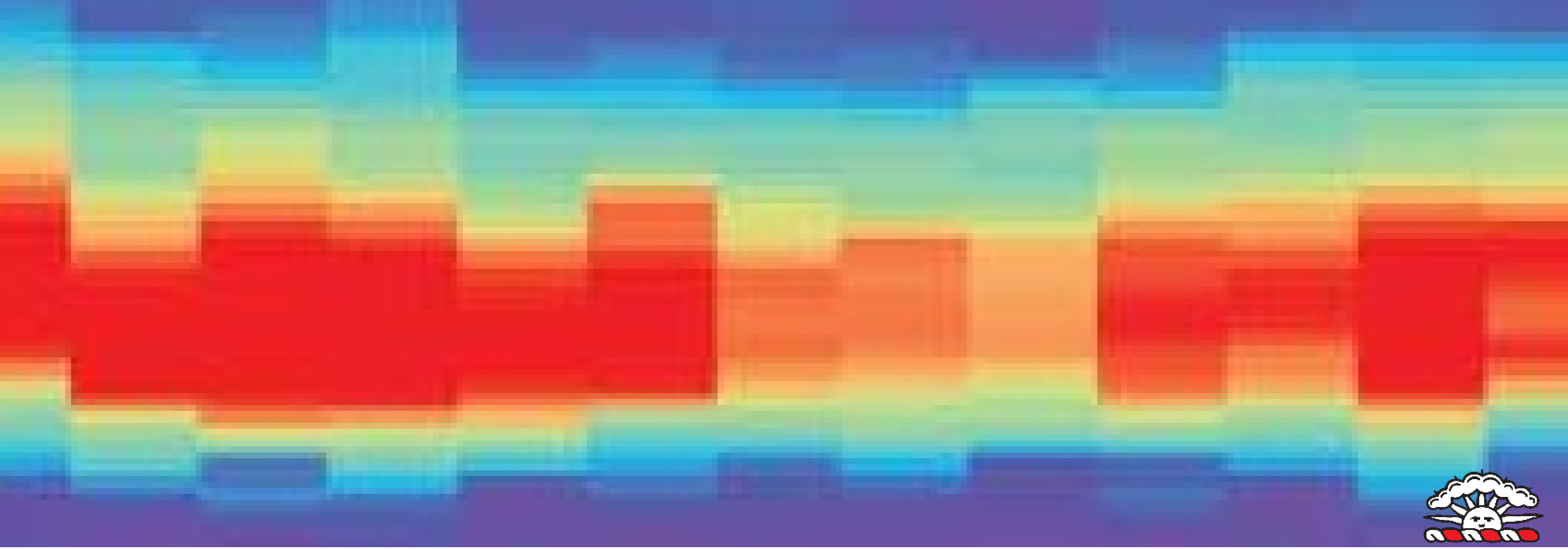
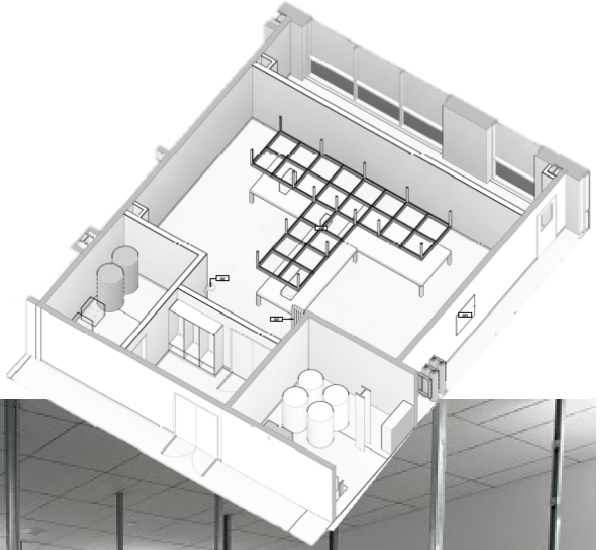
Magneitc excitations and dynamics of the honeycomb iridate $H_3LiIr_2O_6$



Alberto de la Torre (he/his)
Northeastern University

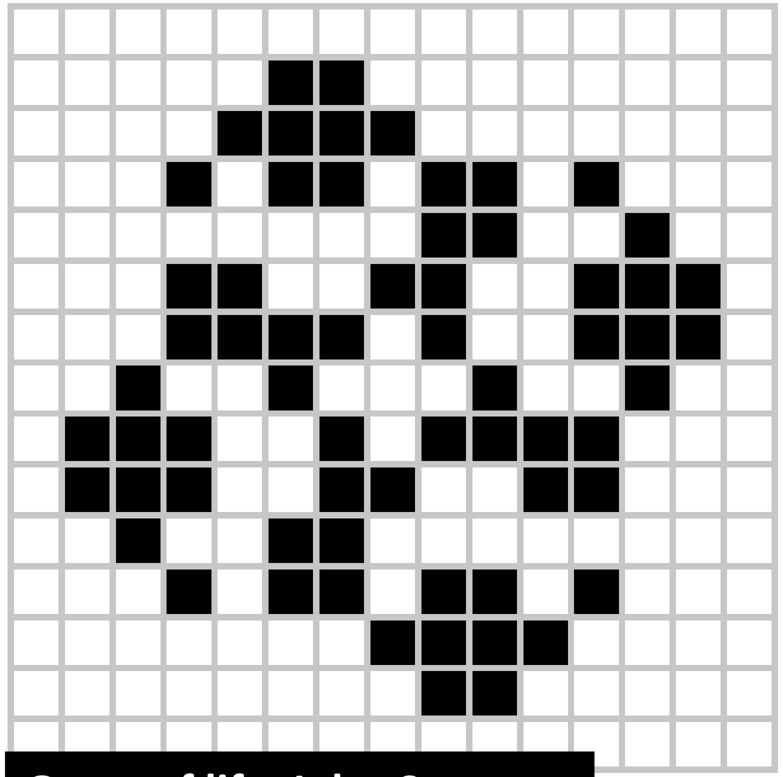
U.S. DEPARTMENT OF ENERGY
Office of Science

DeLTA lab at NU

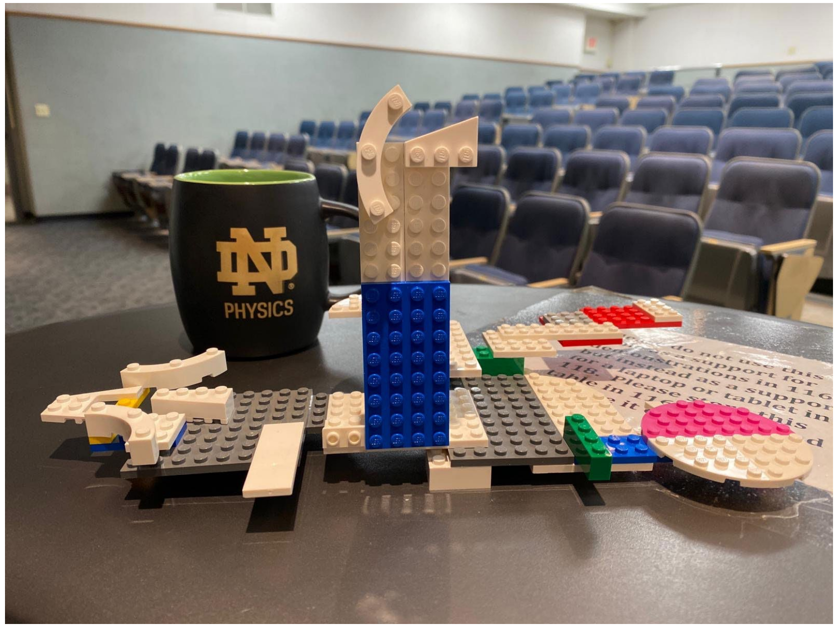


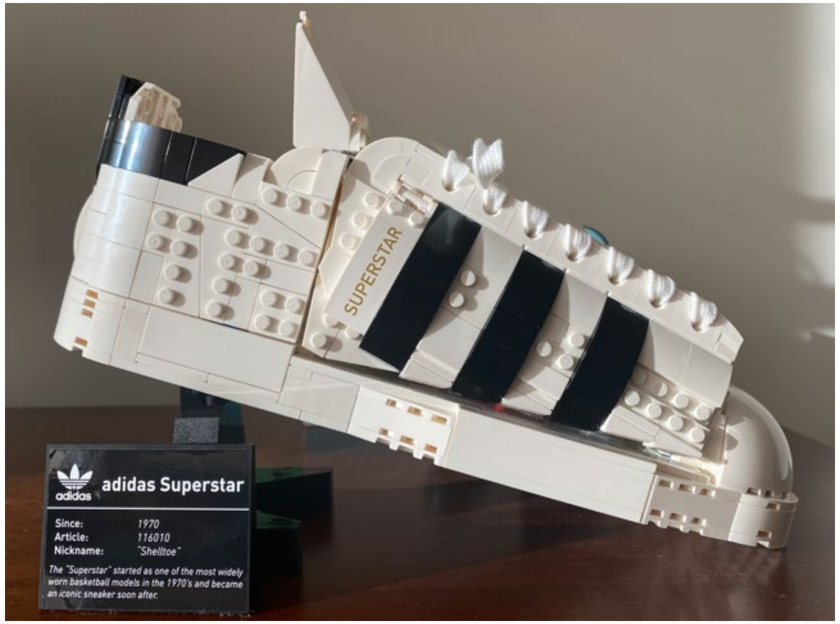
"Emergence"

"More is different"
P. W. Anderson, 1972



Game of life, John Conway

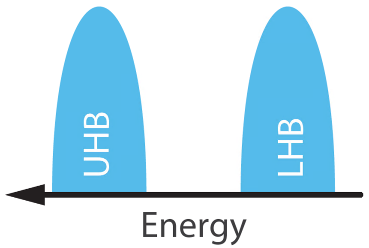




Quantum Materials

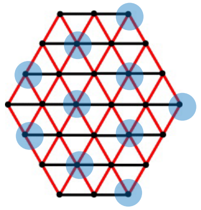
Materials displaying complex emergent phenomena over a wide range of energies and length scales

Correlated insulators

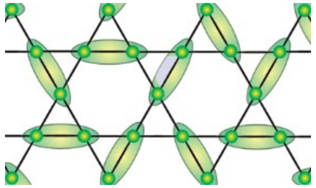


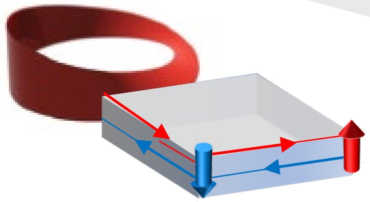
Unconventional superconductivity

Charge orders

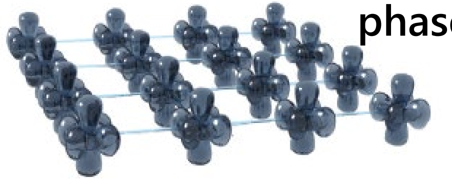


Quantum magnets





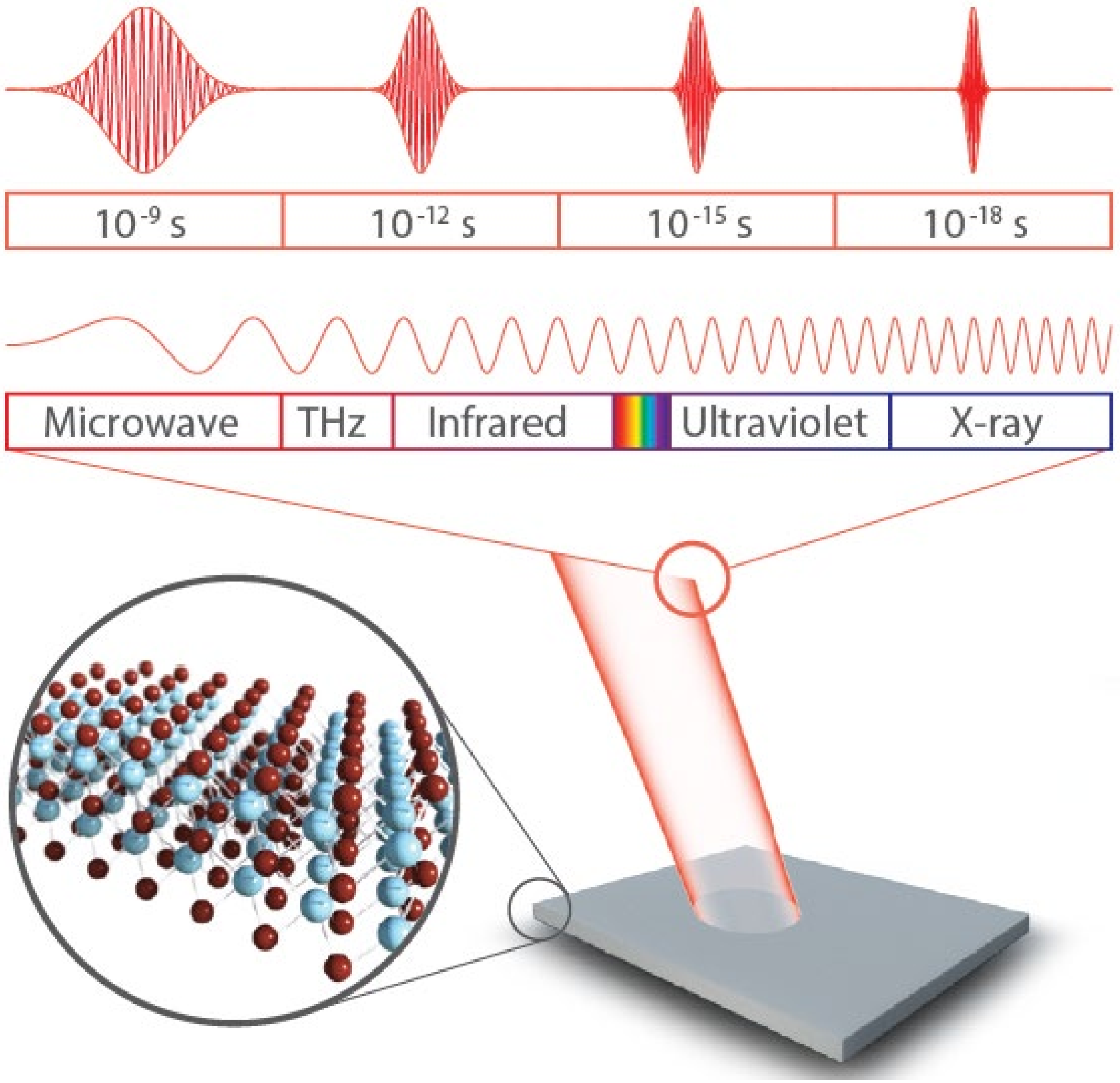
Topological protection



Multipolar ordered phases

From properties by design
to properties by demand
Nat Mat 16, 1077 (2017)

Ultrafast light-matter interaction



Reversible

Timescale of microscopic processes

Flexible implementation

Access p.o.m. without equilibrium counterpart

ADLT et al; Rev. Mod. Phys. 93, 041002 (2021)

Nonthermality as a resource

Colloquium: Nonthermal pathways to ultrafast control in quantum materials

ADLT et al, Rev. Mod. Phys. **93**, 041002 (2021)

