Inelastic light scattering near the Mott metal-insulator transition

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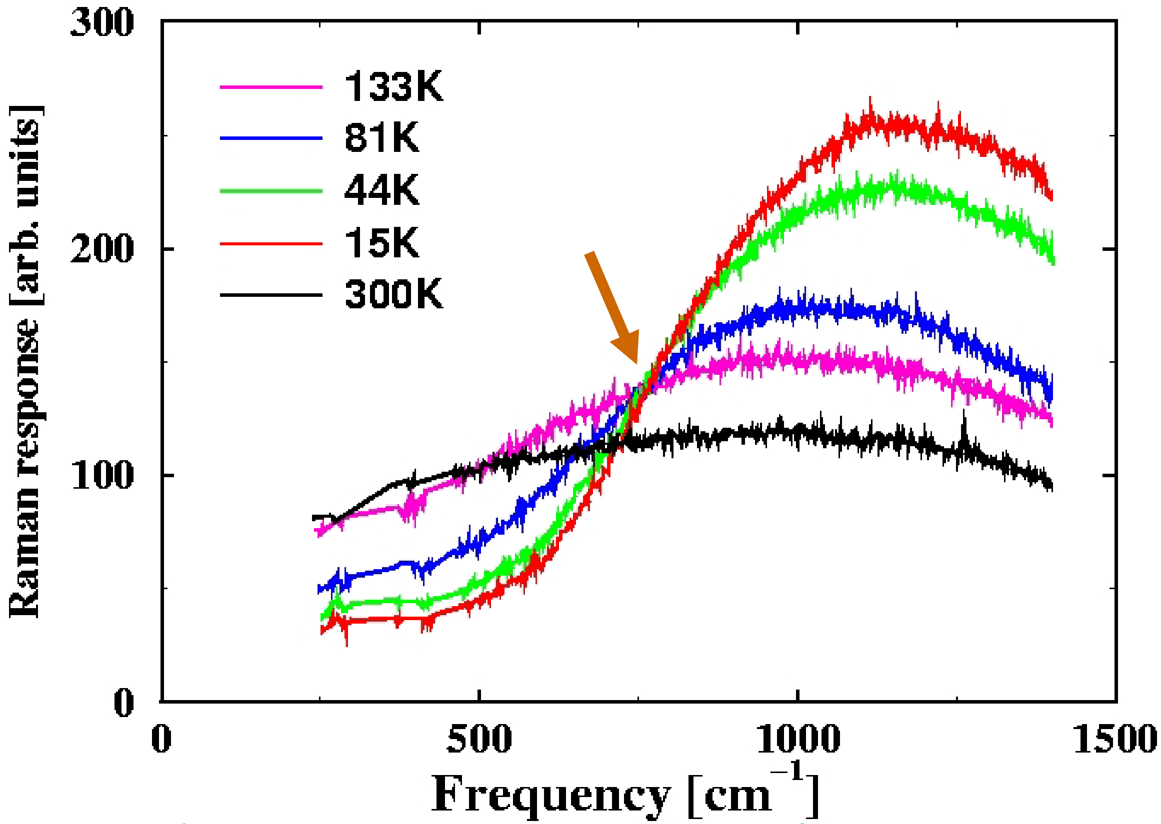
Raman scattering probes electronic excitations

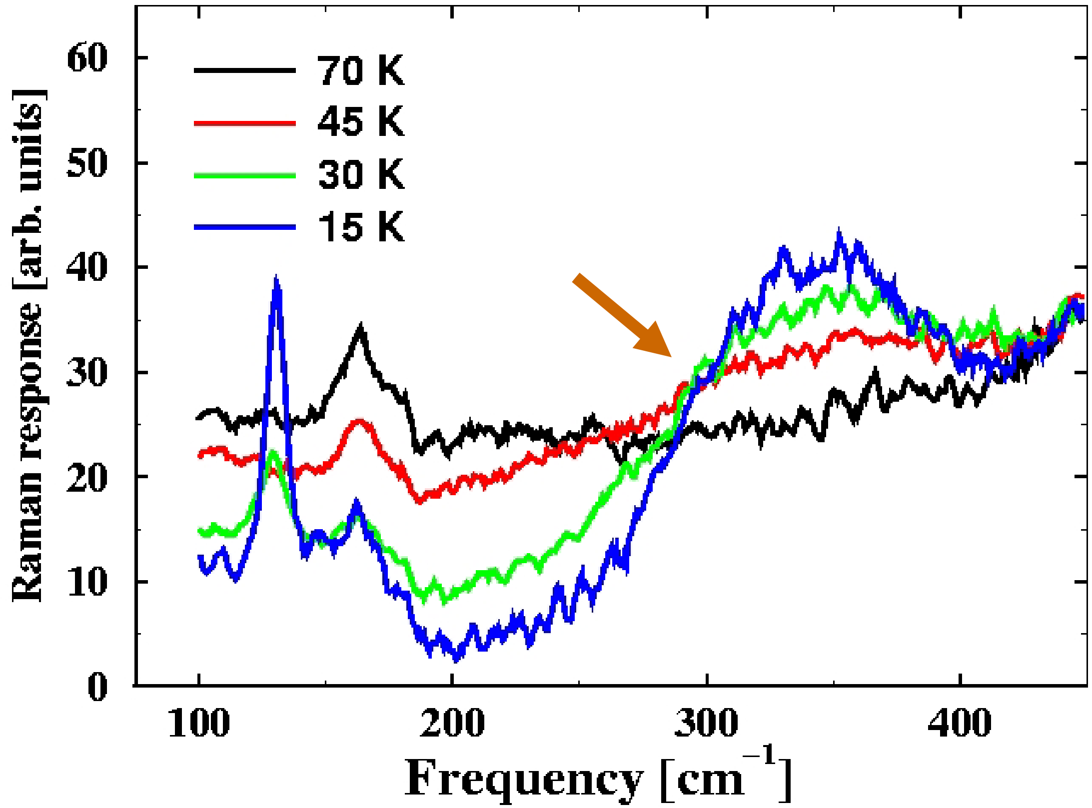
- **Inelastic scattering of light** with electron-hole excitations of the correlated many-body system.
- Use of polarizers for the incident and reflected light allows one to **select different symmetries** of the electron-hole excitations.
- Signal depends on the **Raman scattering amplitude γ(k)**. 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]

Experimental data for Kondo insulators



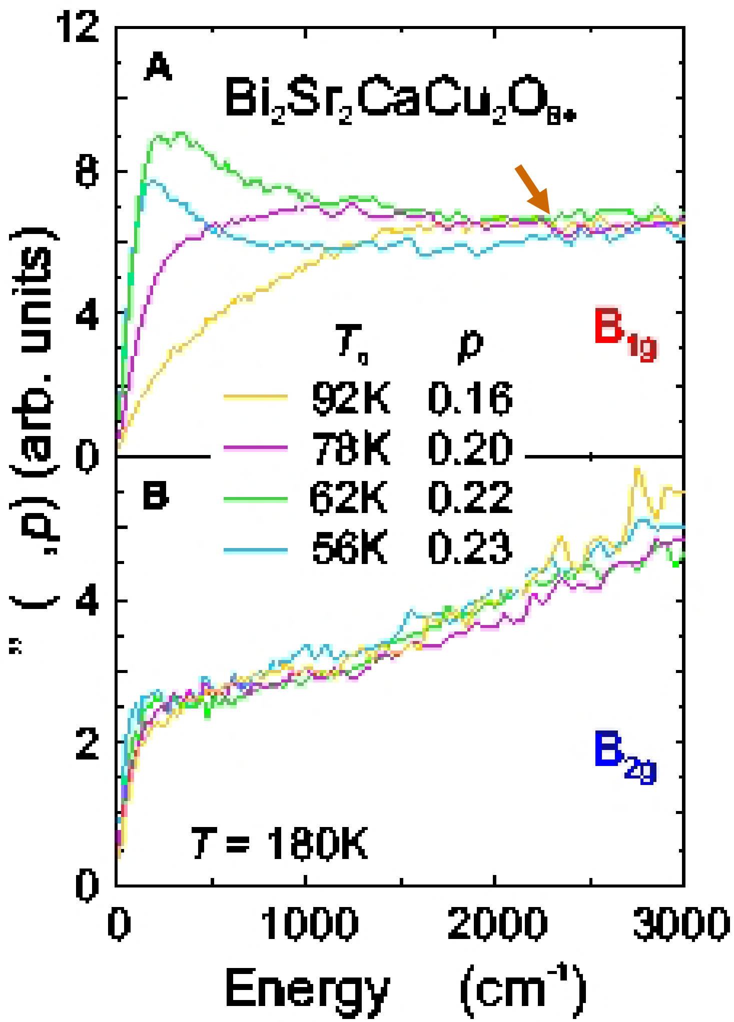
- *Nyhus et al, PRB 95* Raman scattering on **FeSi**. Note the appearance of the **isosbestic point** below about 150K.
- The low frequency spectral weight is **reduced** and the higher frequency weight is **enhanced** as the temperature is lowered.

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}.

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).

