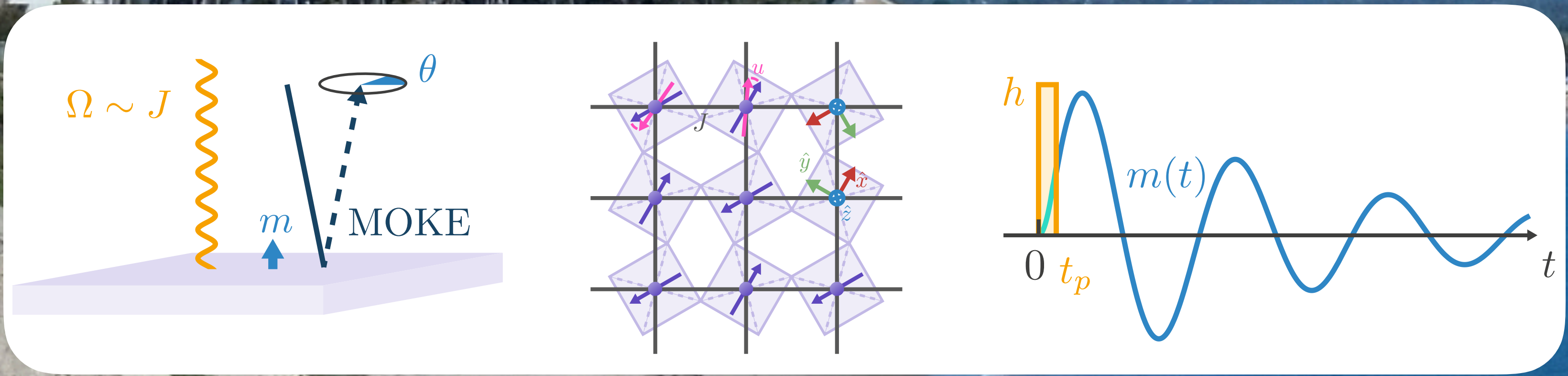
Dynamics of quantum magnets

Leon Balents, KITP, UCSB



CEMS symposium on Emergent Quantum Materials, May 2019, Tokyo

This talk

1. Main subject: dynamics of an antiferromagnet subjected to strong optical fields

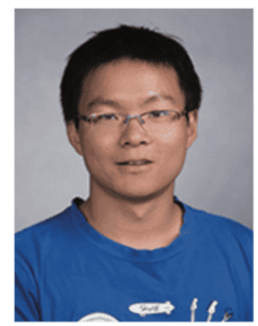
2. If there's time: some new results on excitations of a quantum spin liquid

Collaborators

Urban Seifert, TU Dresden

Oleg Starykh, U. Utah

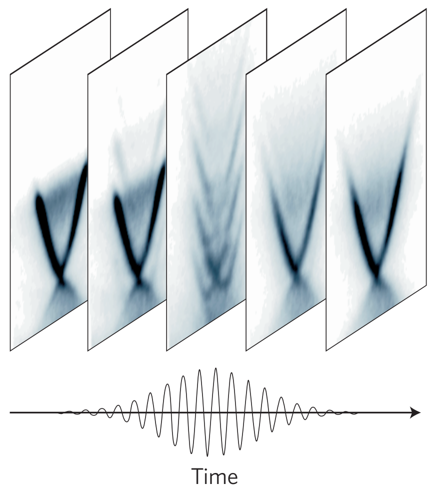
experimental inspiration



Rick Averitt Gufeng Zhang

UCSD

Ultra-fast Manipulation of Quantum Matter



Floquet-Bloch states in Bi_2Se_3

Wang *et al*, Science 2013

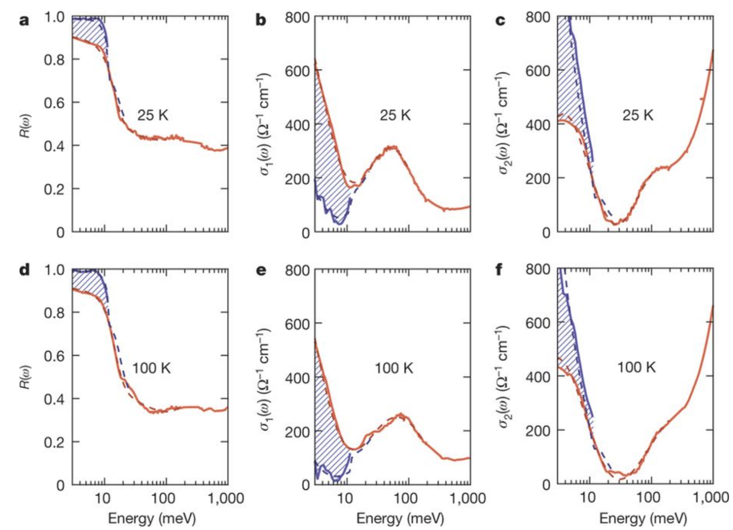


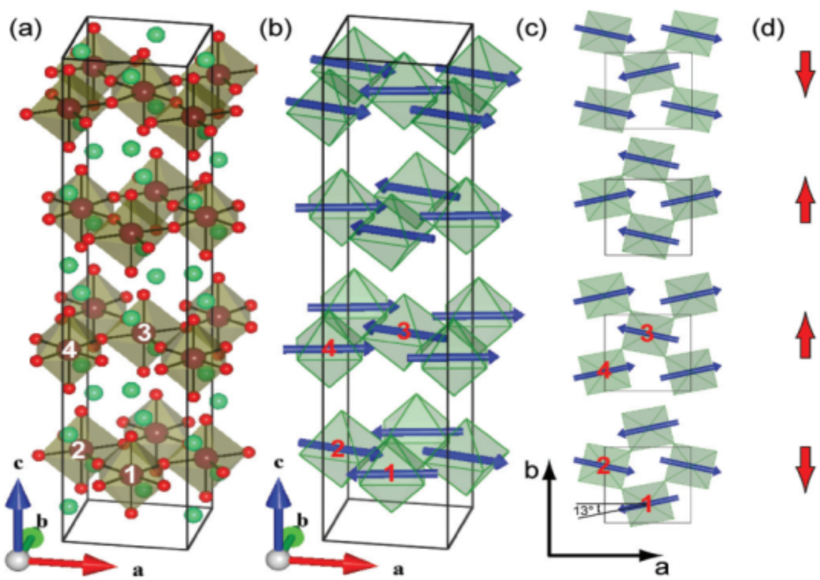
Photo-induced conductivity changes in K_3C_{60}

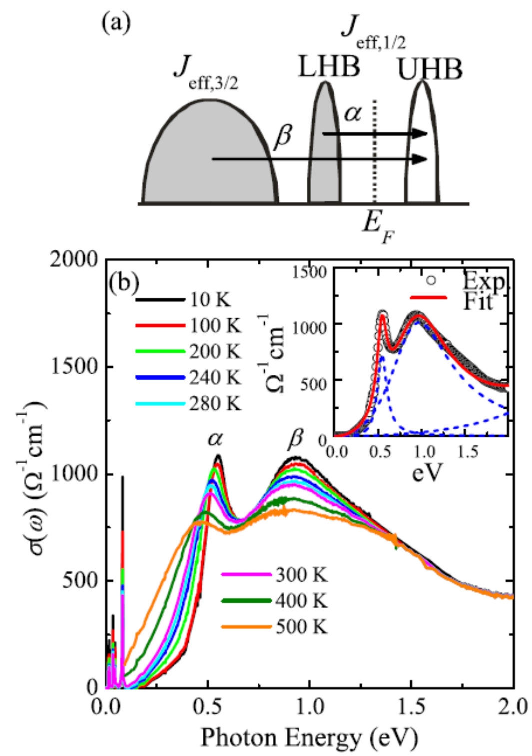
Mitrano *et al*, Nature 2016

Couple to electrons

Couple to phonons?