Dante Kennes

Martin Claassen

Simon Gerber

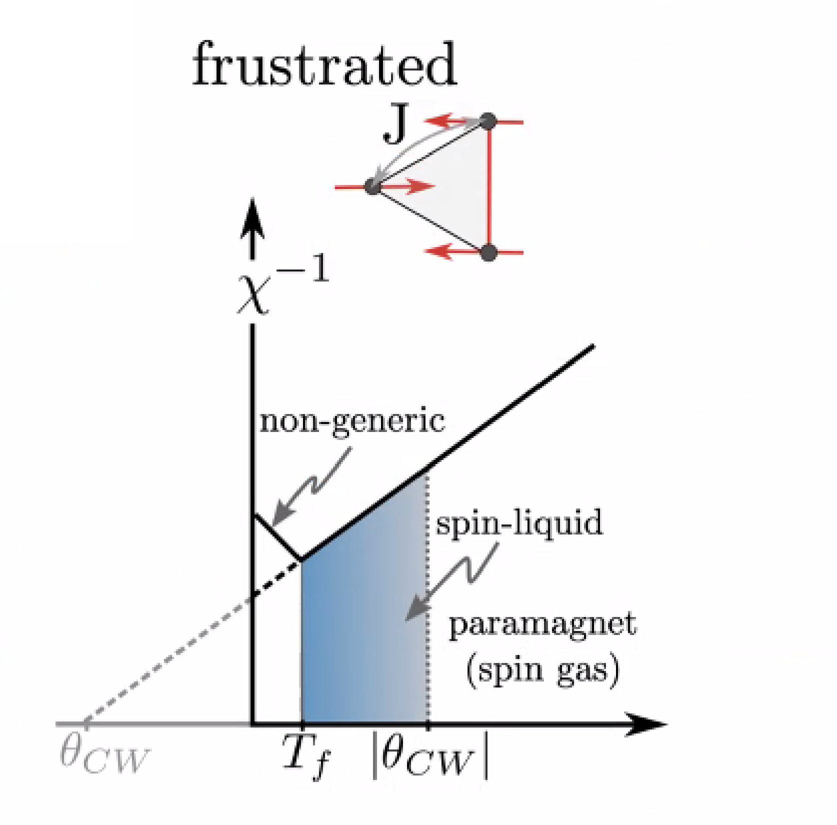
James McIver

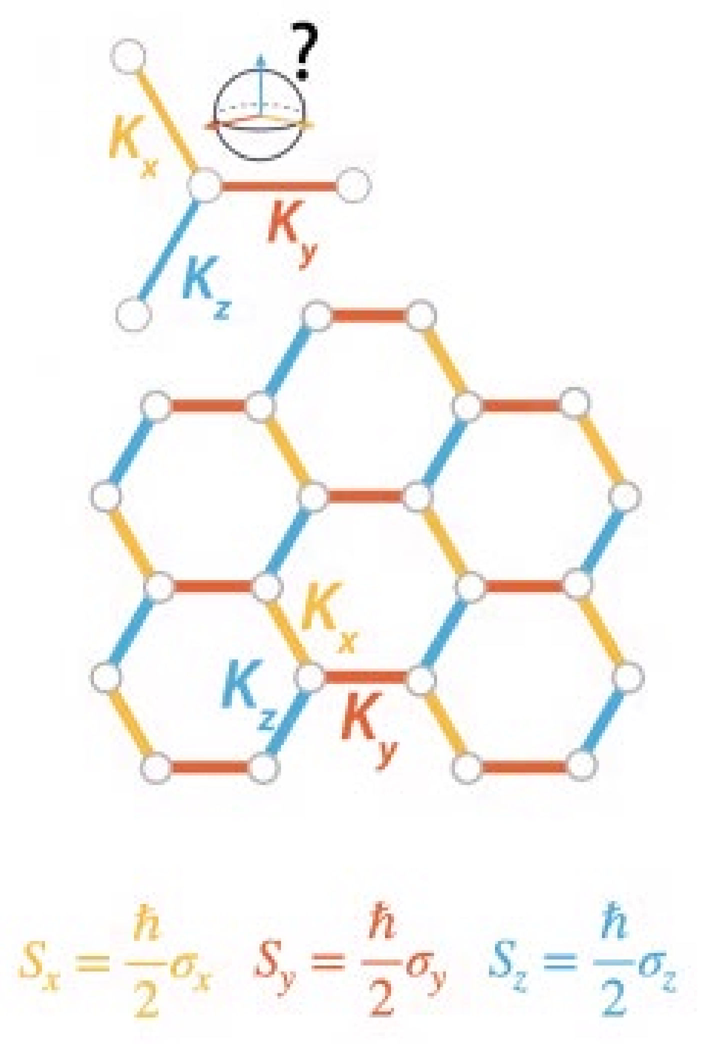
Michael Sentef

Quantum Spin Liquids

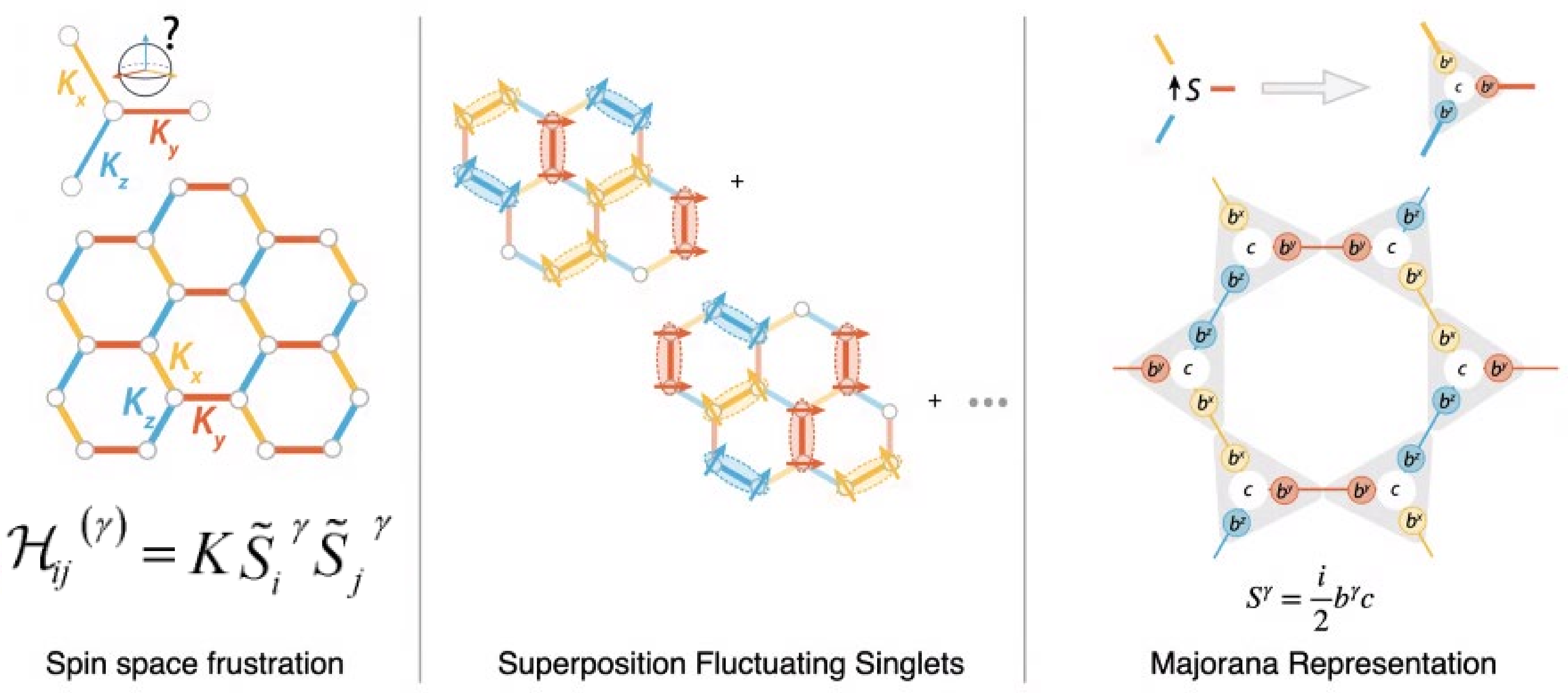
- Highly correlated fluctuating spins down to low temperature without symmetry breaking

- Long range entangled ground states





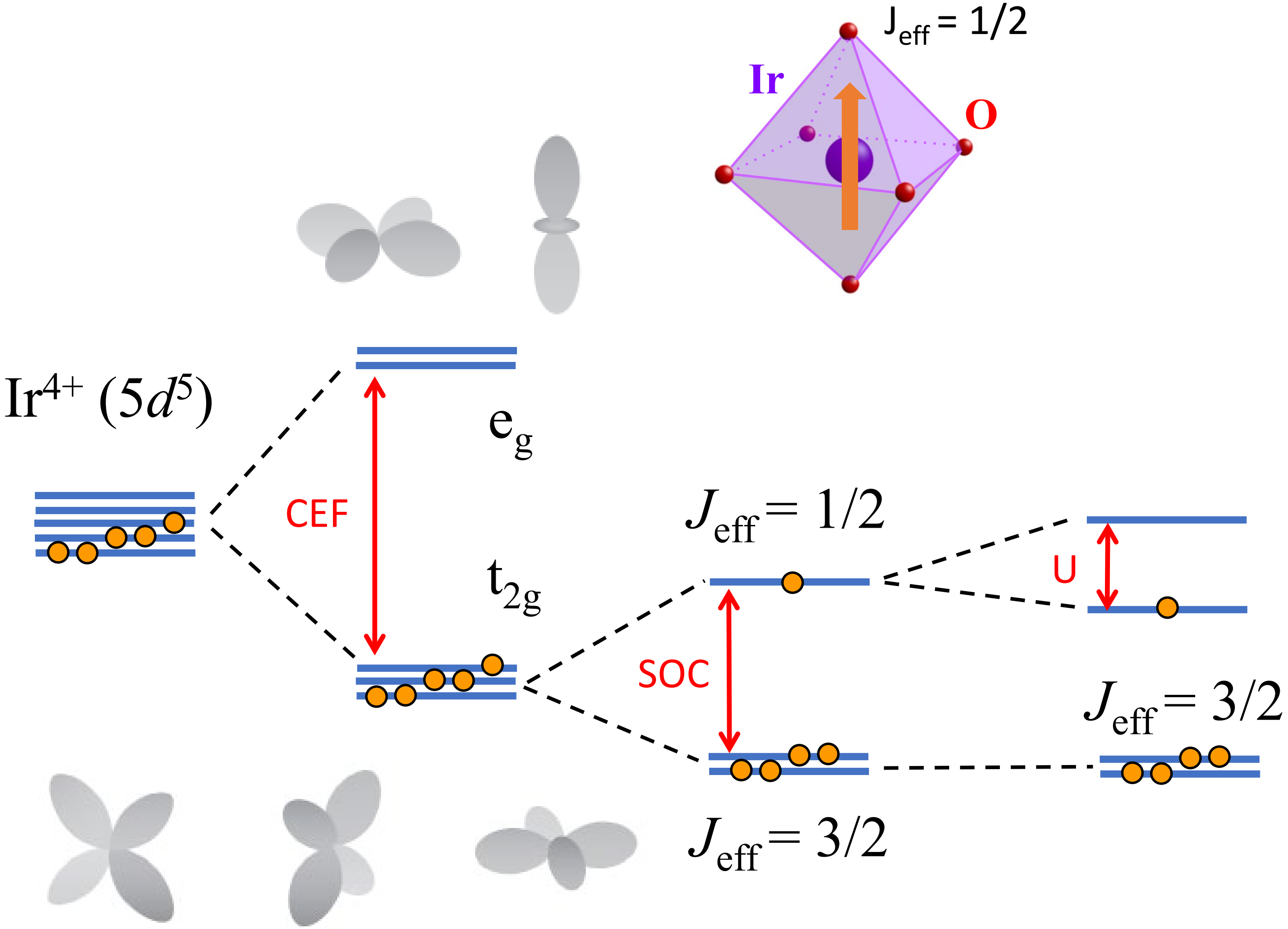
Kitaev model in the honeycomb lattice



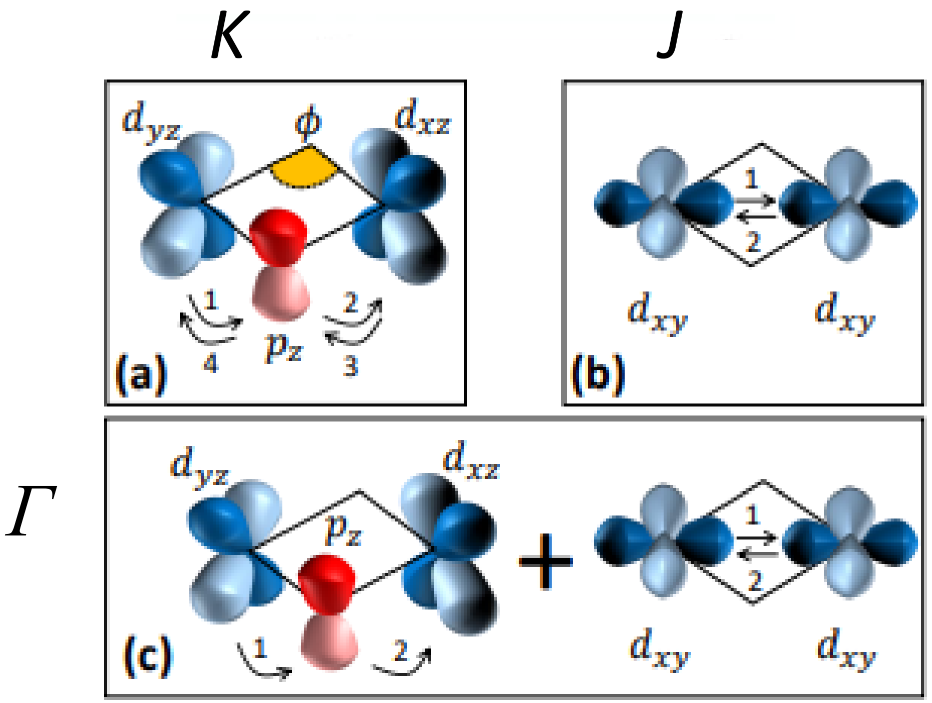
K. Plumb

Iridium oxides

Single ion

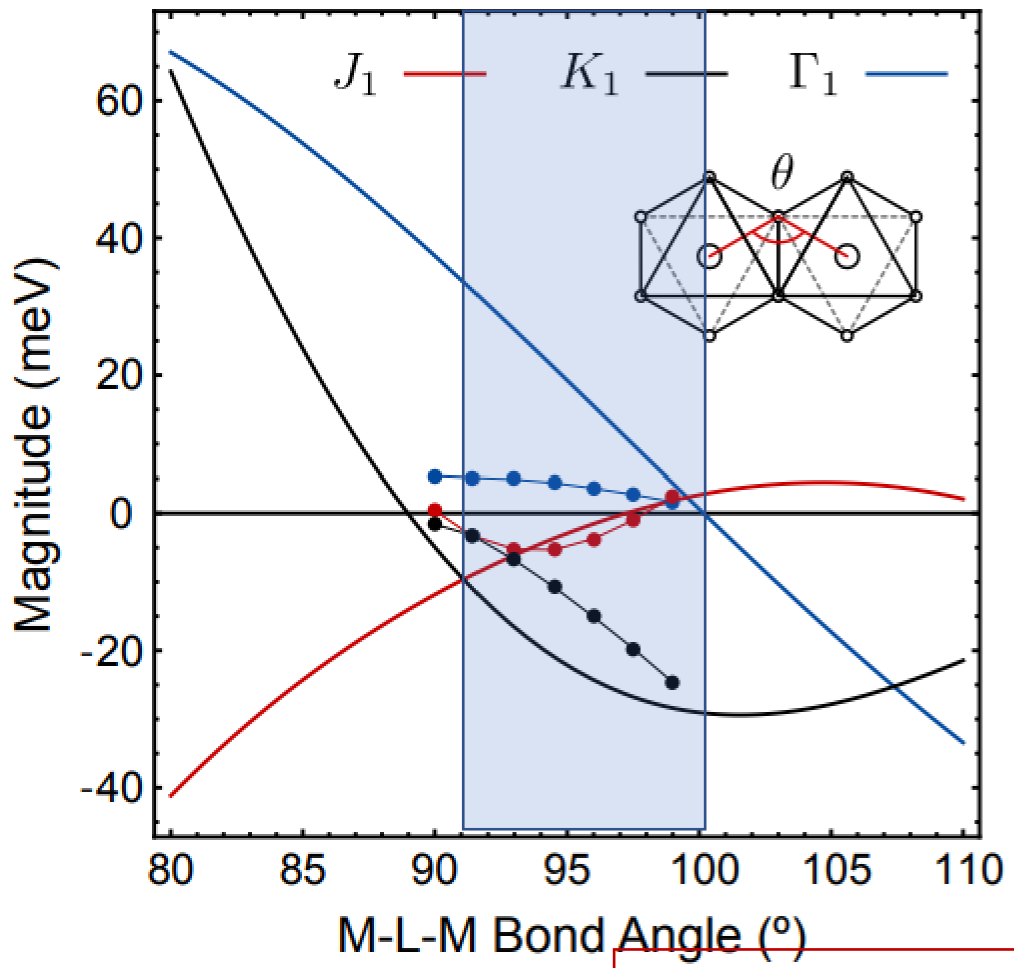


$$\mathcal{H}_{ij}^{(\gamma)} = J\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\tilde{S}_i^{\gamma}\tilde{S}_j^{\gamma} + \sum_{\alpha\neq\beta}\Gamma_{\alpha\beta}\left(\tilde{S}_i^{\alpha}\tilde{S}_j^{\beta} + \tilde{S}_i^{\beta}\tilde{S}_j^{\alpha}\right)$$

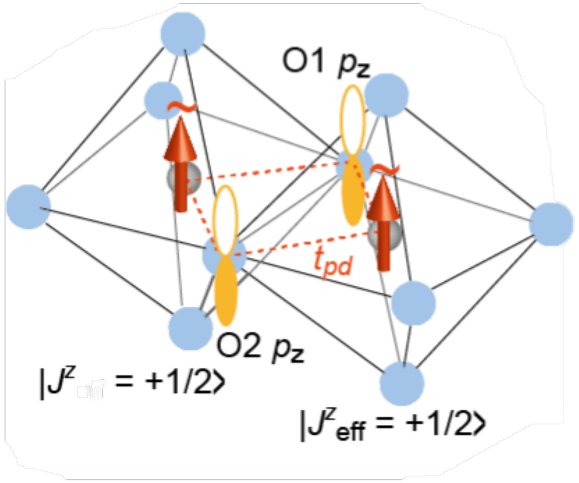


Phys. Rev. Lett. 102, 017205 (2009)

Magnetic interactions



Edge sharing



K

J

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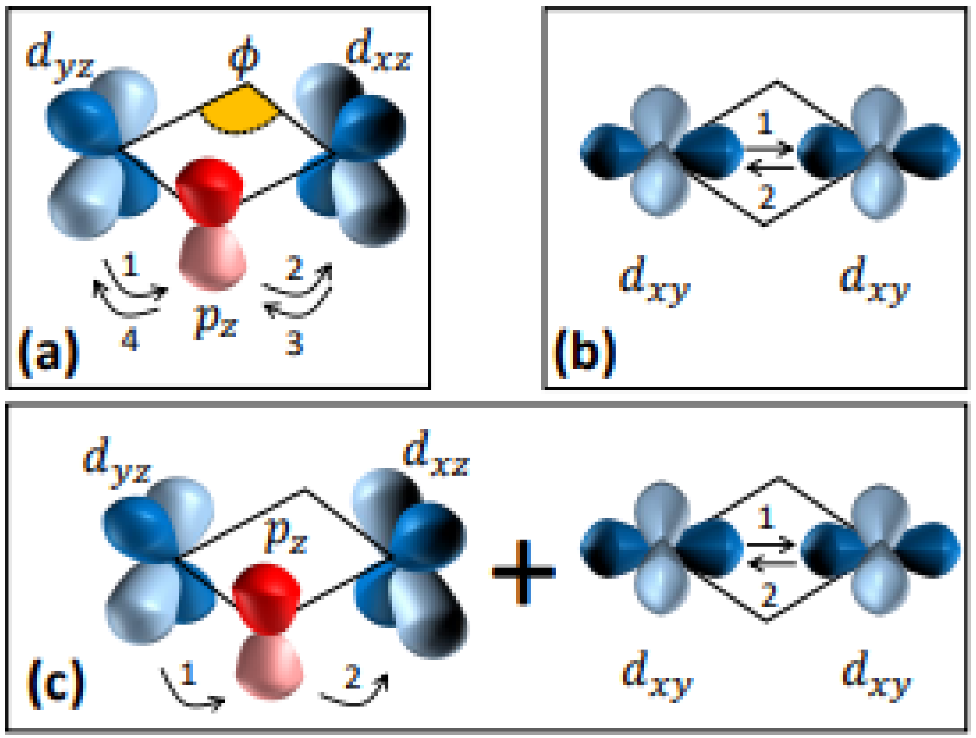
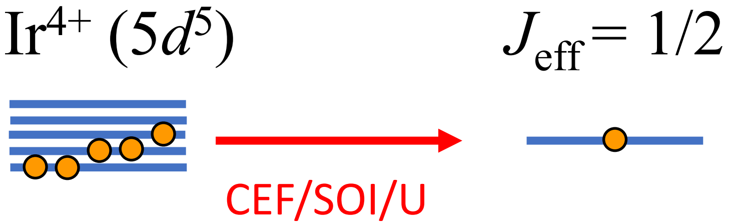
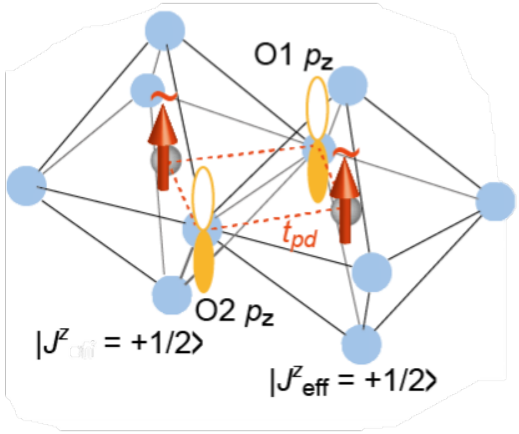


