Raman Responses with and without Topological Defects

Sami Hakani

Group of Itamar Kimchi

A Quantum Many-Body Handshake

Weizmann Institute of Science, 22 December 2022

Georgia Institute of Technology

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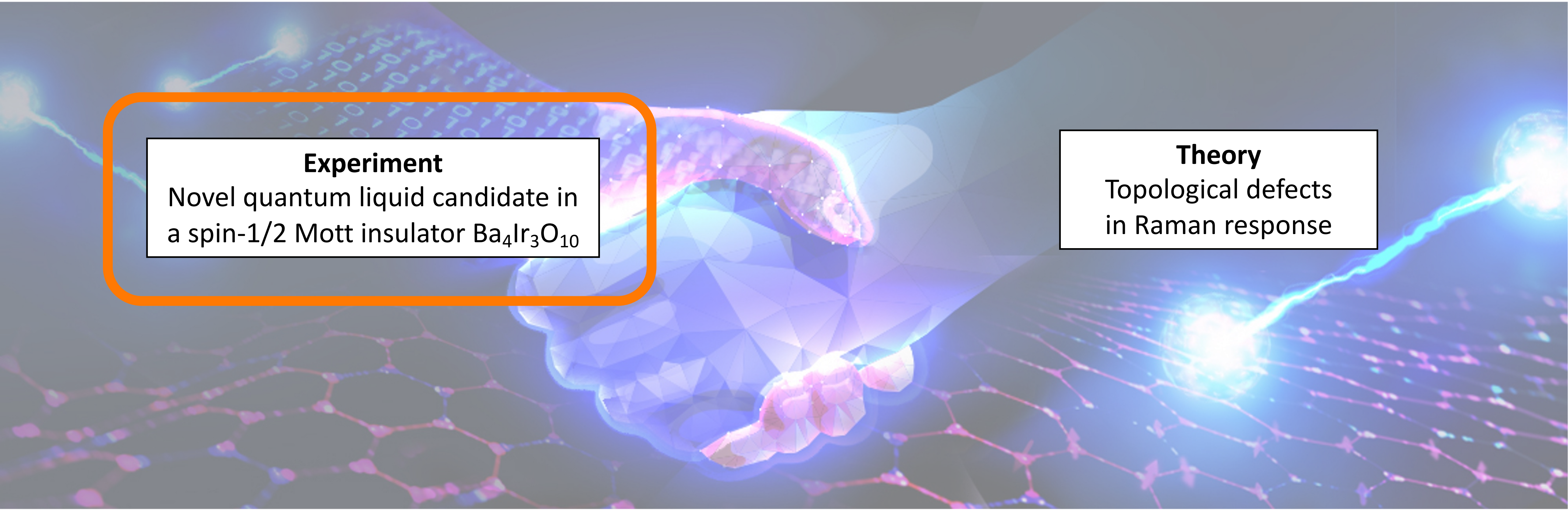
Special thanks to the organizers!

Outline



Experiment
Novel quantum liquid candidate in a spin-1/2 Mott insulator $Ba_4Ir_3O_{10}$

Theory
Topological defects in Raman response

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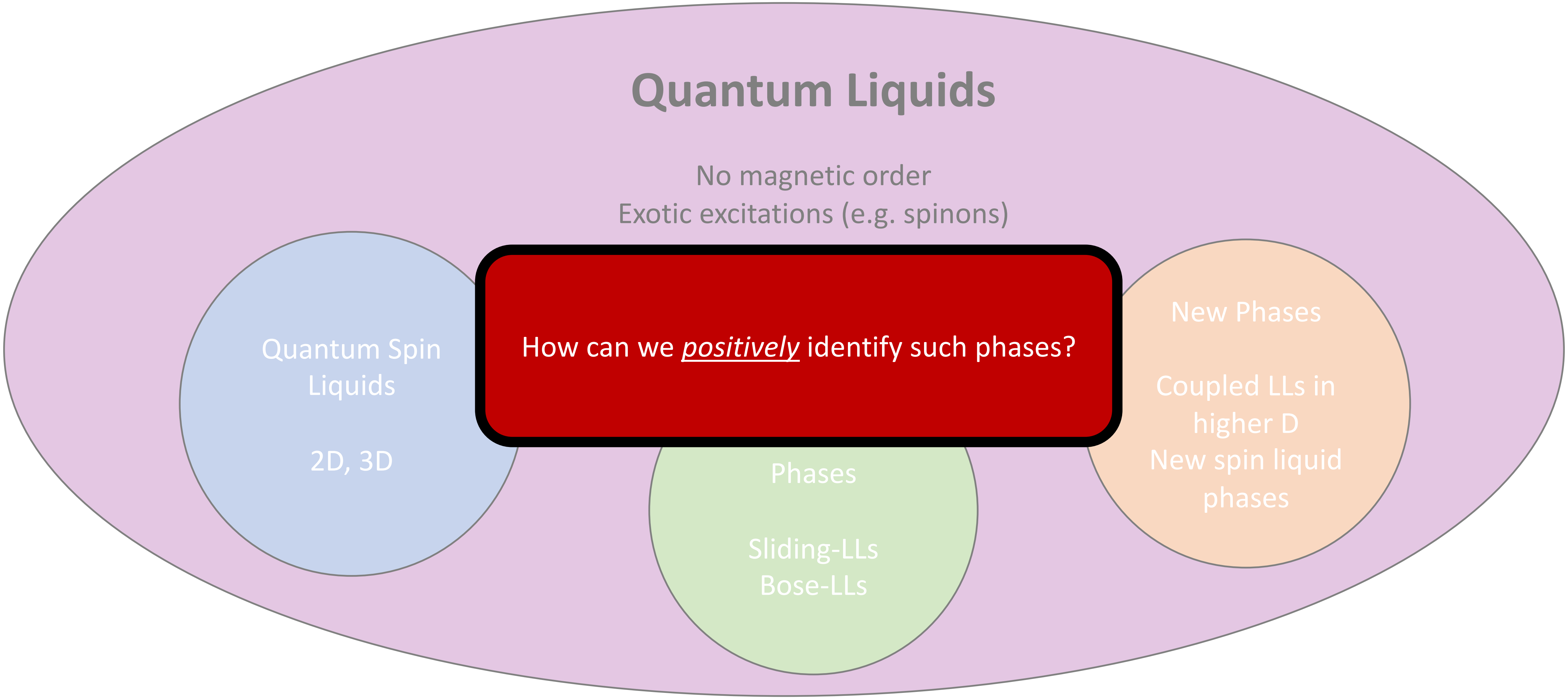


Quantum Liquids

No magnetic order
Exotic excitations (e.g. spinons)

Quantum Spin
Liquids

2D, 3D

How can we *positively* identify such phases?

Phases

Sliding-LLs
Bose-LLs

New Phases

Coupled LLs in
higher D
New spin liquid
phases

Raman spectroscopy probes dynamics of magnetic excitations

Loudon-Fleury superexchange

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

$$\mathbf{r_{12}} = \mathbf{r_1} - \mathbf{r_2}$$

Prefactor
Scales like exchange on bond

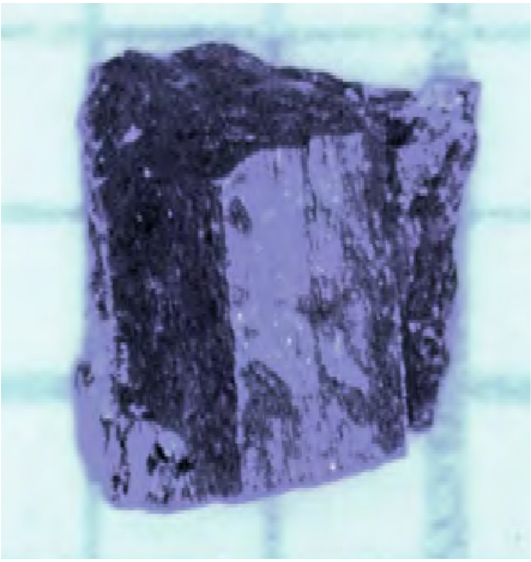
Incident/scattered
Photon polarization

$$I(\omega) = \frac{1}{2\pi}\int dt\, e^{i\omega t}\langle R(t)R(0)\rangle_0$$

Raman spectroscopy can probe various magnetic systems

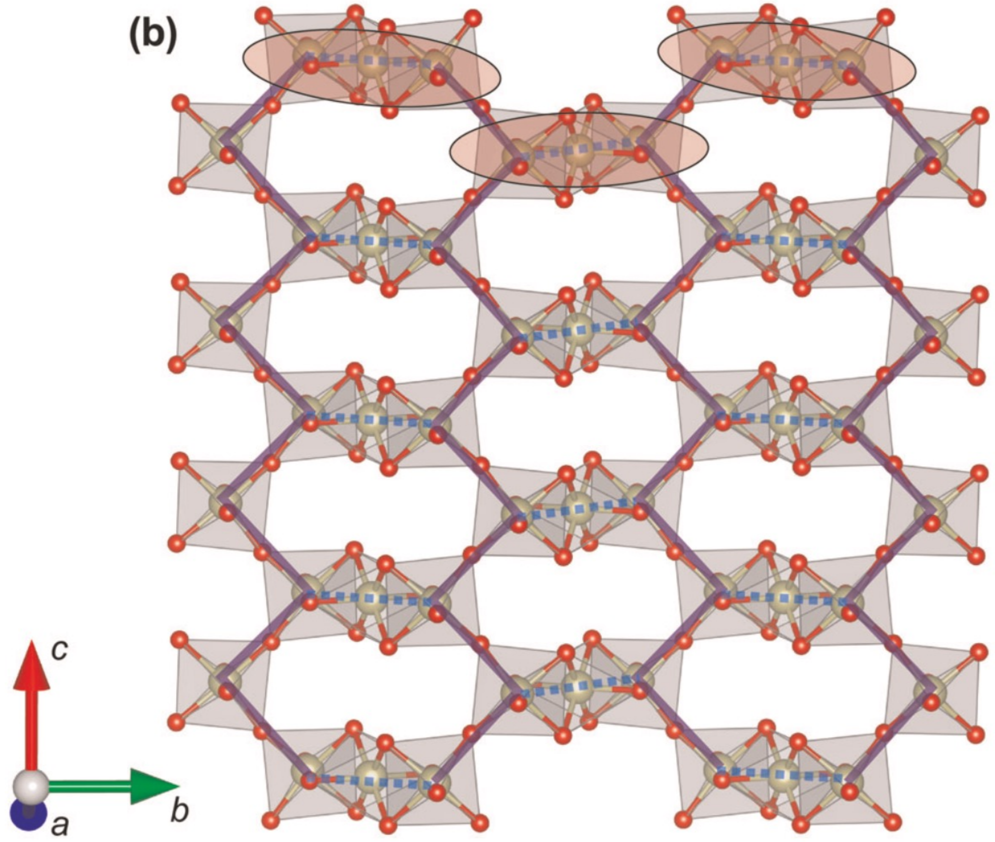
- Geometrically frustrated magnets
  - Phys. Rev. B **77**, 174412, N. B. Perkins and W. Brenig
  - Phys. Rev. B **56**, 2551, W. Brenig
- Spin-Peirels
  - Phys. Rev. B **54**, R9635(R), V. N. Muthukumar, C. Gros, W. Wenzel, R. Valentí, P. Lemmens, B. Eisener, G. Güntherodt, M. Weiden, C. Geibel, and F. Steglich
- 1D magnets
  - Phys. Rev. Lett. **108**, 237401, M. Sato, H. Katsura, and N. Nagaosa
  - Phys. Rev. Lett. **77**, 4086, R. R. P. Singh, P. Prelovšek, and B. S. Shastry
- Kitaev systems
  - Phys. Rev. Lett. **113**, 187201, J. Knolle, Gia-Wei Chern, D. L. Kovrizhin, R. Moessner, and N. B. Perkins
  - Phys. Rev. B **104**, 144412, Y. Yang, M. Li, I. Rousochatzakis, and N. B. Perkins