Sr_2IrO_4



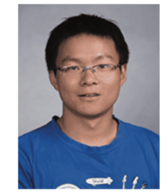


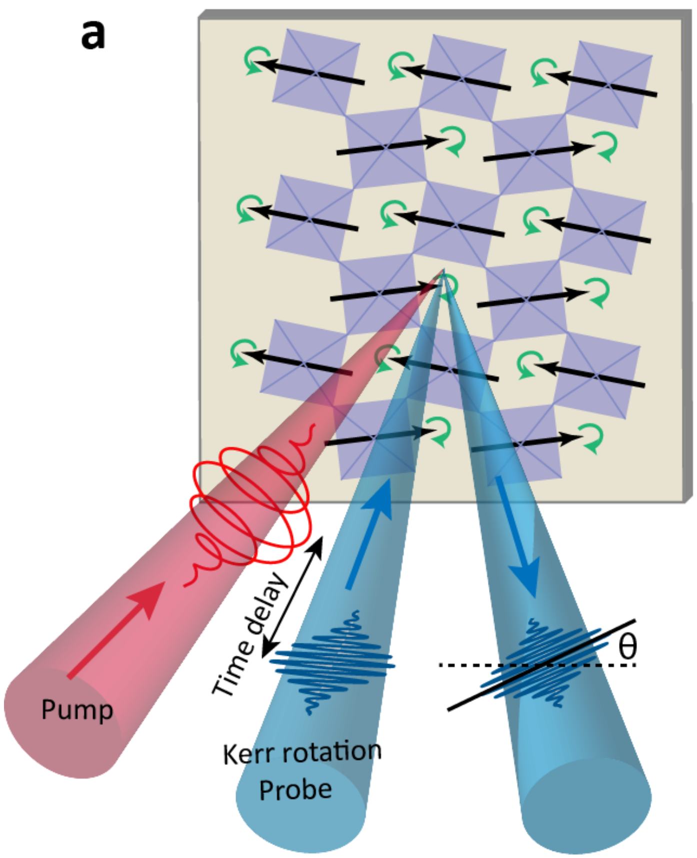
Square lattice antiferromagnet Strong SOC

Good venue for light-spin interactions

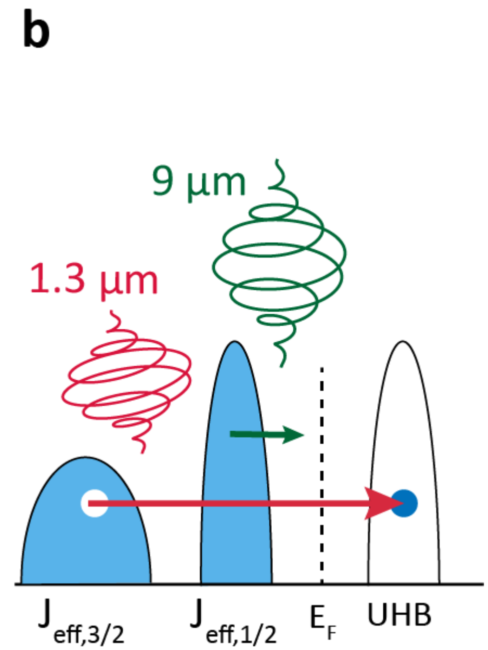
Ultrafast experiments

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



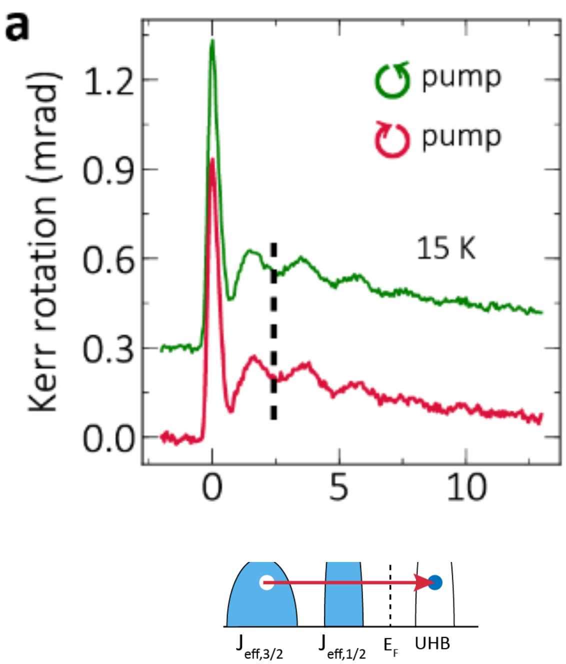
Kerr angle ~ magnetization



"Resonant" versus "non-resonant"