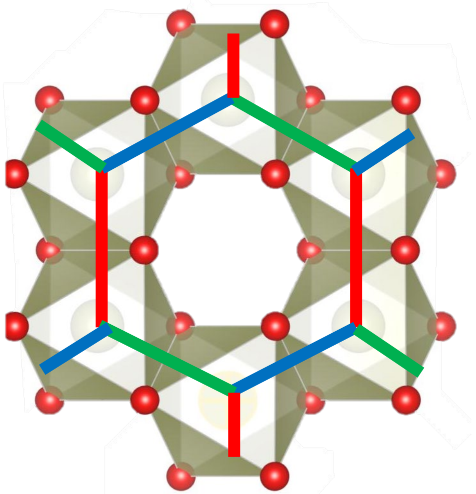
$$\mathcal{H}_{ij}^{(\gamma)} = K\tilde{S}_i^{\gamma}\tilde{S}_j^{\gamma}$$

J. Phys.: Condens. Matter 29, 493002 (2017)

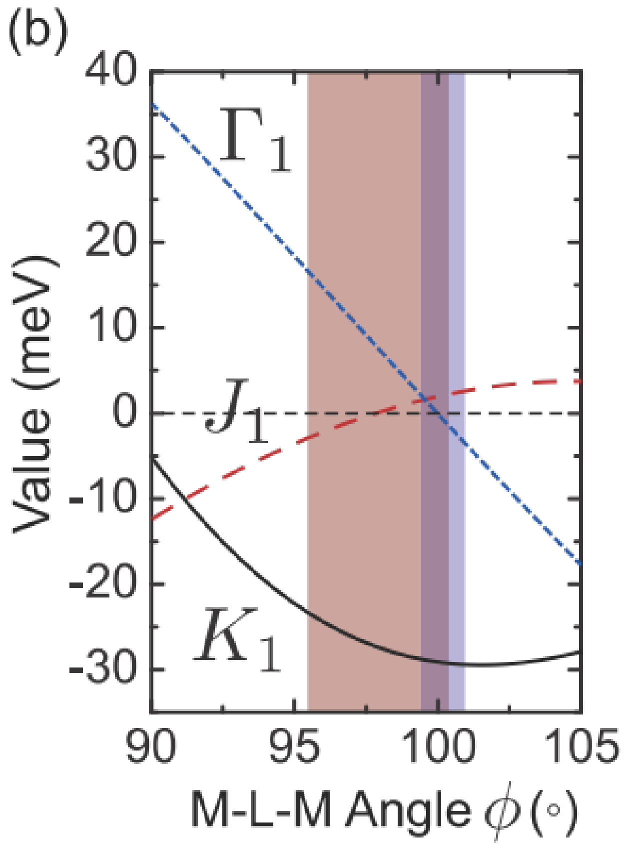
Towards Kitaev Quantum Spin Liquids

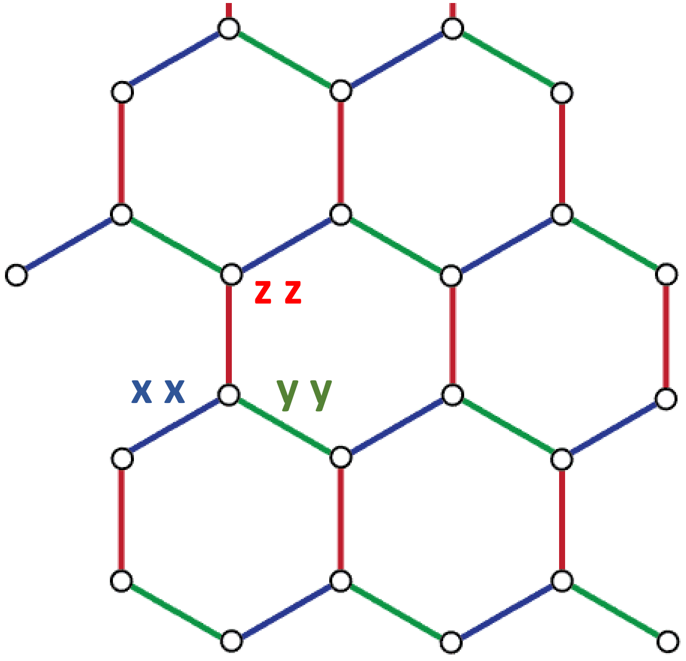






Nature Rev Phys 1, 264–280 (2019)

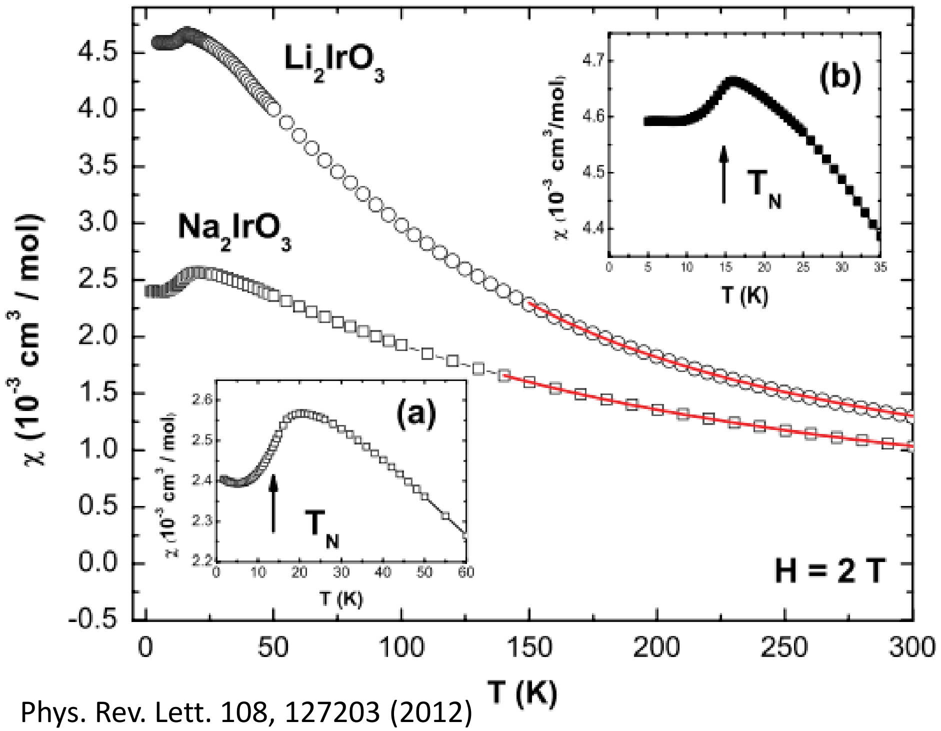




$$\mathcal{H}_{ij}^{(\gamma)} = K \tilde{S}_i^{\gamma} \tilde{S}_j^{\gamma}$$

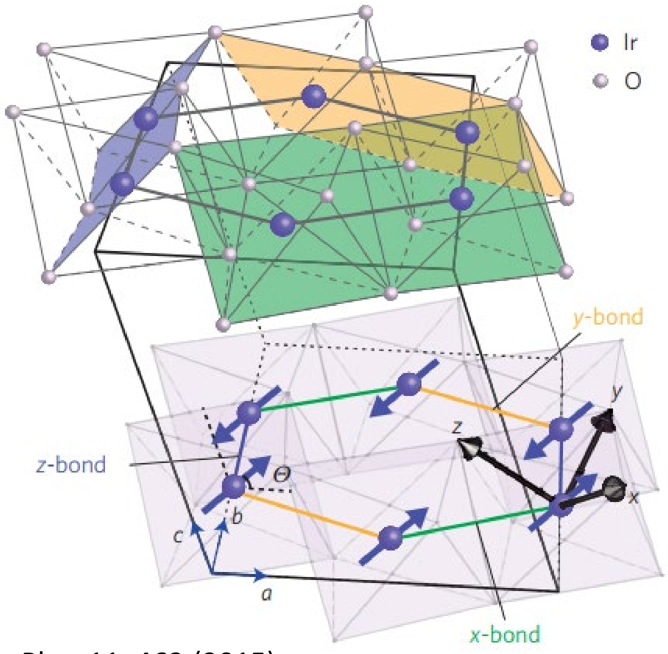
Honeycomb iridates: A_2IrO_3

First experimental realizations



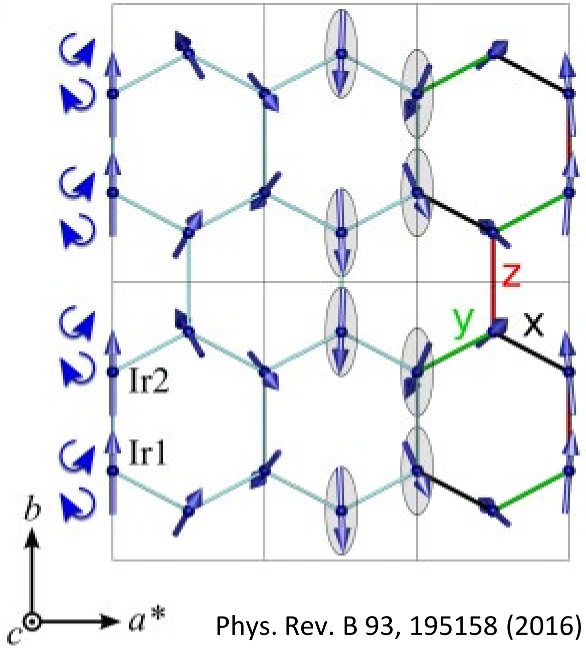
Phys. Rev. Lett. 108, 127203 (2012)

Na_2IrO_3



Nat Phys 11, 462 (2015)

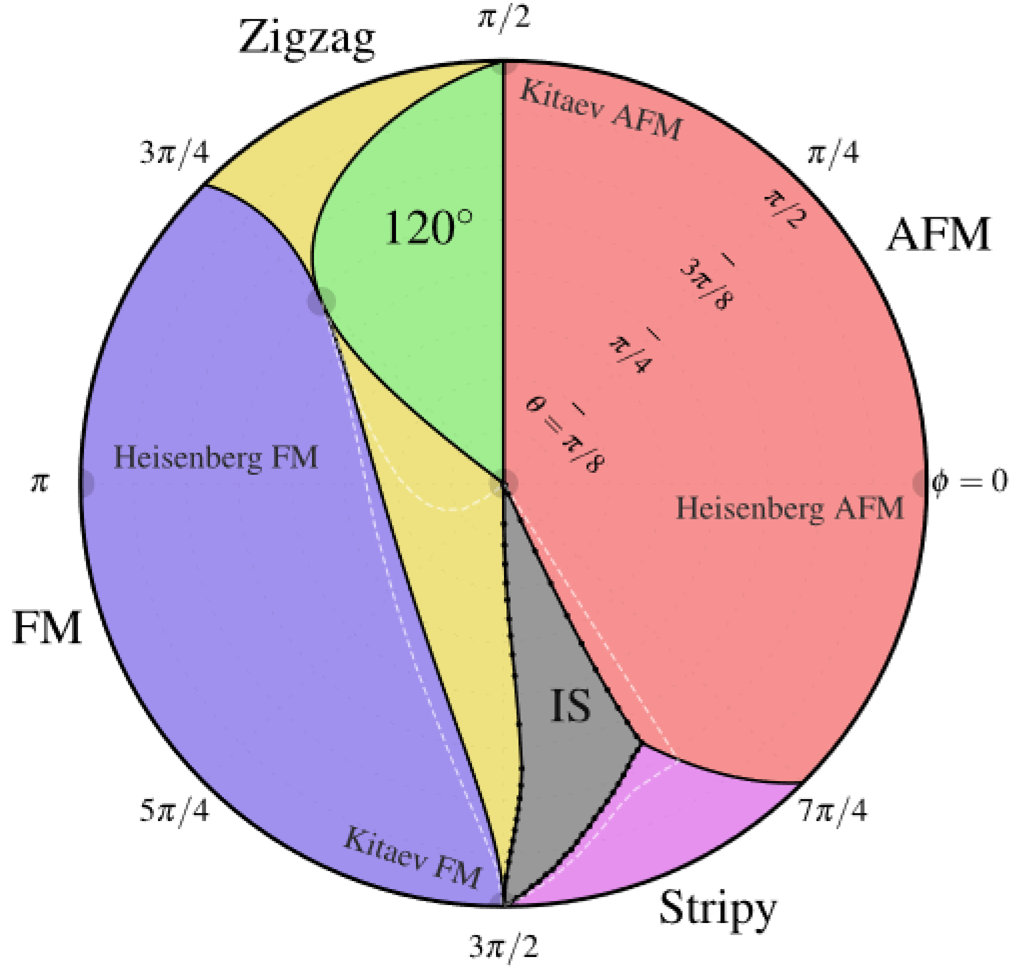
α-Li_2IrO_3

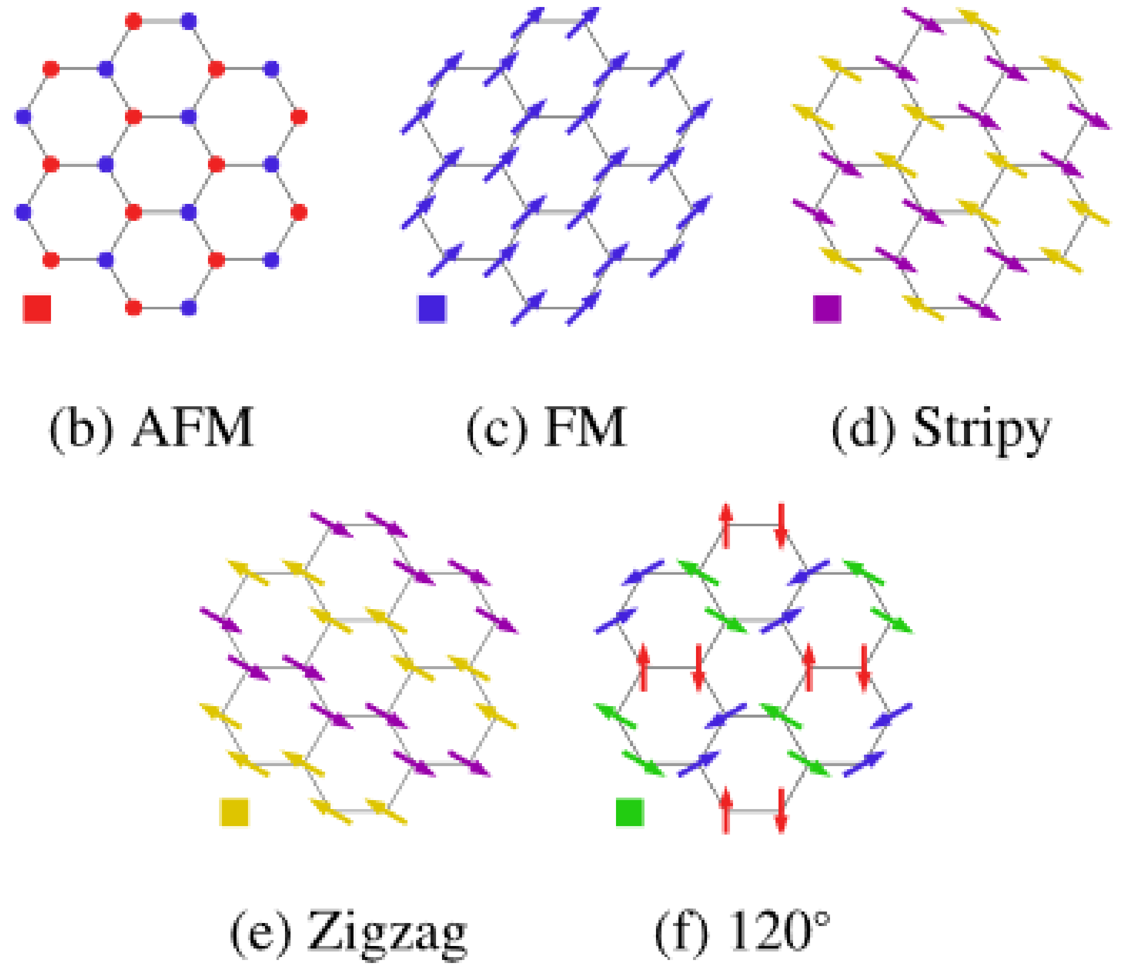


Phys. Rev. B 93, 195158 (2016)

Generic spin model beyond the Kitaev limit

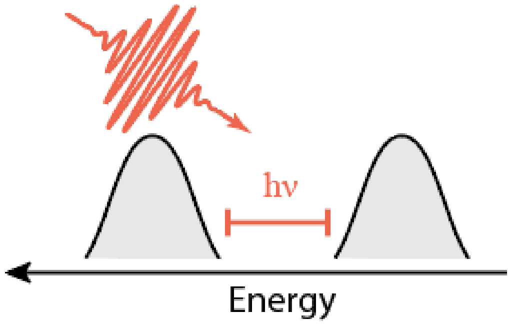
$$\mathcal{H}_{ij}^{(\gamma)} = K\tilde{S}_i^{\gamma}\tilde{S}_j^{\gamma} \longrightarrow \mathcal{H}_{ij}^{(\gamma)} = J\tilde{\boldsymbol{S}}_i\cdot\tilde{\boldsymbol{S}}_j + K\tilde{S}_i^{\gamma}\tilde{S}_j^{\gamma} + \sum_{\alpha\neq\beta}\Gamma_{\alpha\beta}\left(\tilde{S}_i^{\alpha}\tilde{S}_j^{\beta} + \tilde{S}_i^{\beta}\tilde{S}_j^{\alpha}\right)$$





PRL 112, 077204 (2014)