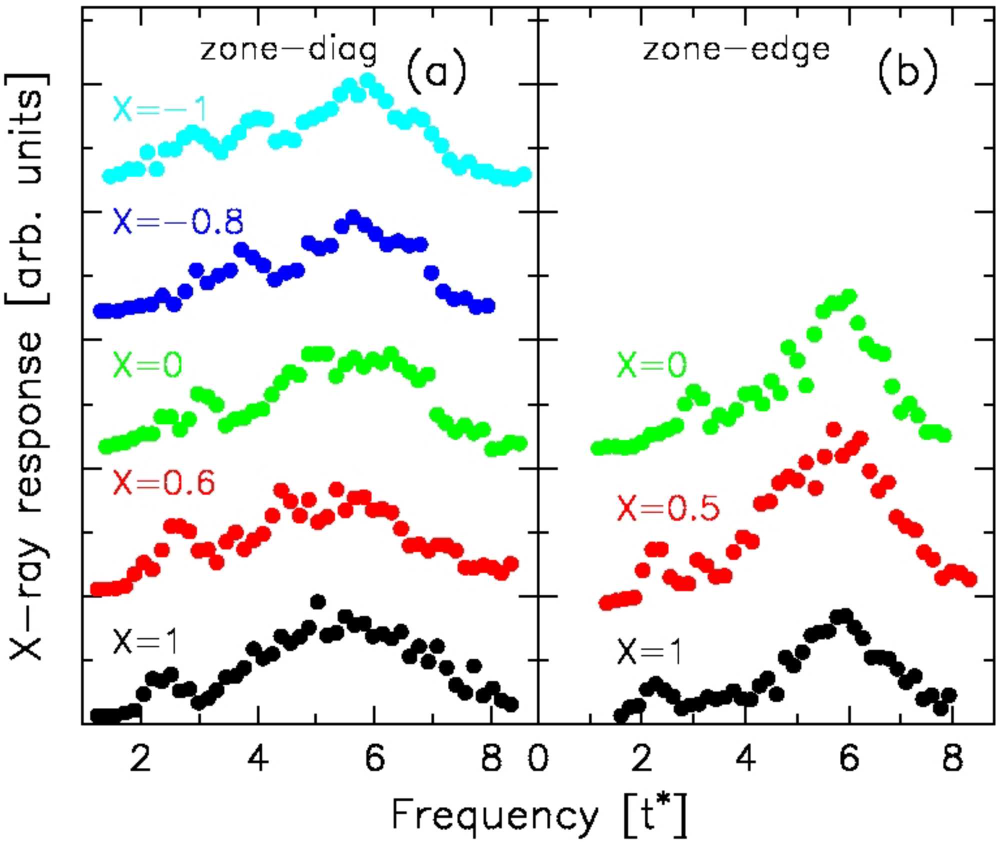
Summary of Experimental Data (Raman)

- 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 Tc superconductor.

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

- By **tuning** the photon energy to the K or L_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 (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 $\gamma(\mathbf{k}+\mathbf{q}/2)$** for transferred momentum **q**.
- The energy resolution in current experiments is poor **(about 0.1ev)** but is expected to improve dramatically with second-generation experiments **(less than 20 meV).**

RIXS on $CaCu_2O_2Cl_2$



RIXS data from Shen's group, Hasan et al., *Science* 2000.

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 $X(\mathbf{q})=[\cos q_x+\cos q_y]/2$. 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)

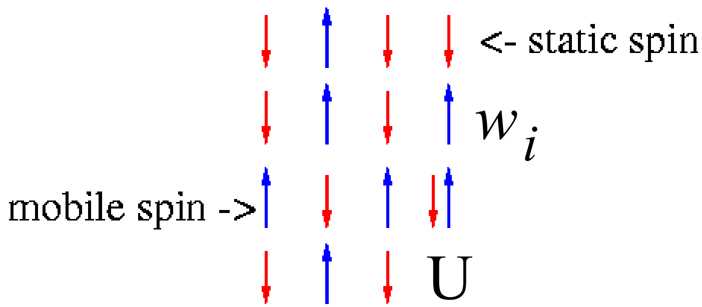
- RIXS experiments on correlated insulators typically show two features---(i) a **large-weight charge-transfer peak** and (ii) a **lower-energy peak**. The charge transfer peak shows **little dispersion** through the Brillouin zone, while the lower-energy peak **does disperse**. The dispersion from the 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.

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.***

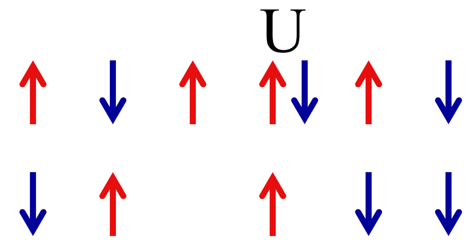
Spinless Falicov-Kimball Model

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



Hubbard Model

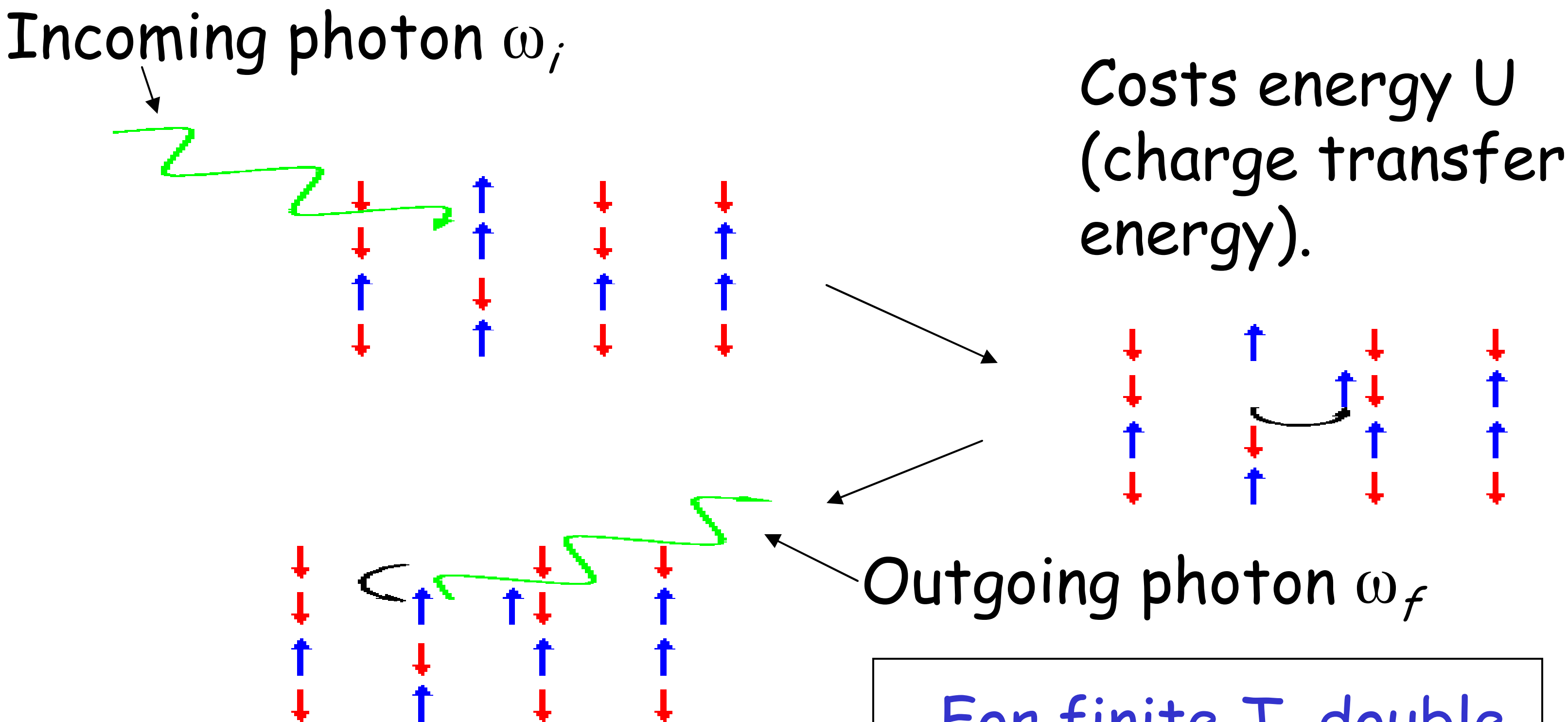
$$H = -\frac{t}{2\sqrt{d}}\sum c_{i\sigma}^{*}\ c_{j\sigma} + U\sum n_{i\uparrow} n_{i\downarrow}$$



Both electrons are now **mobile**

- **exactly solvable model** on a hypercubic lattice in infinite dimensions using dynamical mean field theory.
- possesses homogeneous, commensurate/incommensurate CDW and SDW phases, phase segregation, and **metal-insulator transitions**.
- ***Inelastic light scattering can be constructed formally exactly***.

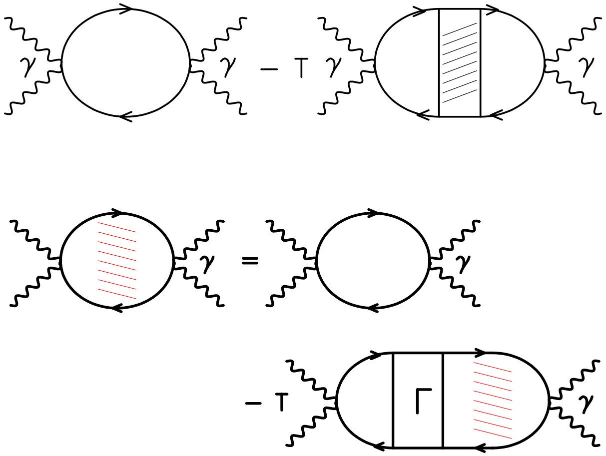
Light scattering processes



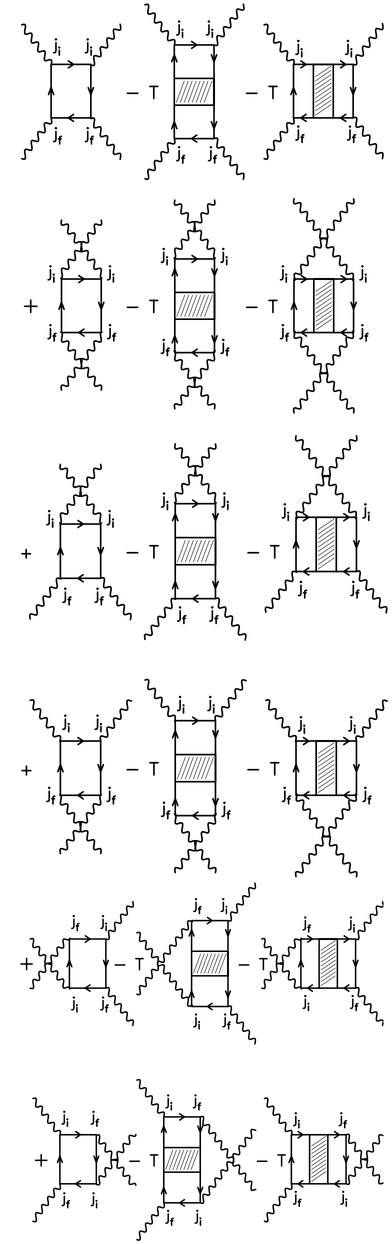
Electron hops, gains t.

For finite T, double occupancies lead to small band of low energy electrons.

