Topological Defects in a Response Theory

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Raman Scattering of Quantum Liquid Candidate $Ba_4Ir_3O_{10}$

Raman scattering has been utilized to demonstrate signatures for fractionalized spinon excitations in quantum liquid candidate $Ba_4Ir_3O_{10}$ [4]. This material is a 2D layered spin- 1/2 magnet featuring 1D zigzag chains coupled via trimers (Fig. 1a). For incident and scattered photon polarizations aligned orthogonal to the chain axis, the inelastic Raman scattering spectrum shows phonon peaks superposed on a broad hump (Fig. 1b). This broad hump is fruitfully captured by a 4-spinon continuum from two equivalent mean field theories (Fig. 1d). In the presence of non-magnetic disorder, the hump disappears, and phonon linewidths narrow (Fig. 1c) which indicate the fragile quantum liquid state is no longer present.

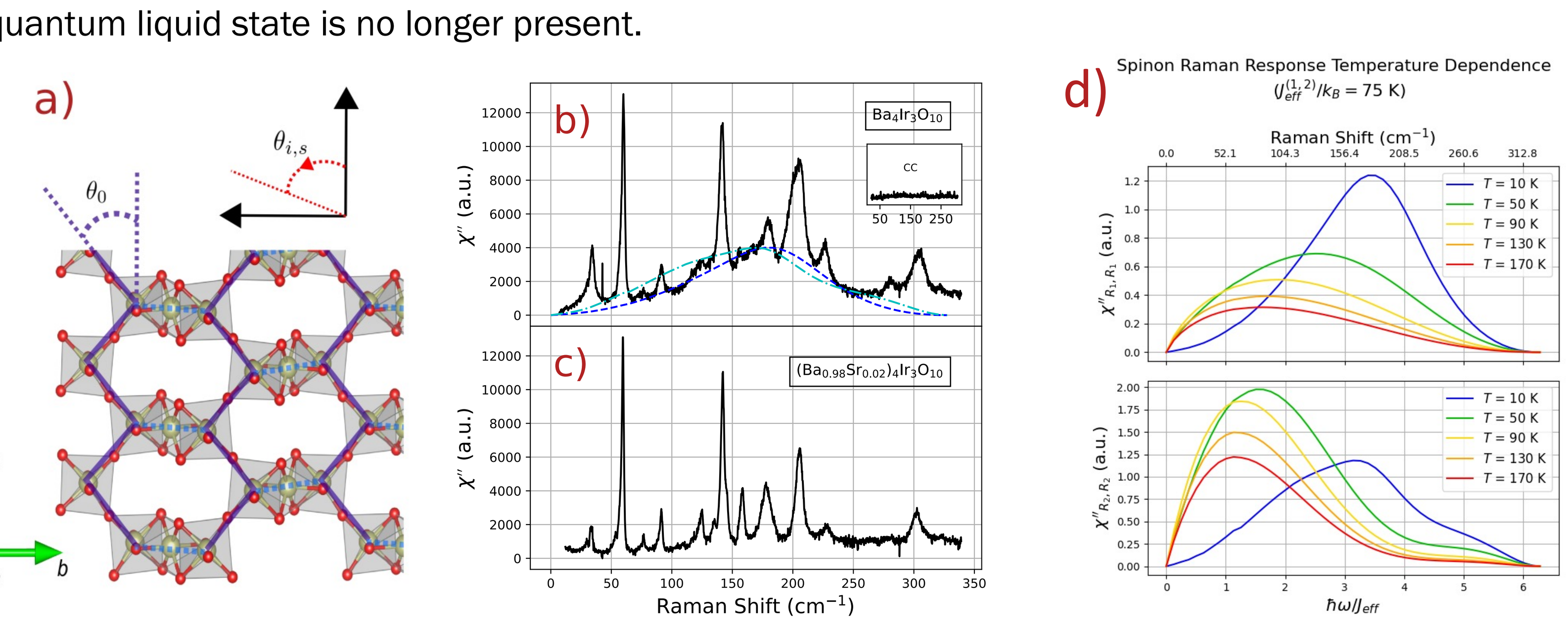


Fig. 1 Raman spectra of quantum liquid $Ba_4Ir_3O_{10}$