Phenomena during excitation



Free moving electron

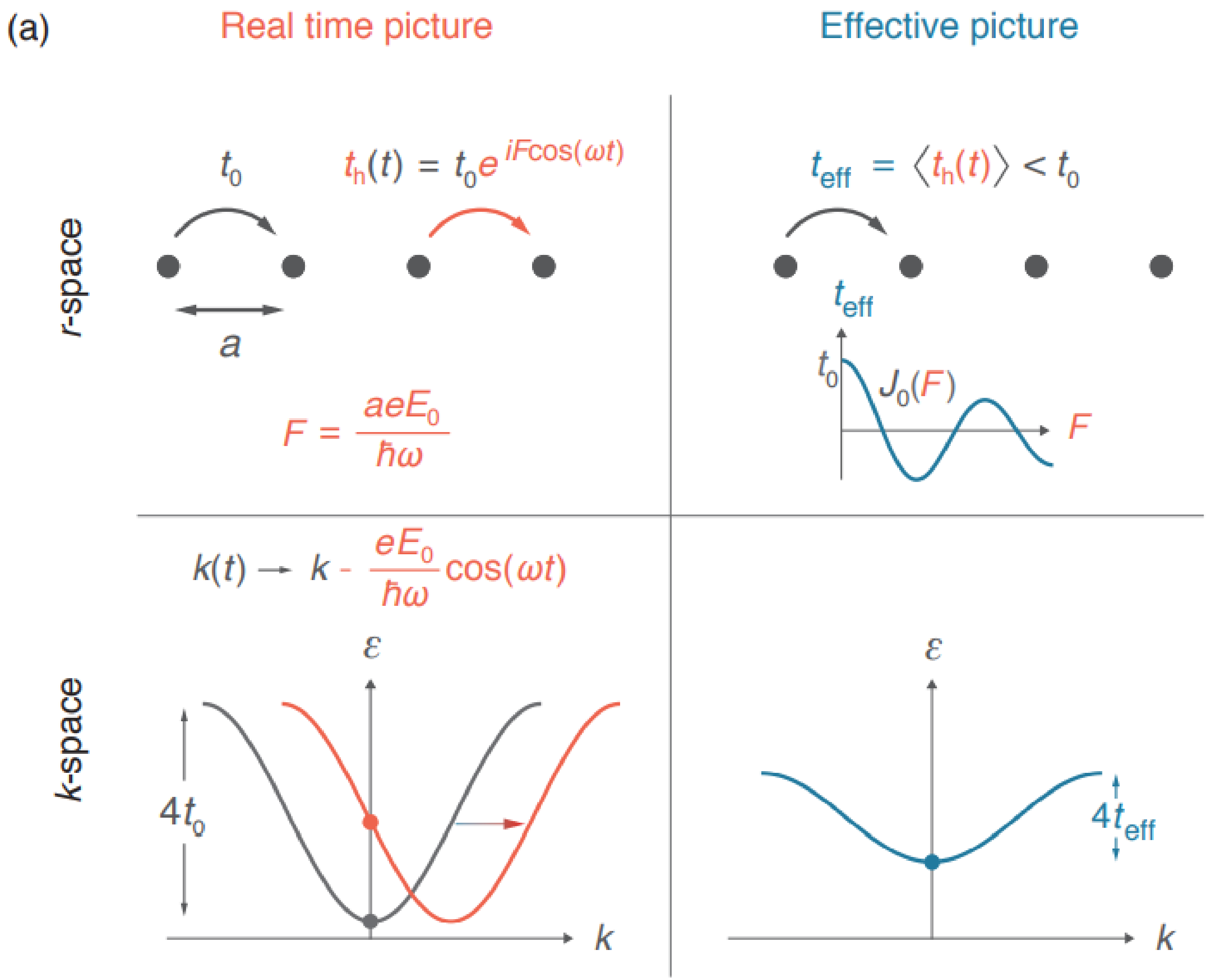
$$p \rightarrow p - eA(t)$$

One dimensional chain (Peierls substitution)

$$t_0 \rightarrow t_\mathrm{h}(t) = t_0 e^{ieaA(t)/\hbar}$$

$$A(t) = E_0/\omega \cos(\omega t)$$

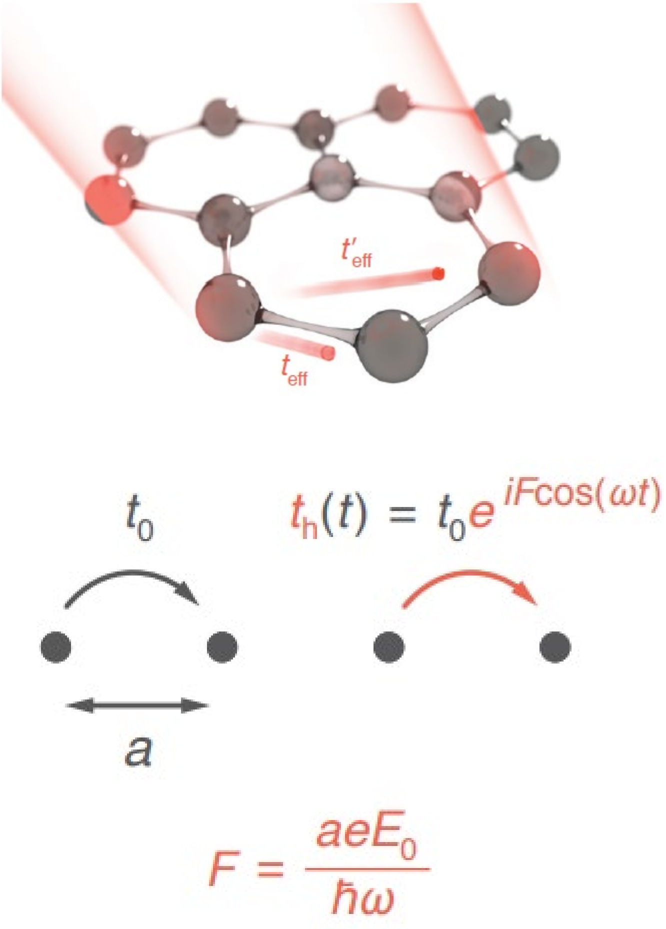
Periodically oscillating field

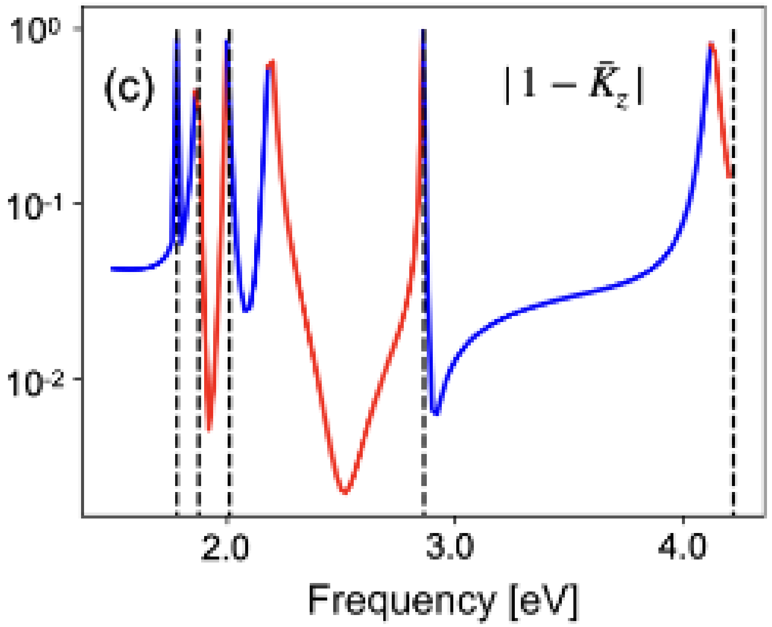


ADLT et al; Rev. Mod. Phys. 93, 041002 (2021)

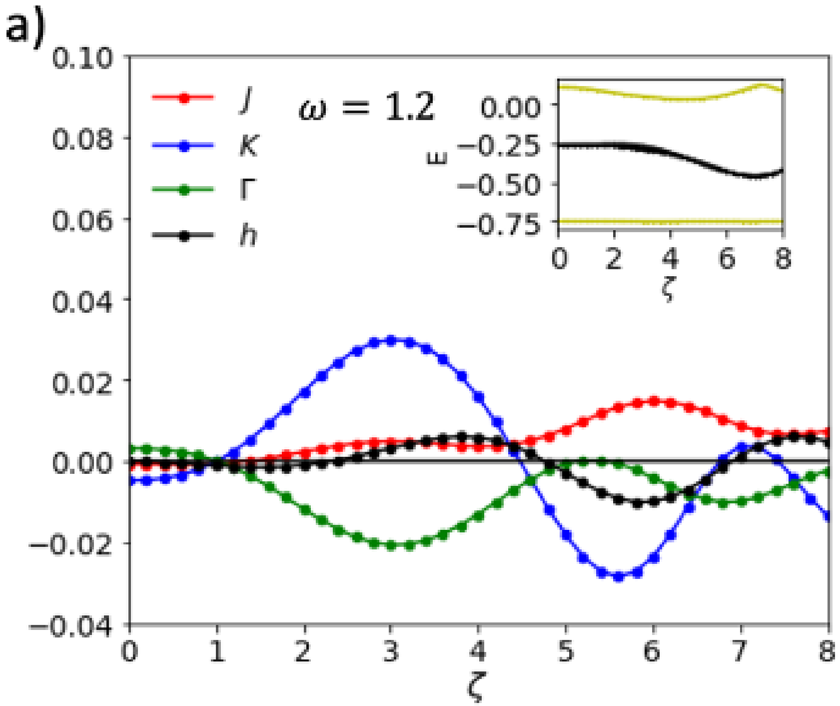
Light induced control of magnetic interactions in Kitaev materials

$$\mathcal{H}_{ij}^{(\gamma)} = J\tilde{\boldsymbol{S}}_i \cdot \tilde{\boldsymbol{S}}_j + K\tilde{S}_i^{\gamma}\tilde{S}_j^{\gamma} + \sum_{\alpha\neq\beta}\Gamma_{\alpha\beta}\left(\tilde{S}_i^{\alpha}\tilde{S}_j^{\beta} + \tilde{S}_i^{\beta}\tilde{S}_j^{\alpha}\right)$$



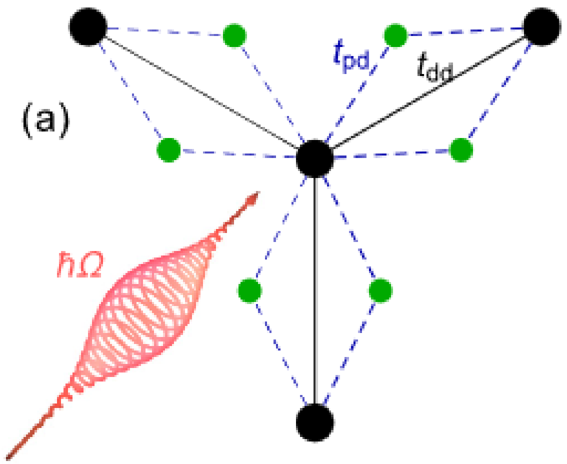


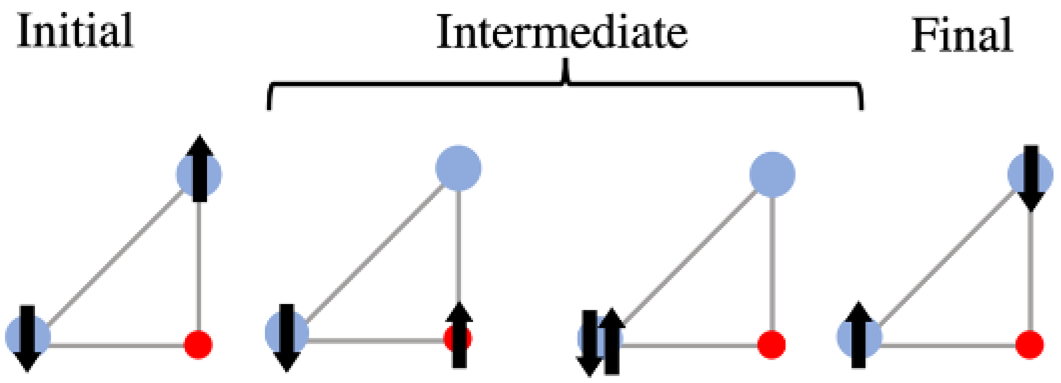
Phys Rev Res 4, L032036 (2022)

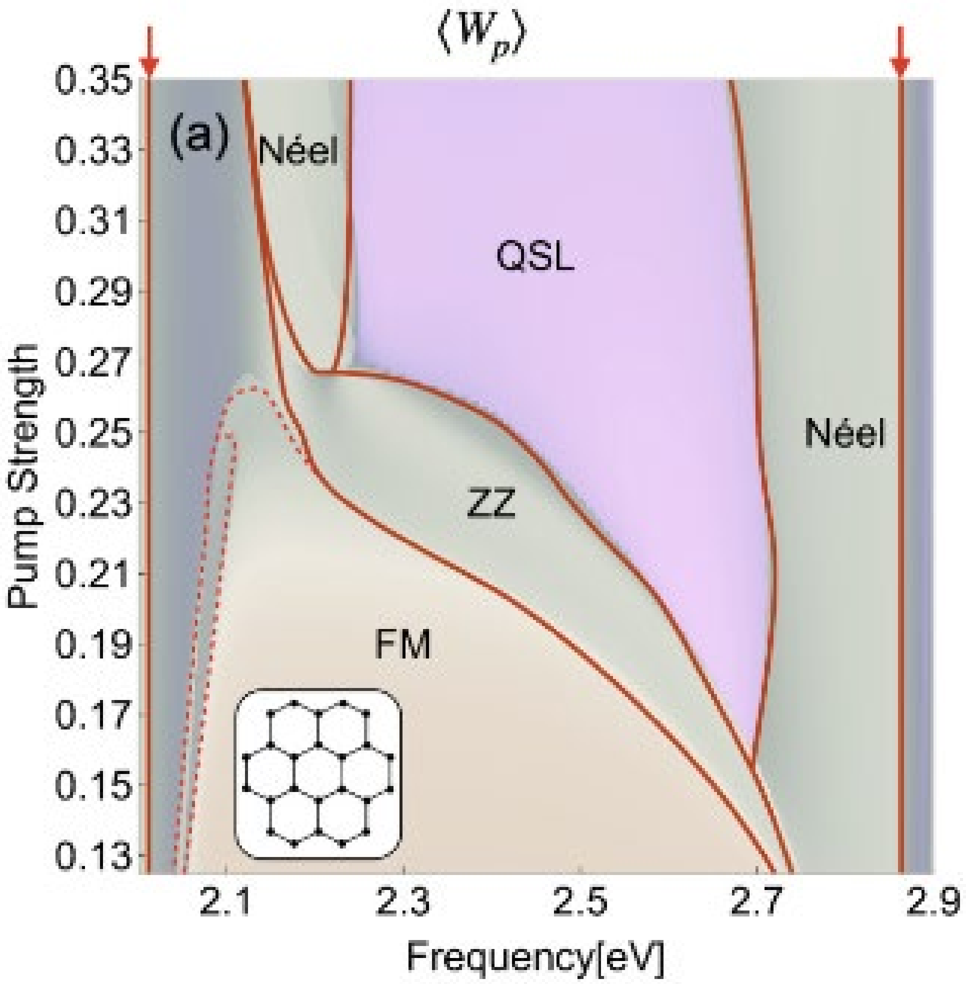


Comm Phys (2022) 5:157

Light induced control of magnetic interactions in Kitaev materials







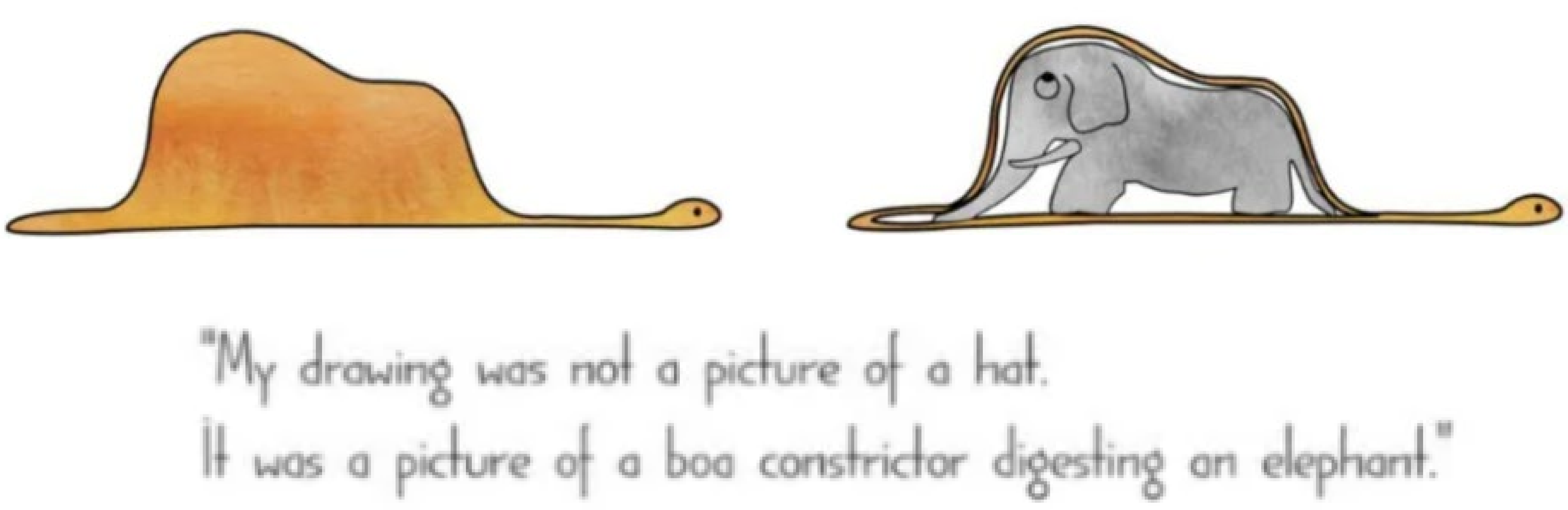
What are the spectroscopic signatures of this transition?

