Lattice tuning and magnetic frustration in Sr_2IrO_4 and $Sr_3Ir_2O_7$ as seen by two-magnon Raman scattering

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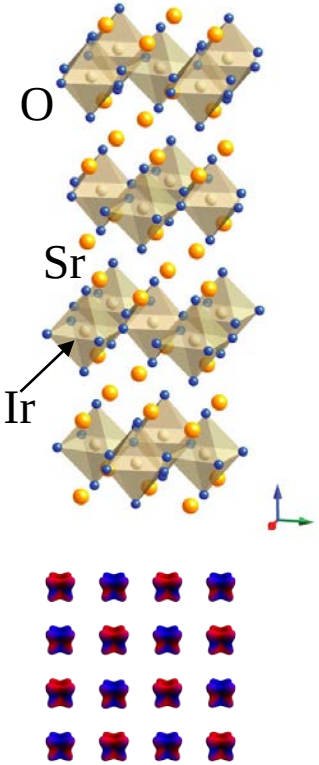
Yao Wang

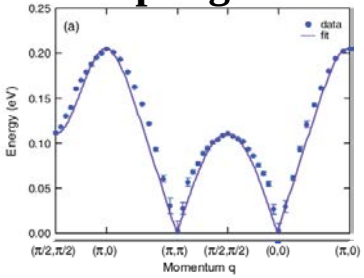
Thomas P. Devereaux

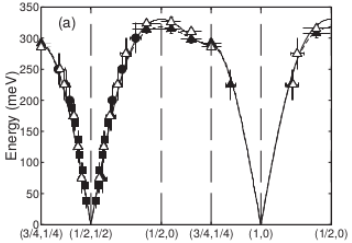
Why Iridates?

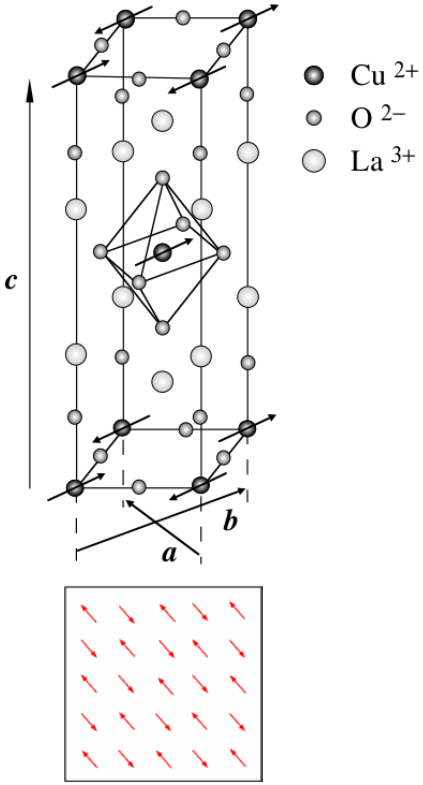


Iridates (Sr_2IrO_4)	Cuprates (La_2CuO_4)
Perovskite	
Quasi 2D square lattice	
Mott insulator AFM	

+ spin-orbit coupling

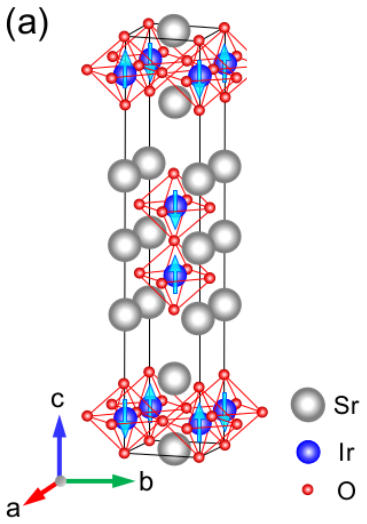






J. Kim *et al*, PRL **108**, 177003 (2012), J. Kim *et al*, Nat.Com **5**, 4453 (2014),
A.B.Harris *et al*, PRB **69**, 035107 (2004), R. Coldea *et al*, PRL **86,** 5377 (2001)

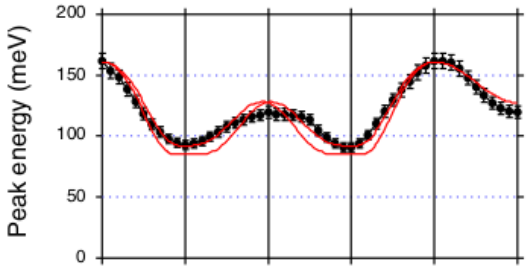
$Sr_3Ir_2O_7$



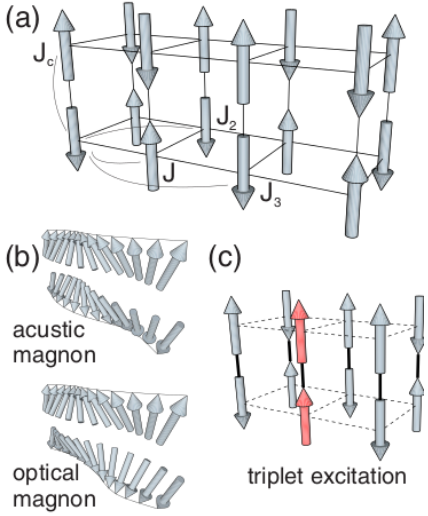
- Spin model

$$H_{ab} = \sum_{\langle i,j \rangle} [J \vec{S}_i \vec{S}_j + \Gamma S_i^z S_j^z + D(S_i^x S_j^y - S_i^y S_j^x)] + \sum_{\langle\langle i,j \rangle\rangle} J_2 \vec{S}_i \vec{S}_j + \sum_{\langle\langle\langle i,j \rangle\rangle\rangle} J_3 \vec{S}_i \vec{S}_j,$$

$$H_c = \sum_i [J_c \vec{S}_i \vec{S}_{i+z} + \Gamma_c S_i^z S_{i+z}^z + D_c(S_i^x S_{i+z}^y - S_i^y S_{i+z}^x) + \sum_{\langle i,j \rangle} J_{2c} \vec{S}_i \vec{S}_{j+z},$$