(a) Crystal structure of $Ba_4Ir_3O_{10}$
(b) Raman scattering spectra in bb photon polarization featuring broadened phonon peaks on top of a broad hump, which is captured well by a four-spinon continuum in quantum liquid mean field theories (dashed, blue and dotted-dashed cyan)
(c) Spectra for sister compound with non-magnetic disorder shows narrower phonon linewidths and absence of broad hump
(d) Spinon Raman response computed within two equivalent mean field choices at $T = 10, 50, 90, 130,$ and 170 K.

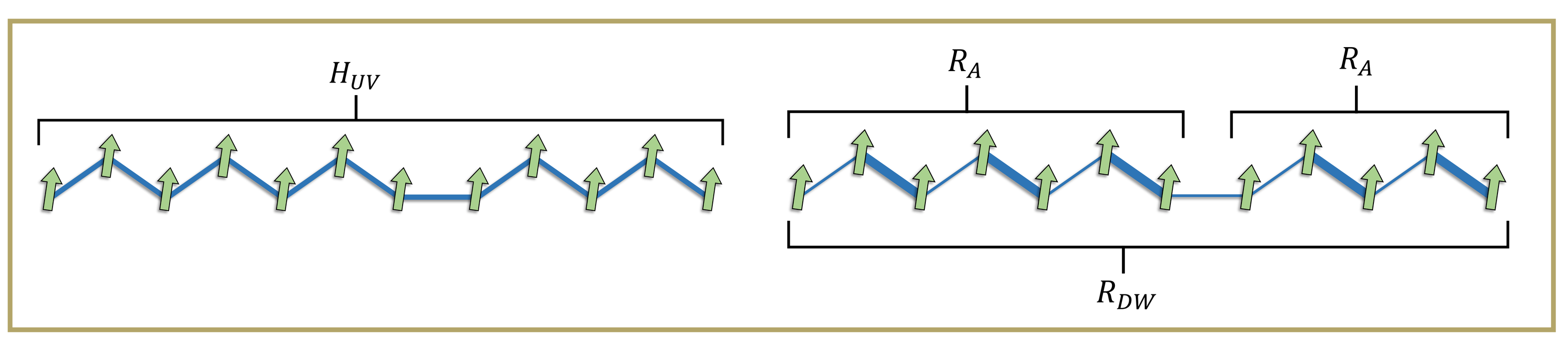
Raman Scattering Probes Magnetism and Geometry

The dynamical response of a system is often specified by its Hamiltonian H and ground state $|\psi\rangle$. A counterexample to this rule of thumb is the inelastic Raman scattering spectrum $I(\omega)$:

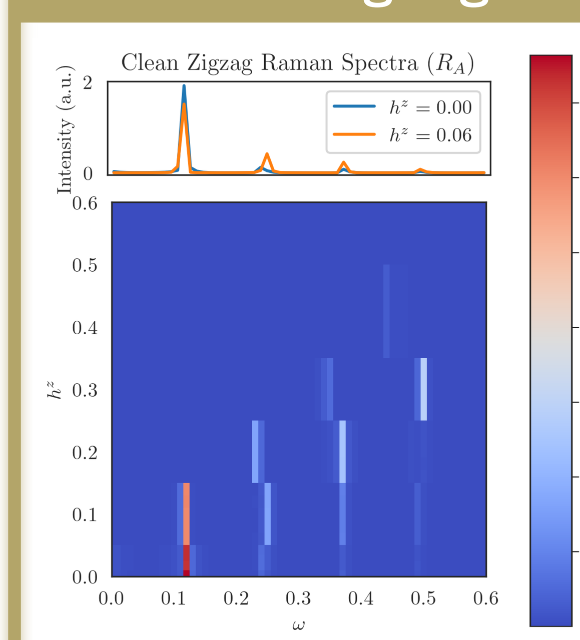
$$I(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\, e^{i\omega t}\langle R(t)R(0)\rangle_0$$

