

Universal features of electronic Raman scattering across the metal-insulator transition

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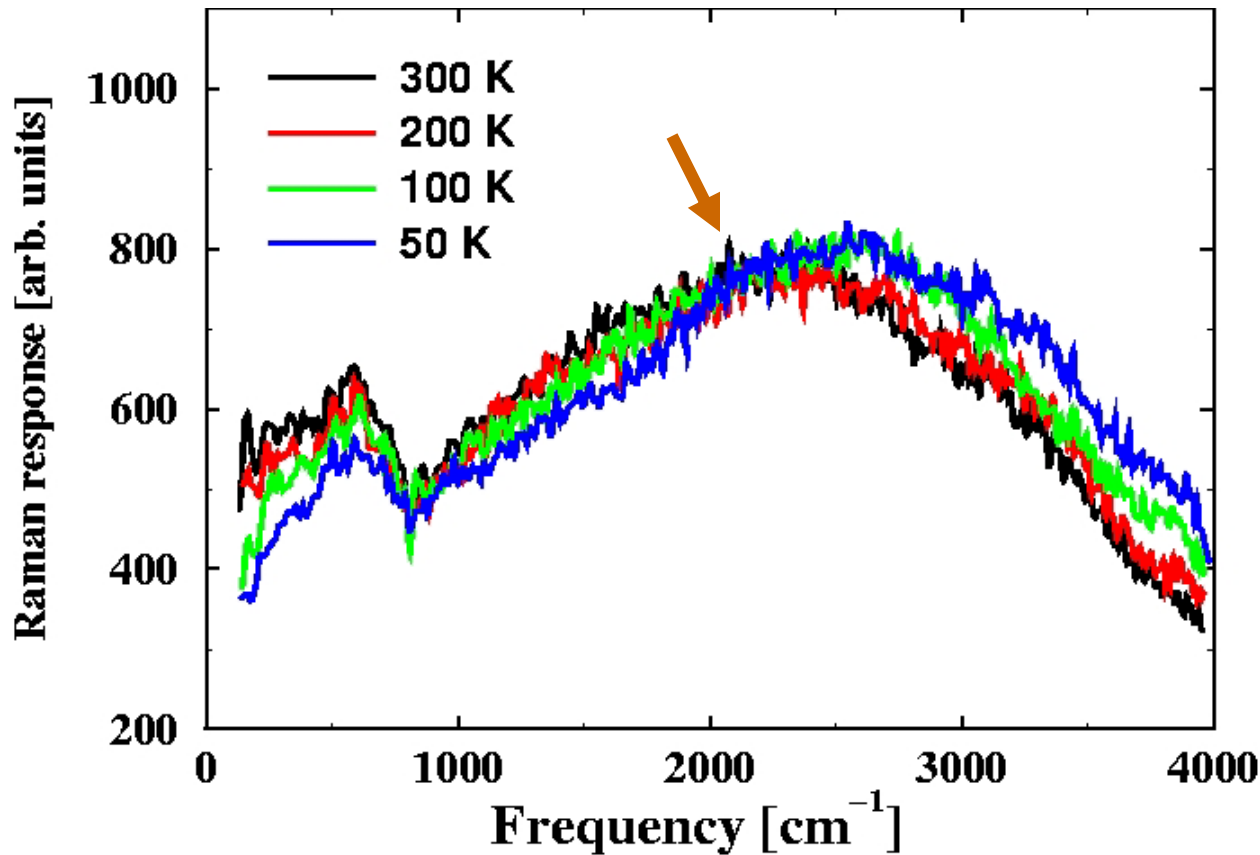
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Raman scattering probes electronic excitations

