Triangular rare-earth quantum magnets

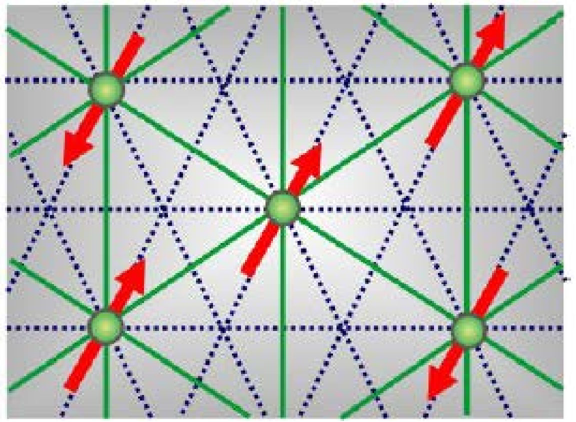
Philipp Gegenwart

Institute of Physics
Center for Electronic Correlations and Magnetism

Augsburg University

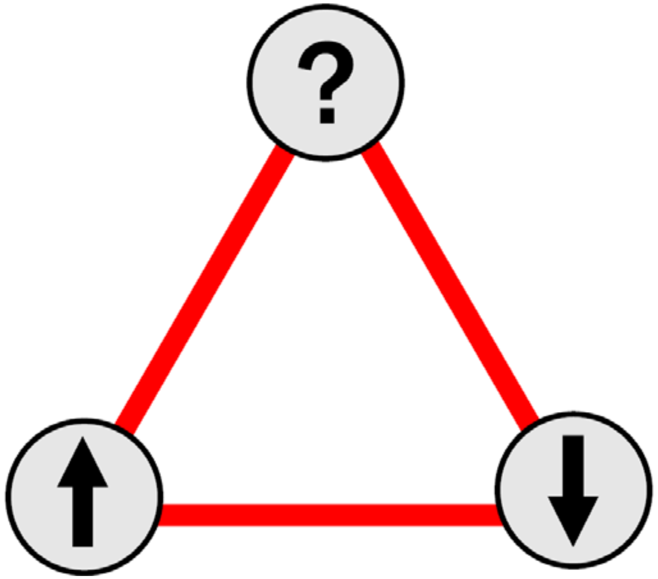
TRR 80

- Magnetic frustration
- Quantum spin liquids and fractionalized excitations
- The triangular lattice quantum magnet $YbMgGaO_4$
- Outlook

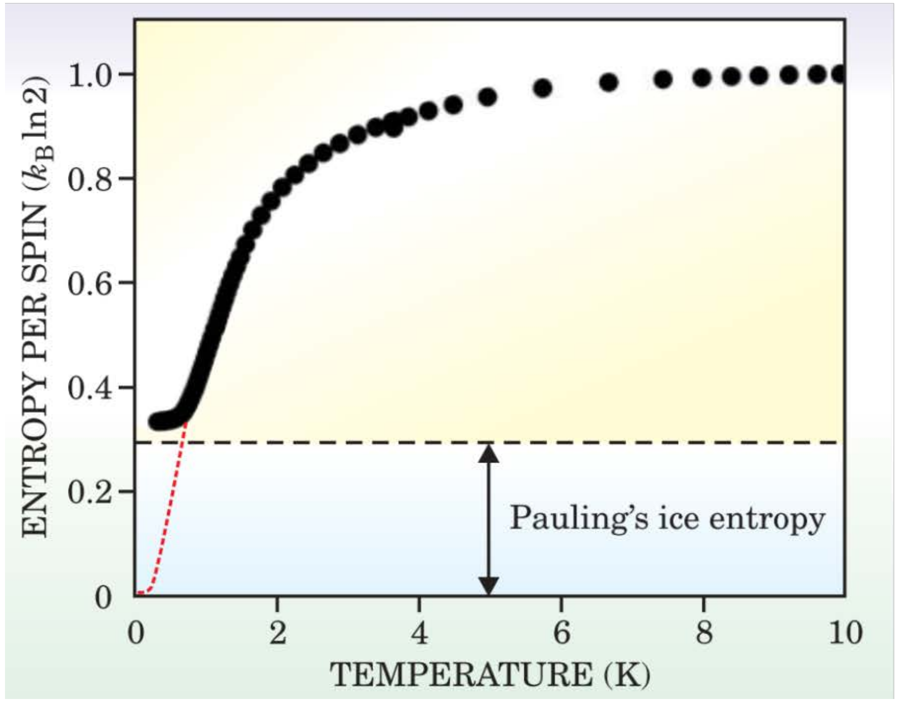


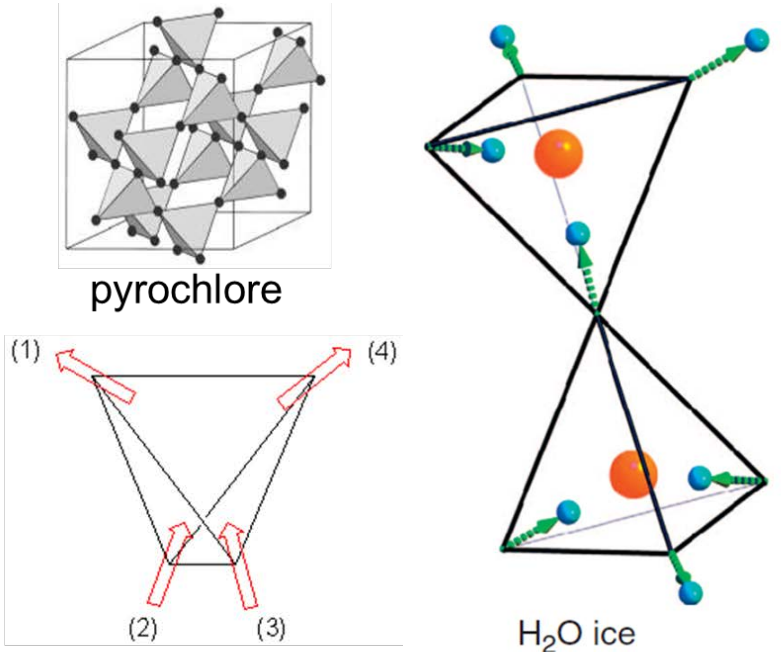
In collaboration with Yuesheng Li, Sebastian Bachus, Franziska Grußler, Yoshi Tokiwa, Alexander Tsirlin,…

Magnetic frustration



A. P. Ramirez et al., Nature 399, 333 (1999)





→ Residual entropy R/2 *ln*(3/2)

- No way to fully satisfy all of pairwise interactions

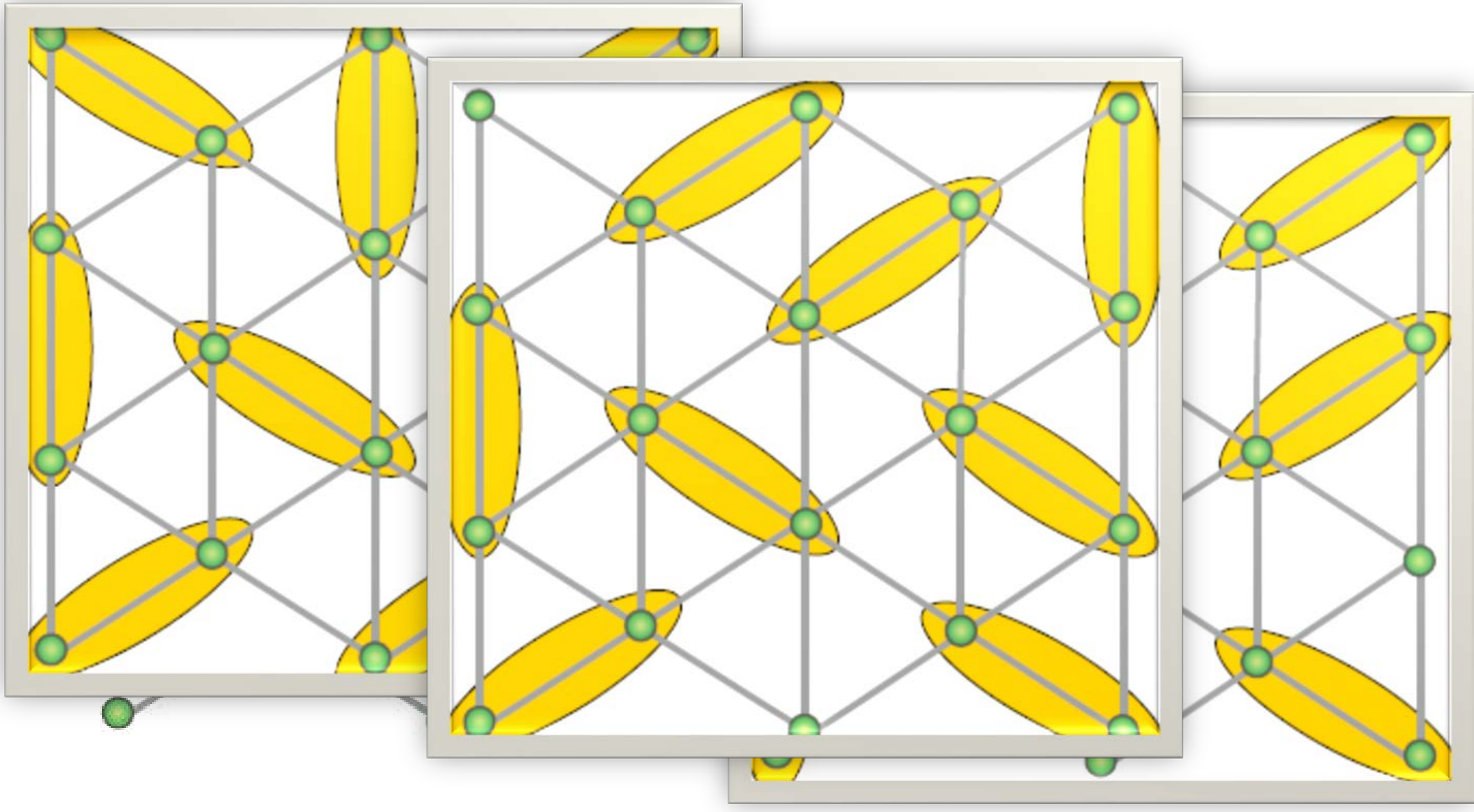
→ macro **degenerate states** → residual entropy, (classical) spin liquid freezes at low T

- **Quantum** spin liquid: quantum entanglement (no residual entropy and spin freezing), **fractionalized** excitations

Resonating Valence Bonds (RVB)

