

Dynamics and transport in quantum spin liquids



Leon Balents, KITP

FRUMAG, PCS IBS, October 2019

Collaborators

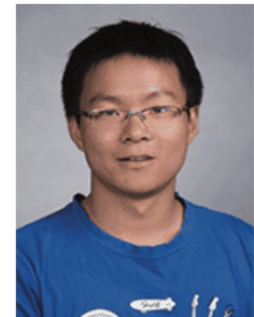


Oleg Starykh, U. Utah



Urban Seifert, TU Dresden

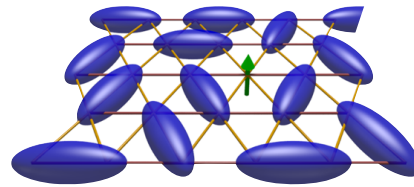
experimental inspiration



Rick Averitt Gufeng Zhang
UCSD

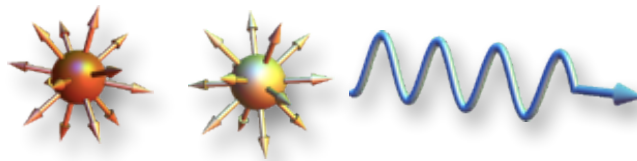
Classes of QSLs

- Topological QSLs



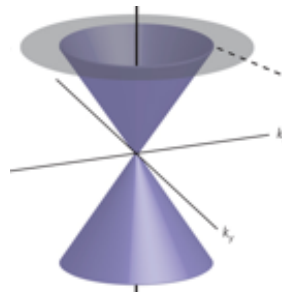
anyons,
spinons

- U(1) QSL



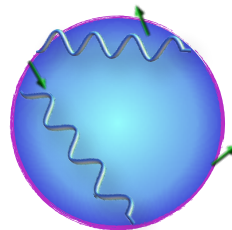
compact U(1)

- Dirac QSLs



QED₃

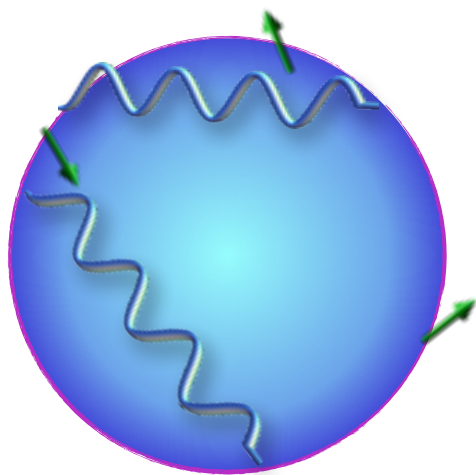
- Spinon Fermi surface



non-Fermi
liquid "spin
metal"

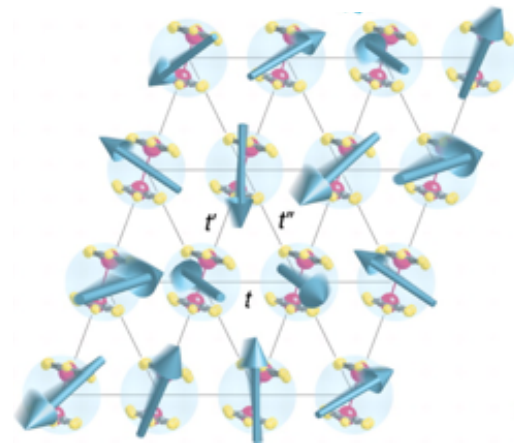
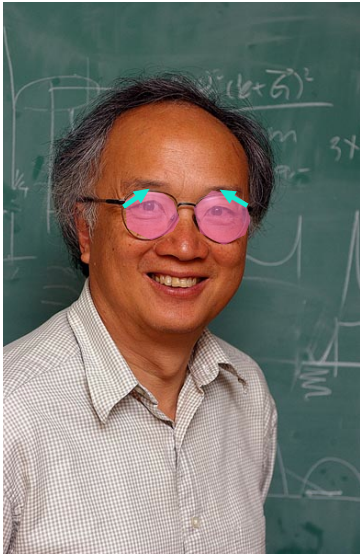
Spinon Fermi surface

$$|\Psi\rangle = \prod_i \hat{n}_i (2 - \hat{n}_i) \prod_{k < k_F} c_{k\uparrow}^\dagger c_{k\downarrow}^\dagger |0\rangle$$

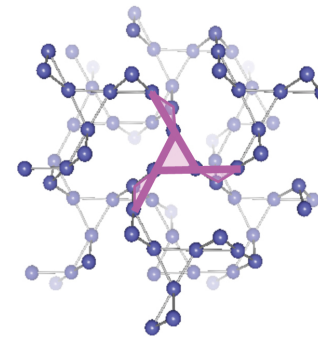


- The most gapless/highly entangled QSL state
- Like a "metal" of neutral fermions w/ a U(1) gauge field
- Prototype "non-Fermi liquid" state of great theoretical interest

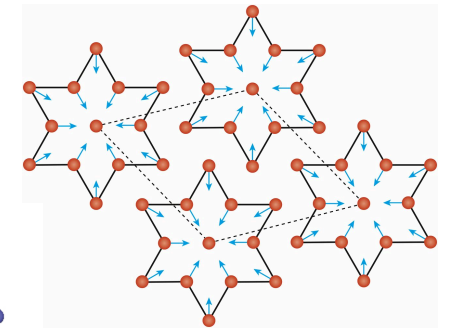
Spinon Fermi surface



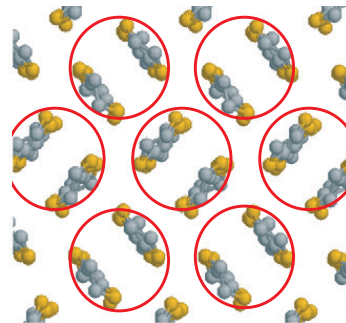
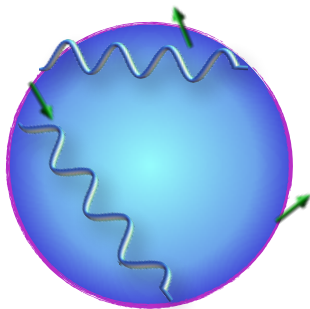
k-ET



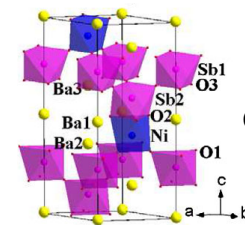
Na₄Ir₃O₈



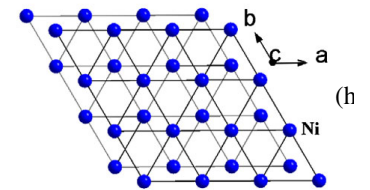
1T-TaS₂



dmit



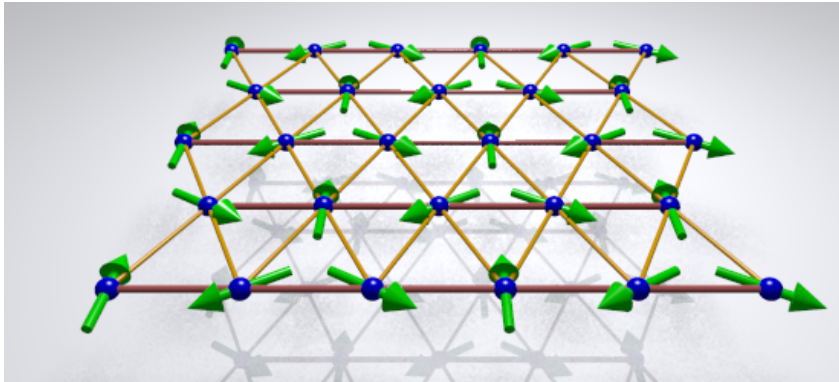
(e)



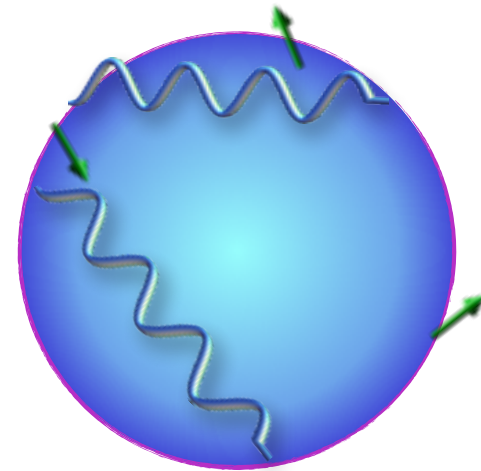
(h)

Ba₃NiSb₂O₉

Triangular lattice w/ ring exchange

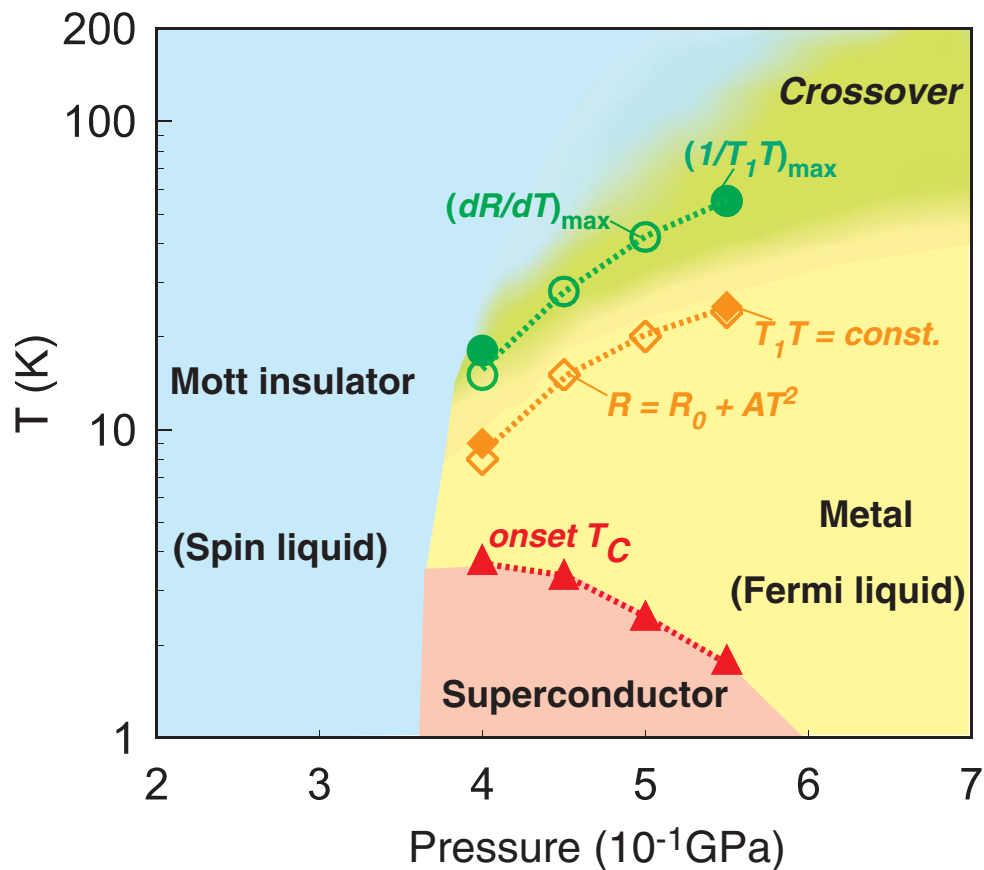
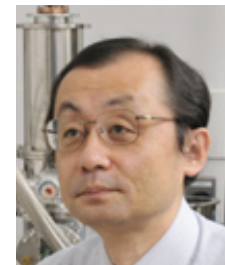
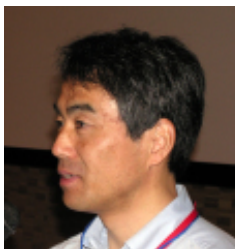


- Motrunich (2005): ring exchange stabilizes a spin liquid

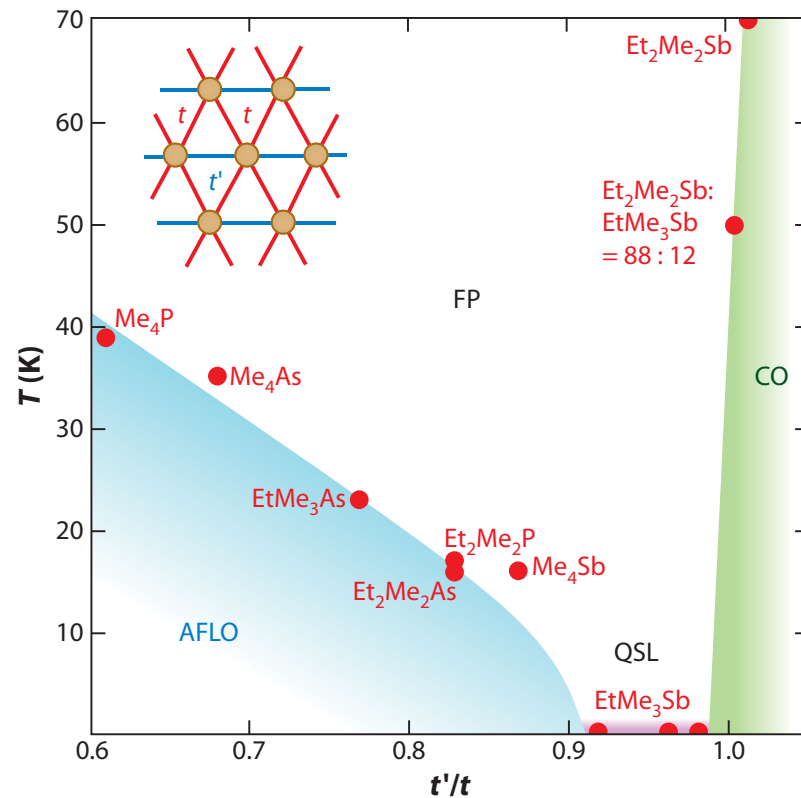


- Motrunich, Lee/Lee: spin liquid state favored by ring exchange is the "spinon Fermi sea" state

triangular organics



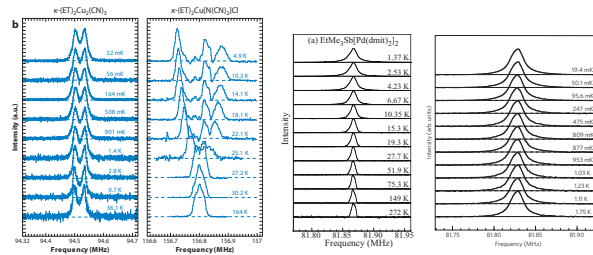
K. Kanoda group (2003-)