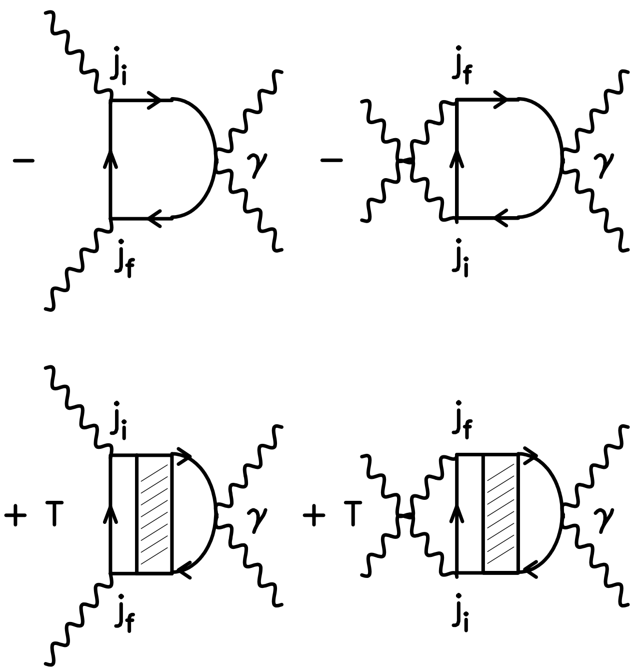
Diagrammatics



Nonresonant



Resonant

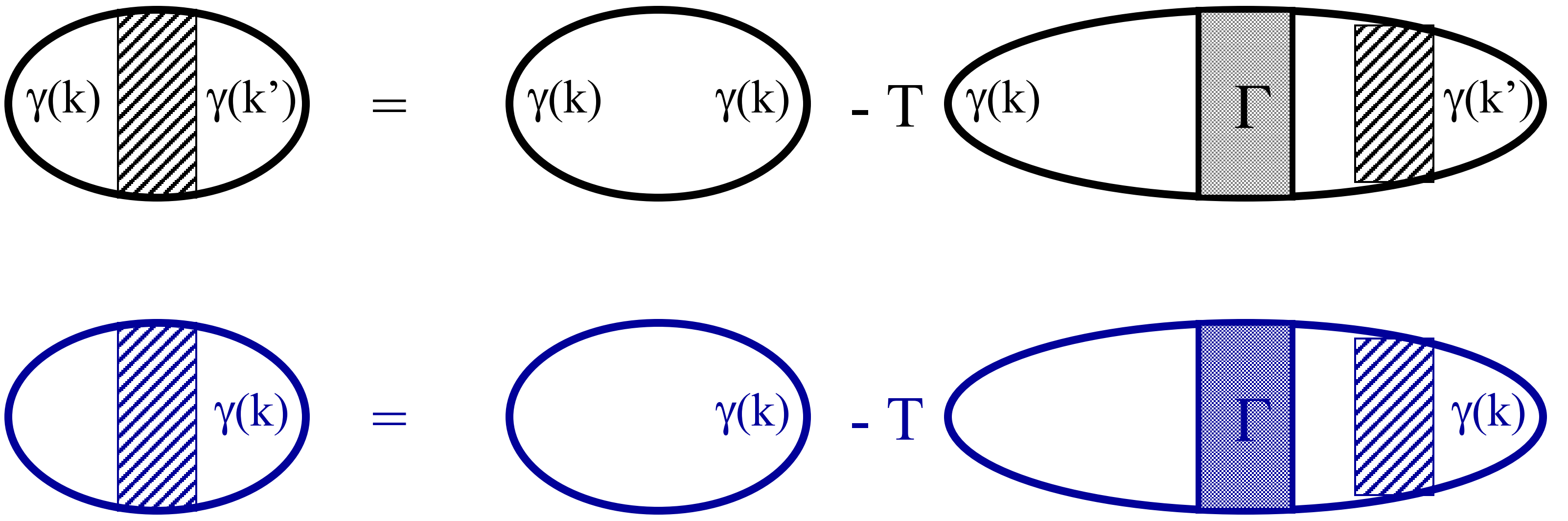


Mixed

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.
- 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, mixed, and resonant** responses can be determined **exactly** by properly solving the relevant Dyson equations.
- We schematically show how to solve this problem using schematic **Feynman diagrams**.

Diagrams for the nonresonant A_{1g} response



$\gamma(k) = -\varepsilon(k)$, Γ is **local** and has no k-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** but not **mixed** scattering is possible in this channel.

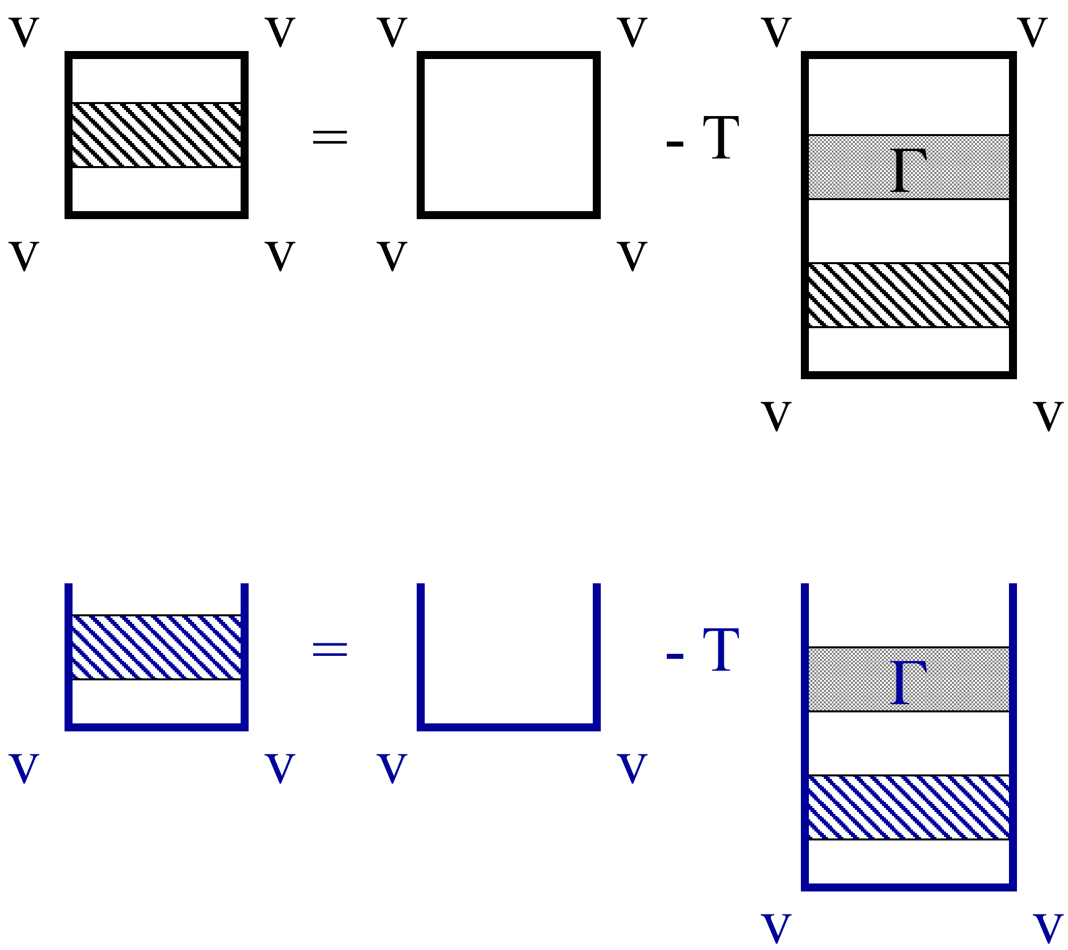
The nonresonant B_{1g} Raman response is closely related to the optical conductivity.

Formal Solution for the Light Scattering Response
B_{2g} 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 B_{2g} 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.

B_{2g} Raman scattering is purely resonant.

Diagrams for the B_{2g} resonant Raman response



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

Solving the many-body problem (FK model)