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

Above, $\hat{\boldsymbol{e}}_{i,s}$ is the incident (scattered) photon polarization. $I(\omega)$ is given by the ground state expectation value (denoted by $\langle\cdots\rangle_0$) of the dynamical correlation function of the Raman operator R [1]. Here, $I(\omega)$ is specified by *three* objects: H, $|\psi\rangle$, and R [2]. Generally, $A(\boldsymbol{r}_{12})$ is difficult to determine, but ratios of A on different bonds are of the order of the ratio of the respective exchange couplings [1]. The Raman response of a system is specified by a *third* non-trivial input, R, which considers photon polarizations that necessarily couple to spatial degrees of freedom. Due to this polarization factor, the Raman operator inherits a rich structure from the spatial geometry of the system. This structure endows R with symmetries that are different from the Hamiltonian. As a proof of principle of this rich structure, we may consider Raman scattering of the zigzag chain.



Clean Zigzags: Gapless Magneto-Raman Response



Consider an effective spin-1/2 zigzag chain with effective 1D nearest-neighbor antiferromagnetic Heisenberg interactions in an applied magnetic field

$$H_0 = J\sum_j \boldsymbol{S}_j\cdot\boldsymbol{S}_{j+1} - h^z\sum_j S_j^z$$

For complementary photon polarizations, the Raman operator for this system is given by an alternating (dimerization) operator

$$R_A \propto \sum_j (-1)^j(\boldsymbol{S}_j\cdot\boldsymbol{S}_{j+1})$$

Fig. 2: (Top) Cartoon depiction of clean zigzag crystal lattice from which R_A may arise. (Left) The corresponding numerically computed inelastic Raman scattering spectra for finite sized clean zigzag spin-1/2 chains ($L = 80$ sites, open boundary conditions) with zero domain walls. Spectral weight shifts linearly with field above $h^z = 0$. [3]

Zigzag Domains Walls: Gap Closes in Applied Field

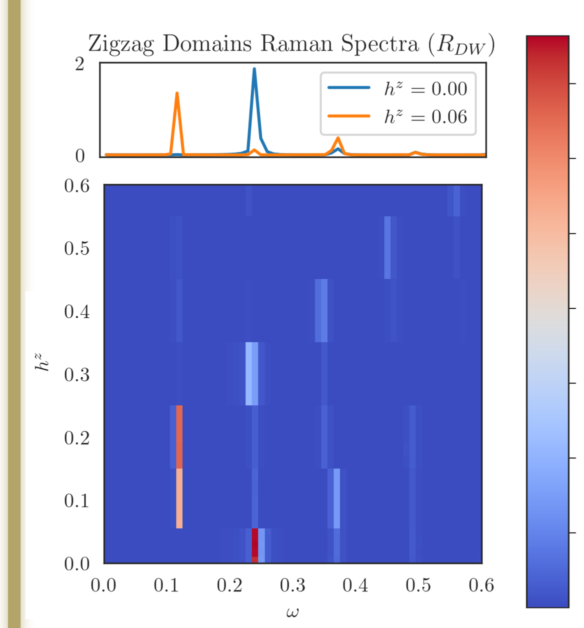


Fig. 3: (Top) Cartoon depiction of clean zigzag crystal lattice from which R_{DW} may arise. (Left) The corresponding numerically computed inelastic Raman scattering spectra for finite sized zigzag spin with two domain walls. At zero field, the presence of domain walls shifts the finite size gap to higher energy. This shift persists up to a small critical field h_c. Spectral weight shifts linearly with field above $h^z = h_c$. [3]

At zero applied field ($h^z = 0$), the mean field Raman spectrum is a *gapped* 2-particle continuum that qualitatively follows the line shape of the R_A spectrum. The gap occurs at a frequency $\omega_c = v_s\,(\pi/L_d)$ where L_d is size of the zigzag domain (unit lattice spacing) and v_s is the spinon velocity. At finite fields ($h^z > 0$), we find a more surprising result: at a small finite field $h_c = \frac{1}{2}\, v_s\,(\pi/L_d)$ the gap in the spectrum closes (Fig. 3).

