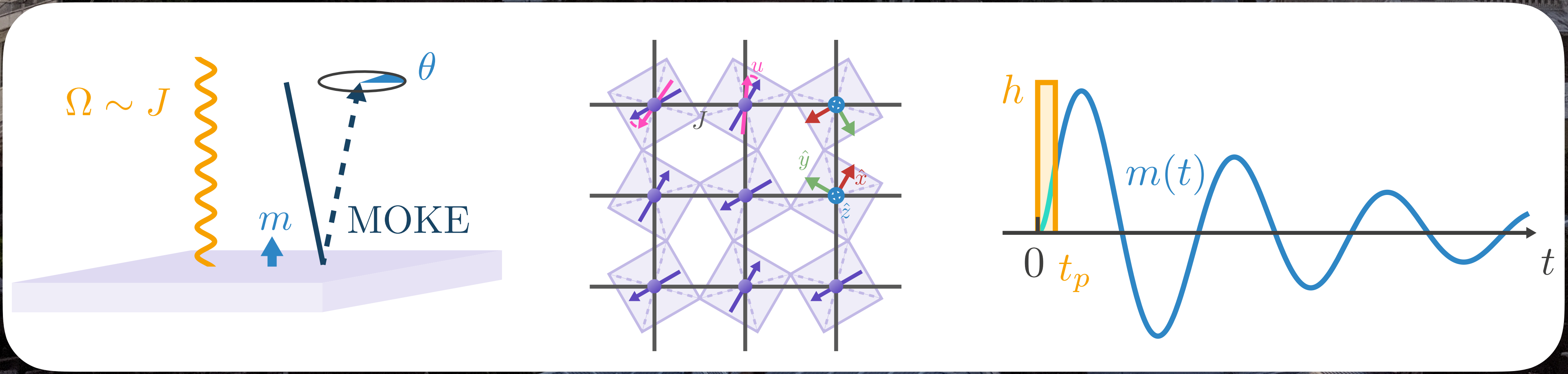
Exciting excitations in quantum magnets

Leon Balents, KITP, UCSB



Tbilisi, Georgia, June 2019

Exciting excitations in quantum magnets

Leon Balents, KITP, UCSB

Rustaveli:

Spending on feasting and wine is better than hoarding our substance. That which we give makes us richer, that which is hoarded is lost.

Tbilisi, Georgia, June 2019

This talk

1. Unusual excitations: bound states in a 1d antiferromagnet: α-$Na_{0.9}MnO_2$

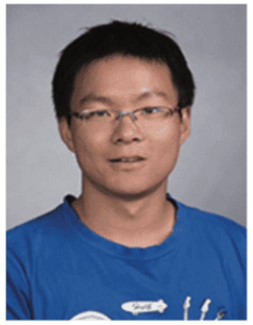
2. Literally exciting excitations in an antiferromagnet subjected by strong optical fields

People

Part I:

Stephen Wilson, UCSB   Rebecca Dally, NIST

Part II:

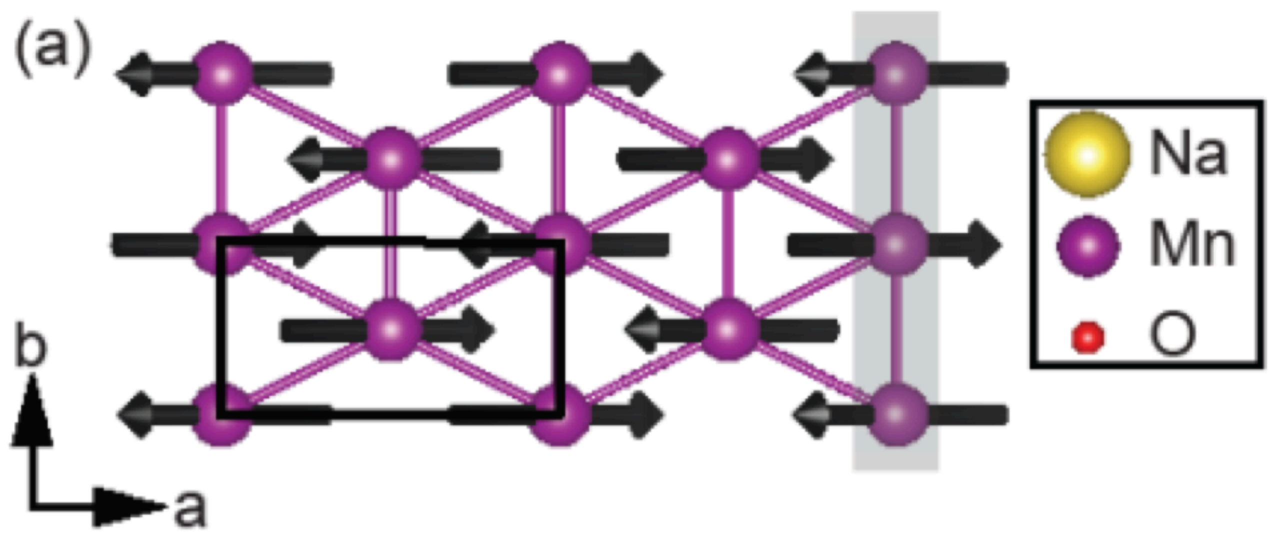


Urban Seifert, TU Dresden

Rick Averitt   Gufeng Zhang

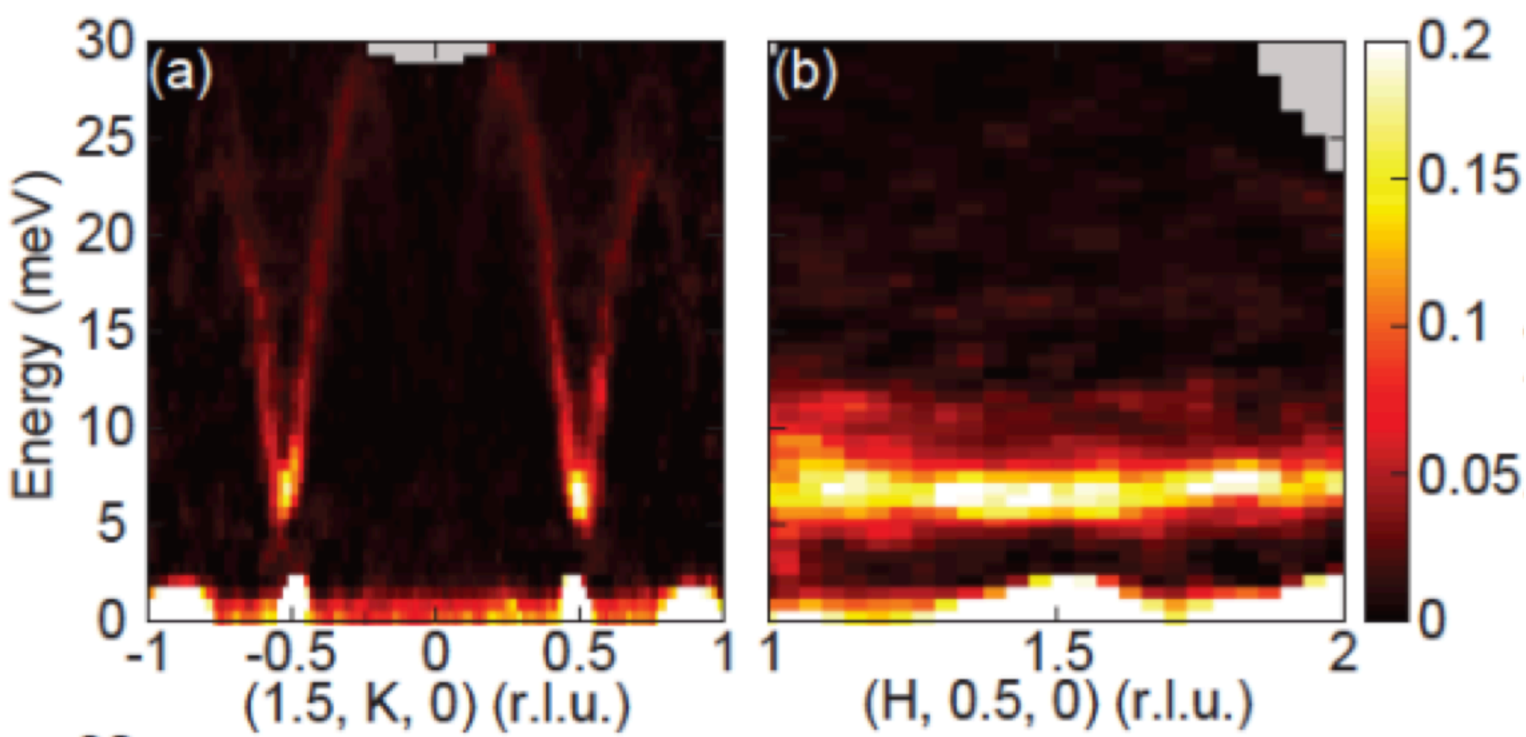
UCSD

$Na_{0.9}MnO_2$



Mn^{3+} S=2 spins
anisotropic
triangular lattice

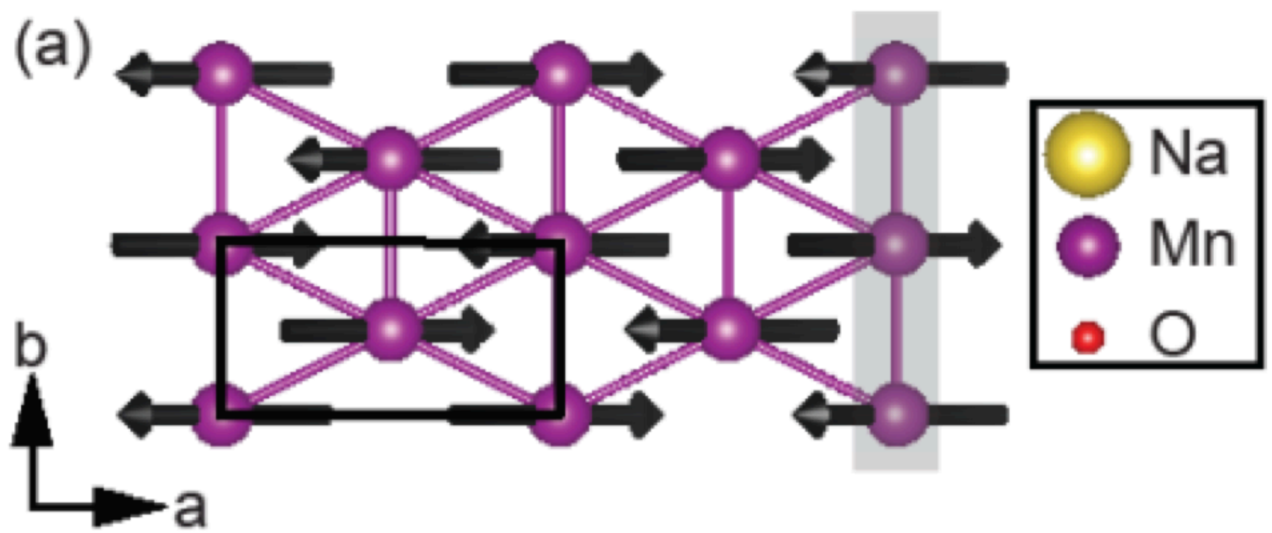
T_N=45K



Quasi-1d:

J_1 = 6.2meV
J_2 = 0.77meV

$Na_{0.9}MnO_2$



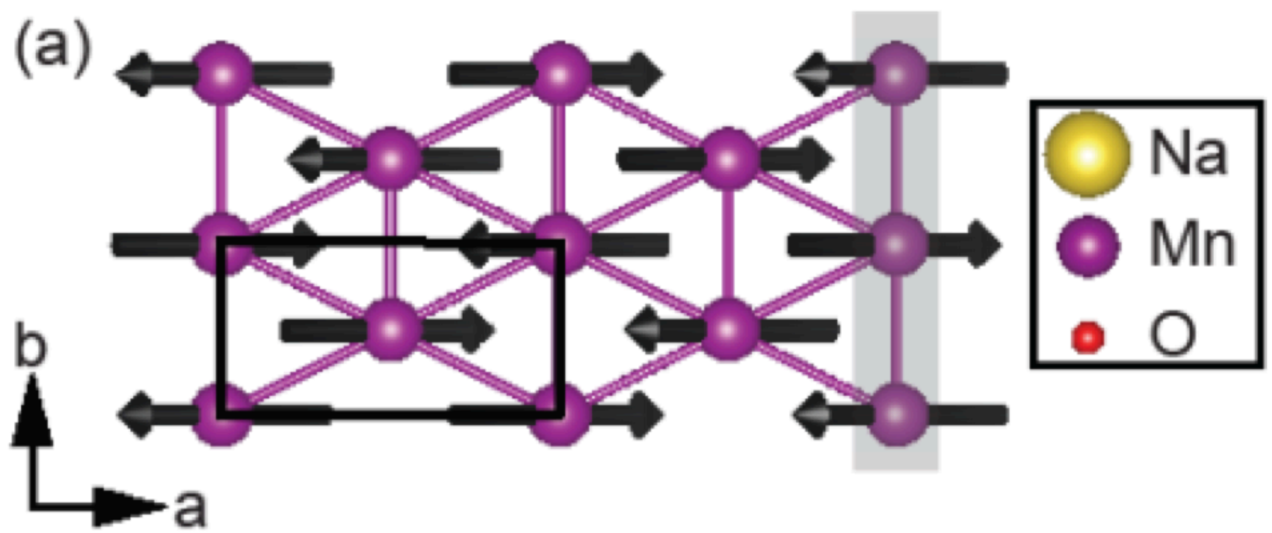
Minimal model: 1d S=2 chain with weak Ising anisotropy

$$H = \sum_n \left[J_1 \boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1} - D (S_n^z)^2 \right]$$

J_1 = 6.2meV D=0.2meV

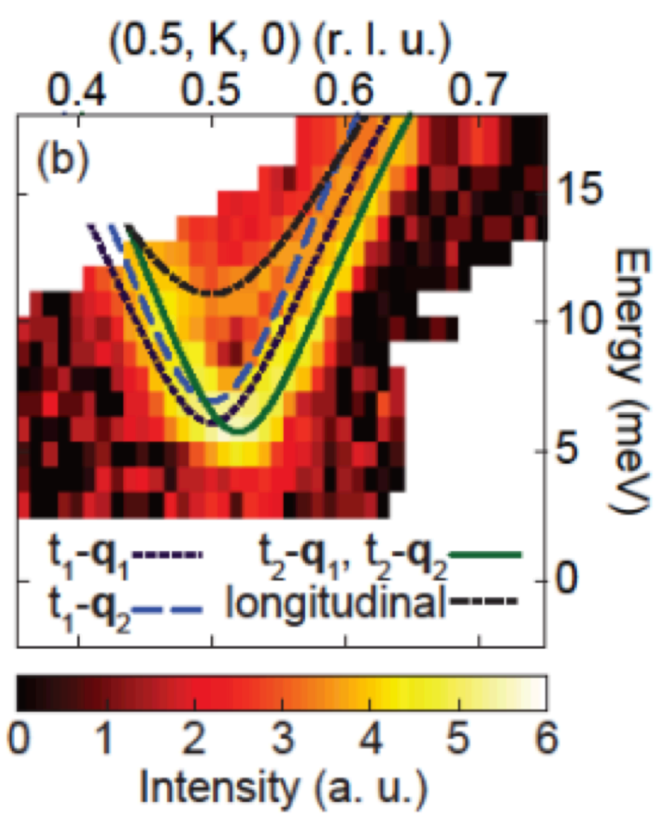
"Exciting" to find anything surprising in this very simple semi-classical ordered system

$Na_{0.9}MnO_2$



Mn^{3+} S=2 spins

anisotropic triangular lattice



INS:

- transverse mode, gap = 6.4meV
- longitudinal mode, gap = 11.0meV

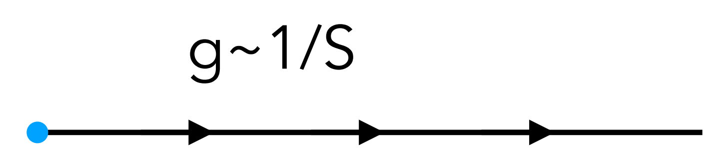
magnon bound state?

non-linear sigma model

- Semiclassical large S description

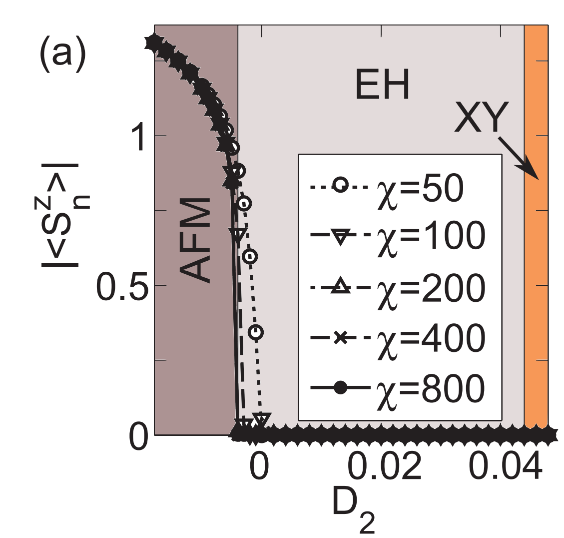
$$\mathcal{L}_E = \frac{1}{g}\left[\frac{1}{v}(\partial_\tau \hat{\boldsymbol{n}})^2 + v(\partial_x \hat{\boldsymbol{n}})^2 - DSn_z^2\right]$$

D=0: Haldane gap $\Delta_H \sim JS^2 e^{-aS}$



D>Δ_H : AF order (D/J>.0046)

Kjäll *et al*, 2013



Anisotropy gap $\Delta_D \sim \sqrt{JDS}$

spin-wave theory (perturbative NLsM) valid

$JSe^{-aS^2} \ll D \ll J \qquad 1/S \ll 1$

Spin waves: ϕ^4 theory

- Expand NLsM $\quad \hat{\boldsymbol{n}} = (\phi_1, \phi_2, \sqrt{1-\phi_1^2-\phi_2^2})$

$$\phi = \phi_1 + i\phi_2$$

- Dimensionless units

$$\mathcal{L}_E = \partial_\tau \bar{\phi} \partial_\tau \phi + \partial_x \bar{\phi} \partial_x \phi + m^2 \bar{\phi}\phi$$

$$+\frac{1}{S}\left\{-m^2\left(\bar{\phi}\phi\right)^2 + \frac{1}{2}\left[\bar{\phi}^2(\partial_x\phi)^2 + (\partial_x\bar{\phi})^2\phi^2\right]\right\}$$

attractive ϕ^4 term

NLsM interactions

dominates for k<<m

ϕ^4 theory to QMs

- Complex scalar boson $\mathcal{L}_E = \partial_\tau \bar{\phi} \partial_\tau \phi + \partial_x \bar{\phi} \partial_x \phi + m^2 \bar{\phi} \phi$

$$\phi(x) = \frac{1}{\sqrt{L}} \sum_k \frac{1}{\sqrt{2\epsilon_k}} \left(a_k e^{ikx} + b_k^\dagger e^{-ikx} \right), \sim_{k \ll m} \frac{1}{\sqrt{2m}} \left(\underbrace{a(x)}_{S^z=1} + \underbrace{b^\dagger(x)}_{S^z=-1} \right)$$

- Delta function attraction

$$\mathcal{L}_{\text{int}} \sim -\frac{1}{4S} \left(a^\dagger + b \right)^2 \left(a + b^\dagger \right)^2$$
$$\sim -\frac{1}{S} a^\dagger a b^\dagger b - \frac{1}{4S} \left(a^\dagger a^\dagger a a + b^\dagger b^\dagger b b \right) + \ldots$$

- First quantization

$$H_{\text{int}} = \sum_{i<j} u_{ij} \delta(x_i - x_j)$$

$$u_{ab} = u_{ba} = -1/S$$
$$u_{aa} = u_{bb} = -1/4S$$

~Lieb-Liniger model

Bound states

- Identical interactions (integrable)

$$H = \sum_{i=1}^{n} -\frac{1}{2m}\frac{\partial^2}{\partial x_i^2} - u\sum_{i<j}\delta(x_i - x_j)$$

$\Rightarrow$

$$\Psi = e^{-\lambda \sum_{i<j} |x_i - x_j|} \qquad \lambda = \frac{mu}{2}$$

$$E_n = -\frac{mu^2}{24}(n^3 - n)$$

"droplets"

Bound states

- Identical interactions (integrable)

$$H = \sum_{i=1}^{n} -\frac{1}{2m}\frac{\partial^2}{\partial x_i^2} - u\sum_{i<j}\delta(x_i - x_j)$$

$$\Psi = e^{-\lambda\sum_{i<j}|x_i - x_j|} \qquad \lambda = \frac{mu}{2}$$

$$E_n = -\frac{mu^2}{24}(n^3 - n)$$

"droplets"

- Here

S^z=1-1 = 0

$$H = -\frac{1}{2m}\left(\frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2}\right) - \frac{1}{S}\delta(x_1 - x_2)$$

$$E_{\text{bind}}^{1,-1} = \frac{m}{4S^2}$$

2 magnon

S^z=1+1-1 = 1

$$H = -\frac{1}{2m}\left(\frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2} + \frac{\partial^2}{\partial x_3^2}\right) - \frac{1}{4S}\delta(x_1 - x_2) - \frac{1}{S}\delta(x_1 - x_3) - \frac{1}{S}\delta(x_2 - x_3)$$

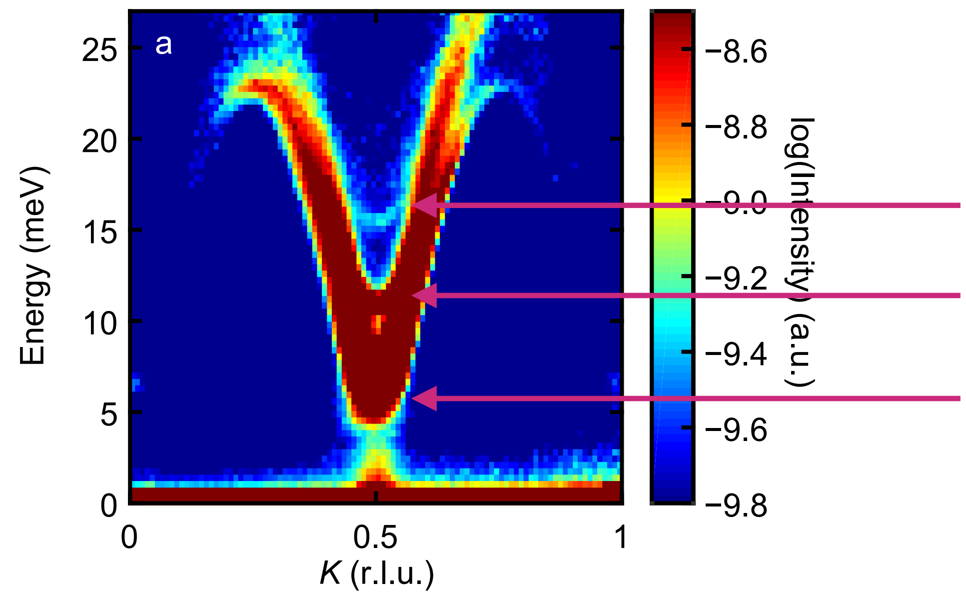
not integrable

$$E_{\text{bind}}^{1,1,-1} = 2.9E_{\text{bind}}^{1,-1}$$

3 magnon

Back to experiment

new experiment



3 magnon!