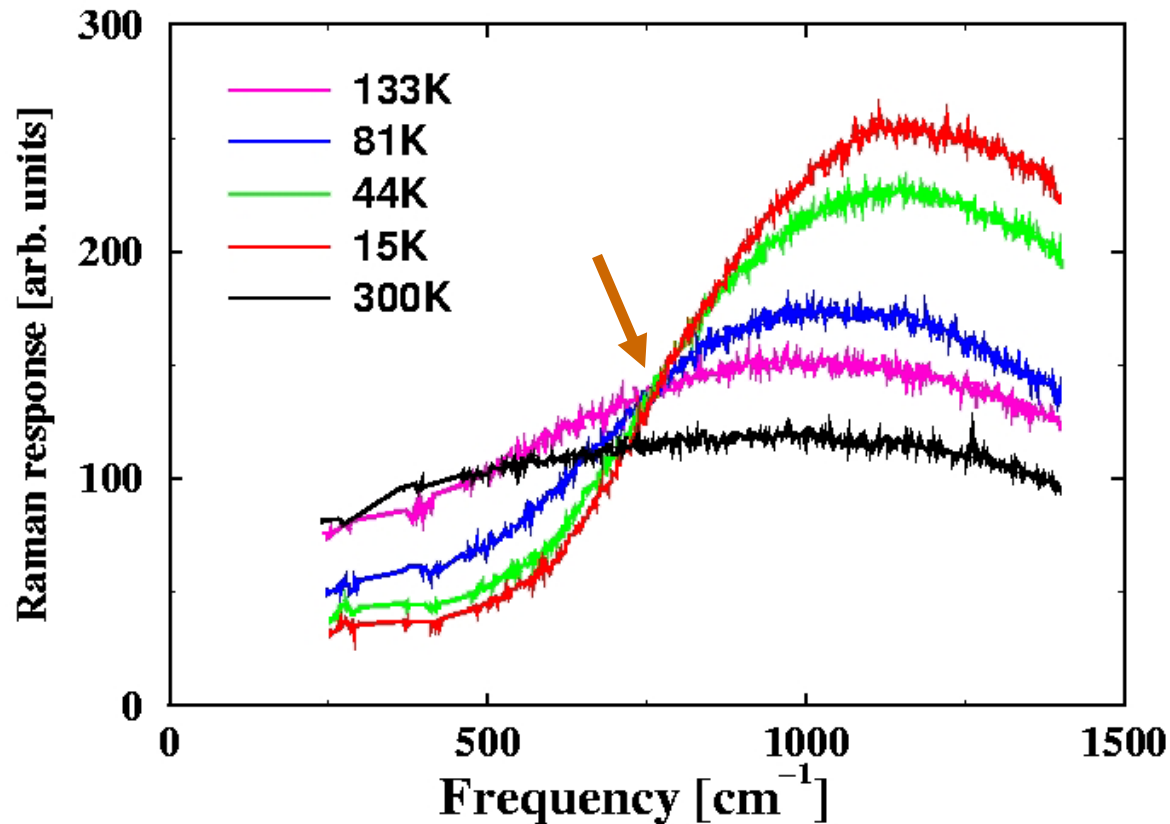
- **Inelastic scattering of light** with electron-hole excitations of the correlated many-body system.
- Use of polarizers for the incident and reflected light allows one to **select different symmetries** of the electron-hole excitations.
- Signal depends on the **Raman scattering amplitude $\gamma(\mathbf{k})$**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

Experimental data for the cuprates



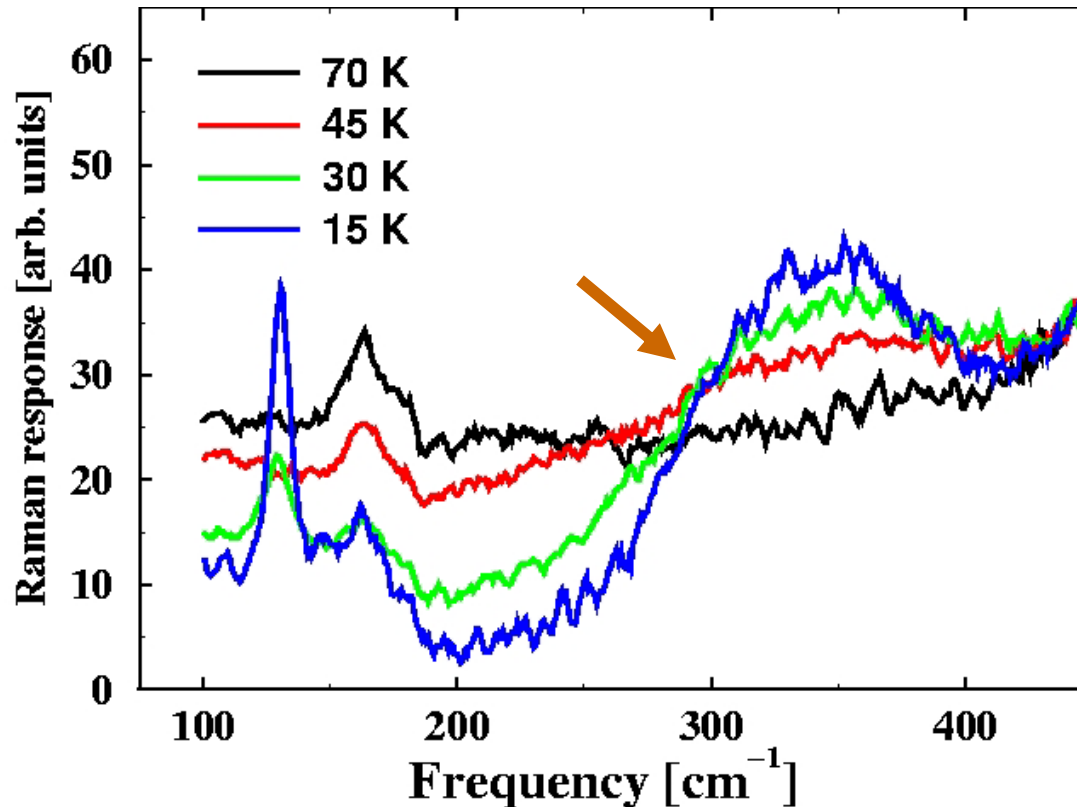
Data on underdoped **Lanthanum Strontium Copper Oxide**, from Chuck Irwin's group, shows the reduction of low-frequency spectral weight and the increase in the charge transfer peak, with an isosbestic point at about 2100 cm^{-1} .