R. Kato group (2008-)

Evidence

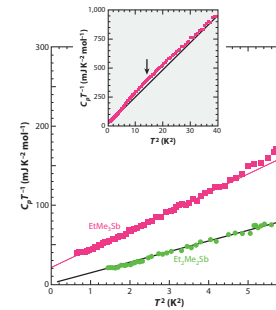
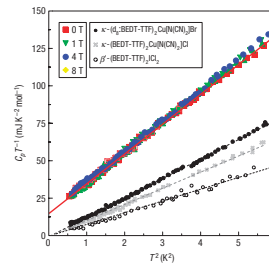
- NMR



no magnetic order

Y. Shimizu *et al*, 2003 T. Itou *et al*, 2008,2010

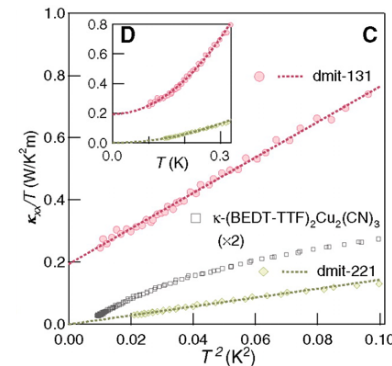
- Specific heat



Sommerfeld law

S. Yamashita *et al*, 2008

- Thermal conductivity

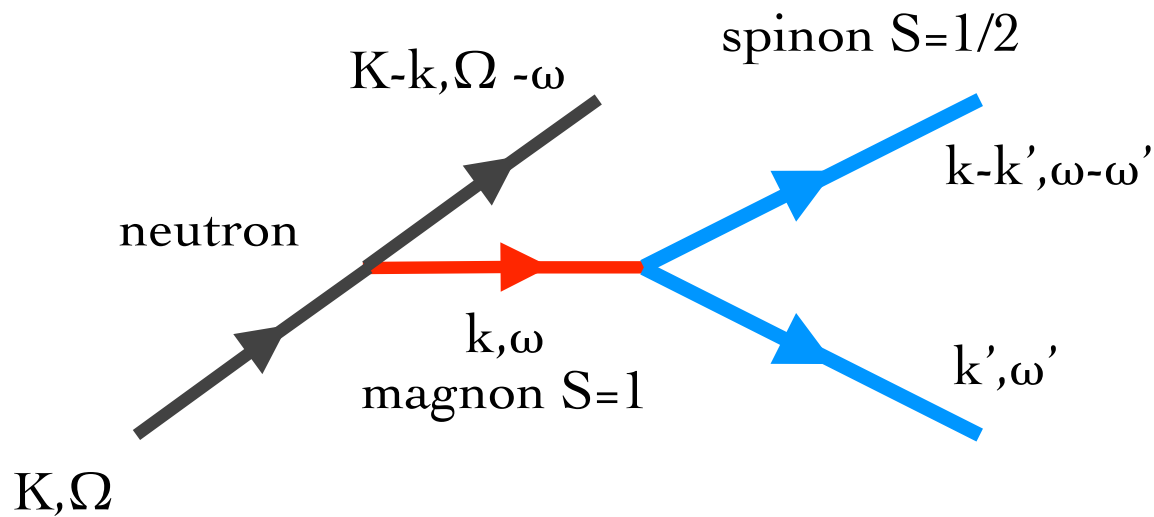


itinerant fermions?

M. Yamashita *et al*, 2010

Spectra

Common expectation in a QSL:
broad continuum scattering

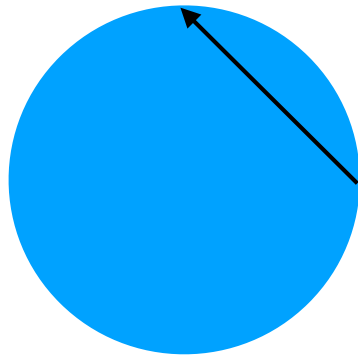


broad peak with
 $\omega = \varepsilon(k') + \varepsilon(k-k')$

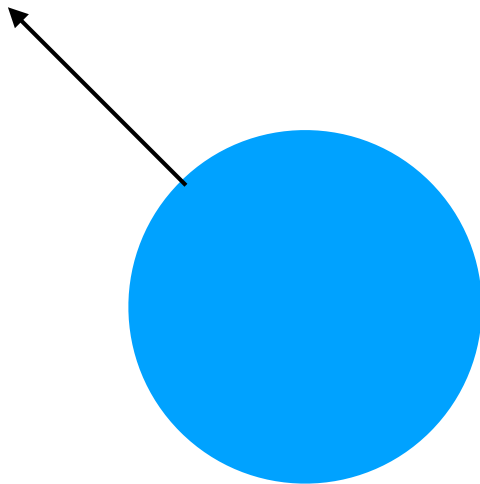
Not very inspiring?

particle-hole continuum

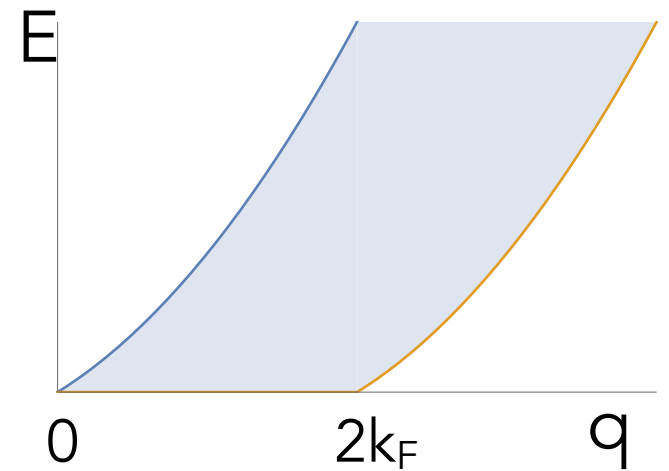
Free fermions



lowest energy for $k < k_F$



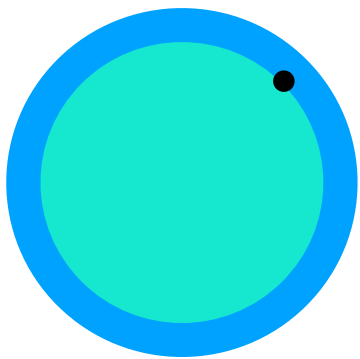
maximum energy



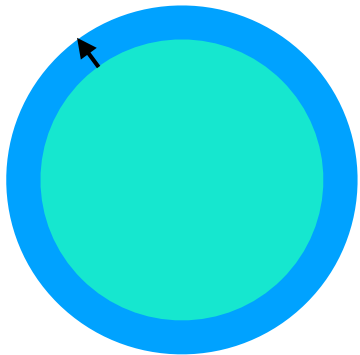
particle-hole continuum

With Zeeman field

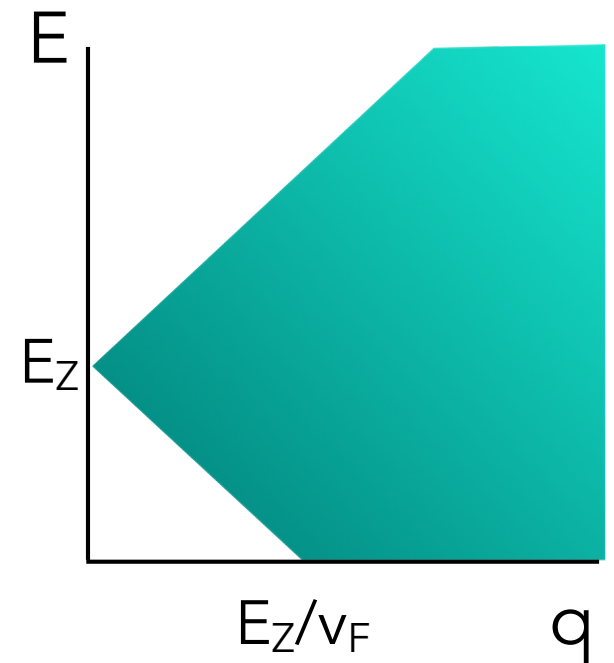
$$\chi_{\pm}(\mathbf{q}, \omega) = i \int_0^{\infty} dt \langle [S_{\mathbf{q}}^{\dagger}(t), S_{-\mathbf{q}}^{-}(0)] \rangle e^{i\omega t}$$



$q=0$ costs Zeeman energy



zero energy when $v_F q$
= Zeeman



Inorganic analogs?

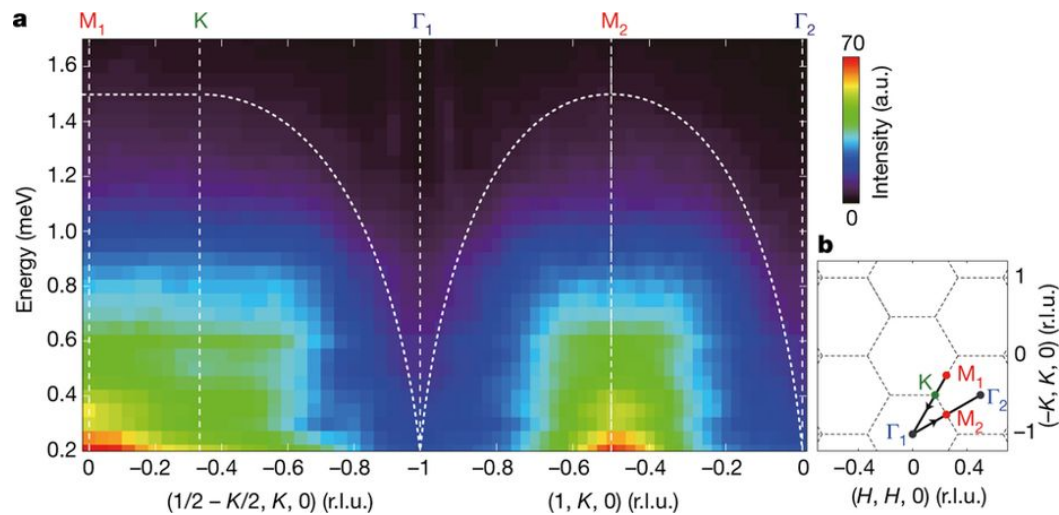
YbMgGaO₄

Letter | Published: 05 December 2016

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

Yao Shen, Yao-Dong Li, Hongliang Wo, Yuesheng Li, Shoudong Shen, Bingying Pan, Qisi Wang, H. C. Walker, P. Steffens, M. Boehm, Yiqing Hao, D. L. Quintero-Castro, L. W. Harriger, M. D. Frontzek, Lijie Hao, Siqin Meng, Qingming Zhang, Gang Chen & Jun Zhao

Nature **540**, 559–562 (22 December 2016) | [Download Citation](#)

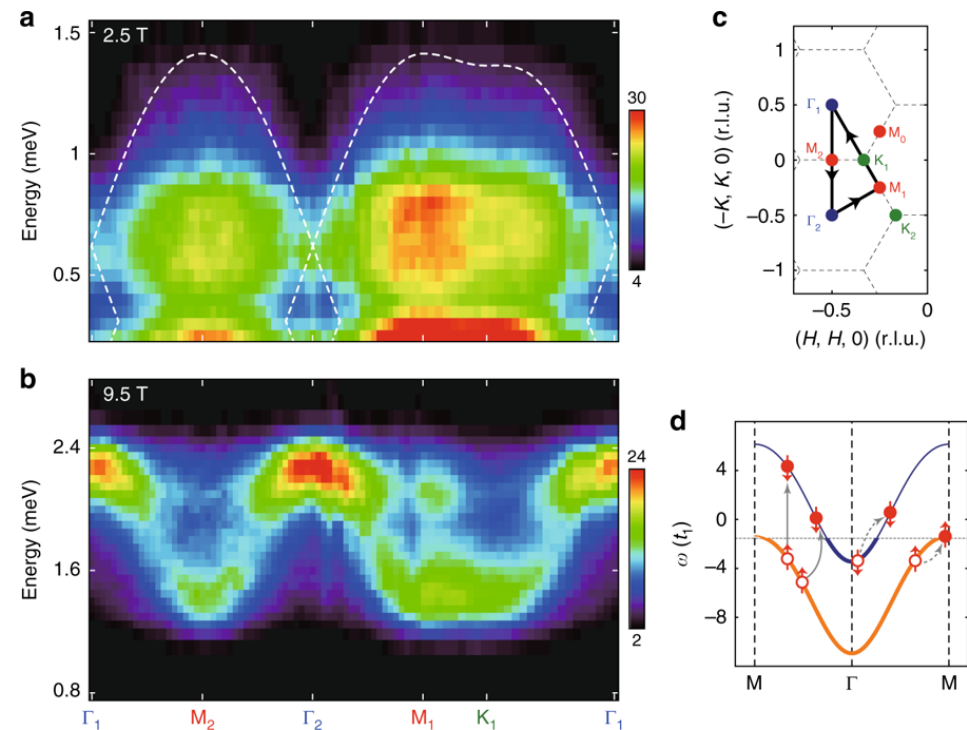


Article | OPEN | Published: 08 October 2018

Fractionalized excitations in the partially magnetized spin liquid candidate YbMgGaO₄