2 magnon

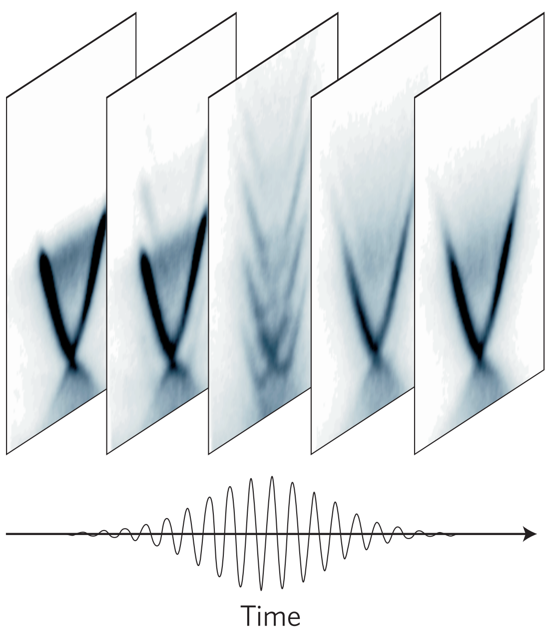
1 magnon

	E_{min} (meV)	E_{bind}		
1 magnon	6.4 (6.15)			
2 magnon	10.9 (11.1)	2*6.4-10.9 = 1.9	$\frac{E_{\mathrm{bind}}^{1,1,-1}}{E_{\mathrm{bind}}^{1,-1}} = 1.9$	(c.f. 2.9)
3 magnon	15.6	3*6.4-15.6 = 3.6		

Quantitative agreement is not great
but many improvements possible

? smaller S ? inter-chain? pair creation?

Ultra-fast Manipulation of Quantum Matter



Floquet-Bloch states in Bi_2Se_3

Wang *et al*, Science 2013

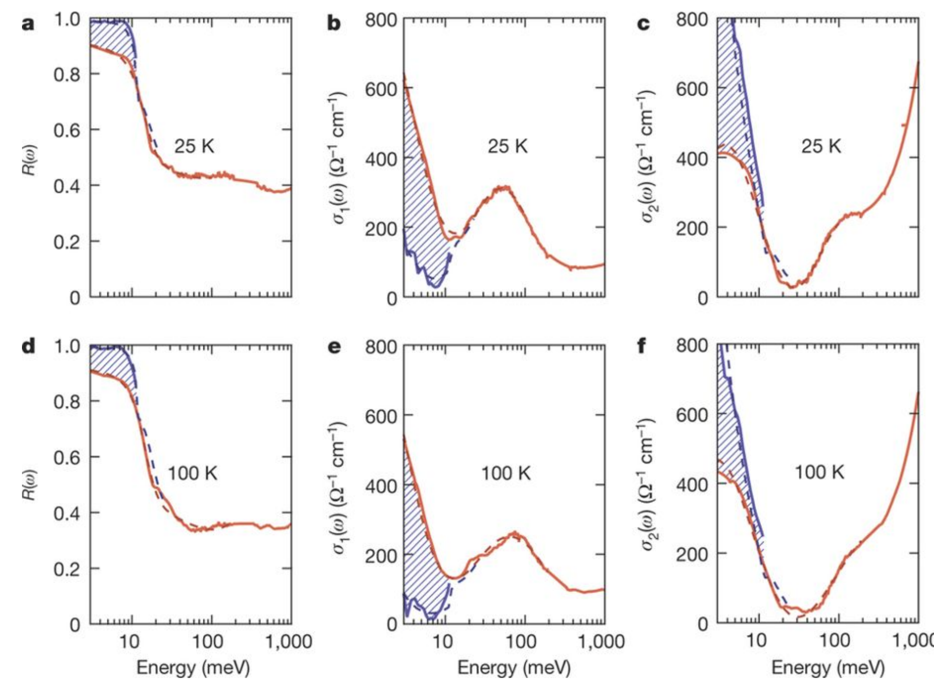


Photo-induced conductivity changes in K_3C_{60}

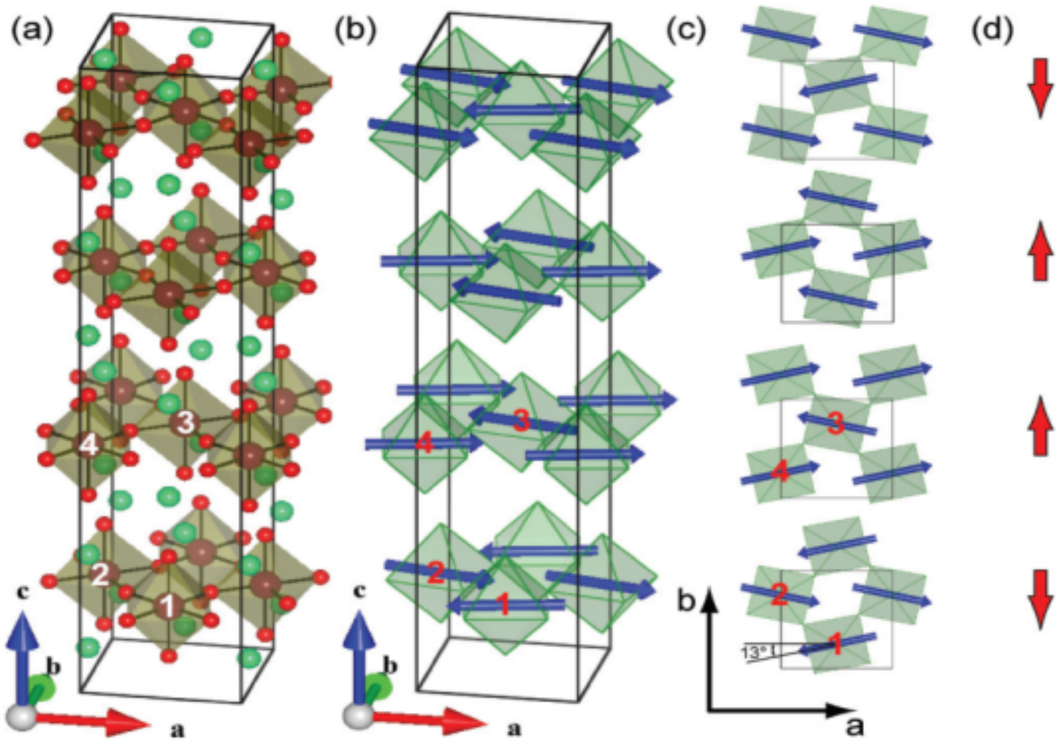
Mitrano *et al*, Nature 2016

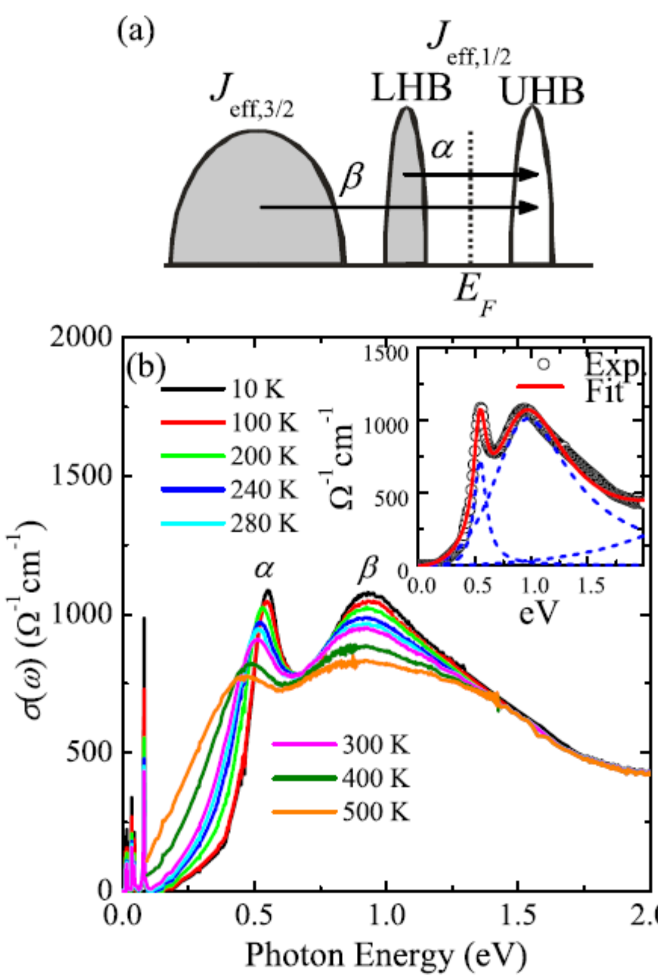
Couple to electrons

Couple to phonons?

How about spins?