Phys Rev Res 4, L032036 (2022)

Outlook

- Broad, momentum-independent, magnetic excitations in $H_3LiIr_2O_6$ reflect Kitaev physics

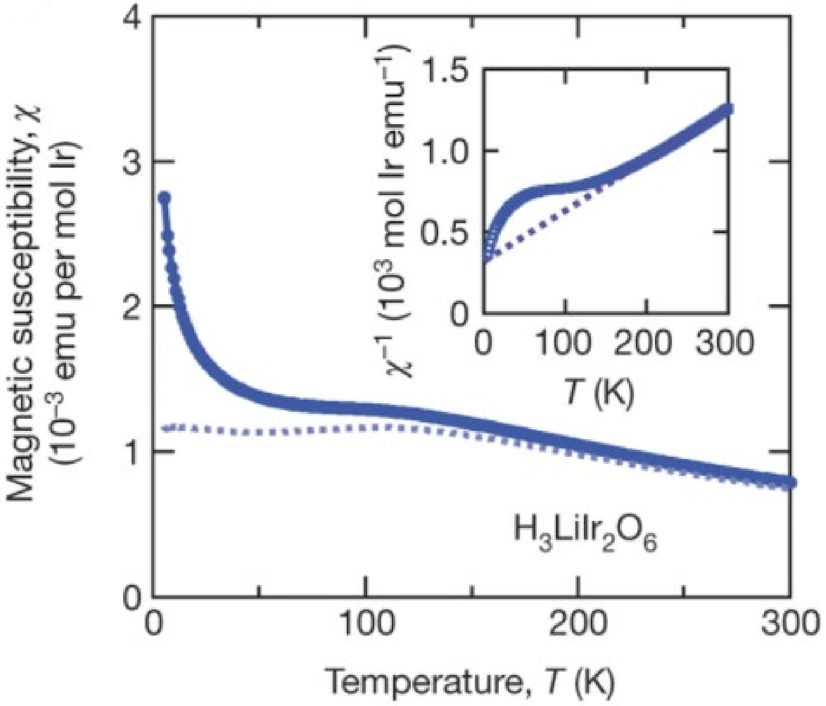
- Ultrafast control of magnetic exchanges (a preview)

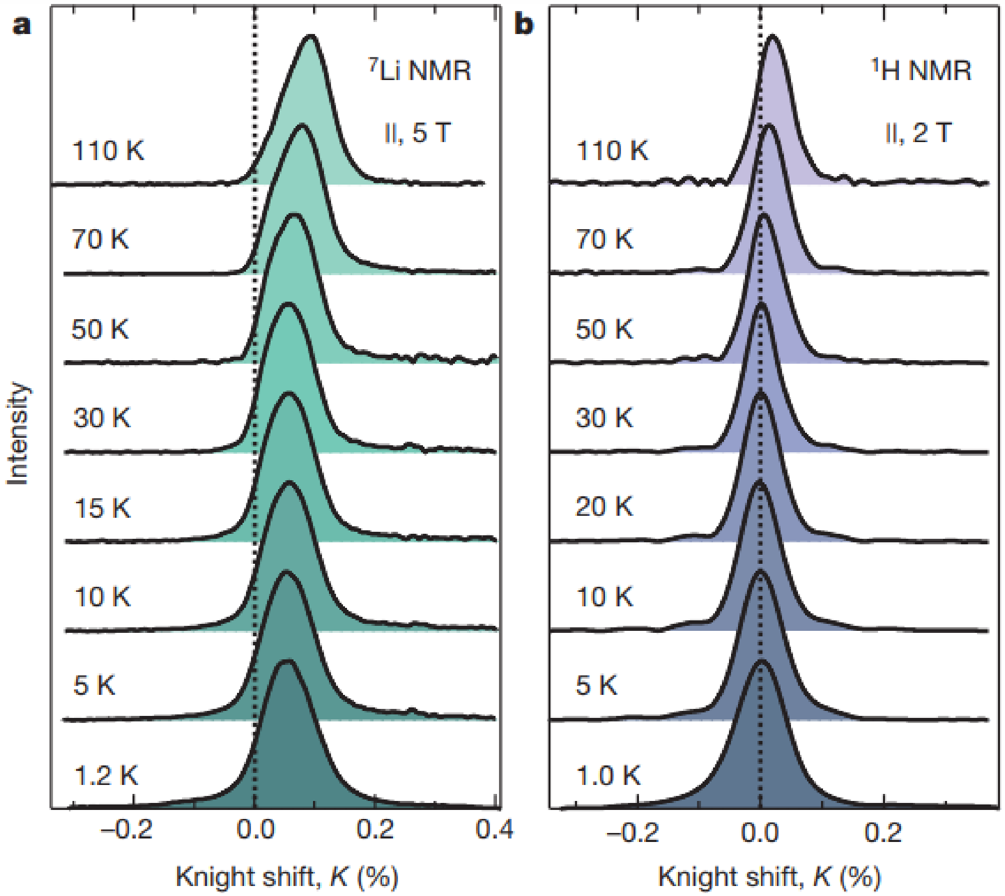


$H_3LiIr_2O_6$

A spin–orbital-entangled quantum liquid on a honeycomb lattice

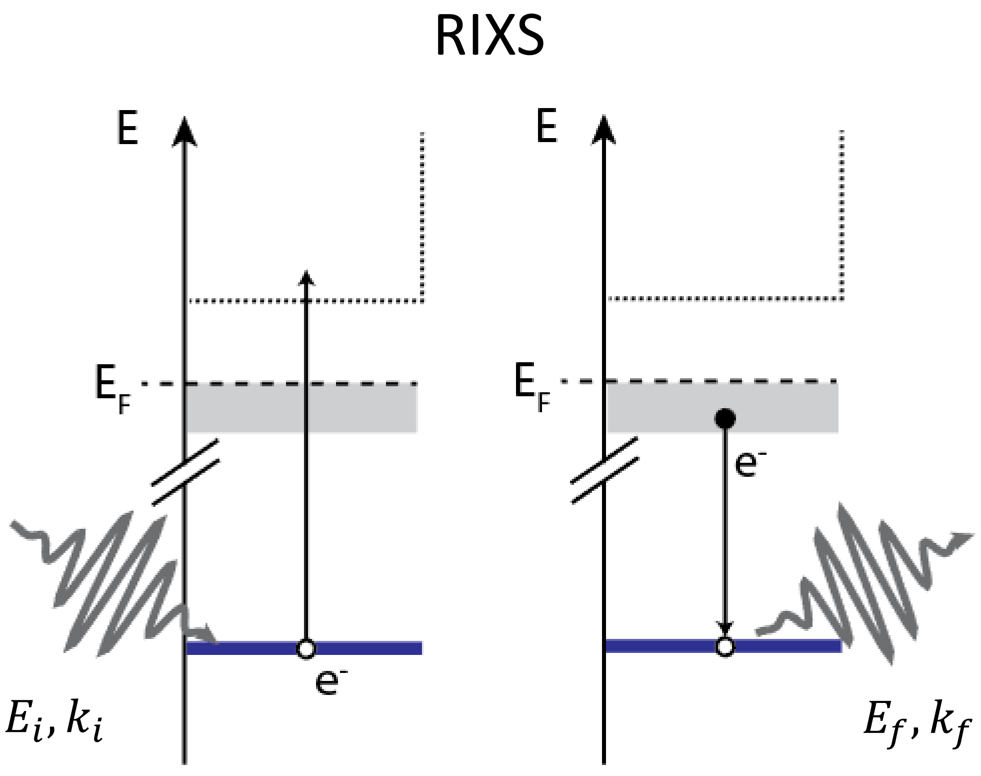
K. Kitagawa[1]*, T. Takayama[2]*, Y. Matsumoto[2], A. Kato[1], R. Takano[1], Y. Kishimoto[3], S. Bette[2], R. Dinnebier[2], G. Jackeli[2,4] & H. Takagi[1,2,4]





Nature 554, 341–345(2018)

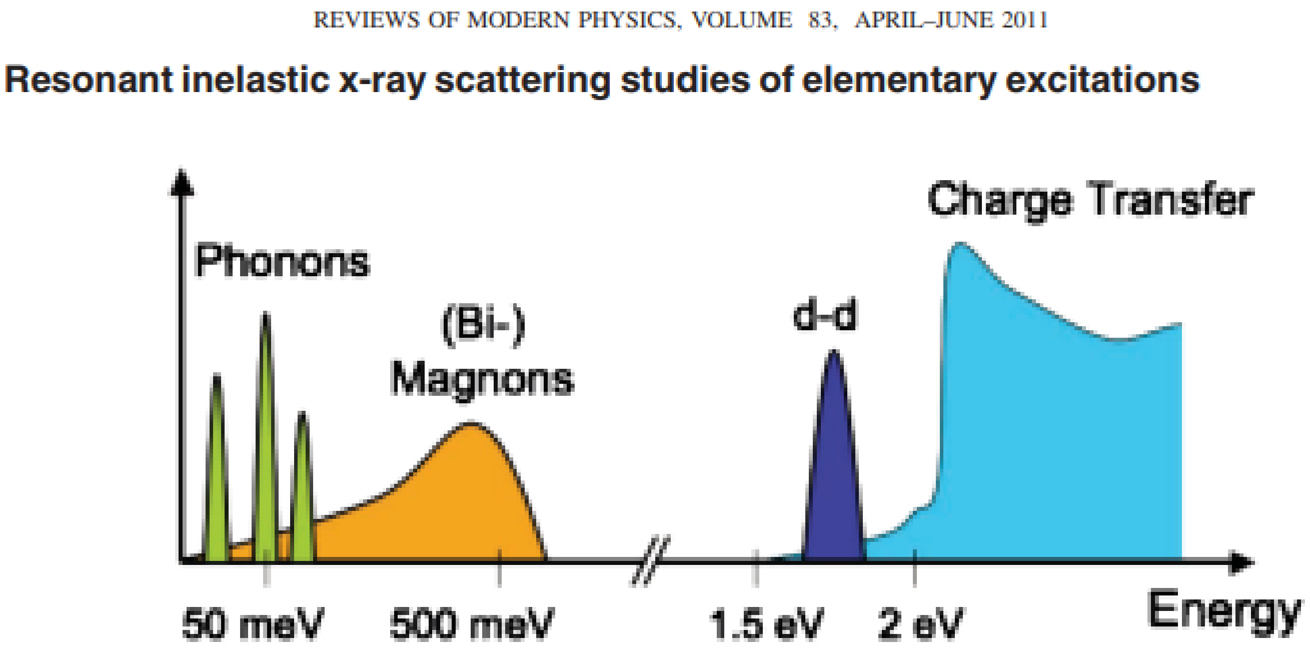
Resonant Inelastic X-ray Scattering (RIXS)



$$E_i = 2p_{3/2} \rightarrow 5d\ (L_3)$$

$$E_{Loss} = E_i - E_f$$

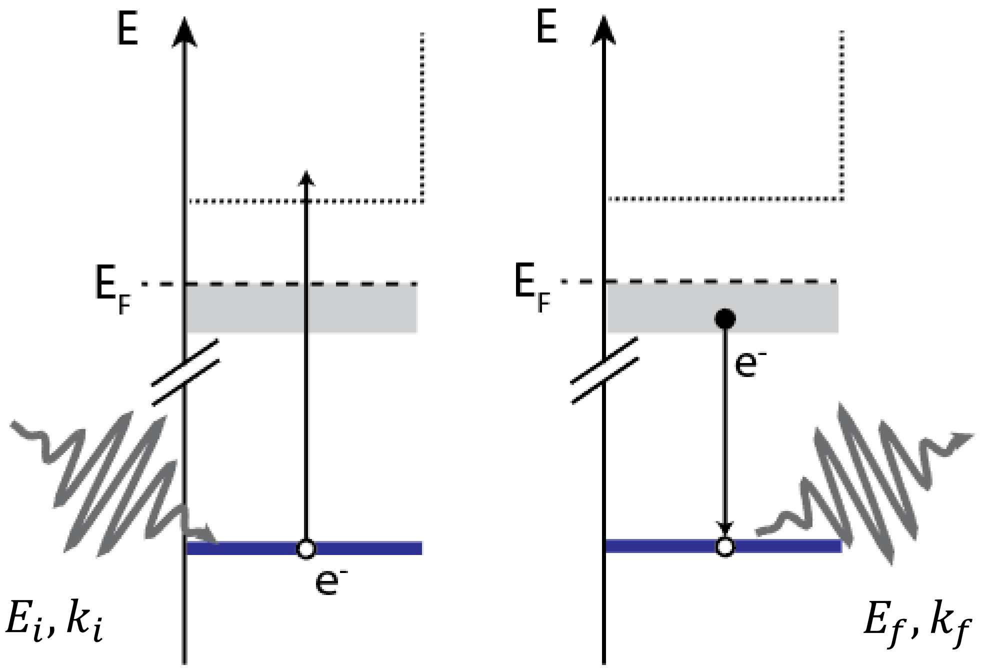
Intermediate state 5d^{5}*



ADLT et al, Phys. Rev. B 104, L100416 (2021)

Resonant Inelastic X-ray Scattering (RIXS)