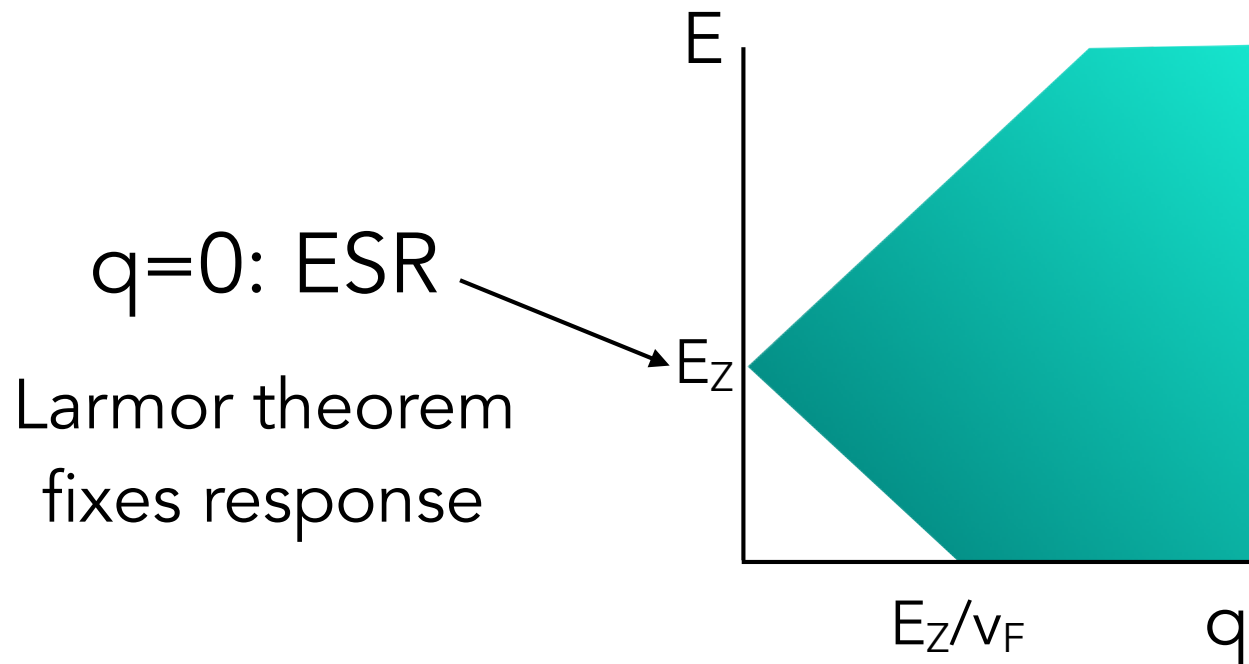
Yao Shen, Yao-Dong Li, H. C. Walker, P. Steffens, M. Boehm, Xiaowen Zhang, Shoudong Shen, Hongliang Wo, Gang Chen & Jun Zhao

Nature Communications **9**, Article number: 4138 (2018) | [Download Citation](#)



particle-hole continuum

With Zeeman field



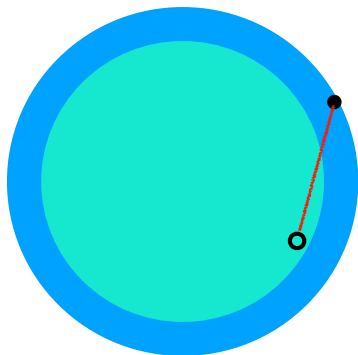
Effects of interactions?

Interactions

- Longitudinal

$$a_0 \psi^\dagger \psi$$

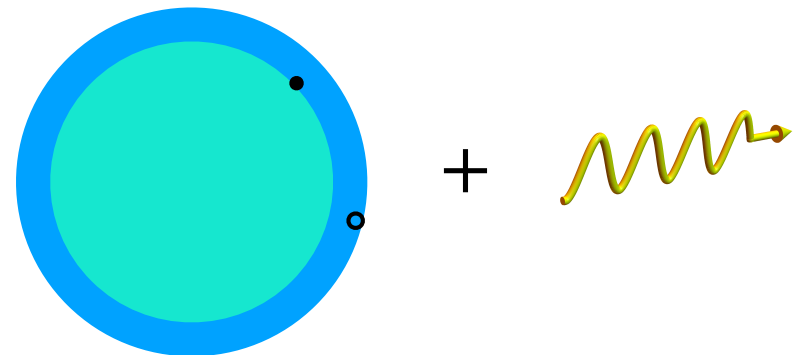
screened Coulomb
interaction



- Transverse

$$i\mathbf{A} \cdot (\psi^\dagger \nabla \psi - \nabla \psi^\dagger \psi)$$

coupling to dynamical
photons



Interactions

• Longitudinal $a_0 \psi^\dagger \psi$  $u \psi_\uparrow^\dagger \psi_\uparrow \psi_\downarrow^\dagger \psi_\downarrow$

$$= -um \left(\psi_\uparrow^\dagger \psi_\uparrow - \psi_\downarrow^\dagger \psi_\downarrow \right) + u : \psi_\uparrow^\dagger \psi_\uparrow \psi_\downarrow^\dagger \psi_\downarrow :$$

self-energy

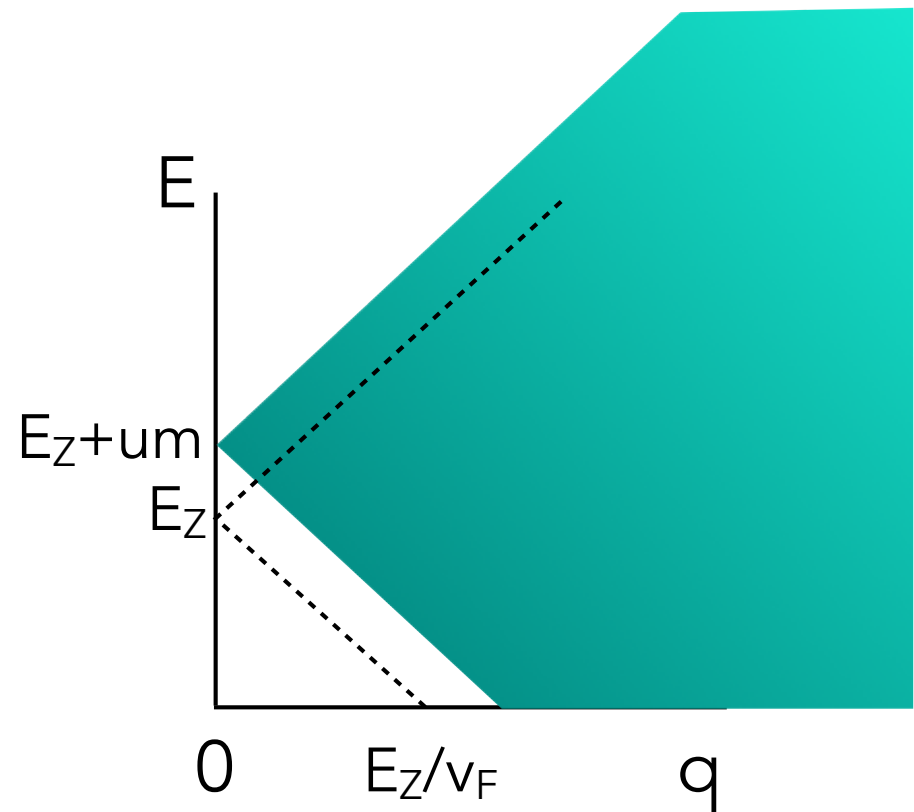
interaction

Self energy

- Longitudinal $a_0 \psi^\dagger \psi$ \rightarrow $u \psi_\uparrow^\dagger \psi_\uparrow \psi_\downarrow^\dagger \psi_\downarrow$

$$= -um \left(\psi_\uparrow^\dagger \psi_\uparrow - \psi_\downarrow^\dagger \psi_\downarrow \right)$$

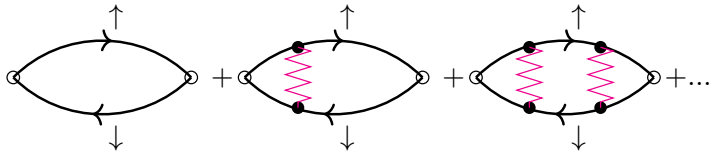
mean field shift



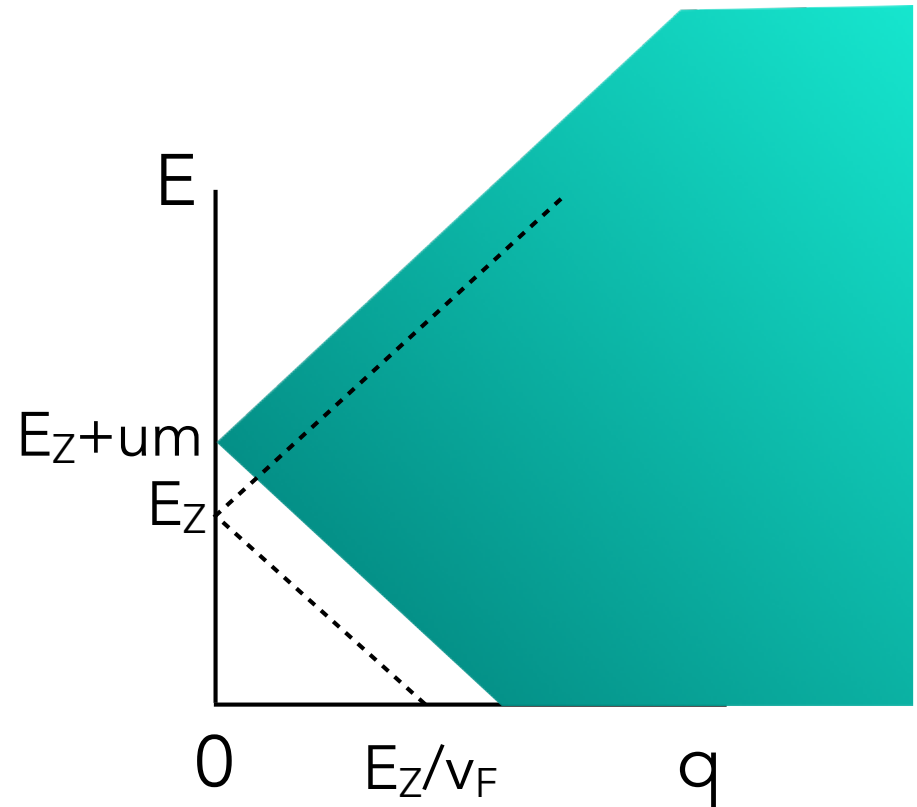
Interaction

Larmor theorem: $q=0$
excitation *must* be at E_Z

RPA



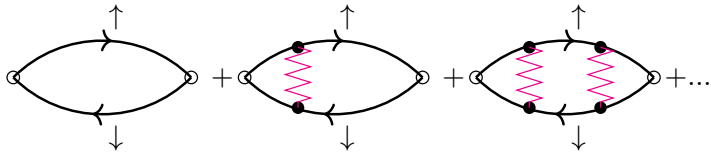
$$\chi(\mathbf{q}, i\omega_n) = \frac{\chi^0(\mathbf{q}, i\omega_n)}{1 + u\chi^0(\mathbf{q}, i\omega_n)}$$



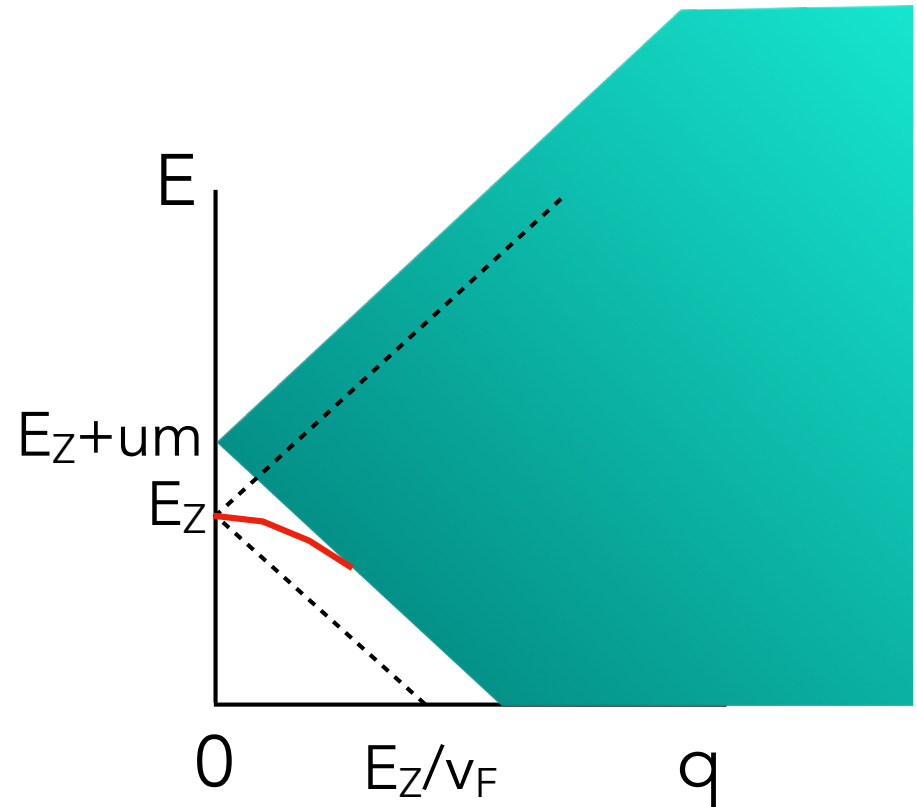
Silin spin wave

Larmor theorem: $q=0$
excitation *must* be at E_Z

RPA



$$\chi(\mathbf{q}, i\omega_n) = \frac{\chi^0(\mathbf{q}, i\omega_n)}{1 + u\chi^0(\mathbf{q}, i\omega_n)}$$