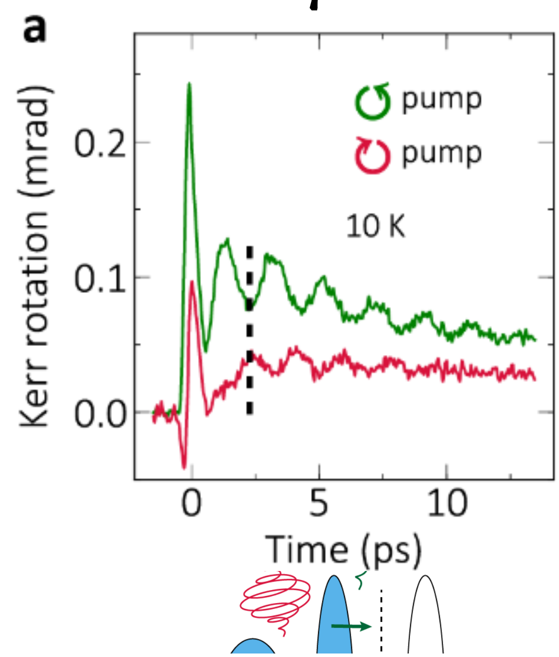
Phenomena

$1.3\ \mu m$



θ_K =.029 mrad mJ^{-1} cm^2

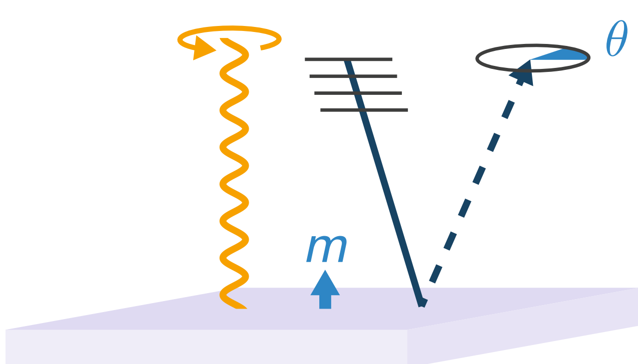
$9\ \mu m$

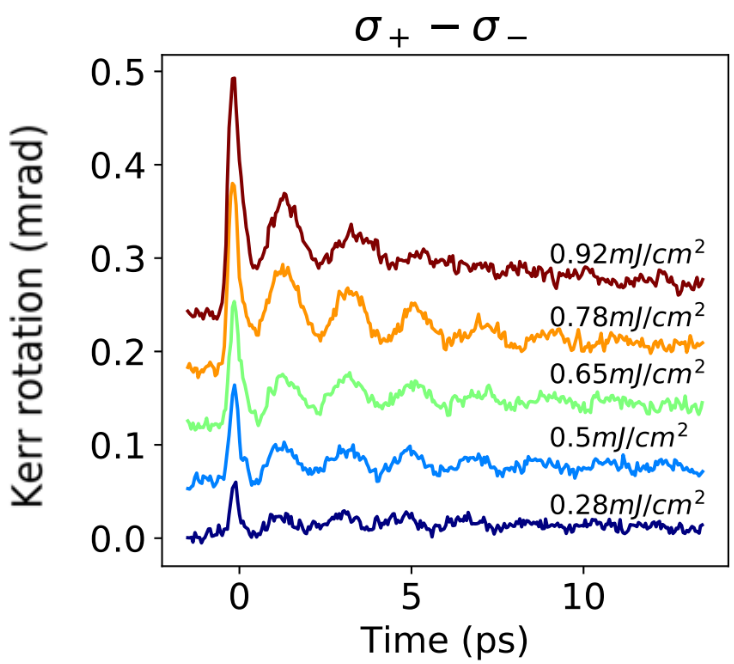


θ_K =.24 mrad mJ^{-1} cm^2

- Oscillation frequency 0.57THz = 2meV independent of details of pump: intrinsic magnon energy
- Pumping <u>much more efficient</u> for 9micron light.

