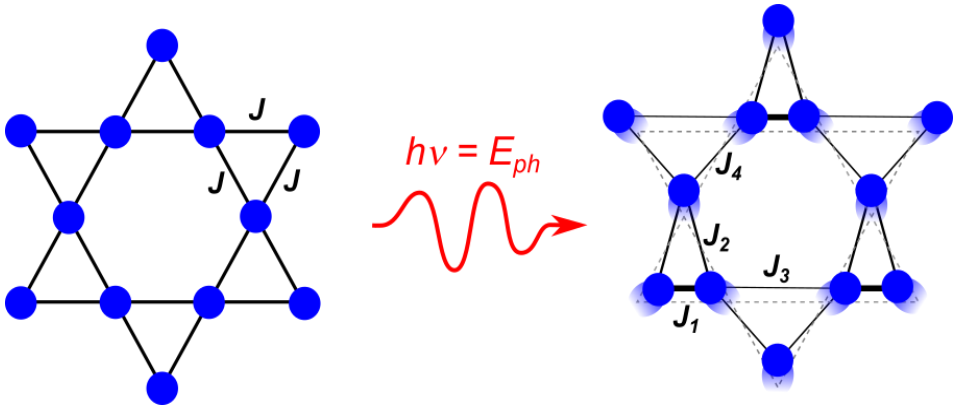
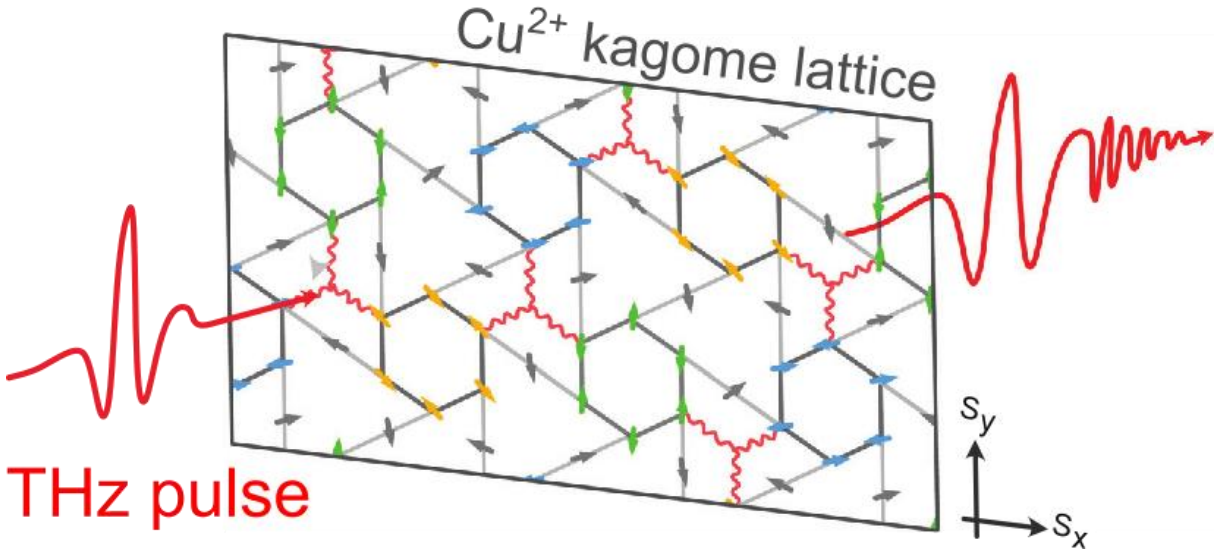


Magnetoelastic Coupling and Terahertz Magnetometry of Kagome Systems

Andrej Pustogow

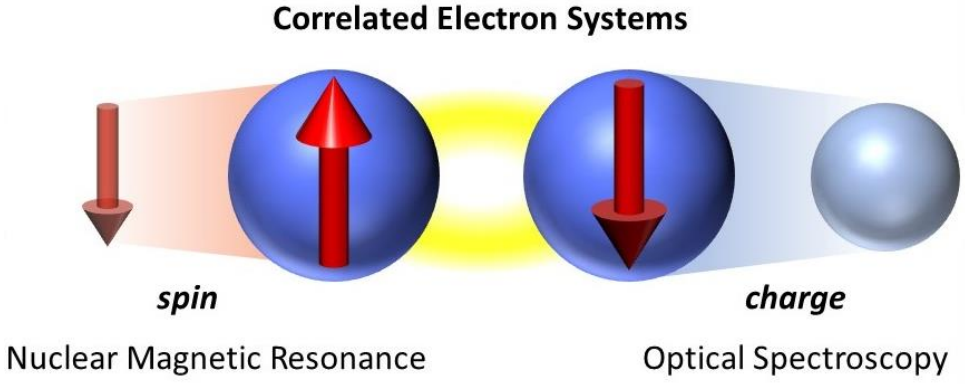
Institute of Solid State Physics
TU Wien, Austria



Collaboration

Alexander von Humboldt Stiftung/Foundation

Andrej Pustogow
TU Wien, Austria



Pascal Puphal
MPI Stuttgart, Germany

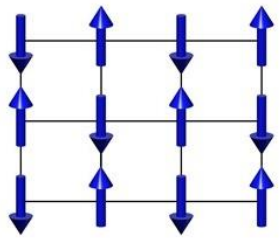
Igor I. Mazin
George Mason Univ., USA

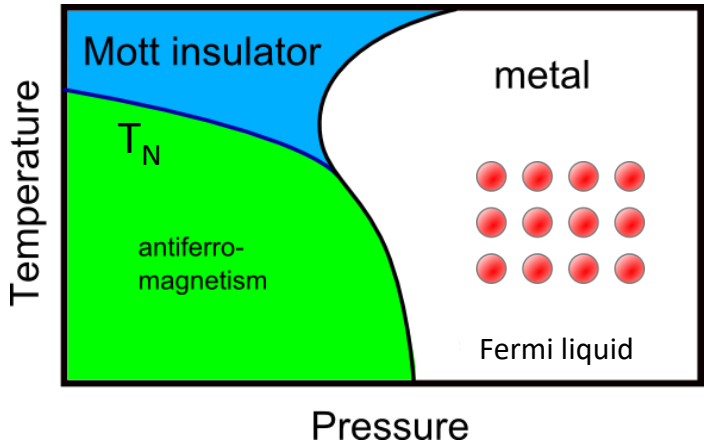
Ying Li
Roser Valentí
Cornelius Krellner
Goethe Univ., Frankfurt, Germany

Tobias Biesner
Seulki Roh
Martin Dressel
1. Physikalisches Institut
Universität Stuttgart

Pustogow Spectroscopy Lab

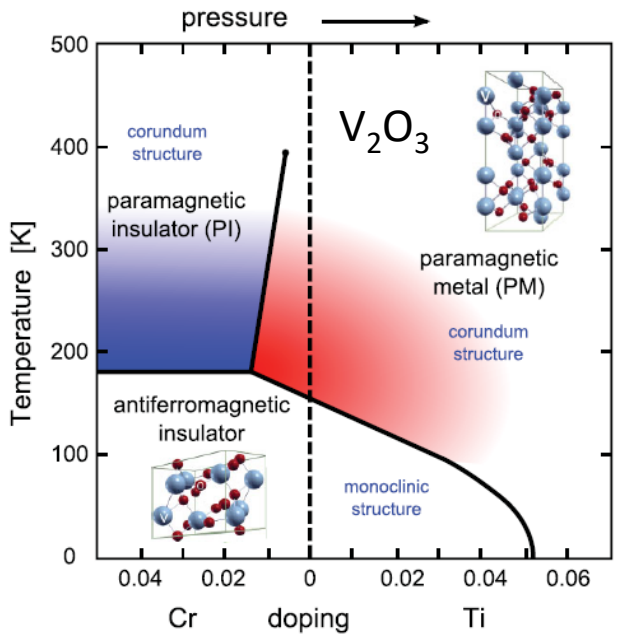
Magnetism in Mott insulators

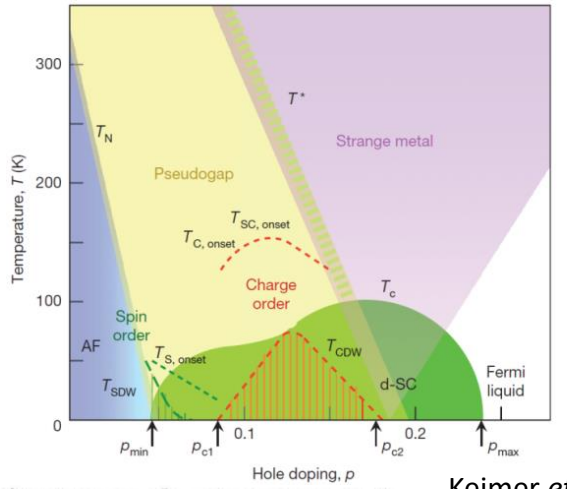




$$J \propto \frac{t^2}{U}$$

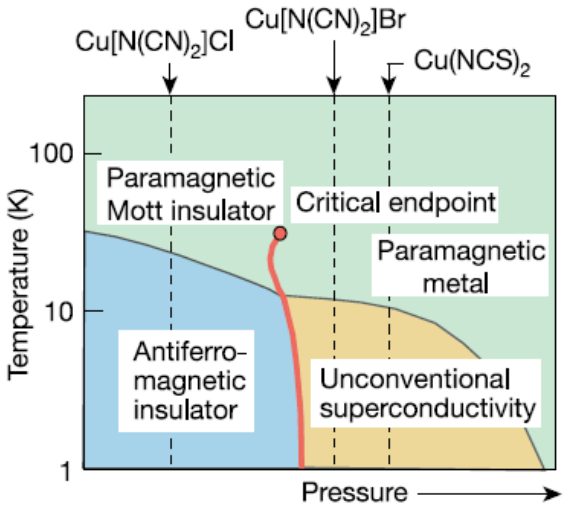
Hansmann *et al.*, *Phys. Status Solidi B* **250**, 1251–1264 (2013)





High-T_c Cuprates

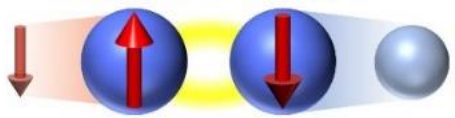
Keimer *et al.*, *Nature* **518**, 179 (2015)



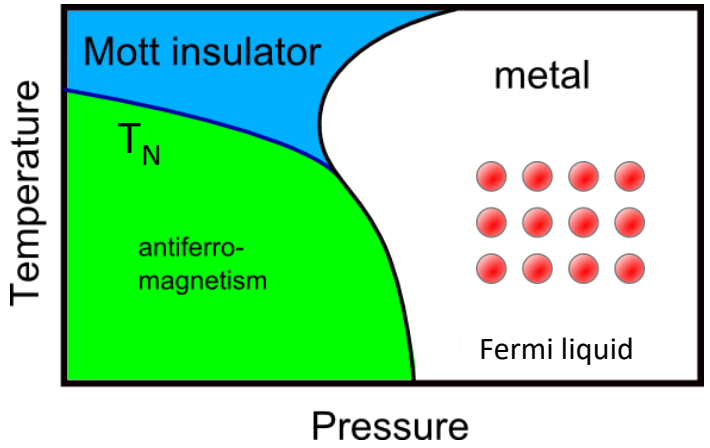
Organic Conductors

Kagawa *et al.*, *Nature* **436**, 534 (2005)

Pustogow Spectroscopy Lab

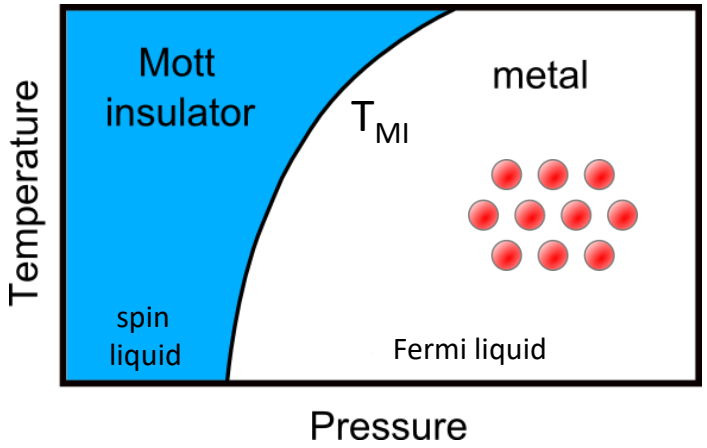


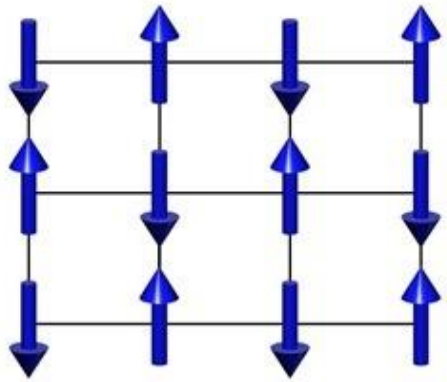
Magnetism in Mott insulators

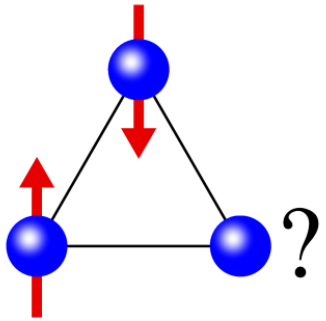


$$J \propto \frac{t^2}{U}$$

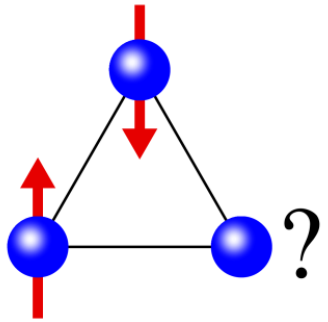
frustrated Mott insulator







Geometrical Frustration



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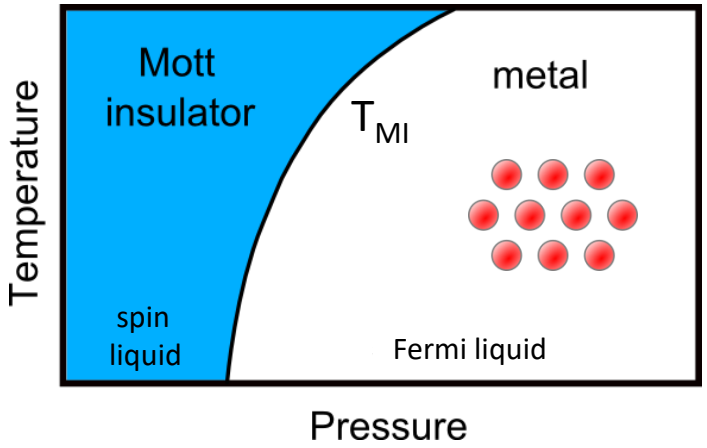


