Raman response via 4-spinon continuum in spin-1/2 quantum liquid $Ba_4Ir_3O_{10}$

arXiv:2110.15916

Sami Hakani[1], Aaron Sokolik[2], Susmita Roy[2],
Nicholas Pellatz[2], Hengdi Zhao[2], Gang Cao[2], Itamar Kimchi[1],
Dmitry Reznik[2]

[1]Georgia Institute of Technology
[2]University of Colorado Boulder

APS March Meeting 2022, Chicago IL

References

[1] Brenig, W. (1997). Raman scattering from frustrated quantum spin chains. *Physical Review B*, *56*(5), 2551–2555. doi:10.1103/PhysRevB.56.2551

[2] Cao, G., Zheng, H., Zhao, H., Ni, Y., Pocs, C. A., Zhang, Y., … Kimchi, I. (2020). Quantum liquid from strange frustration in the trimer magnet $Ba_4Ir_3O_{10}$. *Npj Quantum Materials*, *5*(1), 1–8. doi:10.1038/s41535-020-0232-6

[3] Sato, M., Katsura, H., & Nagaosa, N. (2012). Theory of Raman Scattering in One-Dimensional Quantum Spin-1/2 Antiferromagnets. *Physical Review Letters*, *108*(23), 237401. doi:10.1103/PhysRevLett.108.237401

[4] Fleury, P. A., & Loudon, R. (1968). Scattering of Light by One- and Two-Magnon Excitations. *Physical Review*, *166*(2), 514–530. doi:10.1103/PhysRev.166.514

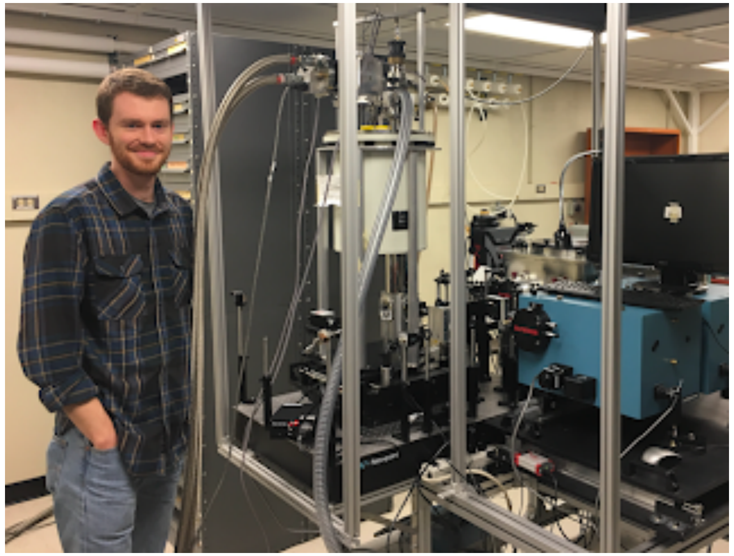
[5] Sokolik, A., Hakani, S., Roy, S., Pellatz, N., Zhao, H., Cao, G., … Reznik, D. (2021). Spinons and damped phonons in spin-1/2 quantum-liquid $Ba_4Ir_3O_{10}$ observed by Raman scattering. *arXiv:2110. 15916 [cond-mat]*. Retrieved from http://arxiv.org/abs/2110.15916

[6] Wulferding, D., Choi, Y., Lee, W., & Choi, K.-Y. (2019). Raman spectroscopic diagnostic of quantum spin liquids. *Journal of Physics: Condensed Matter*, *32*(4), 043001. doi:10.1088/1361-648X/ab45c4

[7] Gretarsson, H., Sung, N. H., Höppner, M., Kim, B. J., Keimer, B., & Le Tacon, M. (2016). Two-Magnon Raman Scattering and Pseudospin-Lattice Interactions in Sr_2IrO_4 and $Sr_3Ir_2O_7$. *Phys. Rev. Lett.*, *116*, 136401. doi:10.1103/PhysRevLett.116.136401

Collaborators on $Ba_4Ir_3O_{10}$

Aaron Sokolik (CU)



Nicholas Pellatz (CU)

Dr. Susmita Roy (CU)

Hengdi Zhao (CU)