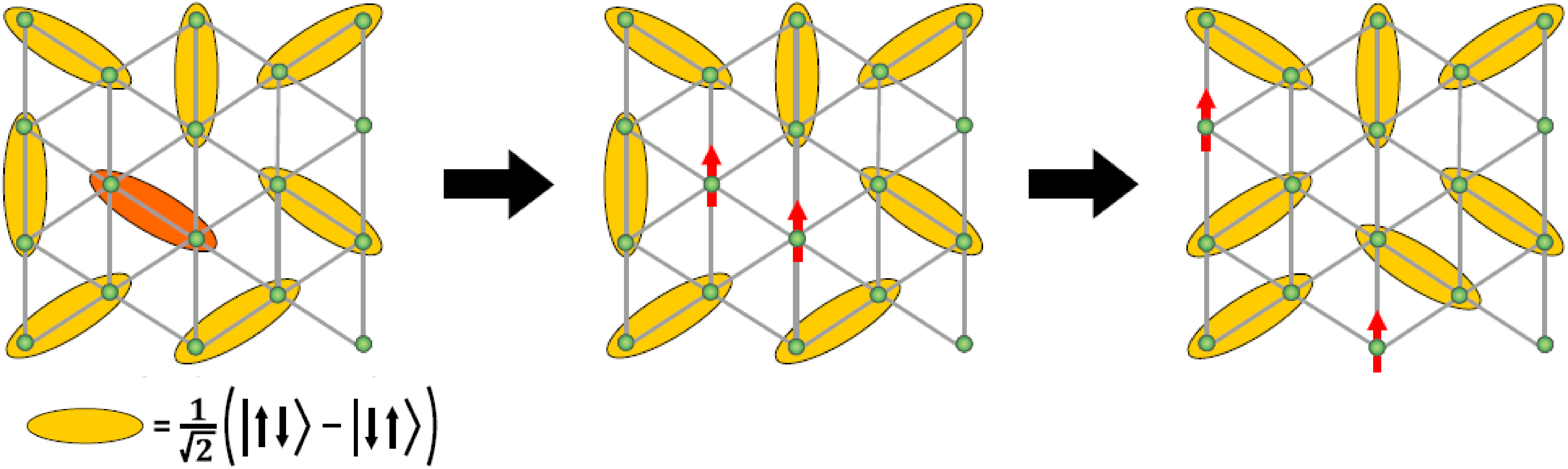
P.W. Anderson (1973):

- Spontaneous spin pairing to singlets
- Ground state as quantum superposition of all possible pair states



$$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$$

Spinon excitations in the RVB scenario



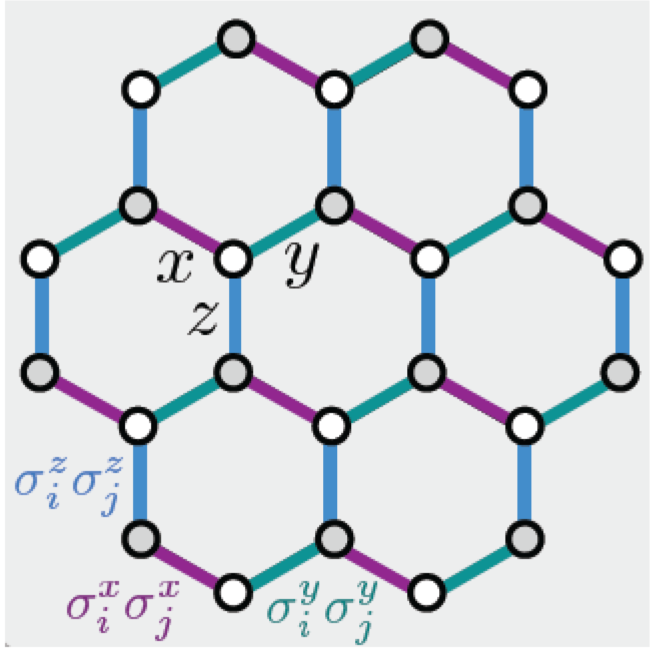
$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$

After breaking a bond, the two unpaired spins can move apart, since rearrangement of valence bonds conserves the energy.

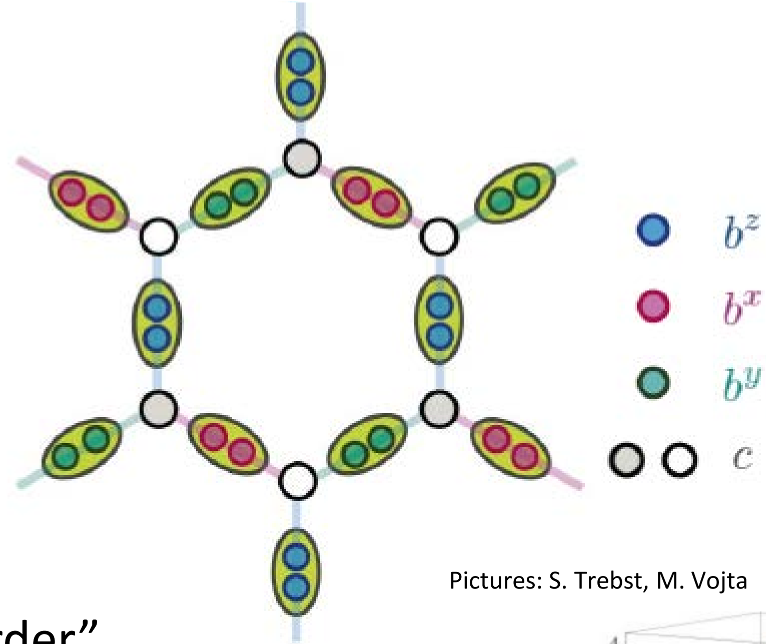
→ spin triplet S=1 excitation breaks into two S=1/2 excitations (“spinons”)

→ Broad excitations in E(**q**), entirely different to sharp magnons

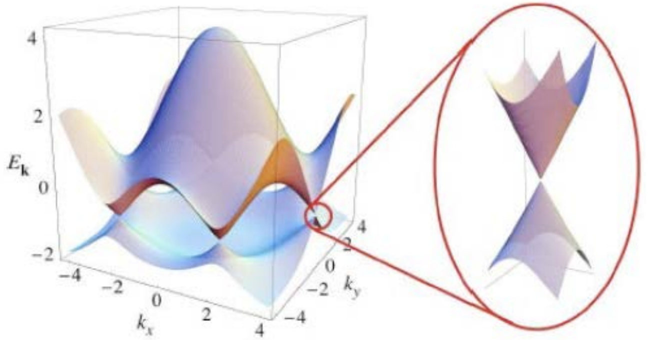
Spin fractionalization in the Kitaev model



$$H_{\text{Kitaev}} = \sum_{\gamma-\text{links}} J_\gamma \sigma_i^\gamma \sigma_j^\gamma$$

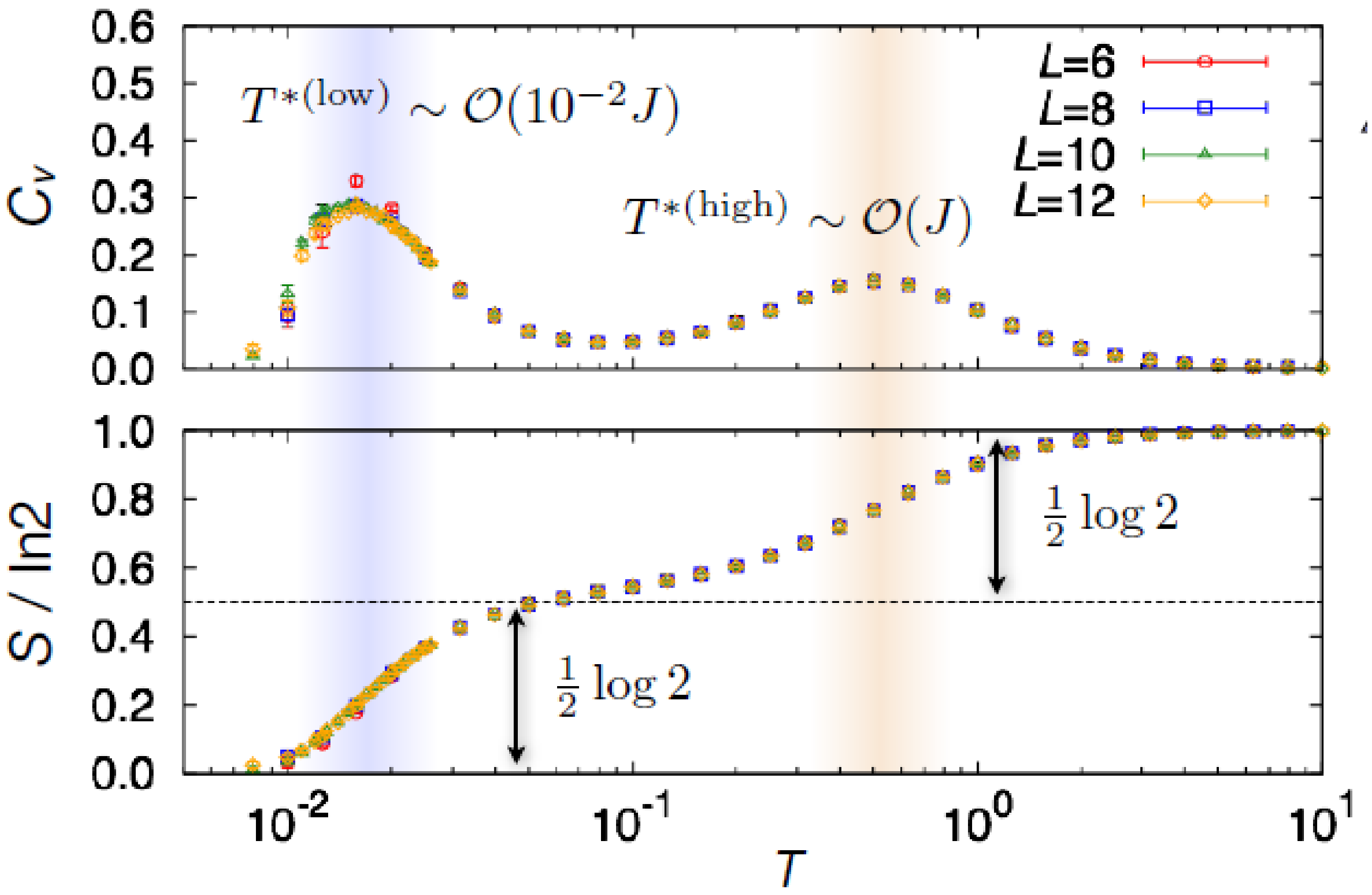


Pictures: S. Trebst, M. Vojta



- No geometrical but **bond** frustration
- Exact solution by mapping spins to **Majorana fermions**
  itinerant M.: Dirac-like dispersion, localized M.: “flux order”
- In presence of magnetic field: Majoranas behave as „anyons“ obeying non-Abelien statistics (application in topological quantum computation)