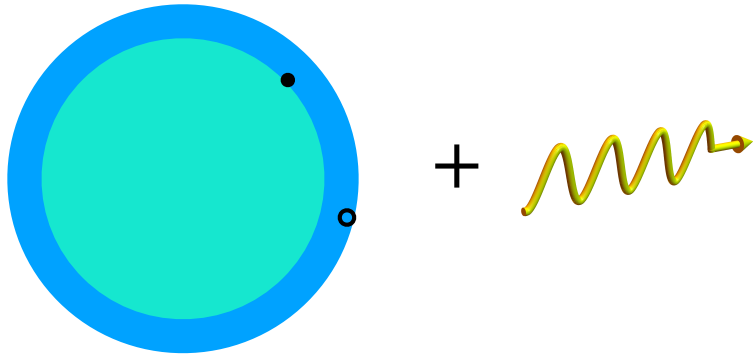


pole: collective mode

“Silin spin wave”

$$\omega = E_Z + um - \sqrt{u^2 m^2 + v_F^2 q^2}$$

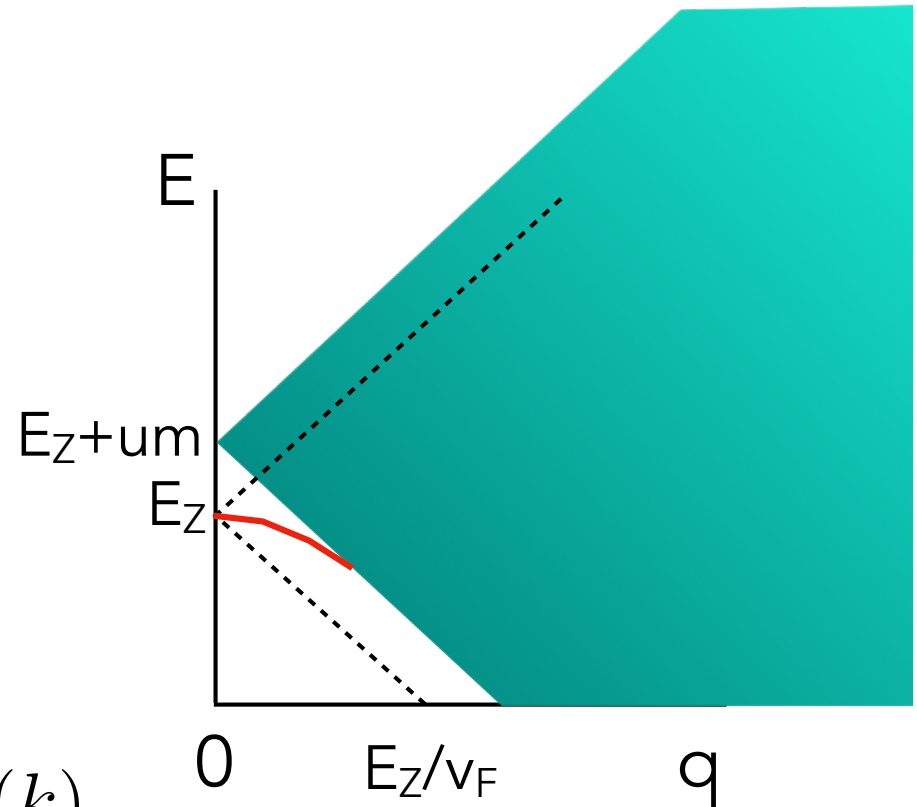
Transverse gauge coupling



Simple picture:

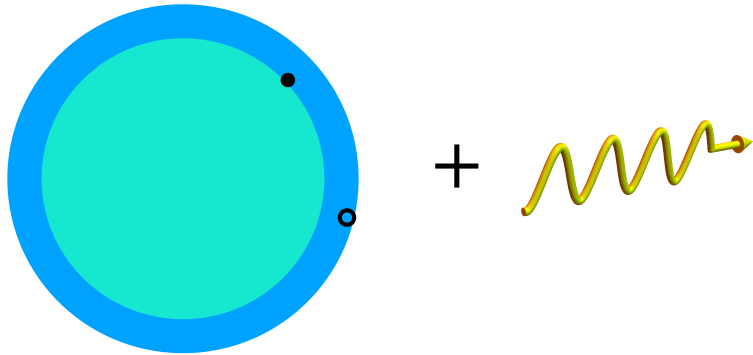
3-particle process:

$$E = E_{p/h}(q - k) + E_{\text{photon}}(k) \\ \sim ck^3$$

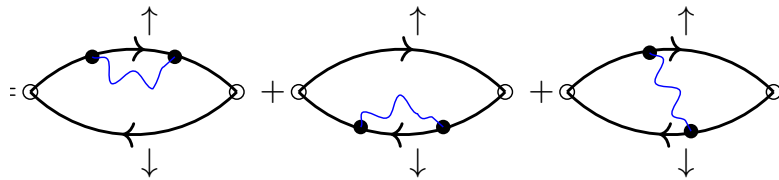


Does this smear out all the Fermi liquid structure?

Transverse gauge coupling

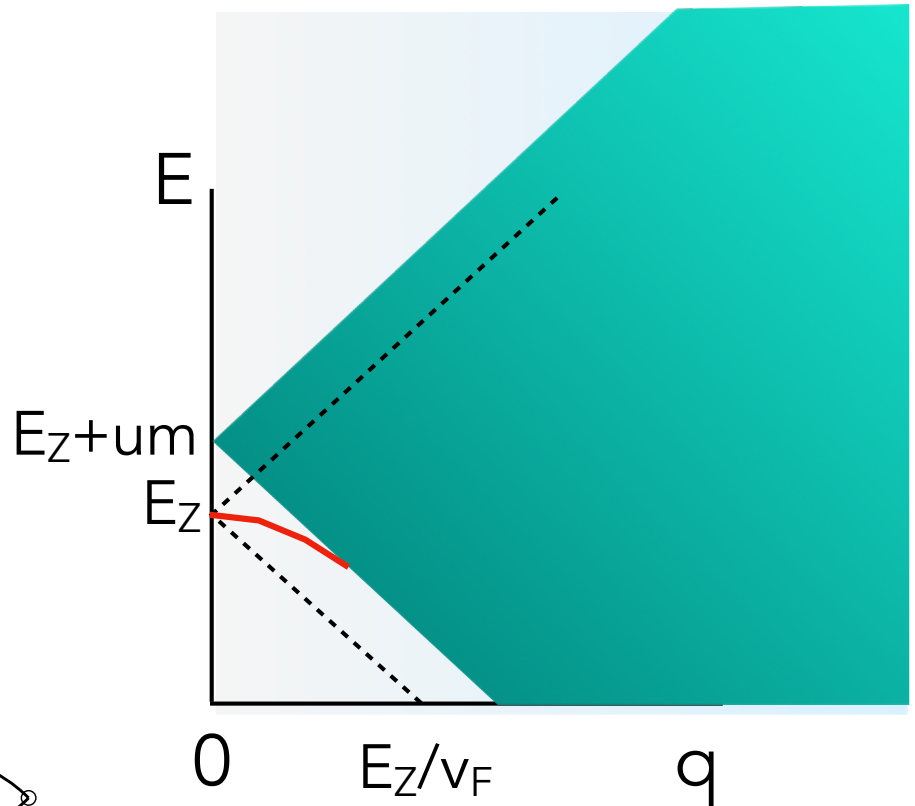


Actual calculation:



c.f. Y. B. Kim, A. Furusaki, X.-G. Wen, and P. A. Lee, *Phys Rev. B* **50**, 17917 (1994).

$$\text{Im}\chi_{\pm} \sim q^2 \omega^{7/3}$$

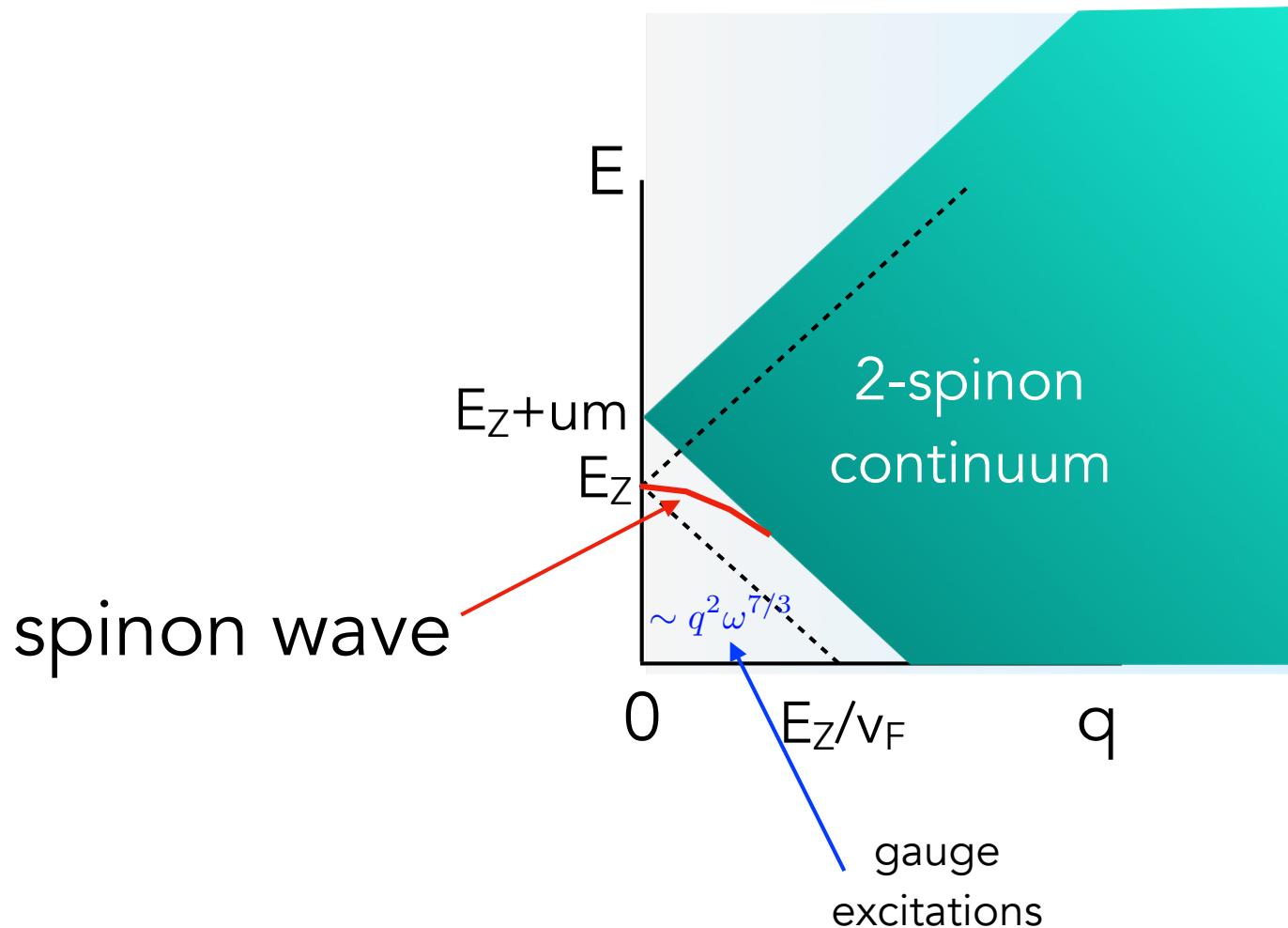


weight at all $q \neq 0$

but weak enough to
preserve structure

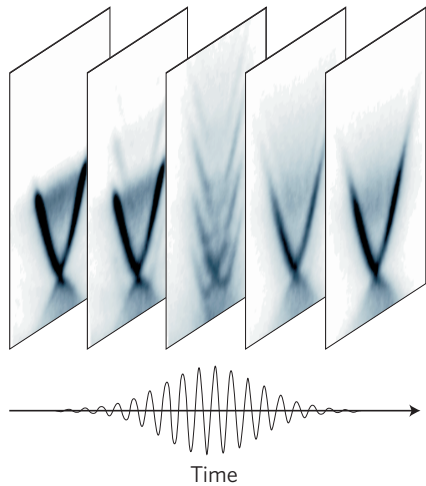
Summary

Distinct signatures of spinons,
interactions, and gauge fields



O.Starykh + LB,
arXiv:1904.02117

Ultra-fast Manipulation of Quantum Matter



Floquet-Bloch states in Bi_2Se_3

Wang *et al*, Science 2013

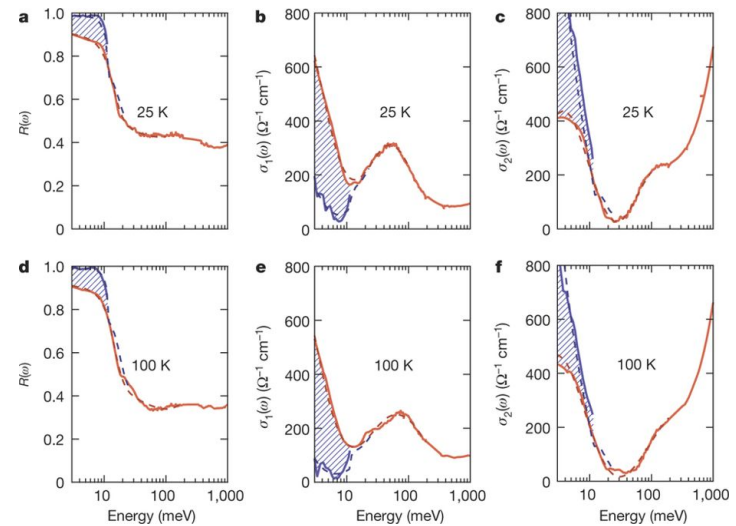


Photo-induced conductivity changes in K_3C_{60}

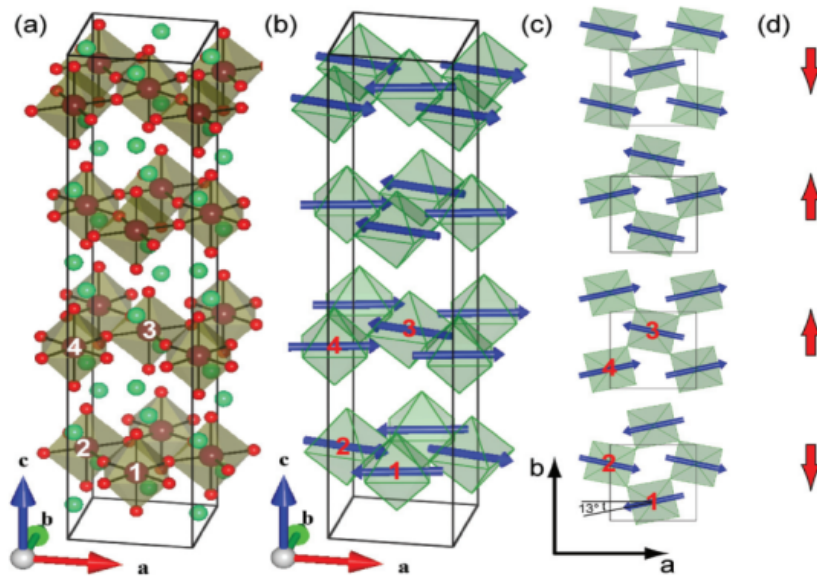
Mitrano *et al*, Nature 2016

Couple to electrons

Couple to phonons?