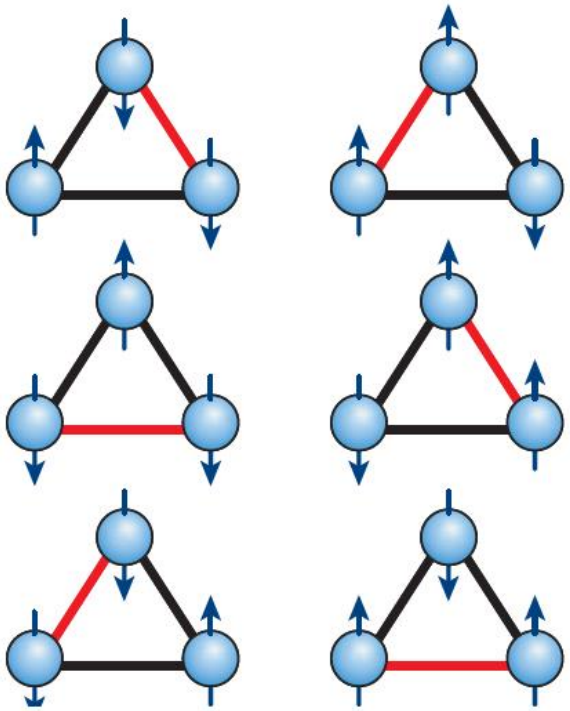
frustration in real life

geometrical frustration in solids

- suppression of magnetic order
- quantum spin liquid
- exotic excitations (spinons)

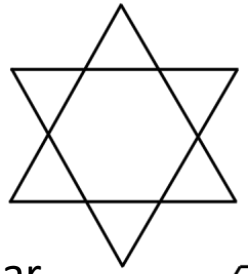
Geometrical Frustration

suppression of long-range order

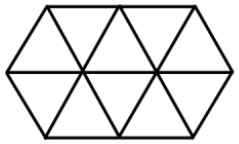


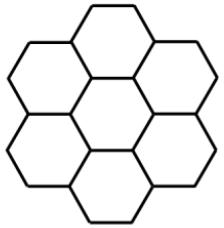
Balents, *Nature* **464**, 199–208 (2010)

kagome



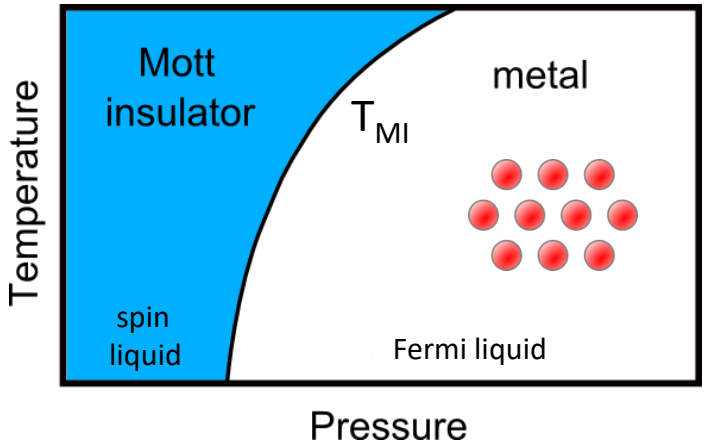
triangular

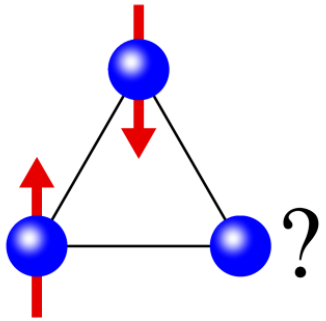




honeycomb

frustrated Mott insulator





geometrical frustration in solids

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- quantum spin liquid
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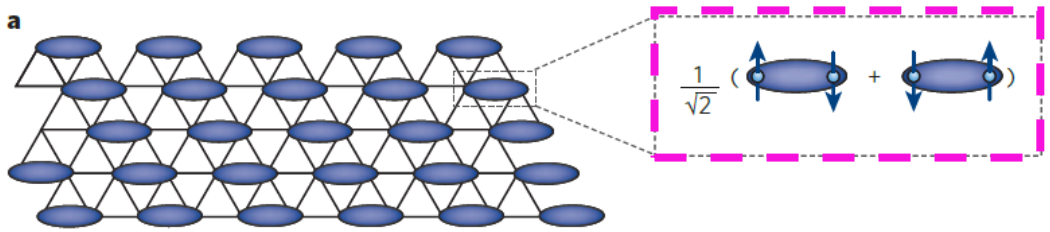
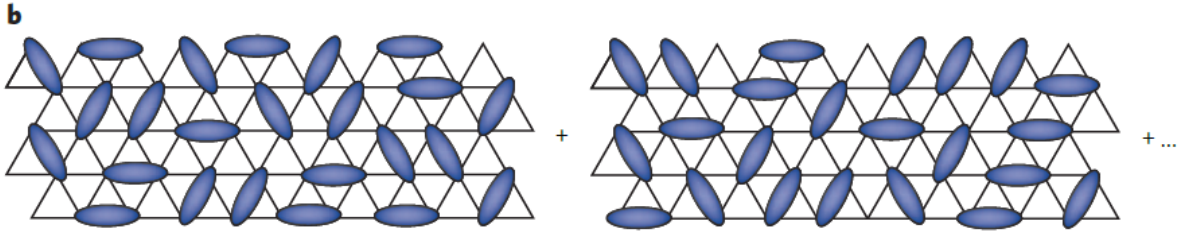
Frustrated Magnetism

Valence Bond Solid
(e.g. Spin-Peierls in 1D)



valence bond

Resonating Valence Bond (RVB) State



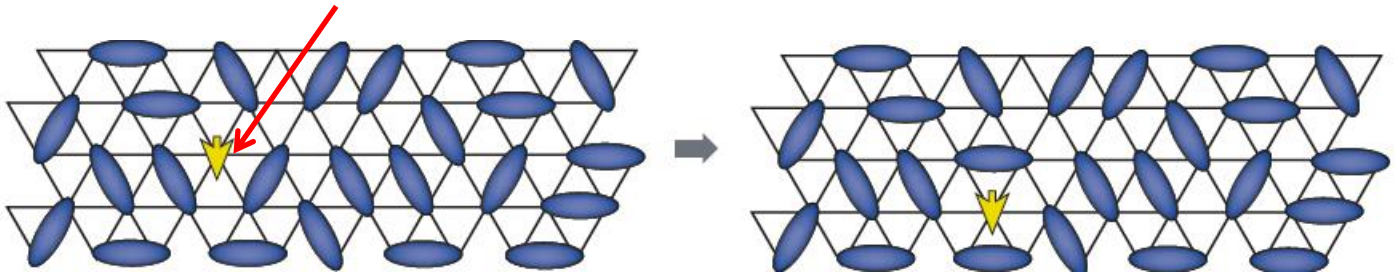
notion of a
Quantum-Spin-Liquid

RESONATING VALENCE BONDS: A NEW KIND OF INSULATOR?

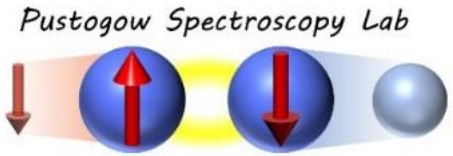
P. W. Anderson

Mater. Res. Bull. **8**, 153–160 (1973)

spinon: neutral $S = ½$ excitation

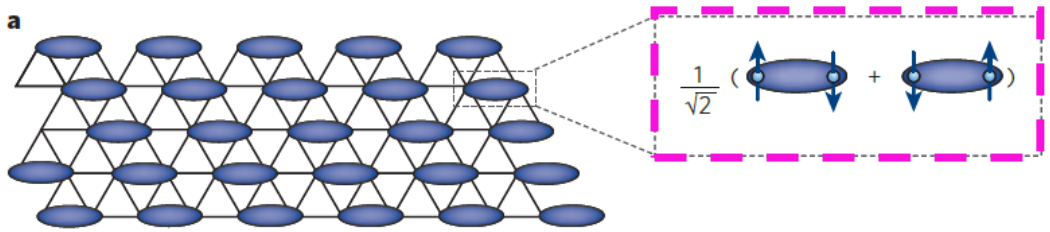


Balents, *Nature* **464**, 199–208 (2010)



Frustrated Magnetism

Valence Bond Solid
(e.g. Spin-Peierls in 1D)



valence bond

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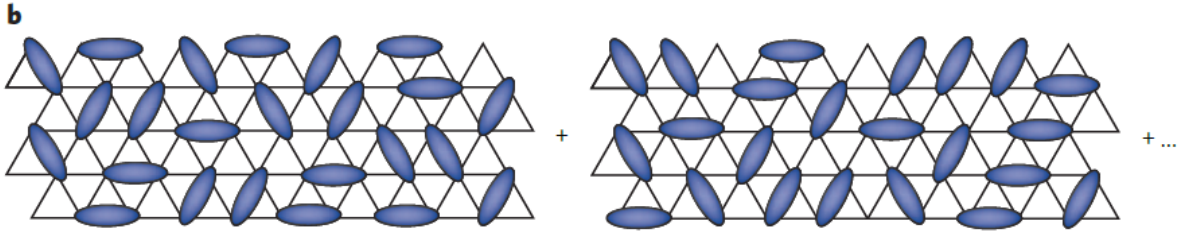


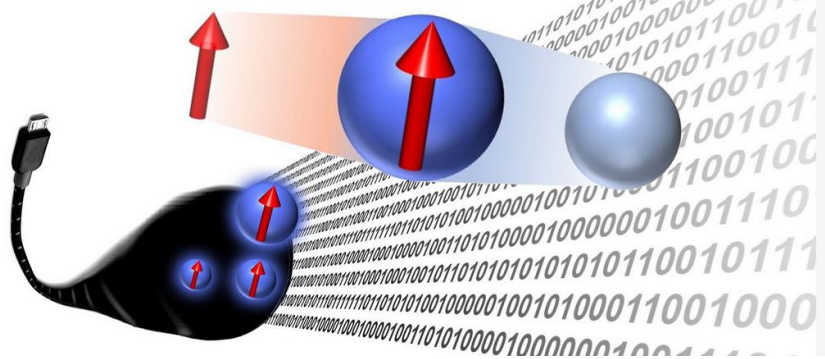
interesting for quantum information

26. April 2021

New measurements call spin liquids into question

Is it possible to transmit information through a material in the form of electron spins? New measurements show: not in the way that scientists had been working on for decades.