Quantum Liquid Candidate $Ba_4Ir_3O_{10}$



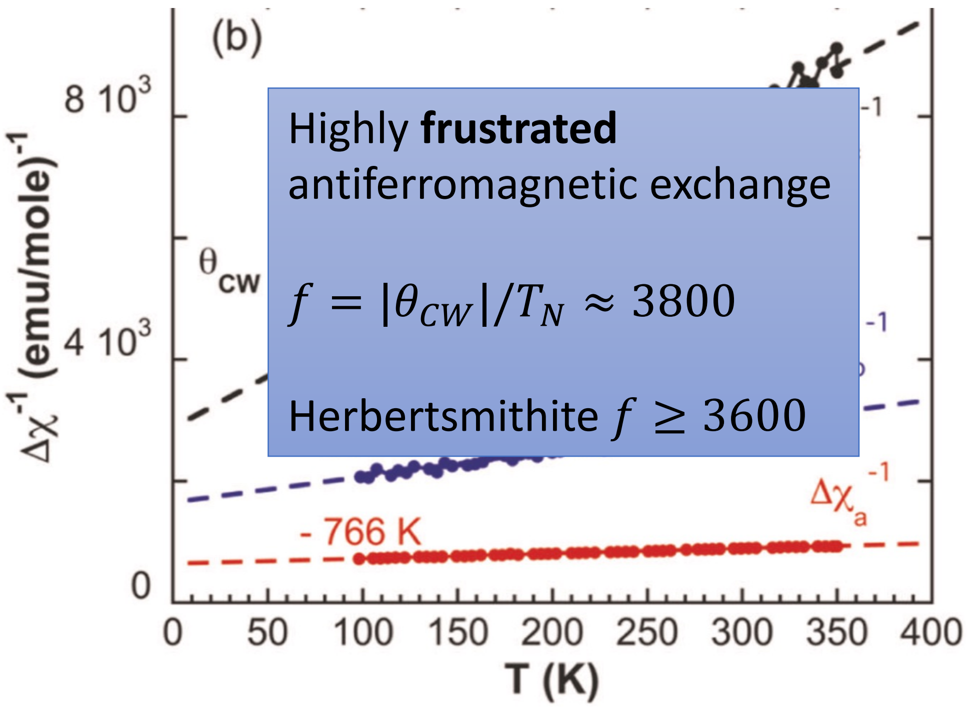
$Ba_4Ir_3O_{10}$ Measurements Suggest Quantum Liquid Candidacy

- 2D or 3D magnetic insulator
- Spin orbit coupled Ir ion with effective s = 1/2
- Magnetic order at $T_N \approx 0.2$ K



npj Quantum Materials, *5*(1), Article 1.
Cao, G., Zheng, H., Zhao, H., Ni, Y., Pocs, C. A., Zhang, Y., Ye, F., Hoffmann, C., Wang, X., Lee, M., Hermele, M., & Kimchi, I. (2020).

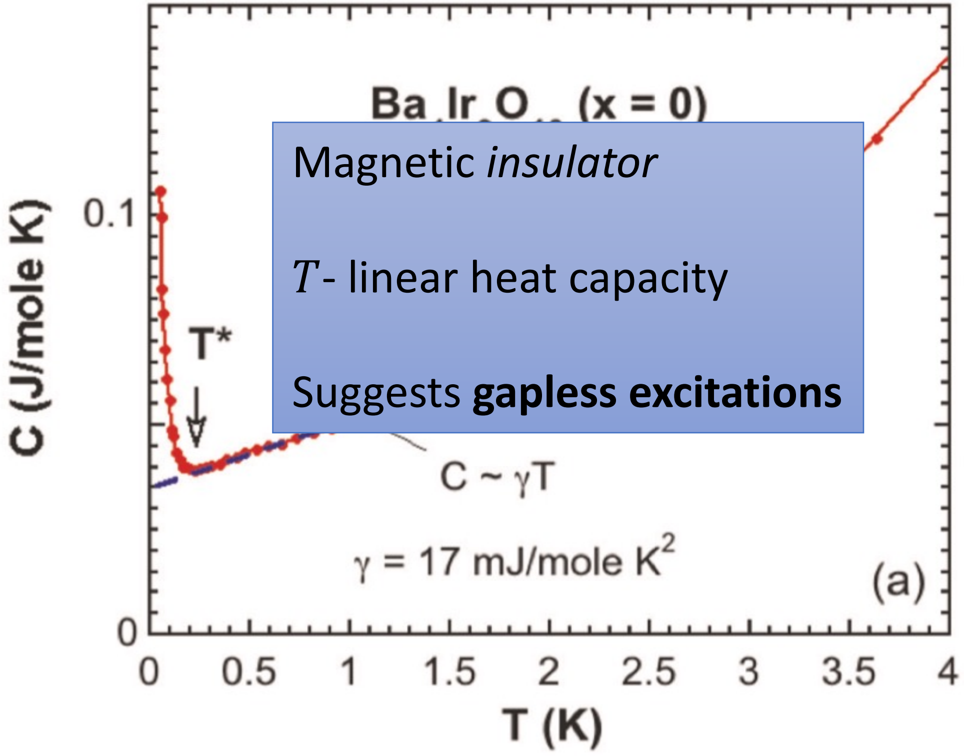
Thermodynamic Measurements Suggest $Ba_4Ir_3O_{10}$ is a Quantum Liquid



Highly **frustrated** antiferromagnetic exchange

$f = |\theta_{CW}|/T_N \approx 3800$

Herbertsmithite $f \geq 3600$



Magnetic *insulator*

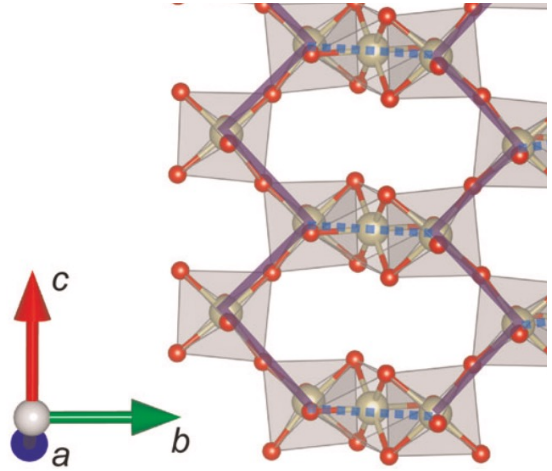
T- linear heat capacity

Suggests **gapless excitations**

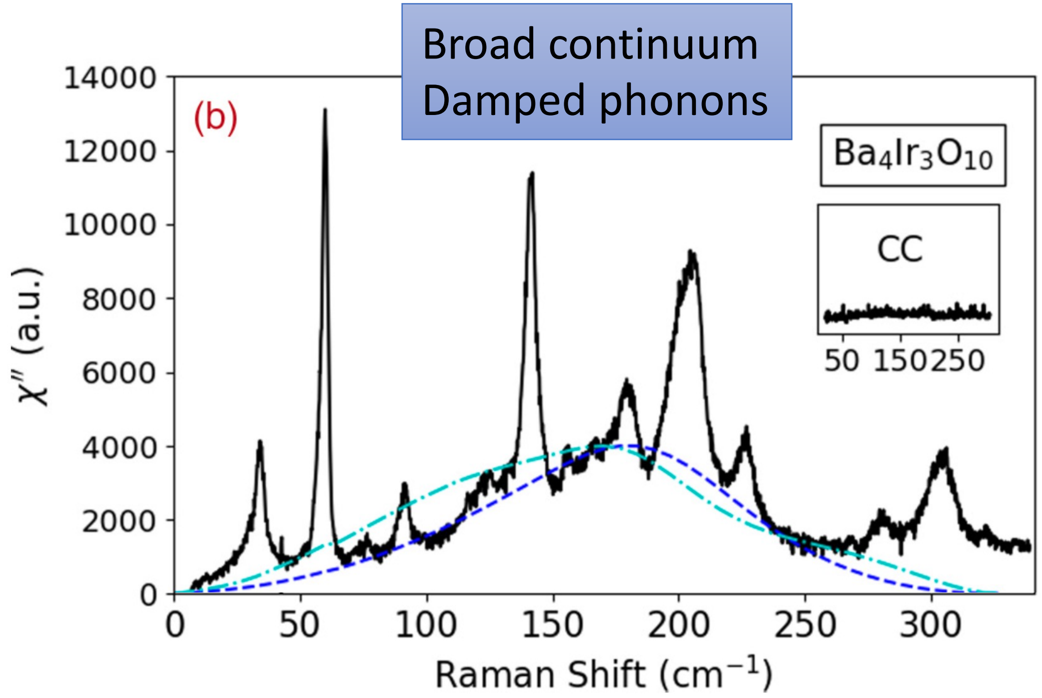
$Ba_4Ir_3O_{10}$ quantum liquid state is destroyed upon adding disorder

- 2% non-magnetic substitution of Ba to Sr
- Magnetic order at 130 K (cf. $T_N \approx 0.2$ K)
- No more linear T features in heat capacity
- Reduced frustration ratio

bb Raman susceptibility ($T = 10$ K)

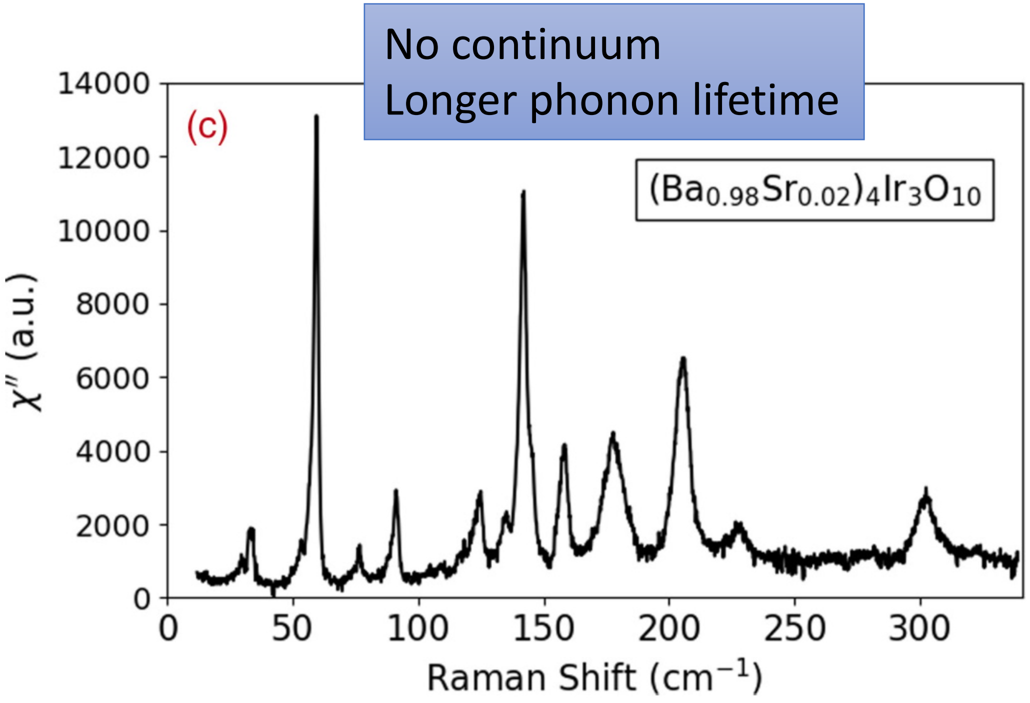


$Ba_4Ir_3O_{10}$
(no magnetic order)