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 **SmB₆**. 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.

Summary of Experimental Data

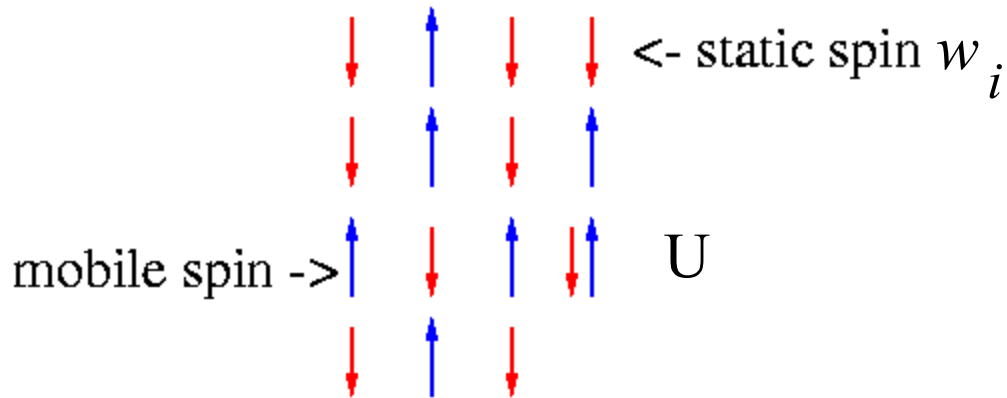
- Three characteristic behaviors are seen: (i) as T is lowered, there is a **redistribution of spectral weight** from low-frequency to high frequency; (ii) these regions are separated by an isosbestic point, where **the Raman response is independent** of T; (iii) the ratio of the twice spectral range 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 high T_c superconductor, a Kondo insulator, or an intermediate-valence material.

Theories of Raman 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).
- But no theory exists that can connect these two regimes and carry 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 scattering through a metal-insulator transition in both the Falicov-Kimball model and the Hubbard model.*

Spinless Falicov-Kimball Model

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



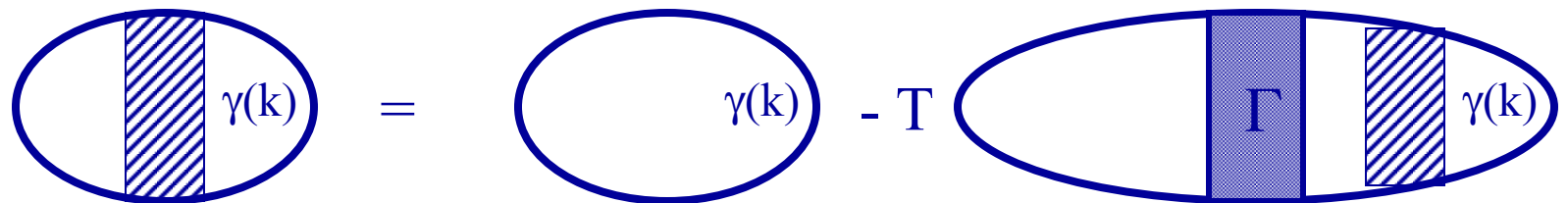
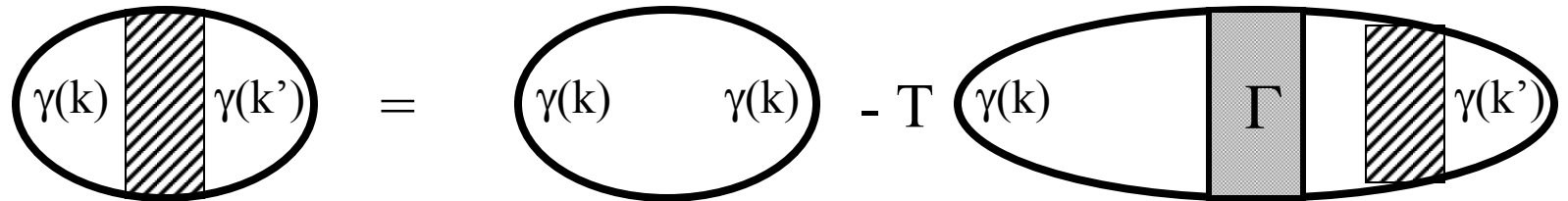
- **exactly solvable model** on a hypercubic lattice in infinite dimensions using dynamical mean field theory.
- possesses homogeneous, commensurate/incommensurate CDW phases, phase segregation, and **metal-insulator transitions**.
- *Raman response can be constructed formally exactly.*