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

Impurity solver

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

Self-energy

Hilbert transform

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

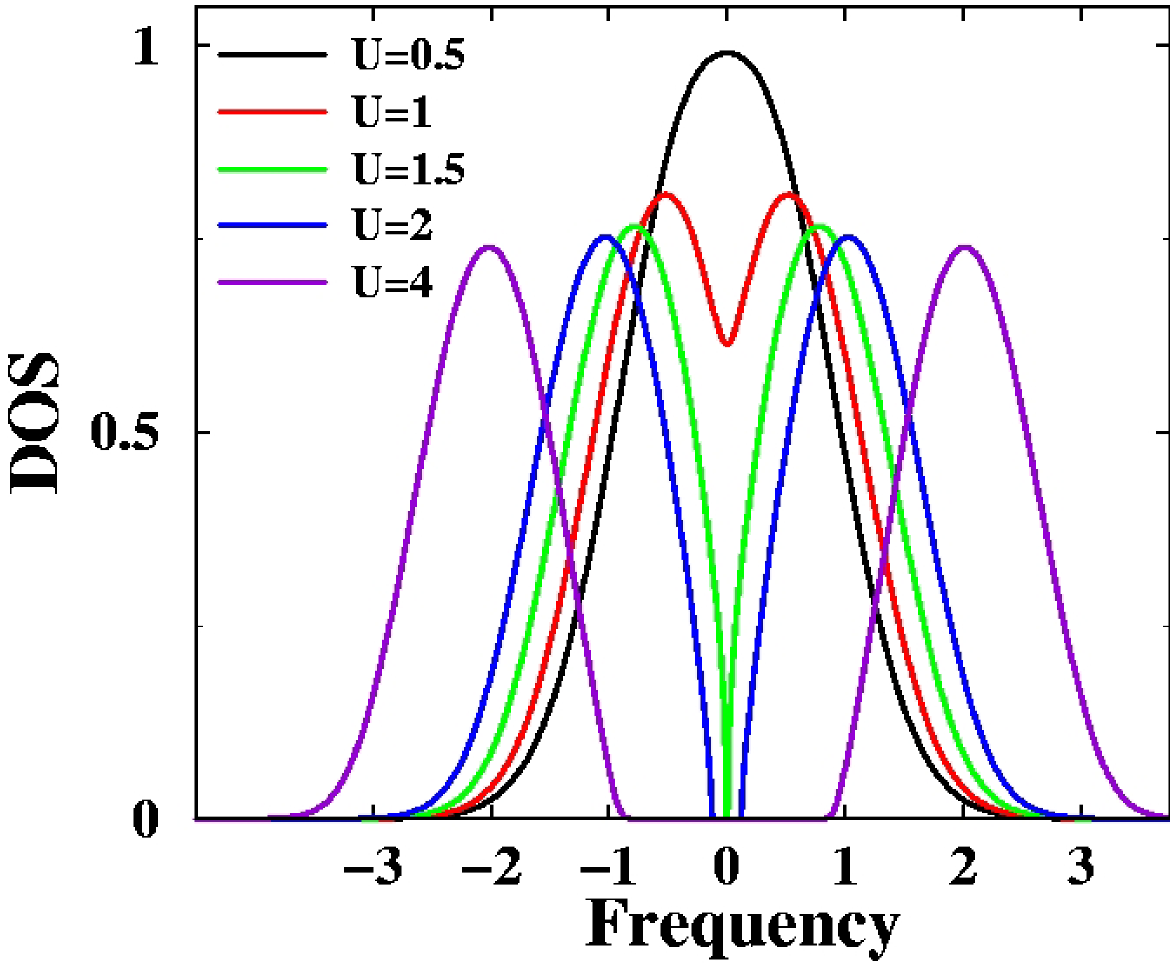
Local Green's function

$G_0^{-1}(\omega) = G^{-1}(\omega) + \Sigma(\omega)$

Effective Medium

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

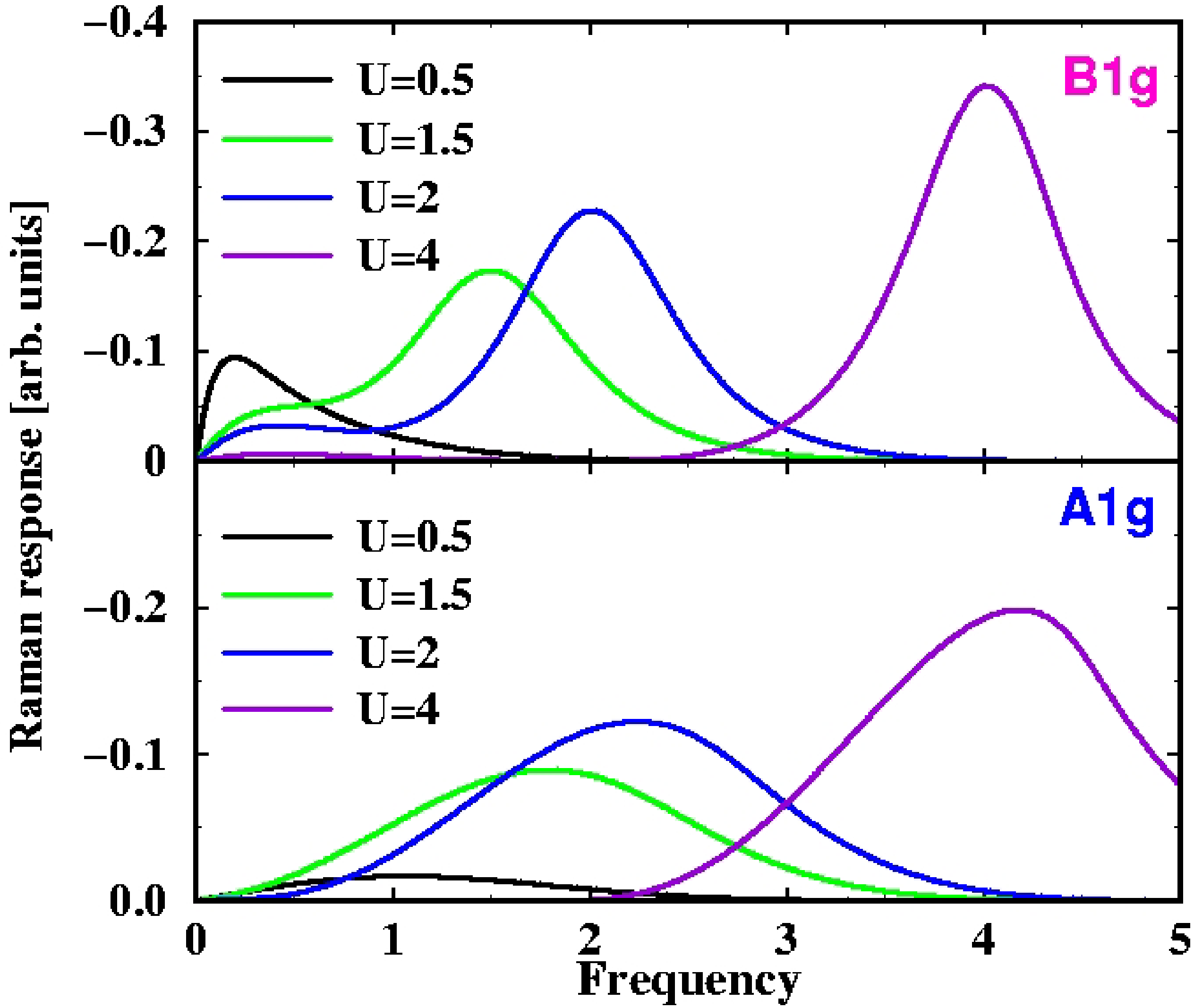
FK model Metal-Insulator transition (NFL)



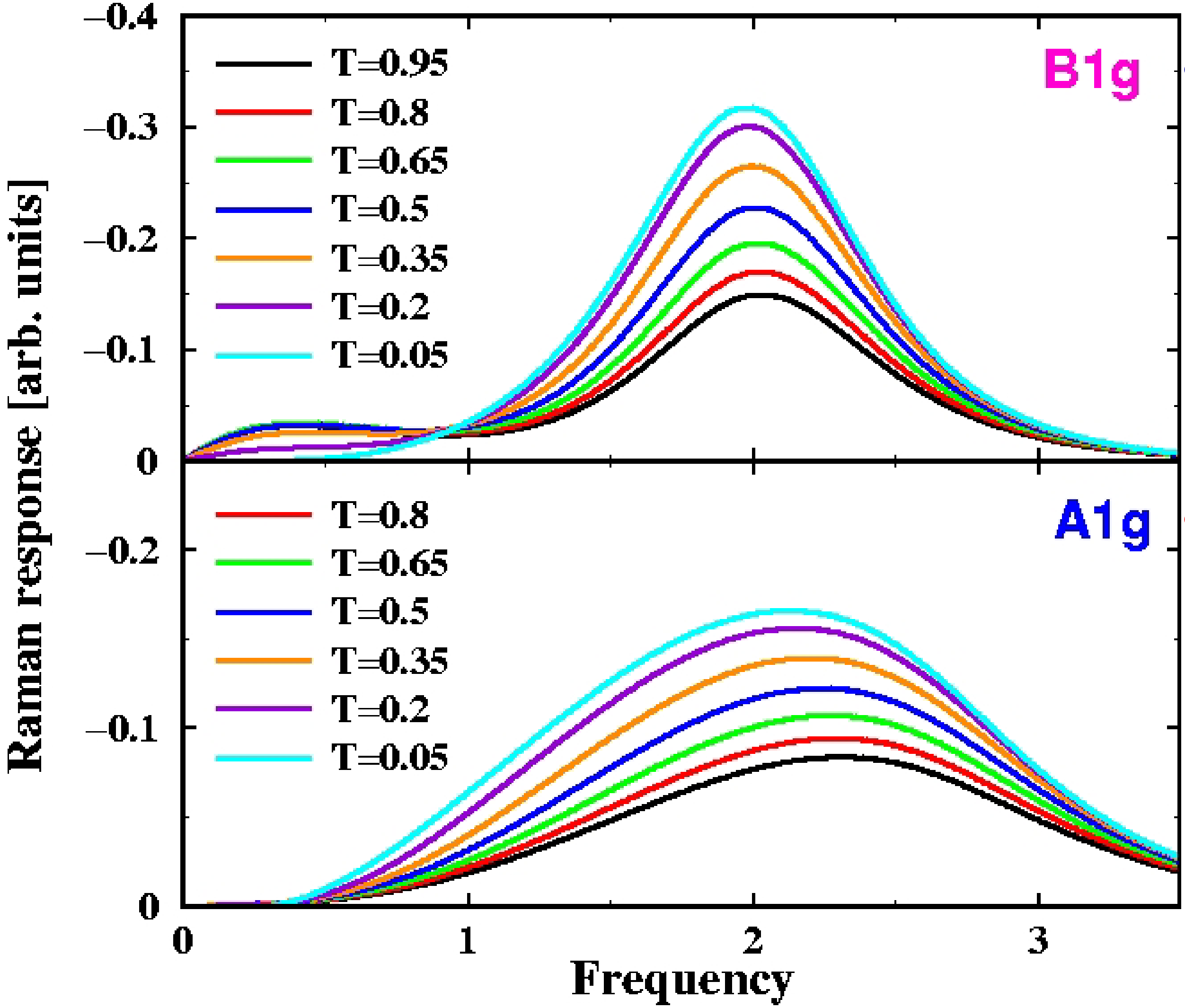
- **Correlation-induced** gap drives the single-particle DOS to zero at ω=0 for U>√2
- Interacting DOS is **independent of T** in DMFT (Van Dongen, PRB, 1992)
- ***Examine Raman response through the (T=0) quantum phase transition.***

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.