Sr_2IrO_4





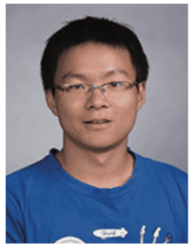
Square lattice antiferromagnet

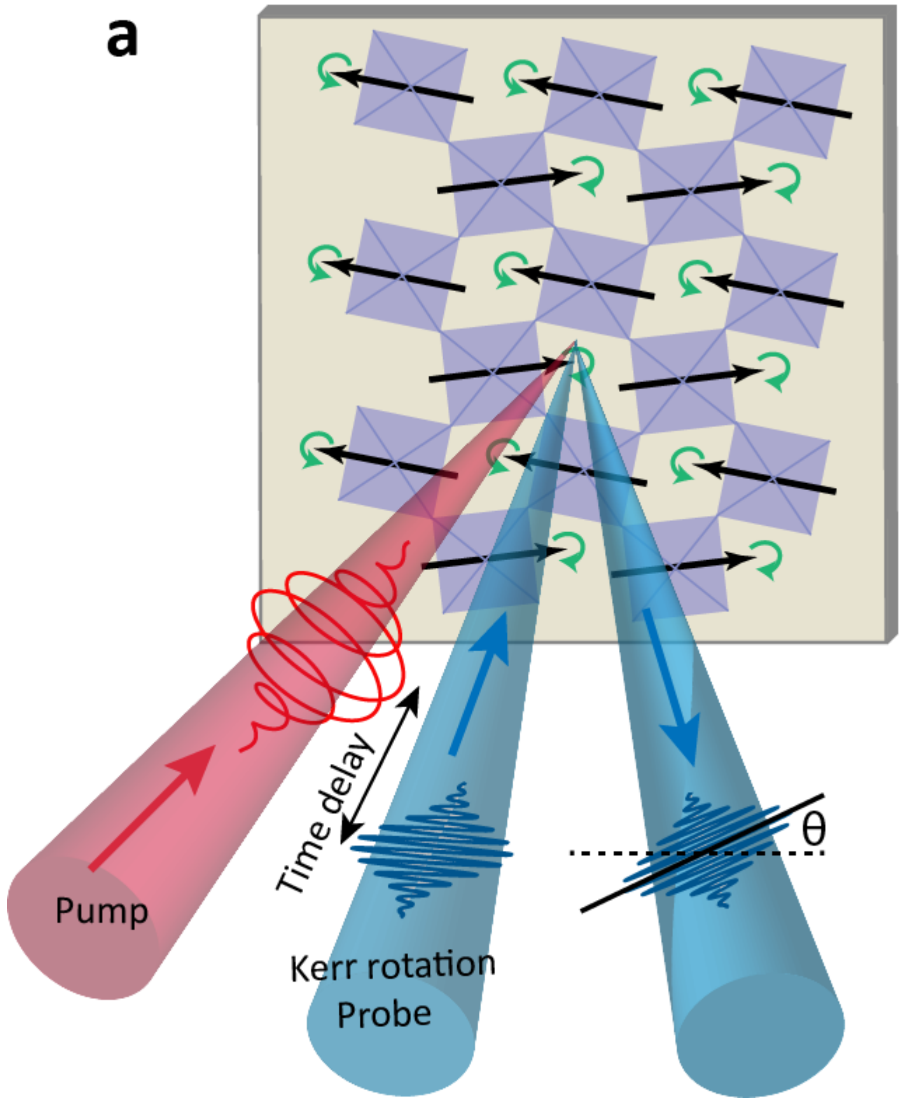
Strong SOC

Good venue for light-spin interactions

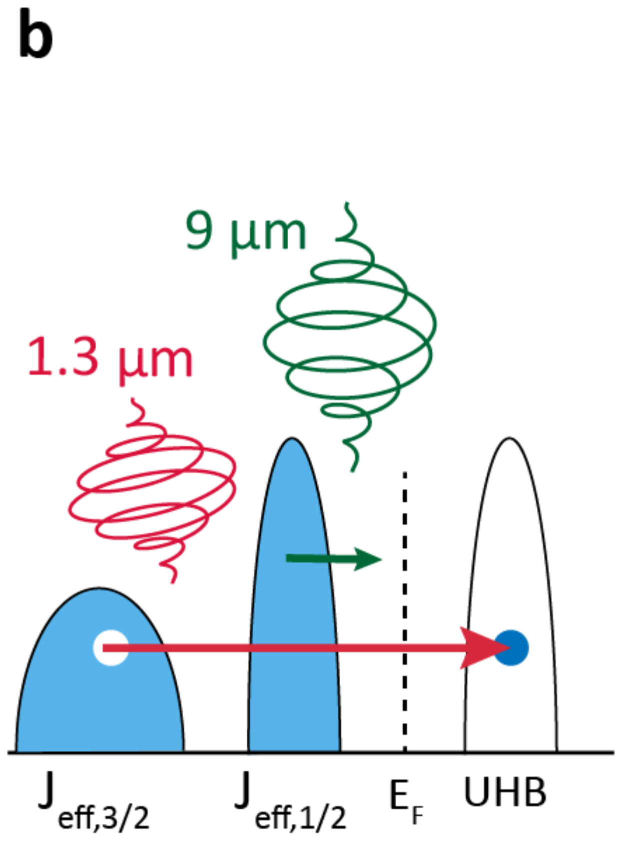
Ultrafast experiments

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





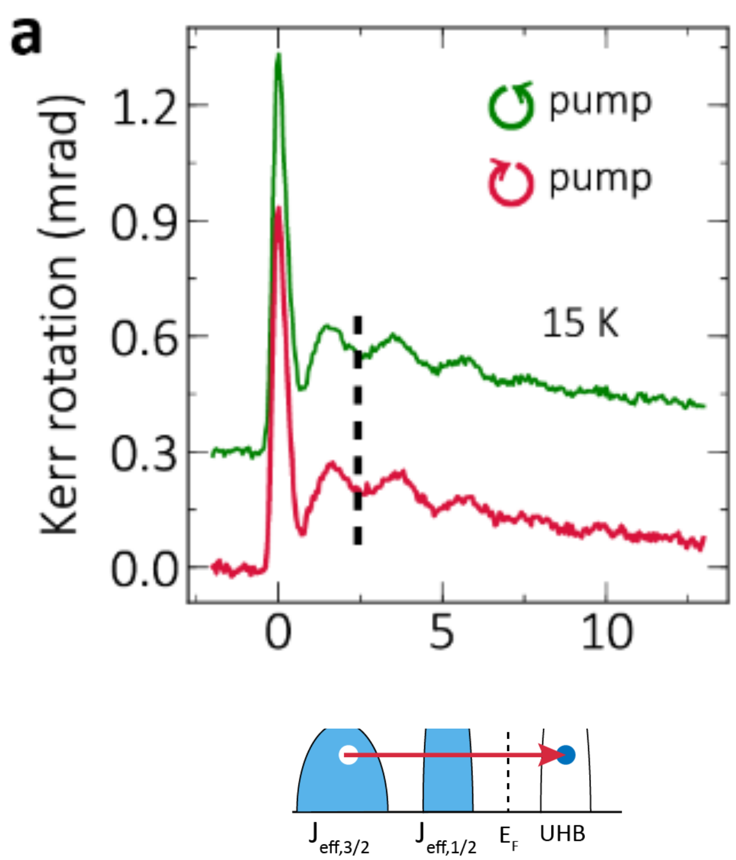
Kerr angle ~ magnetization



"Resonant" versus "non-resonant"

