Inelastic Light scattering and the correlated metal-insulator transition

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Raman scattering probes electronic excitations

- Inelastic scattering of light with electron-hole excitations of the correlated many-body system.
- Use of polarizers for the incident and reflected light allows one to select different symmetries of the electron-hole excitations.
- Signal depends on the Raman scattering amplitude $\gamma(k)$. We consider three different symmetries here:
  - $A_{1g}$: $\gamma(k) \sim \cos(k_x a) + \cos(k_y a)$
  - $B_{1g}$: $\gamma(k) \sim \cos(k_x a) - \cos(k_y a)$
  - $B_{2g}$: $\gamma(k) \sim \sin(k_x a) \sin(k_y a)$
Experimental data for Kondo insulators

- Nyhus et al, PRB 95 Raman scattering on FeSi. Note the appearance of the isosbestic point below about 150K.
- The low frequency spectral weight is **reduced** and the higher frequency weight is **enhanced** as the temperature is lowered.

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Experimental data for intermediate-valence materials

- Nyhus et al, 1995 and 1997 Raman scattering on \( \text{SmB}_6 \).
  Note the appearance of the isosbestic point near 300 cm\(^{-1}\).
- Below 30K, there is an increase in low frequency spectral weight in a narrow peak at about 130 cm\(^{-1}\).

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Summary of Experimental Data (Raman)

- Three characteristic behaviors are seen: (i) as $T$ is lowered, there is a **redistribution of spectral weight** from low-frequency to high frequency; (ii) these regions are separated by an isosbestic point, where the **Raman response is independent** of $T$; (iii) the ratio of the twice spectral range where spectral weight is depleted to the onset temperature, where it first is reduced, is **much larger than 3.5** (typically 10-30).

- For correlated insulators this behavior is “universal” in the sense that it **does not depend** on the microscopic properties of the insulating phase, be it a Kondo insulator or an intermediate-valence material.
Resonant Inelastic X-ray Scattering probes momentum and energy dependent charge excitations

• By tuning the photon energy to the K or L\textsubscript{3} edge of a core state, one finds large enhancements to the inelastic scattering.

• Advanced light sources have linearly polarized light, but experiments to date have not used polarizers on the detectors. Hence different symmetry channels are mixed together in the experimental results.

• The scattered signal depends on the Raman scattering amplitude $\gamma(k+q/2)$ for transferred momentum $q$.

• The energy resolution in current experiments is poor (about 0.1 ev) but is expected to improve dramatically with second-generation experiments (less than 20 meV).

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RIXS on CaCu$_2$O$_2$Cl$_2$

Experimental data on a Mott insulator shows a **broad charge-transfer peak** and a **dispersive low-energy peak**.

We label the transferred momentum by the parameter $X(q)=[\cos q_x + \cos q_y]/2$. When plotted in this fashion, the dispersion along the zone diagonal and zone center is **similar**.

The difference for $X=1$ along the different zone axes occurs due to the relation between the polarization vector and $q$, which **differs** for the different directions.

Summary of Experimental Data (RIXS)

- RIXS experiments on correlated insulators typically show two features—-(i) a large-weight charge-transfer peak and (ii) a lower-energy peak. The charge transfer peak shows little dispersion through the Brillouin zone, while the lower-energy peak does disperse. The dispersion from the zone center to zone corner is usually about twice the dispersion from the zone center to the zone edge boundary.

- Experimental results project onto different weights of the different symmetry channels due to a locking of the photon momentum direction to the polarization of the electric field.

- Systematic changes in temperature have not been carried out yet.

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Spinless Falicov-Kimball Model

\[ H = - \frac{t}{2\sqrt{d}} \sum_{\langle i, j \rangle} c_i^\dagger c_j + E \sum_i w_i + U \sum_i c_i^\dagger c_i w_i \]

- \textit{exactly solvable model} on a hypercubic lattice in infinite dimensions using dynamical mean field theory.
- possesses homogeneous, commensurate/incommensurate CDW phases, phase segregation, and \textbf{metal-insulator transitions}.
- \textit{Raman response can be constructed formally exactly}.

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Formal Solution for the Raman Scattering Response

$A_{1g}$ channel

- This channel has the **full symmetry** of the lattice
- The Raman response contains **resonant, mixed** and **nonresonant** terms. (We consider only the **nonresonant** terms here).
- The irreducible charge vertex for the Falicov-Kimball model is a **simple function** of the electronic self energy and Green’s function (Shvaika, Physica C, 2000; Freericks and Miller, PRB, 2000). This is **not true for the Hubbard model**.
- The **nonresonant** Raman response can be determined **exactly** by properly solving the relevant Dyson equations.
- We illustrate how to solve this problem using **Feynman diagrams**.

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Diagrams for the $A_{1g}$ Raman response

\[ \gamma(k) - \gamma(k') = -T \gamma(k) \]

\[ \gamma(k) = -\varepsilon(k), \quad \Gamma \text{ is local and has no } k\text{-dependence} \]

Solving these coupled equations allows for the full nonresonant Raman response to be determined.
Formal Solution for the Raman Scattering Response

\textbf{B}_{1g} \text{ channel}

- This channel is \textit{orthogonal} to the lattice.
- There are \textit{no vertex corrections} (Khurana, PRL, 1990), so the Raman response is represented by the \textit{bare bubble}.
- This response is \textit{identical} to that of the optical conductivity multiplied by one factor of frequency (Shastry and Shraiman, PRL, 1990).
- \textbf{Resonant} Raman scattering is possible in this channel, but won’t be analyzed here.