The presence of the gap in the 2-particle continuum in addition to its closing at small finite fields is especially surprising since in computing the scattering spectrum, neither the Hamiltonian nor the ground state are altered. Indeed, the only difference in these responses is the presence of zigzag domain walls. Such domain walls are non-magnetic crystal defects, and naively one would not expect such a drastically different response in the presence of applied magnetic field.

Although this astonishing response to magnetic fields is well captured within the mean field theory, a valid concern is that the response is an artifact of the theory rather than a novel physical response. Using beyond mean field approaches, however, one can show that this is indeed a novel magnetic response rather than an artifact of the theory.

Zigzag Domain Walls have a $\mathbb{Z}_2$ Character

Zigzag domain walls not only possess an anomalous magnetic field response, but they are further characterized by a $\mathbb{Z}_2$ character. When the spatial separation between domain walls becomes small, the anomalous magnetic field response is no longer observed.

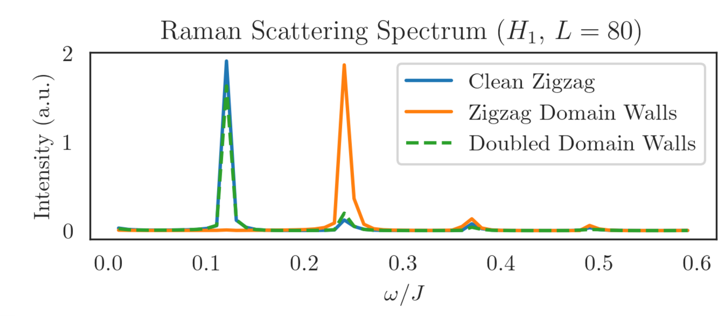


Fig. 4: Numerically computed inelastic Raman scattering spectra for a finite size ($L = 80$) zigzag chain with zero domain walls (solid, blue), two domain walls (solid, orange), and doubled domain walls (dashed, green). At zero applied magnetic field, doubled domain walls do not open the same gap as isolated domain walls. [3]

Raman Correlator

To capture the effects of defects in the Raman operator, we consider the Raman response of a general class of Raman operators R with couplings given by an arbitrary function of space g_j, $R = \sum_j g_j R_j$ with $R_j \equiv \boldsymbol{S}_j\cdot\boldsymbol{S}_{j+1}$. The operator R can be rewritten as a weighted sum of its Fourier modes as $R = \sum_q \tilde{g}_q R_q$ where $R_q = \sum_j e^{iqj}R_j$ where $\tilde{g}_q$ are the Fourier modes of g_j.

The Raman response of R is then given by weighted sums over the Raman correlation function $C_{RR}(q,\omega)$:

$$I(\omega) = \sum_q |\tilde{g}_q|^2 C_{RR}(q,\omega), \quad C_{RR}(q,\omega) = \int dt\, e^{i\omega t}\langle R_q(t)R_{-q}(0)\rangle_0$$

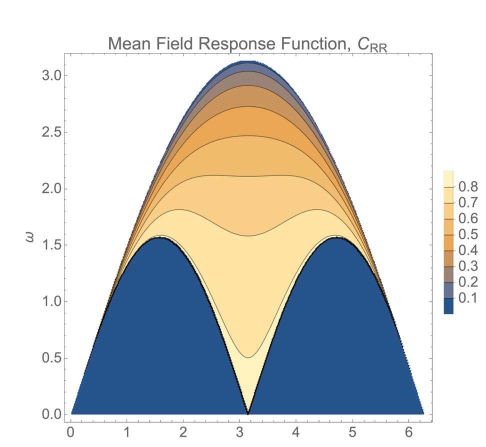


Fig. 5: Finite momentum Raman response for free spinons, $C_{RR}(q,\omega)$. At the 2-particle mean field level, the Raman spectrum is a weighted sum over C_{RR}. The Raman response of the clean zigzag spin chain is a cut at $q = \pi$. In the presence of defects, the Raman spectrum contains cuts at Fourier modes of the disorder profile $q \neq \pi$. For domains of size $L/3$ in a finite chain of size L, the most significant contributions come from $q = \pi \pm 3\pi/L$. [3]

Singular Magnetic Response of Non-magnetic Defects Indicates Presence of Spinon Liquid

Our mean field free spinon picture approach can also be seen as a limiting case of a bosonized theory with Luttinger parameter $K = 1$. To extend our results to interacting Hamiltonians, we also consider $K \neq 1$. The gap associated with domain walls persists at all K.