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**.

Diagrams for the A_{1g} Raman response



$\gamma(k) = -\epsilon(k)$, Γ is **local** and has no k -dependence

Solving these coupled equations allows for the full nonresonant Raman response to be determined.

Formal Solution for the Raman Scattering Response

B_{1g} channel

- This channel is **orthogonal** to the lattice.
- There are **no vertex corrections** (Khurana, PRL, 1990), so the Raman response is represented by the **bare bubble**.
- This response is **identical** to that of the optical conductivity multiplied by one factor of frequency (Shastry and Shraiman, PRL, 1990).
- **Resonant** Raman 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.

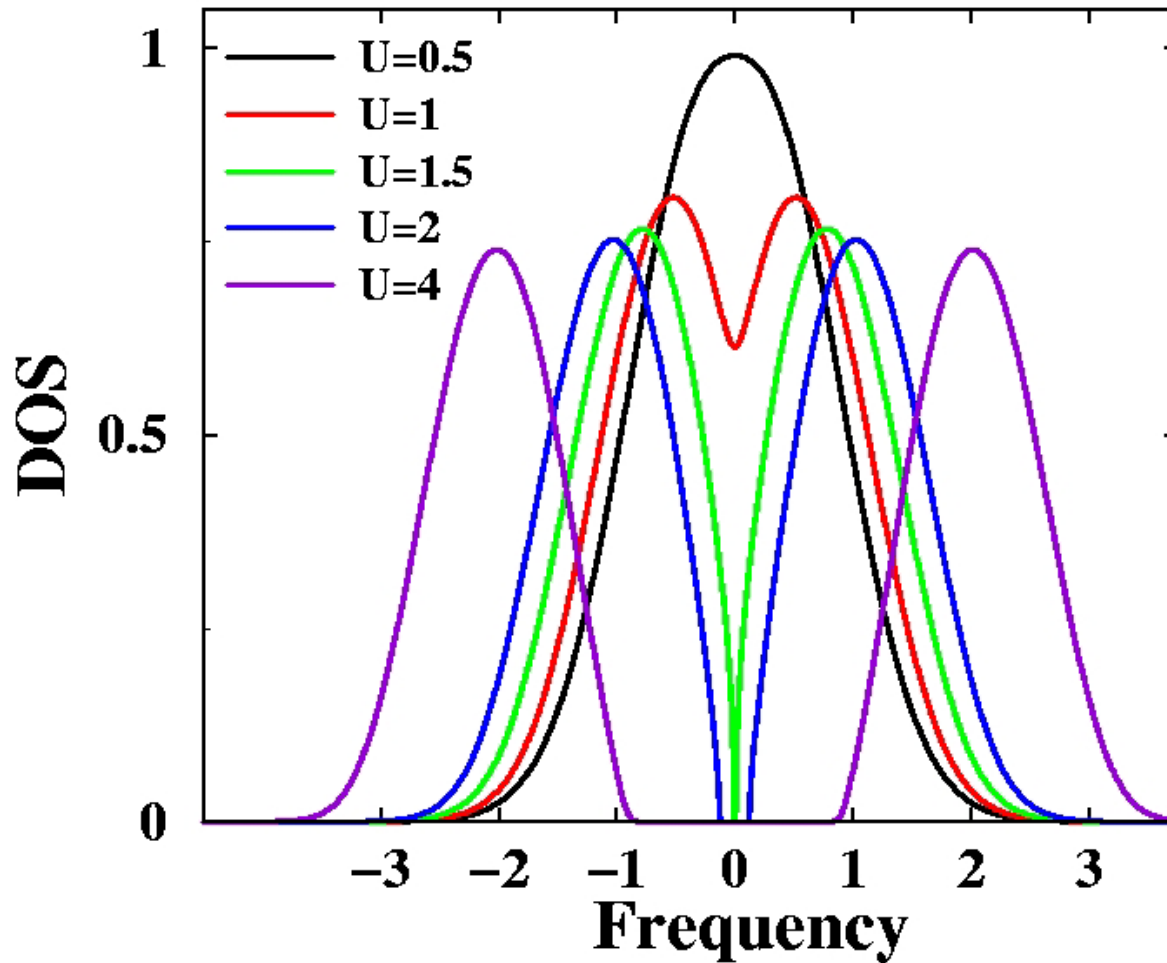
Formal Solution for Raman the Scattering Response