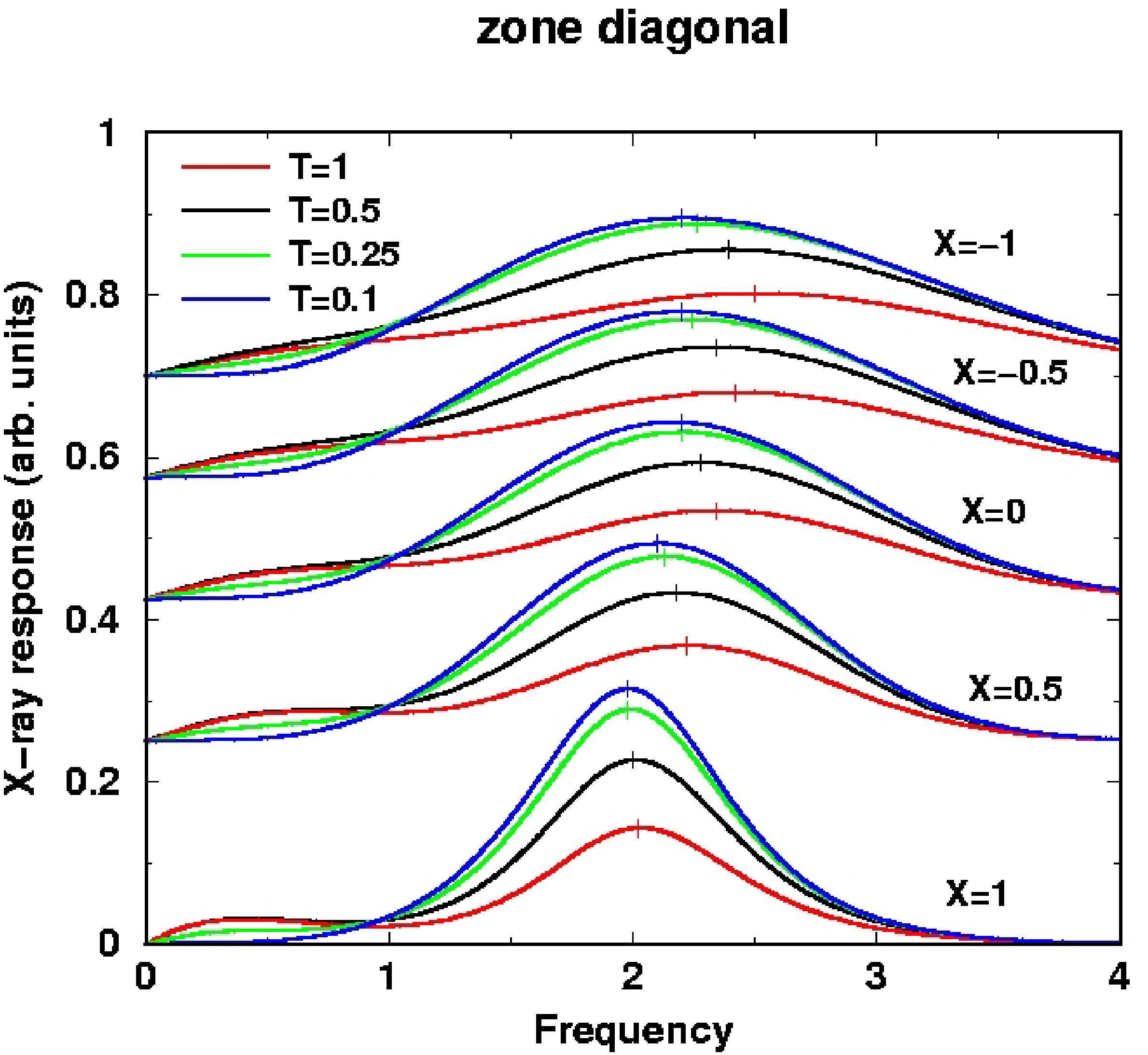


Nonresonant Raman Response (U=2)



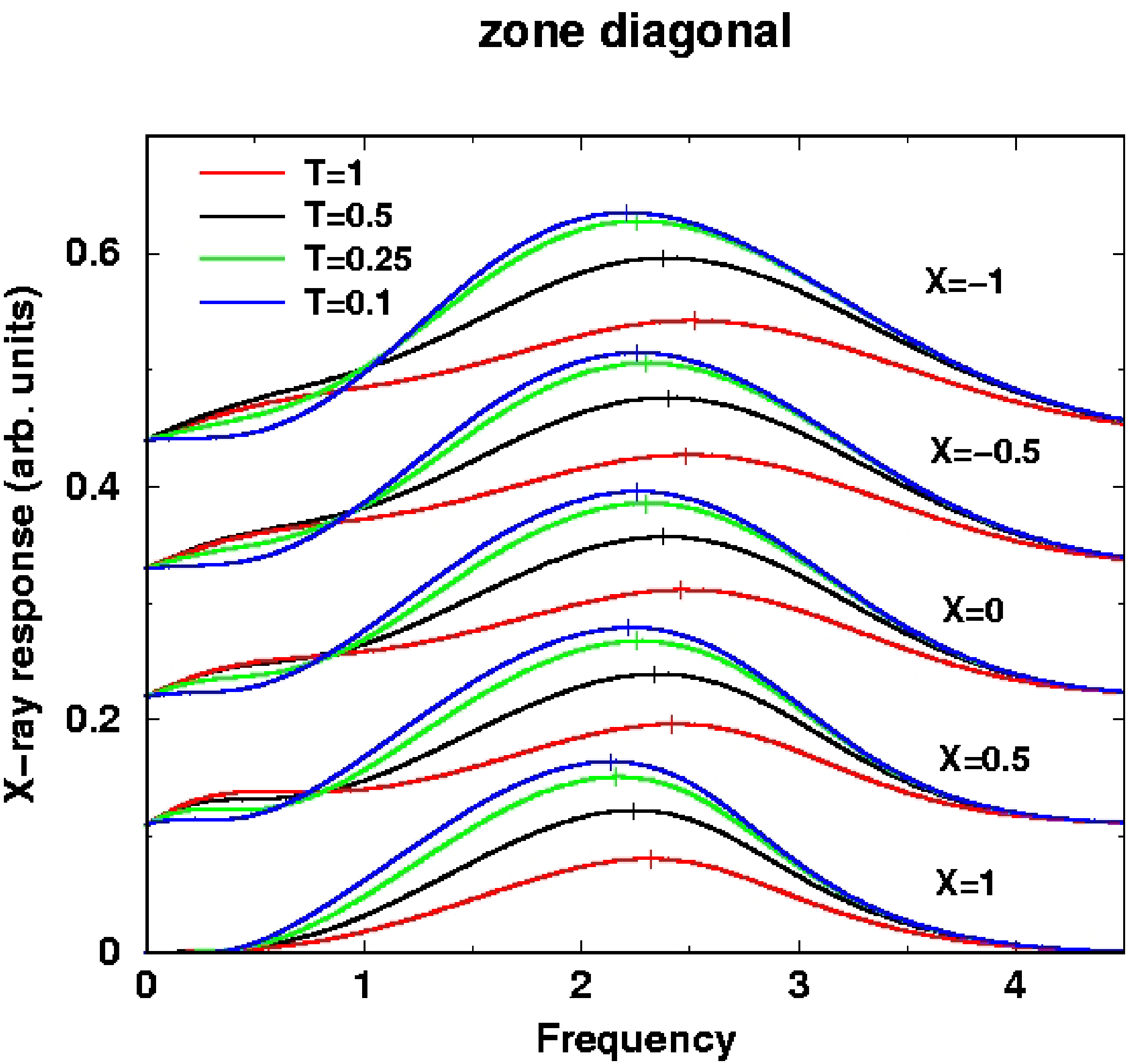
- The low-frequency B_{1g} response develops at a low temperature over a **wide frequency range** of O(1).
- An **isosbestic point** divides where spectral weight increases or decreases as T is lowered (B_{1g}).

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.

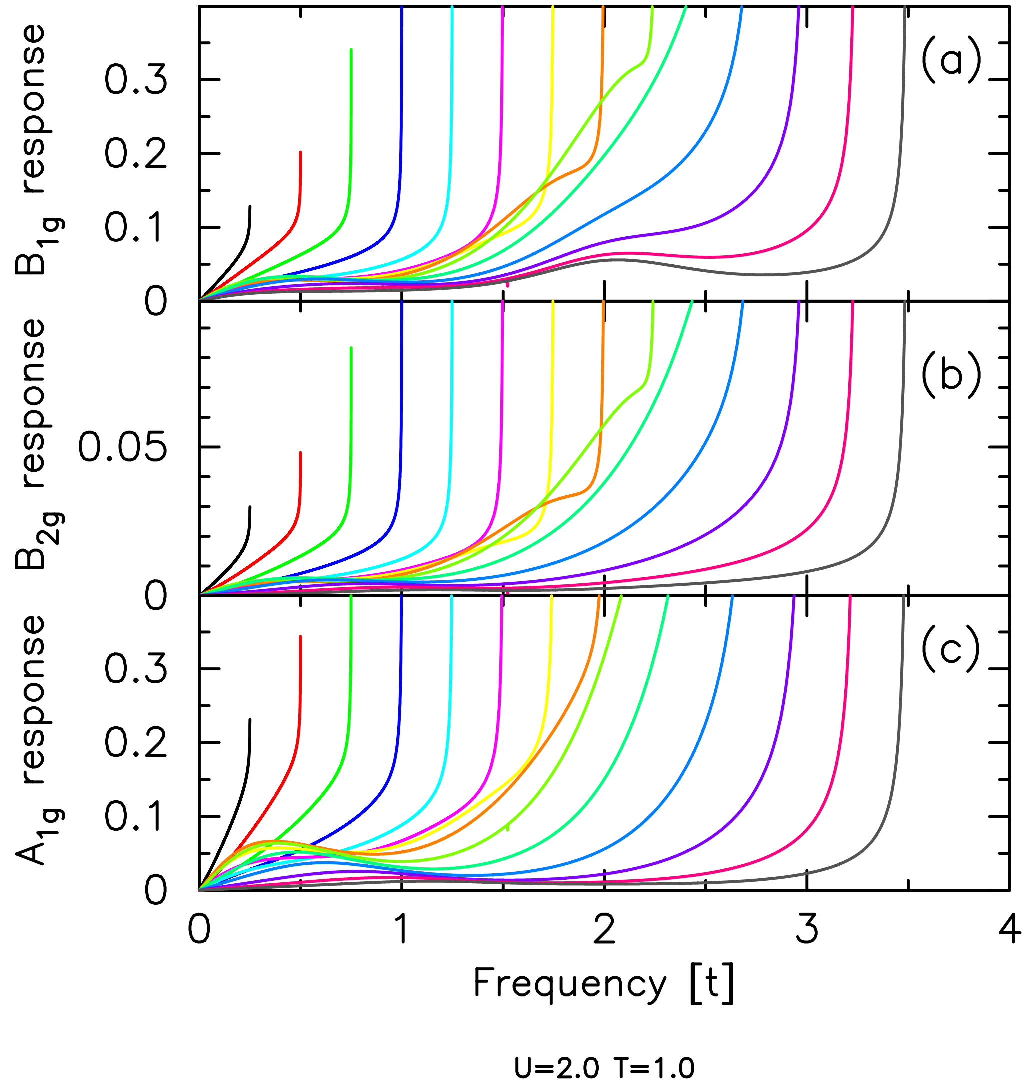
Inelastic X-ray scattering (A_{1g})