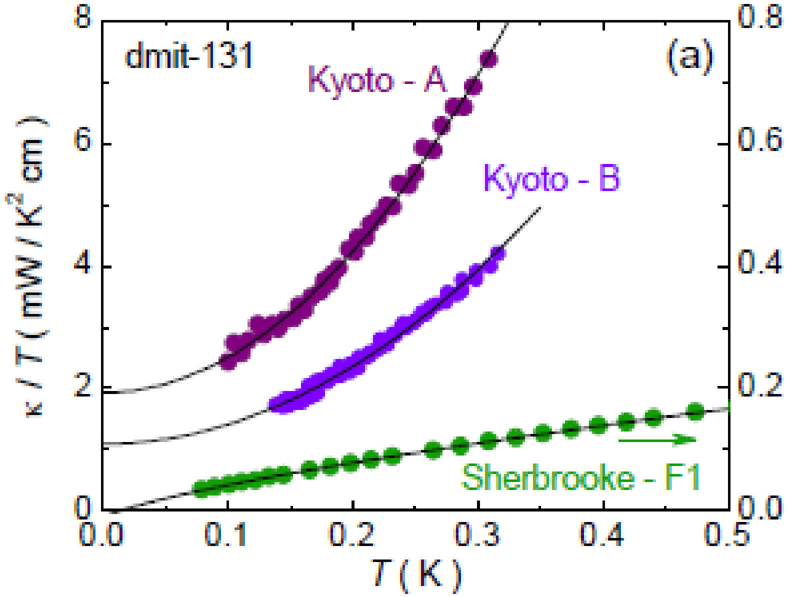
Fractionalization in the Kitaev model

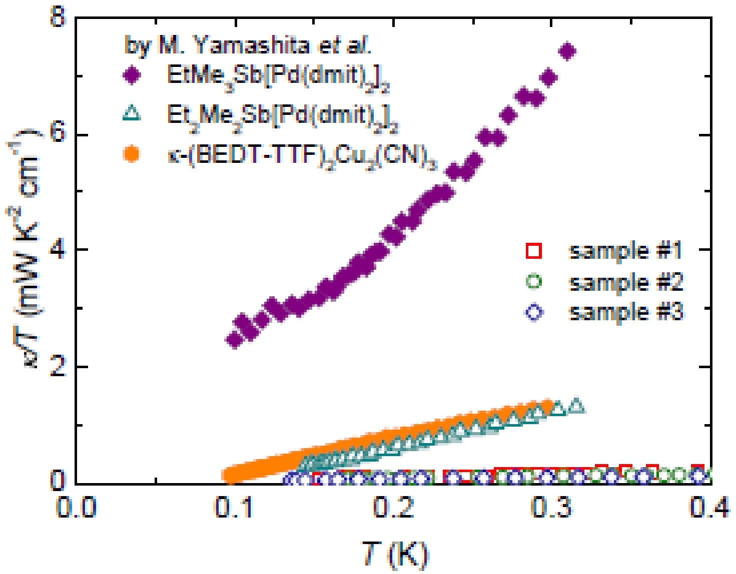


Y. Motome et al. (2015,2016)

Two-step entropy release

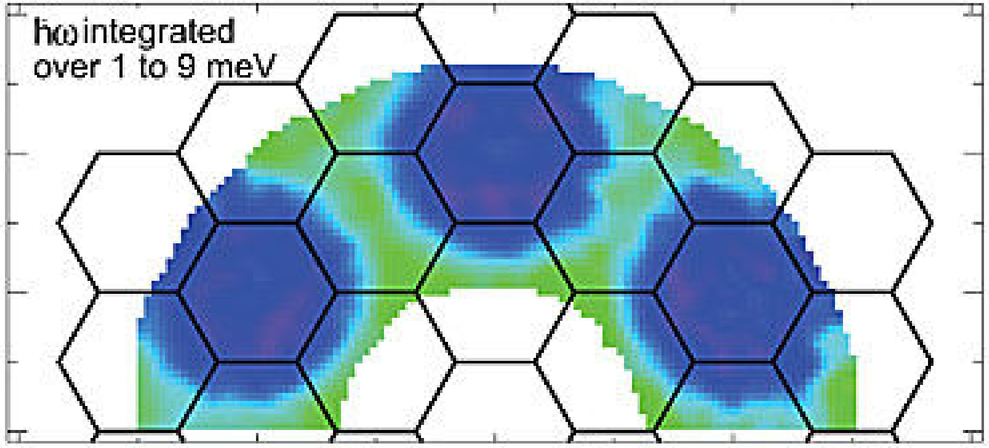
Observation of spin fractionalization?





P. Bourgeois-Hope et al. and J.M. Ni et al, arXiv 2019:
Sherbrooke and Shanghai groups in disagreement with previous Kyoto results on $EtMe_3Sb[Pd(dmit)_2]_2$ (M. Yamashita et al., Science 2010)

$ZnCu_3(OH)_6Cl_2$: ~15% of Zn-sites filled by Cu spins



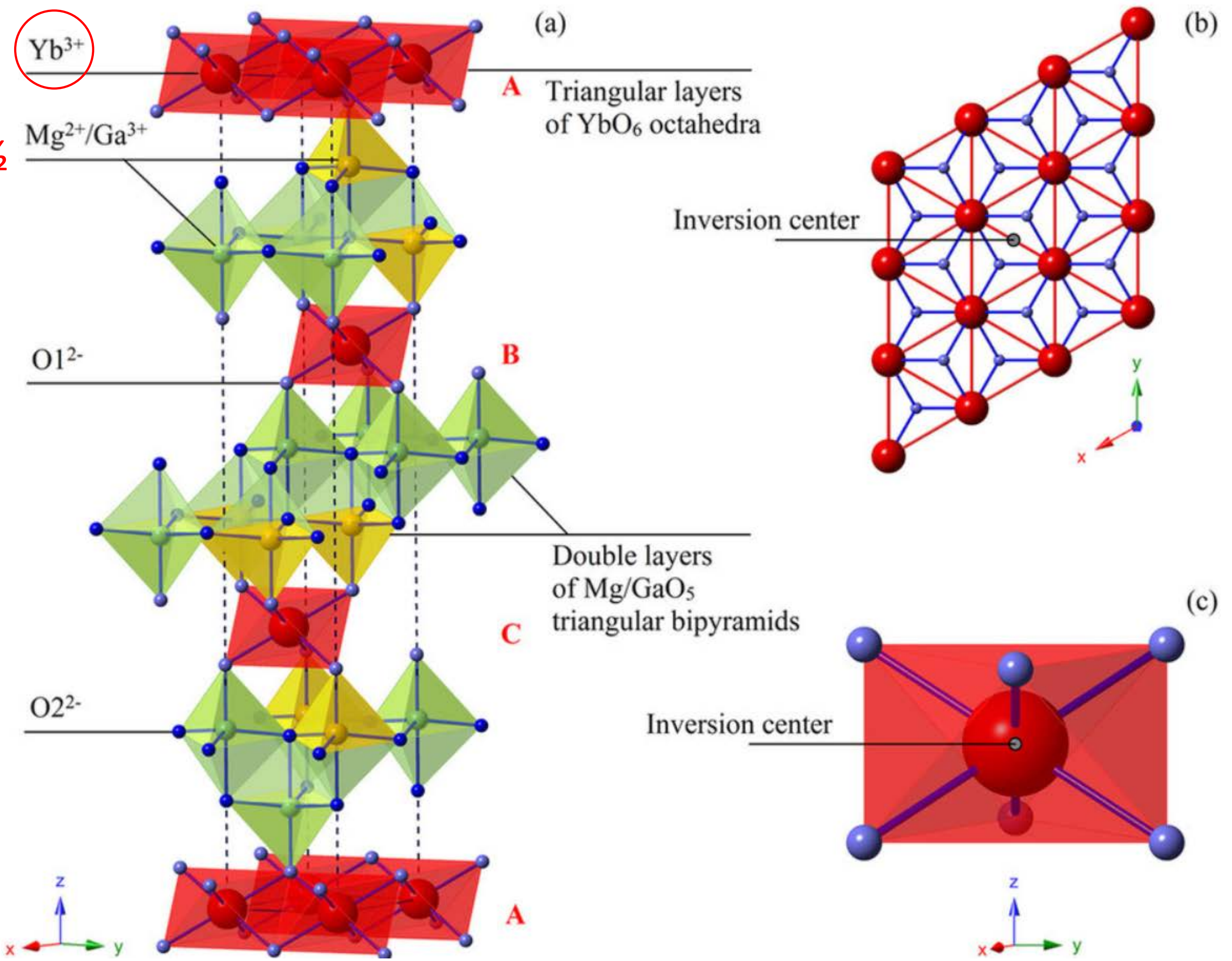
Han et al., Nature 2012

Majorana Fermions in Kitaev materials?

- Half-integer quantized thermal Hall effect (chiral edge current of charge neutral Majoranas)?
- Broad excitation continua (Raman, INS): role of off-diagonal Γ exchange?
- Hydrogenated iridate: strong bond randomness

Triangular rare-earth AFs

one 4f hole →
J=7/2, low-T:
effective spin ½



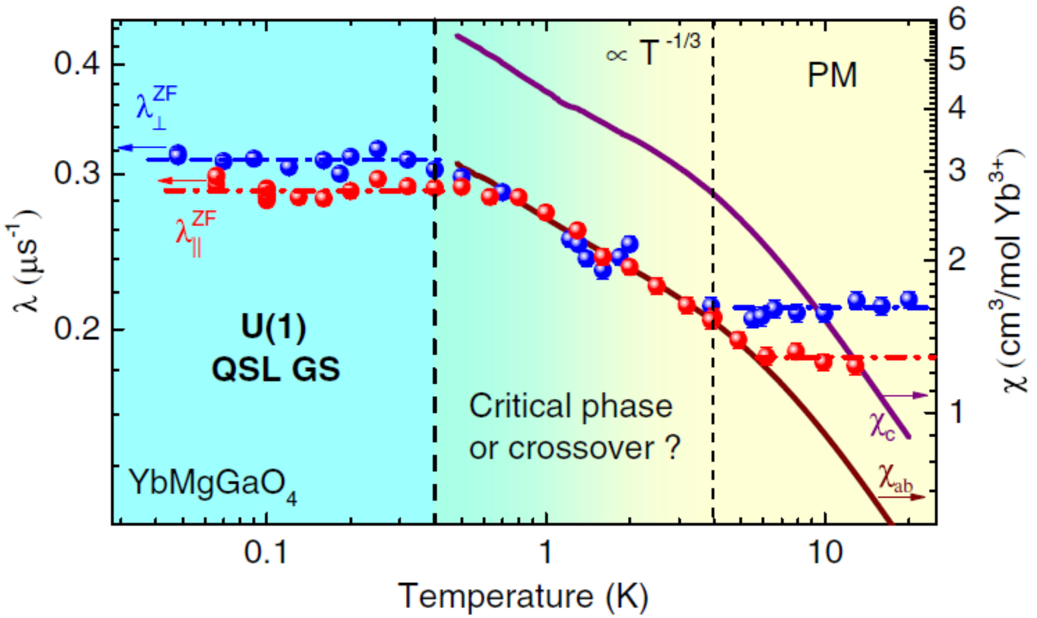
YbMgGaO$_4$

- Free from defects, spatial anisotropy, antisymmetric DM anisotropy
- Large single crystals for INS