- Dimer model



J. Kim, *et. al.*, Phys. Rev. Lett. **109** (2012), M. Moretti Sala, *et. al.*, Phys. Rev. B **92**, 024405 (2015)

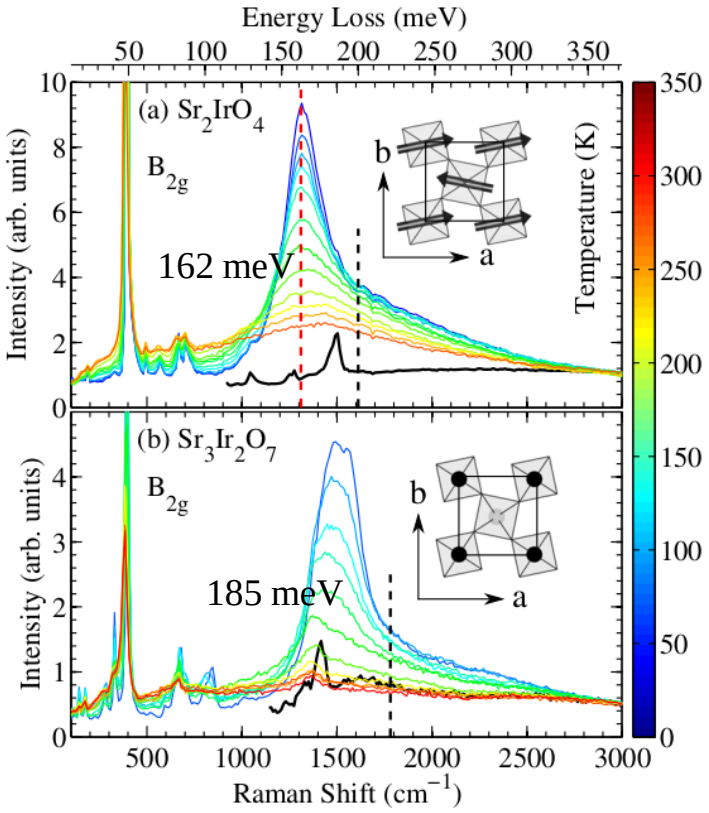
Experimental motivation: Two-magnon Raman scattering on iridates

- linear spin wave theory overestimates the energy of the two-magnon peak:

Sr_2IrO_4 : $3J_1 - 4J_2 - 4J_3 \approx 200$ meV

$Sr_3Ir_2O_7$: $3J - 4J' - 4J'' + J_C - 4J_{2C} = 221$ meV

- strong Fano asymmetries: pseudospin-lattice coupling?



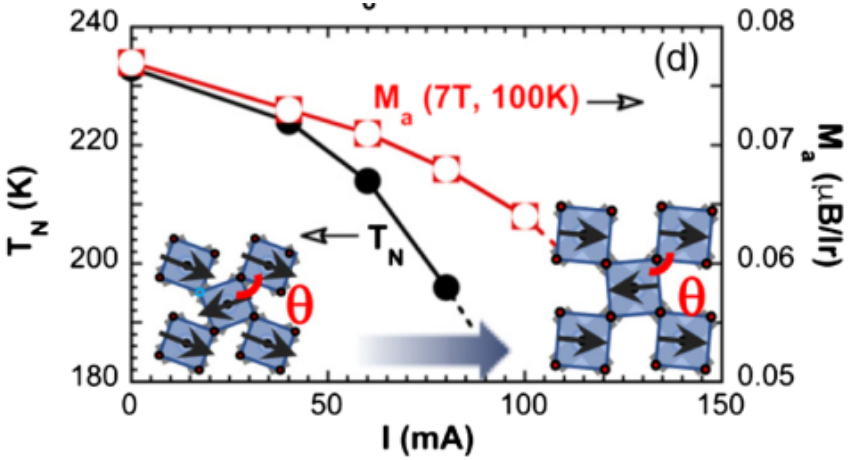
J. Kim, *et. al.*, Phys. Rev. Lett. **109** (2012), M. Moretti Sala, *et. al.*, Phys. Rev. B **92**, 024405 (2015), H. Gretarsson, *et. al.*, Phys. Rev. Lett. **116**, 136401 (2016)

Experimental motivation: non-quenched lattice dynamics

Sr_2IrO_4

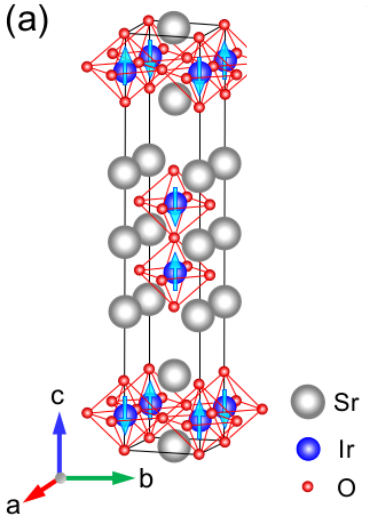
- Pseudospins rotating together with the octahedra under applied electric current

[G. Cao, *et. al.*, Phys. Rev. Lett. **120**, 017201 (2018)]