B_{2g} channel

- The Raman 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 don't discuss this channel further here.

B_{2g} Raman scattering is purely resonant.

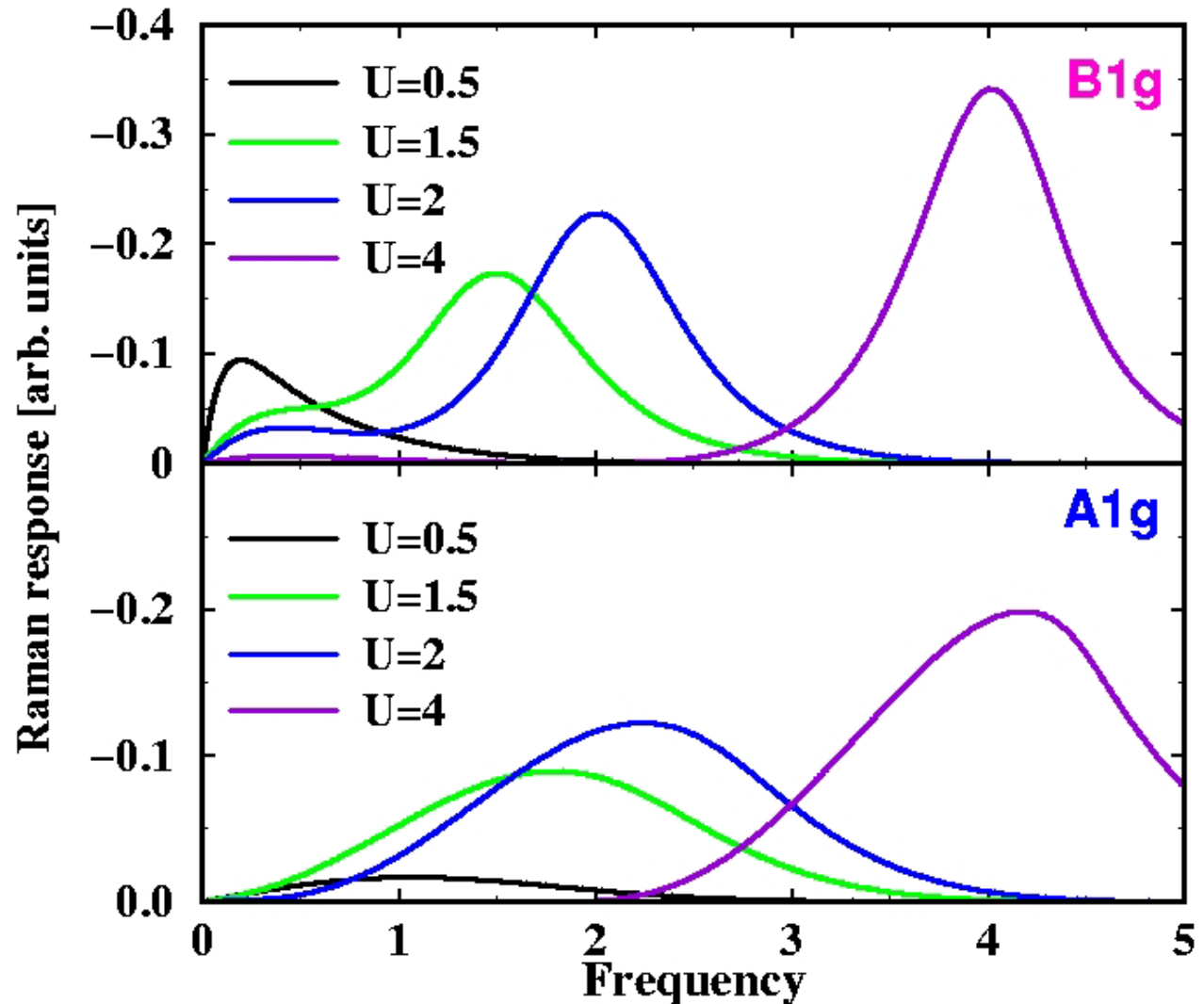
Metal-Insulator transition



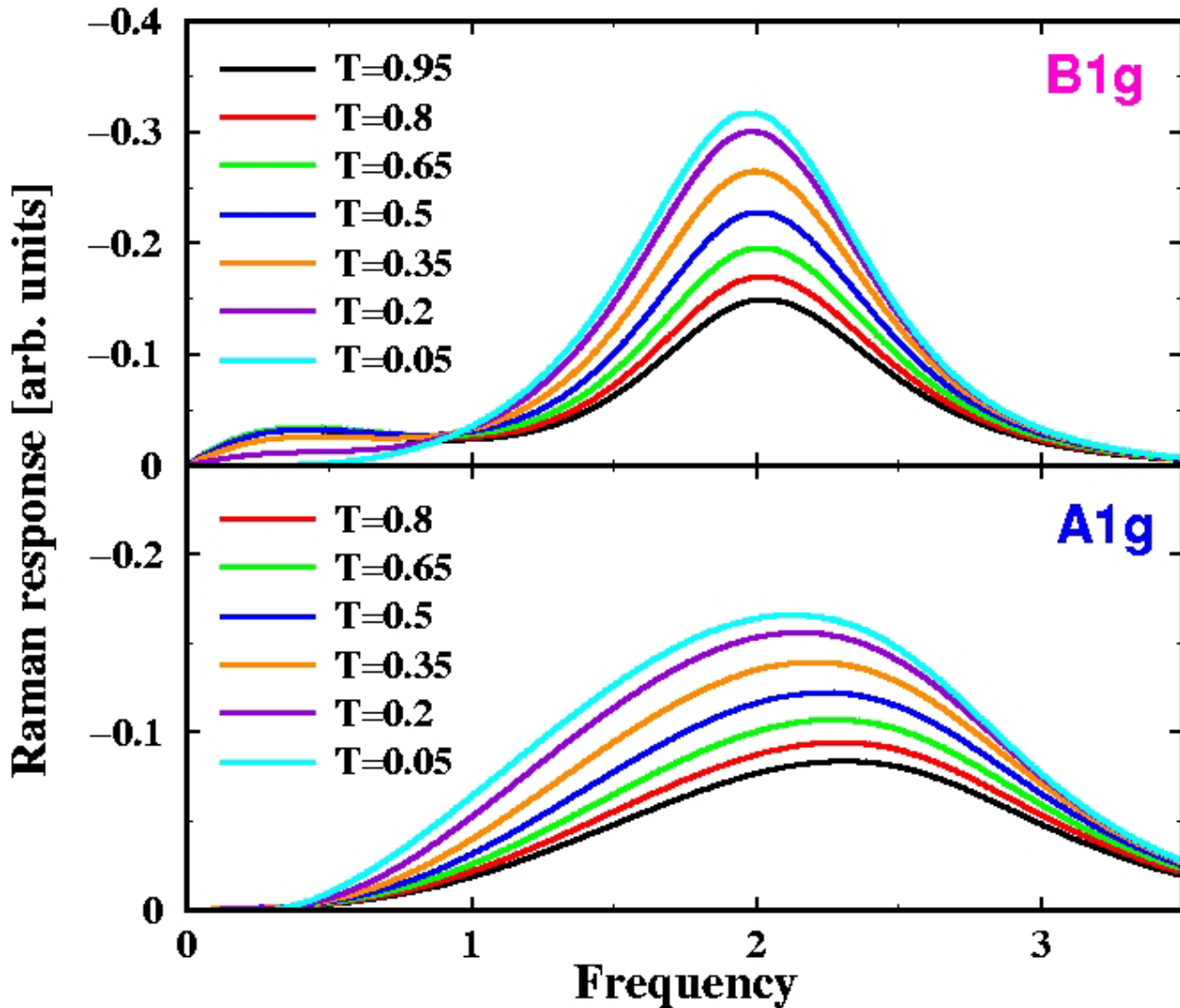
- **Correlation-induced** gap drives the single-particle DOS to zero at $U=1.5$
- Interacting DOS is **independent of T** in DMFT (Van Dongen, PRB, 1992)
- *Examine Raman response through the ($T=0$) quantum phase transition.*

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 $O(1)$.