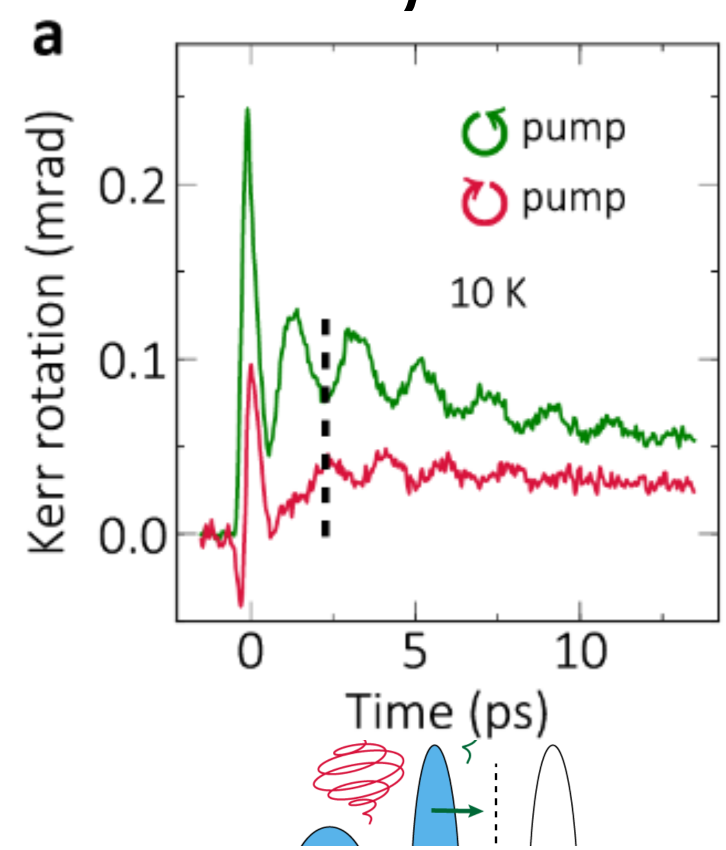
Phenomena

1.3 μm



θ_K =.029 mrad mJ^{-1} cm^2

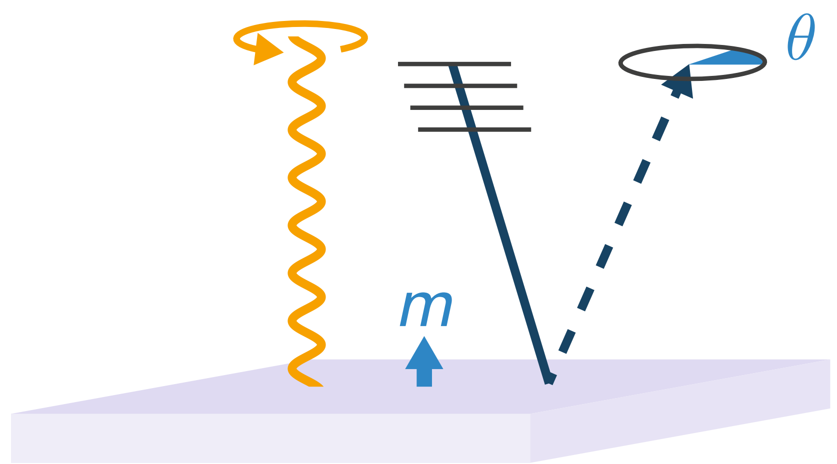
9 μm

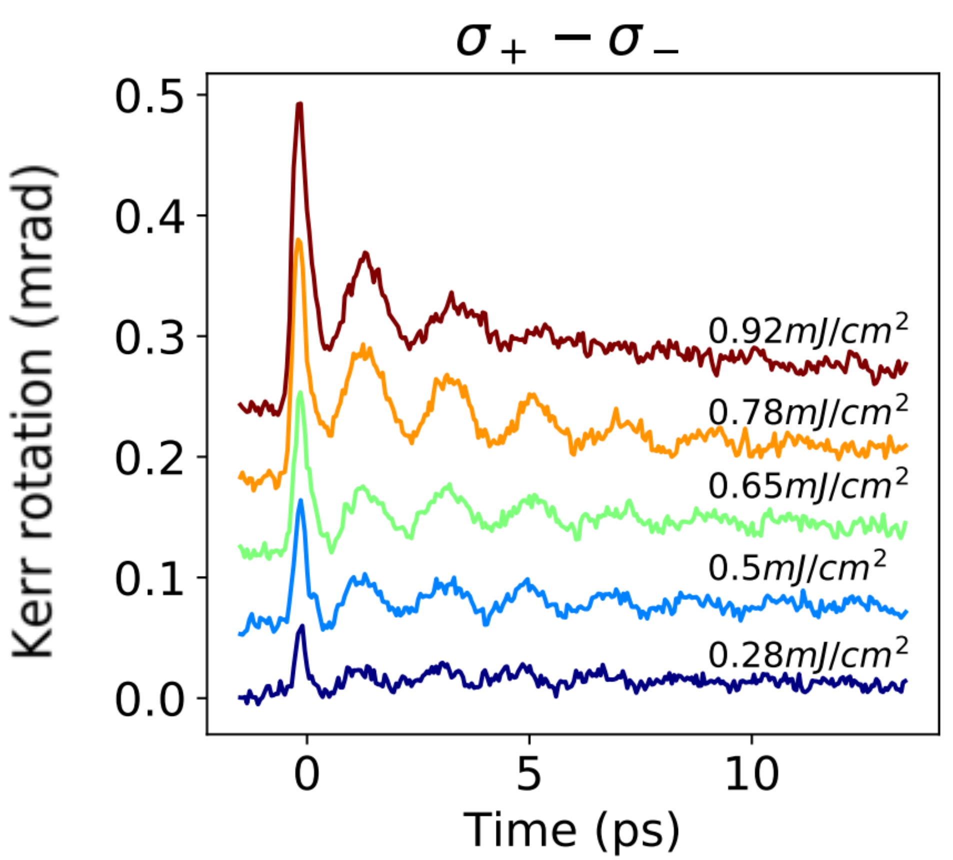


θ_K =.24 mrad mJ^{-1} cm^2

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

Phenomena

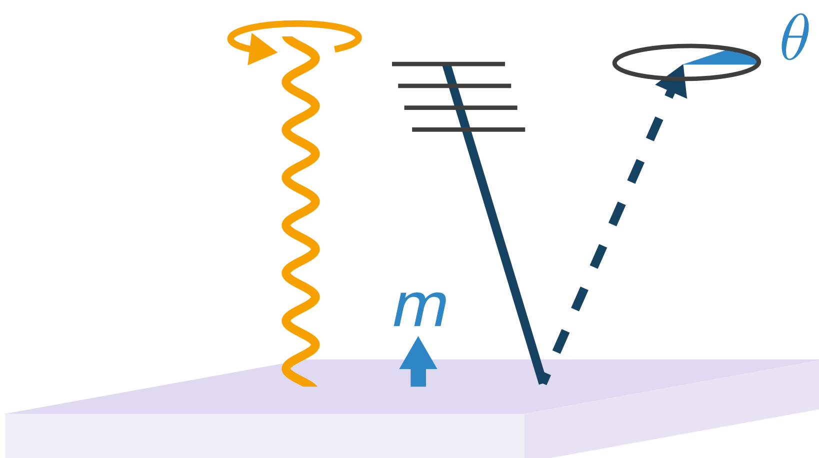


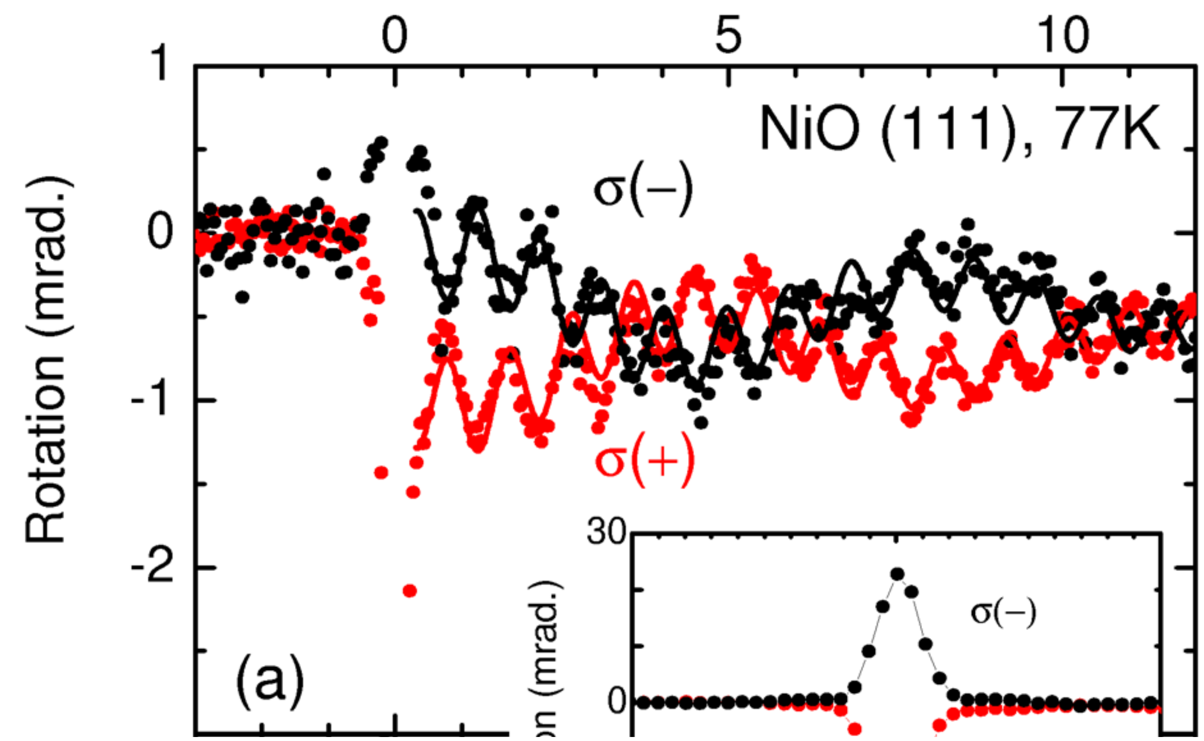


Questions

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

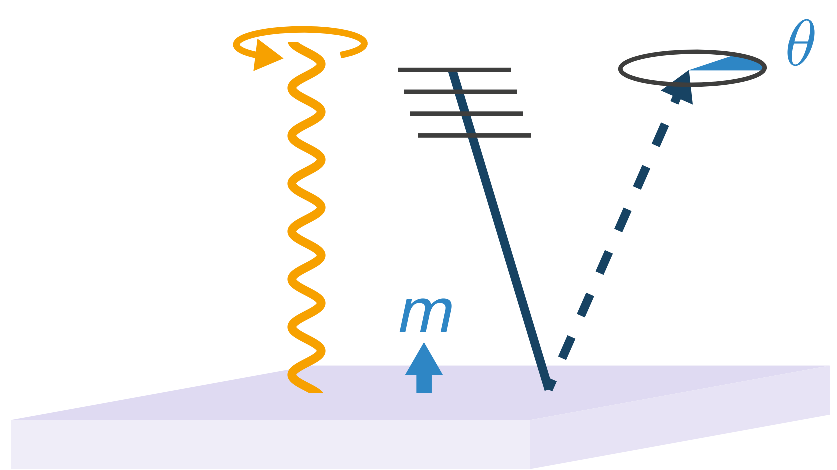
Phenomena

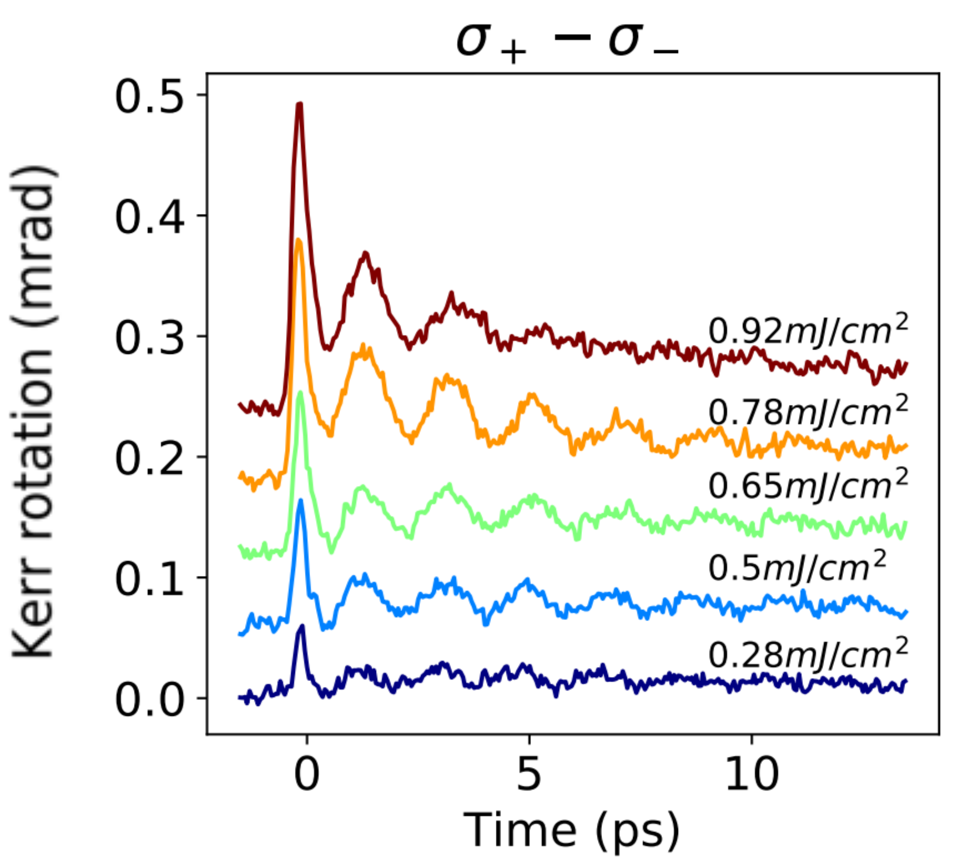




c.f. T. Satoh *et al*, 2010 - NiO

Phenomena



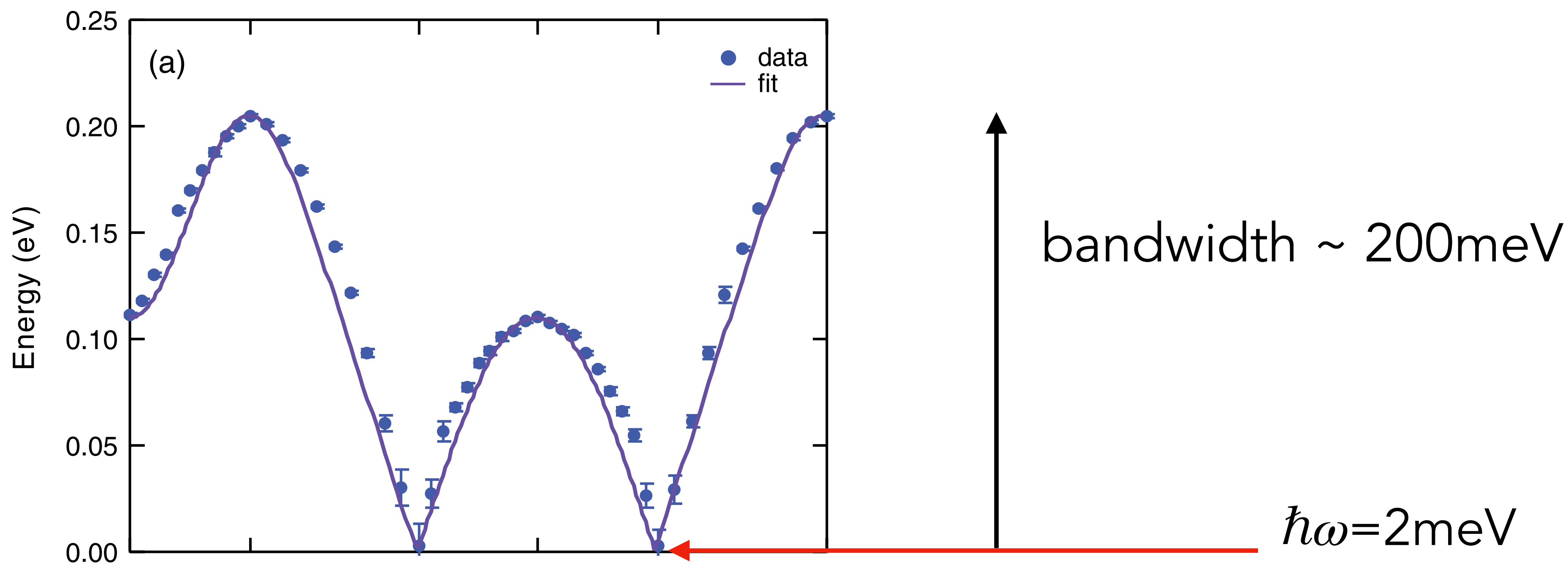


Questions

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

Magnons

- Gross features: square lattice Heisenberg antiferromagnet



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

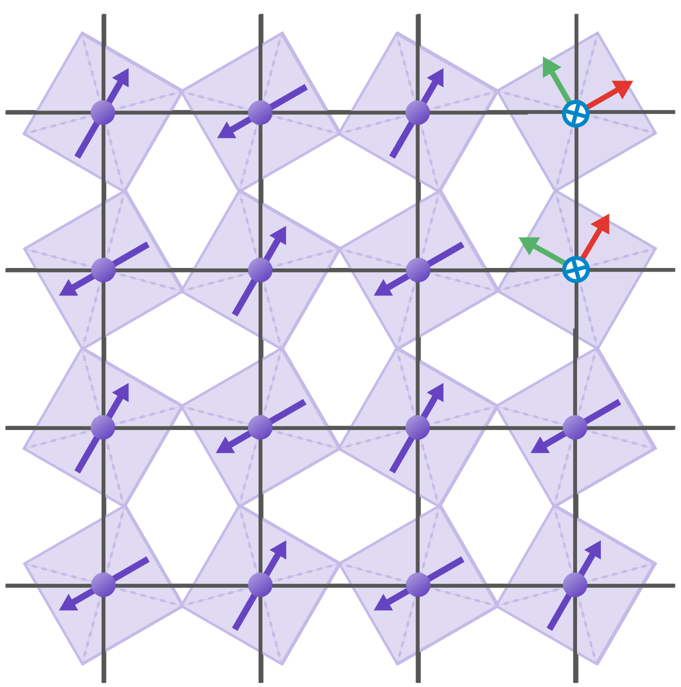
Magnon is a *very* low energy feature

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

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

Fits to RIXS give J~60meV and δ~.05

Small easy-plane anisotropy

Spin wave theory

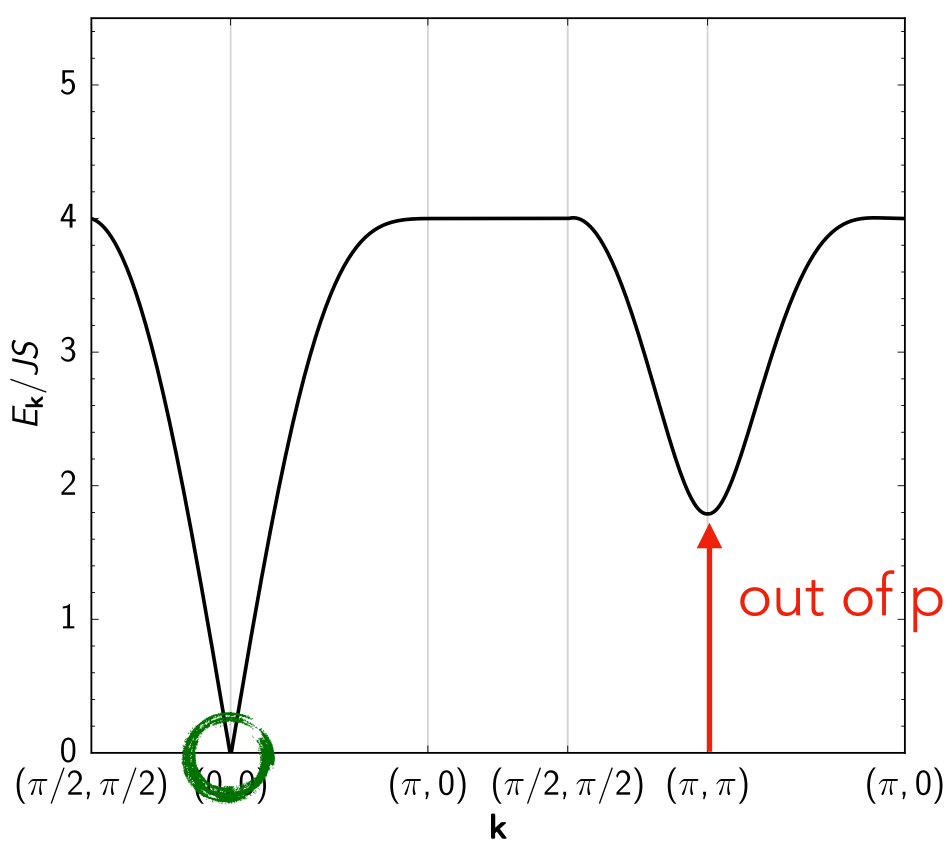
- Holstein-Primakoff

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

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

E_{cl} E_{sw} magnon interactions

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

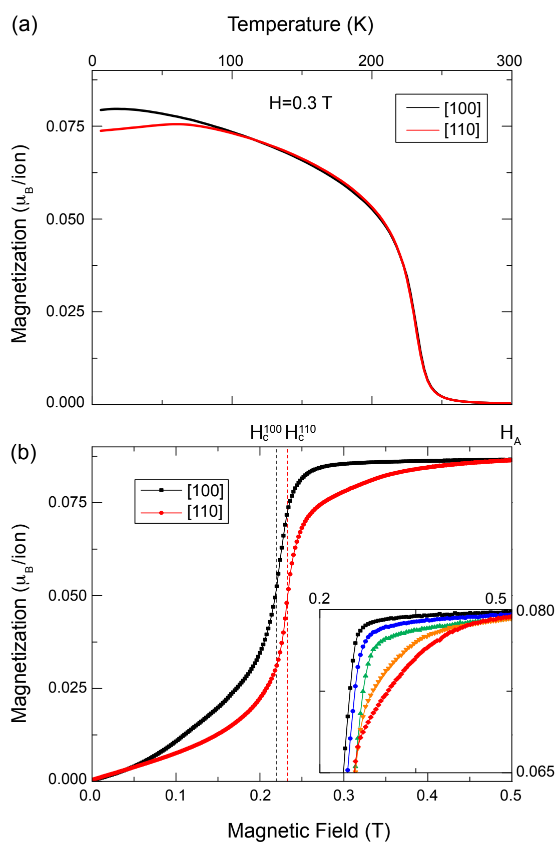


out of plane mode. Gap ~ 30meV! Too big to be the oscillating magnon

gapless Goldstone mode of in-plane rotations

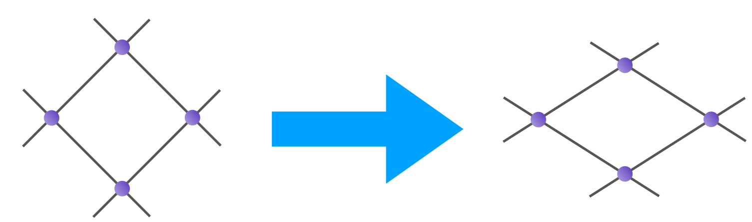
In-plane anisotropy