Broad continuum
Damped phonons

$(Ba_{0.98}Sr_{0.02})_4Ir_3O_{10}$
(magnetically ordered)



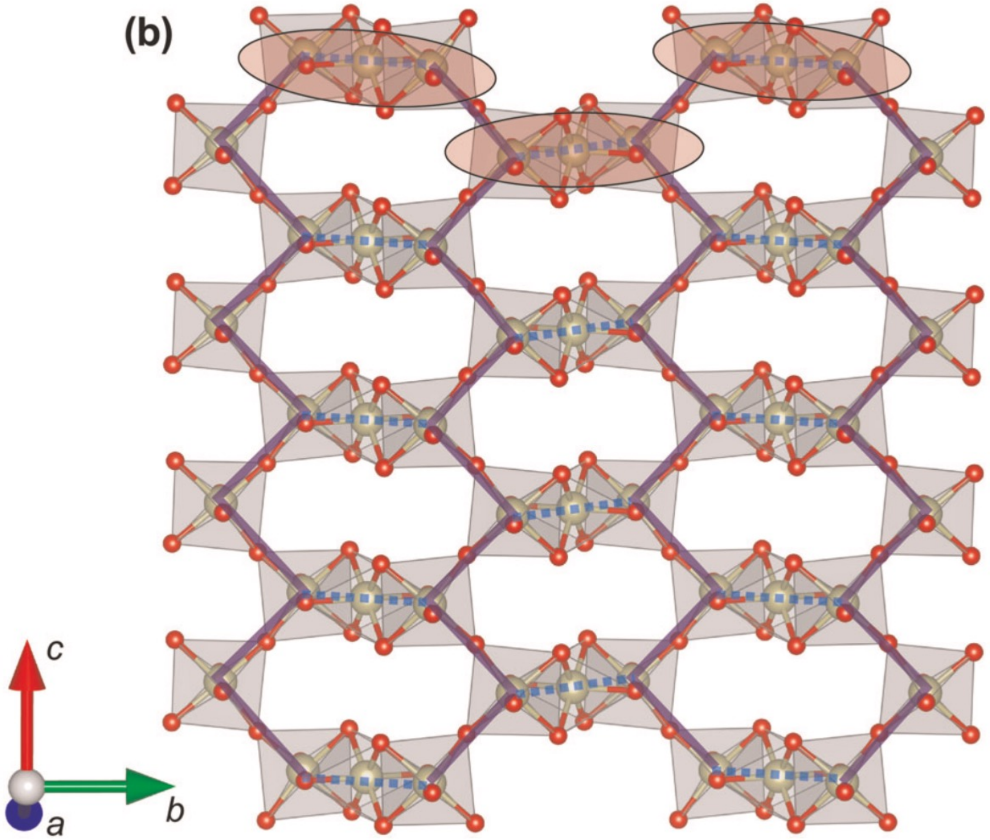
No continuum
Longer phonon lifetime

Phys. Rev. B **106**, 075108 (2022)
A. Sokolik, **SH**, S. Roy, N. Pellatz, H. Zhao, G. Cao, I. Kimchi, and D. Reznik

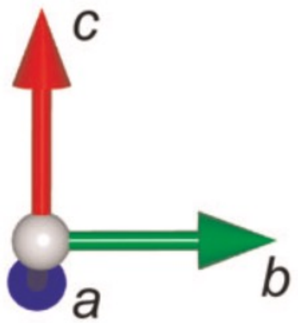
Theoretical model: decoupled 1D chains

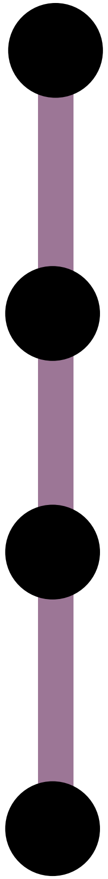
- 1D is tractable
- NB: $Ba_4Ir_3O_{10}$ is neither 1D nor consists of 1D chains
- **Claim: at low T, 1D spinons fruitfully capture dynamics**
- Ground state of H has fractional excitations

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



No *bb* Raman operator for straight chains





$$R = \sum_{\mathbf{r}_1,\mathbf{r}_2} A(\mathbf{r}_{12})(\hat{\mathbf{e}}_i \cdot \mathbf{r}_{12})(\hat{\mathbf{e}}_s \cdot \mathbf{r}_{12})\mathbf{S}_{\mathbf{r}_1}\mathbf{S}_{\mathbf{r}_2}$$
$$= 0$$

$$\hat{b} \cdot \boldsymbol{r}_{12} = 0$$

bb Raman operator for zig-zag needs J_2

- For $J_2 = 0$, $R \propto H$ (only elastic response)
- Minimal model for a continuum in spectrum needs both zig-zag **AND** $J_2 > 0$

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

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

Fermionization of Raman operator

- Jordan-Wigner fermionization of spin operators
- Spinons are correct excitations in quantum liquid & in 1D toy model
- Approximate to free 1D spinon gas
- Fermionize Raman operator and FT
- NB: wavevectors normalized to projection of bonds onto longitudinal chain axis

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

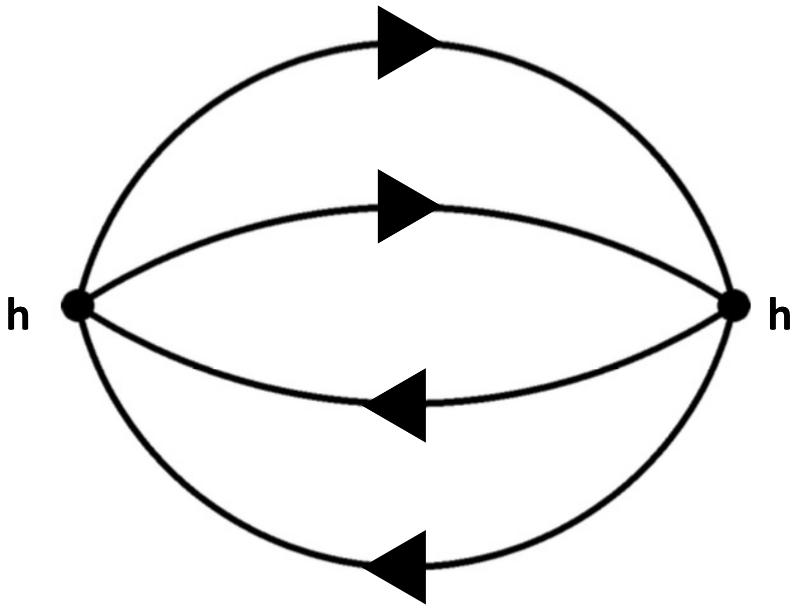
$$R_{\nu=1,2} \propto \sum_{k,k',q} h^{(\nu)}_{kk'q} c^\dagger_k c_{k+q} c^\dagger_{k'} c_{k'-q}$$

$$H_{MF} = \sum_k \epsilon_k c^\dagger_k c_k$$

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

Spinon Mean Field Theory

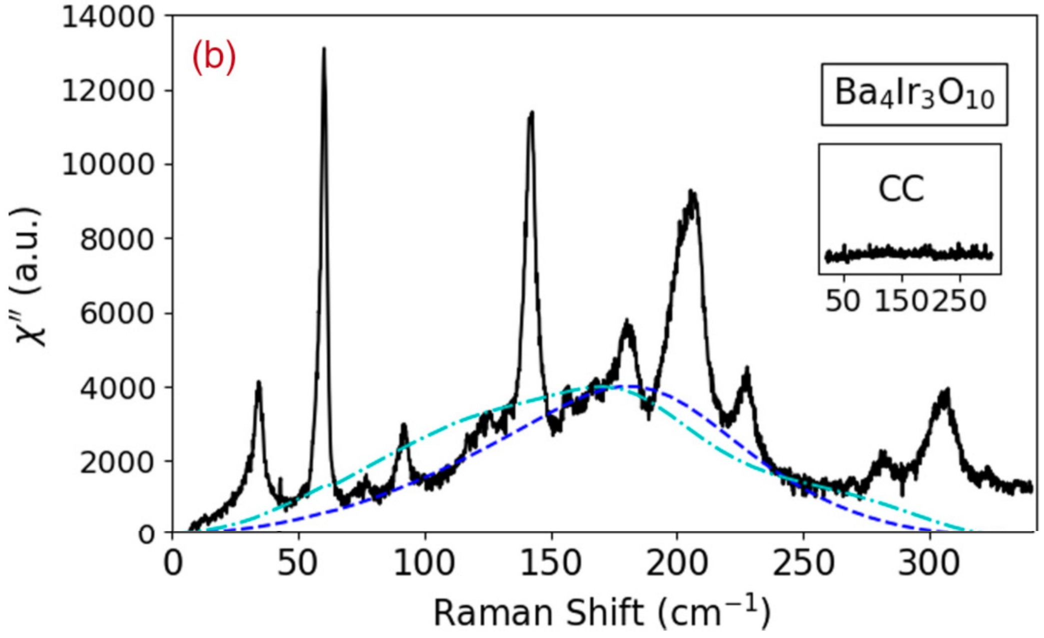
- Time evolve spinons
- Diagramatically expand 8 spinon correlation function on ground state with free propagator
- Most diagrams give elastic contributions



These do not

Mean field spectra of $Ba_4Ir_3O_{10}$

- 4-spinon continuum from two equivalent mean fields
- R_1 (dashed, blue)
- R_2 (dot-dashed, cyan)
- Both capture continuum
- Energy scale of bandwidth consistent with CW