Experimental motivation: non-quenched lattice dynamics

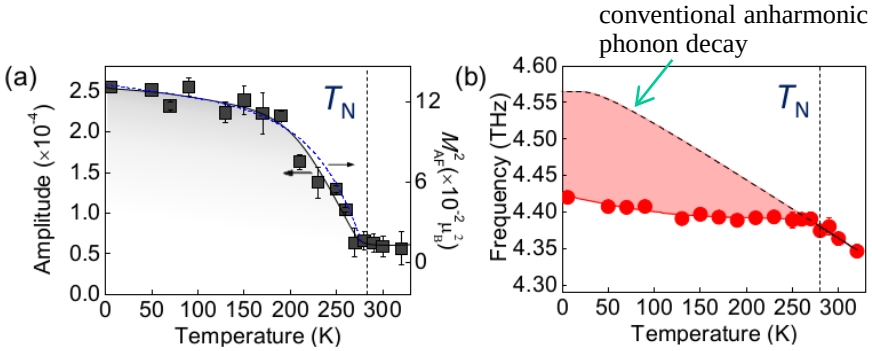
$Sr_3Ir_2O_7$

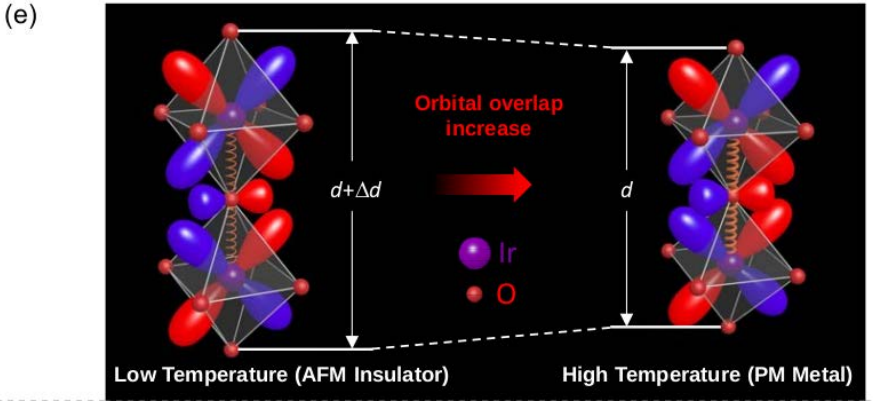


- Pseudospin-lattice coupling:

A_{1g} optical phonon anomaly in ultrafast optical spectroscopy, negative thermal expansion

==> Studying iridates under is very interesting!



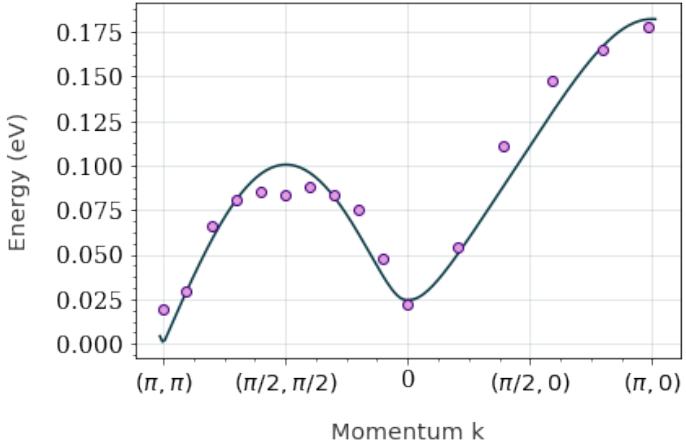


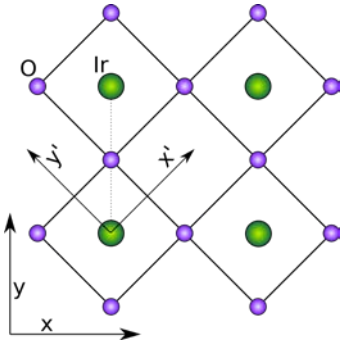
L. Hu, *et. al.*, Phys. Rev. **B** 99, 094307 (2019)

Sr_2IrO_4 two-magnon Raman spectra

- Spin model

$$H = \sum_{\langle i,j \rangle} \tilde{J}\left[S_i^x S_j^x + S_i^y S_j^y + (1-\Delta_\lambda) S_i^z S_j^z\right] + \sum_{\langle\langle i,j \rangle\rangle} J_2 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle\langle i,j \rangle\rangle\rangle} J_3 \vec{S}_i \cdot \vec{S}_j,$$





B_{2g}:

$$\hat{\mathbf{e}}_{\text{in}} = \hat{x}'$$
$$\hat{\mathbf{e}}_{\text{out}} = \hat{y}'.$$

- Exact diagonalization

$$I(\omega) \equiv \left\langle \Psi_0 \middle| \hat{\mathcal{O}}^\dagger \frac{1}{\omega + E_0 + i\epsilon - H} \hat{\mathcal{O}} \middle| \Psi_0 \right\rangle.$$

- Fleury-Loudon Raman scattering operator:

$$\hat{\mathcal{O}} = \sum_{ij} \eta_{ij} (\hat{\mathbf{e}}_{\text{in}} \cdot \hat{\mathbf{d}}_{ij})(\hat{\mathbf{e}}_{\text{out}} \cdot \hat{\mathbf{d}}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j,$$

D. Pincini, *et. al.*, Phys. Rev. B **96**, 075162 (2017)

Renormalizing exchange interaction constants

PHYSICAL REVIEW B VOLUME 40, NUMBER 10 1 OCTOBER 1989

Simple model for the linewidth of the two-magnon Raman feature observed in high-T_c superconductors

W. H. Weber* and G. W. Ford

[In linear spin wave]

if the higher-order terms are treated within a mean-field approximation, the same quadratic Hamiltonian results with the same dispersion curve for the magnons, albeit with a renormalized value $\tilde{J} = \eta J$ for the exchange interaction constant:[22]

RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 39, NUMBER 13 1 MAY 1989

Thermodynamic parameters of the $T=0$, spin-$\frac{1}{2}$ square-lattice Heisenberg antiferromagnet

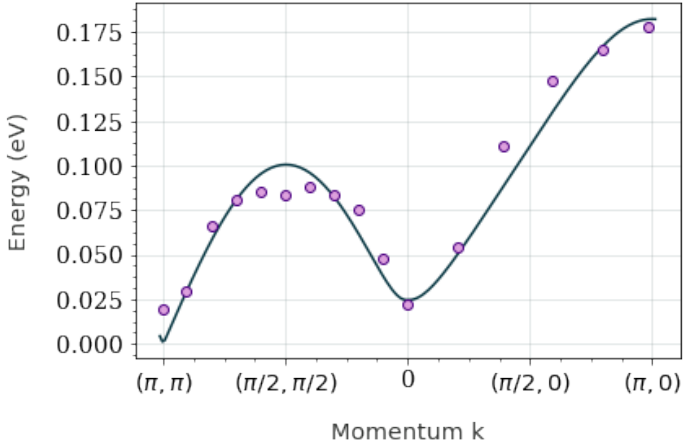
Rajiv R. P. Singh

$$Z_{c_s} = (Z_{\rho_s}/Z_\chi)^{1/2} = 1.18 \pm 0.02 .$$

Sr_2IrO_4 two-magnon Raman spectra

- Spin model

$$\mathcal{H} = \frac{J_1}{Z_c}\sum_{\langle i,j\rangle}\left(\tilde{\mathbf{S}}_i^x\tilde{\mathbf{S}}_j^x + \tilde{\mathbf{S}}_i^y\tilde{\mathbf{S}}_j^y + (1-\Delta_\lambda)\tilde{\mathbf{S}}_i^z\tilde{\mathbf{S}}_j^z\right) + \frac{J_2}{Z_c}\sum_{\langle i,j\rangle}\tilde{\mathbf{S}}_i\cdot\tilde{\mathbf{S}}_j + \frac{J_3}{Z_c}\sum_{\langle i,j\rangle}\tilde{\mathbf{S}}_i\cdot\tilde{\mathbf{S}}_j$$