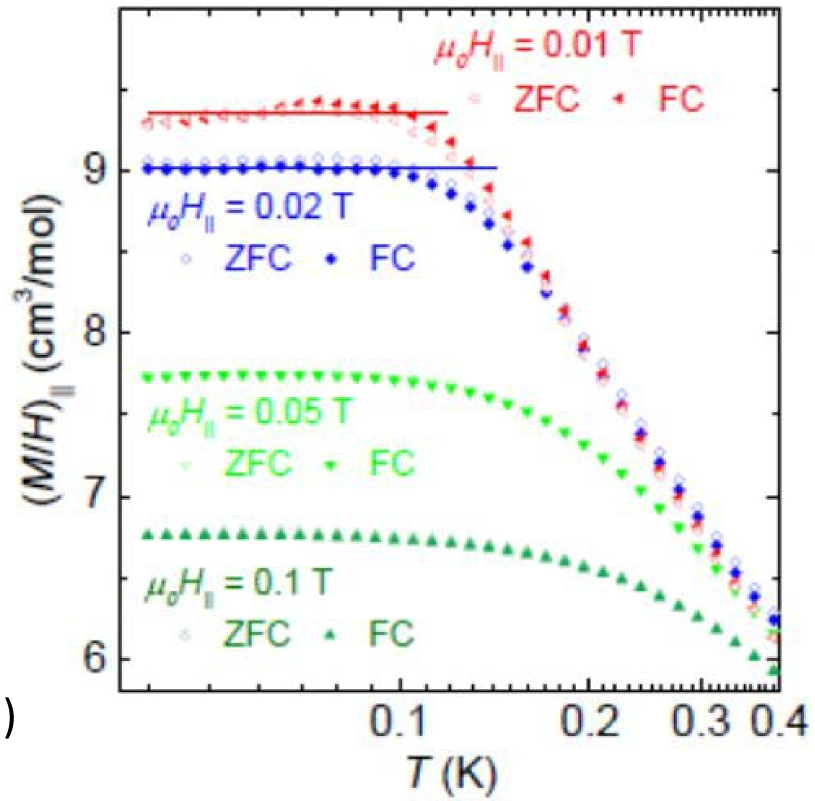
Yuesheng Li et al, Sci. Rep. 5 (2015) 16419, PRL 115 (2015), 167203

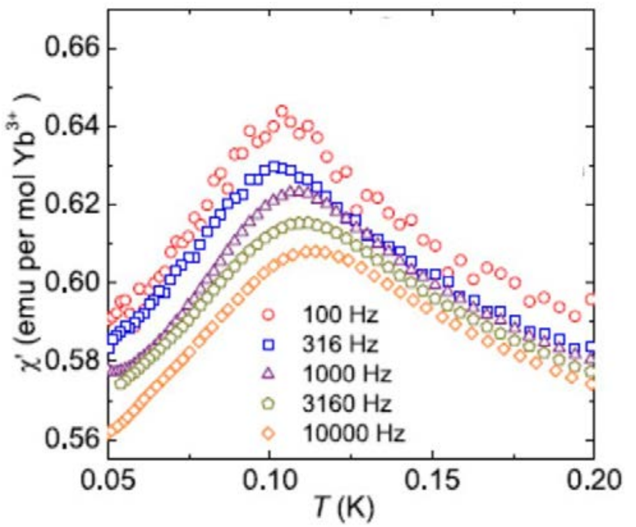
Gapless low-energy excitations in $YbMgGaO_4$

μSR-relaxation rate



Yuesheng Li et al., PRL (2016), PRL (2019)





Z. Ma *et al.* PRL 120, 087201 (2018)

- no order, freezing or gapping seen in M(T), C(T), α(T), μSR down to 50 mK
- Cusp in χ_{ac} involves <1% entropy → (only small fraction of frozen spins)

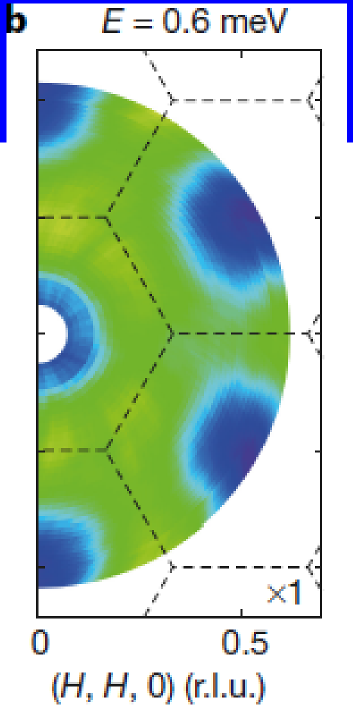
Spinons in $YbMgGaO_4$?

LETTER

doi:10.1038/nature20614

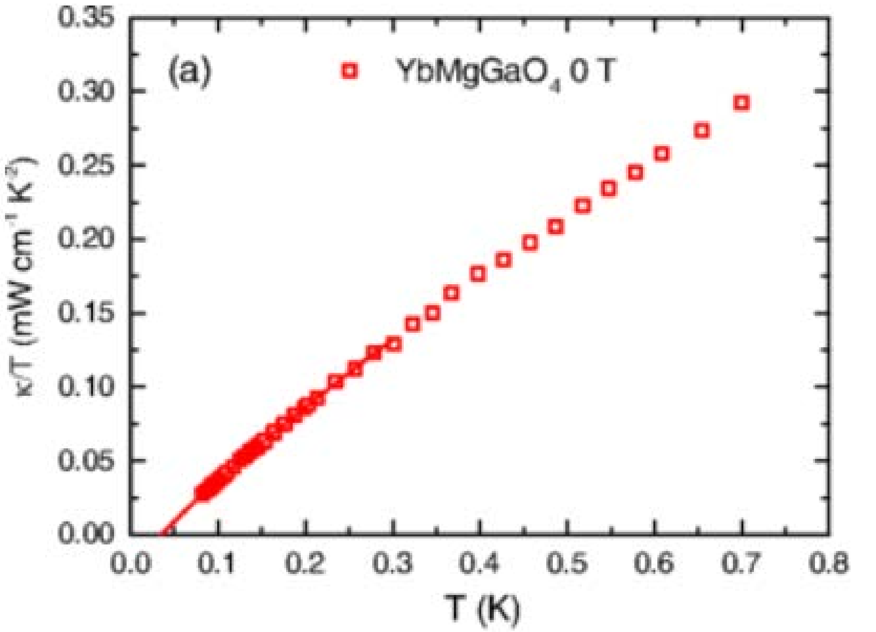
Evidence for a spinon Fermi surface in a triangular-lattice quantum-spin-liquid candidate

Yao Shen[1], Yao-Dong Li[2], Hongliang Wo[1], Yuesheng Li[3], Shoudong Shen[1], Bingying Pan[1], Qisi Wang[1], H. C. Walker[4], P. Steffens[5], M. Boehm[5], Yiqing Hao[1], D. L. Quintero-Castro[6], L. W. Harriger[7], M. D. Frontzek[8], Lijie Hao[9], Siqin Meng[9], Qingming Zhang[3,10,11], Gang Chen[1,11,12] & Jun Zhao[1,11]



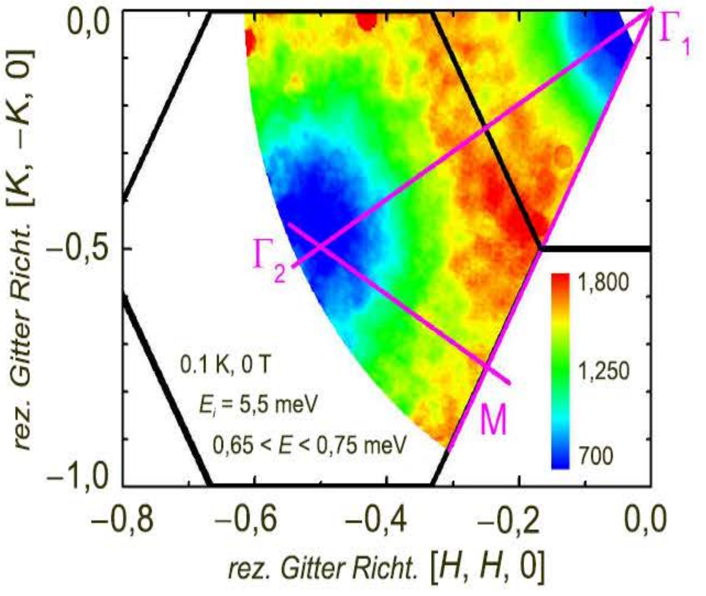
Broad excitations observed between 2 and $10J_0$ ($J_0 \approx 0.2$ meV) (cf. also Paddison et al., Nature Physics 13, 117–122 (2017))

Contradiction to mobile fermionic excitations (spinons):
Xu, Y. et al.: Absence of magnetic thermal conductivity (Phys. Rev. Lett. 117, 267202 (2016))