\textit{The nonresonant B}_{1g} \text{ Raman response is closely related to the optical conductivity.}
Formal Solution for Raman the Scattering Response

\textbf{B}_2^g \text{ channel}

- The Raman scattering amplitude vanishes for nearest neighbor hopping on a hypercubic lattice, so there are \textbf{no nonresonant or mixed responses}.
- The square of the current operator does contain \textbf{B}_2^g \text{ symmetry}, so \textbf{pure resonant processes are possible}.
- \textbf{Vertex corrections} are needed, but are relatively simple to handle.
- We don’t discuss this channel further here.

\textit{B}_2^g \text{ Raman scattering is purely resonant.}

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Metal-Insulator transition

- Correlation-induced gap drives the single-particle DOS to zero at $U=1.5$
- Interacting DOS is independent of $T$ in DMFT (Van Dongen, PRB, 1992)
- Examine Raman response through the $(T=0)$ quantum phase transition.

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Nonresonant Raman Response ($U=2$)

- The low-frequency $B_{1g}$ response develops at a low temperature over a wide frequency range of $O(1)$.

- An isosbestic point divides where spectral weight increases or decreases as $T$ is lowered ($B_{1g}$).

J. K. Freericks, Georgetown University, Raman scattering talk, 2002
Inelastic X-ray scattering ($B_{1g}$)

- Scattering of x-rays allows the photon to exchange both momentum and energy with the electron-hole excitations.

- We see a broadening and dispersion of the peaks, but the same anomalous low-energy behavior and the isosbestic point.

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Inelastic X-ray scattering ($A_{1g}$)

- Here the results at finite-$q$ differ greatly from $q=0$: all of the anomalies appear away from $q=0$!
- A reduced broadening and dispersion of the peaks is seen; but the same anomalous low-energy behavior and the isosbestic points recur for nonzero $q$.

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Summary (Falicov-Kimball model)

• The theoretical results are qualitatively similar to experimental results measured in correlated systems.

• The nonresonant $B_{1g}$ channel displays (i) an isosbestic point that divides the regions where the Raman response increase or decrease as $T$ is lowered; (ii) a sharp depletion of spectral weight in the low-frequency region as $T$ is reduced; and (iii) the temperature where low-frequency spectral features appear is much lower than the range in frequency over which those features appear.

• Results for the Raman scattering are model independent on the insulating side of the MIT.

• Vertex corrections suppress all nontrivial behavior for the $A_{1g}$ channel at $q=0$ only.

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Hubbard Model

\[ H = -\frac{t}{2\sqrt{d}} \sum c_{i\sigma}^* c_{j\sigma} + U \sum n_{i\uparrow} n_{i\downarrow} \]

Both electrons are now mobile

• Exactly solvable model on a hypercubic lattice in infinite dimensions using dynamical mean field theory (but requires NRG calculations to extract real frequency information).

• The irreducible charge vertex is problematic to calculate because it possesses too large a dynamic range for the max-ent techniques.

• Hence, the Raman response can be constructed formally exactly for the nonresonant B_{1g} channel only.

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Nonresonant $B_{1g}$ Raman scattering ($n=1, U=2.1$)

- Note the charge transfer peak as well as the Fermi liquid peak at low energy. As $T$ goes to zero, the Fermi peak sharpens and moves to lower energy, as expected.

- There is no low energy and low-$T$ isosbestic point, rather a high frequency isosbestic point seems to develop.

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Nonresonant $B_{1g}$ Raman scattering ($n=1, U=3.5$)

- This is quite anomalous! A MIT occurs as a function of $T$. Note the appearance of the low-$T$ isosbestic point.
- The low energy Raman response has rich behavior, with a number of low energy peaks developing at low-$T$, but the low energy weight increases as $T$ decreases here.

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Here we see the expected **universal behavior** for the insulator---the low-energy spectral weight is **depleted** as $T$ goes to zero and an **isosbestic point** appears.

- The temperature dependence here is over a **wider range** than for the FK model due to the $T$-dependence of the interacting DOS.
Inelastic X-ray scattering (B$_{1g}$, zone diagonal)

- Nonresonant scattering for a **correlated metal**, a system that undergoes a **metal-insulator transition**, and a **correlated insulator**.
- Note how the Fermi peak **broadens** and remains away from $\omega=0$ as $q$ increases.
- In the MIT case, the scattering results depend **weakly** on $T$.
- For the insulator, the results are quite similar, except for some **broadening**, as one moves through the Brillouin zone.

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Summary Hubbard model

- The Fermi-liquid behavior introduces new features to the $B_{1g}$ Raman response: there is a characteristic Drude like feature that develops at the lowest frequencies (with a width that decreases like $T^2$).

- New behavior occurs on the metallic side of the MIT, where the low-energy spectral weight increases as $T$ decreases and where additional structure is seen, as the system undergoes a temperature-driven insulator-metal transition.

- In the insulating phase we see the expected “universal behavior,” but the temperature dependence is slower here, because the interacting DOS is also $T$-dependent.
Conclusions

• Showed how an exact solution for nonresonant Raman scattering can be constructed for a system that passes through a metal-insulator transition. The solutions displayed both an isosbestic point and a rapid increase in low-frequency spectral weight near the quantum-critical point, just as seen in experiment.

• Results are model independent or “universal” on the insulating side of the metal-insulator transition, explaining why so many different correlated insulators show similar behavior.

• Found the presence of a low frequency Drude peak in the Fermi-liquid metals.

• Showed interesting universal features are to be expected with inelastic x-ray scattering as well.

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