How about spins?

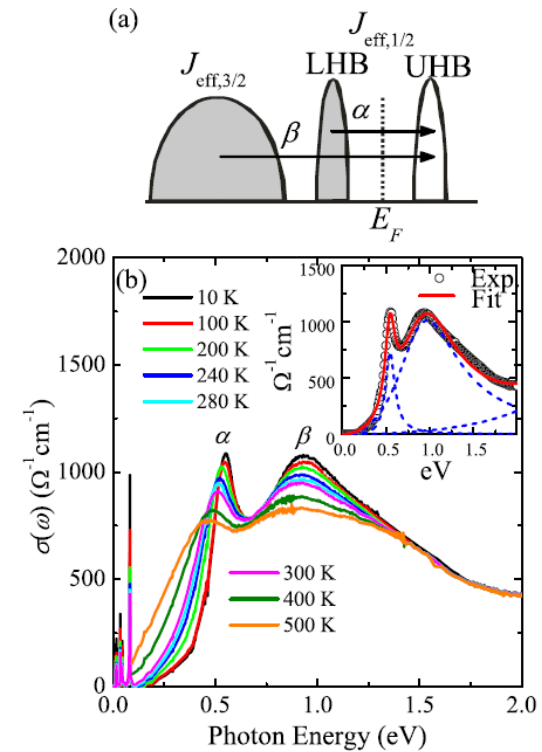
Sr₂IrO₄



Square lattice antiferromagnet

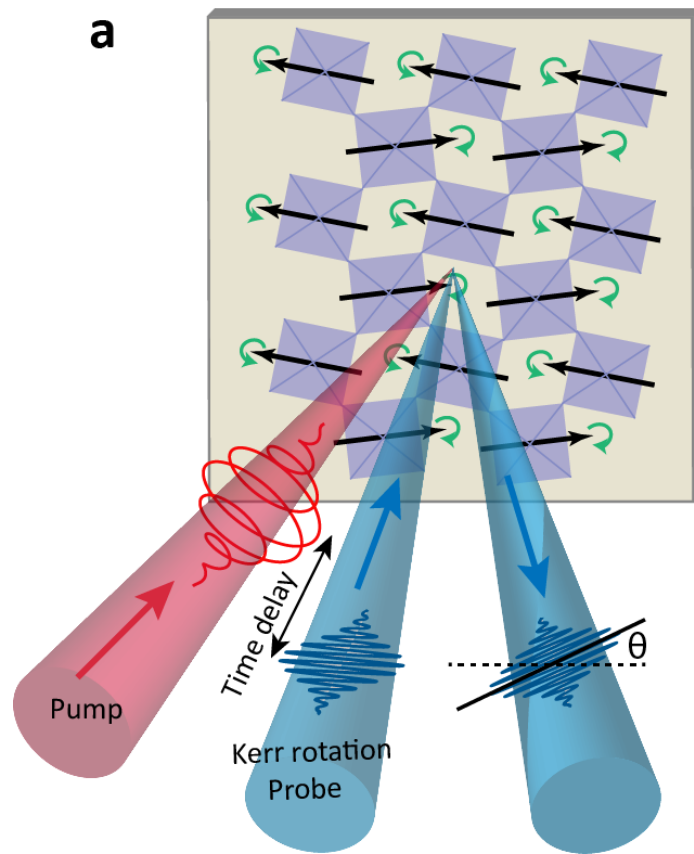
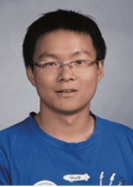
Strong SOC

Good venue for light-spin interactions

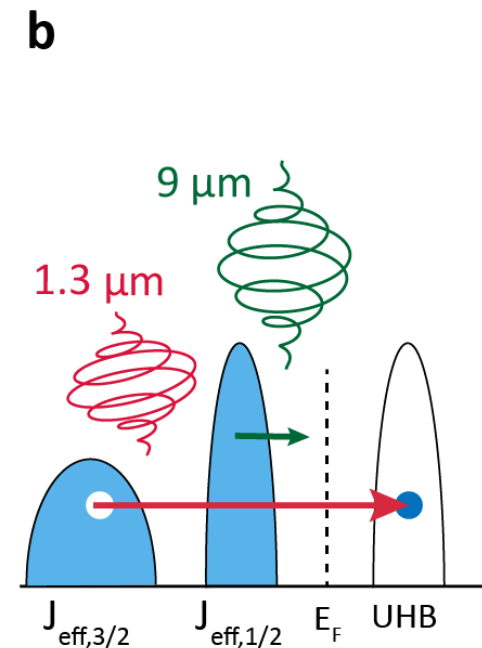


Ultrafast experiments

Gufeng Zhang *et al*, Averitt group, UCSD, in preparation



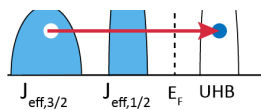
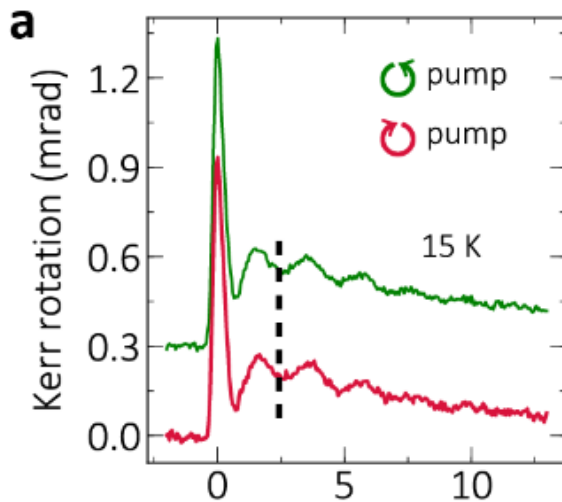
Kerr angle \sim magnetization



“Resonant” versus
“non-resonant”

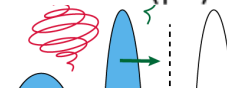
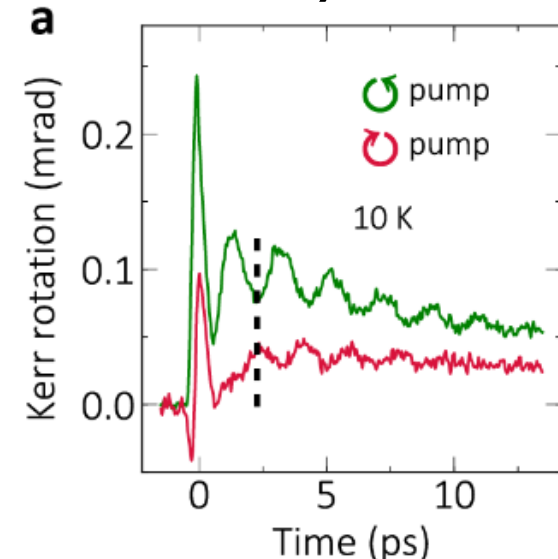
Phenomena

1.3 μm



$$\theta_K = .029 \text{ mrad mJ}^{-1} \text{ cm}^2$$

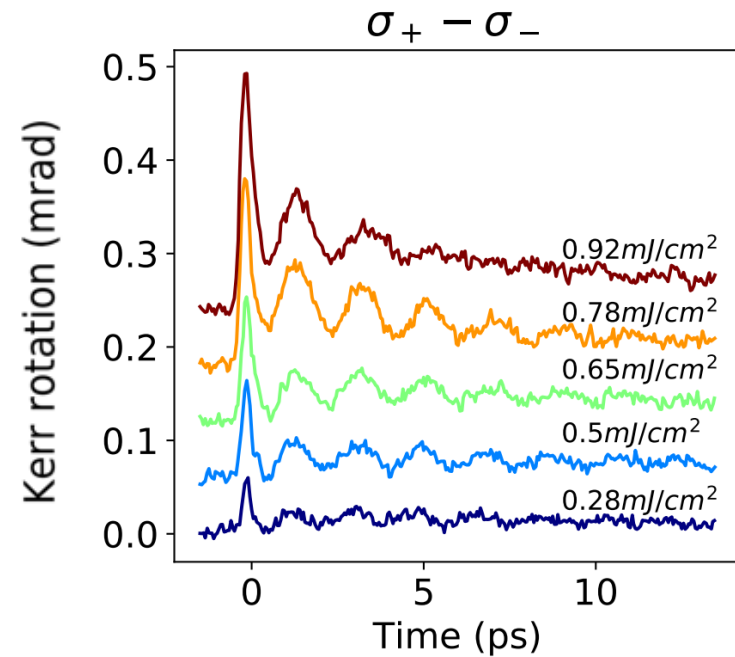
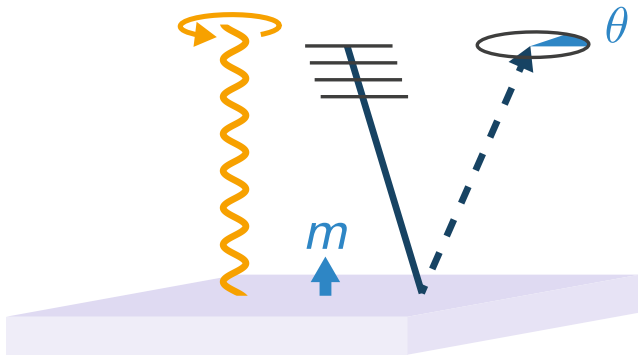
9 μm



$$\theta_K = .24 \text{ mrad mJ}^{-1} \text{ cm}^2$$

- Oscillation frequency $0.57\text{THz} = 2\text{meV}$ independent of details of pump: intrinsic magnon energy
- Pumping much more efficient for 9micron light.

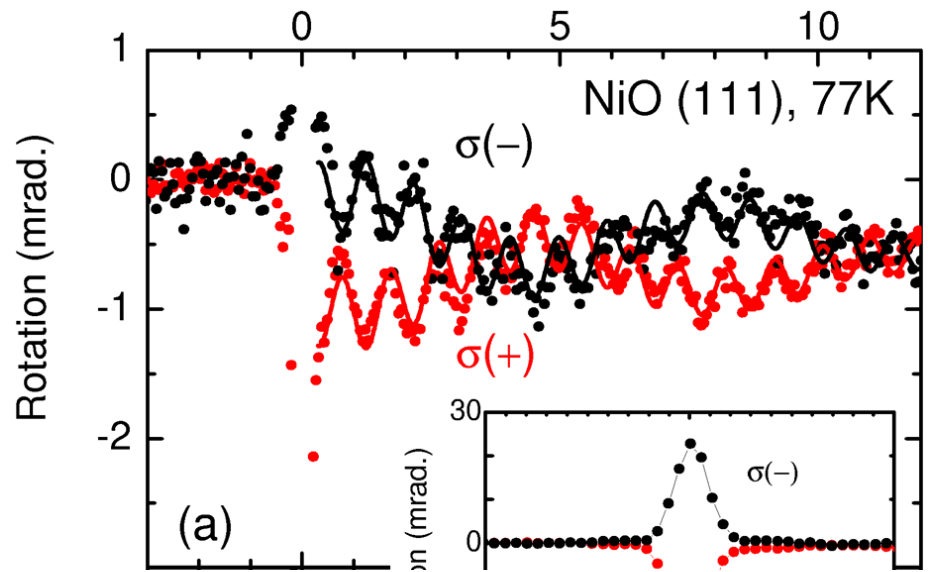
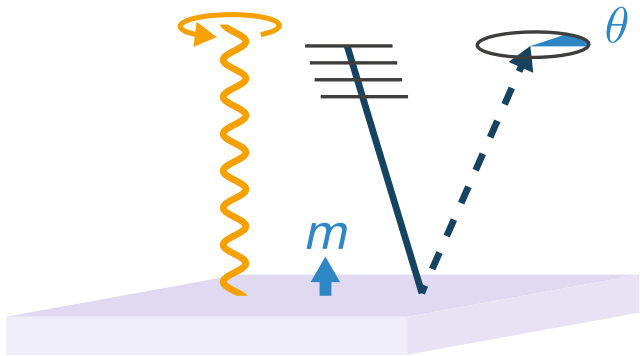
Phenomena



Questions

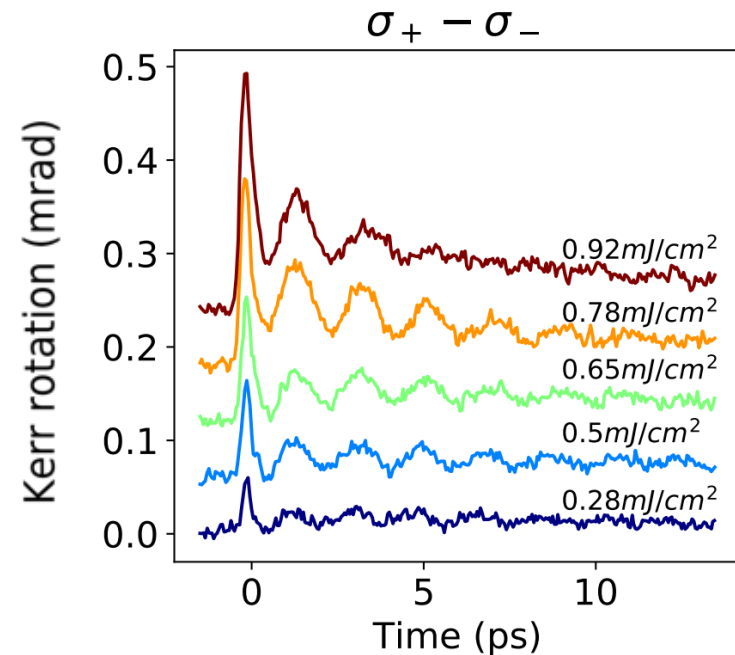
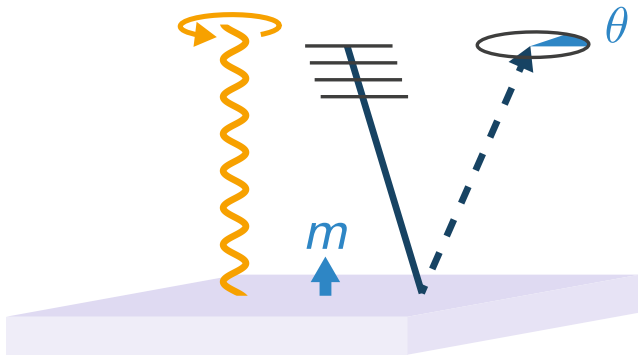
- What is the magnon oscillation and why is it visible?
- How is the magnon excited by sub-gap light?