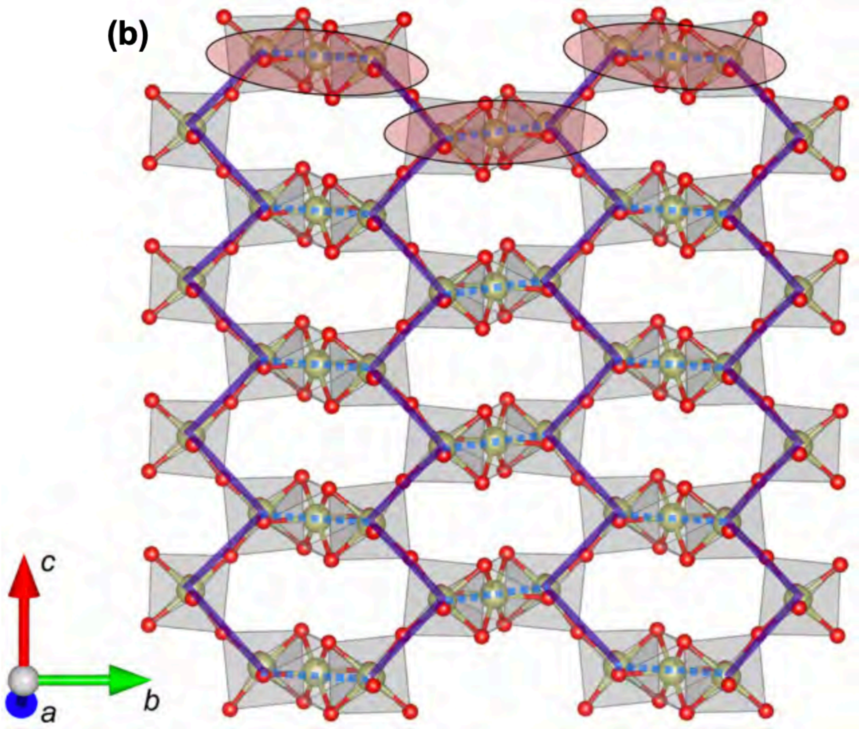
Prof. Itamar Kimchi (GT)

Prof. Dmitry Reznik (CU)

Prof. Gang Cao (CU)

Outline

- Preliminaries
- Raman Spectroscopy
- Spectrum of $Ba_4Ir_3O_{10}$
- Theoretical model
- Spinon mean field theory
- Results of computation



Cao et al. [2]

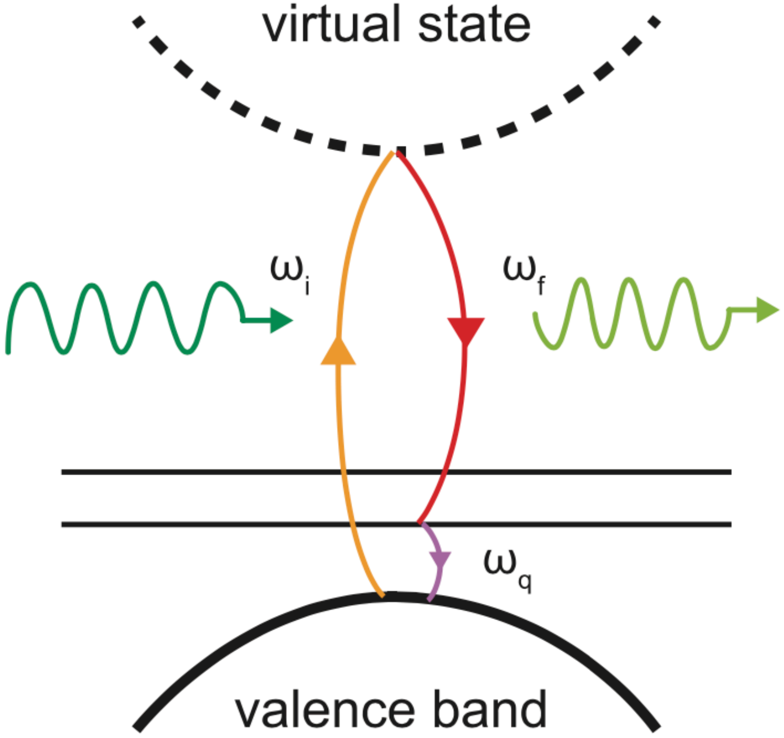
Characterizing Quantum Liquid Phases of Magnetic Insulators

- *Quantum liquids* are a broad family of quantum mechanically induced exotic phases of magnetic insulators
    - Conventional phases: Luttinger Liquids (LLs), quantum spin liquids (QSLs), etc.
    - Novel unconventional: Sliding LLs, Bose-LLs, etc.
- Characterized by **lack of magnetic ordering,** (e.g. via geometric frustration), exotic excitations such as **spinons**
- Highly variable phenomena makes it difficult to positively define a quantum liquid