Miksch *et al.*, *Science* **372**, 276-279 (2021)

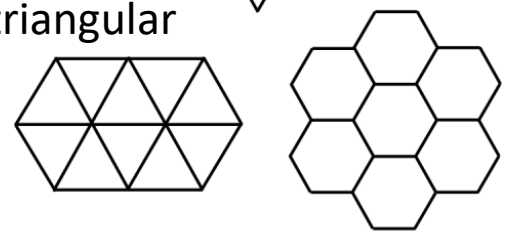


Spin Liquid Candidates

kagome

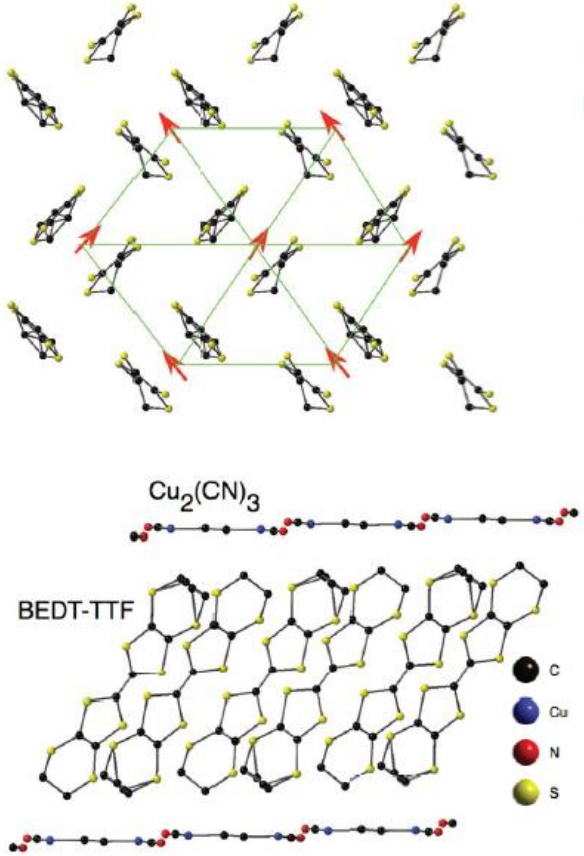
triangular

honeycomb



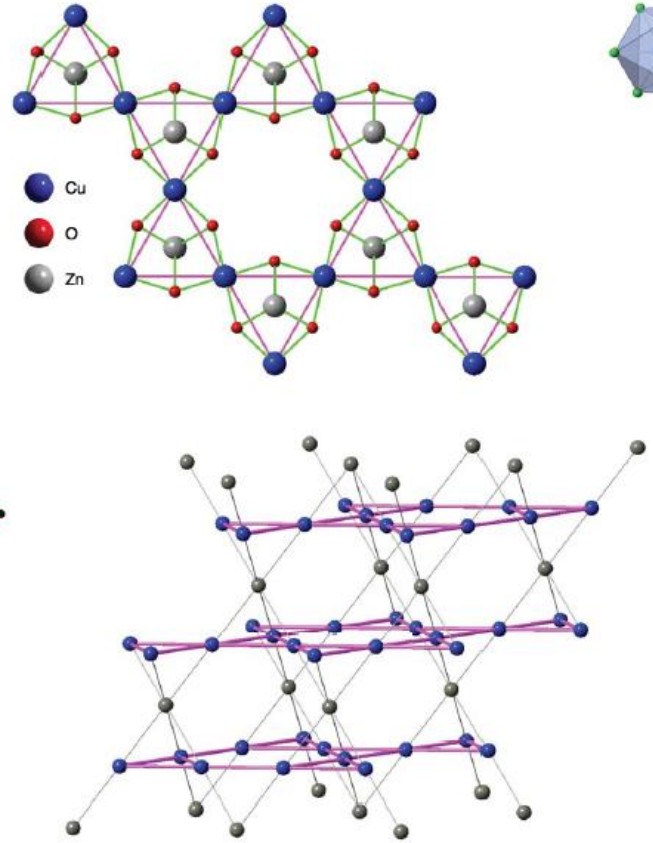
organic charge-transfer salts

A κ-$(ET)_2Cu_2(CN)_3$



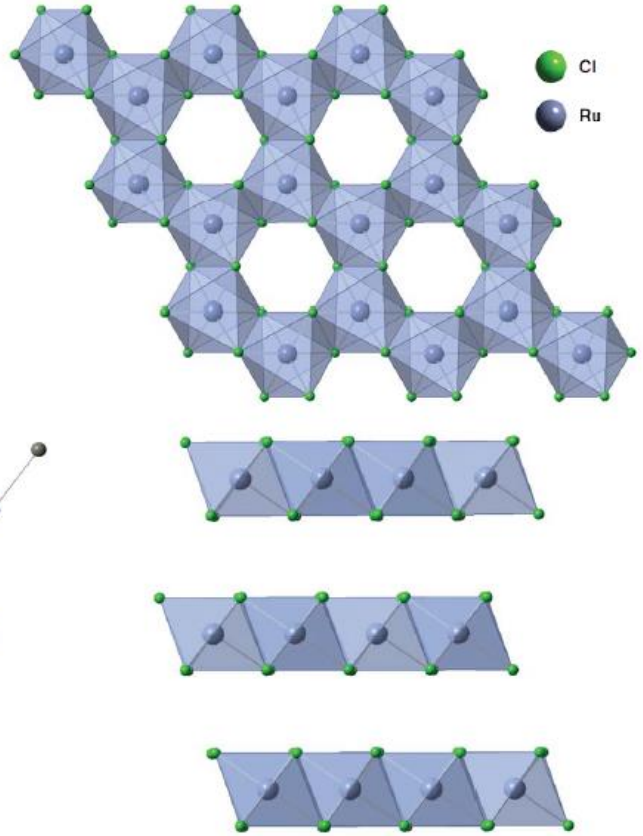
Herbertsmithite

B $ZnCu_3(OH)_6Cl_2$



van der Waals materials

C α-$RuCl_3$



Broholm *et al.*, *Science* **367**, eaay0668 (2020)

Herbertsmithite $ZnCu_3(OH)_6Cl_2$

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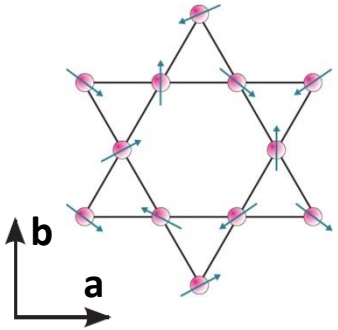
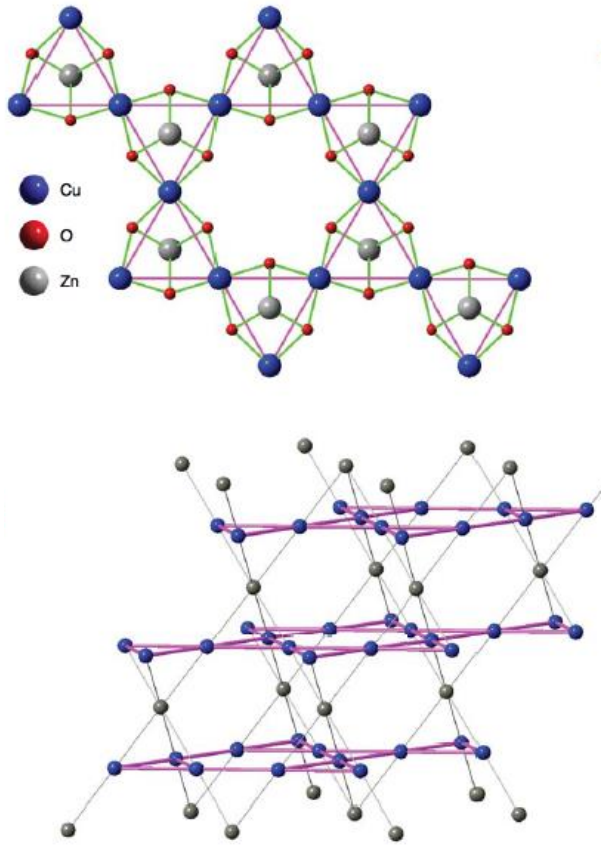
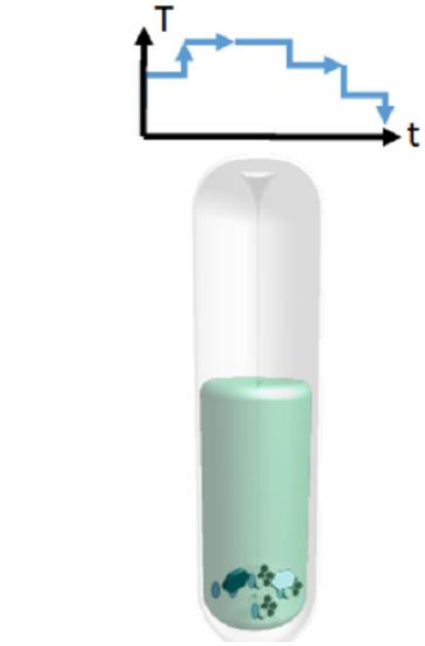
- $S = 1/2$ kagome lattice of Cu^{2+}
- suppression of magnetic order $0 \leq T_N \ll \theta_{CW} \approx 300K$
- exotic magnetic excitations?



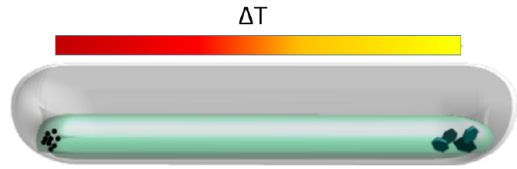
Herbertsmithite



Broholm *et al.*, *Science* **367**, eaay0668 (2020)



hydrothermal synthesis



Herbertsmithite $ZnCu_3(OH)_6Cl_2$

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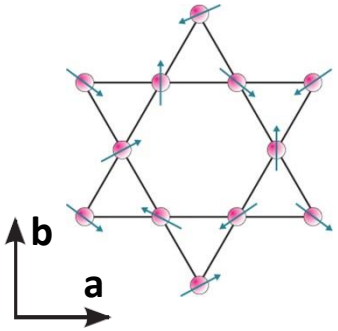
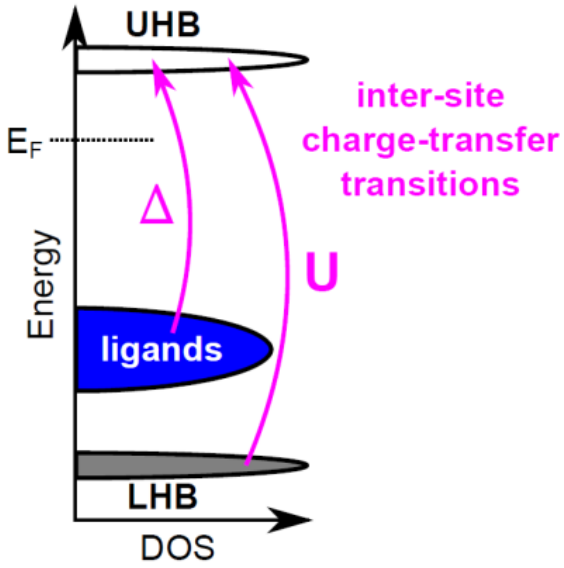
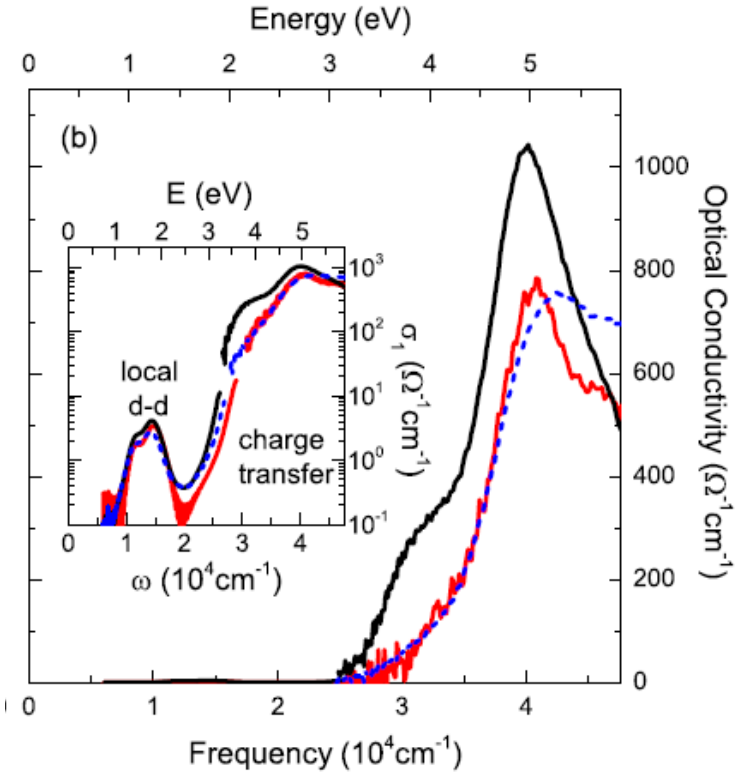




Pustogow *et al.*, *PRB* **96**, 241114(R) (2017)



Kagome Hydroxides XCu$_3$(OH)$_6$Cl$_2$

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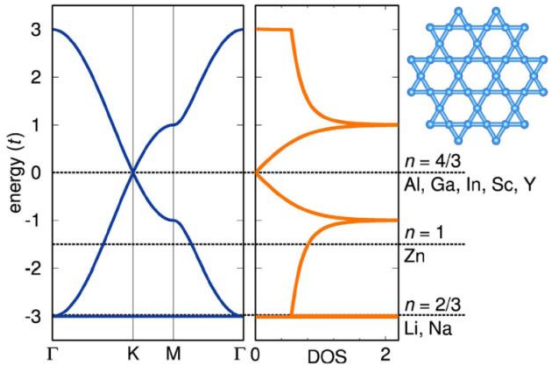

Jeschke *et al.*, *PRB* **88**, 075106 (2013)
Mazin *et al.*, *Nat. Comm.* **5**, 261 (2014)
Guterding *et al.*, *Sci. Rep.* **6**, 25988 (2016)