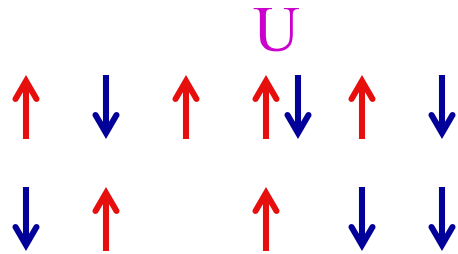
- An **isosbestic point** divides where spectral weight increases or decreases as T is lowered (B_{1g}).

Summary (Falicov-Kimball model)

- 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.

Hubbard Model

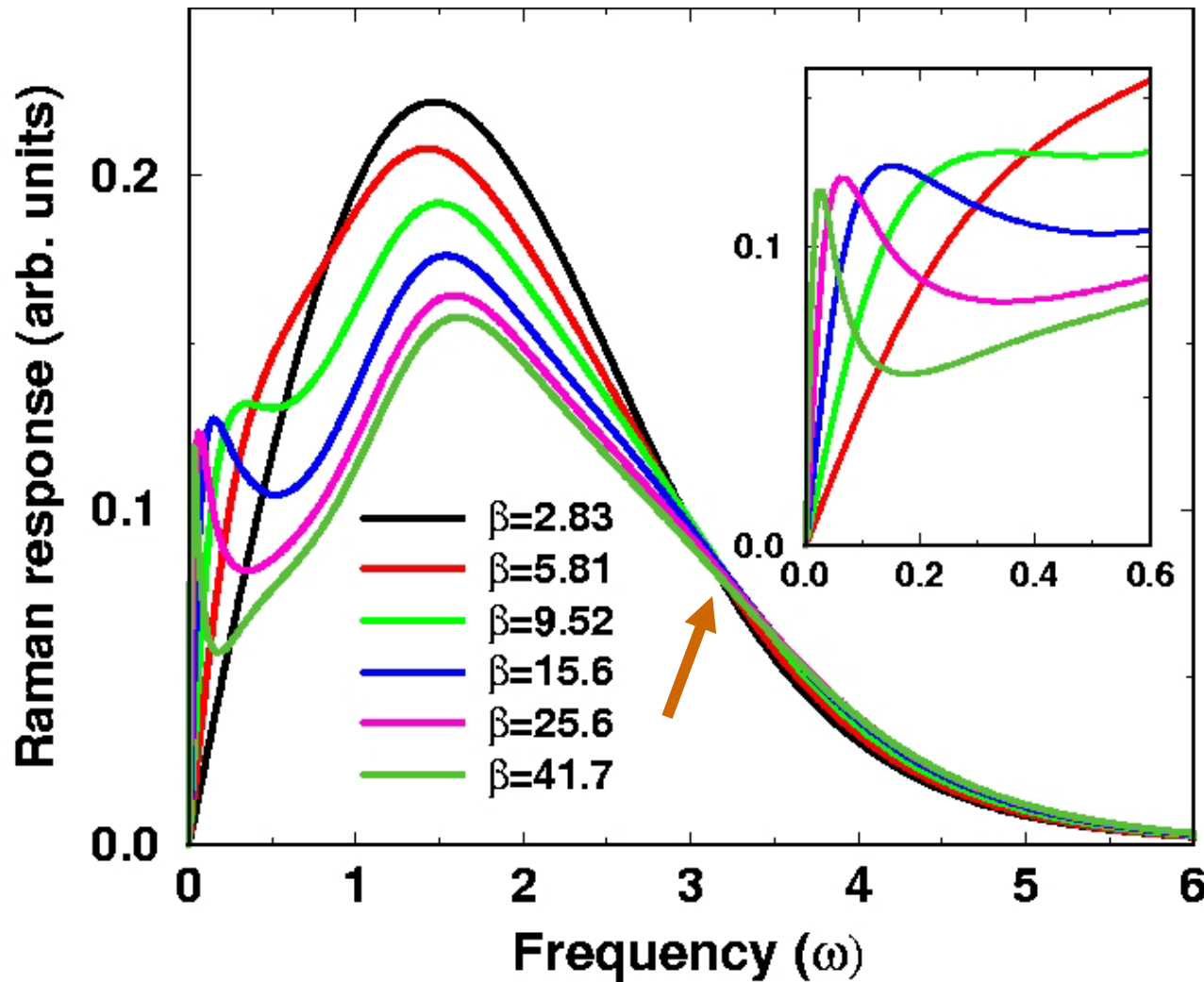
$$H = -\frac{t}{2\sqrt{d}} \sum c_{i\sigma}^* c_{j\sigma} + U \sum n_{i\uparrow} n_{i\downarrow}$$