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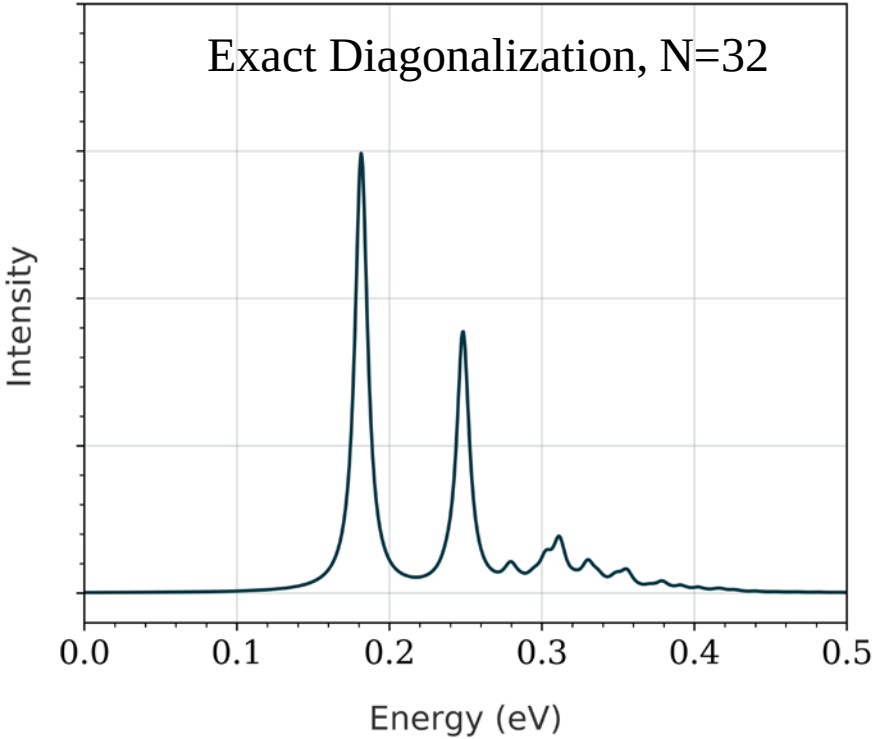
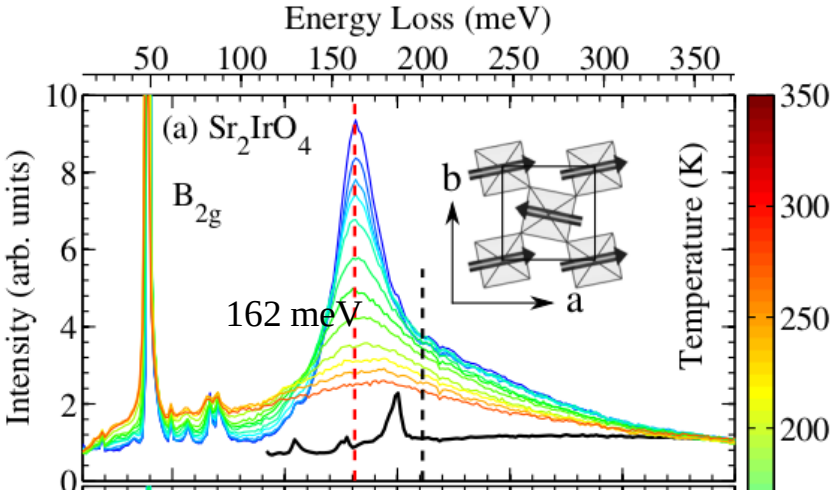
Sr_2IrO_4 two-magnon Raman spectra