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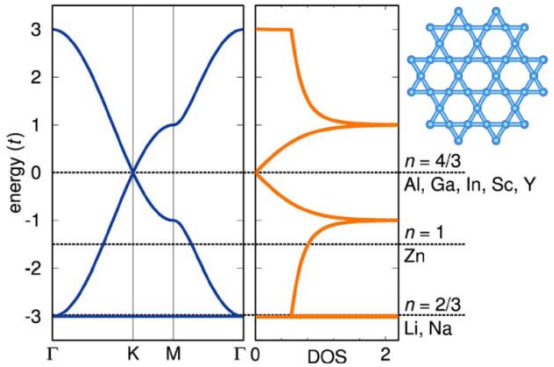
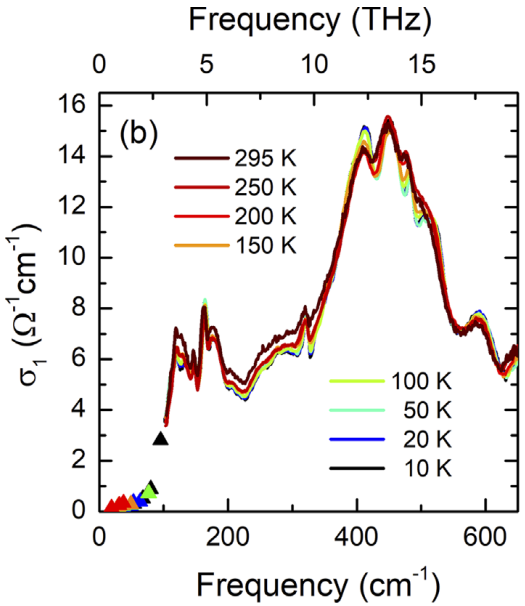
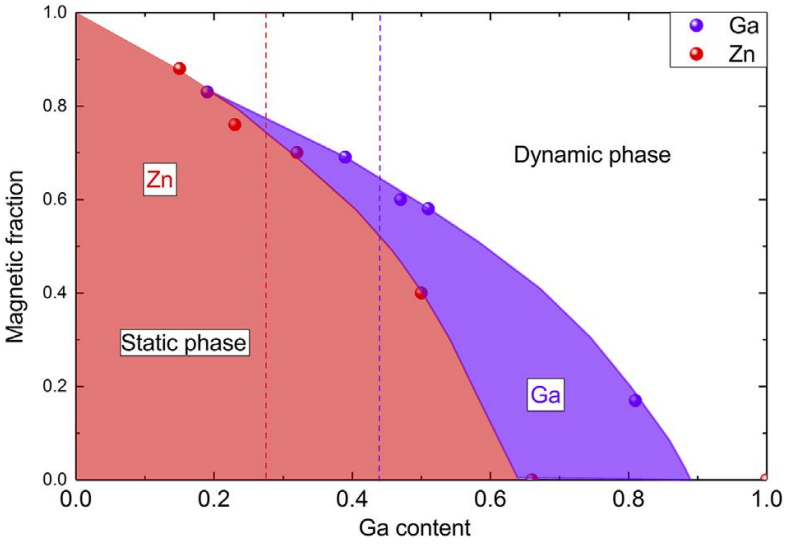




GaCu$_3$(OH)$_6$Cl$_2$

- suppression of magnetic order
- **insulating, not metallic!**
- additional charge upon $Zn^{2+} \rightarrow Ga^{3+}$ compensated by additional OH^-/Cl^-



Jeschke *et al.*, *PRB* **88**, 075106 (2013)
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Puphal *et al.*, *Phys. Status Solidi B* **2019**, 1800663

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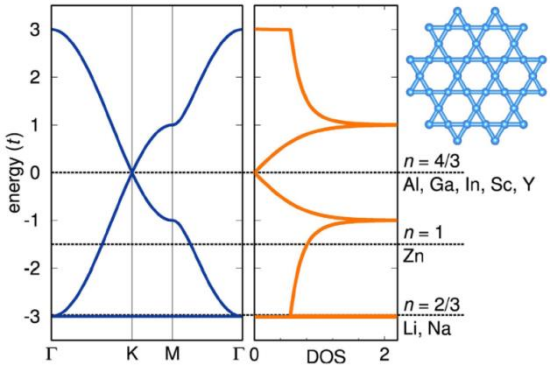

Jeschke *et al.*, *PRB* **88**, 075106 (2013)
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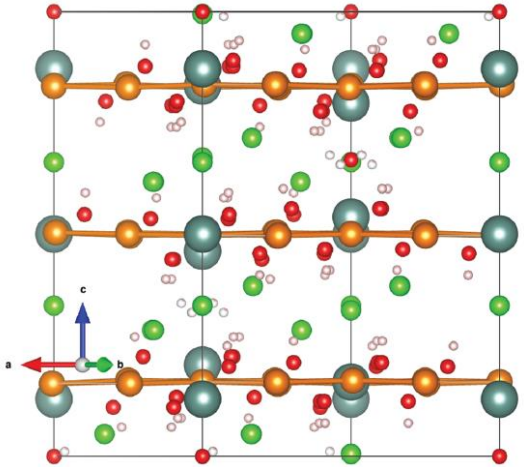
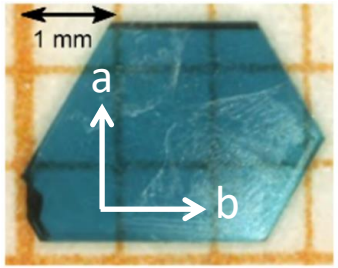
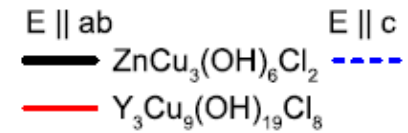
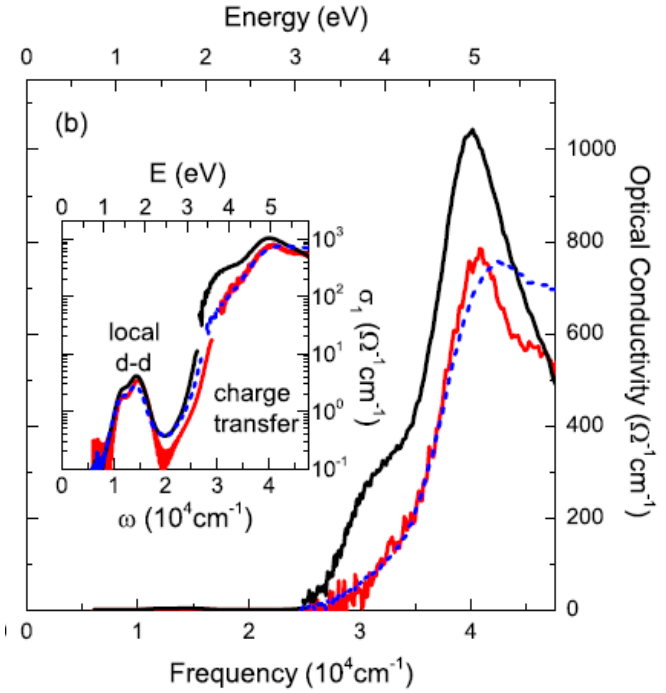




$Y_3Cu_9(OH)_{19}Cl_8$

- slightly distorted Cu^{2+} kagome lattice
- frustrated Mott insulator $\Delta = 3.6$ eV





Pustogow *et al.*, *PRB* **96**, 241114(R) (2017)
Puphal *et al.*, *J. Mater. Chem. C* **5**, 2629 (2017)

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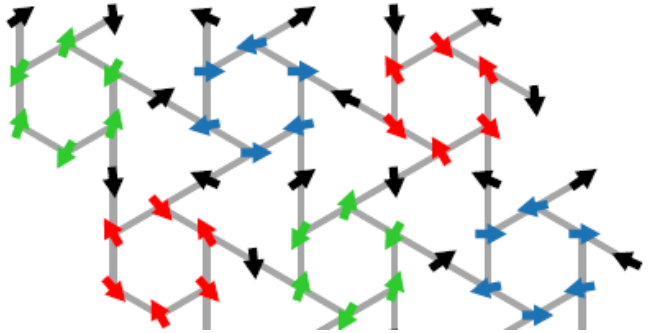
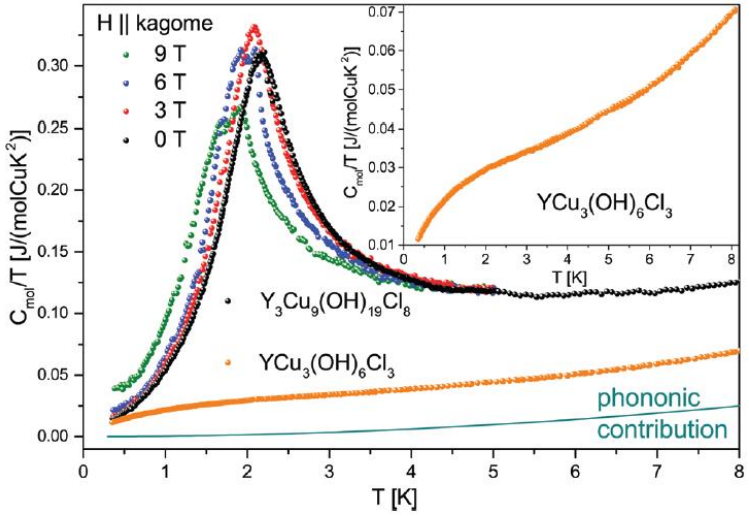
- slightly distorted Cu^{2+} kagome lattice
- frustrated Mott insulator $\Delta = 3.6$ eV
- $\vec{Q} = \left(\frac{1}{3} \times \frac{1}{3}\right)$ antiferromagnetic order

  $T_N = 2.2\ \mathrm{K} \ll \theta_{CW} \approx 100\ \mathrm{K}$



Chatterjee *et al.*, *PRB* **107**, 125156 (2023)
Biesner *et al.*, *Adv. Quantum Technol.* **2022**, 2200023
Hering *et al.*, *npj Comp. Mater.* **8**, 10 (2022)
Wang *et al.*, arXiv:2209.08613

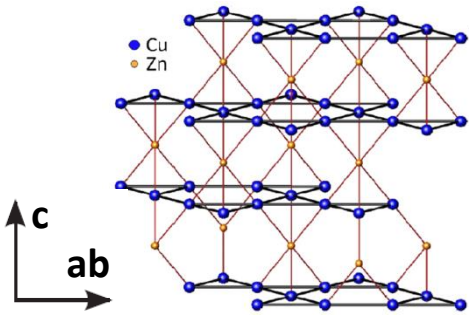


Puphal *et al.*, *J. Mater. Chem. C* **5**, 2629 (2017)

Kagome Hydroxides XCu$_3$(OH)$_6$Cl$_2$

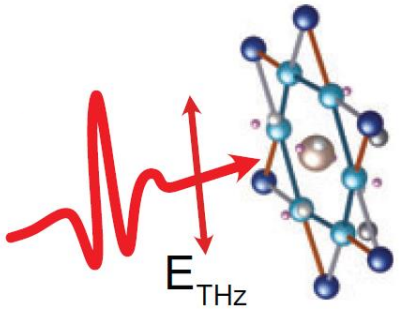
ZnCu$_3$(OH)$_6$Cl$_2$

- $S = 1/2$ kagome lattice of Cu^{2+}
- no magnetic order
- frustrated Mott insulator $\Delta = 3.3$ eV



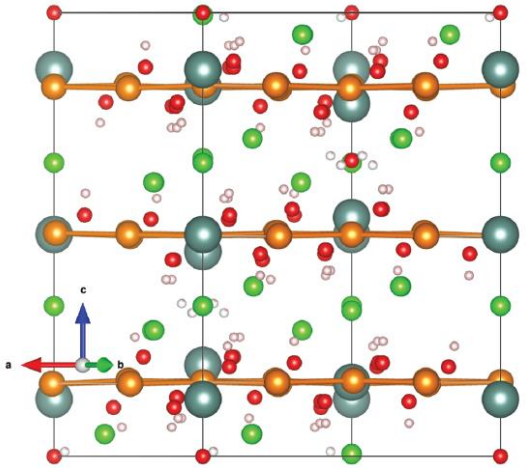
Pilon *et al.*, *PRL* **111**, 127401 (2013)