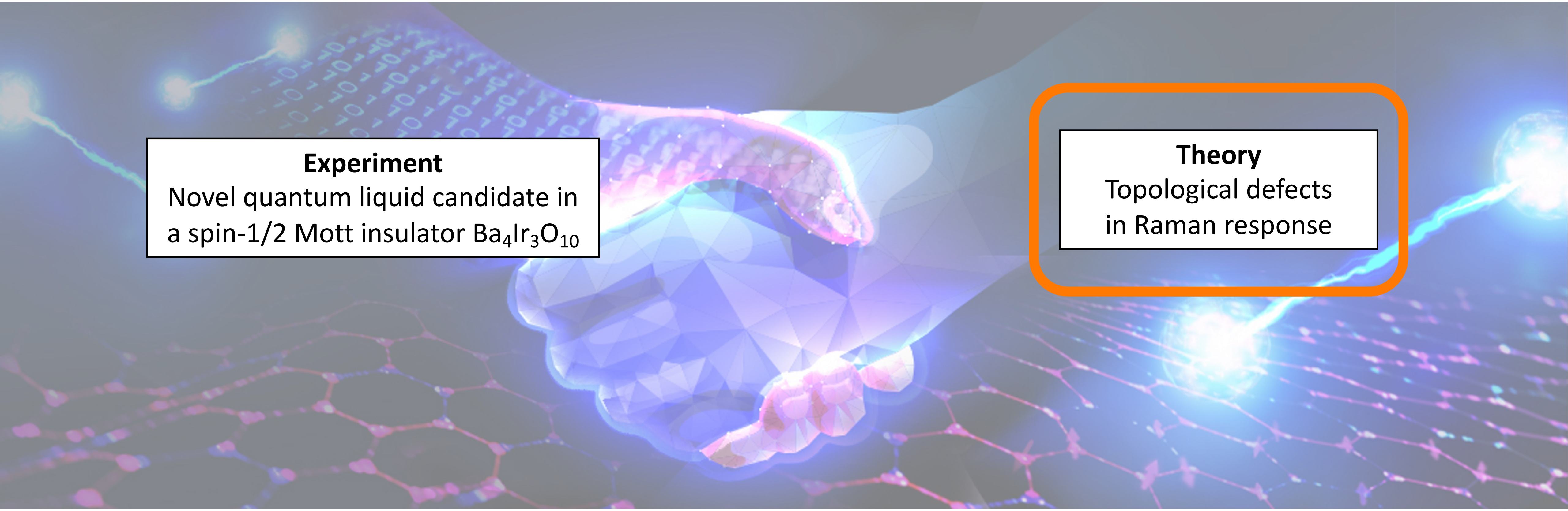


$$I^{(\nu)}(\omega) \propto \int_{-\pi}^{\pi} dk \int_{-\pi}^{\pi} dq \sum_{k'} \frac{h^{(\nu)}(k,k',q)[h^{(\nu)}(k,k',q) - h^{(\nu)}(k,k',k'-k-q)]}{\sqrt{(2t\sin(q/2))^2 + (\epsilon_{k+q} - \epsilon_k - \omega)^2}} \times f(\epsilon_k)(1 - f(\epsilon_{k+q}))f(\epsilon_{k'})(1 - f(\epsilon_{k'-q}))$$

$Ba_4Ir_3O_{10}$ measurements suggest fragile quantum liquid state with gapless spinon excitations

- Broad hump arising from 4-spinon continuum in 1D toy model
  - Zig-zag chain + $J_2 > 0$ needed to capture hump within mean field for *bb* polarization
  - Two equivalent yet distinct mean field approaches (R_1, R_2) capture hump
- 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

Outline



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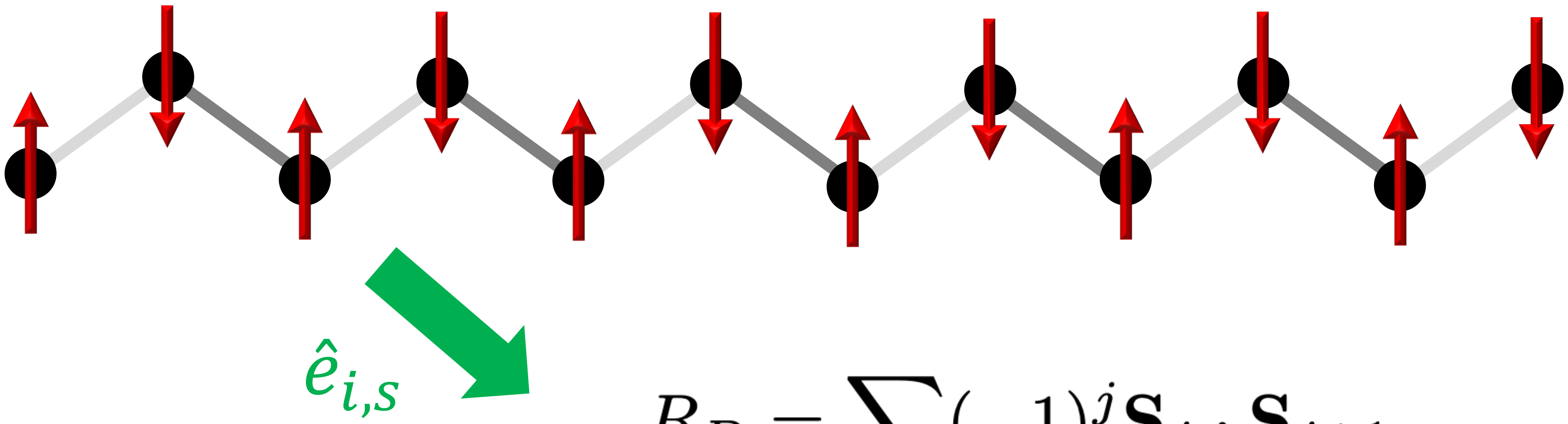
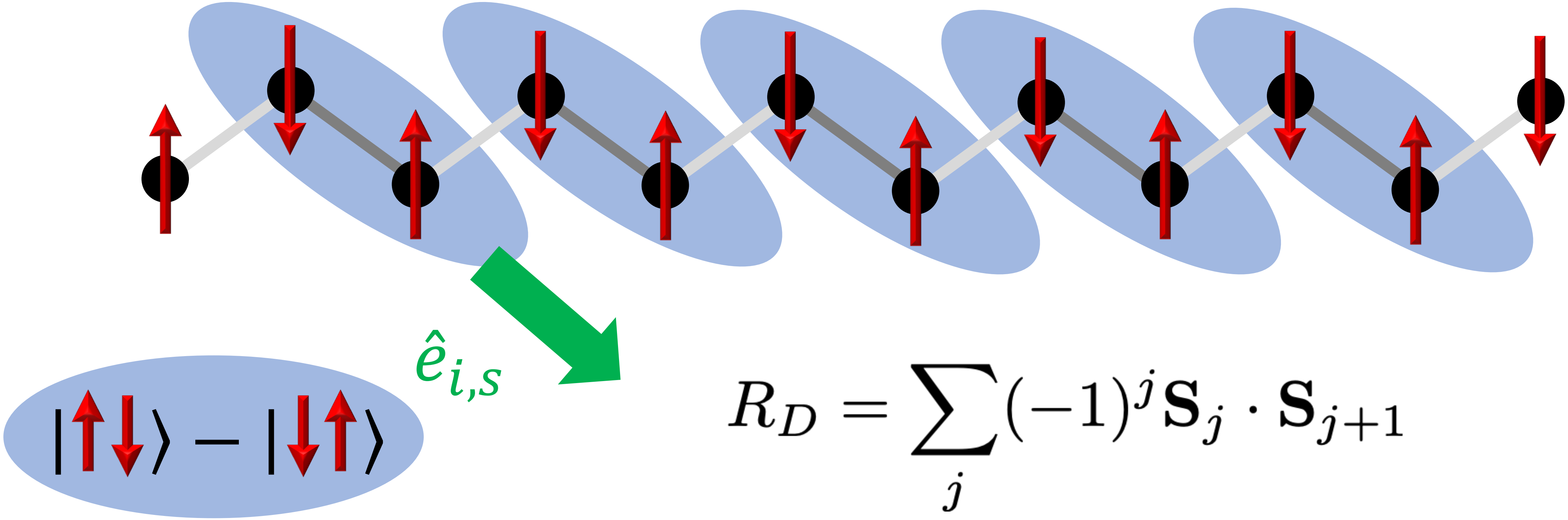
Some Raman operators look like dimerization operators

- Zig-zag chain with *bc* polarization
- Assumes clean zig-zag chain without crystal dislocations

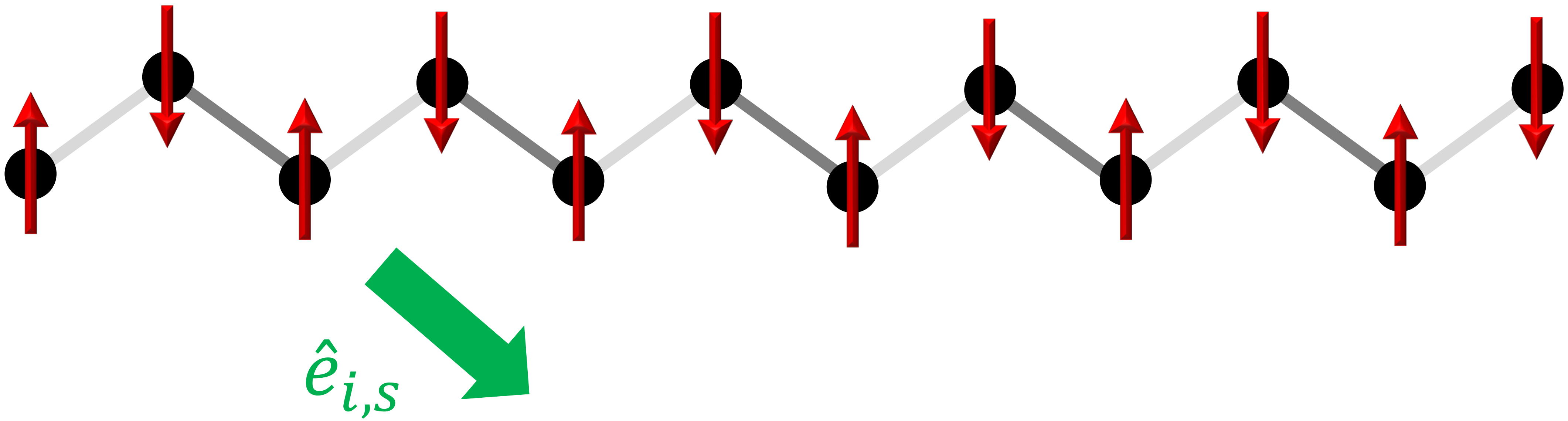


$$R_D = \sum_j (-1)^j \mathbf{S}_j \cdot \mathbf{S}_{j+1}$$

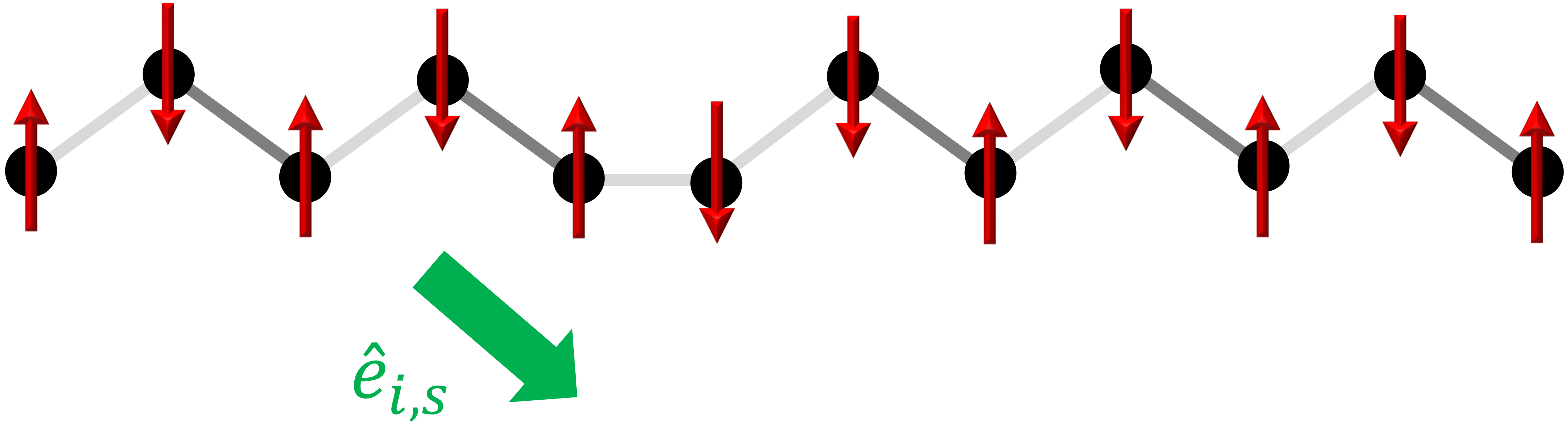
If RD were a Hamiltonian, its ground state would be dimerized