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



Porras *et al*, 2019

argue due to lattice distortion induced by spin order



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

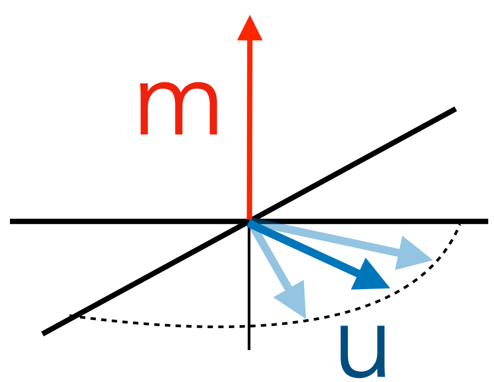
$\mathbf{\Gamma}$~6μeV (!) $\omega_0 \simeq 8S\sqrt{\Gamma J}$ ~2meV

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

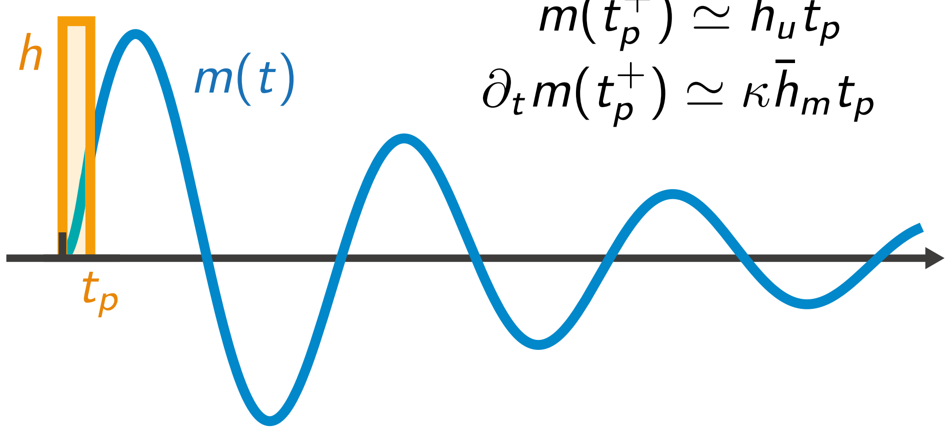
Pumping

- 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

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

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

- 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

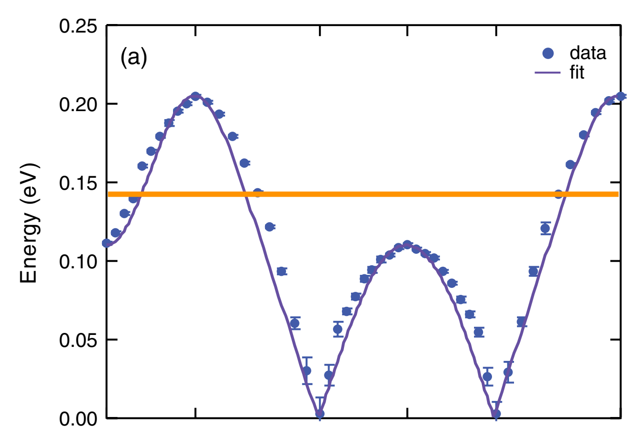
Bosons

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

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

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

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?

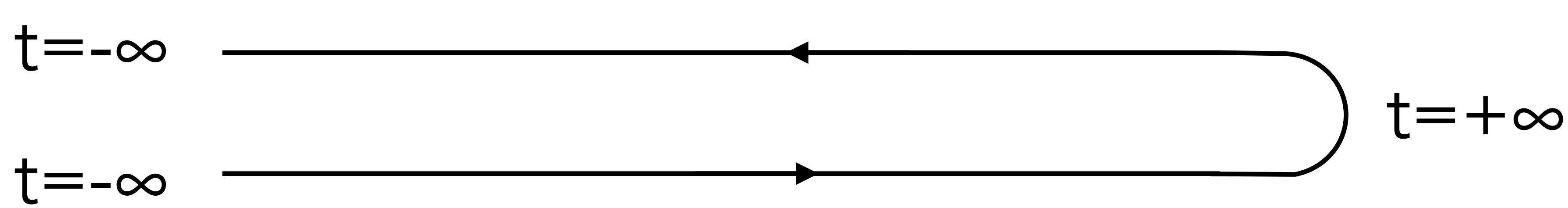
Formalism

- 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



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

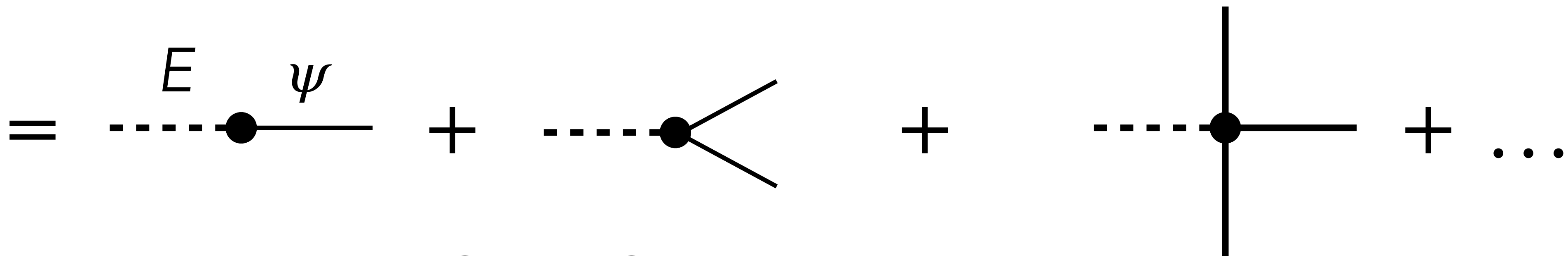
$$\mathcal{S} = \sum_{s=\pm} s \int \mathrm{d}t \left\{ \sum_i \bar{a}_{s,i} \mathrm{i}\partial_t a_{s,i} - \mathcal{H}[\{\bar{a}_{s,i}, a_{s,i}\}] \right\}$$

path integral over doubled fields

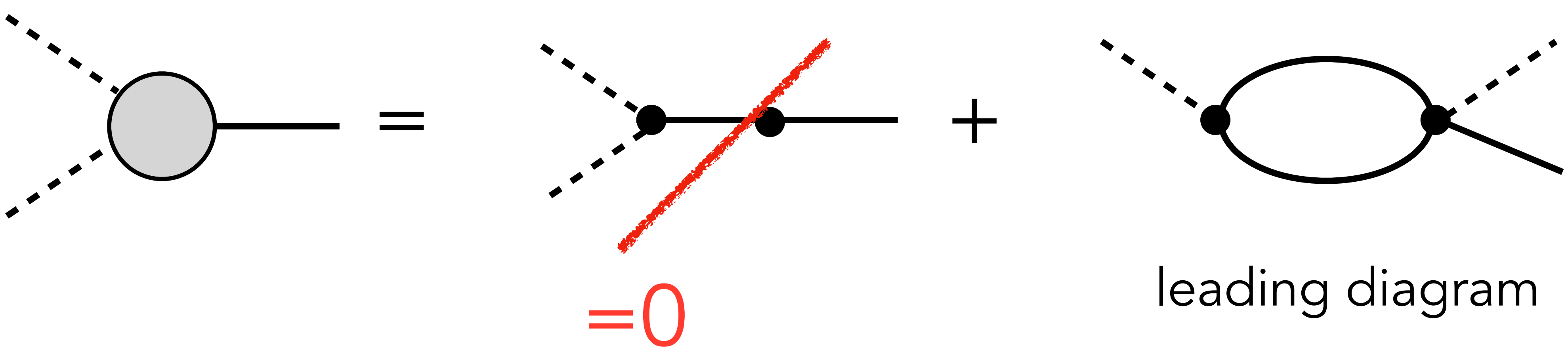
Formalism

- Non-equilibrium Keldysh path integral

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



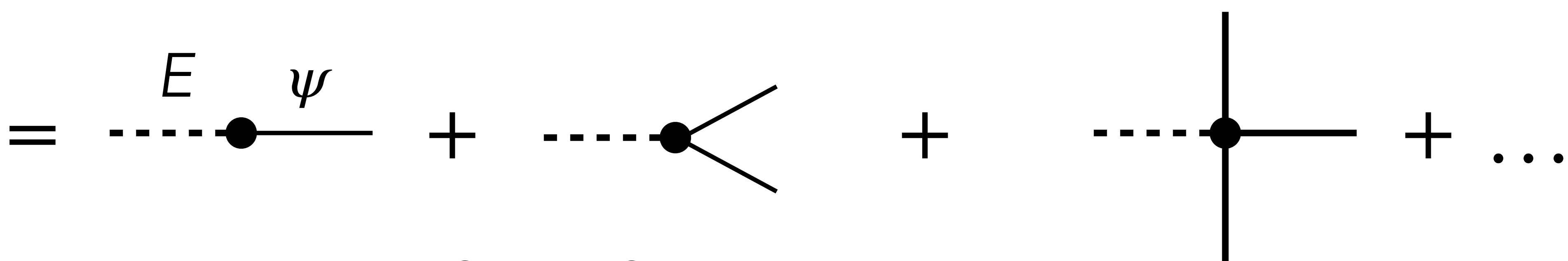
- Now integrate out "fast" fields: internal lines