The anomalous response of non-magnetic topological defects to applied fields indicates the presence of a spinon liquid. We find that bosonic magnon excitations, unlike fermionic spinon excitations, lack the anomalous, singular magnetic field response presented here. Whereas an applied magnetic field shifts the Fermi momentum of a spinon Fermi surface, and may tune gapped excitations to become gapless, bosonic magnon excitations do not experience such an effect. Within linear spin wave theory, we find 1D magnons lack an anomalous magnetic field response probed by Raman scattering.

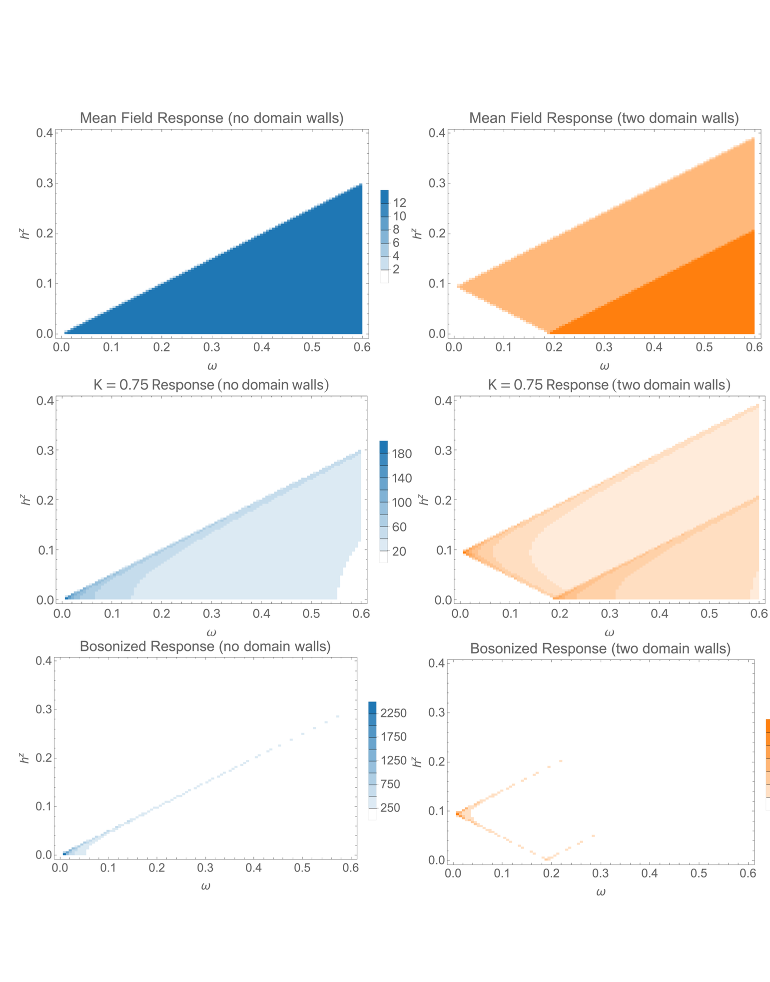


Fig. 6: (Top to bottom) Raman responses from mean field and from the low energy effective theory at Luttinger parameters $K = 0.75, 0.5$ corresponding to XXZ and Heisenberg spin models respectively. The clean zigzag case (left, blue) shows gapless excitations at zero field and a gap which opens in applied field. In the presence of defects (right, orange), a gap opens at zero field. In applied field, however, this gap closes. To best compare with our finite size ($L = 80$) numerics, we show the spectra when $q = \pi \pm 3\pi/80$ is probed. [3]

References

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[4] Sokolik, A. ***et al.*** Spinons and damped phonons in the spin-1/2 quantum liquid $Ba_4Ir_3O_{10}$ observed by Raman scattering. *Phys. Rev. B* **106**, 075108 (2022).