RIXS

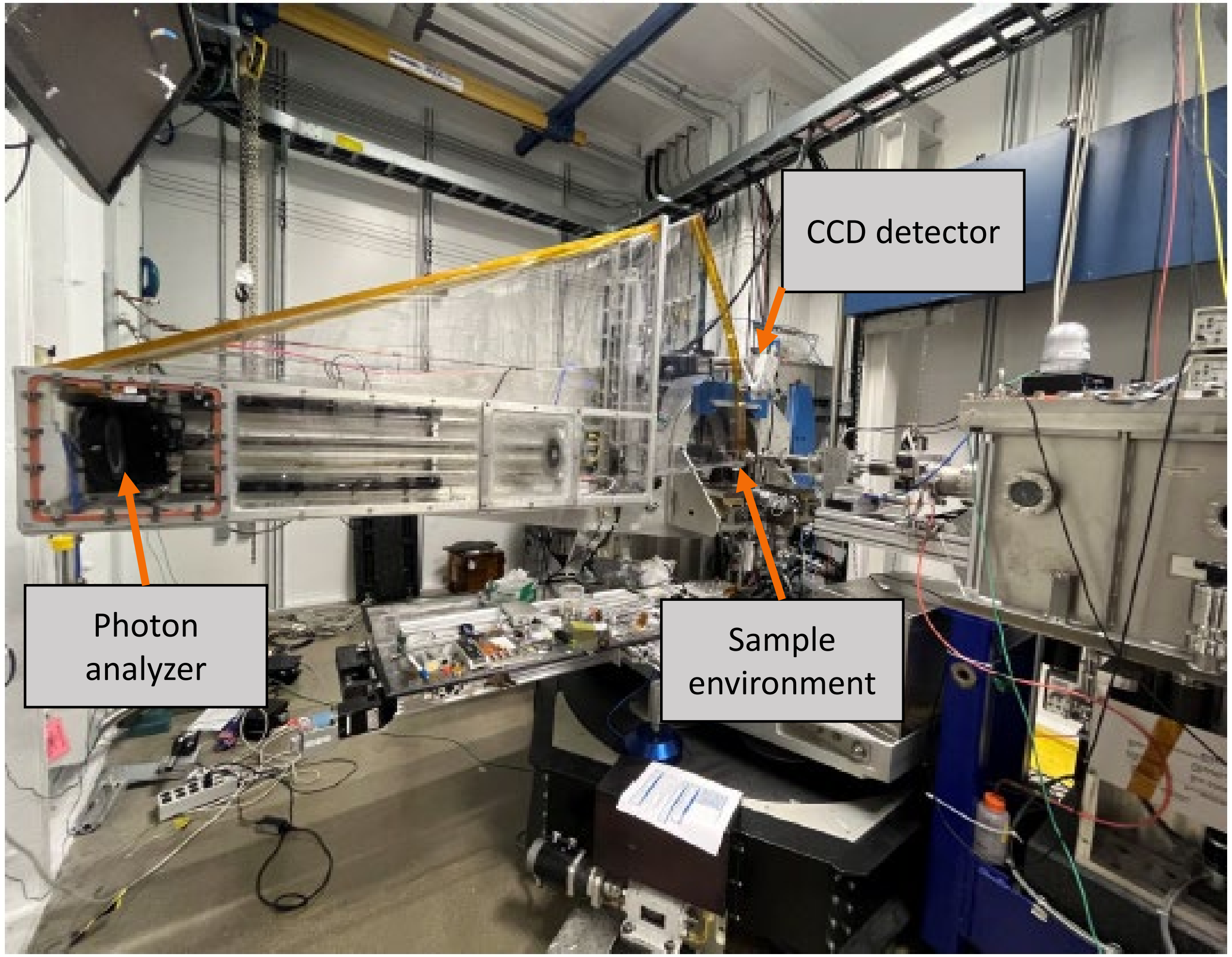


$$E_i = 2p_{3/2} \rightarrow 5d\ (L_3)$$

$$E_{Loss} = E_i - E_f$$

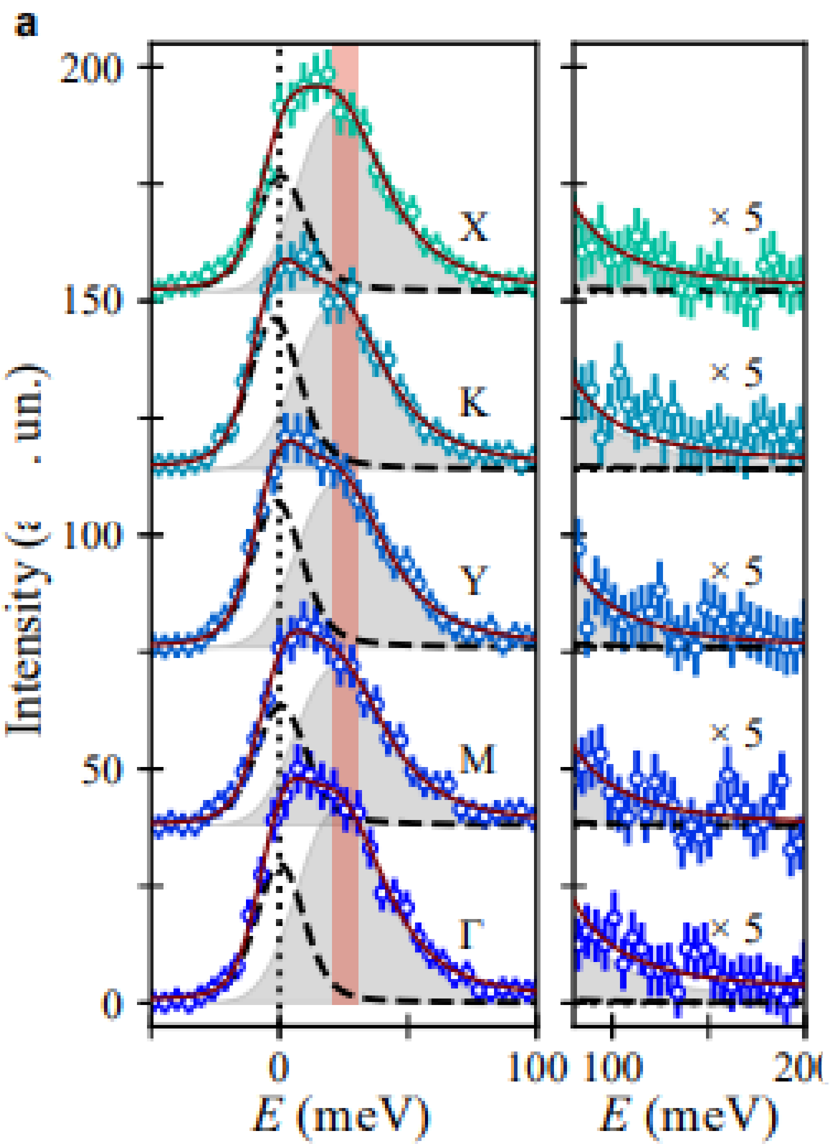
Intermediate state 5d⁵*

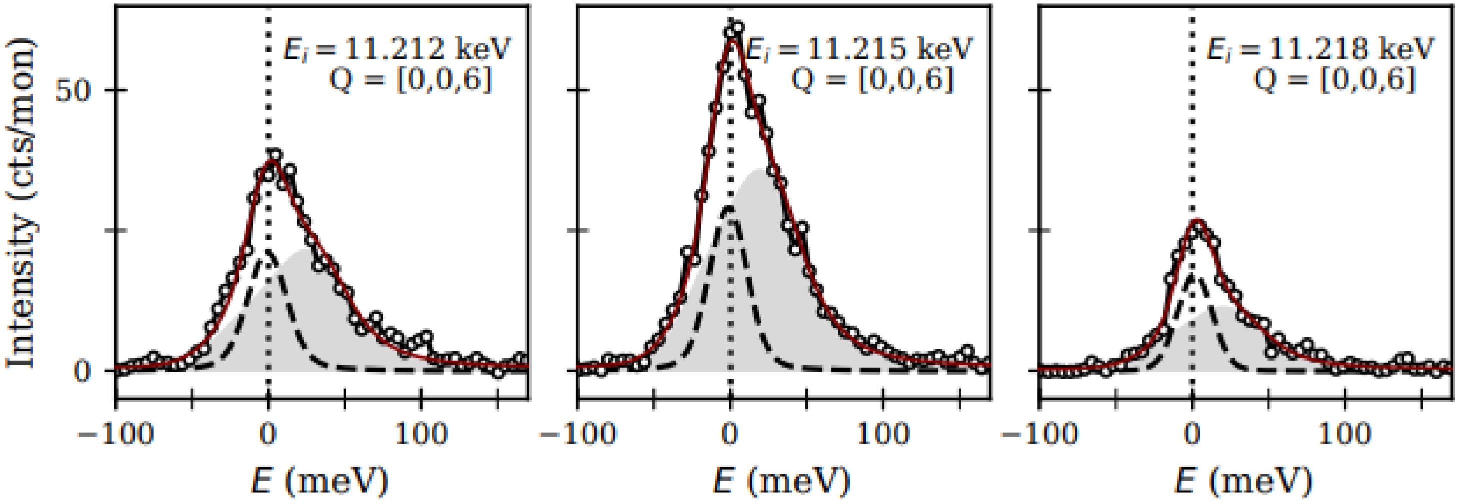
RIXS spectrometer (MERIX) @ APS 27-ID



ADLT et al, Phys. Rev. B 104, L100416 (2021)

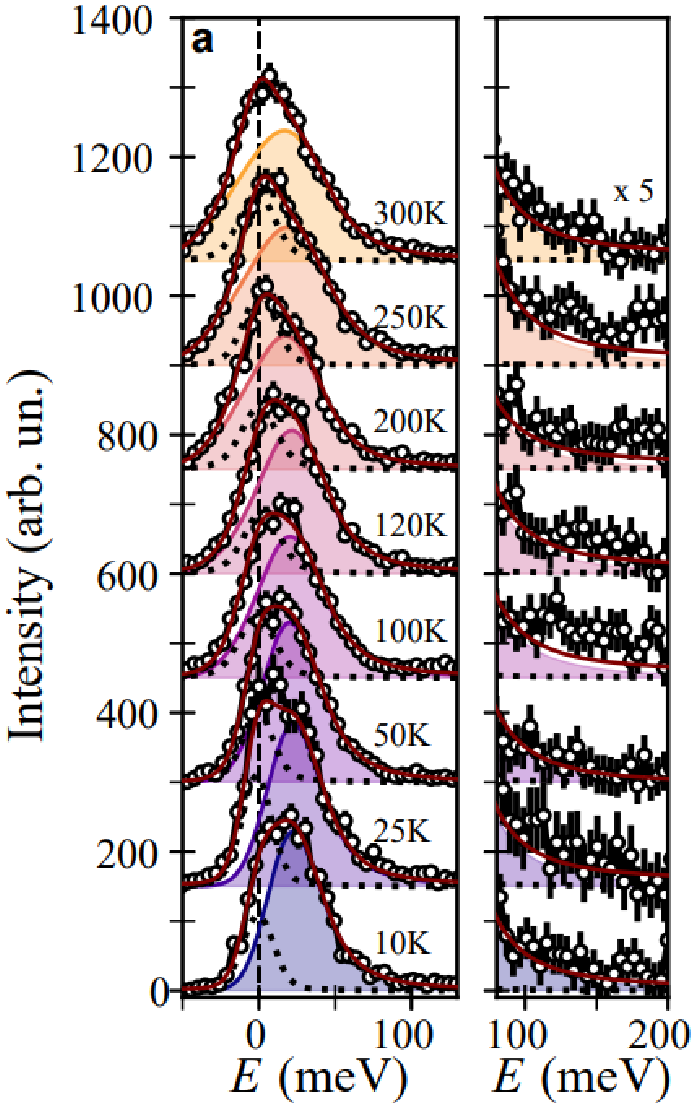
Lowe energy RIXS excitations in $H_3LiIr_2O_6$

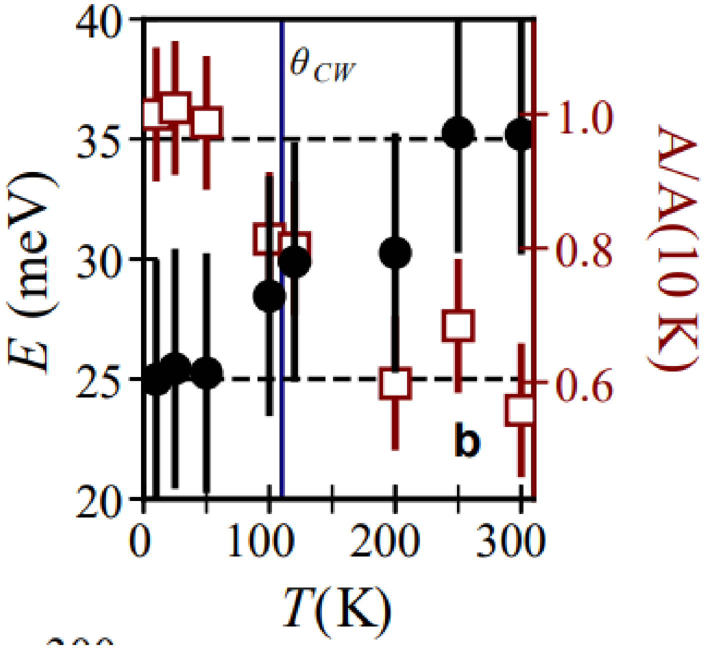


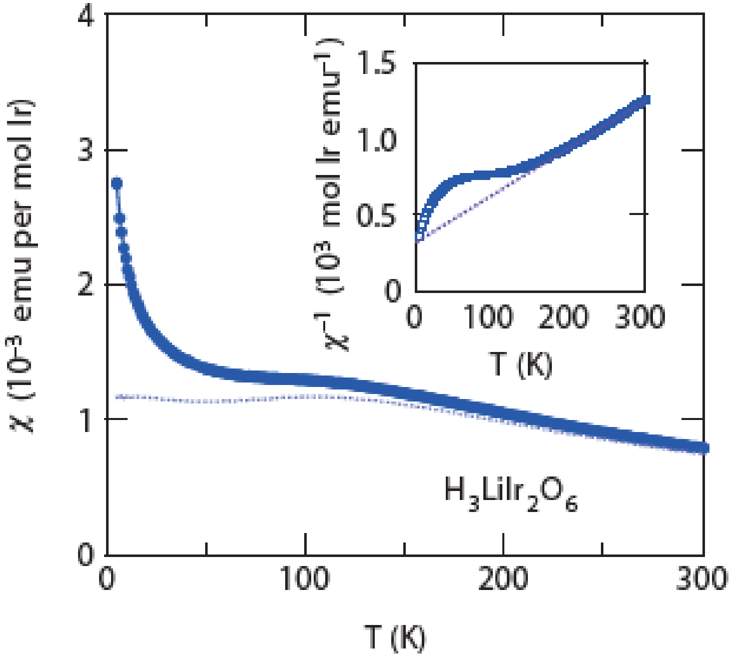


ADLT et al, arxiv: 2302.07907

Continuum of magnetic excitations

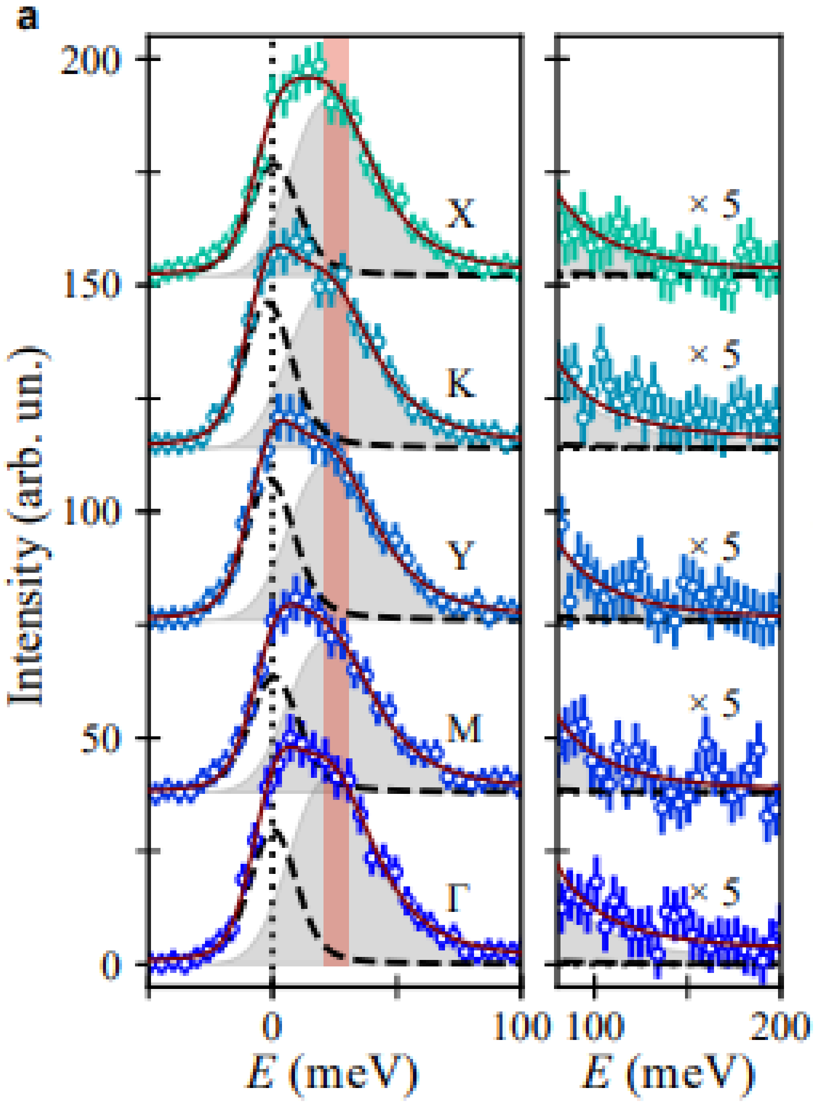


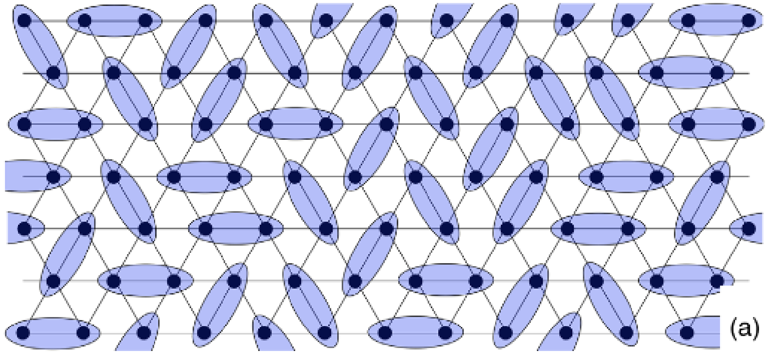


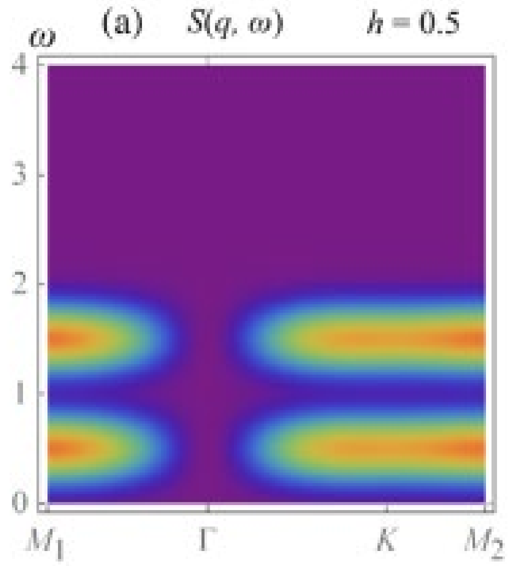


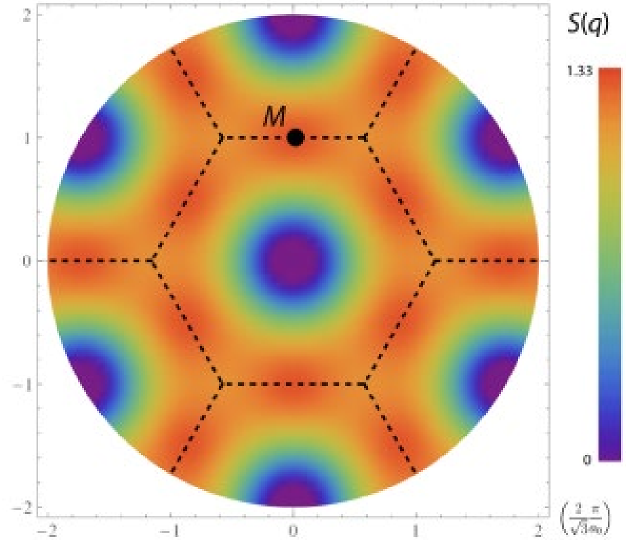
ADLT et al, arxiv: 2302.07907

Data is not consistent with a random valance bond model





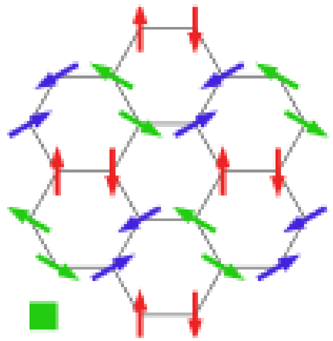




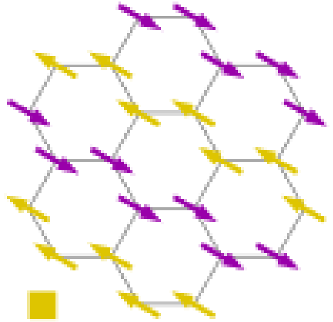
Phys Rev X 8, 031028 (2018)

ADLT et al, arxiv: 2302.07907

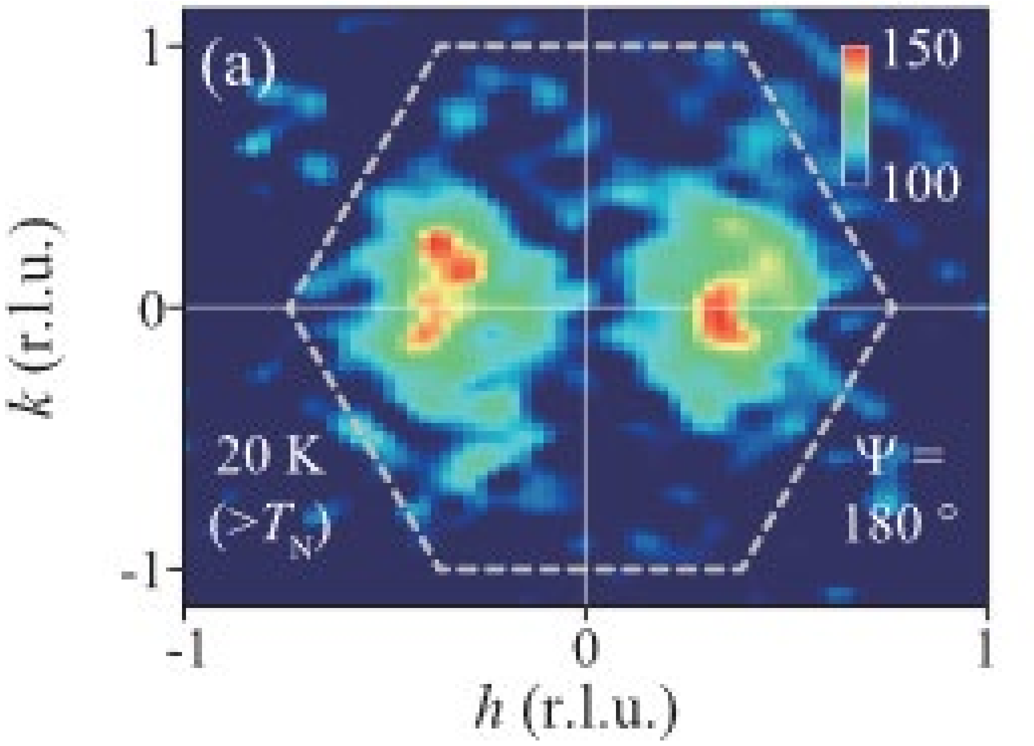
AFM short range correlations in Kitaev magnets