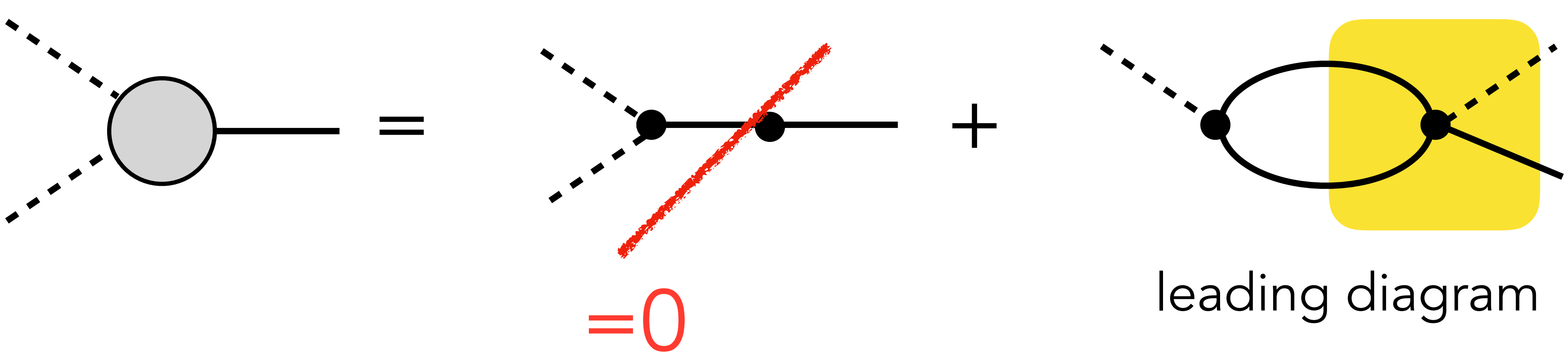
Formalism

- Non-equilibrium Keldysh path integral

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



- Now integrate out “fast” fields: internal lines



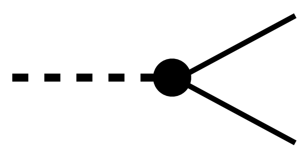
leading diagram

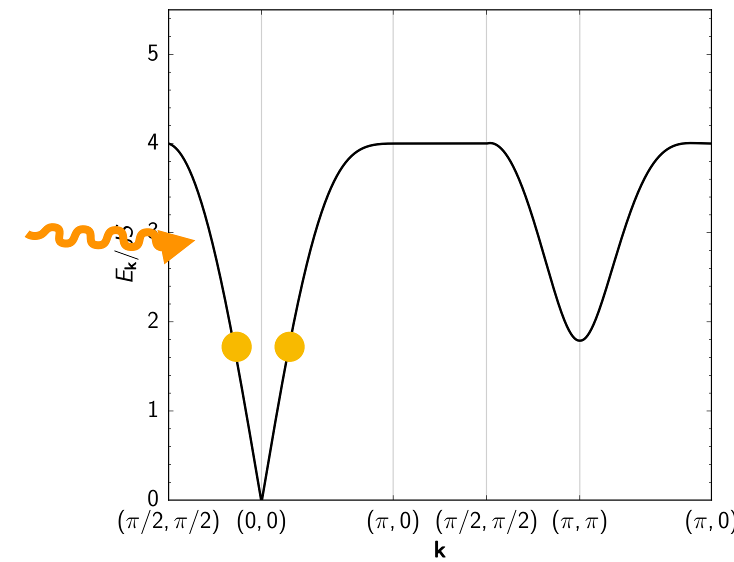
involves boson interactions!

Physical picture

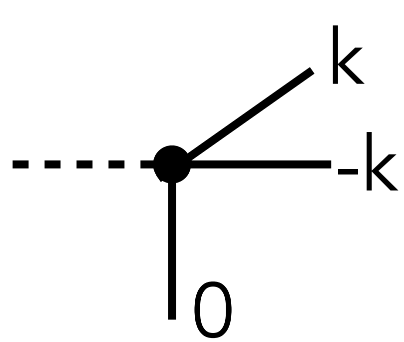
- Step 1:

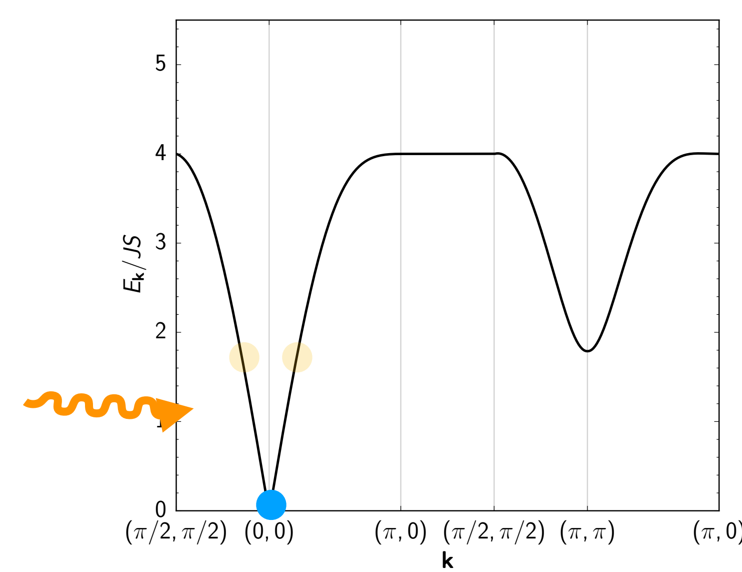
  photon generates coherent pairs





- 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

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

virtual

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

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

	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

- 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

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

virtual

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

- Structure of contributions to effective fields

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

total intensity

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

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

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

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

- 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
(ϕ=π/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

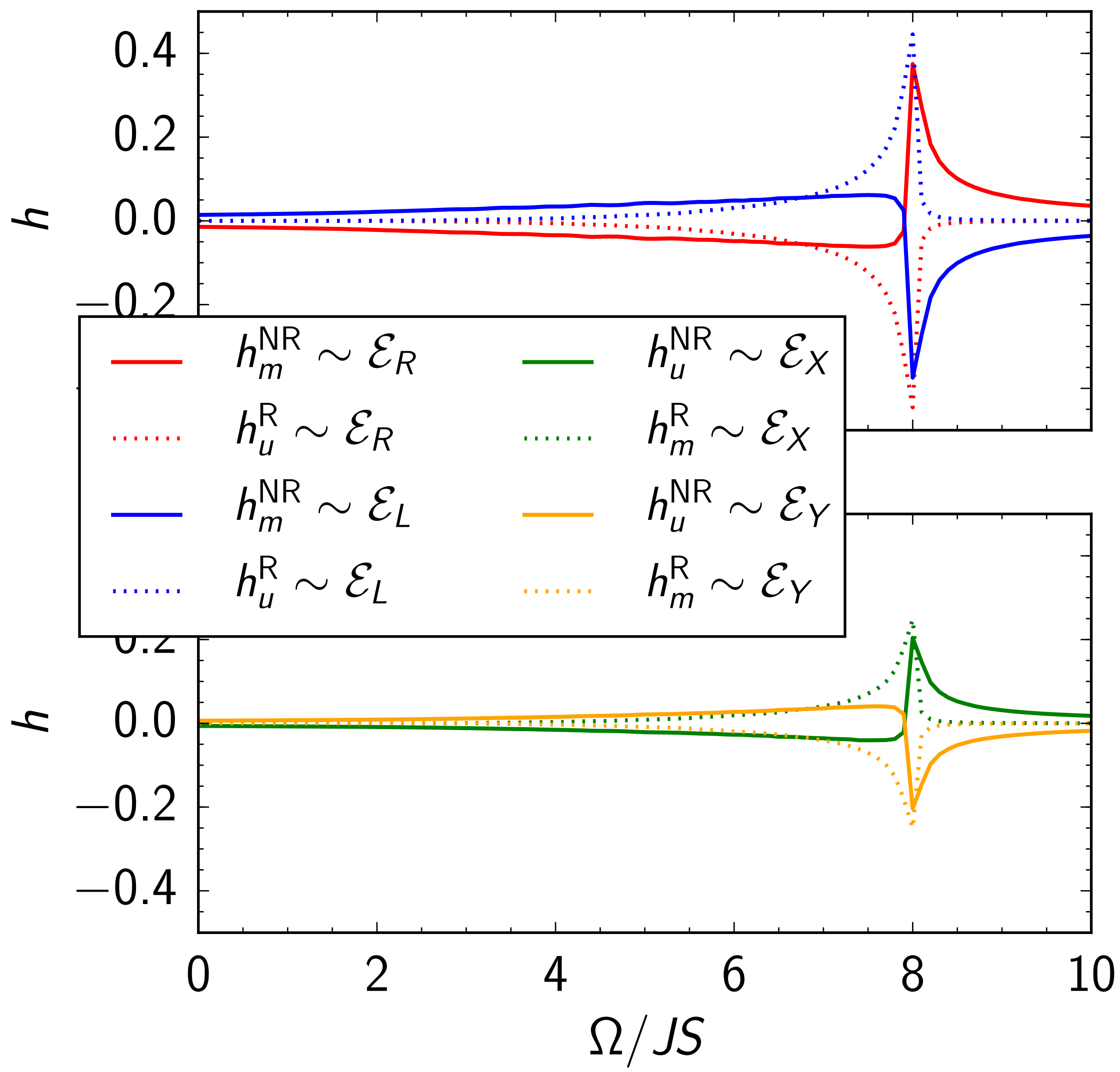
$$\begin{aligned}\mathcal{H}_{\text{eff}} = (\mathcal{E}_R\mathcal{E}_R^* - \mathcal{E}_L\mathcal{E}_L^*)J_zA \\ +\{(\mathcal{E}_R\mathcal{E}_R^* + \mathcal{E}_L\mathcal{E}_L^*)[J_z^2 - \tfrac{1}{3}J(J+1)] \\ - \mathcal{E}_L\mathcal{E}_R^* J_-^2 - \mathcal{E}_L^*\mathcal{E}_R J_+^2\}C,\end{aligned}$$