- **spin-phonon coupling**
- **magnetic THz response**



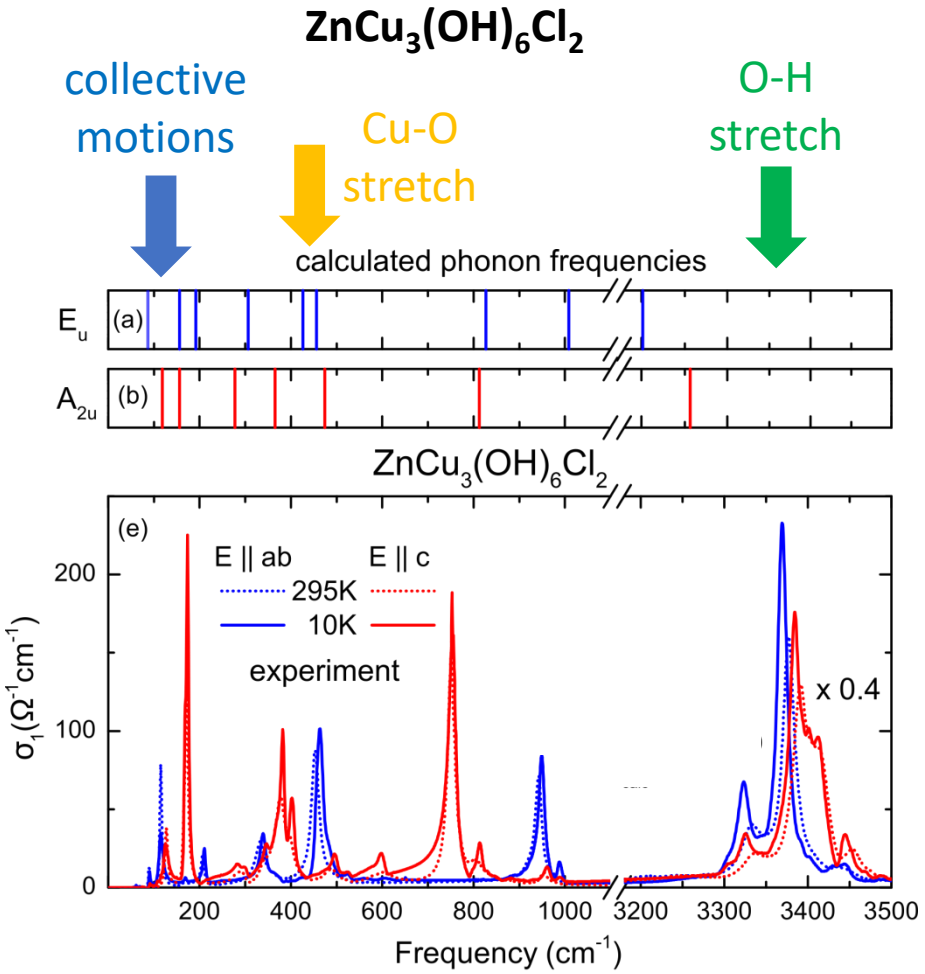
Y$_3$Cu$_9$(OH)$_{19}$Cl$_8$

- slightly distorted Cu^{2+} kagome lattice
- $T_N = 2.2\ \mathrm{K} \ll \theta_{CW} \approx 100\ \mathrm{K}$
- frustrated Mott insulator $\Delta = 3.6$ eV



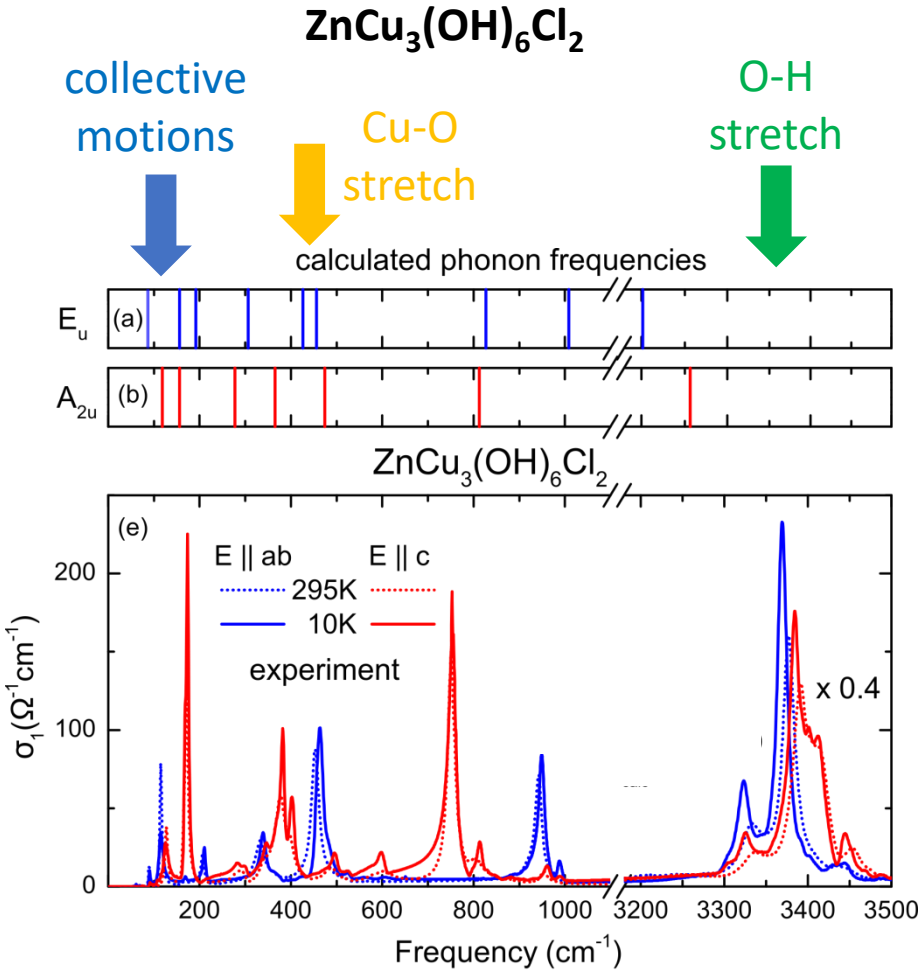
Puphal *et al.*, *J. Mater. Chem. C* **5**, 2629 (2017)

Optical Response - Phonons



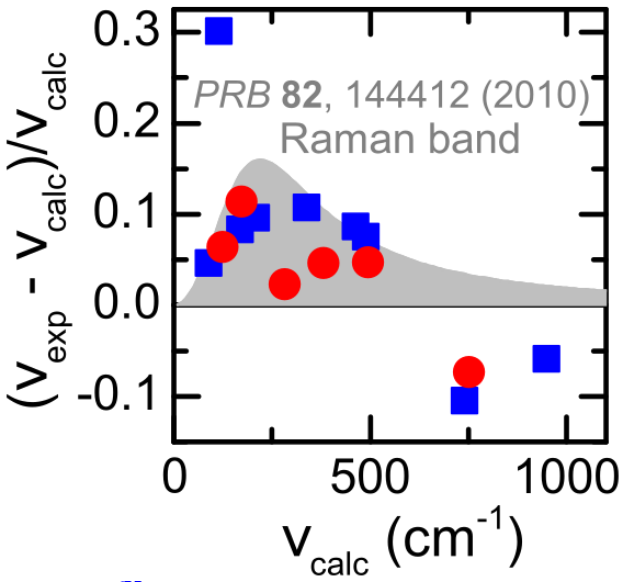
Li *et al.*, *PRB* **101**, 161115(R) (2020) **Editors' Suggestion**

Optical Response - Phonons

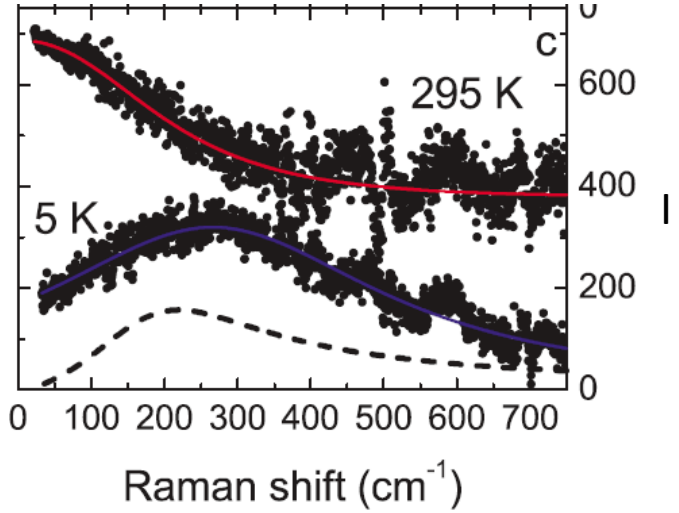


Li *et al.*, *PRB* **101**, 161115(R) (2020) **Editors' Suggestion**

phonon frequencies enhanced around 2-magnon Raman band



Wulferding *et al.*, *PRB* **82**, 144412 (2010)

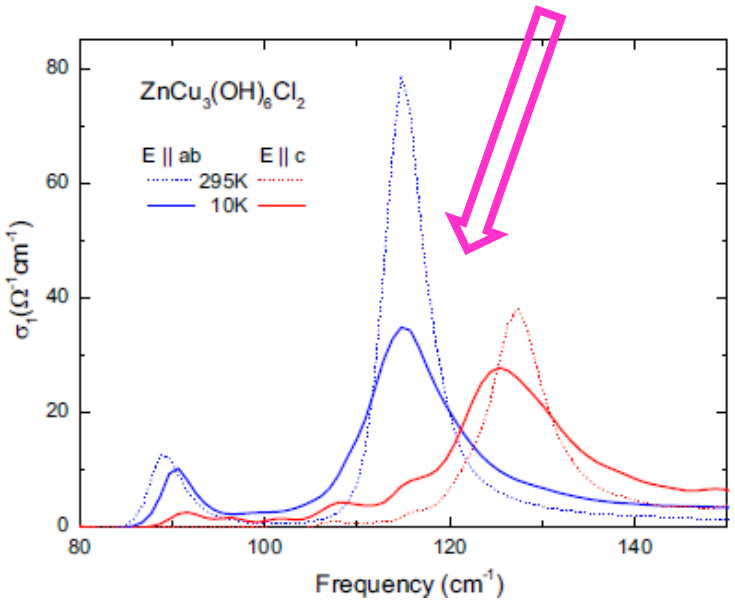


Magnetoelastic Coupling

$ZnCu_3(OH)_6Cl_2$

- non-thermal phonon behavior

broadening & red shift upon cooling



Li *et al.*, *PRB* **101**, 161115(R) (2020) **Editors' Suggestion**

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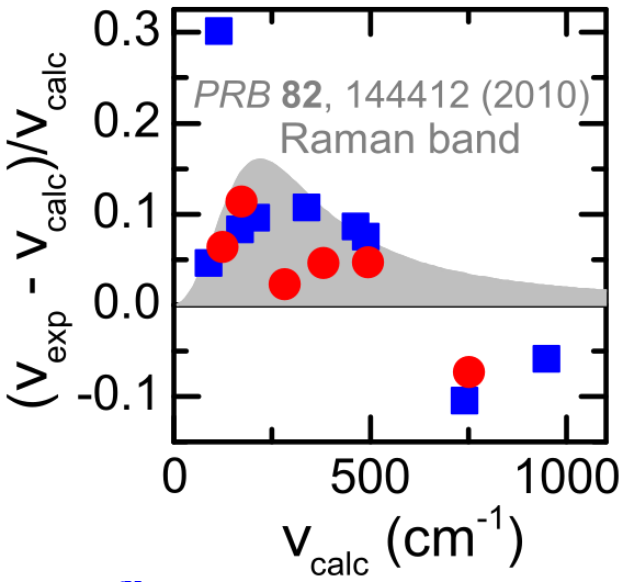
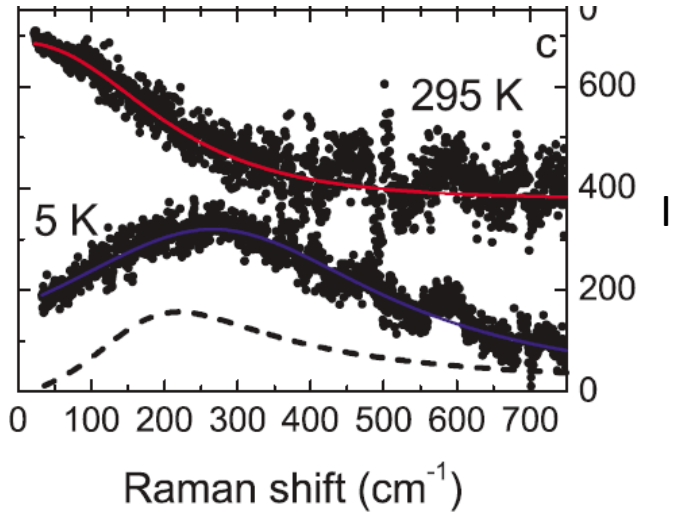


Wulferding *et al.*, *PRB* **82**, 144412 (2010)



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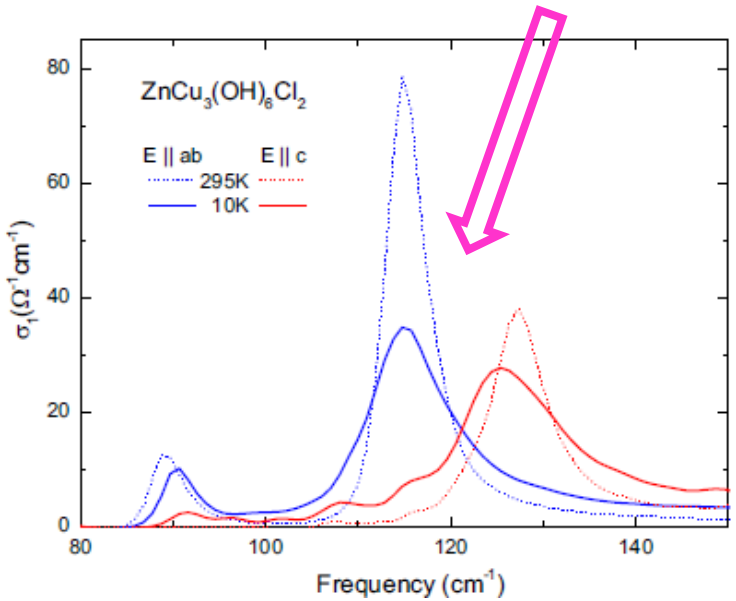
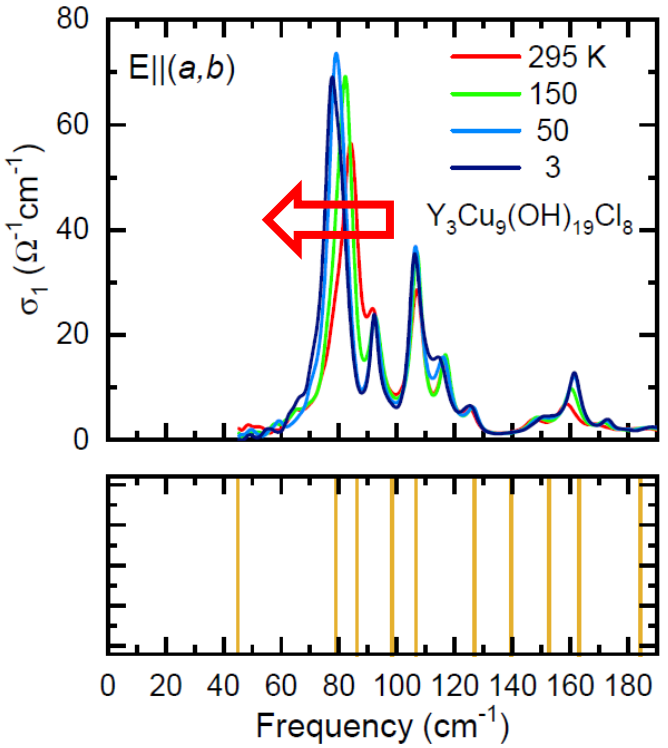


