surface

  Y. Shen *et al.*, Nature **540**, 559-562 (2016) + Y.-D. Li *et al.*, arXiv:1612.03447 & arXiv:1703.01876

  Recent 1/S+DMRG analysis suggests a "spin-liquid mimicry" mechanism

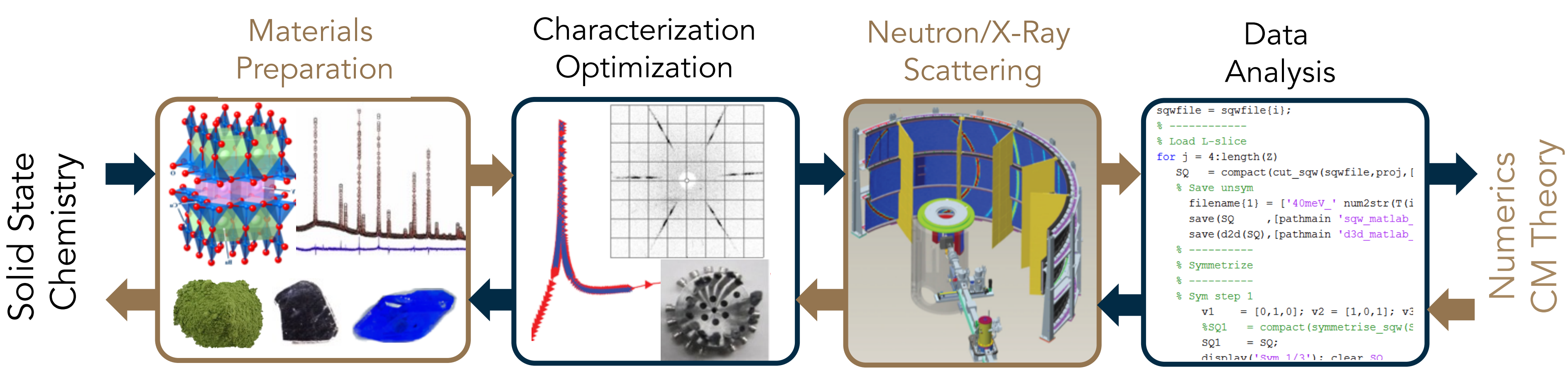
  Z. Zhu *et al.*, arXiv:1703.0297 (2017)

- Explore many related materials with other rare-earths ions

  Difficult in the *RE*$MgGaO_4$ structure … M. B. Sanders *et al.*, arXiv:1611.08548 (2016)

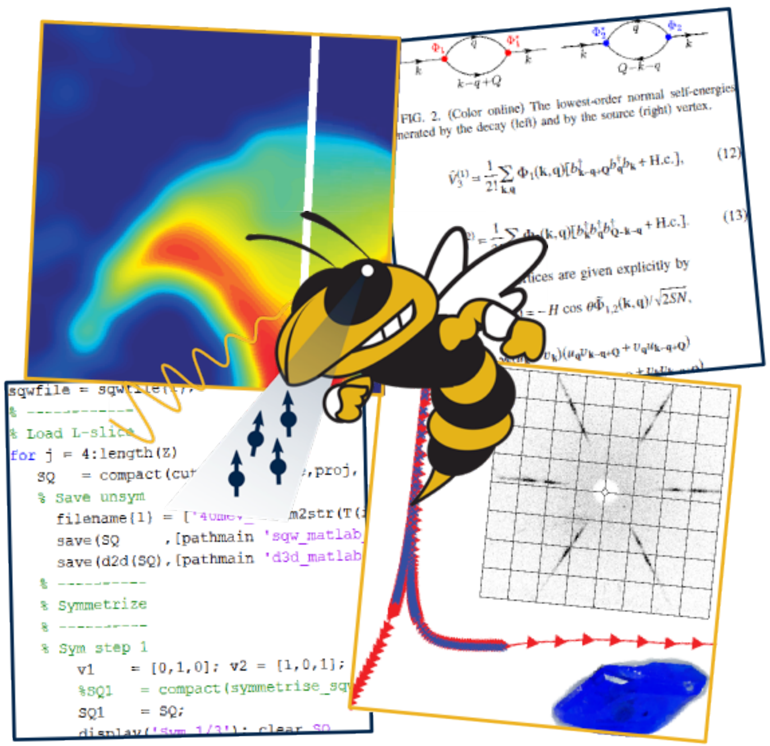
  but very recent discovery of KBa*RE*$(BO3)_2$ (*RE*=Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu)

- Coupling between sample growth, spectroscopy and theory crucial



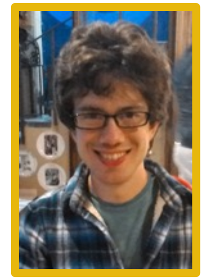
Thank you for your attention!

Quantum Materials Spectroscopy
@ Georgia Tech

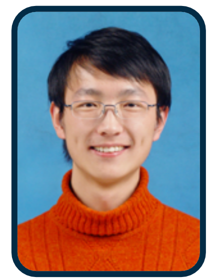


web mourigal.gatech.edu

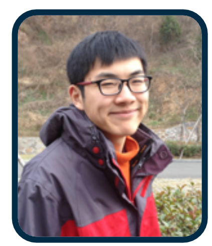
Lab Members



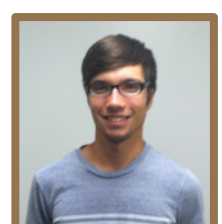
Joseph Paddison
Postdoctoral Fellow
Now in Cambridge



Xiaojian Bai
3rd year Graduate Student



Luwei Ge
3rd year Graduate Student



Marcus Daum
2nd year Graduate Student