Phenomena

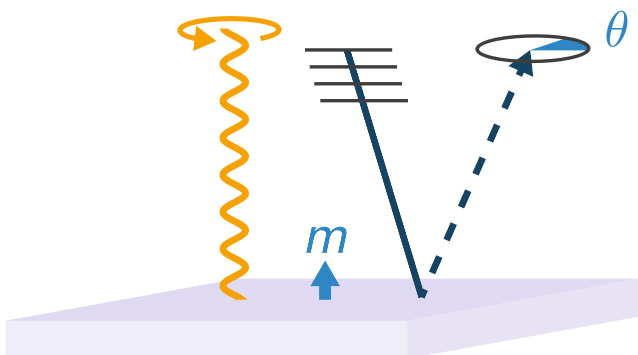


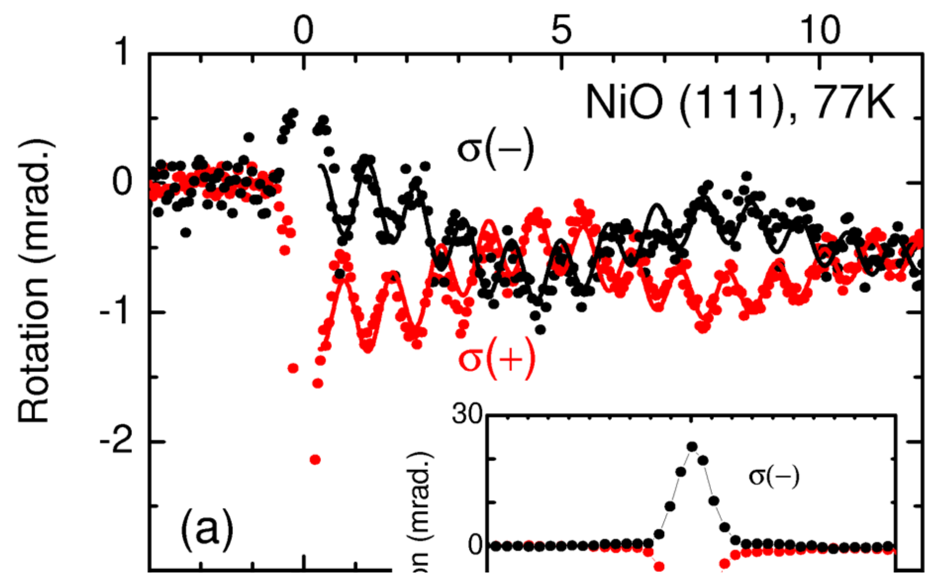


Questions

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

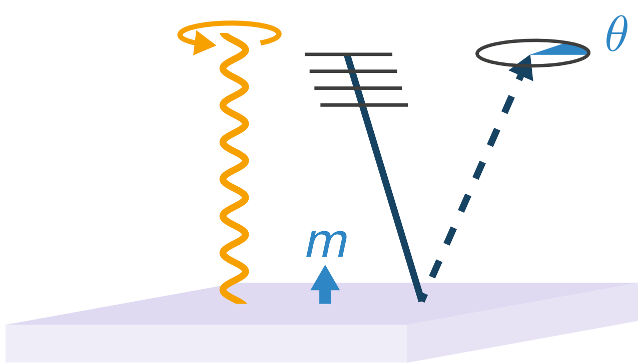
Phenomena

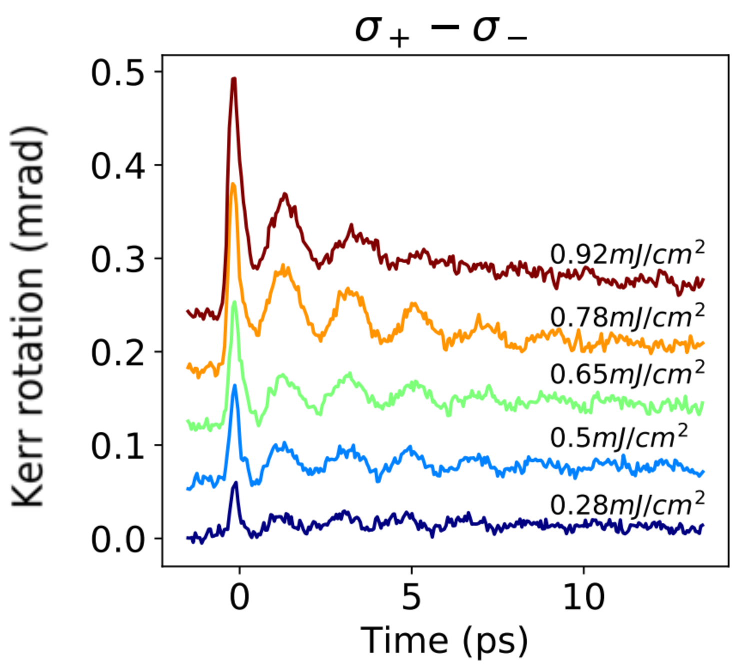




c.f. T. Satoh *et al*, 2010 - NiO

Phenomena





Questions

- What is the magnon oscillation and why is it visible?
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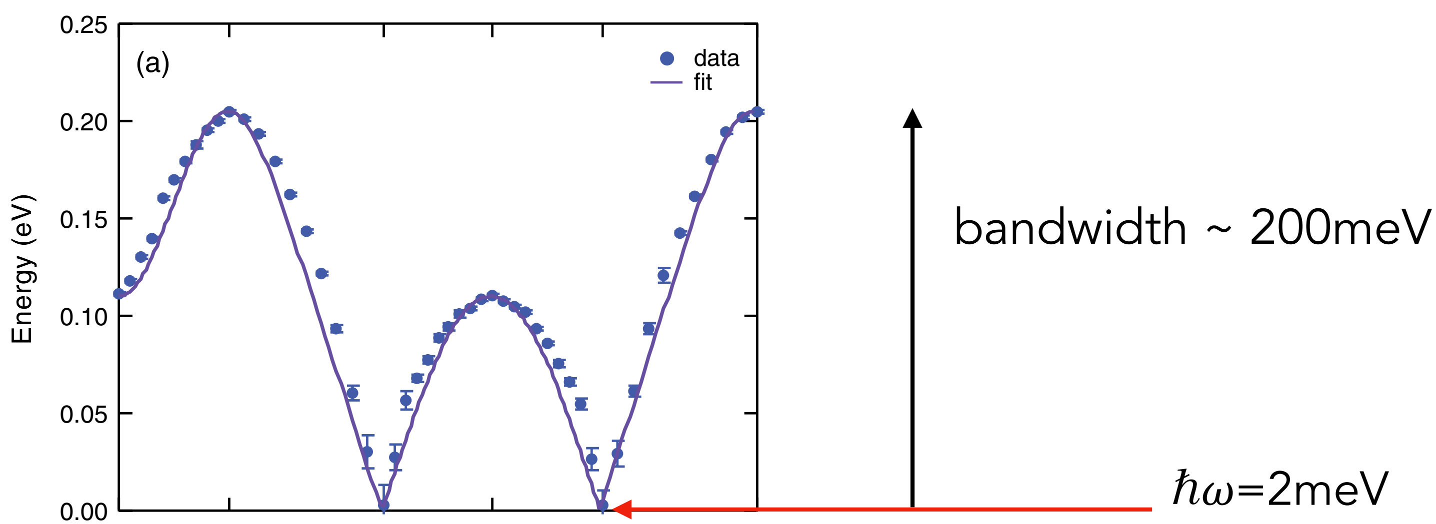
Magnons

- Gross features: square lattice Heisenberg antiferromagnet



RIXS: J. Kim *et al*, PRL 2012

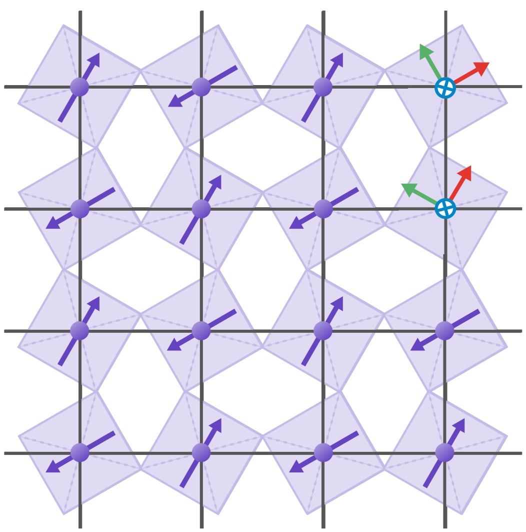
Magnon is a *very* low energy feature

Anisotropy

Unit cell doubled by octahedral rotation

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

XXZ+DM



DM is removed in local frame

G. Jackeli+G.Khaliullin, 2009

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

Fits to RIXS give J~60meV and δ~.05

Small easy-plane anisotropy

Spin wave theory

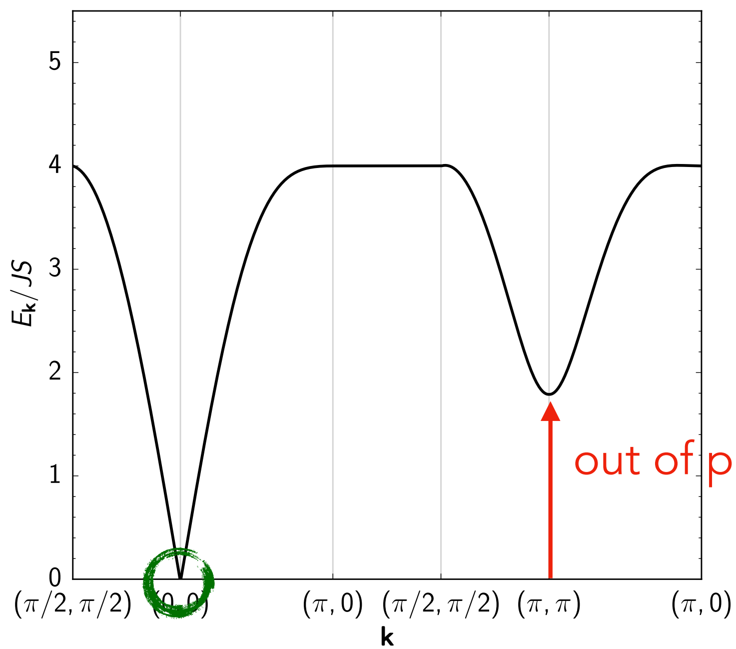
- Holstein-Primakoff

$$S^+ = S^z - \mathrm{i}S^y = \sqrt{2S - a^\dagger a}\, a$$
$$S^- = (S^+)^\dagger \qquad S^x = S - a^\dagger a$$

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

E_{cl} E_{sw} magnon interactions

$$\mathcal{H}^{(2)}_{\mathrm{eq}} = \sum_{\mathbf{k}} E_{\mathbf{k}}\, \alpha^\dagger_{\mathbf{k}} \alpha_{\mathbf{k}}$$

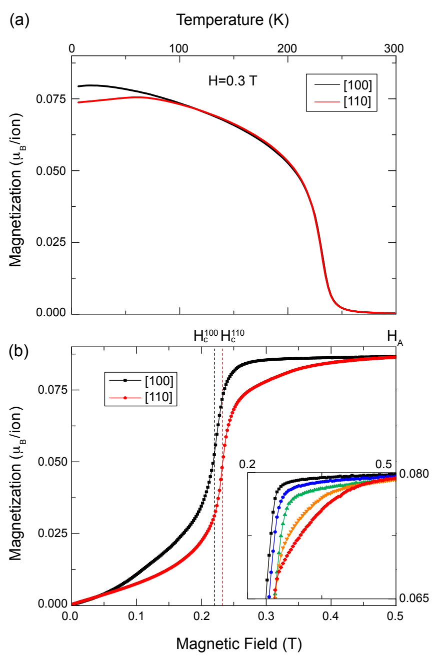


out of plane mode. Gap ~ 30meV! Too big to be the oscillating magnon

gapless Goldstone mode of in-plane rotations

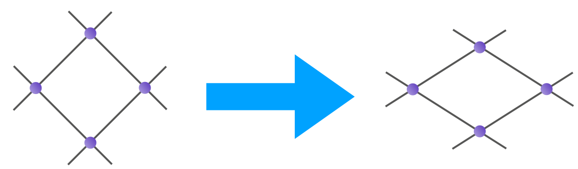
In-plane anisotropy

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



Porras *et al*, 2019

argue due to lattice distortion induced by spin order



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

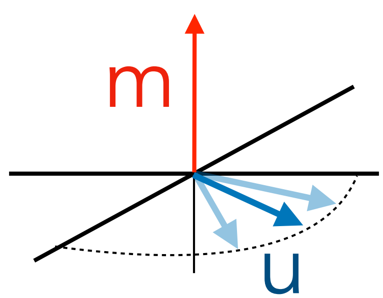
$\mathbf{\Gamma}$~6μeV (!) $\omega_0 \simeq 8S\sqrt{\Gamma J}$ ~2meV

Oscillation matches in-plane magnon.