Li *et al.*, *PRB* **101**, 161115(R) (2020) **Editors' Suggestion**

$Y_3Cu_9(OH)_{19}Cl_8$

- non-thermal phonon behavior

broadening & red shift upon cooling

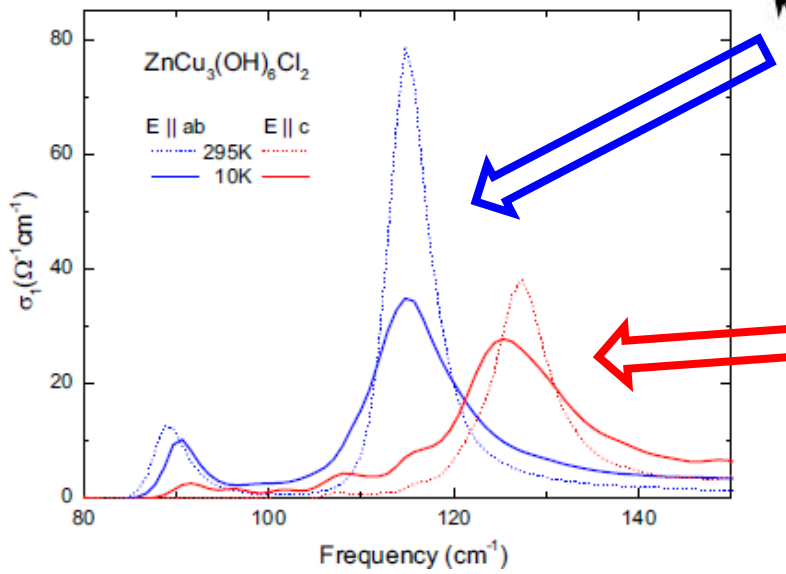


Biesner *et al.*, *Adv. Quantum Technol.* **2022**, 2200023

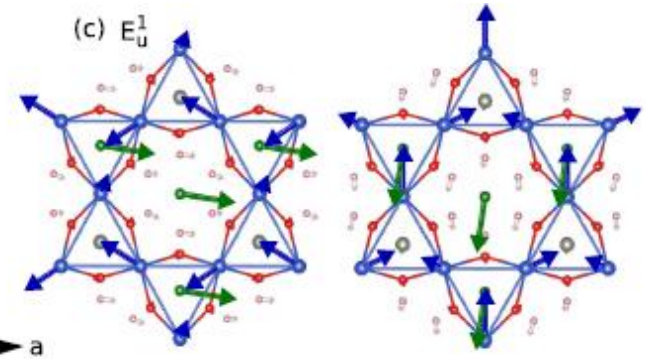
Magnetoelastic Coupling

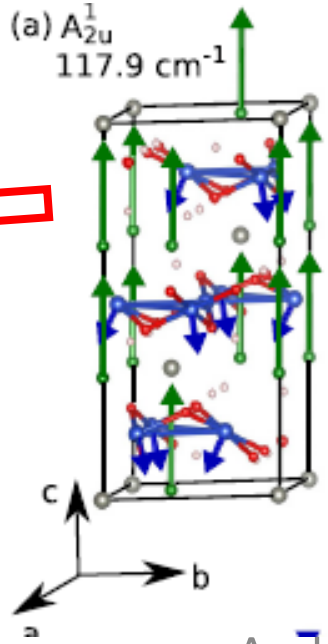
$ZnCu_3(OH)_6Cl_2$

- non-thermal phonon behavior



Li *et al.*, *PRB* **101**, 161115(R) (2020) **Editors' Suggestion**

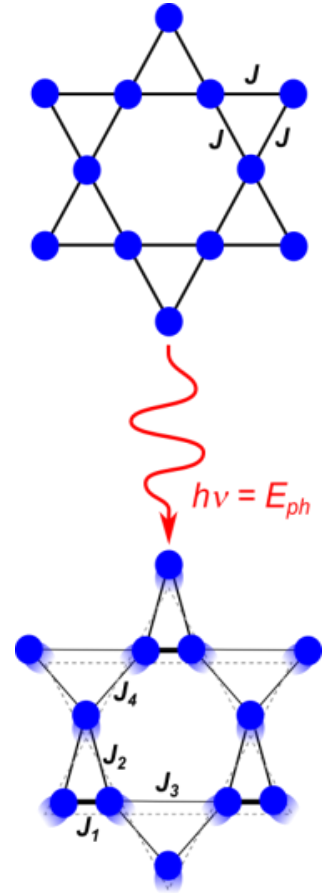




$Y_3Cu_9(OH)_{19}Cl_8$

- non-thermal phonon behavior

magnetoelastic coupling most pronounced for modes that deform the Cu^{2+} kagome layer



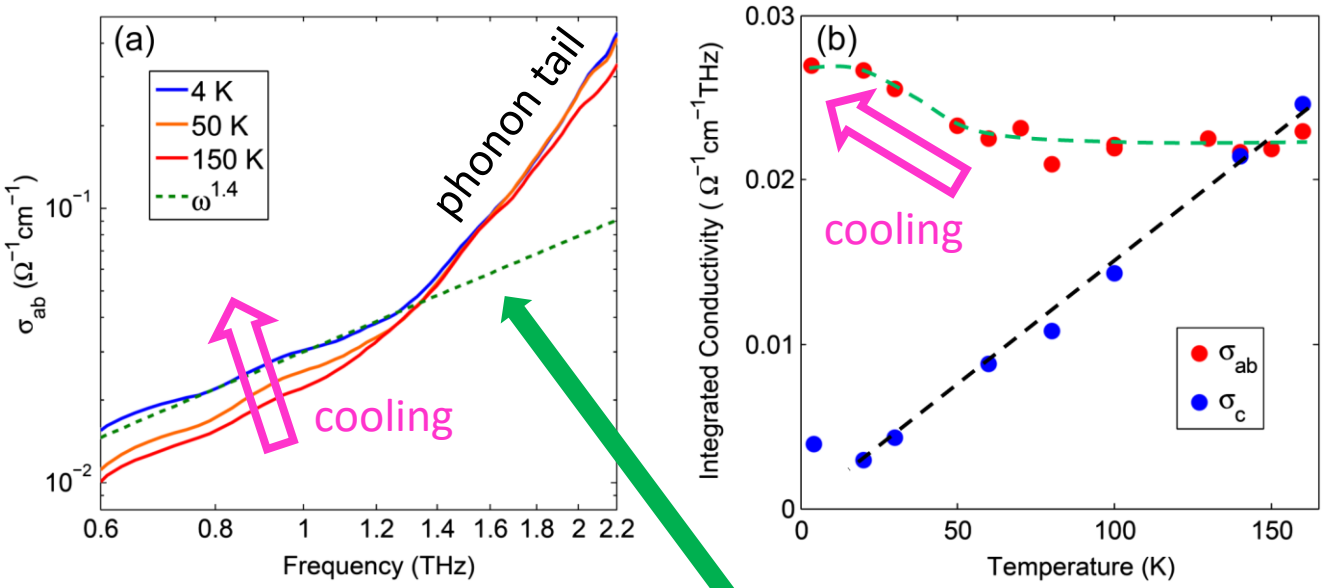
Broadband THz Response

$ZnCu_3(OH)_6Cl_2$

- non-thermal phonon behavior
- non-thermal THz conductivity

$Y_3Cu_9(OH)_{19}Cl_8$

- non-thermal phonon behavior



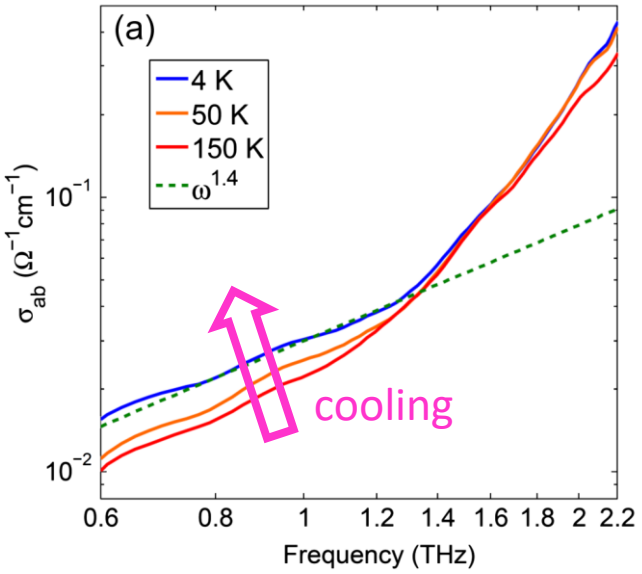
Pilon *et al.*, *PRL* **111**, 127401 (2013)

power law $\sigma_1 \propto \omega^{1.4}$
does not fit theory

Broadband THz Response

$ZnCu_3(OH)_6Cl_2$

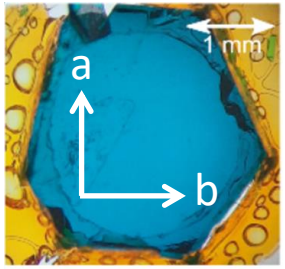
- non-thermal phonon behavior
- non-thermal THz conductivity

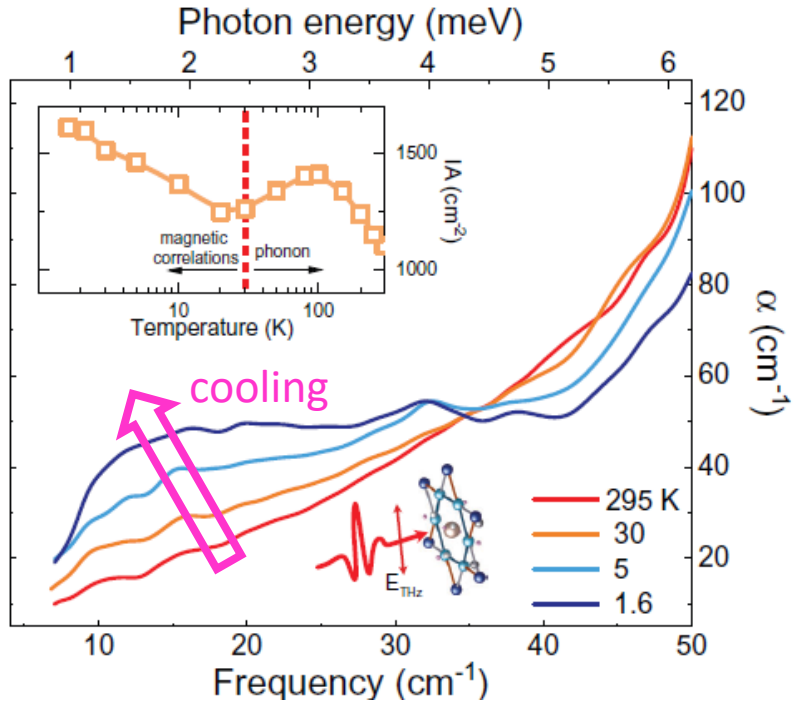


Pilon *et al.*, *PRL* **111**, 127401 (2013)