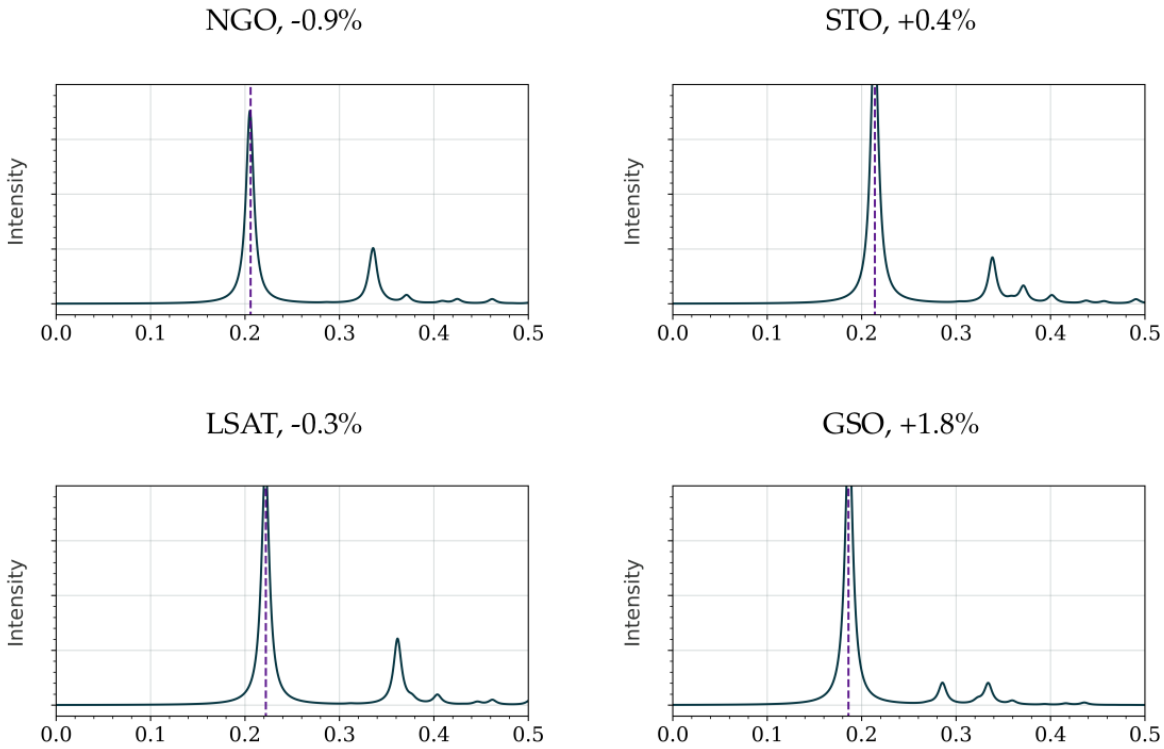
Raman Scattering

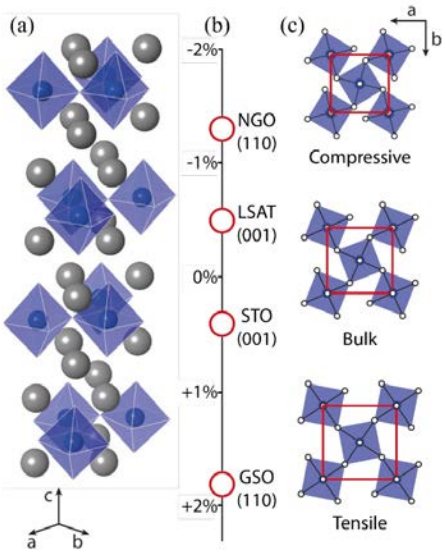


H. Gretarsson, *et. al.*, Phys. Rev. Lett. **116**, 136401 (2016)

Sr_2IrO_4 two-magnon Raman spectra (strained)

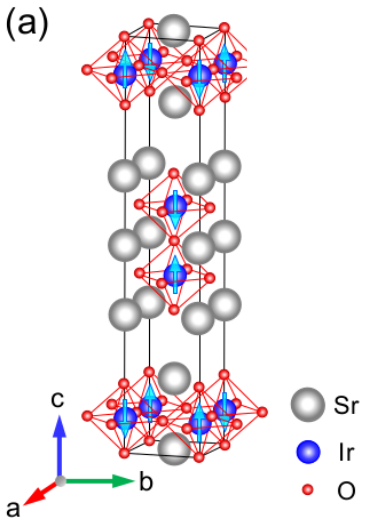
Exact diagonalization, N=16 cluster





A. Seo, , *et. al.* unpublished; E. Pärschke, *et. al.* unpublished; E. Paris, Y. Tseng, E. Pärschke, *et. al.* unpublished (2019)

$Sr_3Ir_2O_7$



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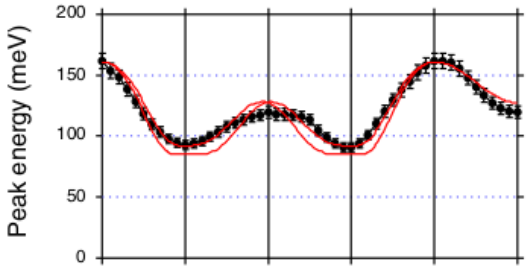
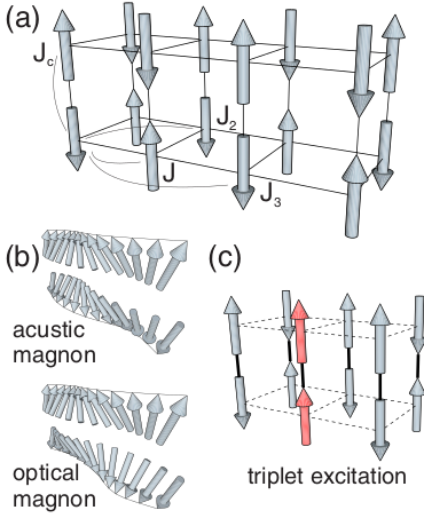
$$H_c = \sum_i [J_c \vec{S}_i \vec{S}_{i+z} + \Gamma_c S_i^z S_{i+z}^z + D_c(S_i^x S_{i+z}^y - S_i^y S_{i+z}^x) + \sum_{\langle i,j \rangle} J_{2c} \vec{S}_i \vec{S}_{j+z},$$