Magnetization oscillation

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

In-plane spin rotation ⟷ 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

$$u = \sqrt{S/2}\, \mathrm{i}\left(a - a^\dagger\right)$$
$$m = \sqrt{S/2}\left(a + a^\dagger\right)$$

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

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

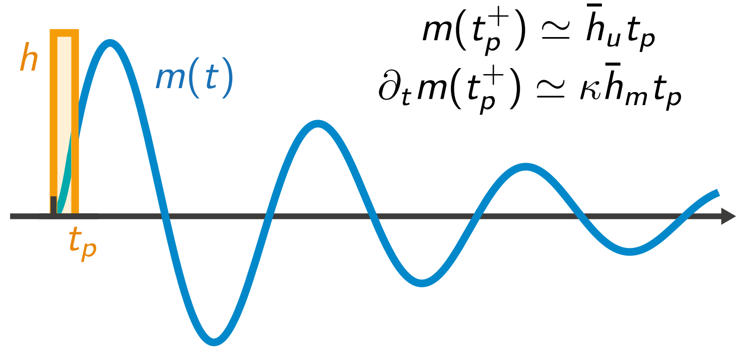
Pumping

- Strategy: light creates *source terms* for EOM

$$\partial_t u = \chi^{-1} m + h_m(t)$$
$$\partial_t m = -\kappa\, u + h_u(t)$$

effective fields drive during pump pulse

decay/relaxation negligible during pump



$$m(t_p^+) \simeq \bar{h}_u t_p$$
$$\partial_t m(t_p^+) \simeq \kappa \bar{h}_m t_p$$

effective initial conditions

<u>Theory</u>: 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

$$\mathcal{T}: \vec{E} \to \vec{E},\ \vec{S} \to -\vec{S} \qquad \Longrightarrow \qquad \mathcal{H}_{\mathrm{E}} \sim g\, E^{\alpha} S_i^{\beta} S_j^{\gamma}$$

- General symmetry allowed couplings

$$\begin{aligned}\mathcal{H}_{\mathrm{E}} = \sum_i \Big[& g_1 \epsilon_i \left[E_x \left(\mathrm{S}_i^y \mathrm{S}_{i+x}^x + \mathrm{S}_i^x \mathrm{S}_{i+x}^y \right) - (x \leftrightarrow y) \right] \\ & + g_2 \epsilon_i \left(E_y \mathrm{S}_i^x \mathrm{S}_{i+x}^x - E_x \mathrm{S}_i^y \mathrm{S}_{i+y}^y \right) \\ & + g_3 \epsilon_i \left(E_y \mathrm{S}_i^y \mathrm{S}_{i+x}^y - E_x \mathrm{S}_i^x \mathrm{S}_{i+y}^x \right) \\ & + g_4 \epsilon_i \mathrm{S}_i^z \left(E_y \mathrm{S}_{i+x}^z - E_x \mathrm{S}_{i+y}^z \right) \\ & + g_5 \left(E_y\, \hat{\mathbf{z}} \cdot \mathrm{S}_i \times \mathrm{S}_{i+x} - E_x\, \hat{\mathbf{z}} \cdot \mathrm{S}_i \times \mathrm{S}_{i+y} \right) \Big]\end{aligned}$$

staggering due to octahedral rotations important!

microscopic calculations confirm these terms

Katsura, Nagaosa, Balatsky, PRL 2005
Bolens, PRB 2018

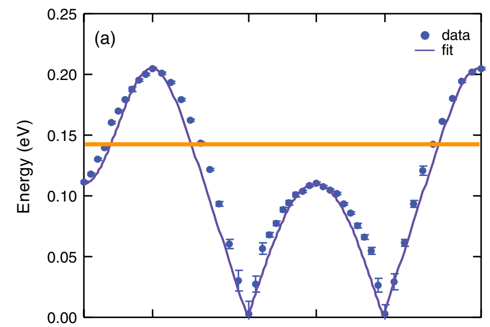
Bosons

- Holstein-Primakoff: $\psi_{\mathbf{k}} = (a_{\mathbf{k}}, a^{\dagger}_{-\mathbf{k}})^T$

$$\mathcal{H}_{\mathrm{E}} = \boxed{E_\mu(t)}\left[\Phi^{1,\mu}_{A}\,\psi_A \;+\; \Phi^{2,\mu}_{A,B}\,\psi_A\psi_B \;+\; \Phi^{3,\mu}_{A,B,C}\,\psi_A\psi_B\psi_C \;+\; \mathcal{O}(1/S^0)\right]$$

$$\sim \mathrm{Re}\left(\mathcal{E}_\mu e^{i\omega t}\right)$$

Floquet drive



- Cannot excite single magnon due to momentum conservation
- Two magnon process is resonant: generates pairs with **k** and **-k**

?? How is **k=0** magnetization created?

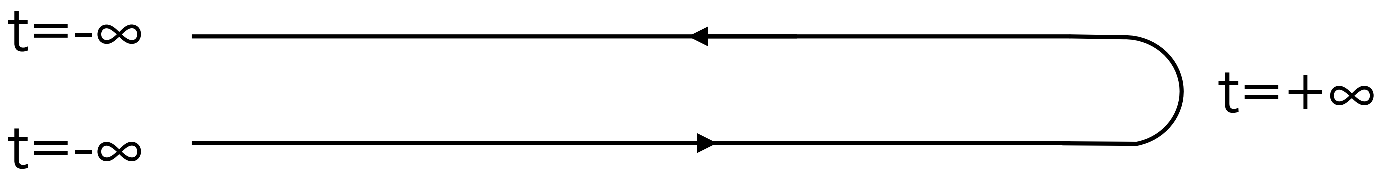
Formalism

- 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



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

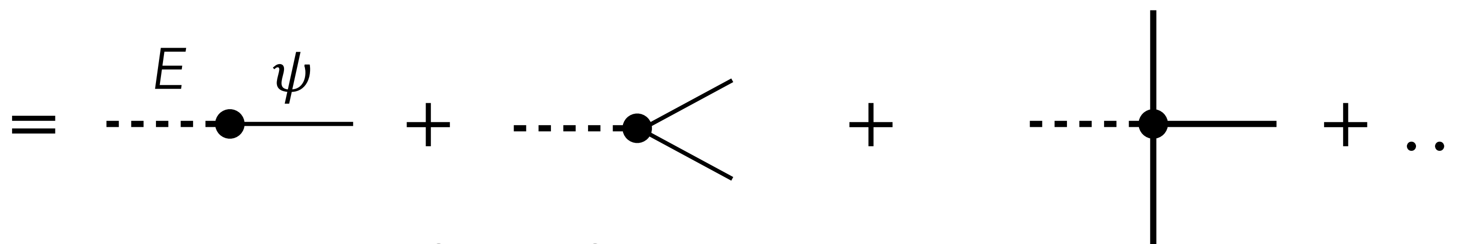
$$\mathcal{S} = \sum_{s=\pm} s \int \mathrm{d}t \left\{ \sum_i \bar{a}_{s,i}\mathrm{i}\partial_t a_{s,i} - \mathcal{H}[\{\bar{a}_{s,i}, a_{s,i}\}] \right\}$$