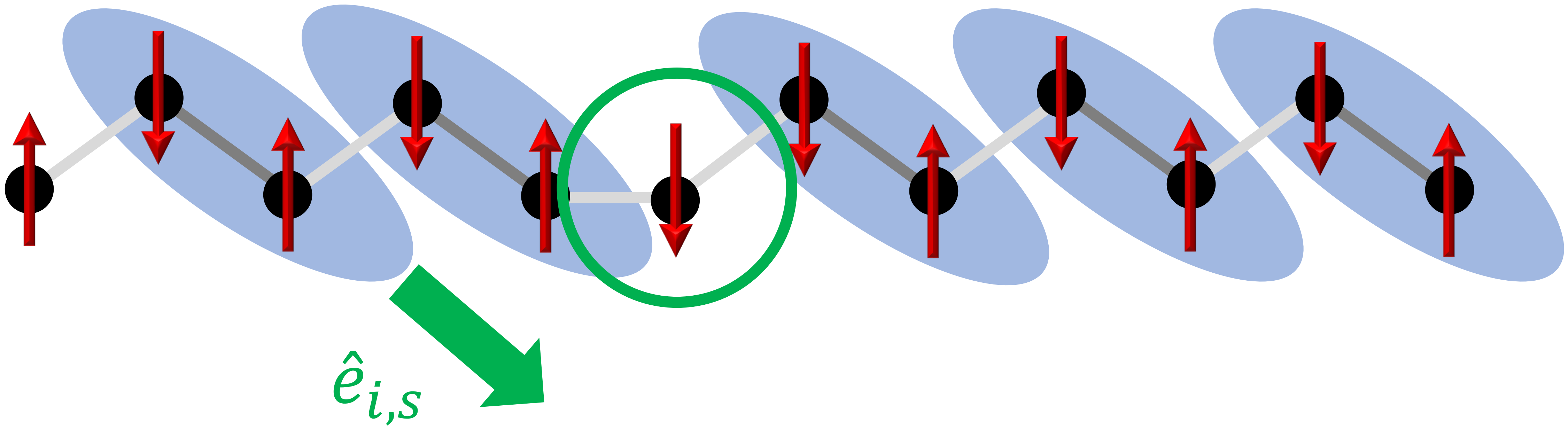
$$R_D = \sum_j (-1)^j \mathbf{S}_j \cdot \mathbf{S}_{j+1}$$


$\hat{e}_{i,s}$

Crystal dislocations induce topological defects in dimerization domains of Raman operator



Crystal dislocations induce topological defects in dimerization domains of Raman operator



Do domain walls respond to magnetic field?

- Use tensor networks to find out

DMRG

- Build H, R as MPOs
- $H = J \sum_j \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+1}$
- Find ground state $|\psi\rangle$ of H

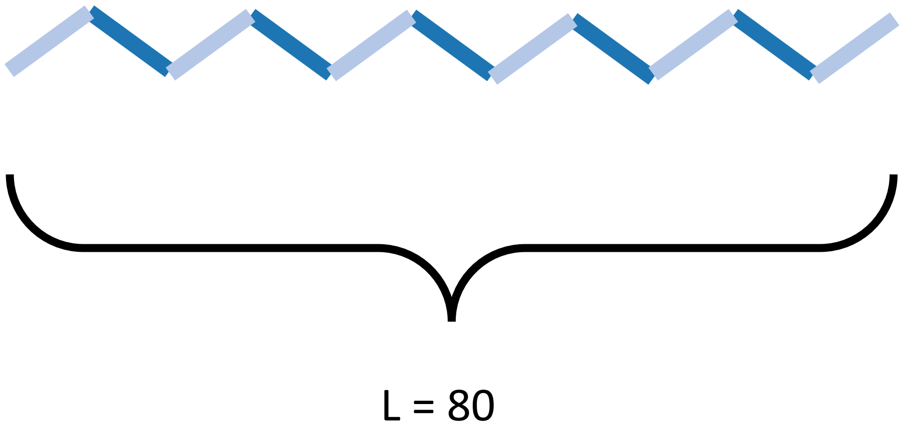
TEBD

- Time evolve to compute $\langle\psi|R(t)R(0)|\psi\rangle$
- Measure at Δt time steps