Francis Pratt/ISIS/Science and Technology Facilities Council

Raman Spectroscopy Probes Magnetic Excitations

- Inelastic Raman scattering with London-Fleury exchange
- Incident photon ω_i excites virtual particle-hole pair
- Pair annihilates to scattered photon ω_f and magnetic excitation)
- Raman shift $\omega = \omega_f - \omega_i$



Wulferding et al. [6]

Raman Spectroscopy Probes Dynamics

- Low energy Raman shifts well below charge gap (electronic DoF frozen out)
- Raman operator R
- Raman spectrum $I(\omega)$
  - Fourier transform of time autocorrelation of R on ground state
  - Analogous to neutron scattering at $q = 0$
- **Spectral equivalence**: R and $R' = R + CH$ give same inelastic spectrum; $R \cong R'$

$$I(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \; e^{i\omega t} \langle R(t) R(0) \rangle_0$$

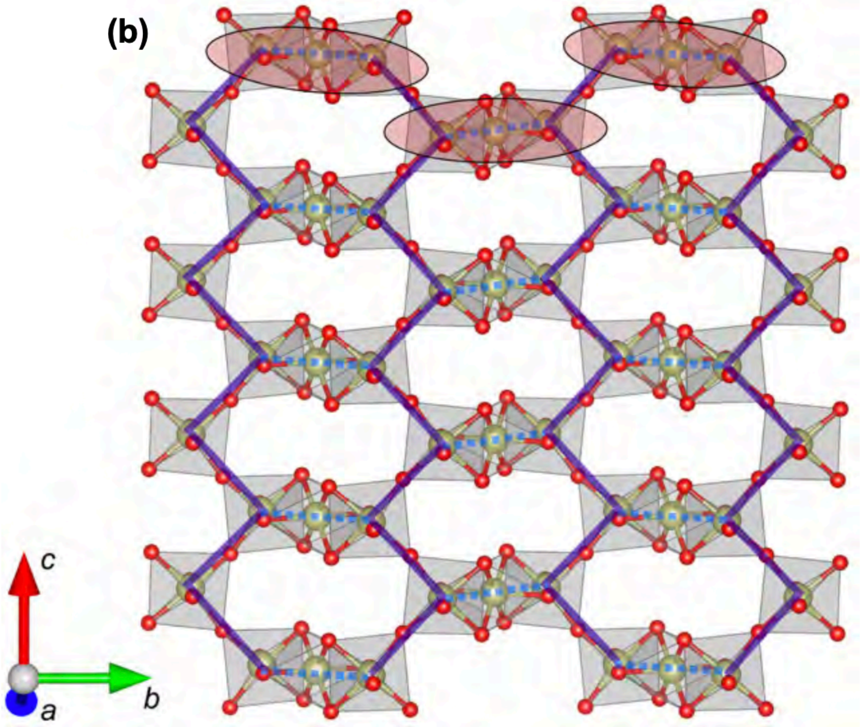
Raman Spectroscopy Probes Spin Dynamics

- London-Fleury photon-induced superexchange operator [4]
- $\hat{\mathbf{e}}_{i,s}$: incident/scattered photon polarization
- $\mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2$
- $A(\mathbf{r}_{12})$: prefactor, on order of exchange couplings J
- Dynamics of R give insight into spin-spin correlations

$$R = \sum_{\mathbf{r}_1, \mathbf{r}_2} (\hat{\mathbf{e}}_i \cdot \mathbf{r}_{12})(\hat{\mathbf{e}}_s \cdot \mathbf{r}_{12}) A(\mathbf{r}_{12}) \mathbf{S}_{r_1} \cdot \mathbf{S}_{r_2}$$