path integral over doubled fields

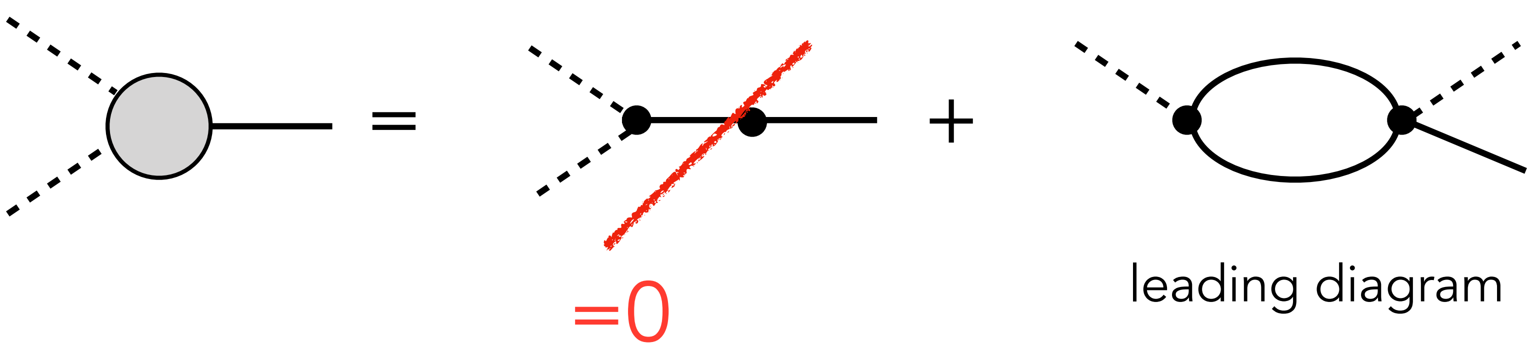
Formalism

- Non-equilibrium Keldysh path integral

$$\mathcal{H}_{\mathrm{E}} = E_\mu(t)\left[\Phi^{1,\mu}_{A}\,\psi_A \;+\; \Phi^{2,\mu}_{A,B}\,\psi_A\psi_B \;+\; \Phi^{3,\mu}_{A,B,C}\,\psi_A\psi_B\psi_C \;+\; \mathcal{O}(1/S^0)\right]$$



- Now integrate out "fast" fields: internal lines



Formalism

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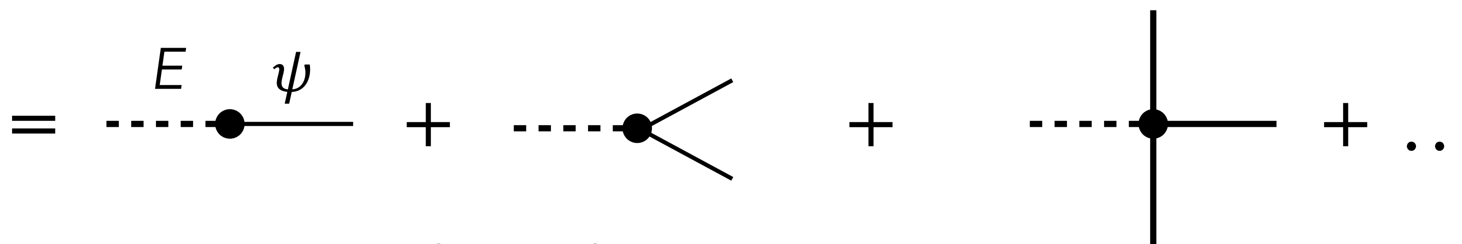
$$\mathcal{H}_{\mathrm{E}} = E_\mu(t)\big[\Phi^{1,\mu}_{A}\,\psi_A \;+\; \Phi^{2,\mu}_{A,B}\,\psi_A\psi_B \;+\; \Phi^{3,\mu}_{A,B,C}\,\psi_A\psi_B\psi_C \;+\; \mathcal{O}(1/S^0)\big]$$



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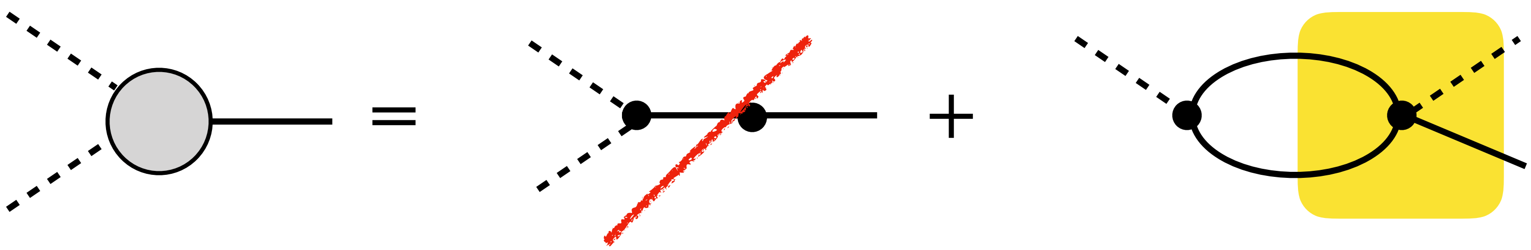


=0

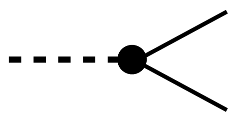
leading diagram

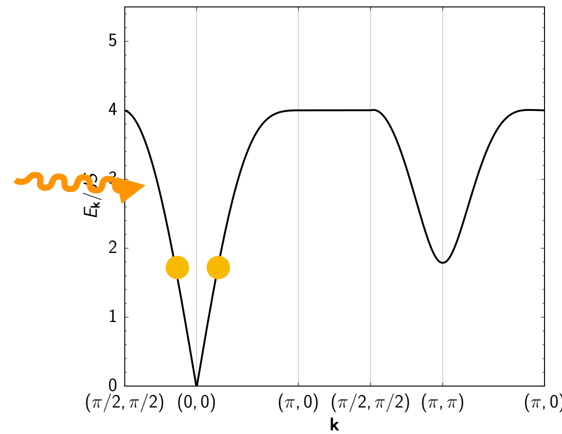
involves boson interactions!

Physical picture

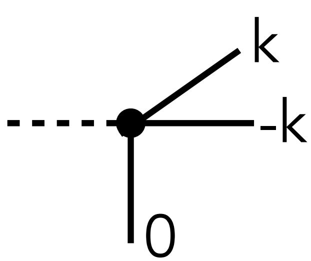
- Step 1:

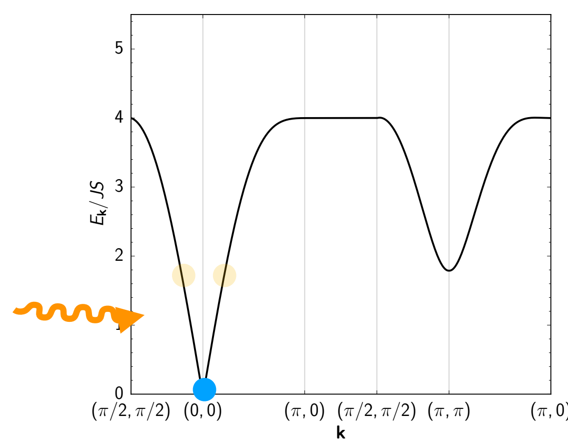
  photon generates coherent pairs





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





Results

- Effective fields given by loop integrals

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

separates into:

	dissipative	virtual
	$\sim \delta(\Omega - E_{\mathbf{k}} - E_{\mathbf{Q}-\mathbf{k}})$	$\sim \frac{P}{\delta - E_{\boldsymbol{k}} - E_{\boldsymbol{Q}-k}}$