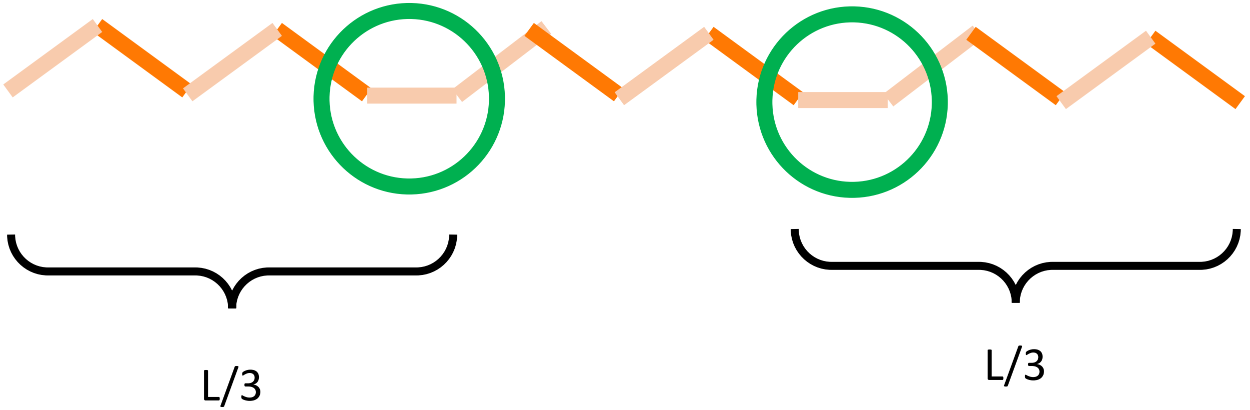
Fourier Transform

Yes: Domain walls respond to magnetic field

No defects

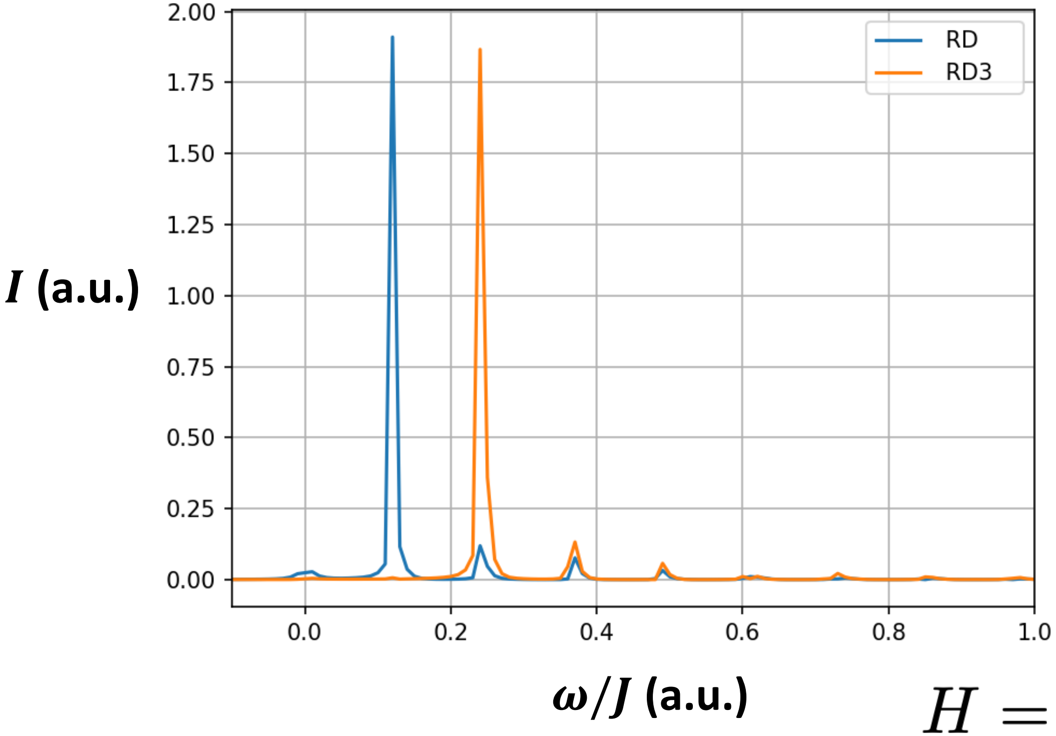


Two defects

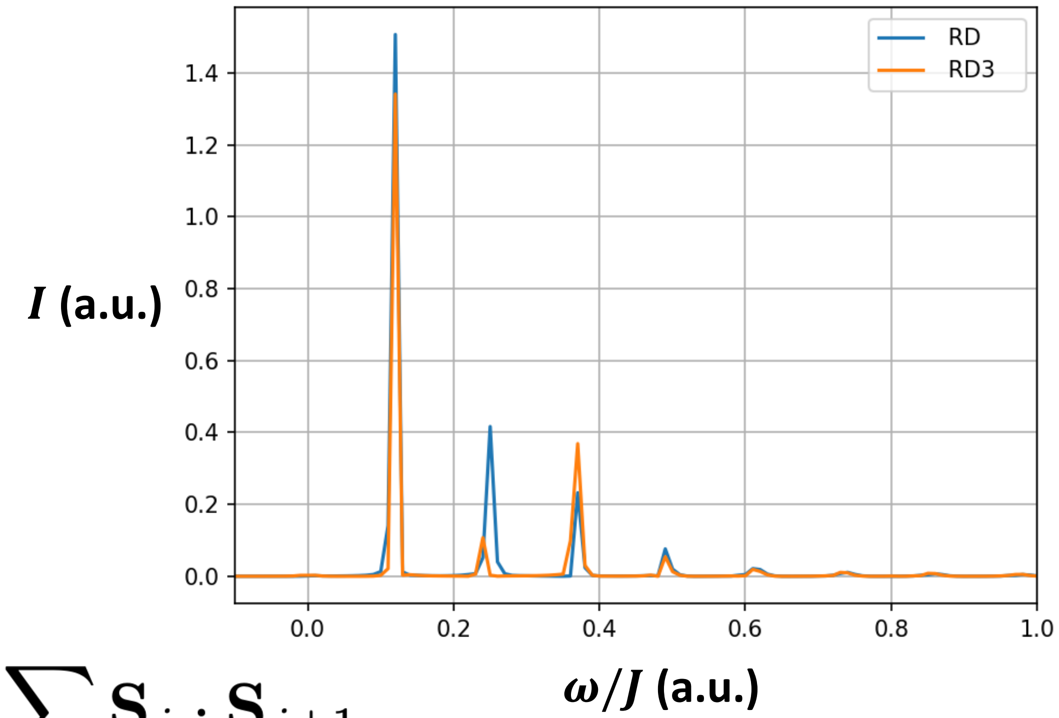


Domain wall (orange) responds to magnetic field

Zero Field



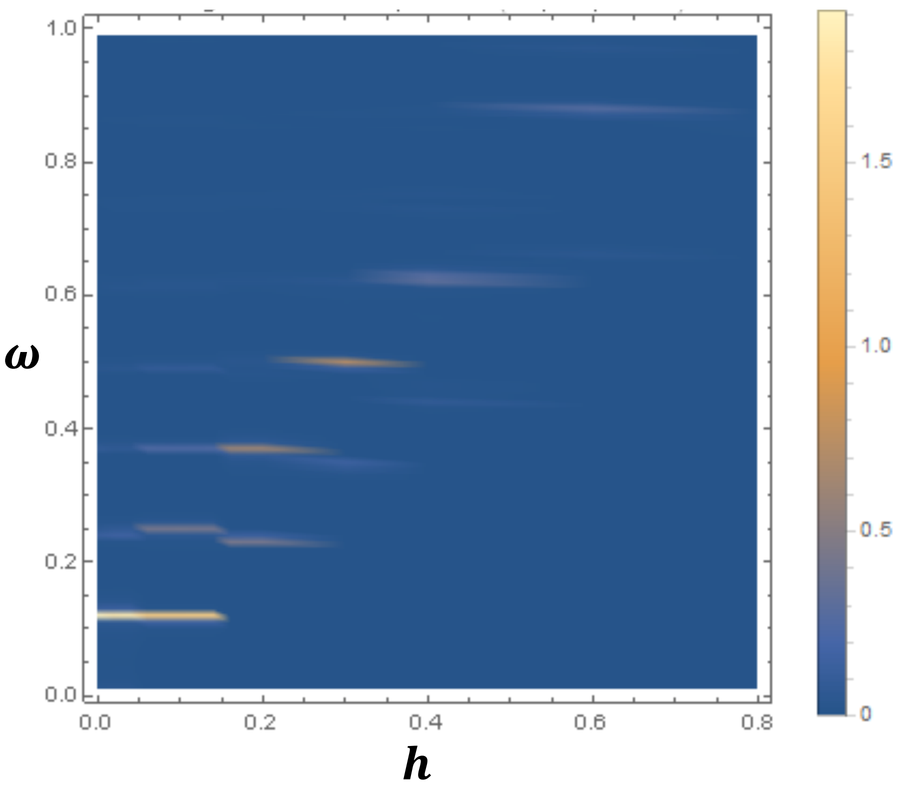
Small Field



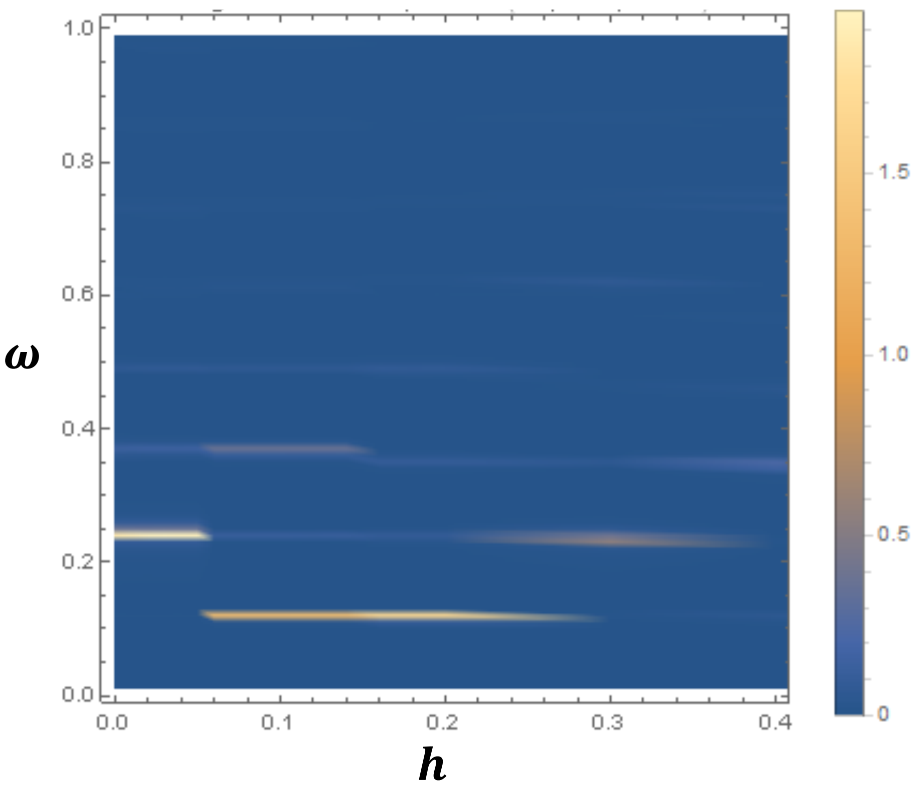
$$H = J \sum_j \mathbf{S}_j \cdot \mathbf{S}_{j+1}$$

Magneto Raman Spectra from TEBD

No defects



Two defects



Raman operator bond profile probes $q \neq 0$ response

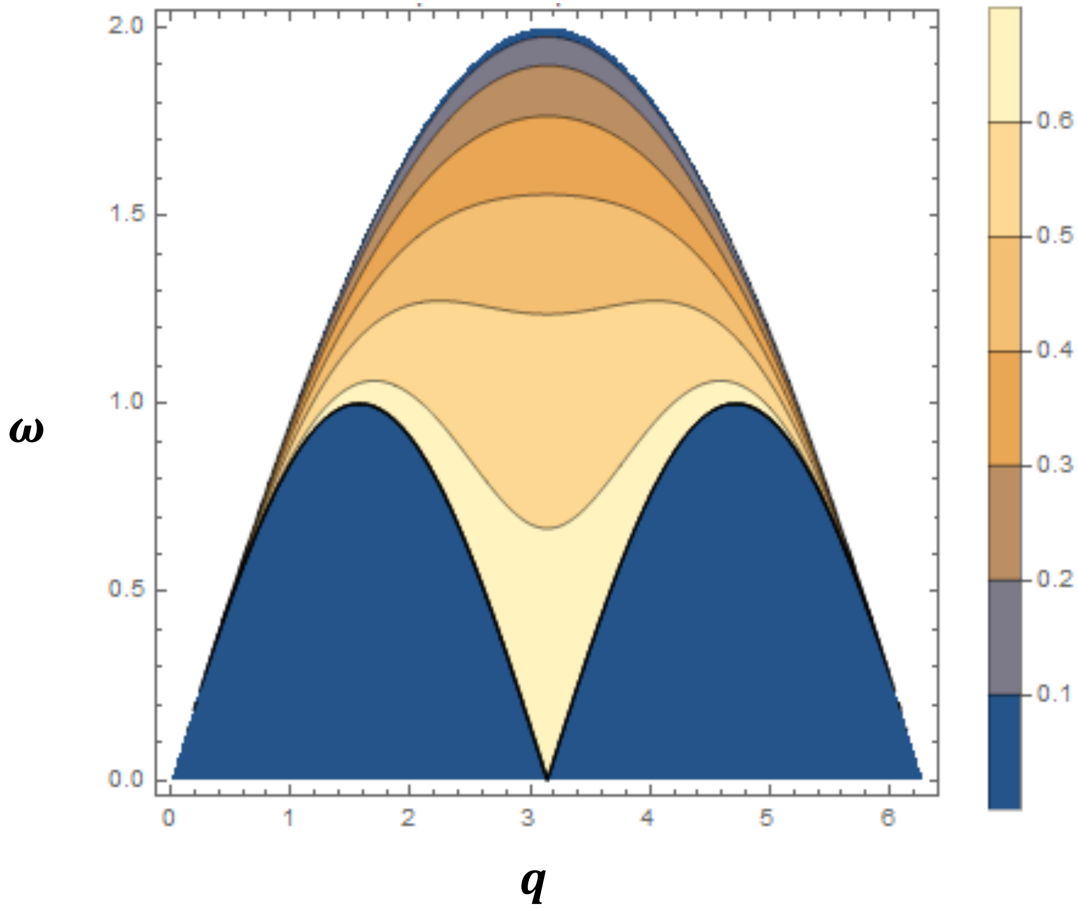
$$
\begin{aligned}
R &= \sum_{j} f(j)\mathbf{S}_j \cdot \mathbf{S}_{j+1} \\
&= \sum_{j,q} \tilde{f}_q \cos(qj)\mathbf{S}_j \cdot \mathbf{S}_{j+1} \\
&= \sum_{q} \tilde{f}_q \left(\sum_{j} \cos(qj)\mathbf{S}_j \cdot \mathbf{S}_{j+1} \right) \\
&= \sum_{q} \tilde{f}_q R_q
\end{aligned}
$$

Raman operator bond profile probes $q \neq 0$ response

$$\int dt\ e^{i\omega t}\langle R_q(t) R_{q'}(0)\rangle = \sum_q |\tilde{f}_q|^2 \boxed{\chi''(q,\omega)}$$

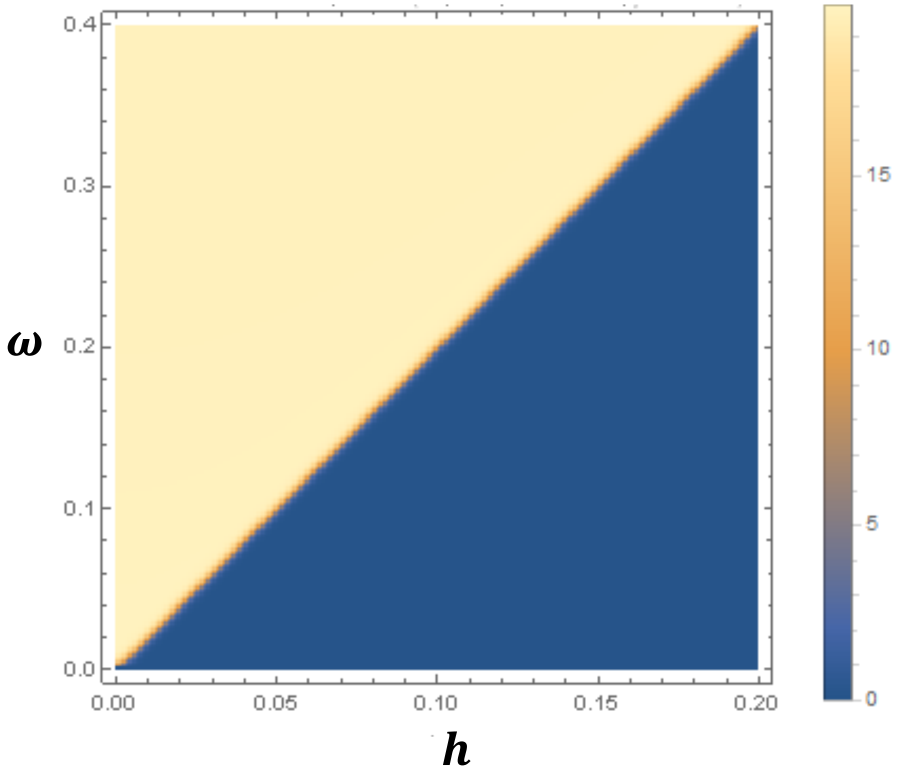
Mean Field: Free Spinon Response follows $\boxed{\chi_{\rho\rho}''}$