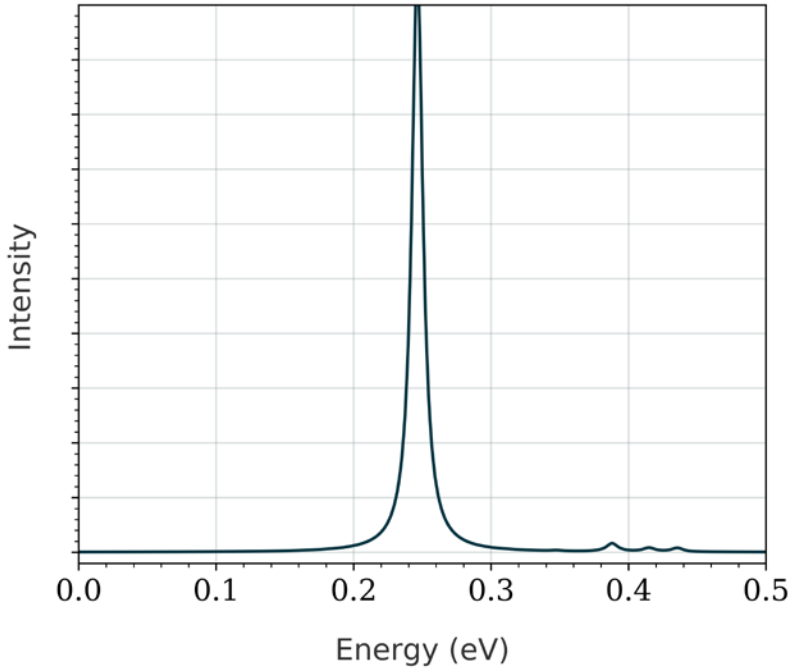
- Dimer model



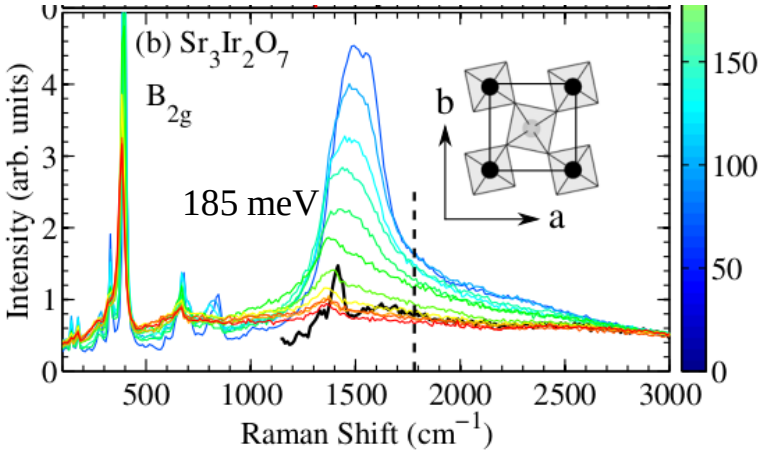
J. Kim, *et. al.*, Phys. Rev. Lett. **109** (2012), M. Moretti Sala, *et. al.*, Phys. Rev. B **92**, 024405 (2015)

$Sr_3Ir_2O_7$ two-magnon Raman spectra

Exact Diagonalization, N=16



Raman Scattering



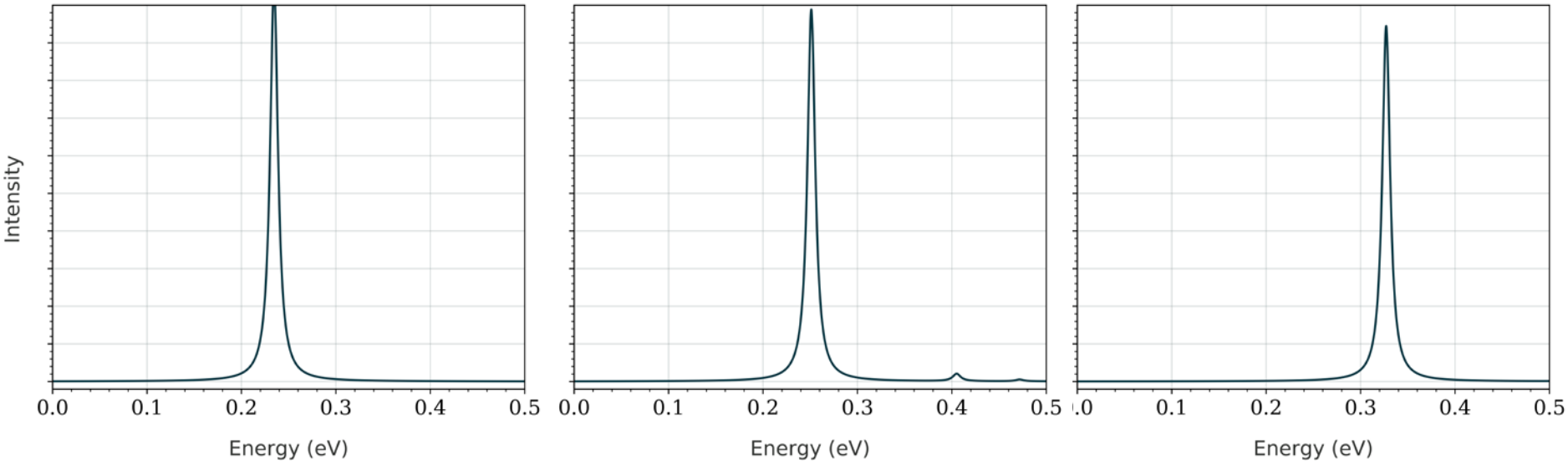
H. Gretarsson, *et. al.*, Phys. Rev. Lett. **116**, 136401 (2016)

Exact diagonalization on $Sr_3Ir_2O_7$: “Manually” dimerized

J_1-J_2 model
(J_1=0.093, J_2 =0.119)

+ J_{1c}=0.05 meV

+ J_{1c}=0.15 meV



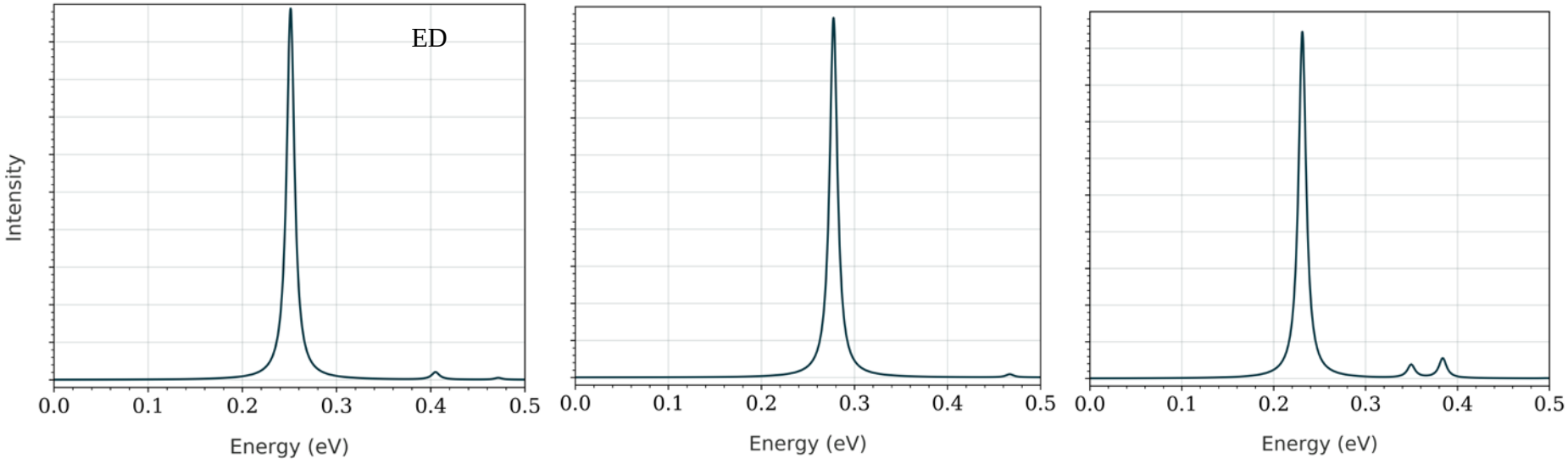
$Sr_3Ir_2O_7$: first VS second neighbor interlayer interaction