Results

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

- Structure of contributions to effective fields

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

Results

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

separates into:

dissipative

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

virtual

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

- Structure of contributions to effective fields

$$\boxed{\sin 2\phi \times \begin{cases} \mathcal{E}_x\bar{\mathcal{E}}_y + \mathcal{E}_y\bar{\mathcal{E}}_x \\ \mathcal{E}_x\bar{\mathcal{E}}_x - \mathcal{E}_y\bar{\mathcal{E}}_y \end{cases} \qquad \text{total intensity} \quad \sin 4\phi \times \left(\mathcal{E}_x\bar{\mathcal{E}}_x + \mathcal{E}_y\bar{\mathcal{E}}_y\right)}$$

chiral intensity

$$\mathrm{i}\left(\mathcal{E}_x\bar{\mathcal{E}}_y - \mathcal{E}_y\bar{\mathcal{E}}_x\right)$$

linear polarization

Results

- Effective fields given by loop integrals

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

separates into:

dissipative	virtual
$\sim \delta(\Omega - E_{\mathbf{k}} - E_{\mathbf{Q}-\mathbf{k}})$	$\sim \dfrac{P}{\delta - E_{\boldsymbol{k}} - E_{\boldsymbol{Q}-k}}$

- Structure of contributions to effective fields

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

circular polarization

Results

- Effective fields given by loop integrals

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

separates into:

dissipative	virtual
$\sim \delta(\Omega - E_{\mathbf{k}} - E_{\mathbf{Q}-\mathbf{k}})$	$\sim \frac{P}{\delta - E_{\boldsymbol{k}} - E_{\boldsymbol{Q}-k}}$

- 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
$\sin 4\phi \times \left(\mathcal{E}_x\bar{\mathcal{E}}_x + \mathcal{E}_y\bar{\mathcal{E}}_y\right)$	$\mathrm{i}\left(\mathcal{E}_x\bar{\mathcal{E}}_y - \mathcal{E}_y\bar{\mathcal{E}}_x\right)$

=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

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

really correct for low frequency magnetization (not short pulse)

- Carried out for quantum mechanical few-level system

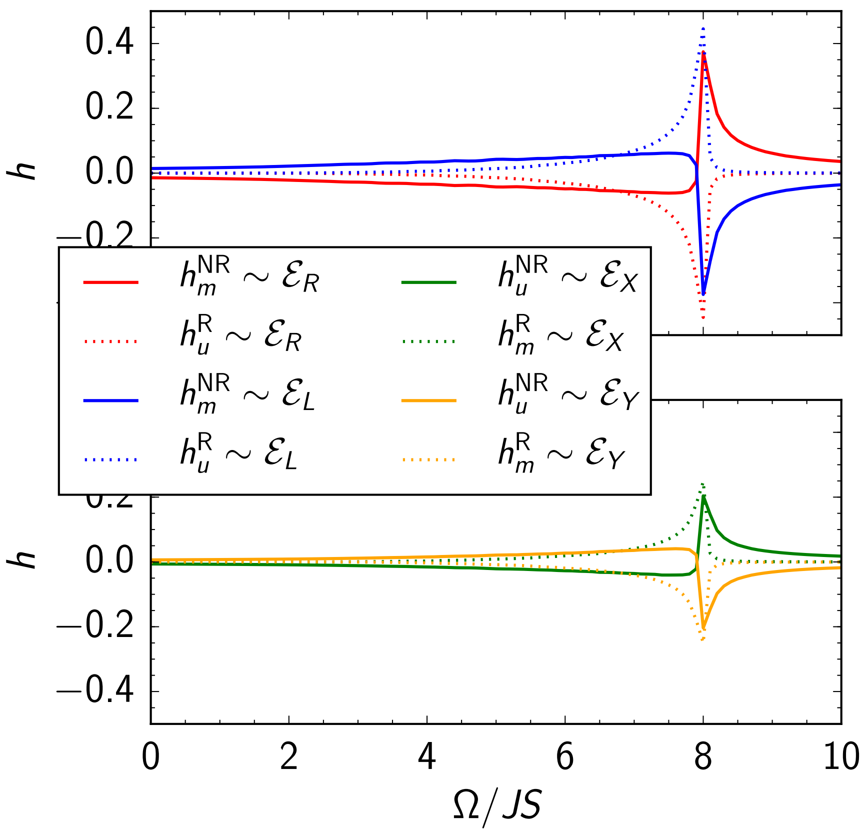
$$\begin{aligned}\mathcal{H}_{\text{eff}} = {} & (\mathcal{E}_R\mathcal{E}_R^* - \mathcal{E}_L\mathcal{E}_L^*)J_z A \\ & + \{(\mathcal{E}_R\mathcal{E}_R^* + \mathcal{E}_L\mathcal{E}_L^*)[J_z^2 - \tfrac{1}{3}J(J+1)] \\ & - \mathcal{E}_L\mathcal{E}_R^* J_-^2 - \mathcal{E}_L^*\mathcal{E}_R J_+^2\}C,\end{aligned}$$