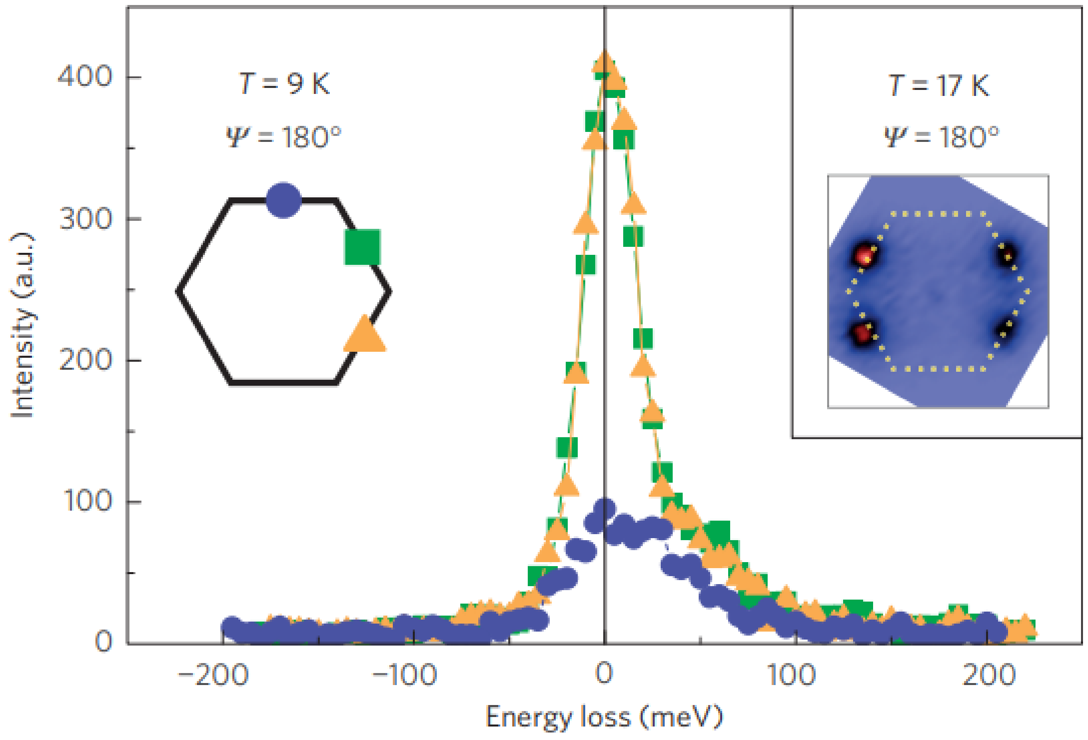


α - Li_2IrO_3
$T > T_N$



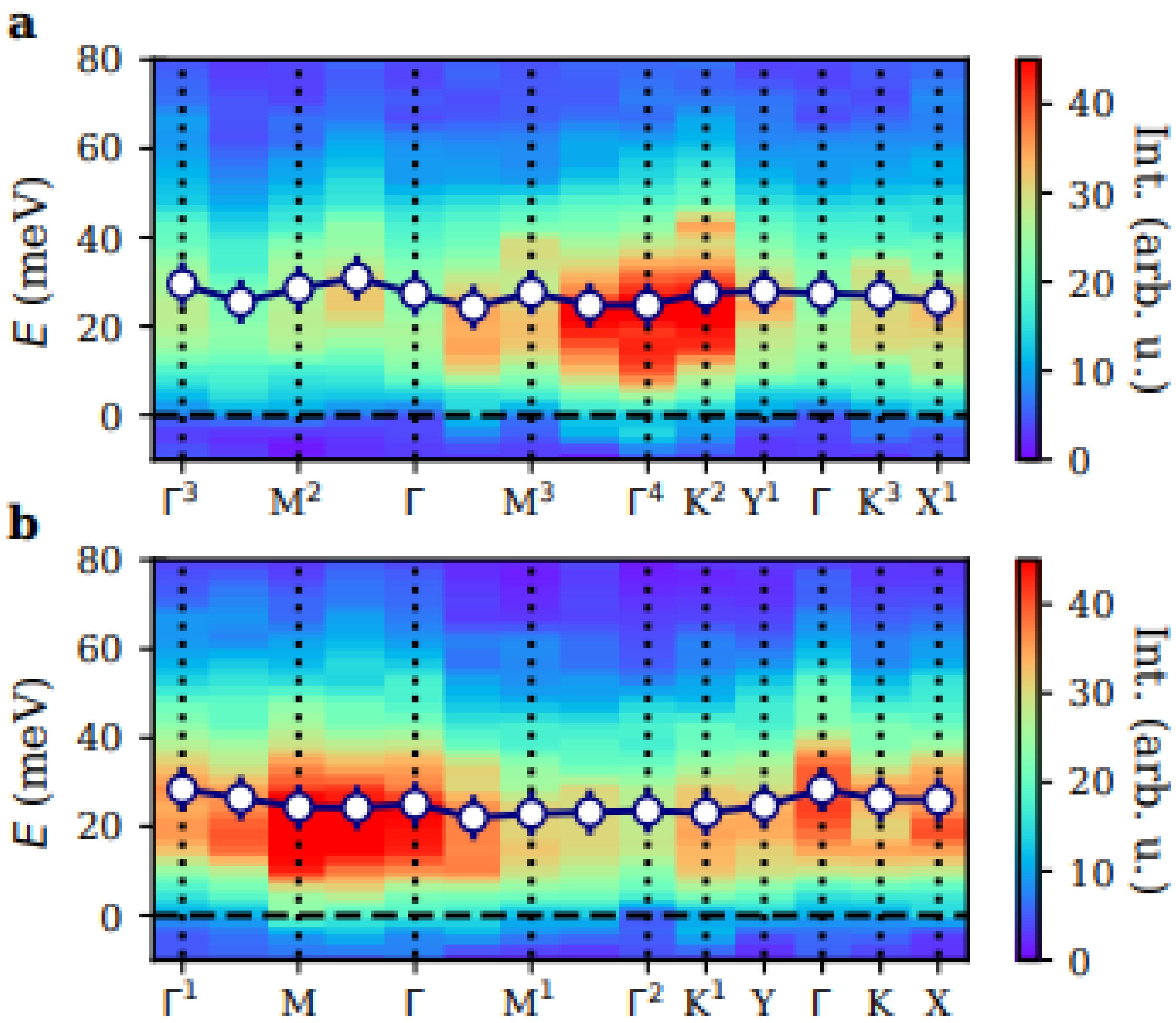
Na_2IrO_3
$T > T_N$

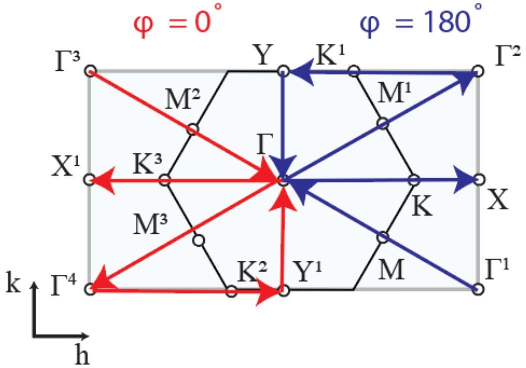




Nat Phys 11, 462 (2015)

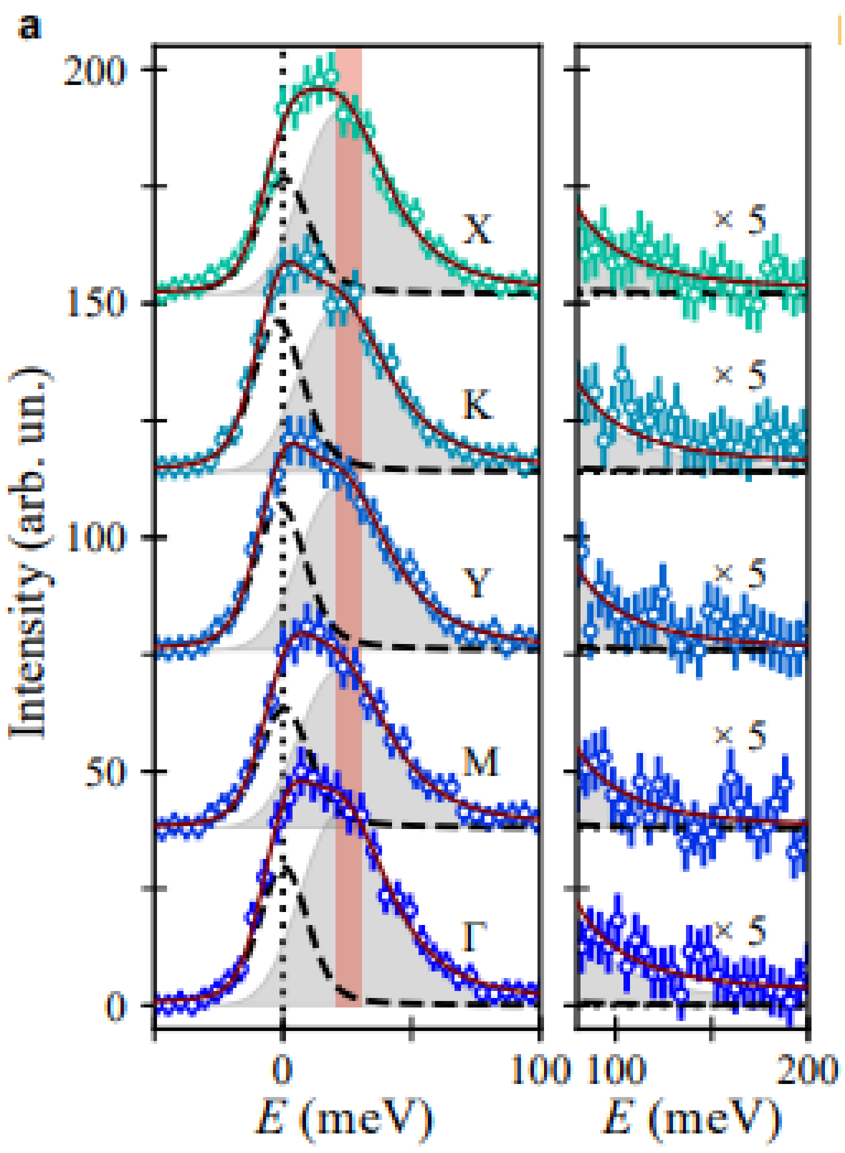
Absence of short-range correlations

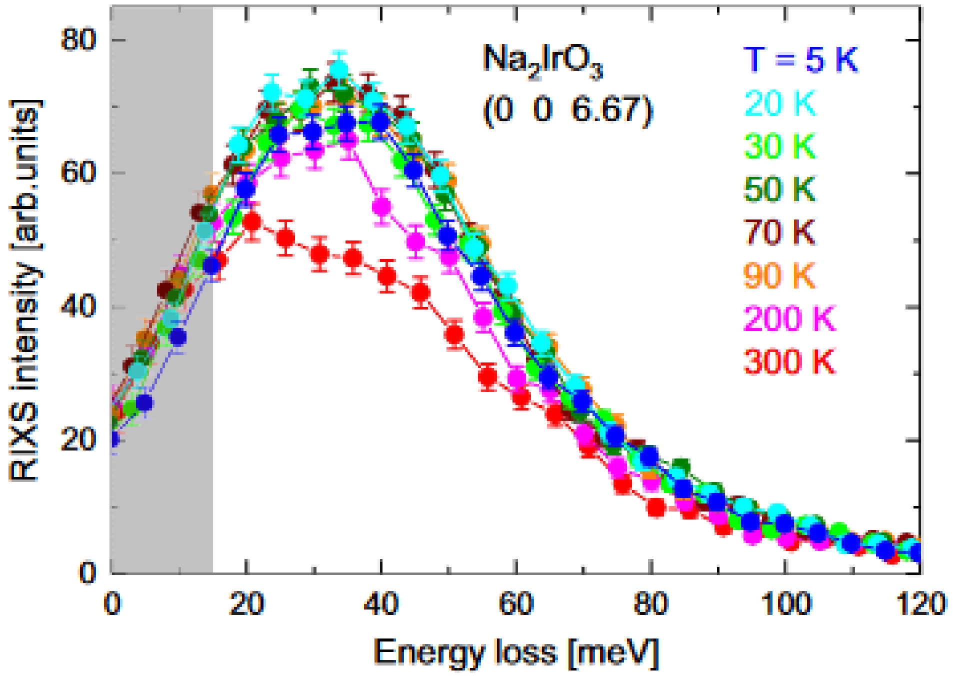




ADLT et al, arxiv: 2302.07907

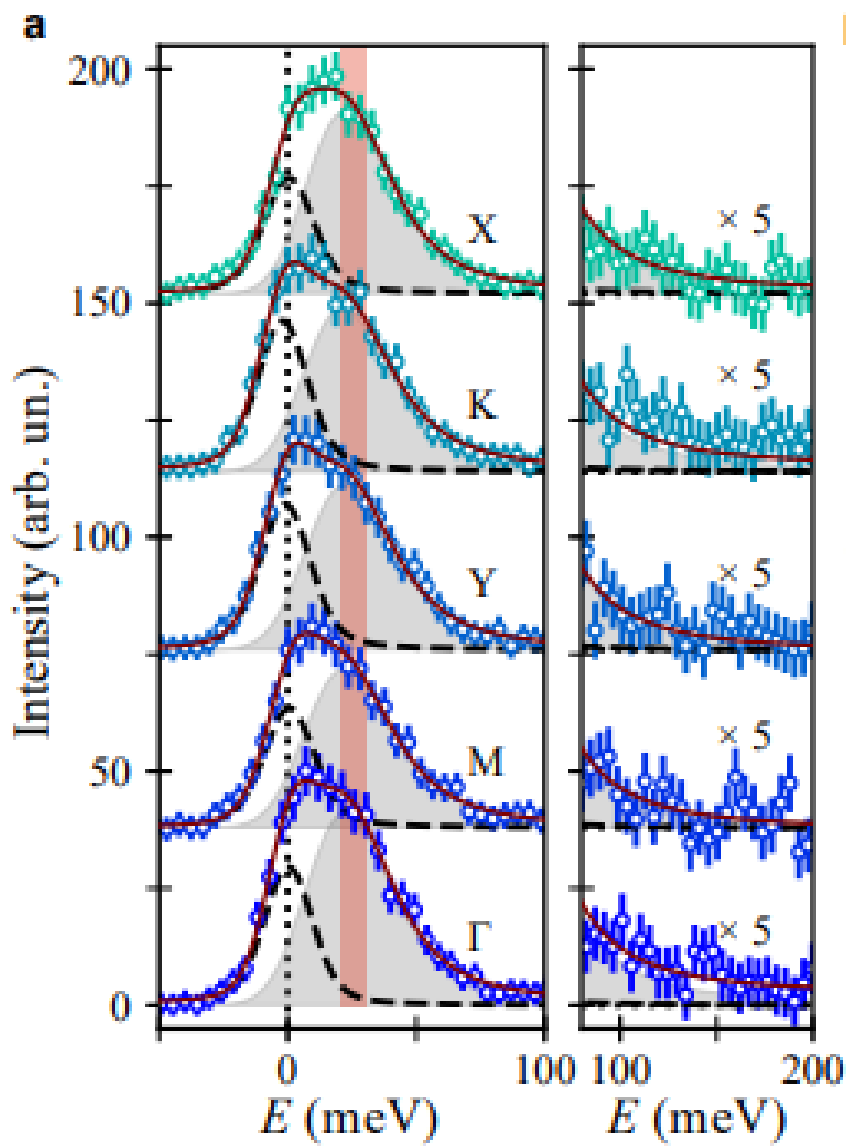
Comparison to Kitaev QSL

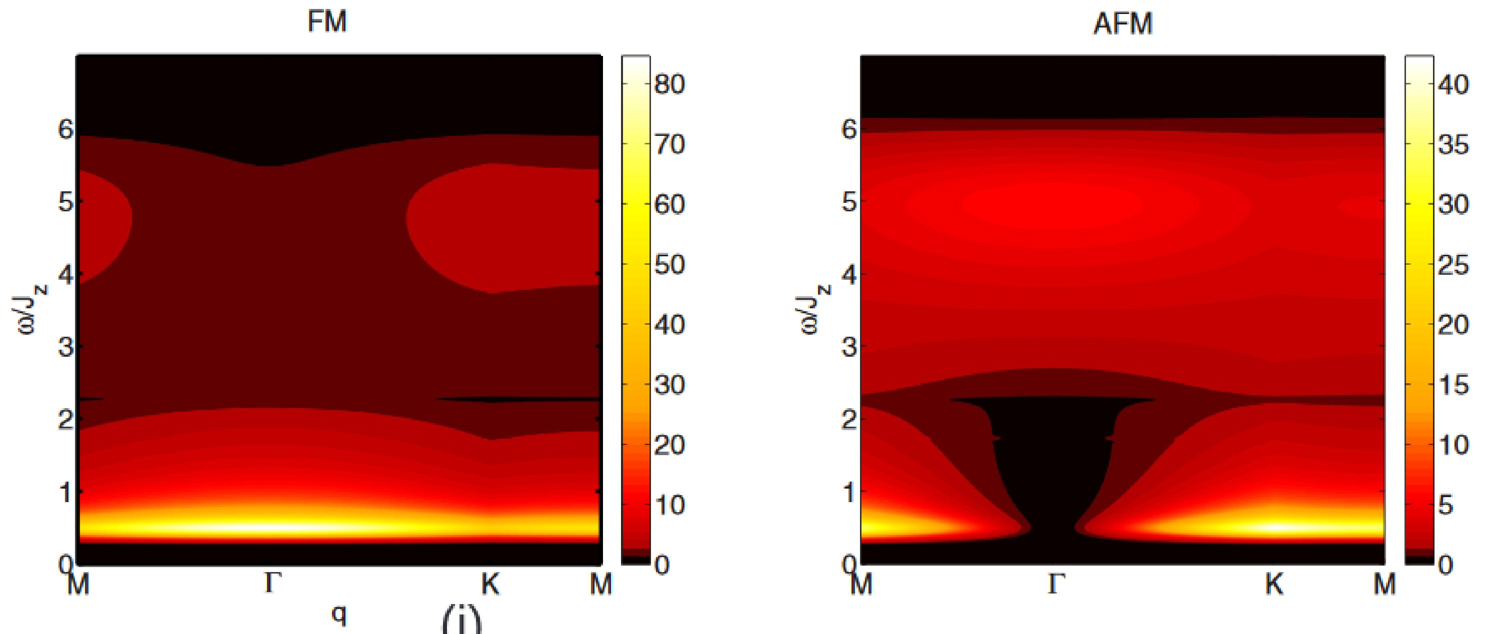




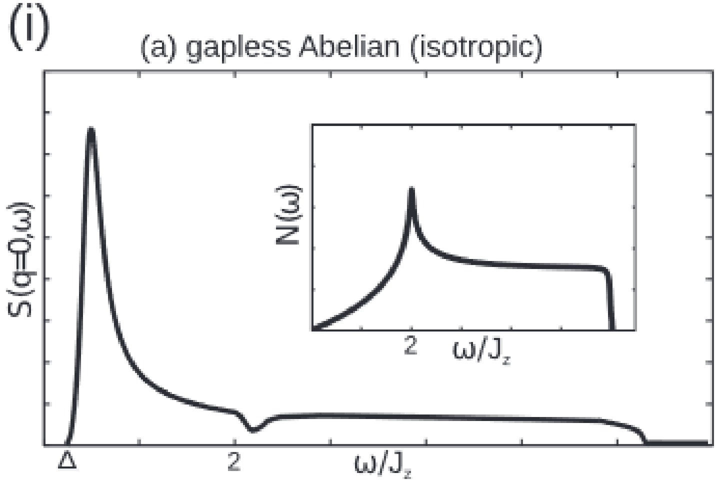
PRL 112, 207203 (2014)
PHYSICAL REVIEW B 92, 115127 (2015)
PHYSICAL REVIEW B 97, 134432 (2018)