J_1-J_2 model (J_1=0.093, J_2 =0.119)

+ J_{1c}=0.05 meV

+ J_{2c}=0.02 meV

+ J_{1c}=0.05, J_{2c}=0.02 meV

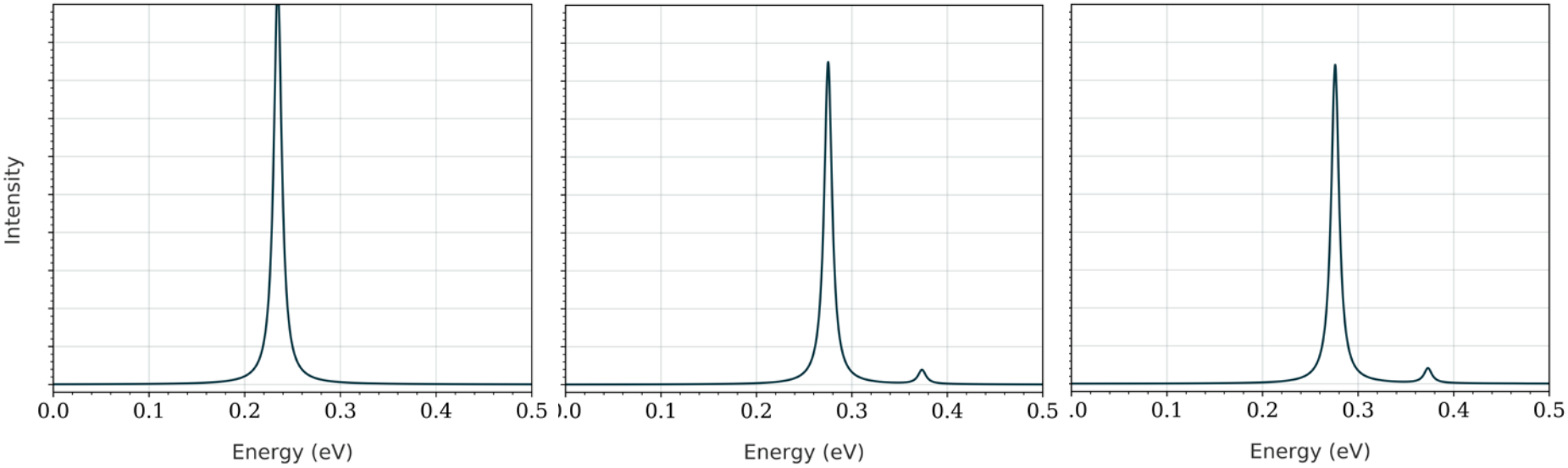


Exact diagonalization on $Sr_3Ir_2O_7$: “Manually” dimerized

- J_1-J_2 model
(J_1=0.093, J_2 =0.119)