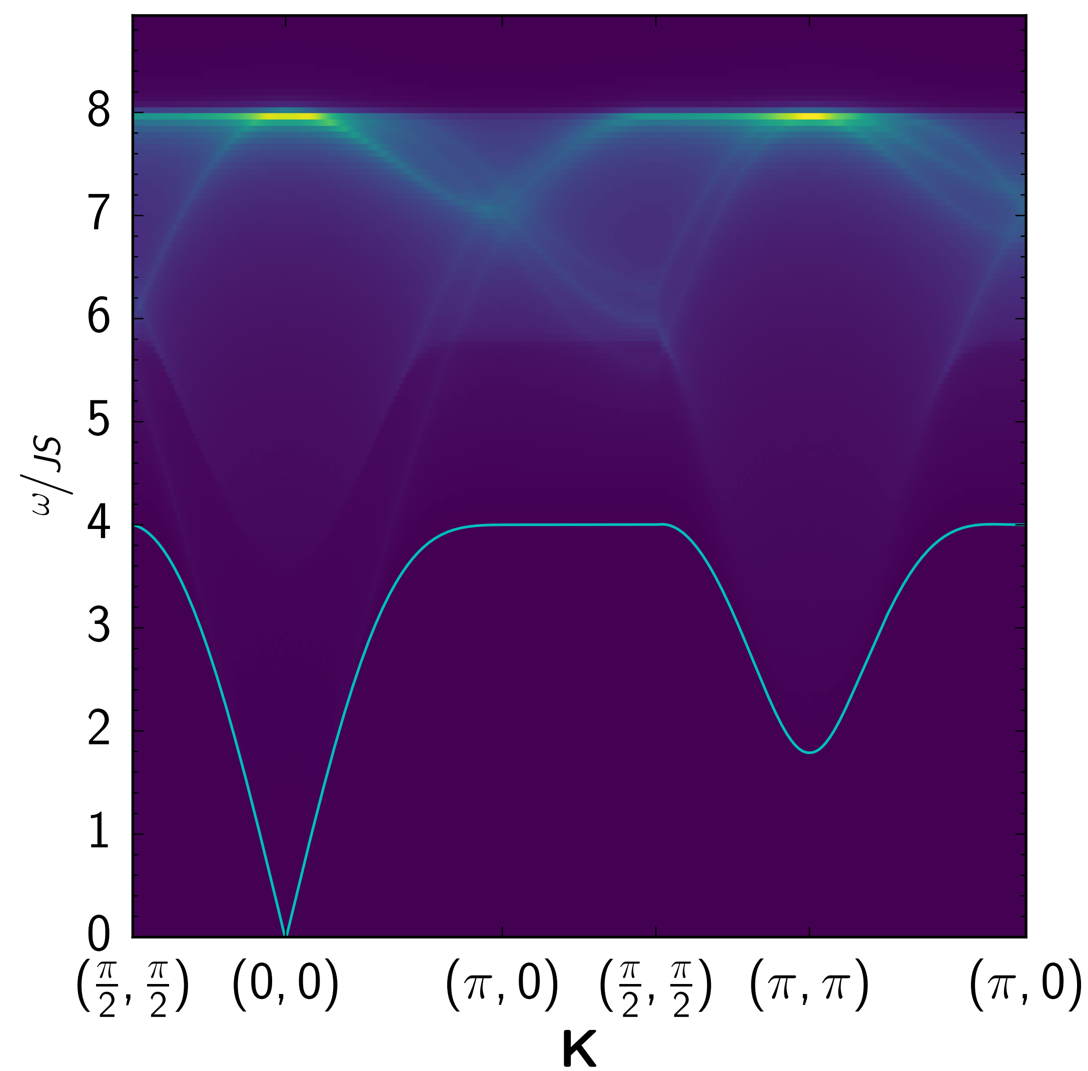
Purely virtual excitations

- 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

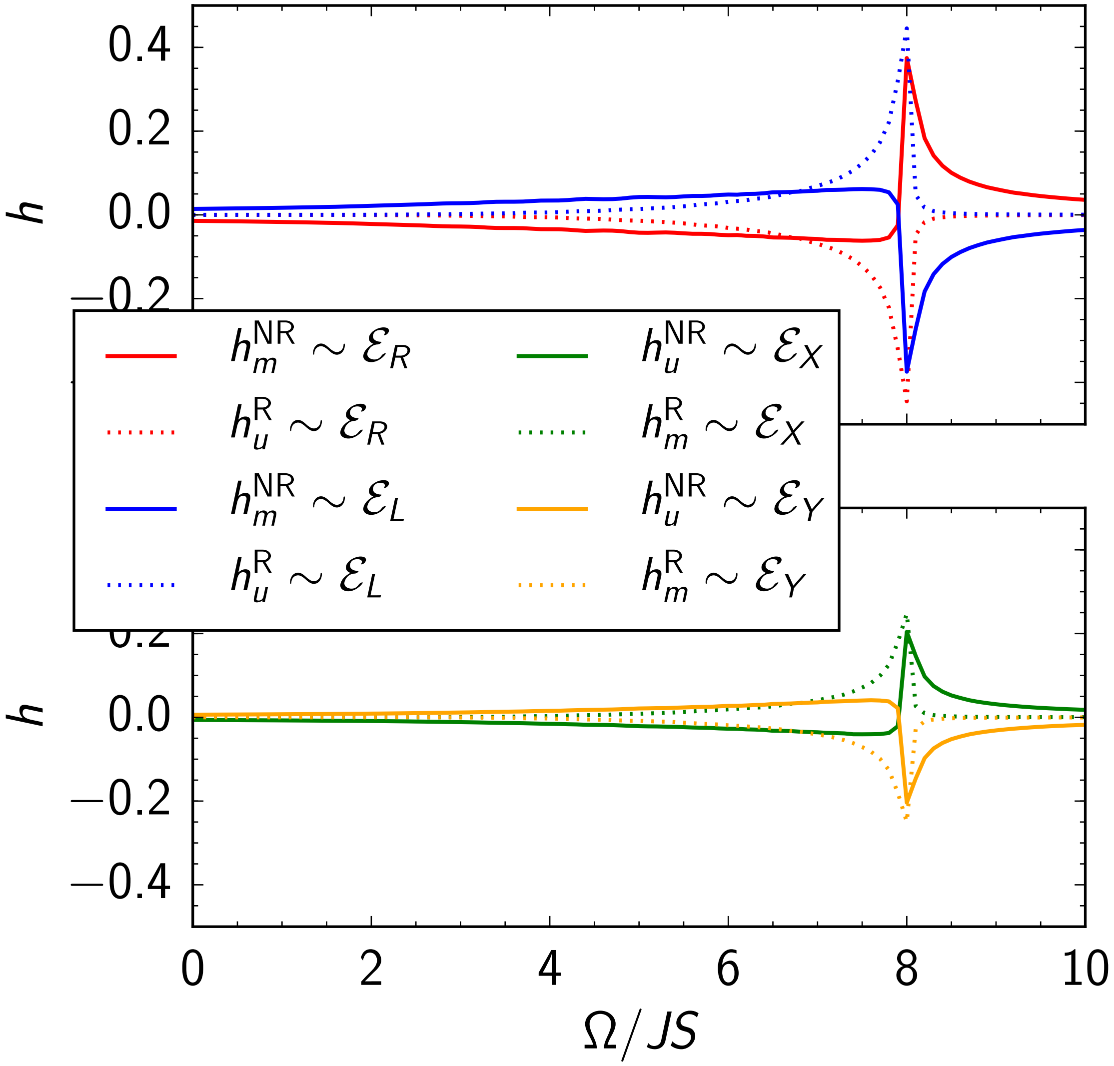




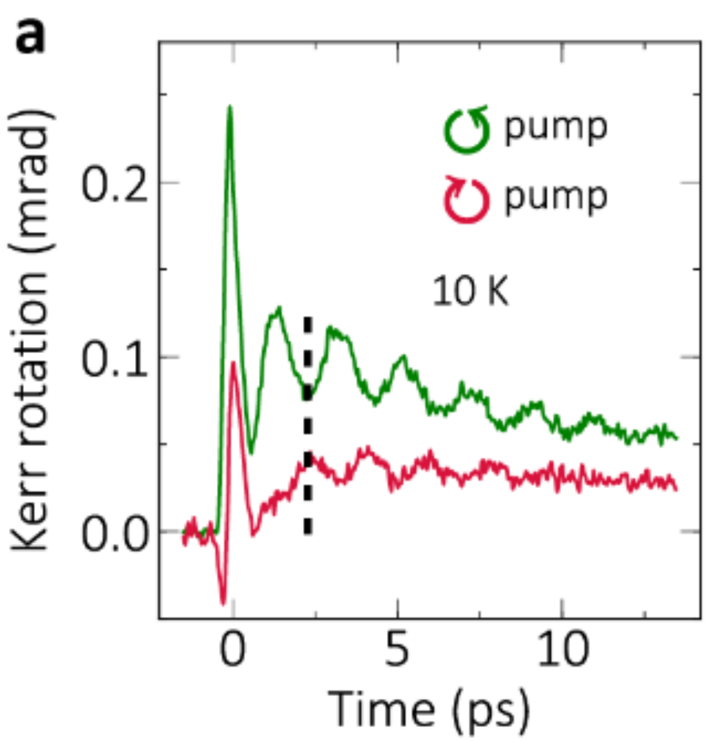
Significant enhancement still possible!

Results

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



Gufeng Zhang *et al*

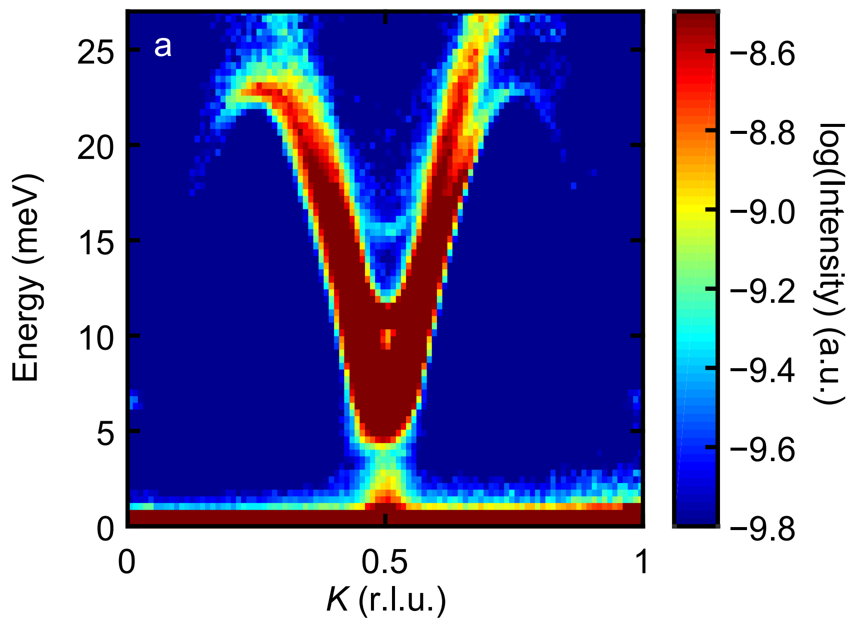


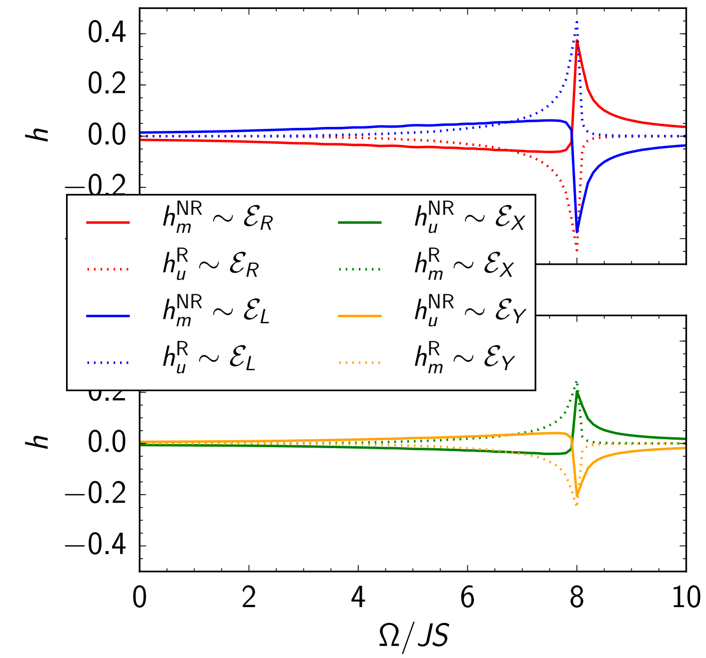
Dominant chiral intensity: magnetization of opposite sign for two circular polarizations

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?

Recap





- Semiclassical Heisenberg chain with weak anisotropy has many bound states described by bosons with delta-function attractions

R.Dally *et al*, Nat. Comm. (2018)

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

SFB 1143

arXiv:1905.01313