$Y_3Cu_9(OH)_{19}Cl_8$

- non-thermal phonon behavior
- non-thermal THz conductivity



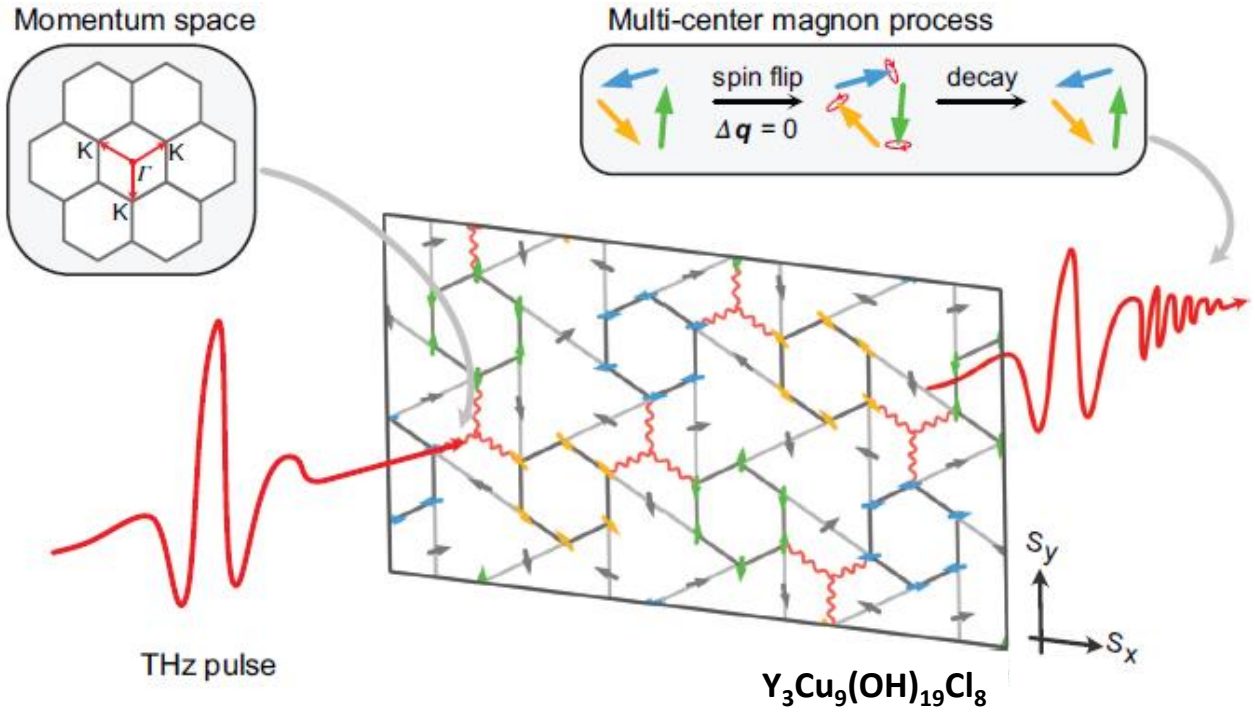


Biesner *et al.*, *Adv. Quantum Technol.* **2022**, 2200023

THz Magnetometry

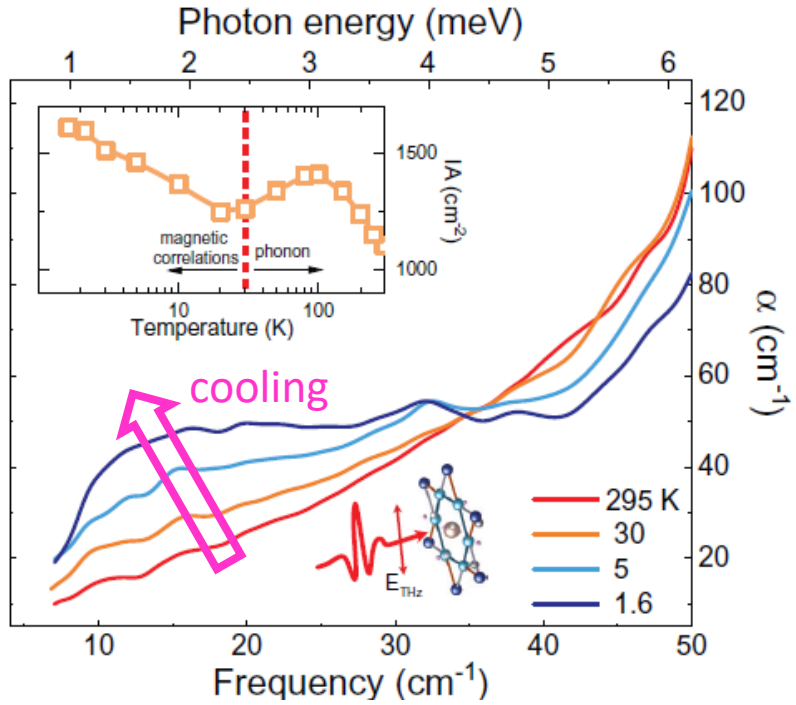
Multicenter Magnon Excitations

- $\vec{Q} = \left(\frac{1}{3} \times \frac{1}{3}\right)$ antiferromagnetic order
- 3 different magnetic sublattices lift zone-center constraint of photons



$Y_3Cu_9(OH)_{19}Cl_8$

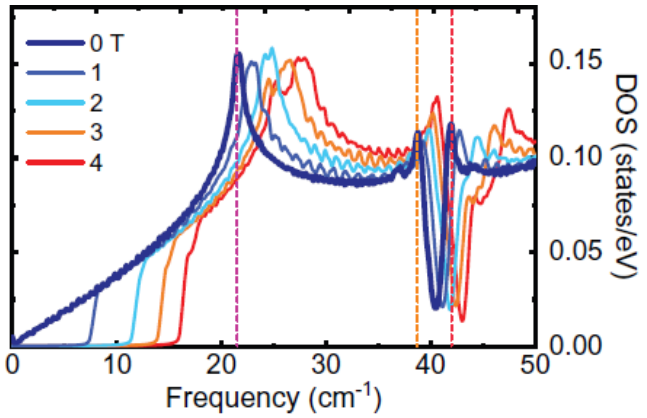
- non-thermal phonon behavior
- non-thermal THz conductivity



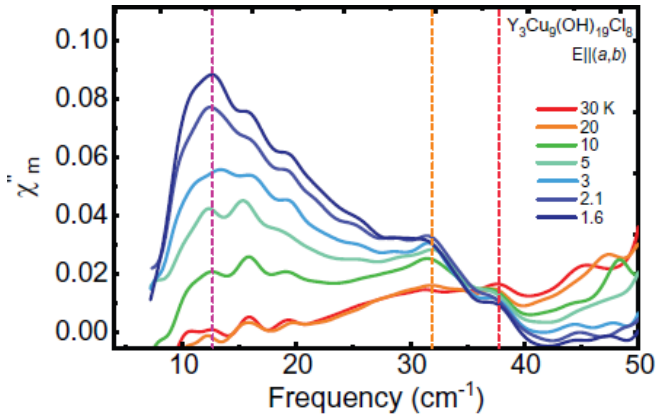
Biesner *et al.*, *Adv. Quantum Technol.* **2022**, 2200023

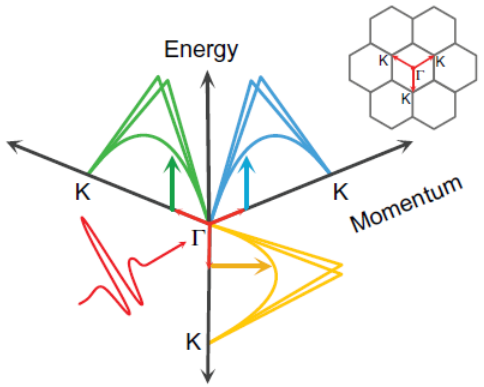
THz Magnetometry

linear spin-wave theory



experiment

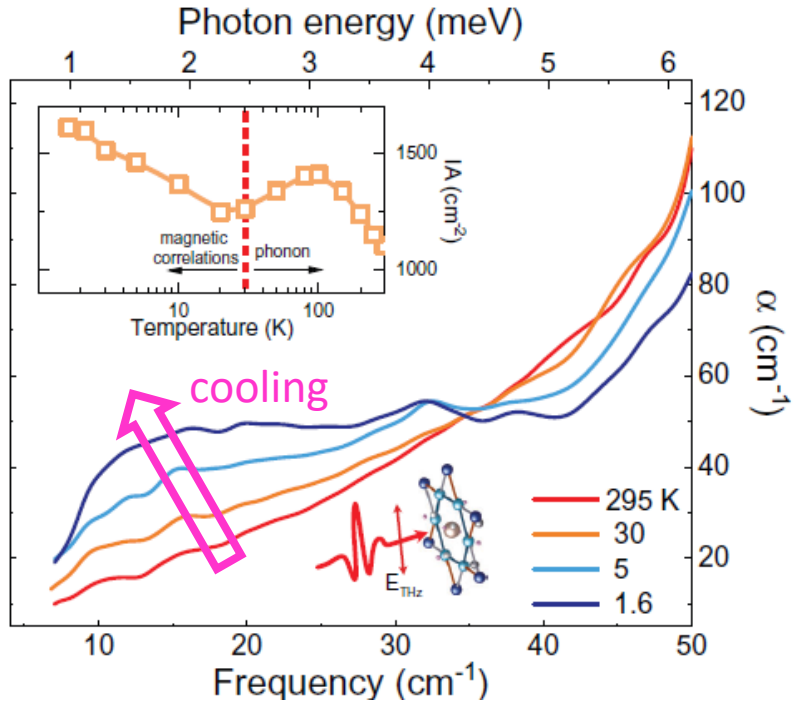




- extract dynamic susceptibility from α
- assume $\varepsilon = const$ below $T = 80$ K

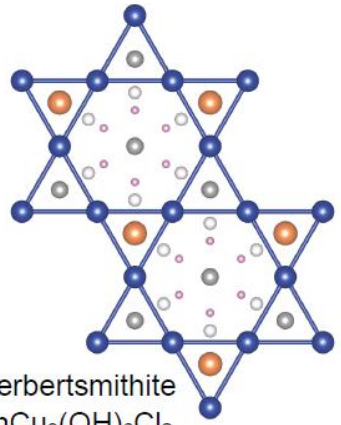
$Y_3Cu_9(OH)_{19}Cl_8$

- non-thermal phonon behavior
- non-thermal THz conductivity

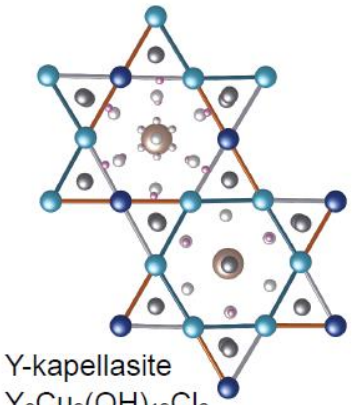


Biesner *et al.*, *Adv. Quantum Technol.* **2022**, 2200023

Unfrustrating the Kagome Layer



Herbertsmithite
$ZnCu_3(OH)_6Cl_2$

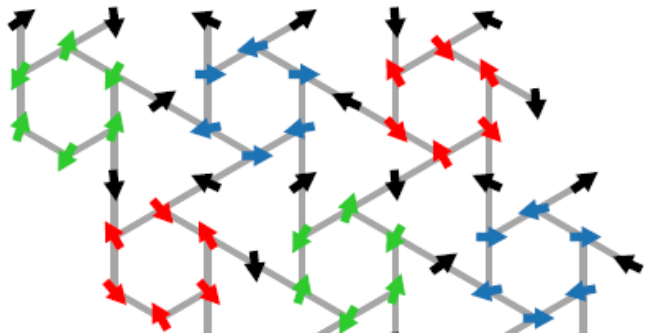


Y-kapellasite
$Y_3Cu_9(OH)_{19}Cl_8$

- ideal kagome
- no magnetic order

- distorted kagome
- $T_N = 2.2$ K

- **magnetic order stabilized upon increased kagome-plane distortion**
- **broken symmetry activates multi-magnon THz response**



Biesner *et al.*, *PRB* **105**, L060410 (2022)
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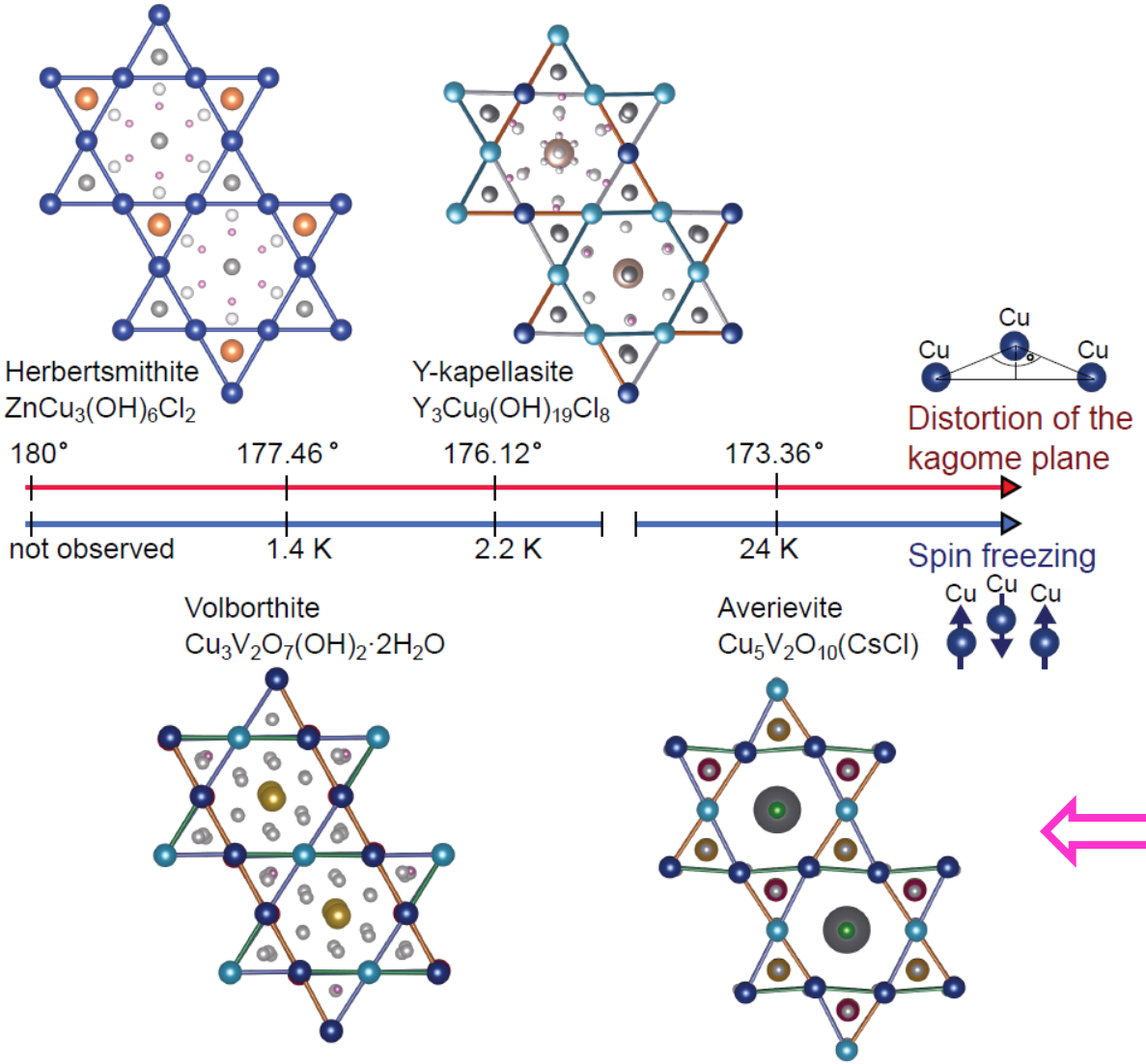
Unfrustrating the Kagome Layer