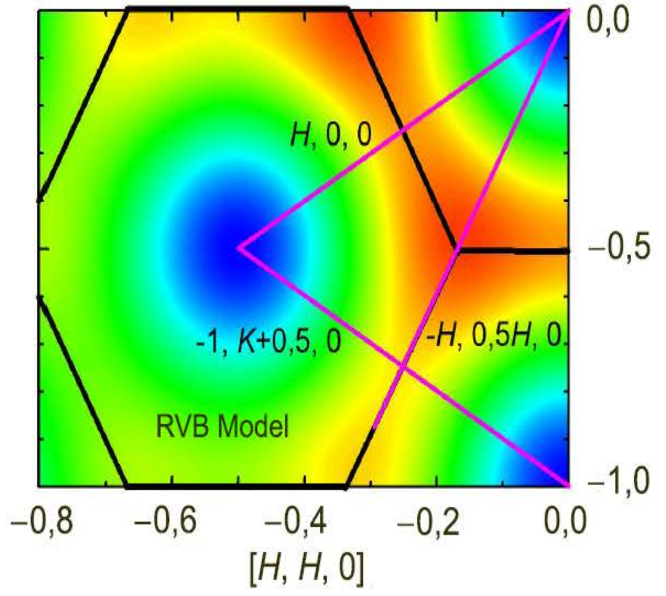


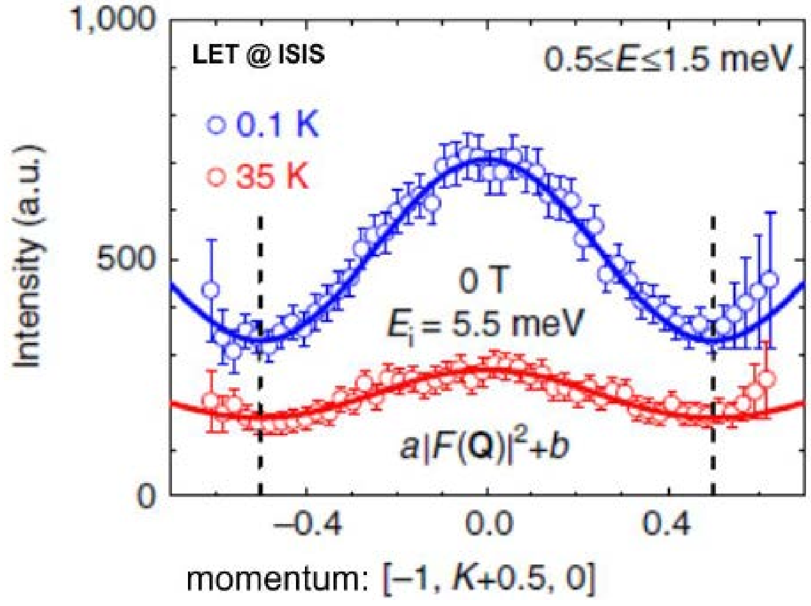
ISIS LET data for $YbMgGaO_4$

experiment



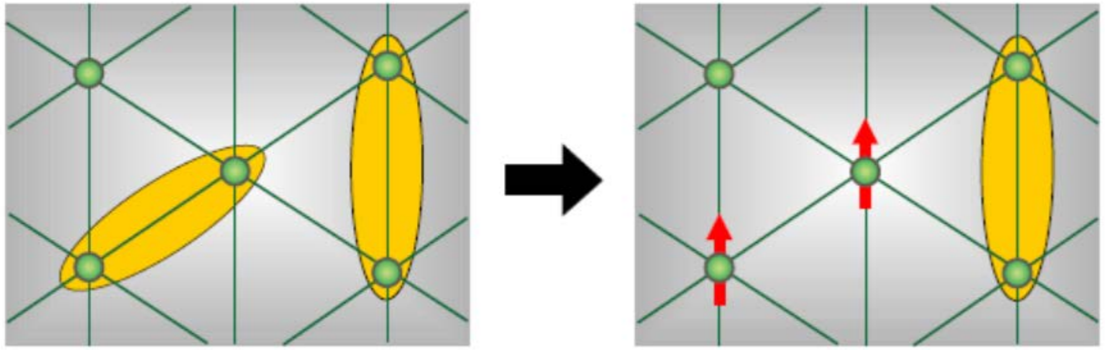
model calculation





- continuum persists up to 35 K and 2 meV
- $N|F(\mathbf{Q})|^2 = \frac{2}{3} N|f(\mathbf{Q})|^2 \{3 - \cos(2\pi H) - \cos(2\pi K) - \cos[2\pi(H+K)]\}.$

$\rightarrow$ breaking of nearest-neighbor valence bonds
on triangular lattice

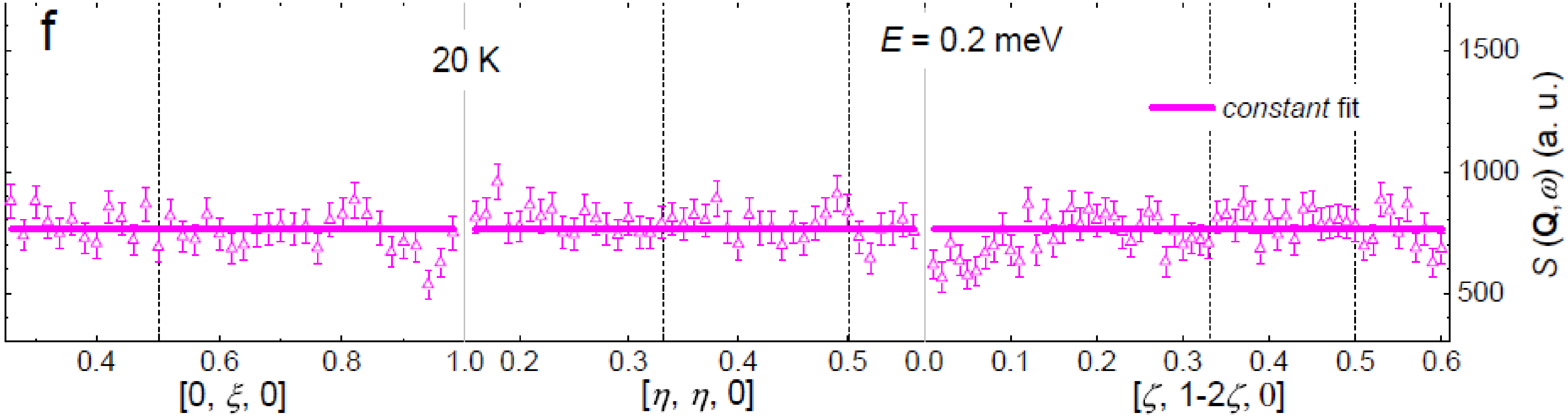


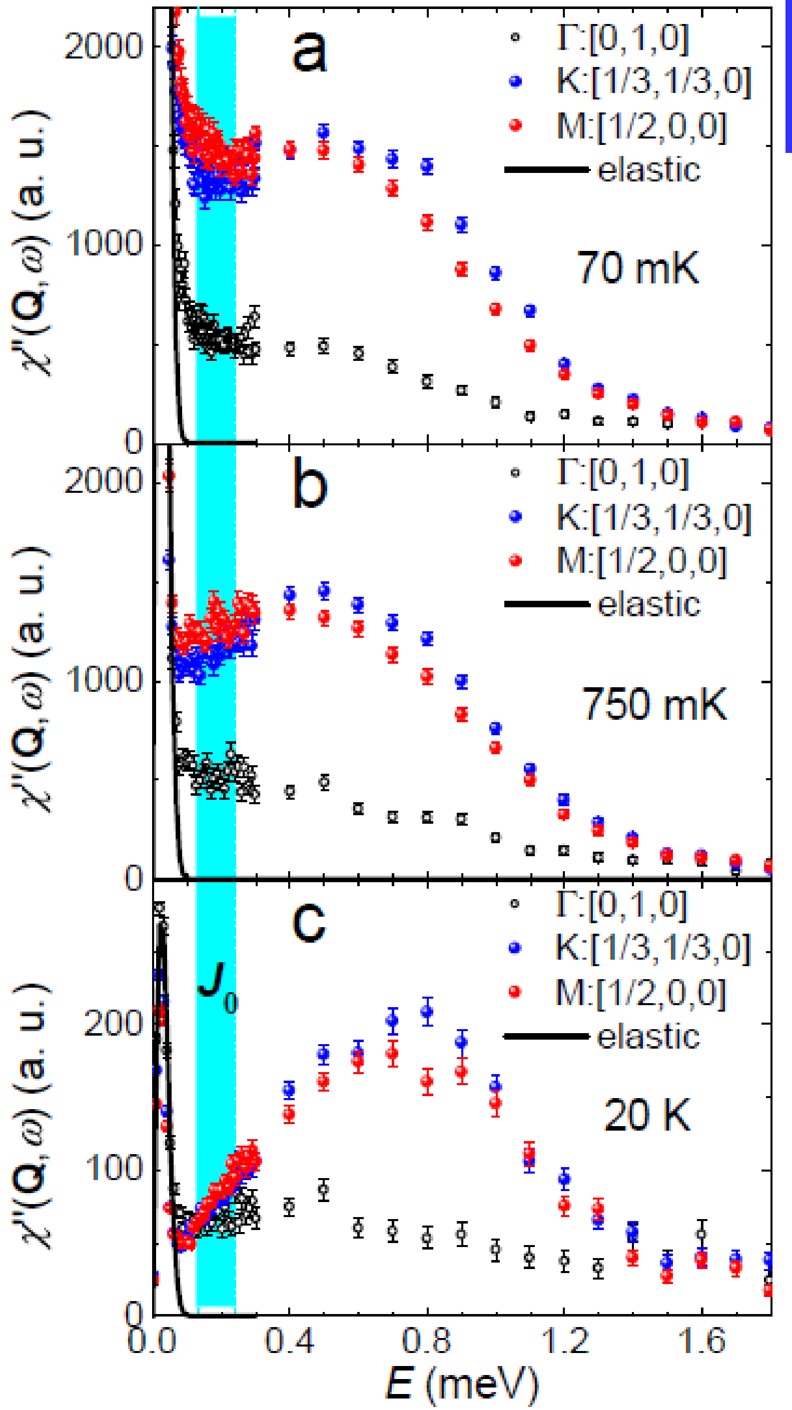
$\rightarrow$ expect different excitations at lower energy

Yuesheng Li, D. Adroja, D. Voneshen, R.I. Bewley, Q. Zhang, A.A. Tsirlin, P. Gegenwart, Nat. Commun. **8**, 15814 (2017)