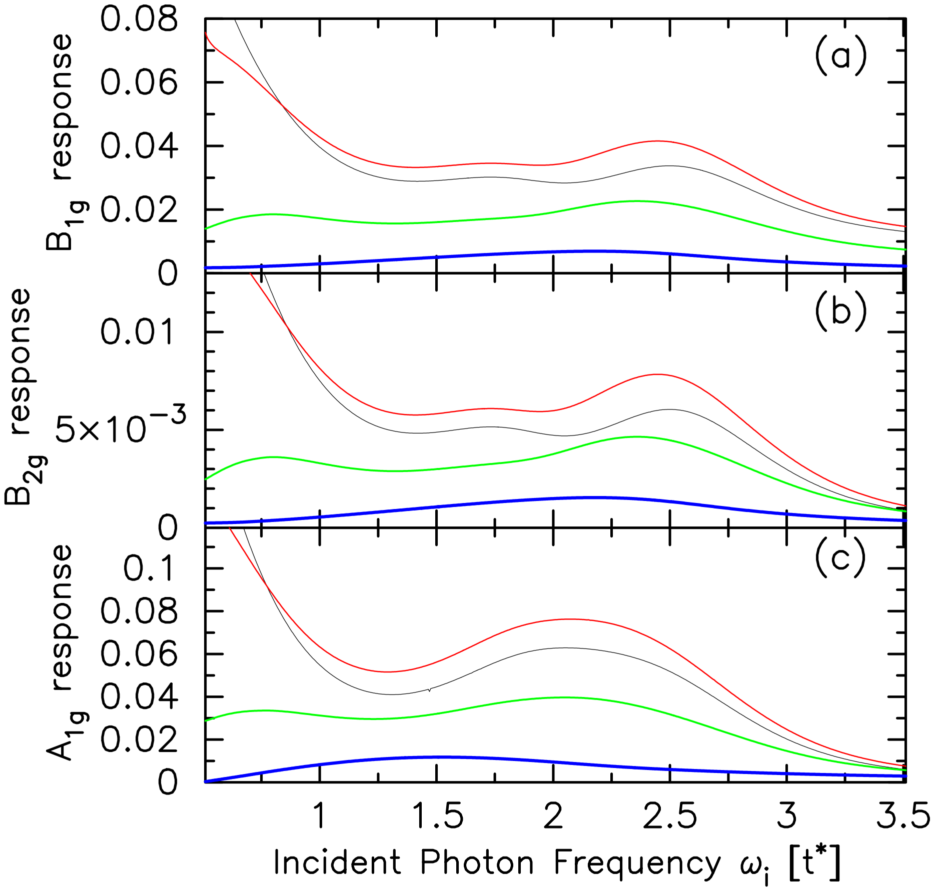
- Here the results at finite-**q** differ greatly from **q=0**: all of the anomalies appear away from **q=0**!
- A reduced **broadening** and **dispersion** of the peaks is seen; but the **same** anomalous low-energy behavior and the isosbestic points recur for nonzero **q**.

Resonant effects

- There is a large **double resonance** when the transferred frequency approaches the incident photon frequency.
- The resonant effects can be large, and can **change the shape** of the nonresonant results when the photon frequency is close to either Ω or U.



Joint resonance effect



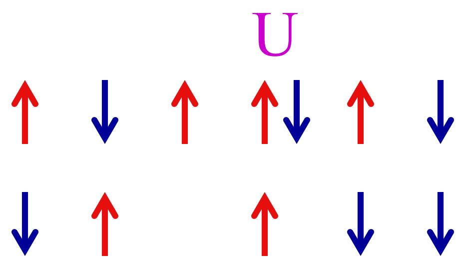
- When the examine the Raman signal at the low-energy peak, we find that it resonates when the photon frequency is close to it, and when it is close to U. The different colors are different temperatures.

Summary (FK 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 $\mathbf{q=0}$.
- There is an interesting **joint resonance** of the high and low energy peaks when the initial photon frequency is close to U.

Hubbard Model

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



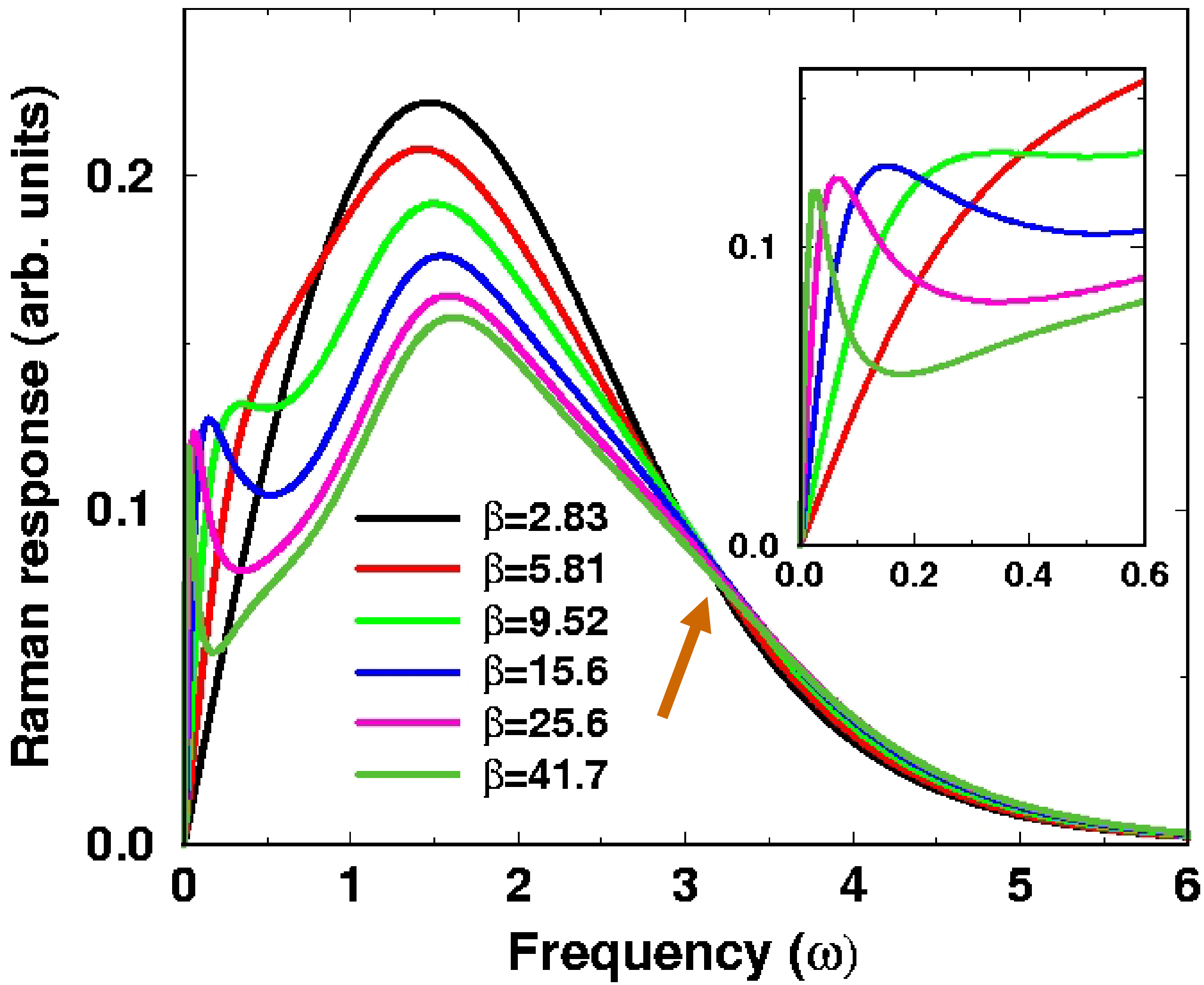
Both electrons are now **mobile**

•**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 max-ent techniques.

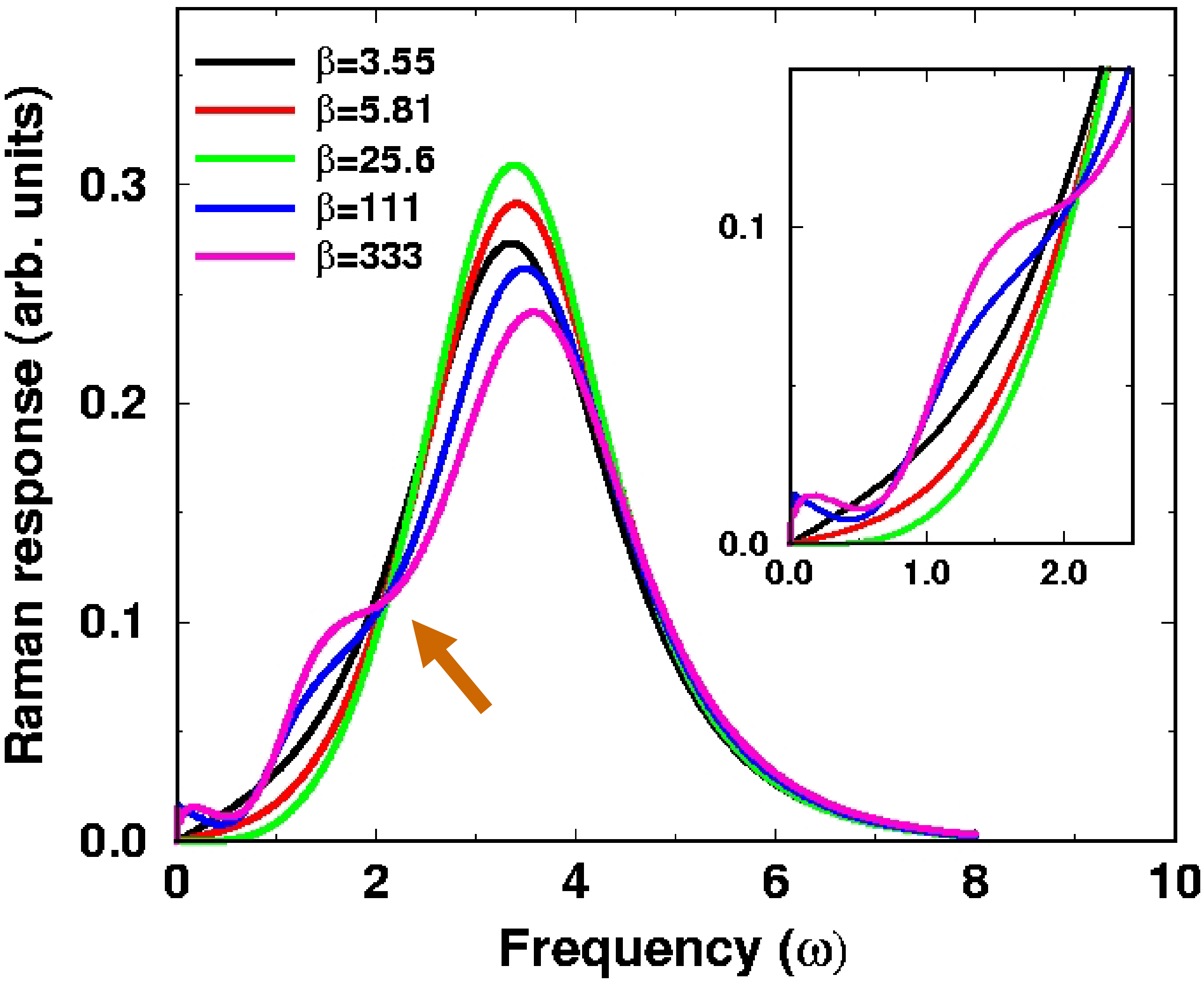
•Hence, the inelastic light scattering response can be constructed formally exactly for the nonresonant B_{1g} channel only (zone diagonal).