Comparison to Kitaev QSL





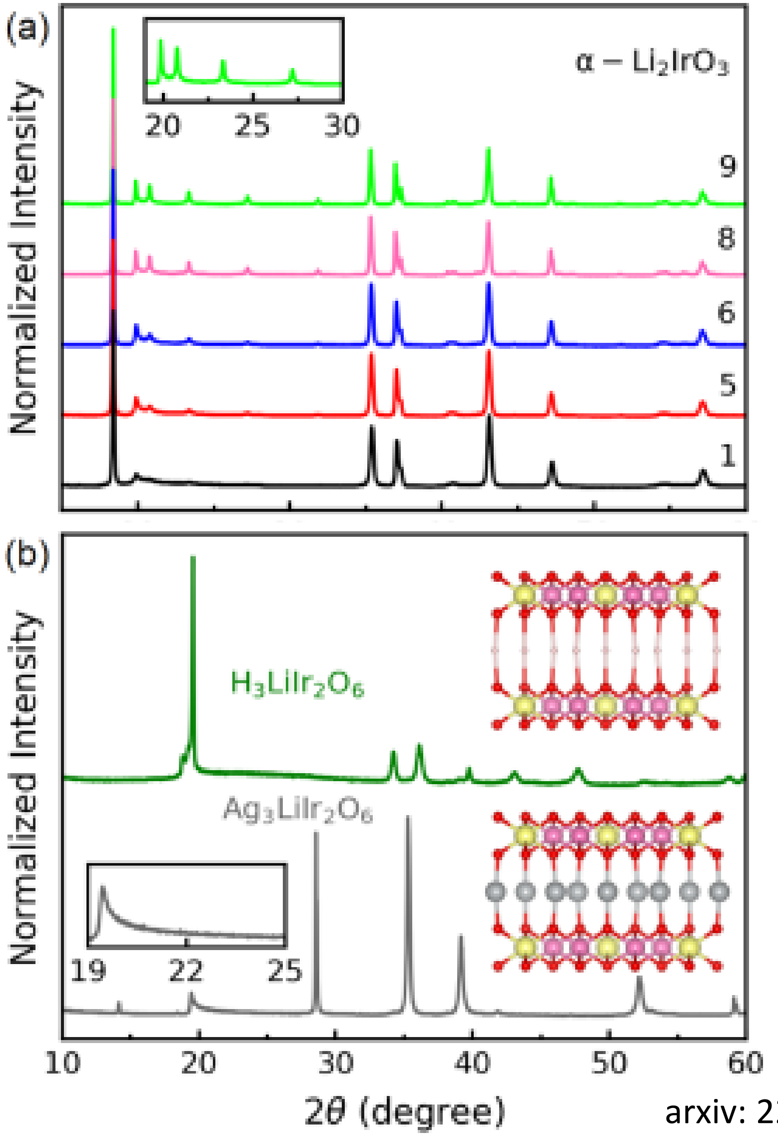
|K| = 25 meV,
Γ,J < 5meV



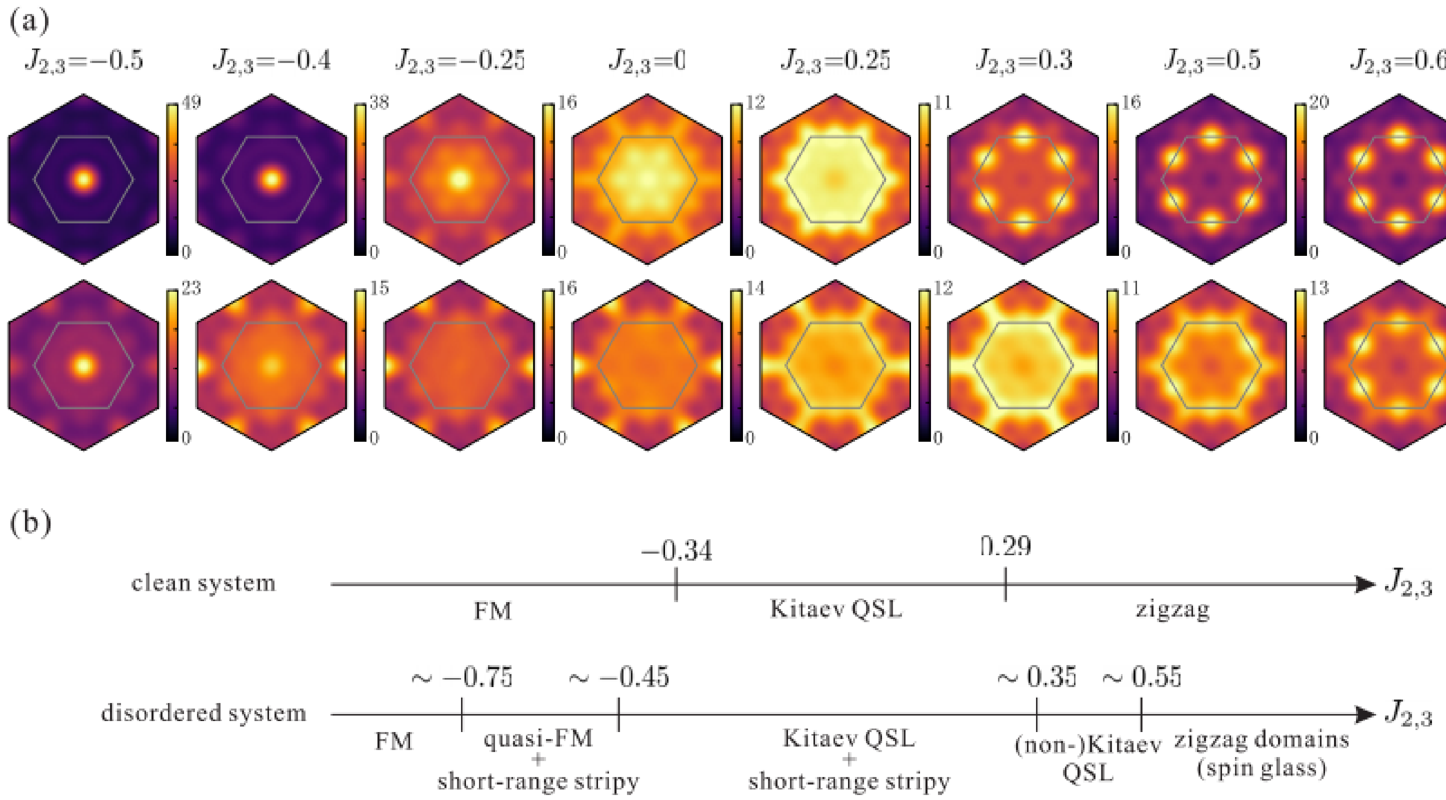
PRL 112, 207203 (2014)
PHYSICAL REVIEW B 92, 115127 (2015)
PHYSICAL REVIEW B 97, 134432 (2018)

Effect of disorder in S(Q)

Stacking faults

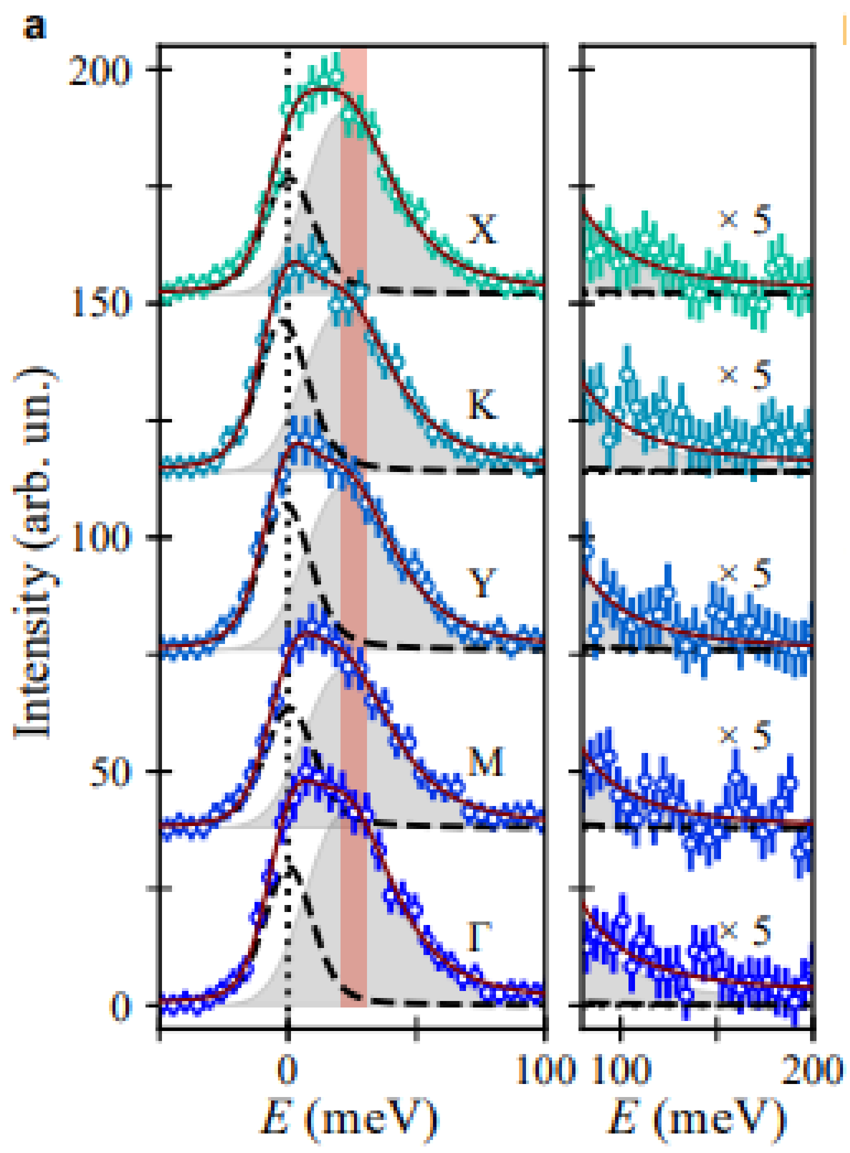


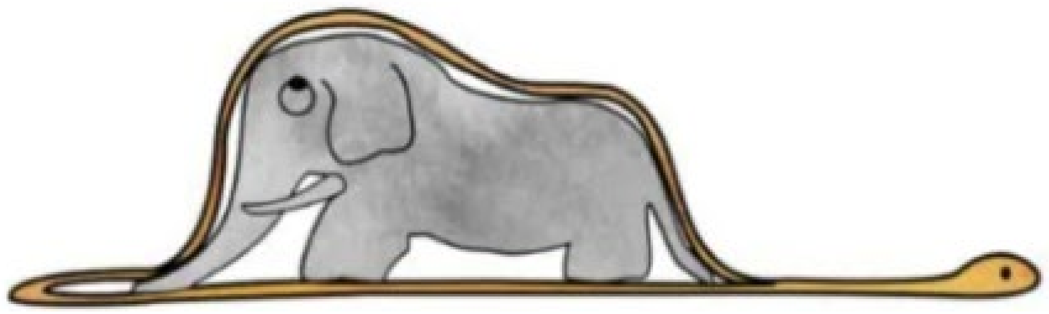
arxiv: 2201.06085



Phys. Rev. Research 5, 023009 (2023)

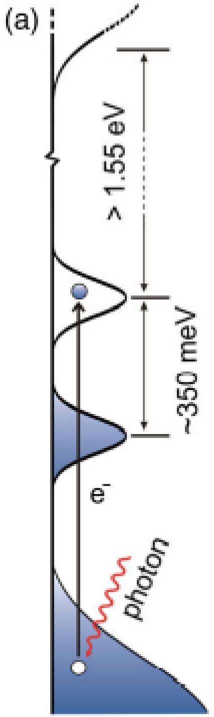
Take home message



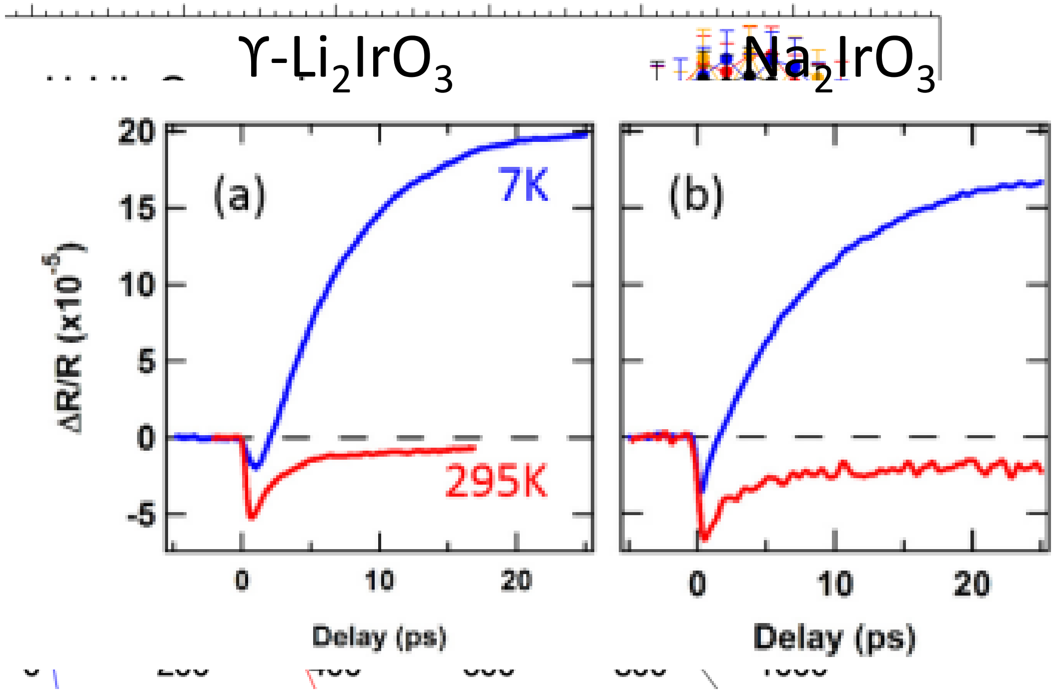


- Spin excitation is characterized by a broad continuum of comparable intensity to the elastic line, centered at E ~ 25 meV with a tail extending up to 150 meV
- Disorder induced absence of momentum dependence of the magnetic excitations
- $H_3LiIr_2O_6$ is proximal to bond-disordered KQSL with dominant K ~ 25 meV

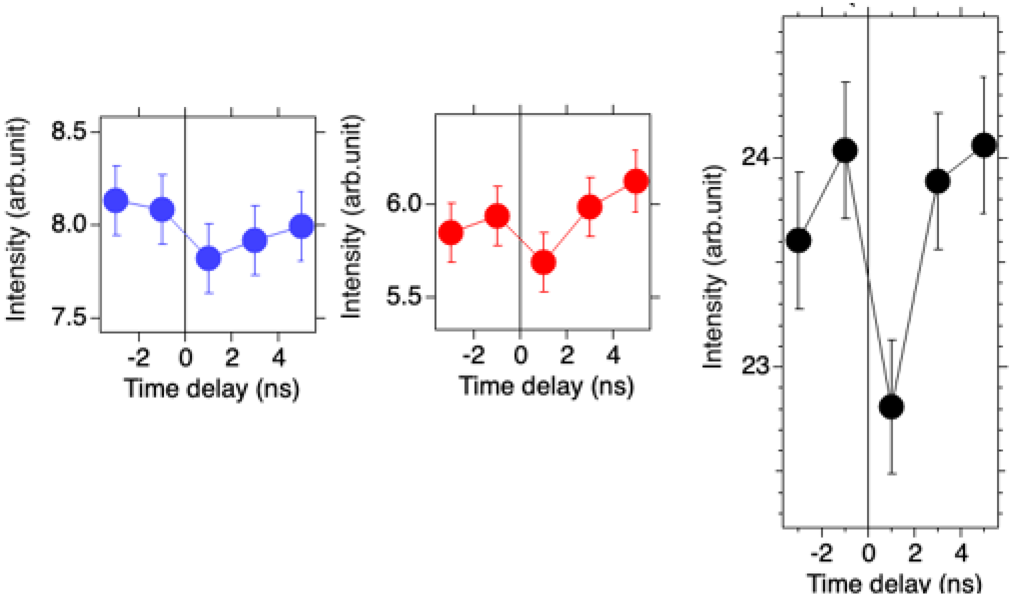
Some preliminary synchrotron tr-RIXS results



Υ-Li_2IrO_3 Na_2IrO_3



E (mev)



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