Cold triple axes data (PANDA

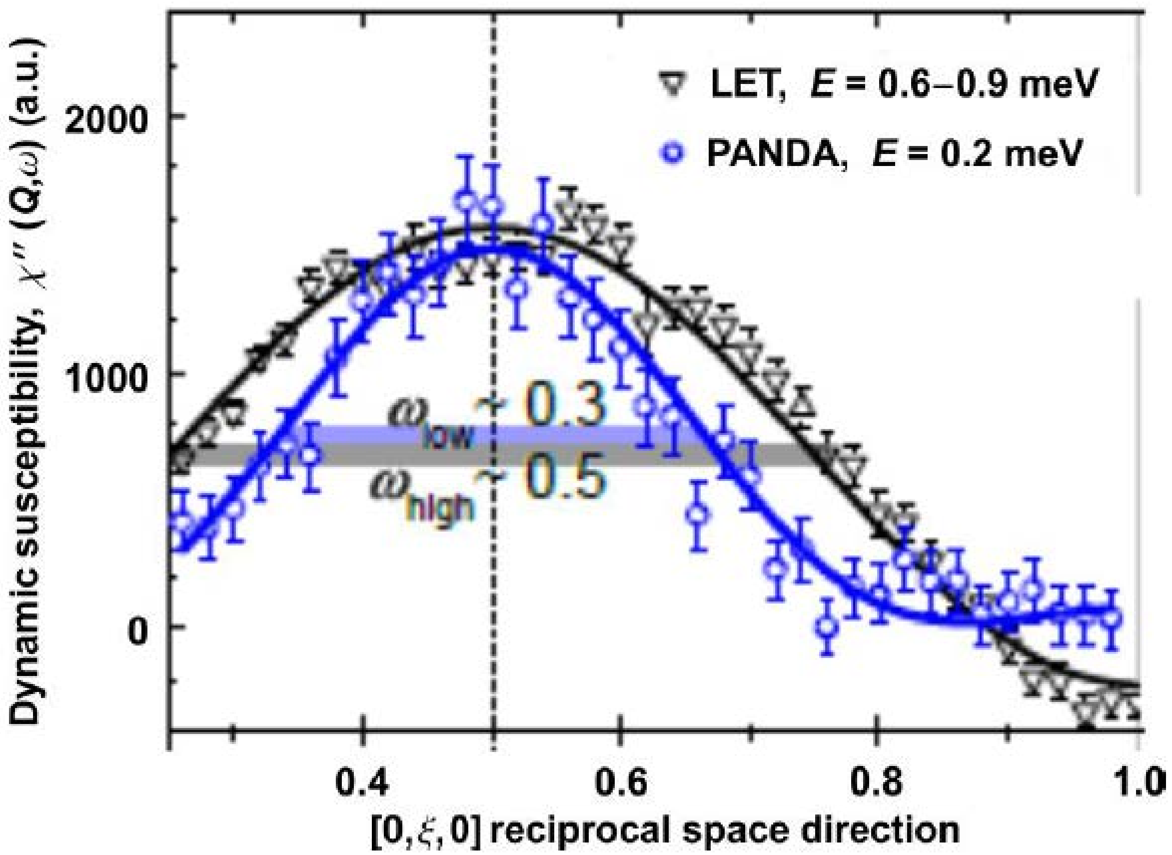
Q-dependence from form factor anisotropy (cf. Tóth et al)?
→ low-energy 20 K data show no q-dependence (form factor E-independent)





- gapless low-energy excitations at low T (consistent with heat cap and μSR relaxation rate)
- Nature of low-energy excitations??
  → analysis of q-dependence at 0.2 meV…

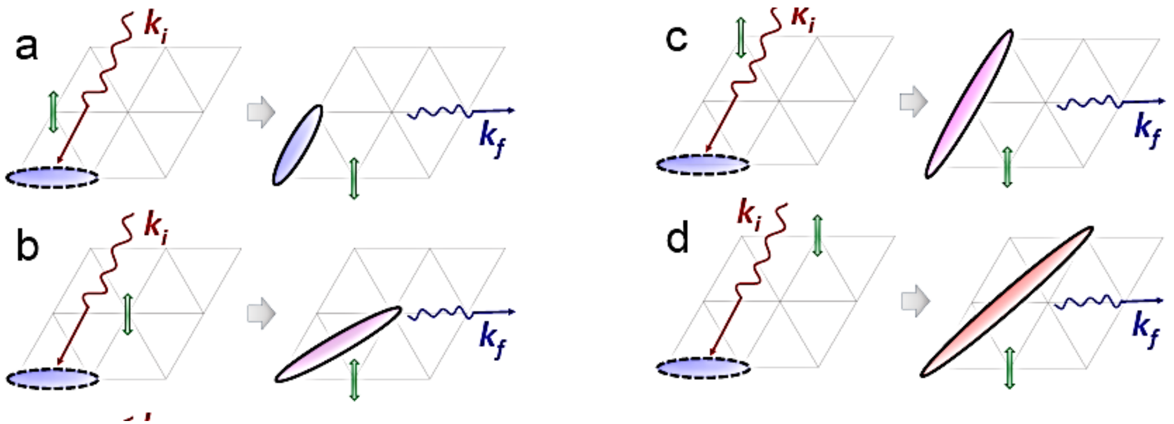
Low-energy INS on $YbMgGaO_4$



Narrowing of q-dep
→ increase in correlation length of excitations

$$S(\mathbf{Q}) = A \sum_{j=1}^{\infty} \sum_{l=1}^{Z_j} \frac{f_j}{Z_j} \sin^2\left(\frac{\mathbf{Q}\cdot\mathbf{R}_l^j}{2}\right) + B$$

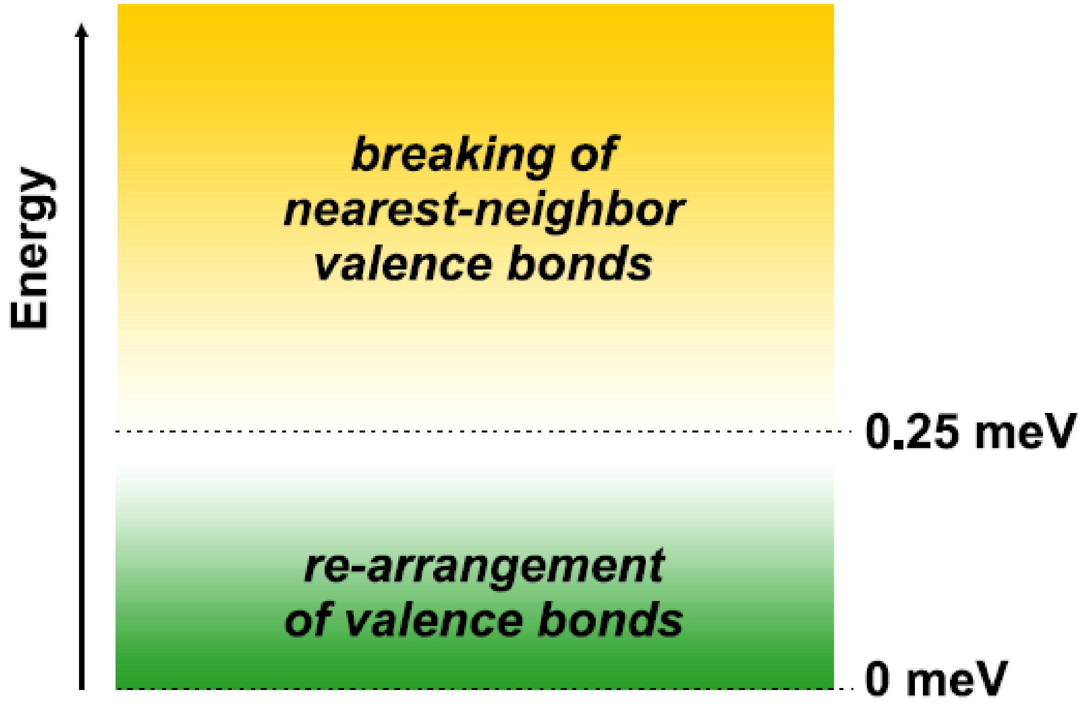
equal time correlation function, f_j: from j^{th} neighbor

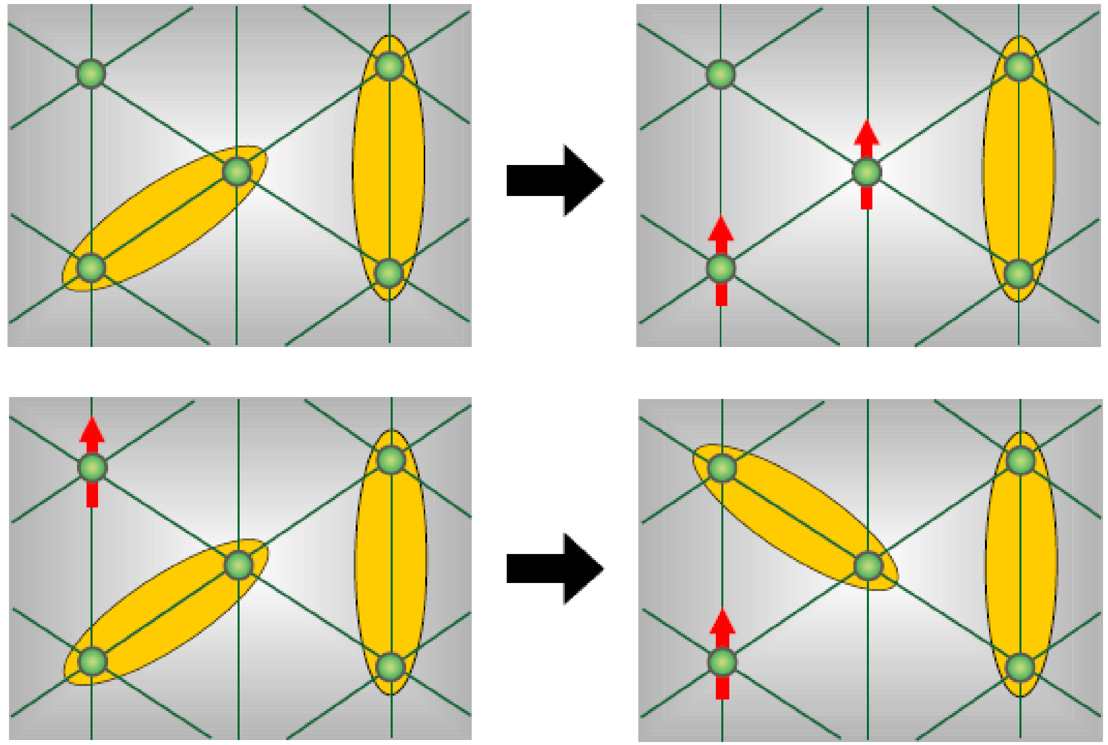


→ re-arrangement of valence bonds & propagation of unpaired spins

Y. Li, S. Bachus, B. Liu, I. Radelytskyi, A. Bertin, A. Schneidewind, Y. Tokiwa, A.A. Tsirlin, P. Gegenwart, PRL **122**, 137201 (2019)