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



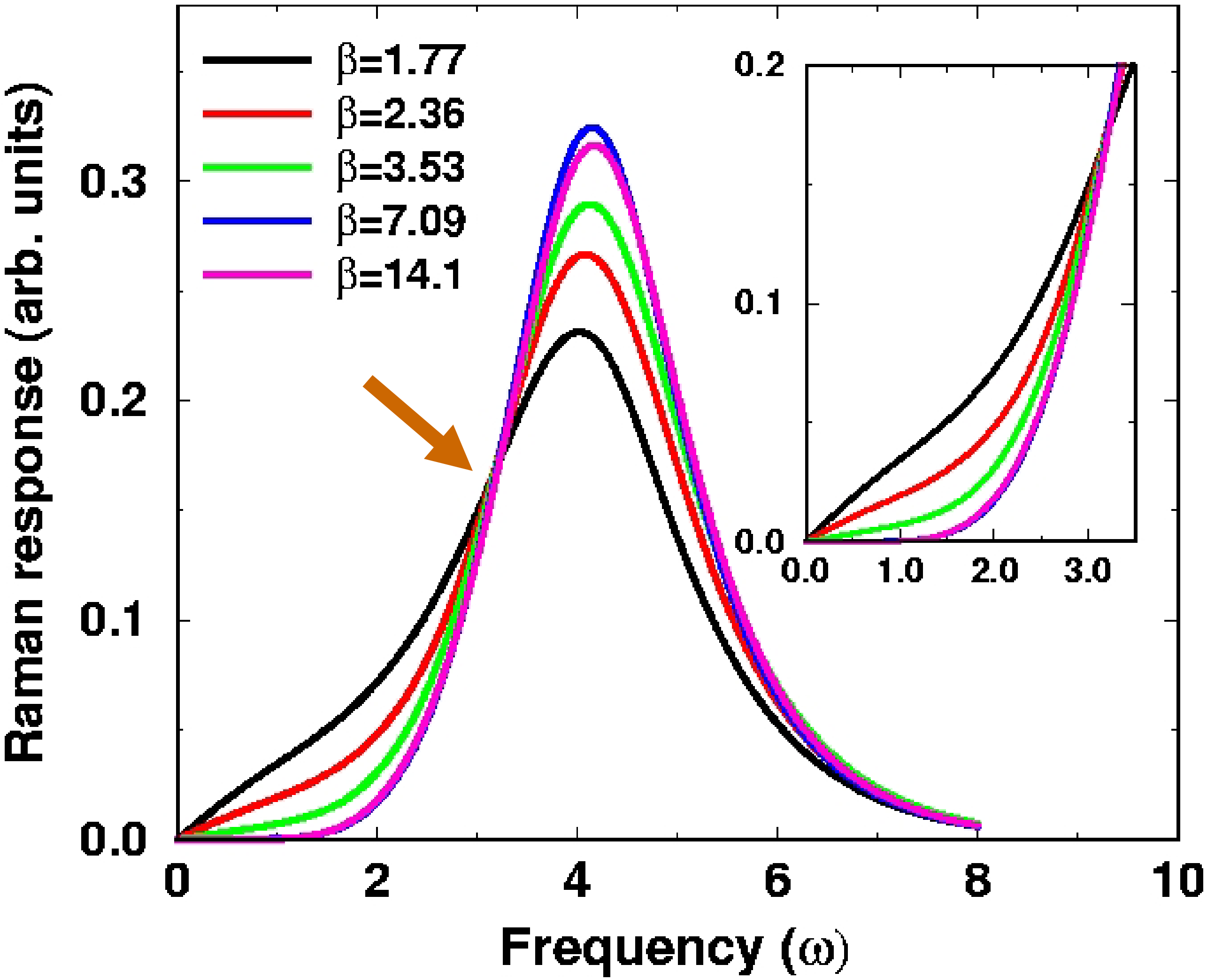
- Note the **charge transfer peak** as well as the **Fermi liquid peak** at low energy. As T goes to zero, the Fermi peak **sharpens** and **moves to lower energy**, as expected.
- There is **no low energy and low-T isosbestic point**, rather a high frequency isosbestic point seems to develop.

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**.

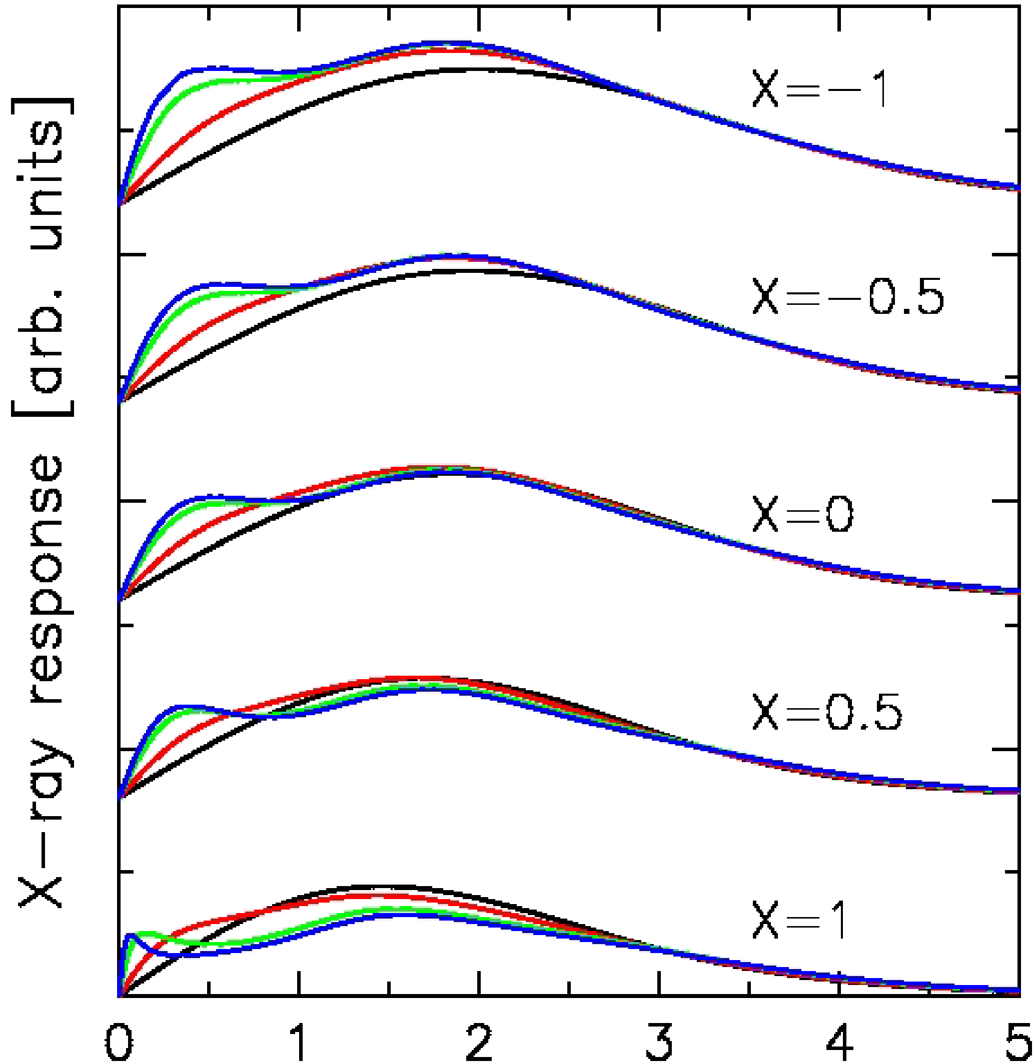
Nonresonant B_{1g} Raman scattering (n=1,U=4.2)



- 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.