$Ba_4Ir_3O_{10}$ Measurements Suggest Quantum Liquid Candidacy

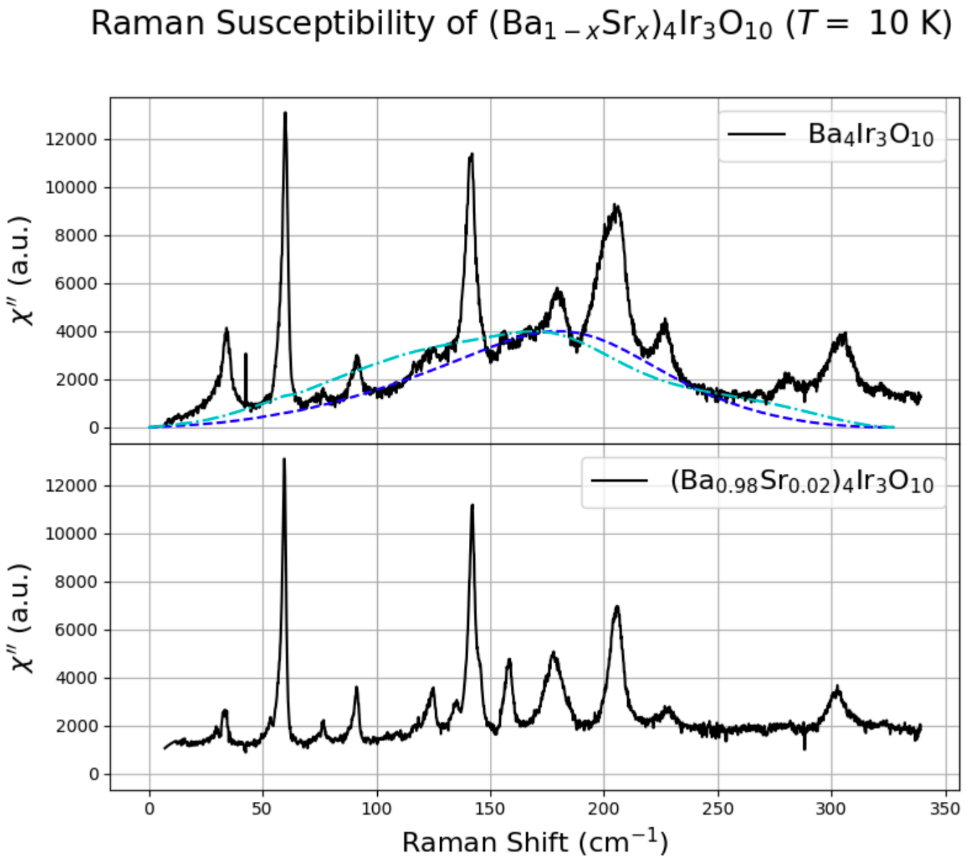
- Magnetic insulator
- Spin-orbit coupled Ir ion with effective spin-1/2
- θ_{CW} between -766 K and -169 K (AFM; $J > 0$)
- Linear heat capacity at low T
- No magnetic order down to 0.2 K
- 2% non-magnetic substitution of Ba to Sr
  - Precipitates long range order at 130 K
  - No more linear T features



Ir_3O_{12} trimers (red ovals) seen as zig-zag chains (purple) with interchain couplings (blue, dashed) [2]

Raman Spectrum of $(Ba_{1-x}Sr_x)_4Ir_3O_{10}$

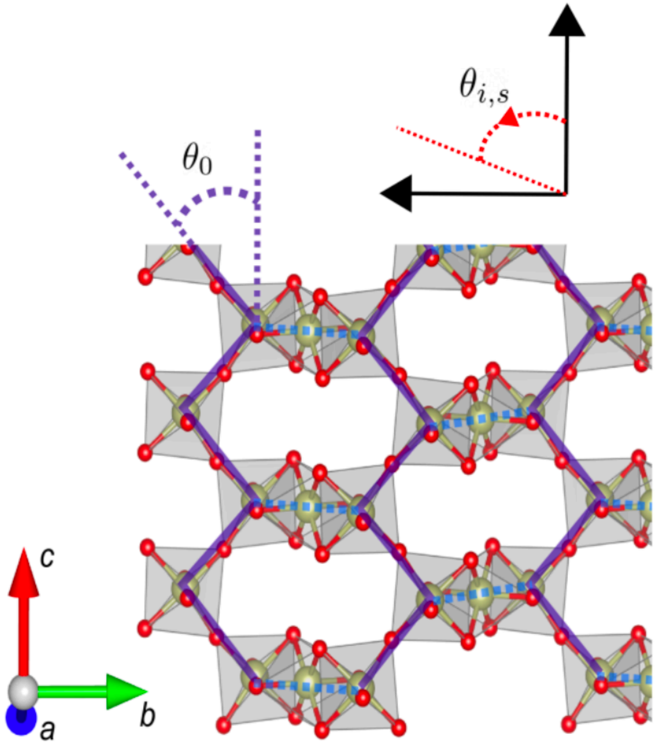
- $I(\omega) = \chi''(\omega)/(1 - e^{-\beta\omega})$
- Imaginary part of Raman operator susceptibility
- Spectrum taken in bb polarization (perpendicular to chain axis)
- Peaks are phonon modes
  - Narrower linewidth means longer phonon lifetime
  - Usually, impurities damp phonon lifetime
  - Here, 2% substitution gives ***longer*** phonon lifetime!
- **Broad hump feature** in pure sample spectrum (fractional excitations?)



