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:
  - $A_{1g}$: $\gamma(k) \sim \cos(k_x a) + \cos(k_y a)$
  - $B_{1g}$: $\gamma(k) \sim \cos(k_x a) - \cos(k_y a)$
  - $B_{2g}$: $\gamma(k) \sim \sin(k_x a) \sin(k_y a)$ [vanishes for nn hopping]

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J. K. Freericks, Georgetown University, Raman scattering talk, 2003
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.

J. K. Freericks, Georgetown University, Raman scattering talk, 2003
Experimental data for intermediate-valence materials

- Nyhus et al, 1995 and 1997 Raman scattering on 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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Experimental data for high Tc superconductors

- *Venturini et al.* PRL 2002, Raman scattering on BSCO as a function of doping at constant temperature (180 K).

- Note how the $B_{1g}$ and $B_{2g}$ results agree in the overdoped regime, but they differ as the system becomes more underdoped (and hence more correlated).

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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 or a high $T_c$ superconductor.

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Resonant Inelastic X-ray Scattering probes momentum and energy dependent charge excitations

• By tuning the photon energy to the K or L₃ 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 (crossed) 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 show a **broad charge-transfer peak** and a **dispersive low-energy peak**.

We label the transferred momentum by the parameter $X(q) = \frac{\cos q_x + \cos q_y}{2}$. 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 $\mathbf{q}$, which **differs** for the different directions.


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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.
Theories of inelastic light scattering

- The **insulating limit** has been analyzed by Chubukov and Frenkel (PRL, 1995).

- The **antiferromagnetically correlated metal** has been described by Devereaux and Kampf (PRB, 1999).

- Here we develop a theory that connects these two regimes and carries one through the **quantum critical point** of a metal-insulator transition.

- Experimental results exist for a variety of materials that pass through a **metal-insulator transition** as a function of doping.

- **Here we show how one can solve for Raman and inelastic X-ray scattering through a metal-insulator transition in both the Falicov-Kimball model and the Hubbard model.**

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Spinless Falicov-Kimball Model

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

• exactly solvable model on a hypercubic lattice in infinite dimensions using dynamical mean field theory.

• possesses homogeneous, commensurate/incommensurate CDW phases, phase segregation, and metal-insulator transitions.

• Inelastic light scattering can be constructed formally exactly.

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Formal Solution for the Light Scattering Response

$A_{1g}$ channel

- This channel has the **full symmetry** of the lattice
- The scattering response function 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** response can be determined **exactly** by properly solving the relevant Dyson equations.
- We illustrate how to solve this problem using **Feynman diagrams**.
Diagrams for the $A_{1g}$ response

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

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

$B_{1g}$ channel

- This channel is **orthogonal** to the lattice.
- There are **no vertex corrections** (Khurana, PRL, 1990), so the response is represented by the **bare bubble** (Raman response and X-ray response along the zone diagonal only).
- This Raman ($q=0$) response is **identical** to that of the optical conductivity multiplied by one factor of frequency (Shastry and Shraiman, PRL, 1990).
- **Resonant** scattering is possible in this channel, but won’t be analyzed in detail here.

The nonresonant $B_{1g}$ Raman response is closely related to the optical conductivity.
Formal Solution for the Light Scattering Response

**B₂g channel**

- The scattering amplitude vanishes for nearest neighbor hopping on a hypercubic lattice, so there are no nonresonant or mixed responses.
- The square of the current operator does contain B₂g symmetry, so pure resonant processes are possible.
- **Vertex corrections** are needed, but are relatively simple to handle.
- We describe how the resonant calculations can be performed in this channel.

*B₂g Raman scattering is purely resonant.*

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Diagrams for the $B_{2g}$ resonant Raman response

- In these diagrams, the vertex $v$ is the **velocity operator** $v(k) = \frac{d\varepsilon(k)}{dk}$ dotted into the photon polarization.
- These **coupled** Dyson equations must be solved together in order to get the resonant Raman response.

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Solving the many-body problem (FK model)

\[ G(\omega) = (1-w_1)G_0(\omega) + w_1 G_0(\omega)/[1+G_0(\omega)\Sigma(\omega)] \]

**Impurity solver**

\[ \Sigma(\omega) = G_0^{-1}(\omega) - G^{-1}(\omega) \]

**Self-energy**

**Effective Medium**

\[ G_0^{-1}(\omega) = G^{-1}(\omega) - \Sigma(\omega) \]

**Local Green’s function**

\[ G(\omega) = \int \frac{\rho(\varepsilon) d\varepsilon}{\omega + \mu - \Sigma(\omega) - \varepsilon} \]

DMFT algorithm is iterated until a self-consistent solution is achieved

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Metal-Insulator transition (NFL)

- Correlation-induced gap drives the single-particle DOS to zero at \( U=1.414 \)
- 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 (Constant T)

- The $A_{1g}$ response is suppressed at low frequencies, but the $B_{1g}$ response displays low-frequency spectral weight as one passes through the metal-insulator transition.

- Note the charge transfer peaks for large $U$. 

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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 ($B_{1g}$).

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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 ($B_{1g}$)

- General shape is the same as on the zone diagonal, even though the scattering is renormalized here.

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

• Once again we see a similarity on the zone edge and the zone diagonal.

• Note that at the X=-1, zone boundary, the result is independent of the symmetry.
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 inelastic light scattering are model independent on the insulating side of the MIT.
- Vertex corrections suppress all nontrivial behavior for the $A_{1g}$ channel at $q=0$. 

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Hubbard Model

\[ H = -\frac{t}{2\sqrt{d}} \sum c^*_i c_j + U \sum n_{i\uparrow} n_{i\downarrow} \]

- 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 max-ent techniques.

- Hence, the inelastic light scattering response can be constructed formally exactly for the nonresonant B_{1g} channel only (zone diagonal).

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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.
Nonresonant $B_{1g}$ Raman scattering ($n=1, U=4.2$)

- 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.

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Inelastic X-ray scattering ($B_{1g}$, zone diagonal)

- Nonresonant scattering for a **correlated metal**, at half filling and $U=2.12$.
- Note how the Fermi peak **broadens** and remains **away from** $\omega=0$ as $q$ **increases**.
- The response functions at finite momentum transfer are all quite **similar**.
- There is a **small dispersion** of the peak locations.

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Inelastic X-ray scattering ($B_{1g}$, zone diagonal)

- Nonresonant scattering for a **correlated metal**, at $n=0.8$ and $U=2.12$.
- The fermi peak is **even less developed** at finite-$q$.
- The charge transfer peak is **much reduced** as expected.
- There is still a **small dispersion** of the peak locations.

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Inelastic X-ray scattering (B$_{1g}$, zone diagonal)

- There is **no fermi peak** here because it is an insulator.
- Note how the main effect of finite-q scattering is to **broaden the charge transfer peak and shift it** to slightly higher energy.
- The isosbestic point **does not disperse** through the Brillouin zone.

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Inelastic X-ray scattering ($B_{1g}$, zone diagonal)

- Nonresonant scattering for a strongly correlated metal, at $n=0.8$ and $U=4.24$.
- Once again, the fermi peak is only seen at $q=0$.
- The temperature dependence of the low-energy scattering response is stronger, especially at the zone boundary.
- The high-energy features are essentially temperature and momentum independent.

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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 $T^2$). This low-energy spectral weight increases as $T$ decreases.

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

- When we transfer both momentum and energy from the photon, we find that the peaks are generically broadened, and there is no evolution of the fermi-peak.

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Conclusions

• Showed how an exact solution for nonresonant inelastic light 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 experimental Raman scattering.

• 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 fermi-liquid metals.

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

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