Complete excitation spectrum of $YbMgGaO_4$

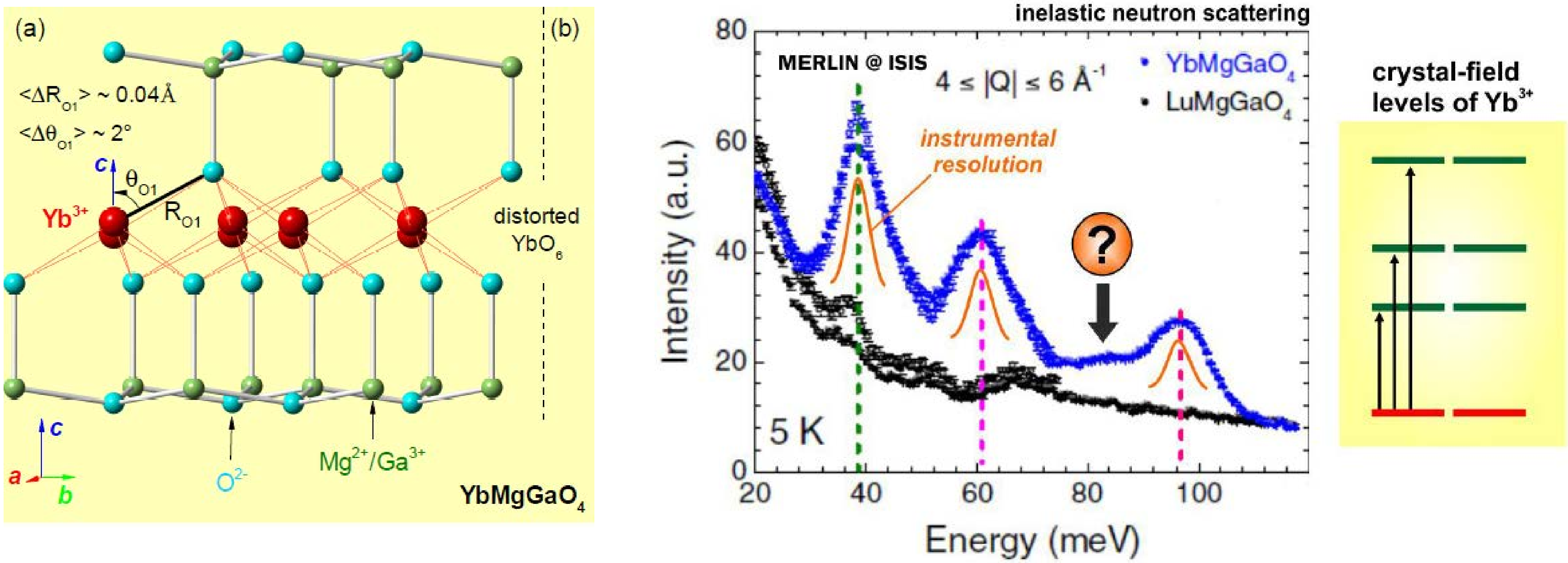




- First direct observation of valence bonds on triangular lattice
- Re-arrangement of VBs reminds to spinons, however, they must be localized (absence of thermal conductivity)

Y. Li et al., Nat. Commun. **8**, 15814 (2017) and Phys. Rev. Lett. **122**, 137201 (2019)

CEF randomness in $YbMgGaO_4$



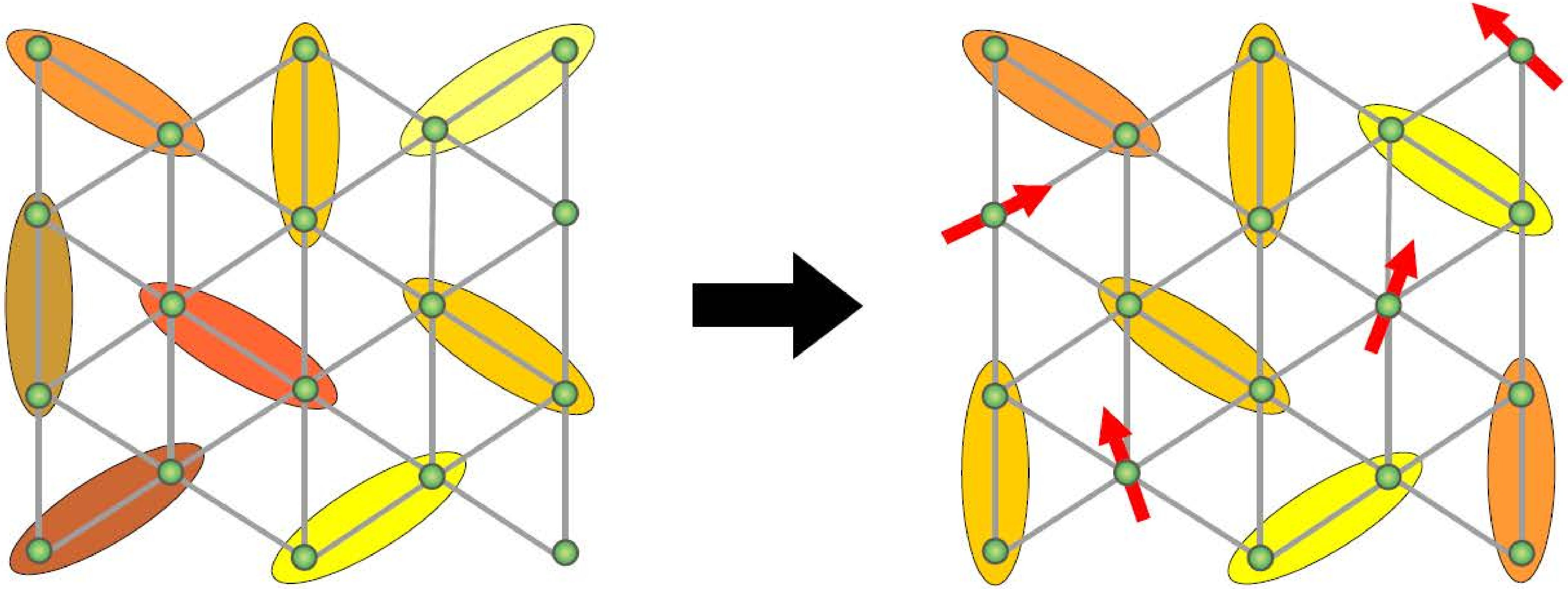
- Randomness in Yb environment (Mg^{2+}/Ga^{3+}), distorted YbO_6 octahedra
  $\rightarrow$ broadening of CEF excitations gives rise to distribution of *g*-values ($g_\perp = 3.2 \pm 0.15$, $g_{||} = 3.45 \pm 0.6$)
  and probably also exchange interactions

Y. Li, D. Adroja, R.I. Bewley, D. Voneshen, A.A. Tsirlin, P. Gegenwart, Q. Zhang, PRL **118**, 107202 (2017)

PHYSICAL REVIEW X **8**, 031028 (2018)

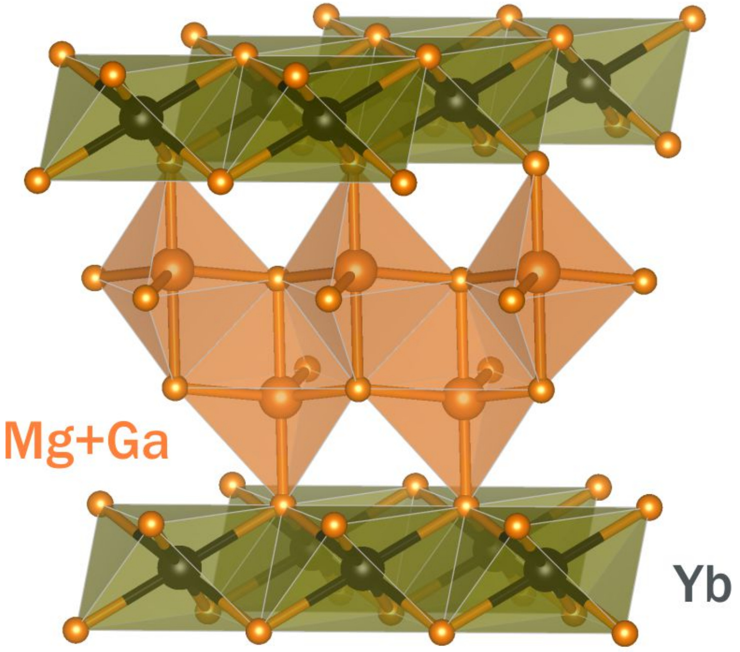
Valence Bonds in Random Quantum Magnets: Theory and Application to $YbMgGaO_4$

Itamar Kimchi,[1,*] Adam Nahum,[1,2] and T. Senthil[1]

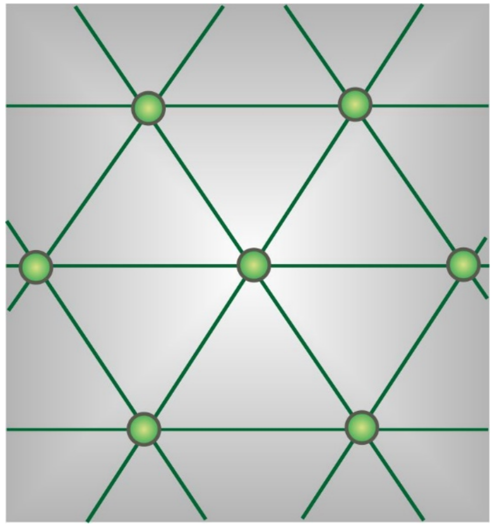


- Valence-bond magnets unstable against the disorder, nucleation of defects (carrying S=1/2)
- Structural randomness enables observed excitations at low energy?!