Sokolik et al. [5]

Theoretical Model: Decoupled 1D Chains

- Tractable theoretical model
- NB: $Ba_4Ir_3O_{10}$ is neither 1D nor consisting of 1D chains
- Claim: at low T, 1D spinons fruitfully capture dynamics
- Frustrated J_1, J_2 AFM model (fractional excitations from g.s.)



$$H = \sum_j J_1 \mathbf{S}_j \cdot \mathbf{S}_{j+1} + J_2 \mathbf{S}_j \cdot \mathbf{S}_{j+2}$$

Raman Operators for Zig-Zag Chains

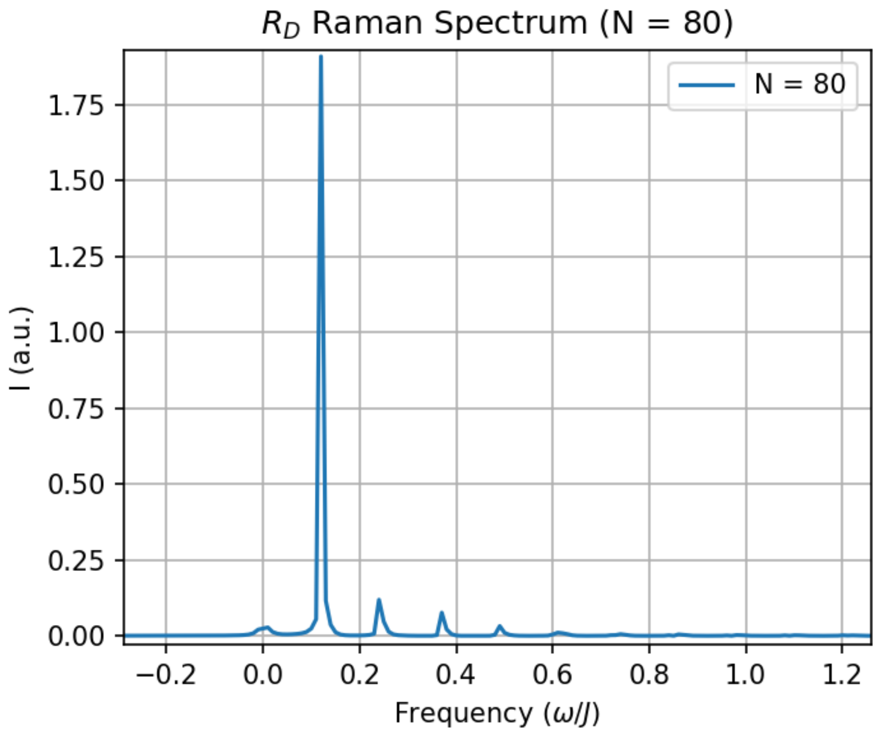
- Compute Raman operator for 1D zig-zag chain with bb polarizations
- Straight chains
  - $J_2 = 0 : R \cong 0$
  - $J_2 > 0 : R \cong R_D$ for bc, cb polarizations, else 0
- Zig-zag chains
  - $J_2 = 0 : R \cong R_1 + R_2$ for cc, else 0
  - $J_2 > 0 : R \cong R_1 + R_2$ for bb

$$R_D = \sum_j (-1)^j \mathbf{S}_j \cdot \mathbf{S}_{j+1}$$

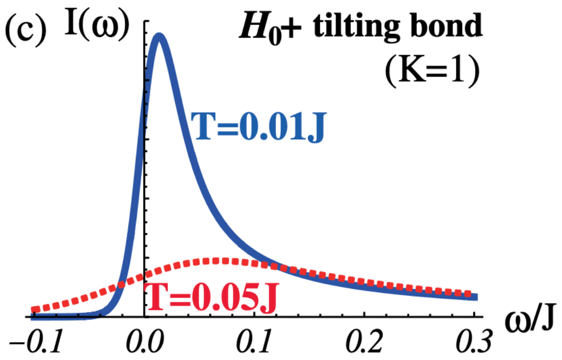
$$R_{\nu=1,2} = \sum_j \mathbf{S}_j \cdot \mathbf{S}_{j+\nu}$$

R_D Spectrum Inconsistent with $Ba_4Ir_3O_{10}$ Spectrum

- R_D produces a sharp, temperature sensitive peak [3]; no broad hump
- Tensor network calculations (DMRG, TEBD) show similar feature