$$H = \sum_k -t\cos(k) c_k^\dagger c_k$$

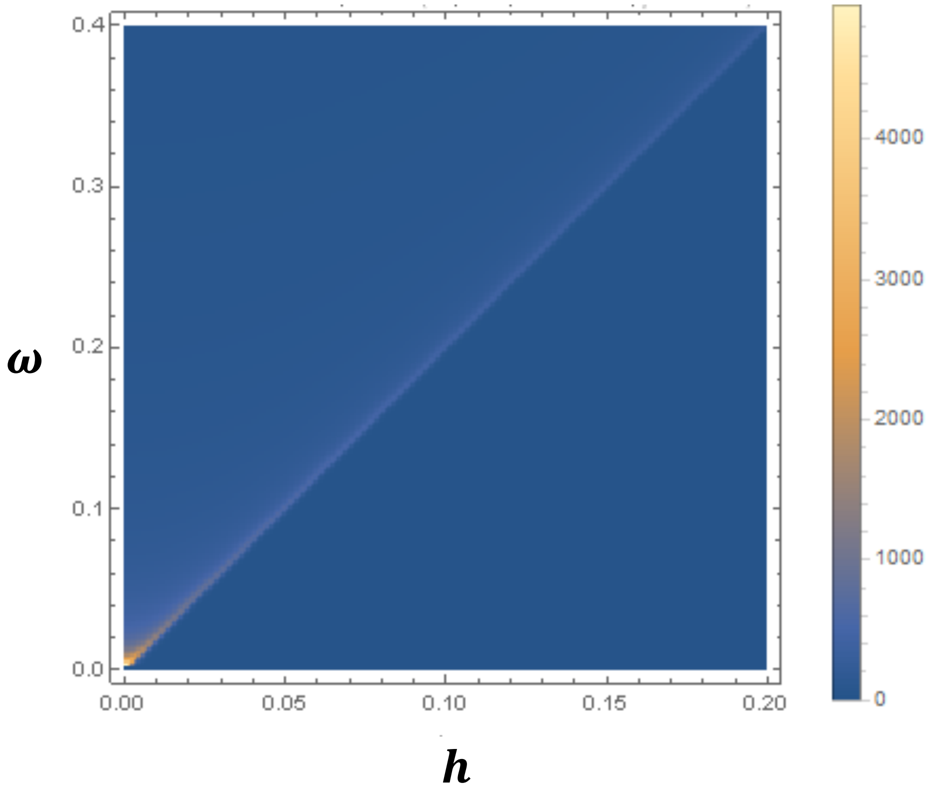


Bosonized density-density response (no defects)

Free Spinons (K = 1)



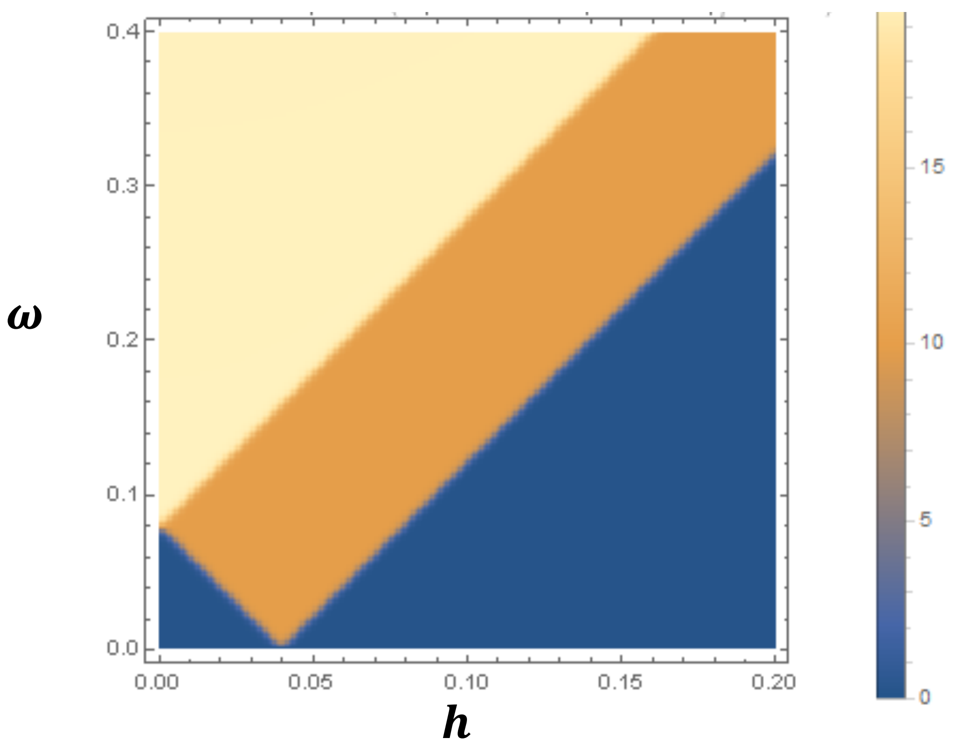
Heisenberg (K = 1/2)



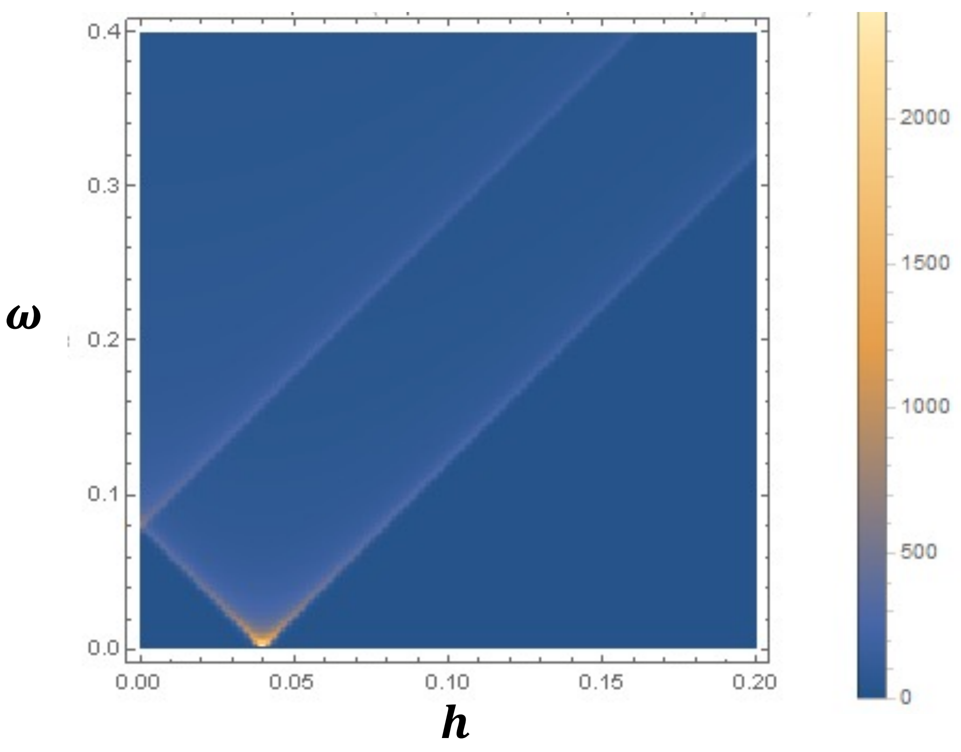
Bosonization: Giamarchi appendix. Also Sato, Katsura, Nagaosa PRL ‘12

Bosonized density-density response (two defects approximated by $\widetilde{f_q} = \delta_{q,\pi\pm\frac{2\pi}{L}}$)

Free Spinons (K = 1)



Heisenberg (K = 1/2)



Summary & Questions

- $Ba_4Ir_3O_{10}$
    - S = 1/2 Mott insulator
    - Magneto Raman suggests quantum (spin) liquid behavior with gapless spin excitations
    - Spinons capture observations; unclear what mechanism produces them
- Topological defects beyond the Hamiltonian
    - Present in Raman response
    - Induced by crystal dislocations
    - Probe $q \neq 0$ response