Phenomena



c.f. T. Satoh *et al*, 2010 - NiO

Phenomena

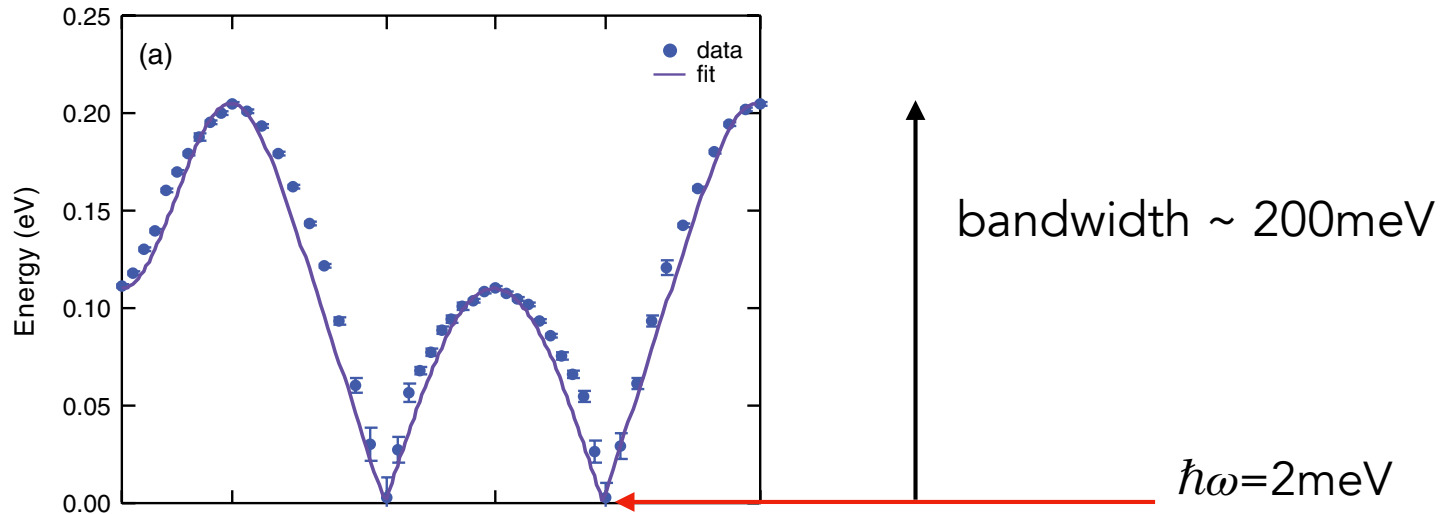


Questions

- What is the magnon oscillation and why is it visible?
- How is the magnon excited by sub-gap light?

Magnons

- Gross features: square lattice Heisenberg antiferromagnet



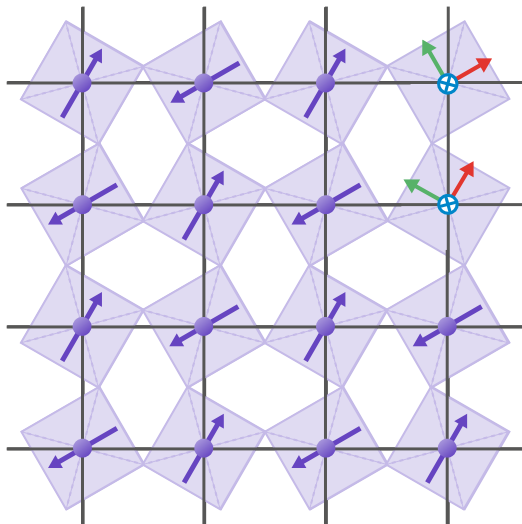
RIXS: BJ Kim *et al*, PRL 2012

Magnon is a very low energy feature

Anisotropy

Unit cell doubled by octahedral rotation

$$\mathcal{H}_{\text{eq}} = \sum_{i \in A} \sum_{\mu = \pm x, \pm y} \left[J_{xy} \left(S_i^x S_{i+\mu}^x + S_i^y S_{i+\mu}^y \right) + J_z S_i^z S_{i+\mu}^z + D \hat{z} \cdot \vec{S}_i \times \vec{S}_{i+\mu} \right] \quad \text{XXZ+DM}$$



DM is removed in local frame

G. Jackeli+G.Khaliullin, 2009

$$\rightarrow \sum_{i \in A} \sum_{\mu = \pm x, \pm y} J \left[S_i^x S_{i+\mu}^x + S_i^y S_{i+\mu}^y + (1 - \delta) S_i^z S_{i+\mu}^z \right]$$

Fits to RIXS give $J \sim 60 \text{ meV}$ and $\delta \sim .05$

Small easy-plane anisotropy

Spin wave theory

- Holstein-Primakoff

$$S^+ = S^z - iS^y = \sqrt{2S - a^\dagger a} a$$

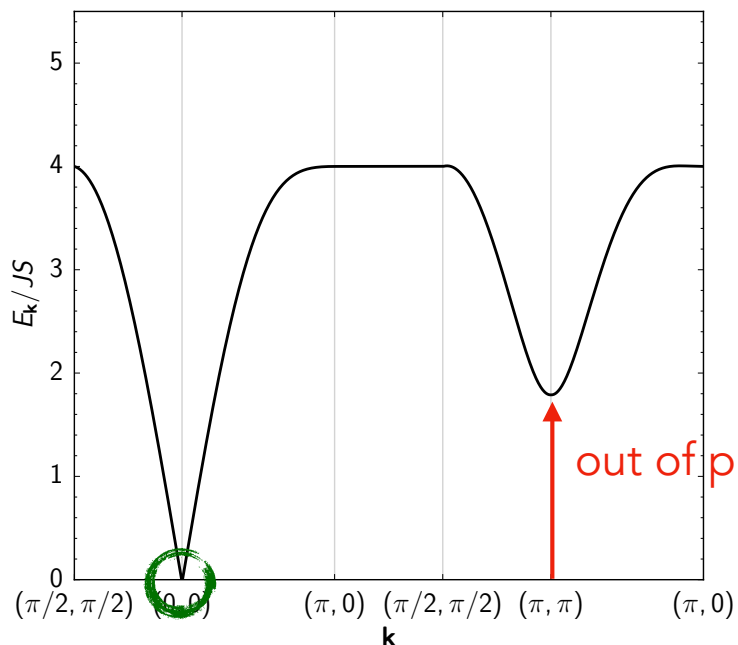
$$S^- = (S^+)^\dagger \quad S^x = S - a^\dagger a$$

$$\mathcal{H}_{\text{eq}} = \mathcal{H}_{\text{eq}}^{(0)} + \mathcal{H}_{\text{eq}}^{(2)} + \mathcal{H}_{\text{eq}}^{(4)} + \mathcal{O}(1/S)$$

E_{cl}

E_{sw}

magnon interactions



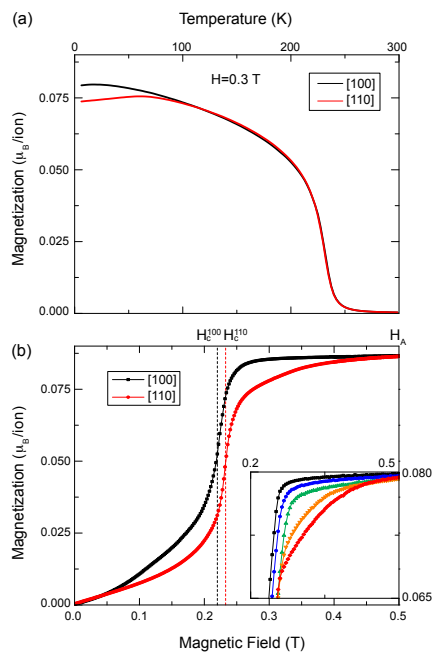
$$\mathcal{H}_{\text{eq}}^{(2)} = \sum_{\mathbf{k}} E_{\mathbf{k}} \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}}$$

out of plane mode. Gap ~ 30meV! Too big to be the oscillating magnon

gapless Goldstone mode of in-plane rotations

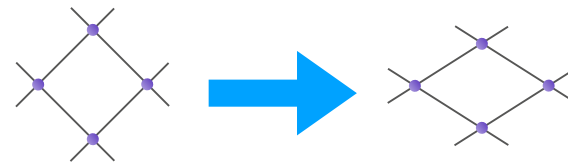
In-plane anisotropy

- Even weaker effect gives tiny gap to in-plane magnon



Porras et al, 2019

argue due to lattice distortion induced by spin order



$$\tilde{\mathcal{H}}_{JT} = \Gamma \sum_{\langle ij \rangle} S_i^x S_j^x - S_i^y S_j^y$$

$$\Gamma \sim 6 \mu\text{eV} (!)$$

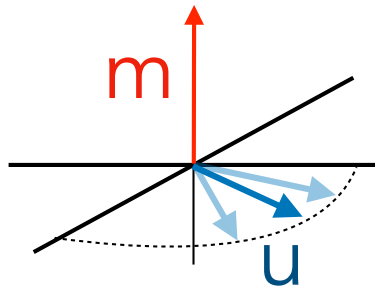
$$\omega_0 \simeq 8S\sqrt{\Gamma J} \sim 2\text{meV}$$

Oscillation matches in-plane magnon.

Magnetization oscillation

- Q: Why does Kerr angle oscillate if magnon is in-plane??

In-plane spin rotation \longleftrightarrow out of plane torque



$$\vec{S}_i = \begin{pmatrix} (-1)^i S \\ (-1)^i u(x_i) \\ m(x_i) \end{pmatrix}$$

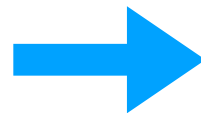
relation to magnons

$$u = \sqrt{S/2} i(a - a^\dagger)$$

$$m = \sqrt{S/2} (a + a^\dagger)$$

$$\frac{dS_i^z}{dt} = h_i^x S_i^y - h_y S_i^z$$

uniform
staggered
staggered



$$\partial_t u = \chi^{-1} m$$

$$\partial_t m = -\kappa u$$

slow modes - hydrodynamic equations valid even when magnons scatter

uniform magnetization oscillates out of phase with in-plane angle u

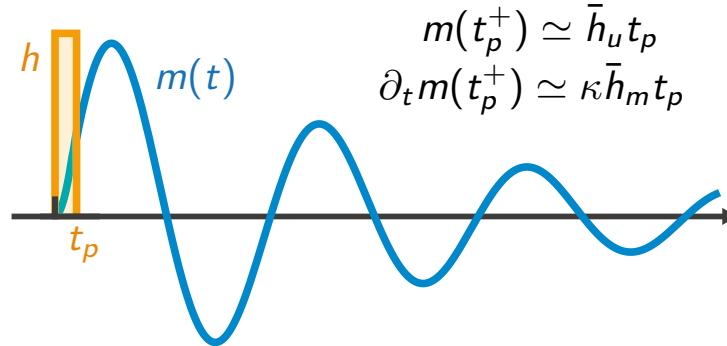
Pumping

- Strategy: light creates *source terms* for EOM

$$\begin{aligned}\partial_t u &= \chi^{-1} m + h_m(t) \\ \partial_t m &= -\kappa u + h_u(t)\end{aligned}$$

effective fields drive during pump pulse

decay/relaxation negligible
during pump



effective initial conditions

Theory: include light-matter interaction and integrate out higher energy modes to obtain h_m, h_u

Coupling to E-field

- Assumption: E-field of light dominates. Strong time-reversal-symmetry constraints

$$\mathcal{T} : \vec{E} \rightarrow \vec{E}, \vec{S} \rightarrow -\vec{S} \quad \longrightarrow \quad \mathcal{H}_E \sim g E^\alpha S_i^\beta S_j^\gamma$$

- General symmetry allowed couplings

$$\begin{aligned} \mathcal{H}_E = \sum_i & \left[g_1 \epsilon_i \left[E_x (S_i^y S_{i+x}^x + S_i^x S_{i+x}^y) - (x \leftrightarrow y) \right] \right. \\ & + g_2 \epsilon_i \left(E_y S_i^x S_{i+x}^x - E_x S_i^y S_{i+y}^y \right) \\ & + g_3 \epsilon_i \left(E_y S_i^y S_{i+x}^y - E_x S_i^x S_{i+y}^x \right) \\ & + g_4 \epsilon_i S_i^z \left(E_y S_{i+x}^z - E_x S_{i+y}^z \right) \\ & \left. + g_5 \left(E_y \hat{z} \cdot S_i \times S_{i+x} - E_x \hat{z} \cdot S_i \times S_{i+y} \right) \right] \end{aligned}$$

staggering due to
octahedral rotations
important!

microscopic calculations confirm these terms

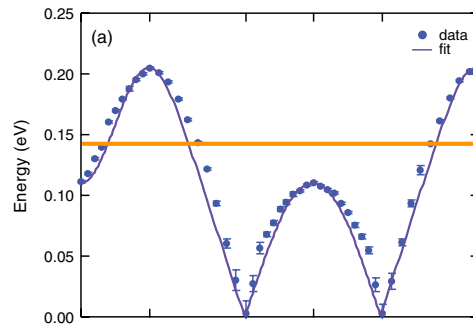
Bosons

- Holstein-Primakoff:

$$\psi_{\mathbf{k}} = (a_{\mathbf{k}}, a_{-\mathbf{k}}^\dagger)^T$$

$$\mathcal{H}_E = E_\mu(t) [\Phi_A^{1,\mu} \psi_A + \Phi_{A,B}^{2,\mu} \psi_A \psi_B + \Phi_{A,B,C}^{3,\mu} \psi_A \psi_B \psi_C + \mathcal{O}(1/S^0)]$$

$\sim \text{Re} (\mathcal{E}_\mu e^{i\omega t})$
 Floquet drive



- Cannot excite single magnon due to momentum conservation
- Two magnon process is resonant: generates pairs with \mathbf{k} and $-\mathbf{k}$
 ?? How is $\mathbf{k}=\mathbf{0}$ magnetization created?