R_D spectrum also show sharp peak ($T = 0$)
(preliminary TEBD calculations using TeNPy)



Sato (bosoniztation) [3]:
R_D gives sharp peak response

Nontrivial bb Spectra Require Zig-Zag chains *and* $J_2 > 0$

- R_D qualitatively inconsistent with experiment
- Spectral equivalence: can choose one of R_ν
- R_ν produce nontrivial broad hump in spectrum within mean field in bb polarization

$$\boxed{R_{\nu=1,2} = \sum_j \mathbf{S}_j \cdot \mathbf{S}_{j+\nu}}$$

Fermionization of Raman Operators

- Jordan-Wigner fermionization of spin operators
- Spinon excitations are the correct excitations in the quantum liquid of this material & the 1D toy model
- Apply to Raman operator and FT
- NB: wavevectors normalized to projection of bonds onto chain axis
- Density-density interaction with some momentum dependence, $h^{(\nu)}_{kk'q}$; e.g. $h^{(1)}_{kk'q} = \cos(q)$
- Approximate to free spinon theory

$$R_\nu \propto \sum_{kk'q} h^{(\nu)}_{kk'q} c^\dagger_k c_{k+q} c^\dagger_{k'} c_{k'-q}$$

$$H = \sum_k \epsilon_k c^\dagger_k c_k$$

$$\epsilon_k = -\frac{\pi}{2} J_{\text{eff}} \cos(k)$$

Spinon Mean Field Theory: Computing $I(\omega)$

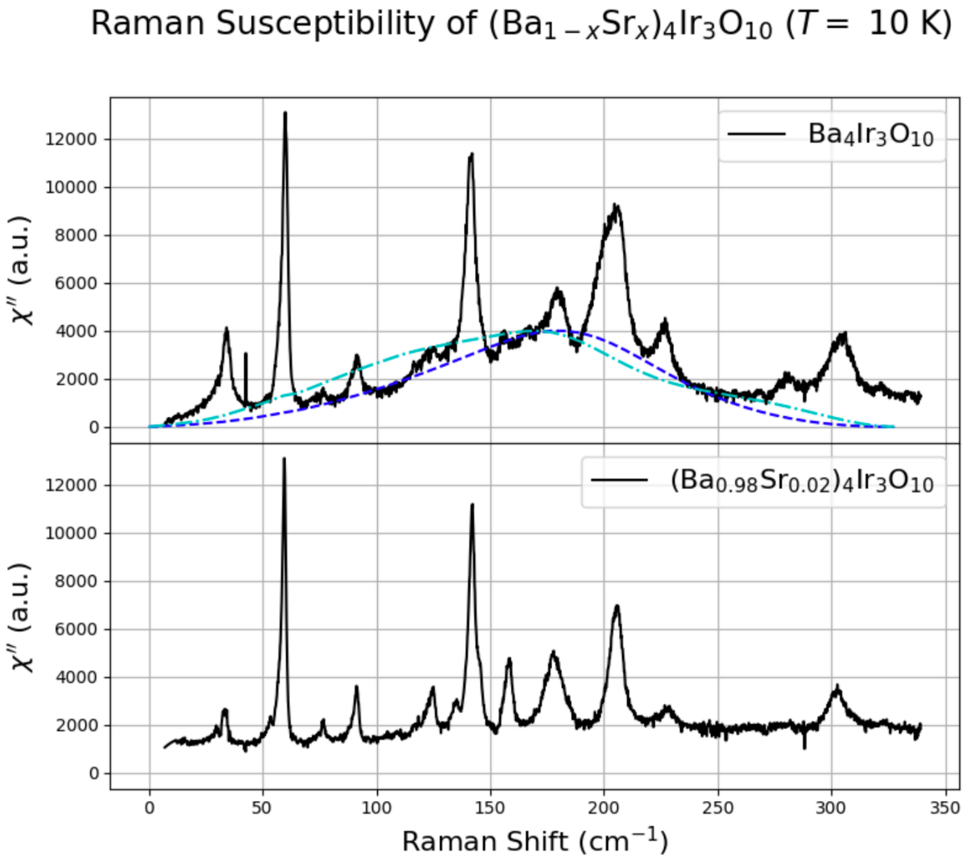
- Time evolve spinon operators
- Evaluate correlation function on g.s
  - R_ν is quadratic in $\mathbf{S}$
  - $\mathbf{S}$ is quadratic in c_k
  - 8 fermionic operators in correlation function
  - one-particle-hole excitations (zero total momentum) do not contribute within mean field
- Wick's theorem: evaluate using 2-pt correlates
- Intensity arising from R_ν given as integrals in momentum space