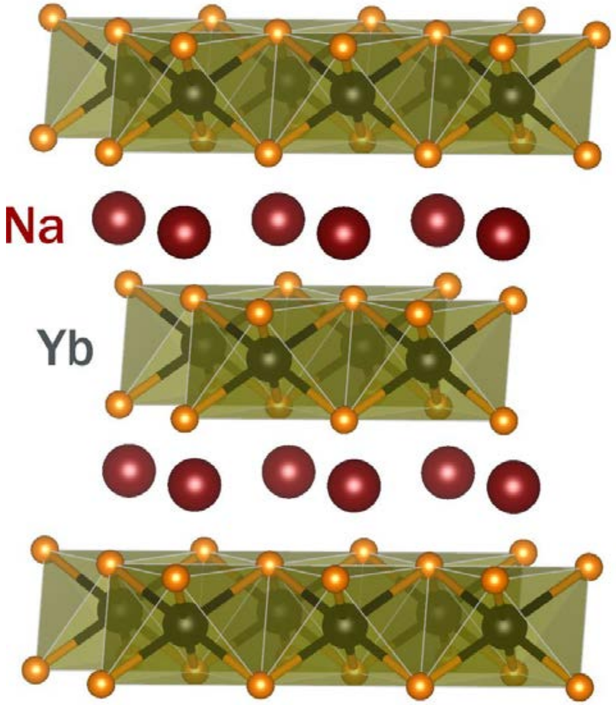
getting rid of structural disorder…



$YbMgGaO_4$



triangular lattice of Yb^{3+}

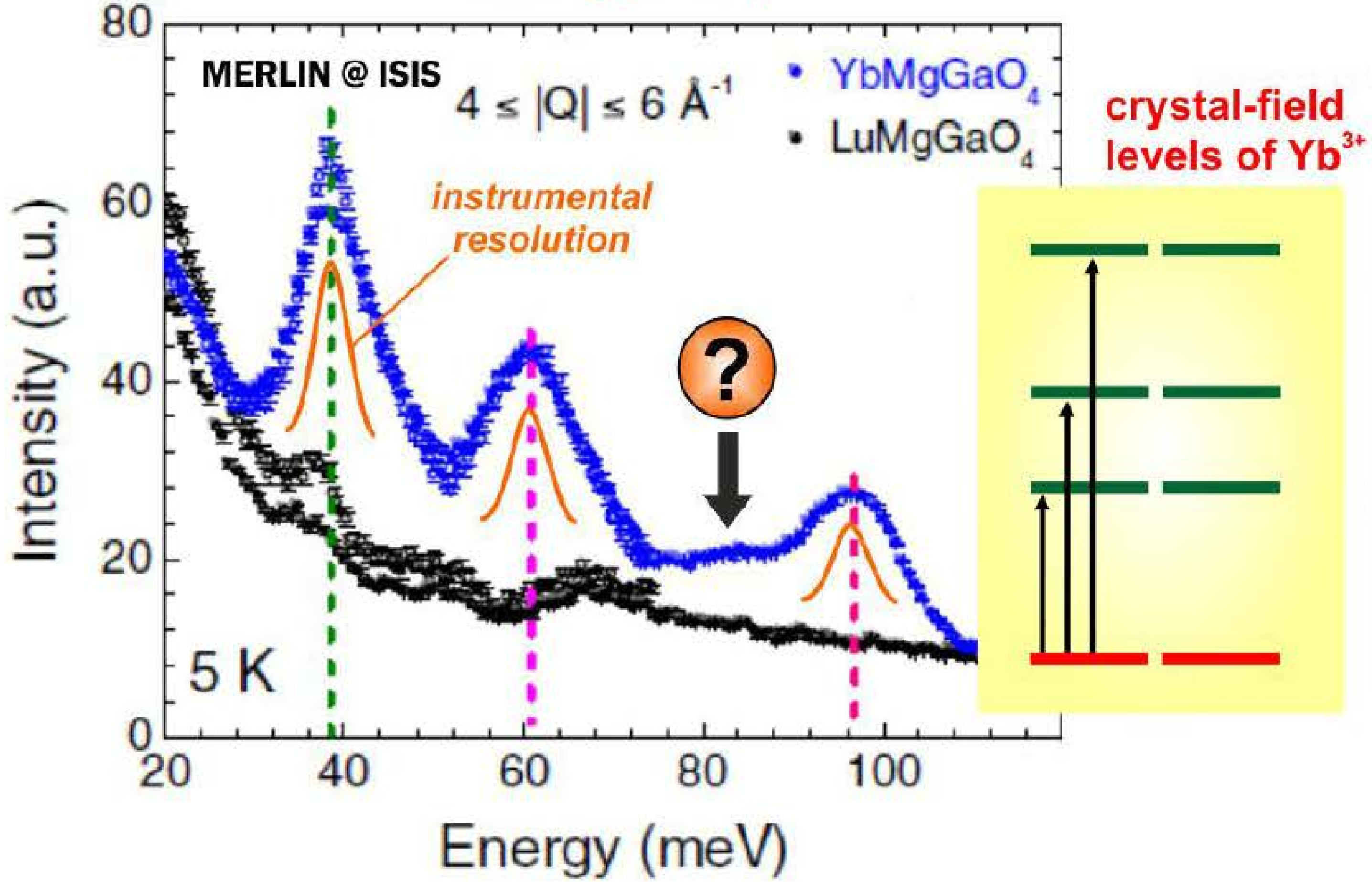


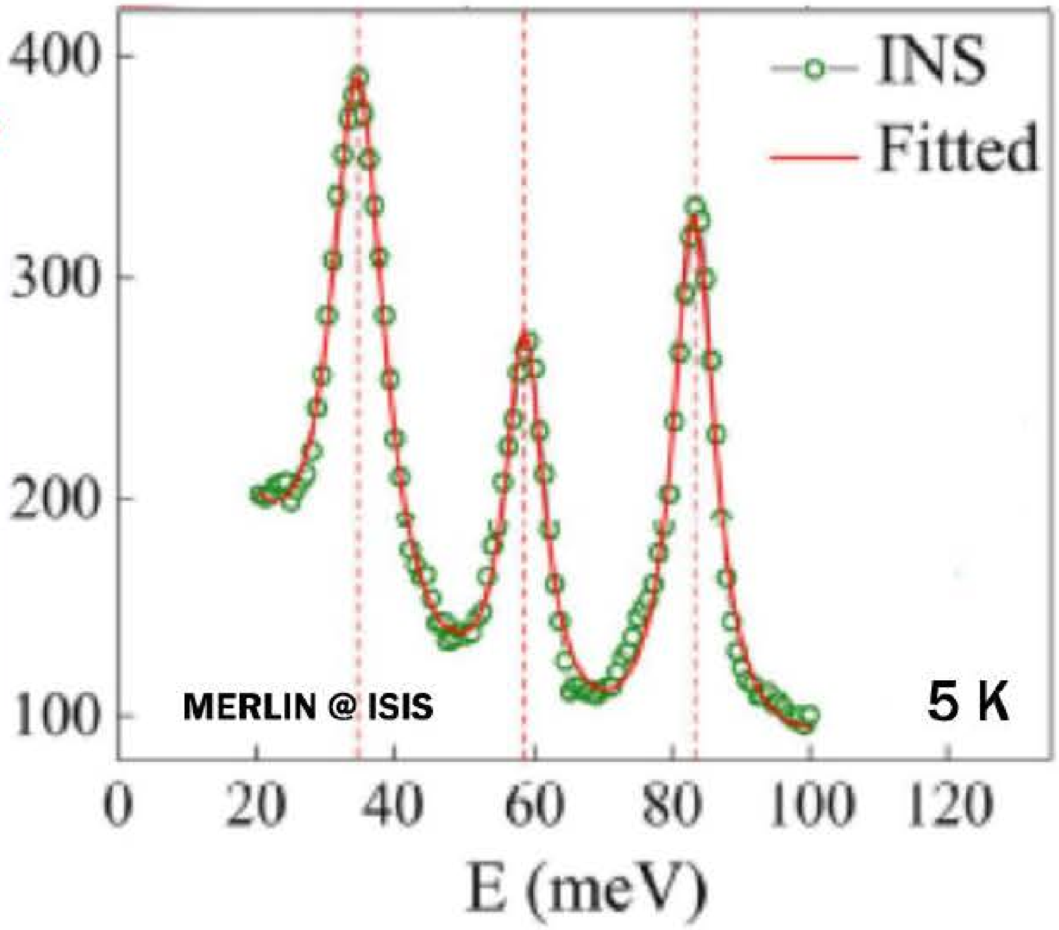
$NaYbO_2$

Same triangular layers of Yb^{3+}, replacing Mg/Ga by Na eliminates the structural disorder

L. Ding, P. Manuel, S. Bachus, F. Grußler, P. Gegenwart, J. Singleton, R.D. Johnson, H.C. Walker, D.T. Adroja, A.D. Hillier, A.A. Tsirlin, arXiv:1901.07810

$YbMgGaO_4$ $NaYbO_2$





- Similar triangular Yb-lattice but no site randomness → sharp CEF excitations
- Absence of order in zero field, broad excitations, however, only polycrystals available yet…

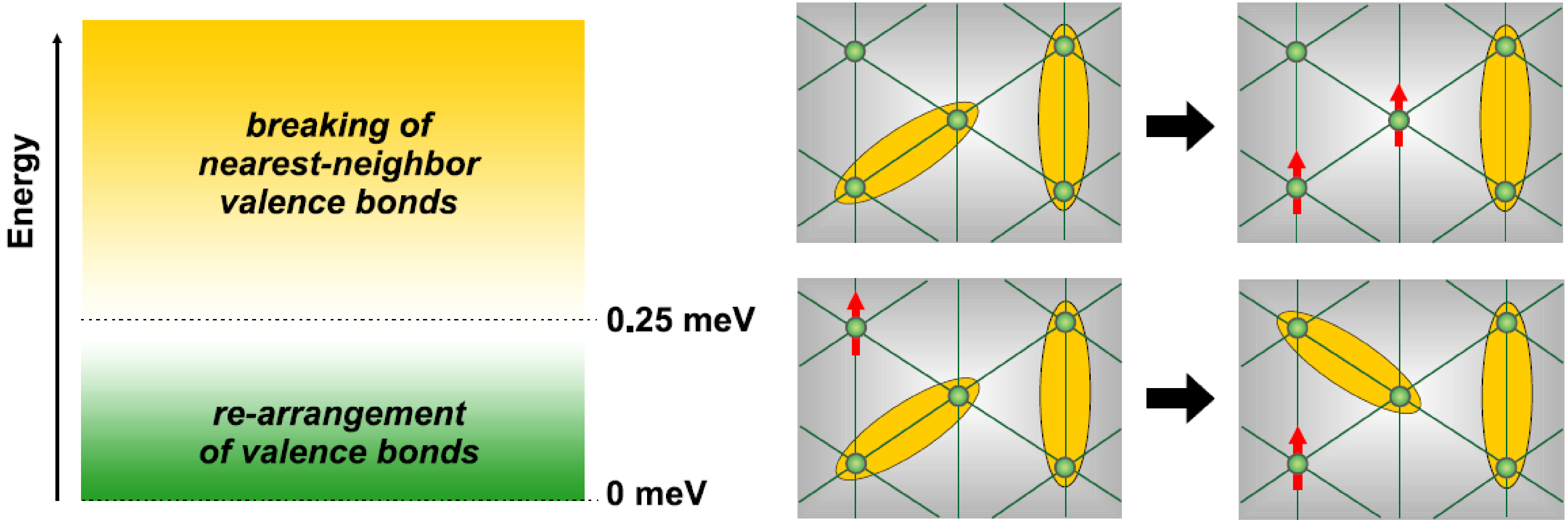
L. Ding, P. Manuel, S. Bachus, F. Grußler, P. Gegenwart, J. Singleton, R.D. Johnson, H.C. Walker, D.T. Adroja, A.D. Hillier, A.A. Tsirlin, arXiv:1901.07810

Summary

- Frustrated magnets promise novel quantum states with fractionalized excitations
- Triangular AF $YbMgGaO_4$: low-energy excitations indicating valence bond breaking and re-arranging, however, structural randomness prevents truly mobile spinons

→ Comparison with disorder-free triangular rare-earth magnets

→ Application of such materials for efficient and easy adiabatic demag cooling



Thanks

Yuesheng Li

Alexander Tsirlin

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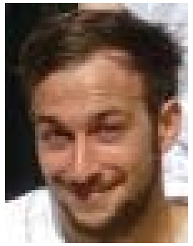
Anton Jesche



Sebastian Bachus

Kavita

Yoshi Tokiwa

Astrid Schneidewind

<u>External collaborators</u>:
Adroja, Baenitz, Schneidewind, Zhang, …and their groups

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