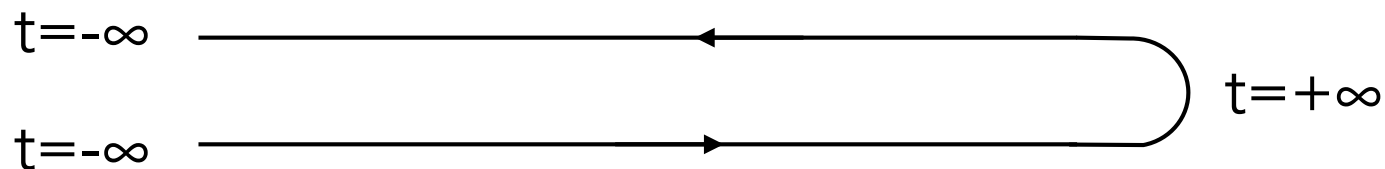
Formalism

- Non-equilibrium Keldysh method: evolve both bra and ket

$$|\Psi(t)\rangle = U(t, -\infty)|\Psi(-\infty)\rangle$$

$$\langle\mathcal{O}(t)\rangle = \langle\Psi(\infty)|U(-\infty, t)\mathcal{O}U(t, -\infty)|\Psi(-\infty)\rangle$$

- Keldysh contour



$$\mathcal{Z} = \int \mathcal{D}[a_+, a_-] \exp(i\mathcal{S})$$

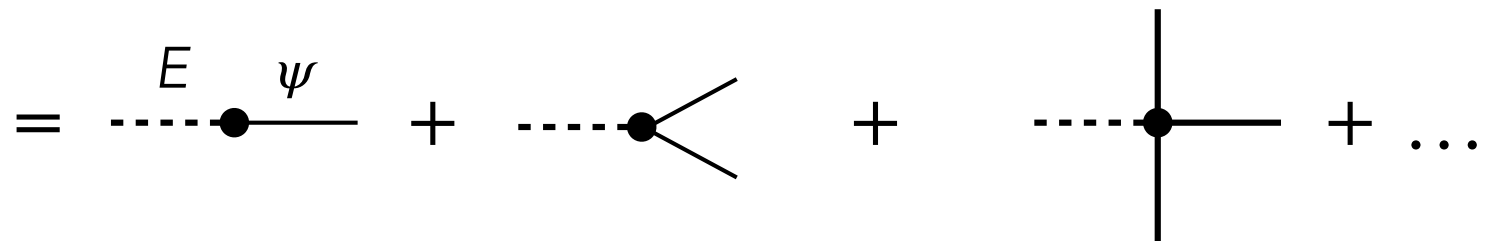
$$\mathcal{S} = \sum_{s=\pm} s \int dt \left\{ \sum_i \bar{a}_{s,i} i \partial_t a_{s,i} - \mathcal{H}[\{\bar{a}_{s,i}, a_{s,i}\}] \right\}$$

path integral over
doubled fields

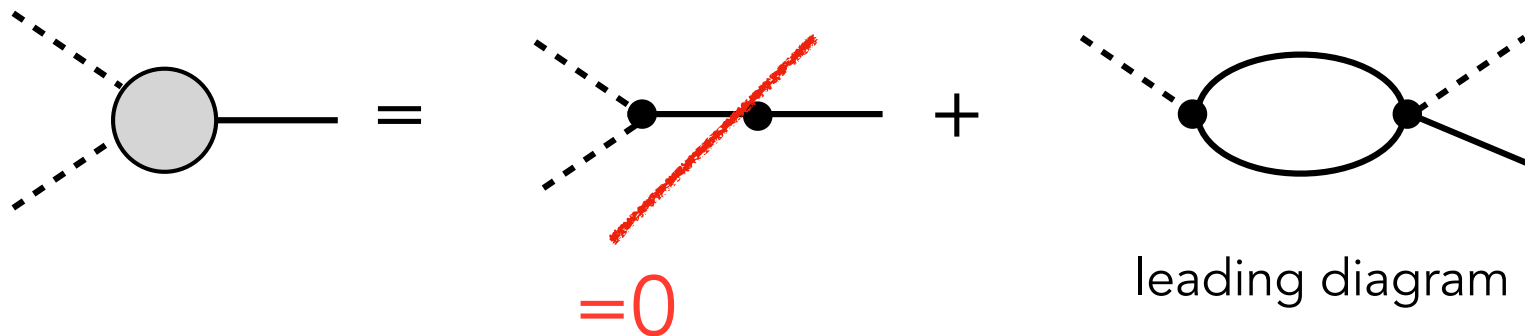
Formalism

- Non-equilibrium Keldysh path integral

$$\mathcal{H}_E = E_\mu(t) [\Phi_A^{1,\mu} \psi_A + \Phi_{A,B}^{2,\mu} \psi_A \psi_B + \Phi_{A,B,C}^{3,\mu} \psi_A \psi_B \psi_C + \mathcal{O}(1/S^0)]$$



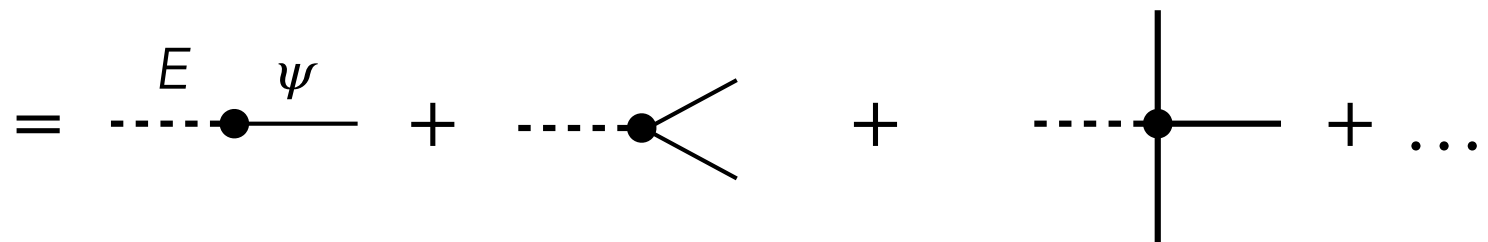
- Now integrate out "fast" fields: internal lines



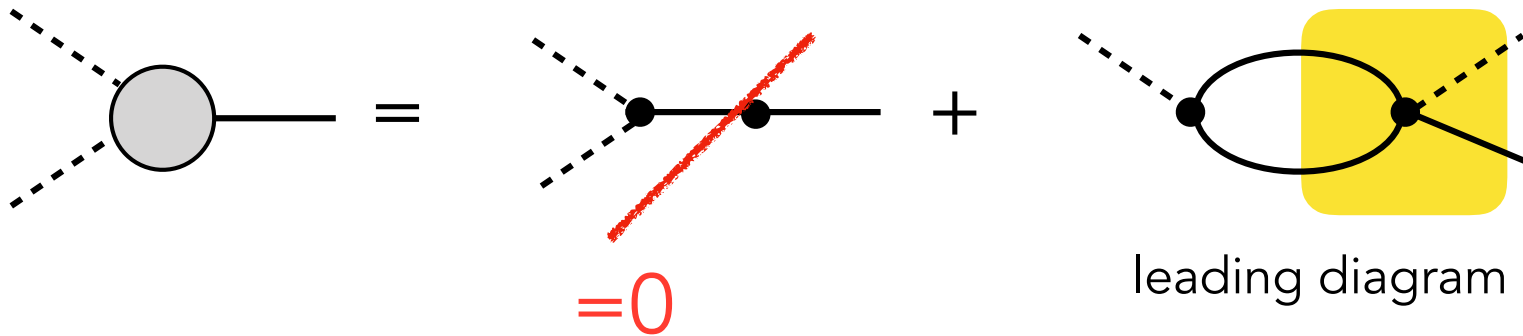
Formalism

- Non-equilibrium Keldysh path integral

$$\mathcal{H}_E = E_\mu(t) [\Phi_A^{1,\mu} \psi_A + \Phi_{A,B}^{2,\mu} \psi_A \psi_B + \Phi_{A,B,C}^{3,\mu} \psi_A \psi_B \psi_C + \mathcal{O}(1/S^0)]$$



- Now integrate out "fast" fields: internal lines

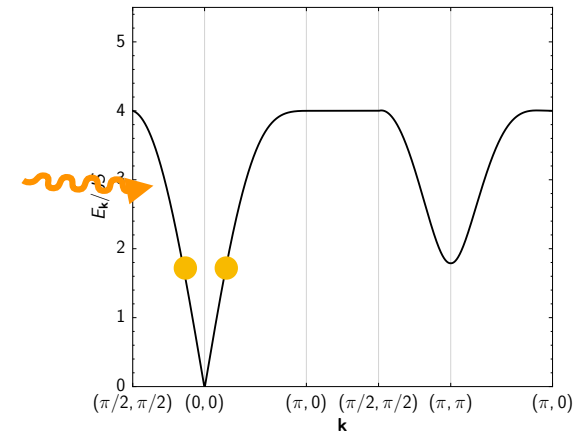
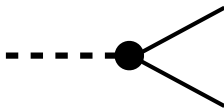


involves boson interactions!

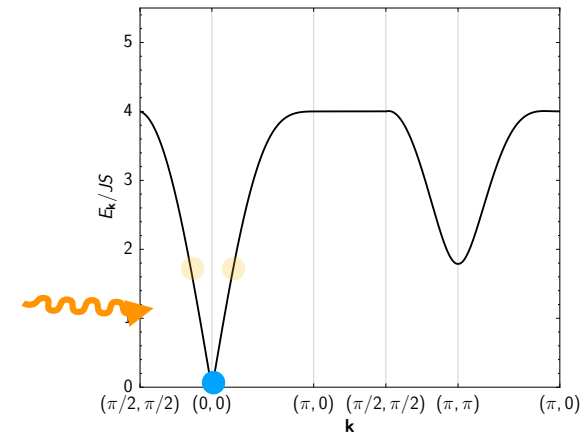
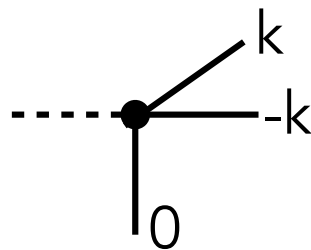
Physical picture

- Step 1:

photon generates coherent pairs



- Step 2: Interaction "proximitizes" low energy condensate = magnons



Results

- Effective fields given by loop integrals

$$h \sim \int d^2\mathbf{k} \mathcal{E}_\mu \bar{\mathcal{E}}_\nu \sum_{\beta, \beta' = \pm 1} \frac{\phi^{2, \mu} \phi^{3, \nu}}{\Omega + \beta E_{\mathbf{k}} + \beta' E_{\mathbf{Q} - \mathbf{k}} + i\eta}$$

separates into:

dissipative

$$\sim \delta(\Omega - E_{\mathbf{k}} - E_{\mathbf{Q} - \mathbf{k}})$$

virtual

$$\sim \frac{P}{\delta - E_{\mathbf{k}} - E_{\mathbf{Q} - \mathbf{k}}}$$

Results

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- Structure of contributions to effective fields

$\sin 2\phi \times \begin{cases} \mathcal{E}_x \bar{\mathcal{E}}_y + \mathcal{E}_y \bar{\mathcal{E}}_x \\ \mathcal{E}_x \bar{\mathcal{E}}_x - \mathcal{E}_y \bar{\mathcal{E}}_y \end{cases}$	total intensity	chiral intensity
	$\sin 4\phi \times (\mathcal{E}_x \bar{\mathcal{E}}_x + \mathcal{E}_y \bar{\mathcal{E}}_y)$	$i (\mathcal{E}_x \bar{\mathcal{E}}_y - \mathcal{E}_y \bar{\mathcal{E}}_x)$
traceless symmetric	identity	antisymmetric

Results

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	$\sin 4\phi \times (\mathcal{E}_x \bar{\mathcal{E}}_x + \mathcal{E}_y \bar{\mathcal{E}}_y)$	

linear polarization

Results

- Effective fields given by loop integrals

$$h \sim \int d^2\mathbf{k} \mathcal{E}_\mu \bar{\mathcal{E}}_\nu \sum_{\beta, \beta' = \pm 1} \frac{\phi^{2, \mu} \phi^{3, \nu}}{\Omega + \beta E_{\mathbf{k}} + \beta' E_{\mathbf{Q}-\mathbf{k}} + i\eta}$$

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$$\sim \delta(\Omega - E_{\mathbf{k}} - E_{\mathbf{Q}-\mathbf{k}})$$

virtual

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- Structure of contributions to effective fields

$$\sin 2\phi \times \begin{cases} \mathcal{E}_x \bar{\mathcal{E}}_y + \mathcal{E}_y \bar{\mathcal{E}}_x \\ \mathcal{E}_x \bar{\mathcal{E}}_x - \mathcal{E}_y \bar{\mathcal{E}}_y \end{cases}$$

total intensity	chiral intensity
$\sin 4\phi \times (\mathcal{E}_x \bar{\mathcal{E}}_x + \mathcal{E}_y \bar{\mathcal{E}}_y)$	$i (\mathcal{E}_x \bar{\mathcal{E}}_y - \mathcal{E}_y \bar{\mathcal{E}}_x)$

circular polarization

Results

- Effective fields given by loop integrals

$$h \sim \int d^2\mathbf{k} \mathcal{E}_\mu \bar{\mathcal{E}}_\nu \sum_{\beta, \beta' = \pm 1} \frac{\phi^{2, \mu} \phi^{3, \nu}}{\Omega + \beta E_{\mathbf{k}} + \beta' E_{\mathbf{Q}-\mathbf{k}} + i\eta}$$

separates into:

dissipative

$$\sim \delta(\Omega - E_{\mathbf{k}} - E_{\mathbf{Q}-\mathbf{k}})$$

virtual

$$\sim \frac{P}{\delta - E_{\mathbf{k}} - E_{\mathbf{Q}-\mathbf{k}}}$$

- Structure of contributions to effective fields

$$\sin 2\phi \times \begin{cases} \mathcal{E}_x \bar{\mathcal{E}}_y + \mathcal{E}_y \bar{\mathcal{E}}_x \\ \mathcal{E}_x \bar{\mathcal{E}}_x - \mathcal{E}_y \bar{\mathcal{E}}_y \end{cases}$$

total intensity	chiral intensity
$\sin 4\phi \times (\mathcal{E}_x \bar{\mathcal{E}}_x + \mathcal{E}_y \bar{\mathcal{E}}_y)$	$i (\mathcal{E}_x \bar{\mathcal{E}}_y - \mathcal{E}_y \bar{\mathcal{E}}_x)$