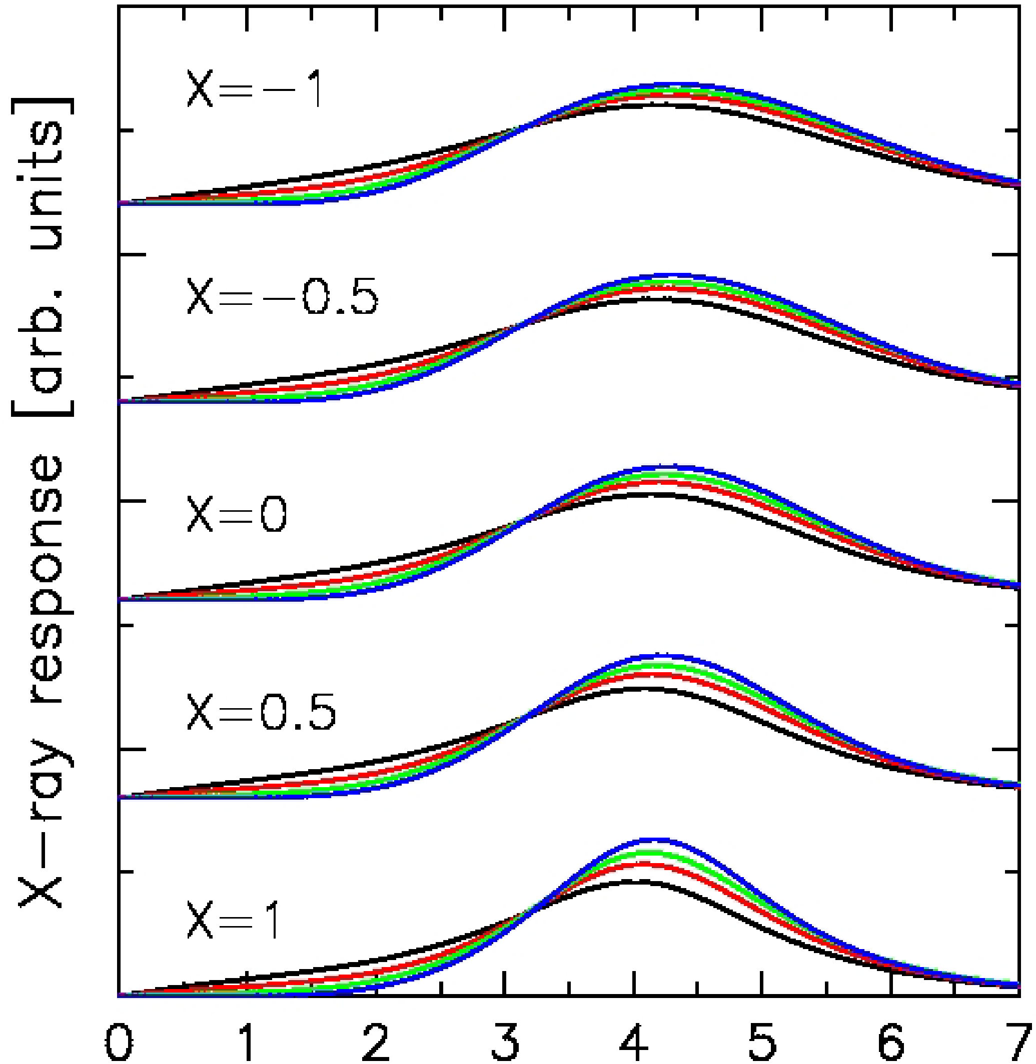
Inelastic X-ray scattering (B_{1g}, zone diagonal)

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



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

- Nonresonant scattering for a **correlated insulator**, at half filling and U=4.24.
- 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.

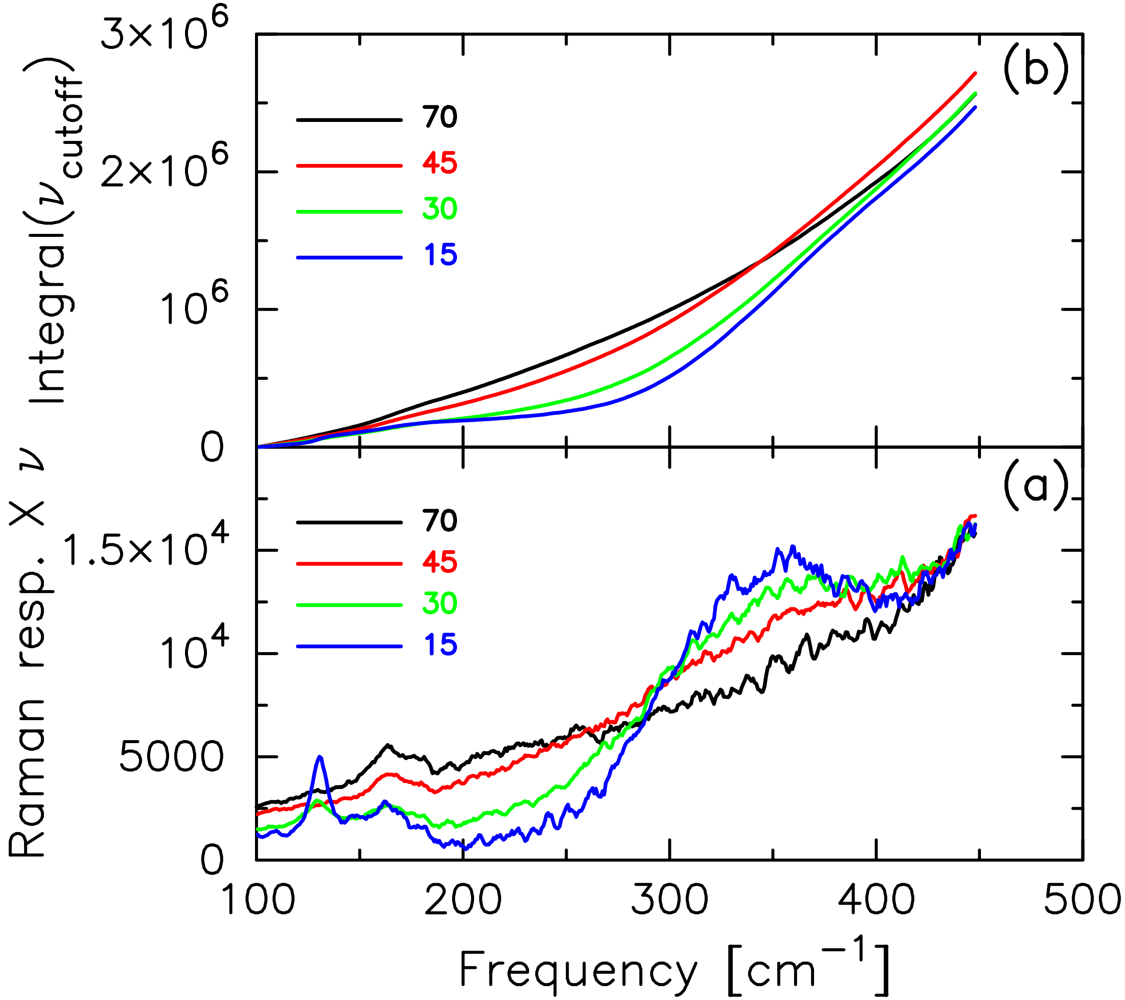


Summary Hubbard model

- 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 smoother 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.

Sum rules for Raman scattering

- Data on SmB_6 shows that the integral of the first moment of the Raman response **satisfies a sum rule**. It is almost a constant as a function of T.

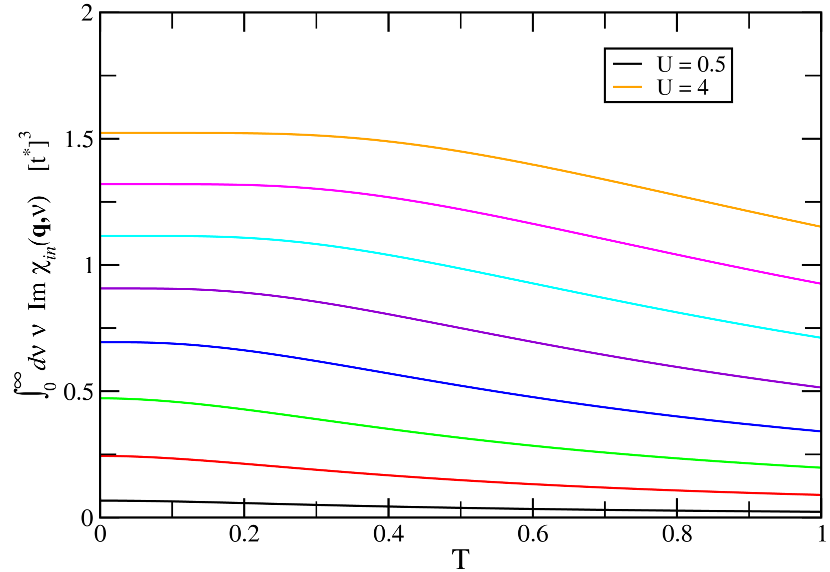


Theory for the sum rule

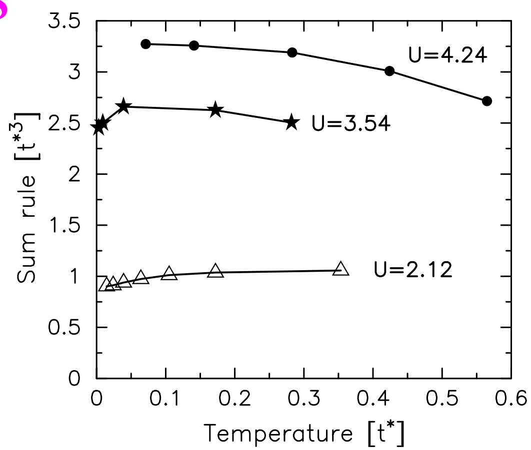
- A simple analysis of the nonresonant response function shows the **first moment** of the Raman signal is proportional to $<[\rho_{Raman},[H,\rho_{Raman}]]>$, with $\rho_{Raman}=\Sigma_{\mathbf{k}}\gamma(\mathbf{k})c^{\dagger}_{\mathbf{k}}c_{\mathbf{k}}$ the Raman density operator (stress tensor).
- Since the kinetic energy **commutes** with the stress tensor, the sum rule depends only on the **potential energy**!

Results f

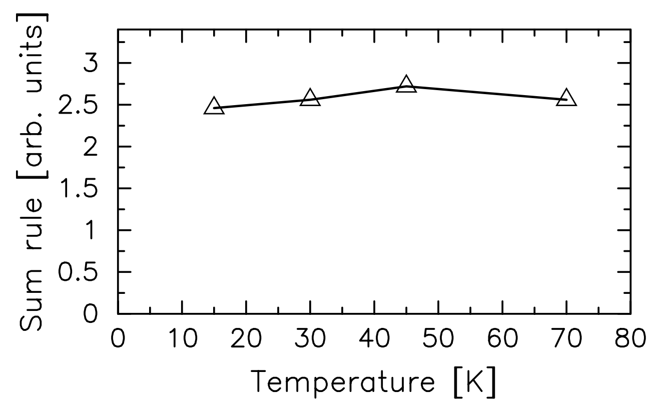
- The sum rule for the FK model is **almost constant** as a function of T at low T.
- For the Hubbard model, it appears to have a **low-T decline**.
- This is similar to SmB_6.



FK



Hubbard



SmB_6

Conclusions

- Showed how an exact solution for **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 experimental Raman scattering. There were interesting **resonant effects** too.
- 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 a new **sum rule** for inelastic light scattering that provides information about the **potential energy** of the material.