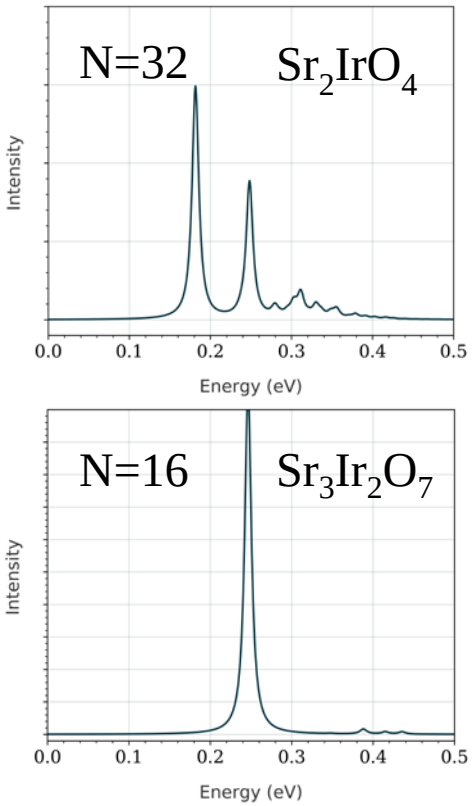
+ D=0.05 meV

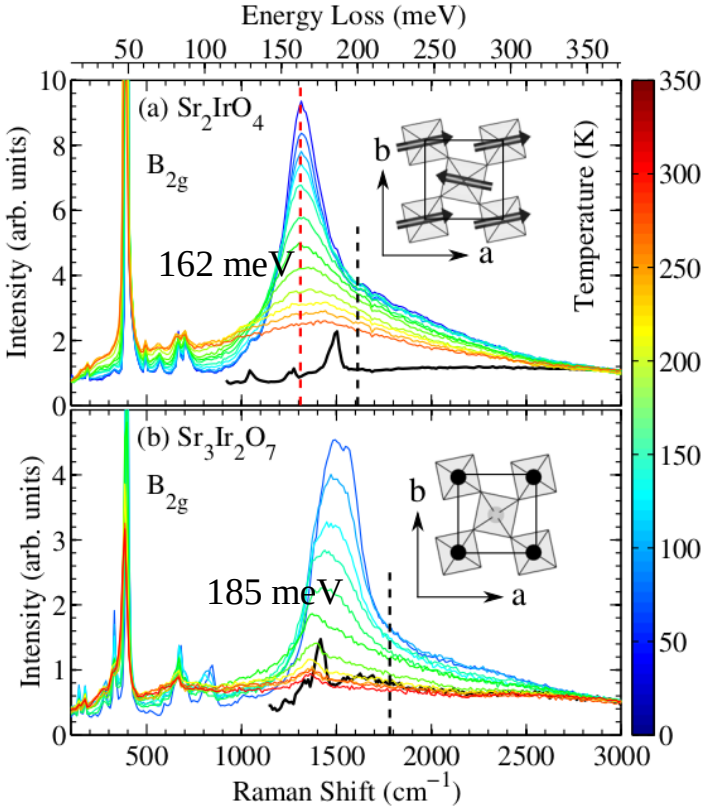
+ D=0.05 meV
D_c=0.02 meV



Sr_2IrO_4 two-magnon Raman spectra

Exact Diagonalization





H. Gretarsson, *et. al.*, Phys. Rev. Lett. **116**, 136401 (2016)

Conclusions and outlook

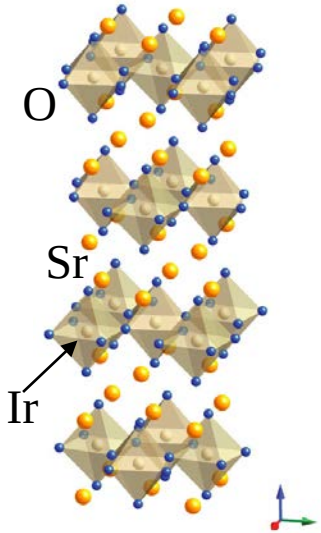
- For Sr_2IrO_4, the energy of the two-magnon Raman peak is lowered due to magnon-magnon interaction and quantum fluctuations
- One can reproduce the changes in two-magnon Raman peak in strained Sr_2IrO_4
- For $Sr_3Ir_2O_7$, two-magnon Raman peak position is governed by intrinsic interplay of the magnetic couplings - understanding interplay of those is a challenge
- Proper estimation of Z_c for models beyond J_1 Heisenberg is needed

Thank you for your attention!

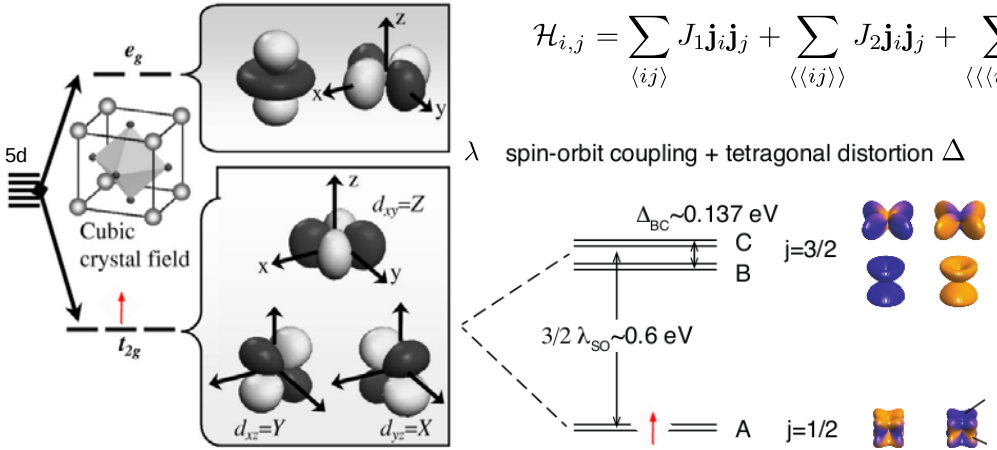
- For Sr_2IrO_4, the energy of the two-magnon Raman peak is lowered due to magnon-magnon interaction and quantum fluctuations
- One can reproduce the changes in two-magnon Raman peak in strained Sr_2IrO_4
- For $Sr_3Ir_2O_7$, two-magnon Raman peak position is governed by intrinsic interplay of the magnetic couplings - understanding interplay of those is a challenge
- Proper estimation of Z_c for models beyond J_1 Heisenberg is needed

All you need to know about Sr_2IrO_4

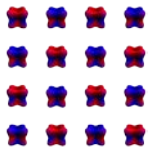
Structure



Single Ir^{4+} ion:
1 hole in 5*d* (t_{2g}) orbitals



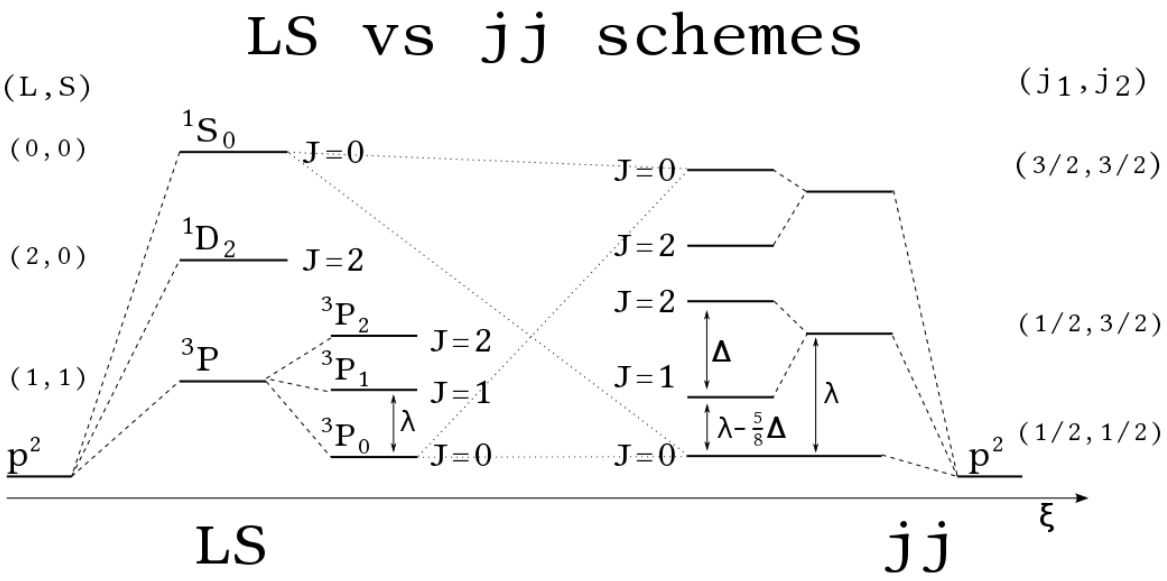
Ground state: j=1/2 antiferromagnet
Heisenberg model



$$\mathcal{H}_{i,j} = \sum_{\langle ij \rangle} J_1 \mathbf{j}_i \mathbf{j}_j + \sum_{\langle\langle ij \rangle\rangle} J_2 \mathbf{j}_i \mathbf{j}_j + \sum_{\langle\langle\langle ij \rangle\rangle\rangle} J_3 \mathbf{j}_i \mathbf{j}_j$$

J. Kim *et al*, PRL **108**, 177003 (2012) & A.B.Harris *et al*, PRB **69**, 035107 (2004)

Spin-orbit coupling in Ir^{3+}: weak and strong limit



(J = L + S) **(J = j_1 + j_2)**

[arXiv:1802.04158]: E. Pärschke and R. Ray (2018)