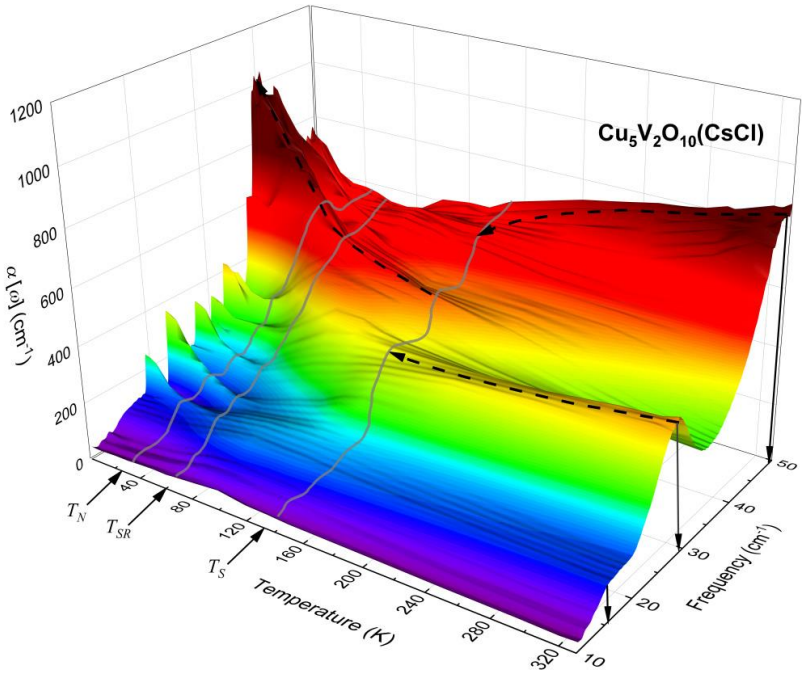
- **magnetic order stabilized upon increased kagome-plane distortion**
- **broken symmetry activates multi-magnon THz response**

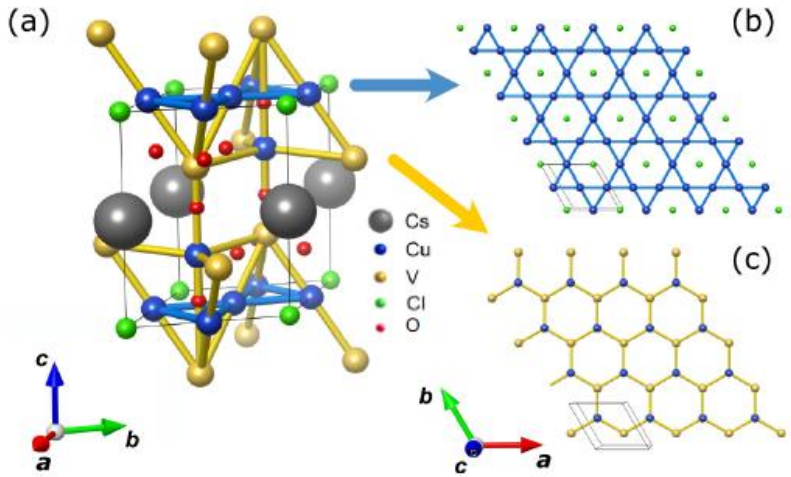
Look at even more distorted kagome lattice in Averievite!

Biesner *et al.*, *PRB* **105**, L060410 (2022)
Adv. Quantum Technol. **2022**, 2200023

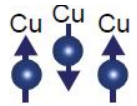
Unfrustrating the Kagome Layer

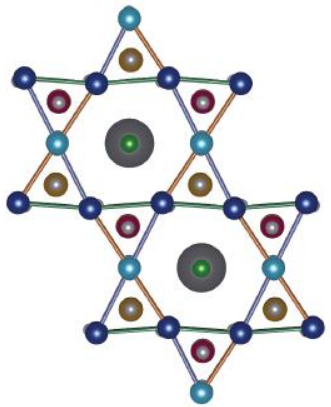
Strong magnetic resonances!





Averievite
$Cu_5V_2O_{10}$(CsCl)



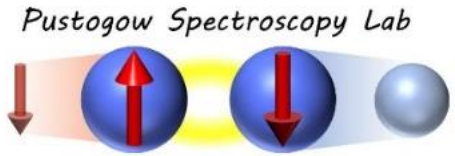


$Cu_{5-x}Zn_xV_2O_{10}$(CsCl)

- replace Cu in honeycomb layers
- decouple kagome layers
- no magnetic order for $x = 1$

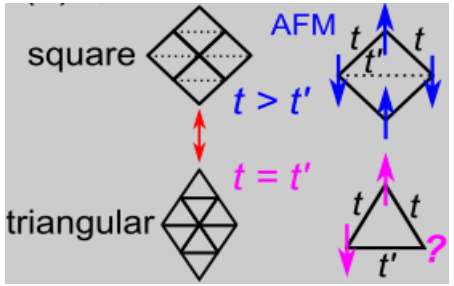
Look at even more distorted kagome lattice in Averievite!

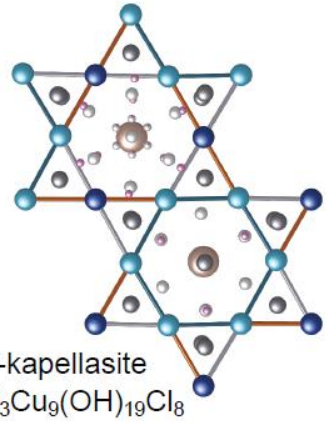
Biesner *et al.*, *PRB* **105**, L060410 (2022)



Unfrustrating the Kagome Layer

tuning magnetic frustration by strain





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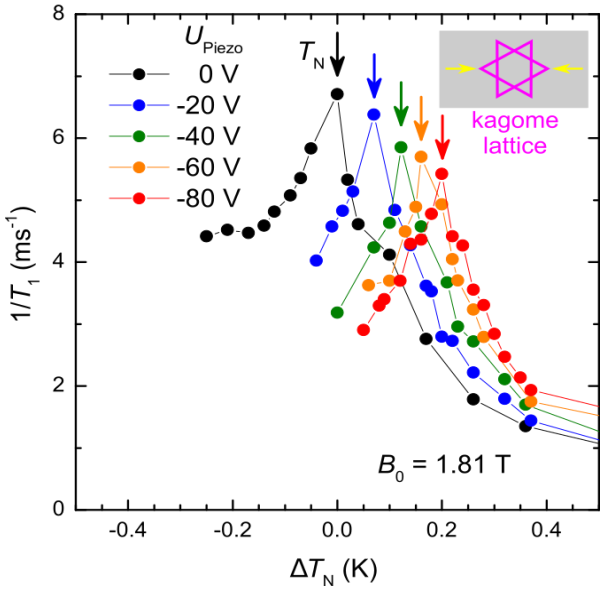
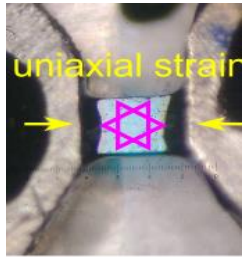




in situ* strain tuning at low *T

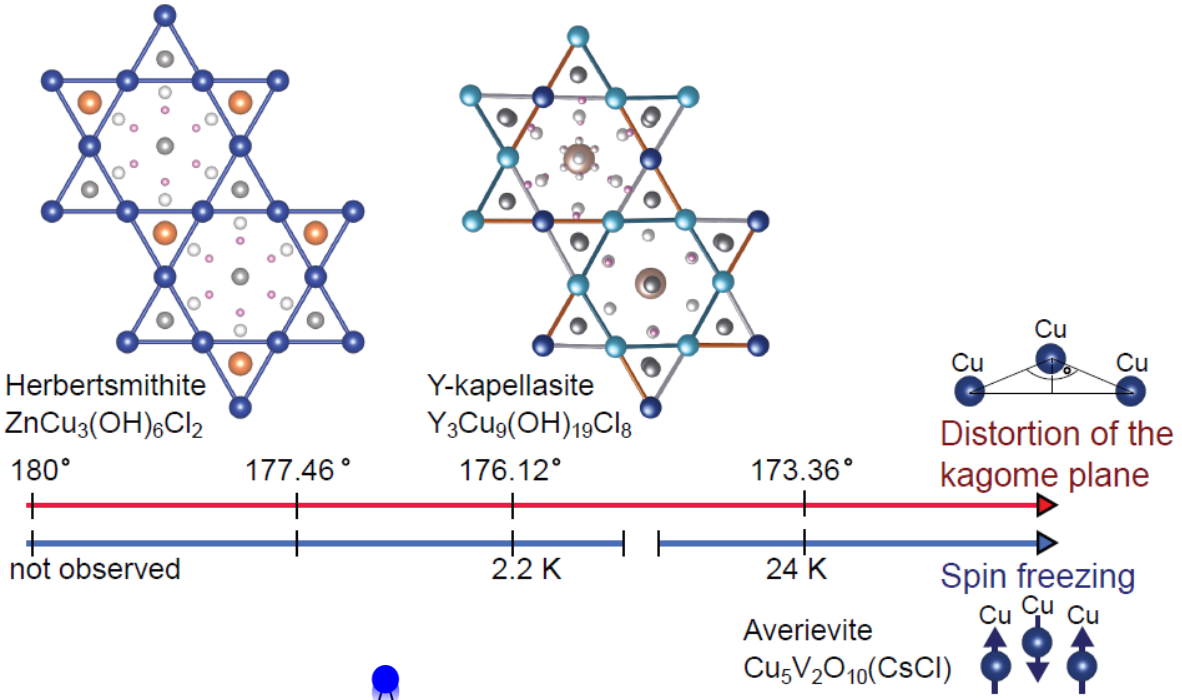
Controlling frustrated magnetism on the kagome lattice by uniaxial-strain tuning

Jierong Wang,[1] M. Spitaler,[2] Y.-S. Su,[1] K.M. Zoch,[3] C. Krellner,[3] P. Puphal,[3,4] S. E. Brown,[1] and A. Pustogow[1,2,*]

arXiv:2209.08613

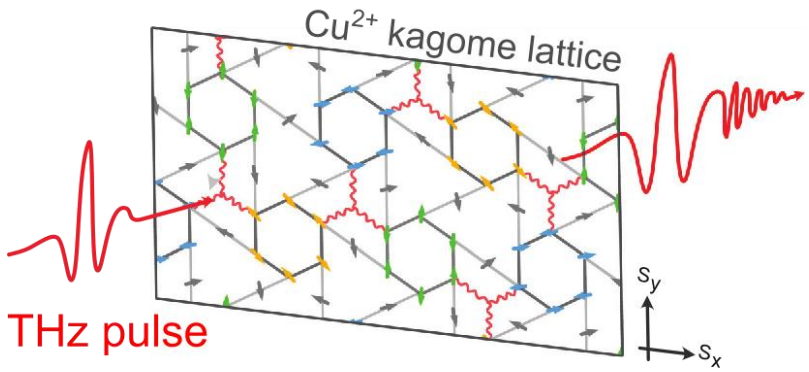
Pustogow Spectroscopy Lab

Conclusion



Li *et al.*, *PRB* **101**, 161115(R) (2020) **Editors' Suggestion**

- **magnetic order stabilized upon increased kagome-plane distortion**
- **broken symmetry activates multi-magnon THz response**
- **strong spin-phonon coupling**



Biesner *et al.*, *PRB* **105**, L060410 (2022)
Adv. Quantum Technol. **2022**, 2200023