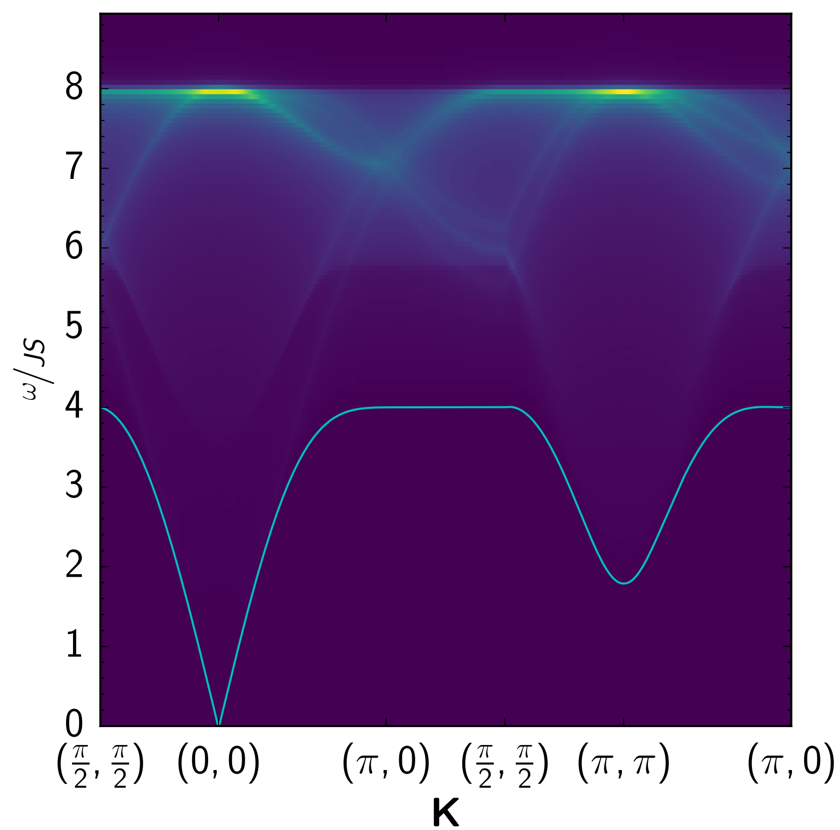
Purely virtual excitations

- 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

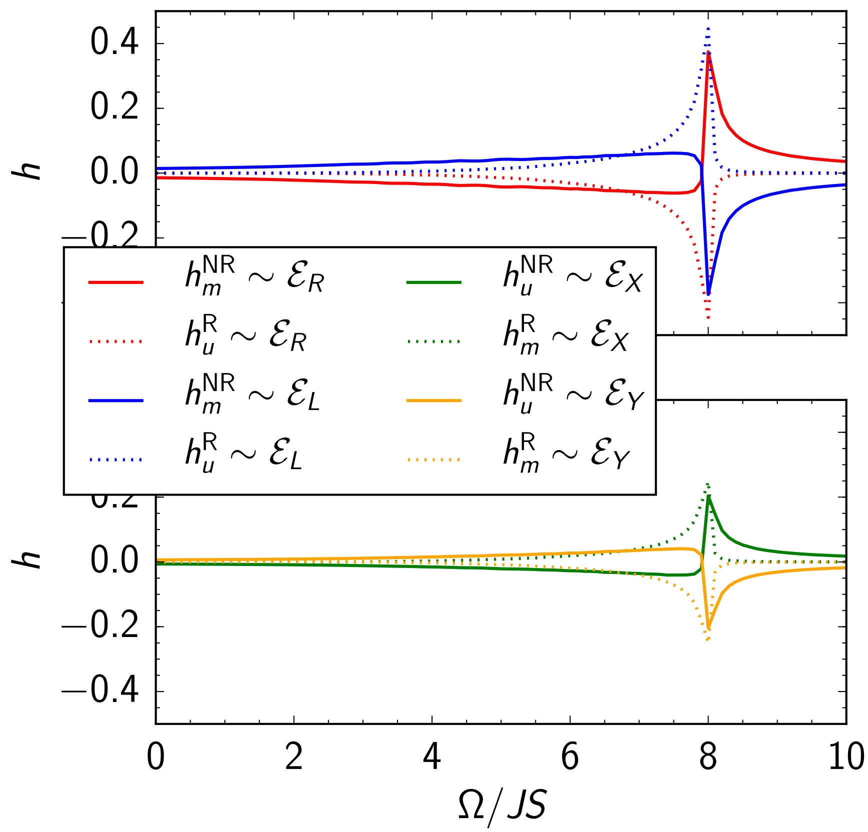




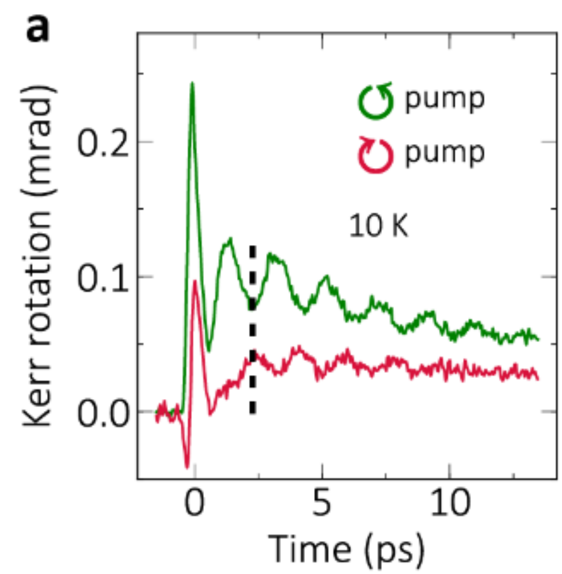
Significant enhancement still possible!

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?

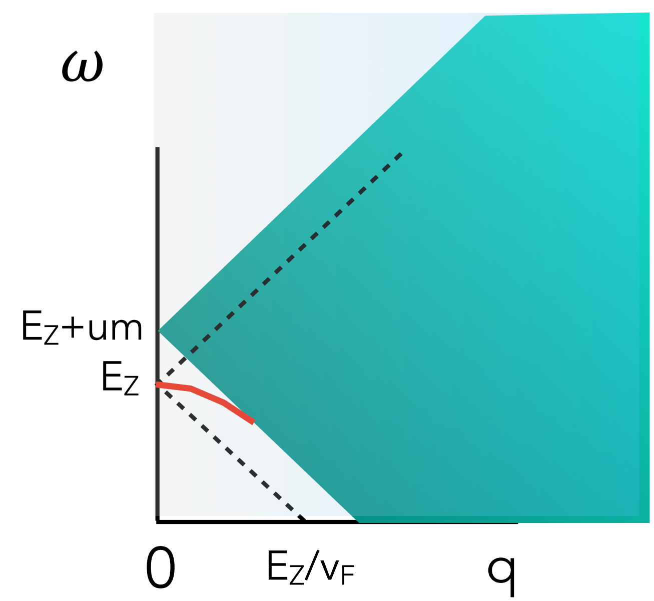
New results

Spinon interactions strongly modify their spectra

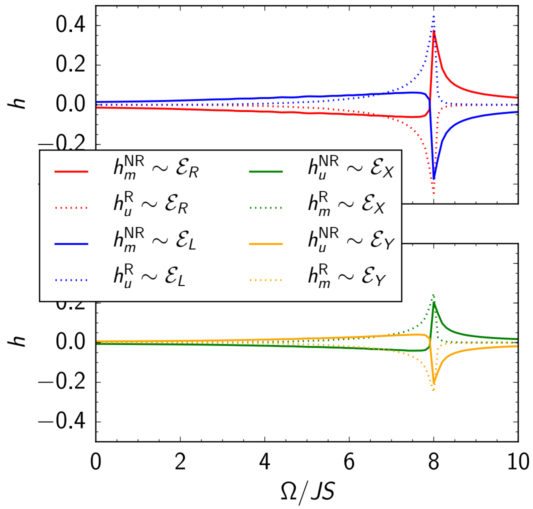
The effect becomes dramatic in an applied Zeeman field:

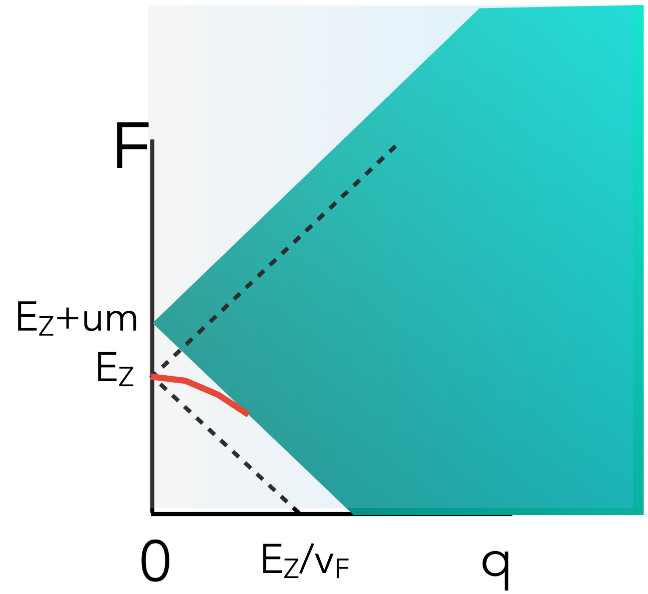
Dominant spectral weight at small momentum is a collective "**Spinon wave**" mode

Full small q-ω structure given in LB+O. Starykh, arXiv:1904.02117



Recap





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

- 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

SFB 1143

arXiv:1905.01313

arXiv:1904.02117