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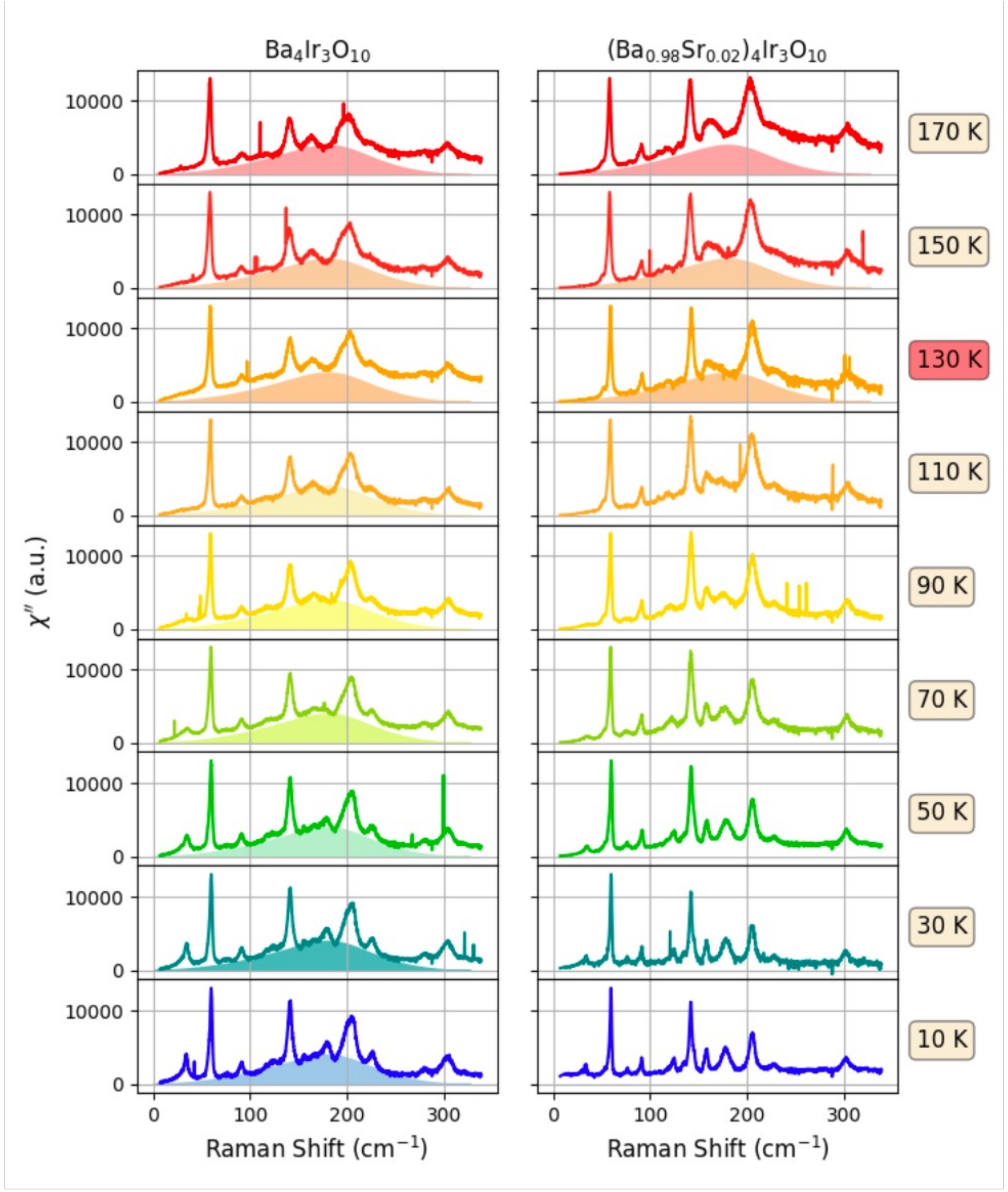
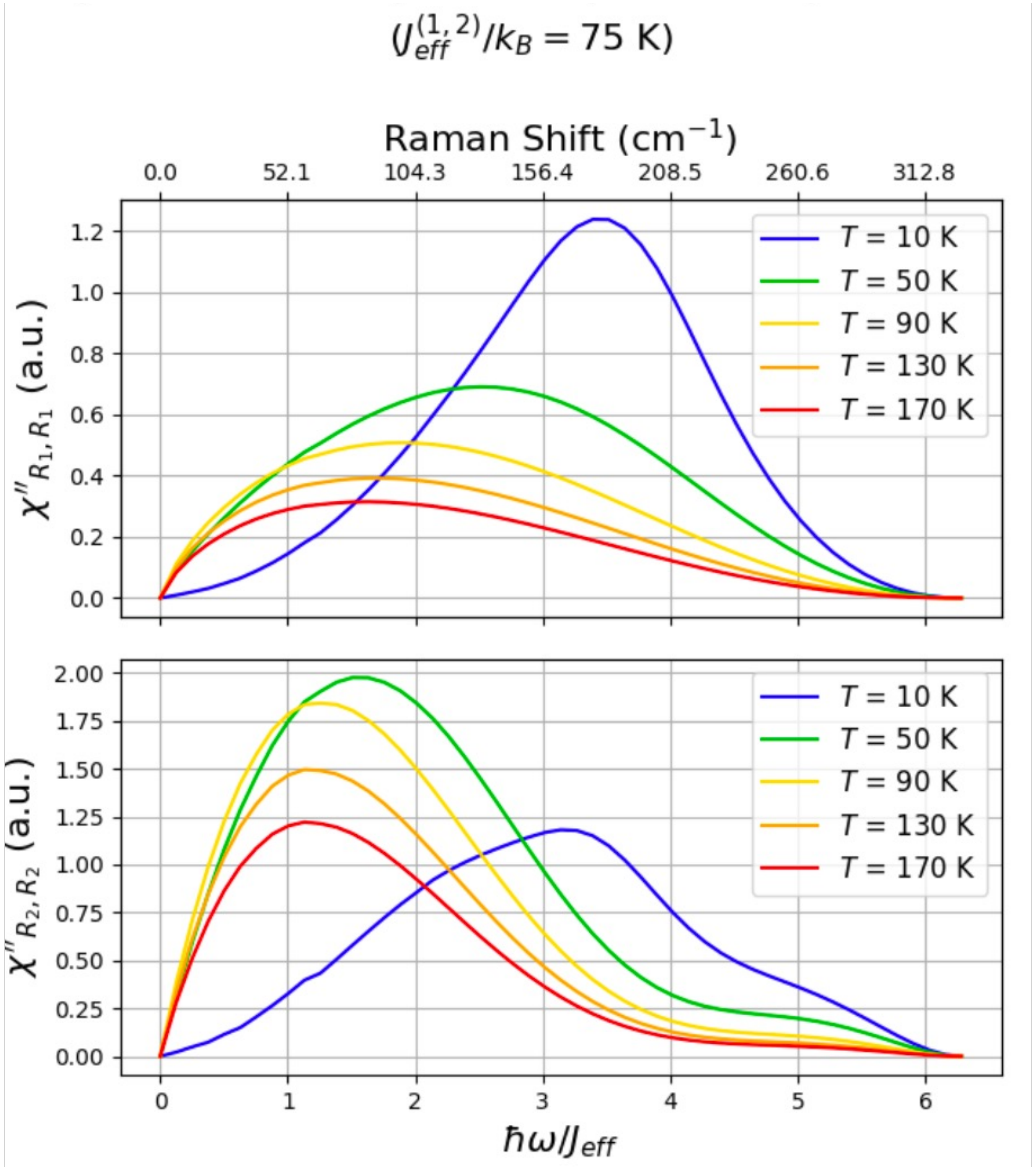
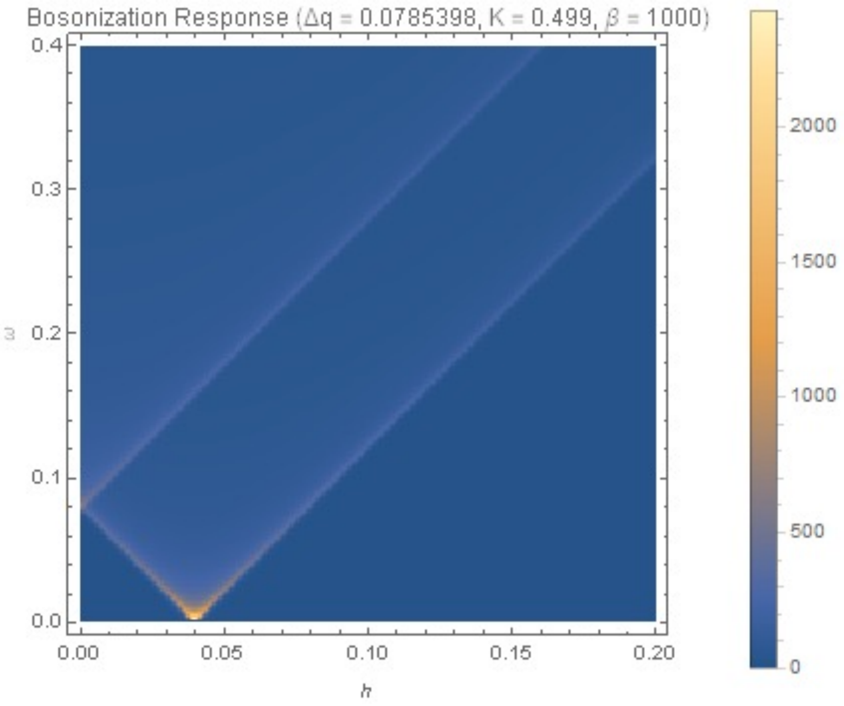


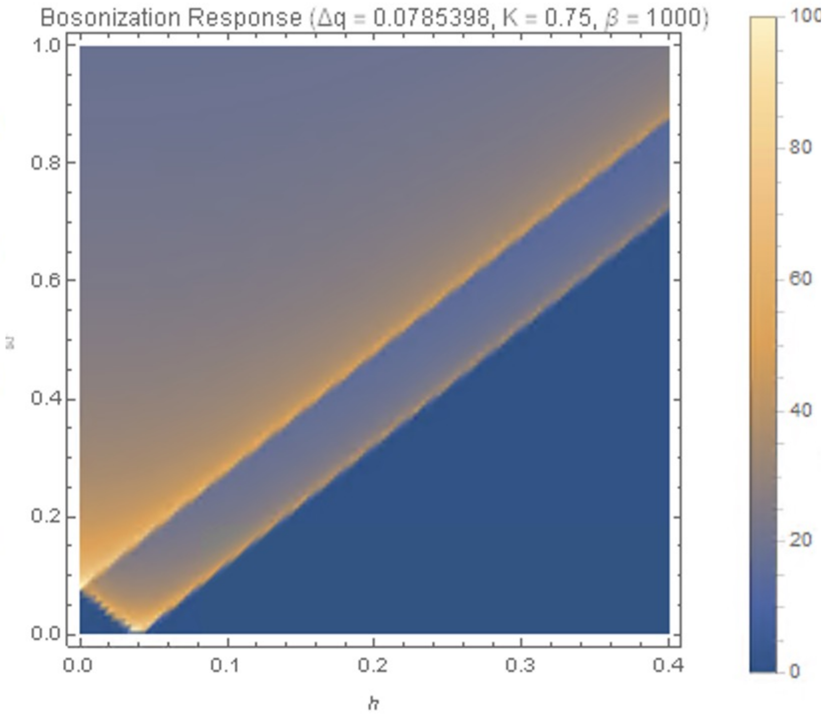
Temperature dependence of $Ba_4Ir_3O_{10}$ Raman susceptibility



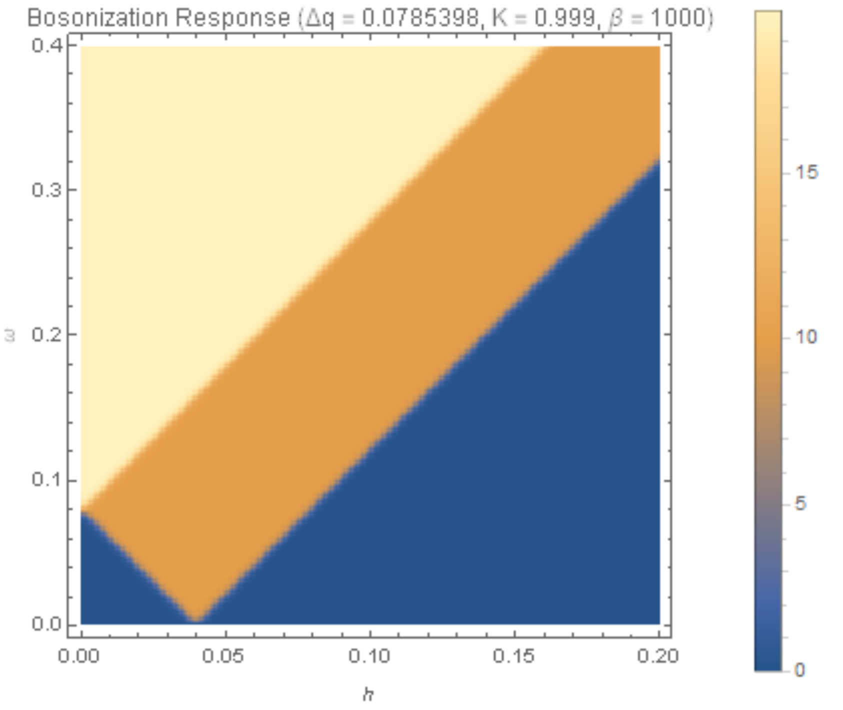
Mean field temperature dependence


Bosonization Response (Δq = 0.0785398, K = 0.499, β = 1000)
ω
0.0
0.1
0.2
0.3
0.4
h
0.00
0.05
0.10
0.15
0.20
0
500
1000
1500
2000

K = 1/2


Bosonization Response (Δq = 0.0785398, K = 0.75, β = 1000)
ω
0.0
0.2
0.4
0.6
0.8
1.0
h
0.0
0.1
0.2
0.3
0.4
0
20
40
60
80
100

K = 3/4


Bosonization Response (Δq = 0.0785398, K = 0.999, β = 1000)
ω
0.0
0.1
0.2
0.3
0.4
h
0.00
0.05
0.10
0.15
0.20
0
5
10
15

K = 1