$$\langle R_\nu(t) R_\nu(0) \rangle$$

$$I^{(\nu)}(\omega) \propto \int_{-\pi}^{\pi} dk \int_{-\pi}^{\pi} dq \sum_{k'} \frac{h^{(\nu)}(k,k',q)[h^{(\nu)}(k,k',q) - h^{(\nu)}(k,k',k'-k-q)]}{\sqrt{(2t\sin(q/2))^2 + (\epsilon_{k+q} - \epsilon_k - \omega)^2}} \times f(\epsilon_k)(1-f(\epsilon_{k+q}))f(\epsilon_{k'})(1-f(\epsilon_{k'-q}))$$

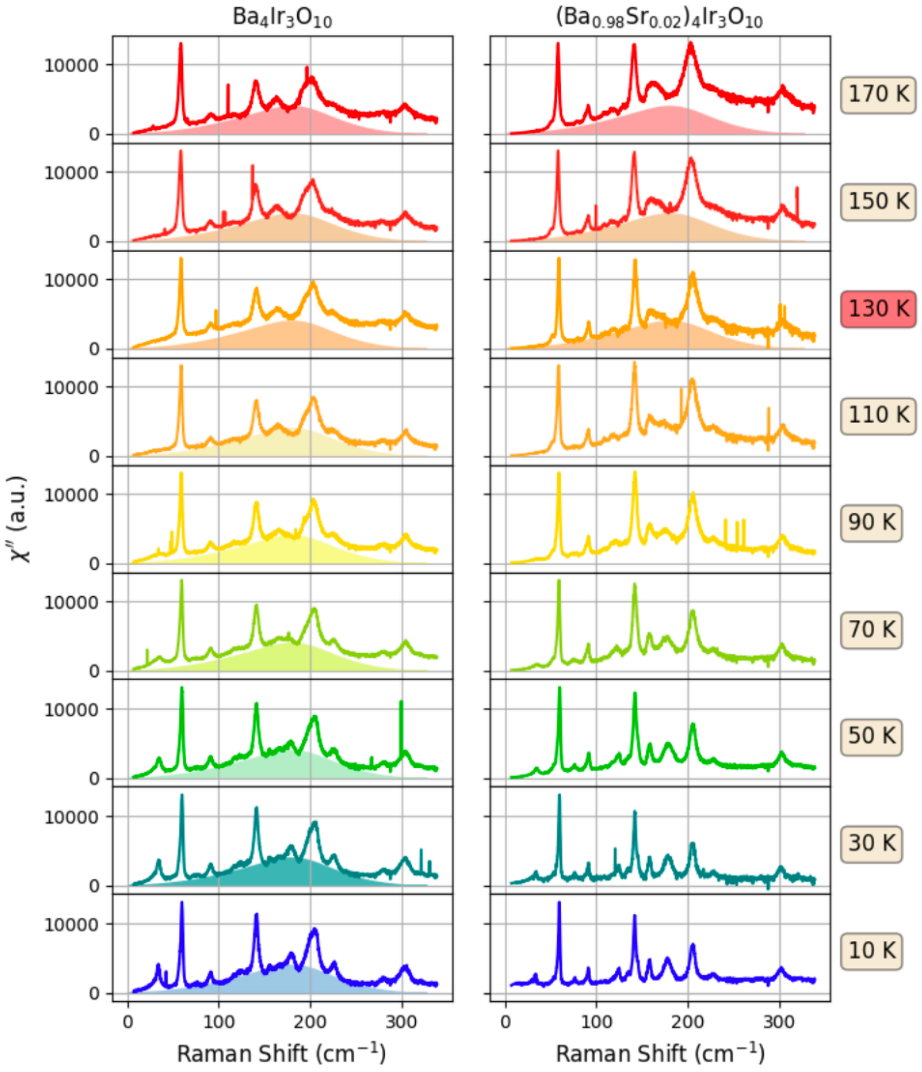
Mean Field Spectrum of $Ba_4Ir_3O_{10}$

- Broad hump feature arising from 4-spinon continuum using 1D spinon mean field at low T
  - R_1 (dashed, blue)
  - R_2 (dot-dashed, cyan)
  - Both mean fields capture hump!
- No hump after substitution
- Captured by $J_{eff}/k_B \approx 75$ K


Raman Susceptibility of $(Ba_{1-x}Sr_x)_4Ir_3O_{10}$ ($T =$ 10 K)

Sokolik et al. [5]