=0 for Sr_2IrO_4
($\phi = \pi/4$)

circular polarization

“inverse Faraday effect”

Prior approach to Inverse Faraday effect

PHYSICAL REVIEW

VOLUME 143, NUMBER 2

MARCH 1966

Theoretical Discussion of the Inverse Faraday Effect, Raman Scattering, and Related Phenomena*

P. S. PERSHAN,[†] J. P. VAN DER ZIEL,[‡] AND L. D. MALMSTROM
Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts
 (Received 25 October 1965)

- Derived effective thermodynamic potential

$$\mathcal{F} \sim \chi^{ijk} M_i(0) E_j(\omega) E_k(-\omega) \quad \longrightarrow \quad h_{\text{eff}}^i = -\frac{\partial \mathcal{F}}{\partial M_i(0)} = \chi^{ijk} E_j(\omega) E_k(-\omega)$$

really correct for low frequency magnetization (not short pulse)

- Carried out for quantum mechanical few-level system

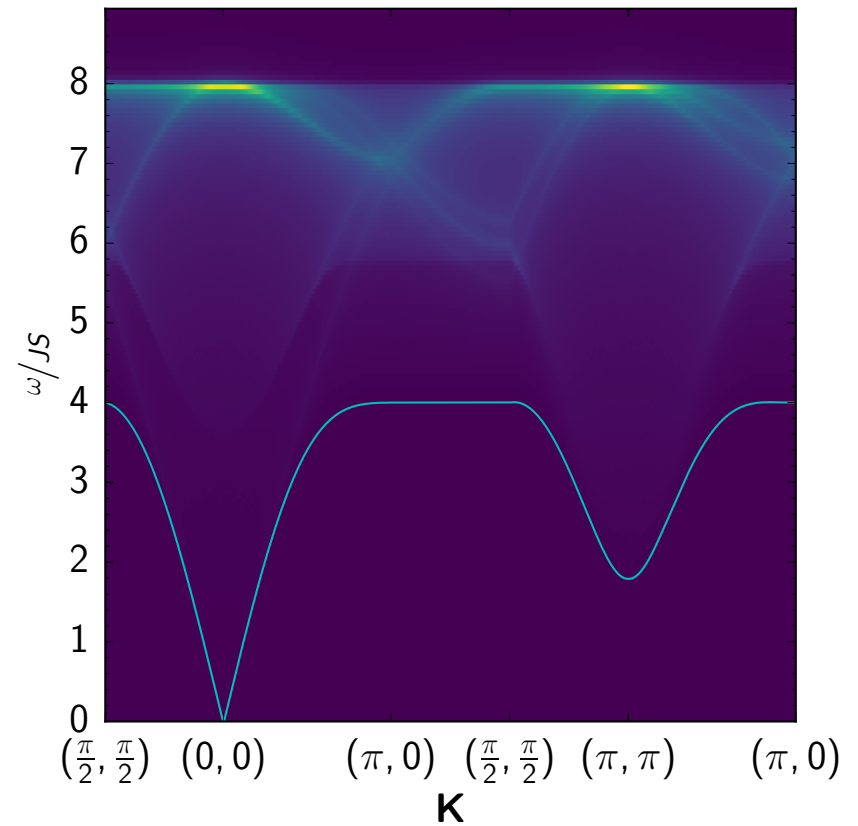
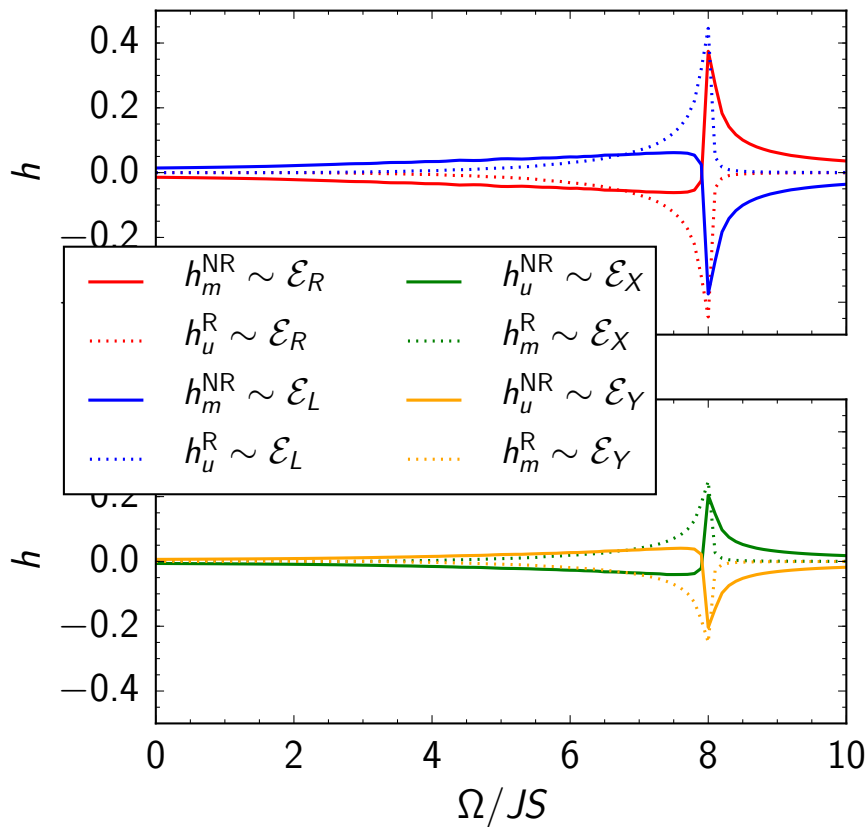
$$\mathcal{H}_{\text{eff}} = (\mathcal{E}_R \mathcal{E}_R^* - \mathcal{E}_L \mathcal{E}_L^*) J_z A + \{ (\mathcal{E}_R \mathcal{E}_R^* + \mathcal{E}_L \mathcal{E}_L^*) [J_z^2 - \frac{1}{3} J(J+1)] - \mathcal{E}_L \mathcal{E}_R^* J_-^2 - \mathcal{E}_L^* \mathcal{E}_R J_+^2 \} C,$$

Purely virtual excitations

- Our results treat general many body situation with fast dynamics and both real and virtual excitations

Results

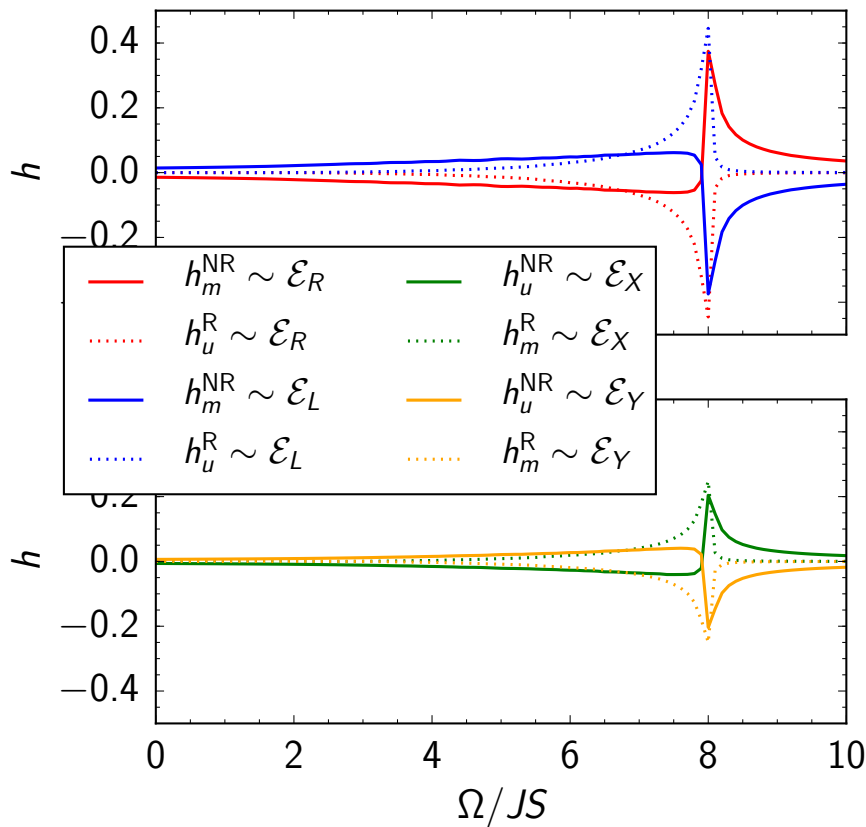
- Effective fields maximized when light is near peak of two-magnon DOS



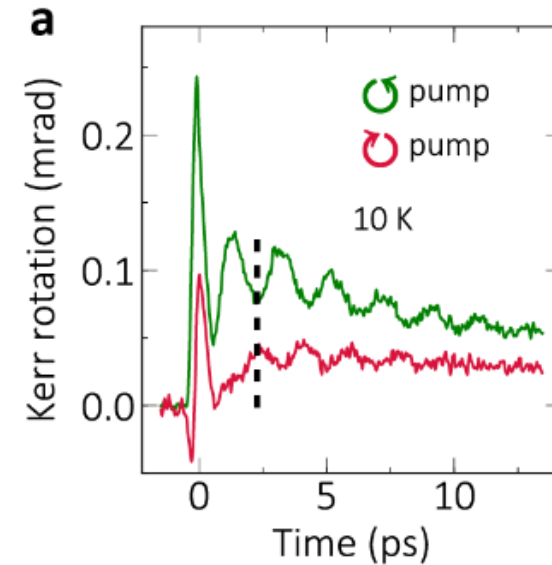
Significant enhancement still possible!

Results

- Effective fields maximized when light is near peak of two-magnon DOS



Gufeng Zhang *et al*



Dominant chiral intensity:
 magnetization of opposite sign
 for two circular polarizations

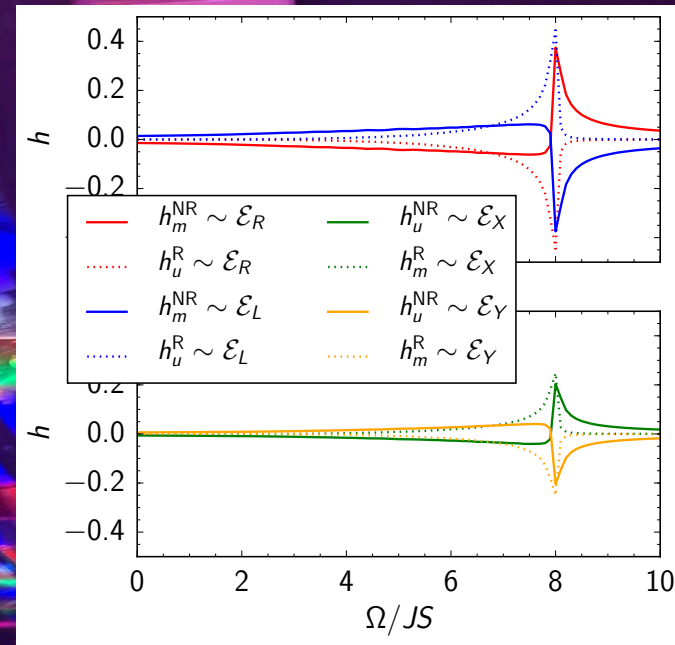
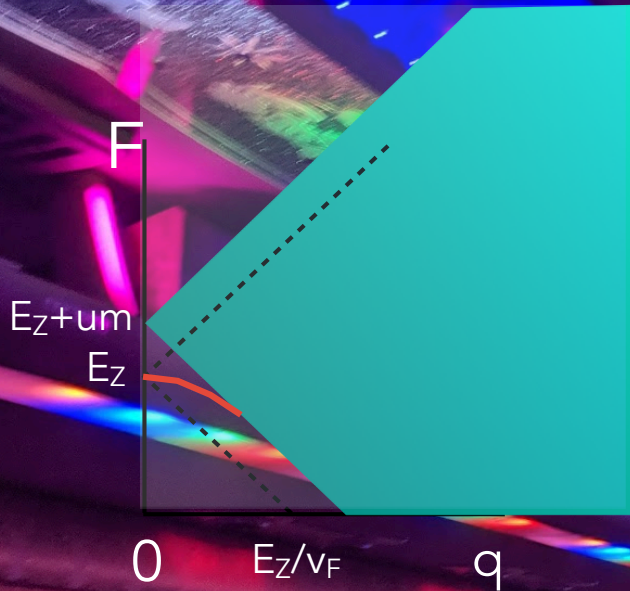
Future directions

- Experimental test of frequency dependence. Many other materials applications.
- Extension to $T > 0$
- Extension to non-collinear magnets: much larger anharmonic effects
- Topological effects: Pump/probe of Dirac/Weyl magnons, analogies to topological effects in SHG?
- Ultrafast dynamics of quantum spin liquids without magnons?

Recap

O.Starykh + LB, arXiv:1904.02117

U.F.P. Seifert+LB, PRB **100**, 125161 (2019)



- Fermionic two-spinon continuum modified by *Silin* spin wave and gauge continuum.
- Further work: modifications near $q=0$ by anisotropies/SOC - needed to understand ESR, extension to Dirac spin liquids

- Theory of light-induced magnetization oscillations in an antiferromagnet, with direct application to Sr_2IrO_4



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