$Ba_4Ir_3O_{10}$ Susceptibility Temperature Dependence

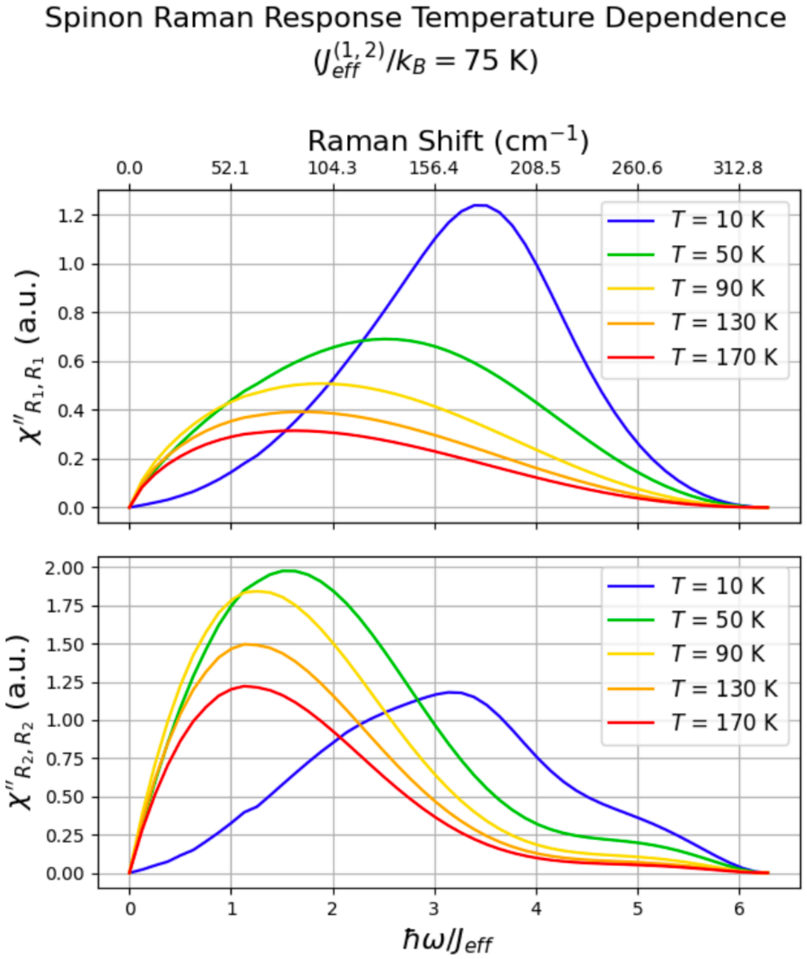


Sokolik et al. [5]

- Temperature dependence of bb Raman spectrum
- Broad hump and broadened (shorter lifetime) phonons persist up to higher temperatures
- Similar hump in sister compound above Néel temperature
- Experiment always at low T relative to true effective spinon theory
- Interactions and 3D nature of material important at intermediate experiment T

Mean Field Susceptibility Temperature Dependence

- Hump feature persists at high temperatures, but shape changes (not seen in experiment)
- Qualitative differences between R_1, R_2 theories quantify breakdown of mean field theory at high temperature $(k_B T/J_{eff} > 1)$



Sokolik et al. [5]

Microscopics and Phonon Broadening

- Self-consistency equation (Brenig [1]) relates J_{eff} to a difference between microscopic J_1, J_2
  - Destructive interference of nn and nnn tunneling
  - J_{eff}/k_B of 75 K permits $J_{1,2}/k_B$ of 324 K
- Range of J_1, J_2 consistent with Curie-Weiss measurements
- Phonon broadening due to scattering of phonons and spinons
  - Observed in spin-orbit coupled materials (e.g. Sr_2IrO_4 [7])
  - Similarly observed here
- Broadened phonons persisting down to low T consistent with magnetic quantum liquid
- Narrowing of phonons after Sr substitution further supports quantum liquid behavior

Conclusion: Raman signatures for spinons in possible spin-orbit coupled quantum liquid $Ba_4Ir_3O_{10}$

- Broad hump arising from 4-spinon continuum in 1D toy model
  - Zig-zag chain + $J_2 > 0$ necessary & sufficient to capture hump within mean field for bb polarization
  - Two equivalent yet distinct mean field approaches (R_1, R_2) both capture hump and temperature dependence
- Strong phonon damping from phonon-spin coupling via spin-orbit interaction
- 2% non-magnetic Ba-to-Sr substitution precipitates magnetically ordered phase without hump, phonon damping: spinon features are fragile to disorder