Both electrons are now **mobile**

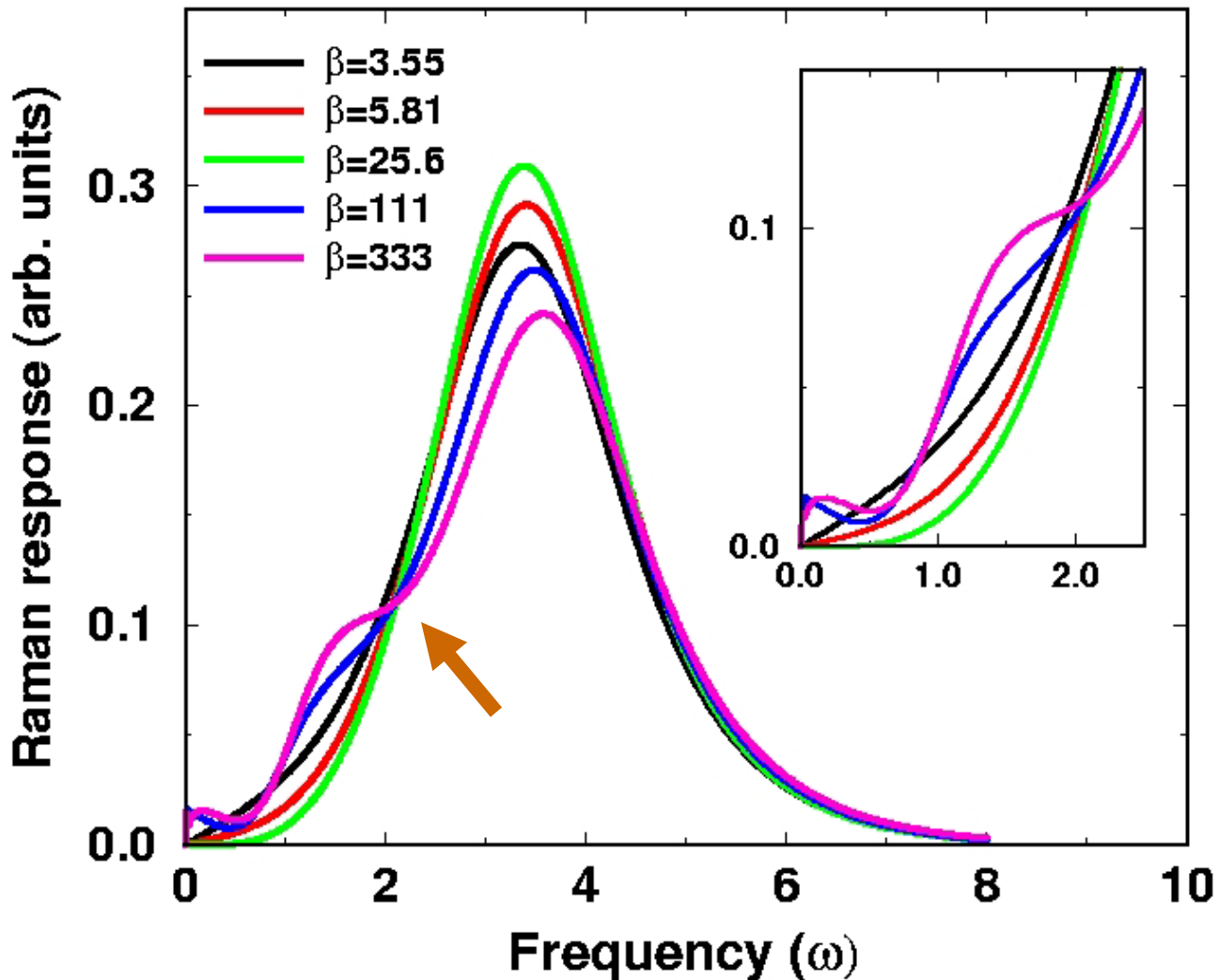
- **Exactly solvable** model on a hypercubic lattice in infinite dimensions using dynamical mean field theory (but requires NRG calculations to extract real frequency information).
- The irreducible charge vertex is **problematic to calculate** because it possesses too large a dynamic range for the max-ent techniques.
- *Hence, the Raman response can be constructed formally exactly for the nonresonant B_{1g} channel only.*

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



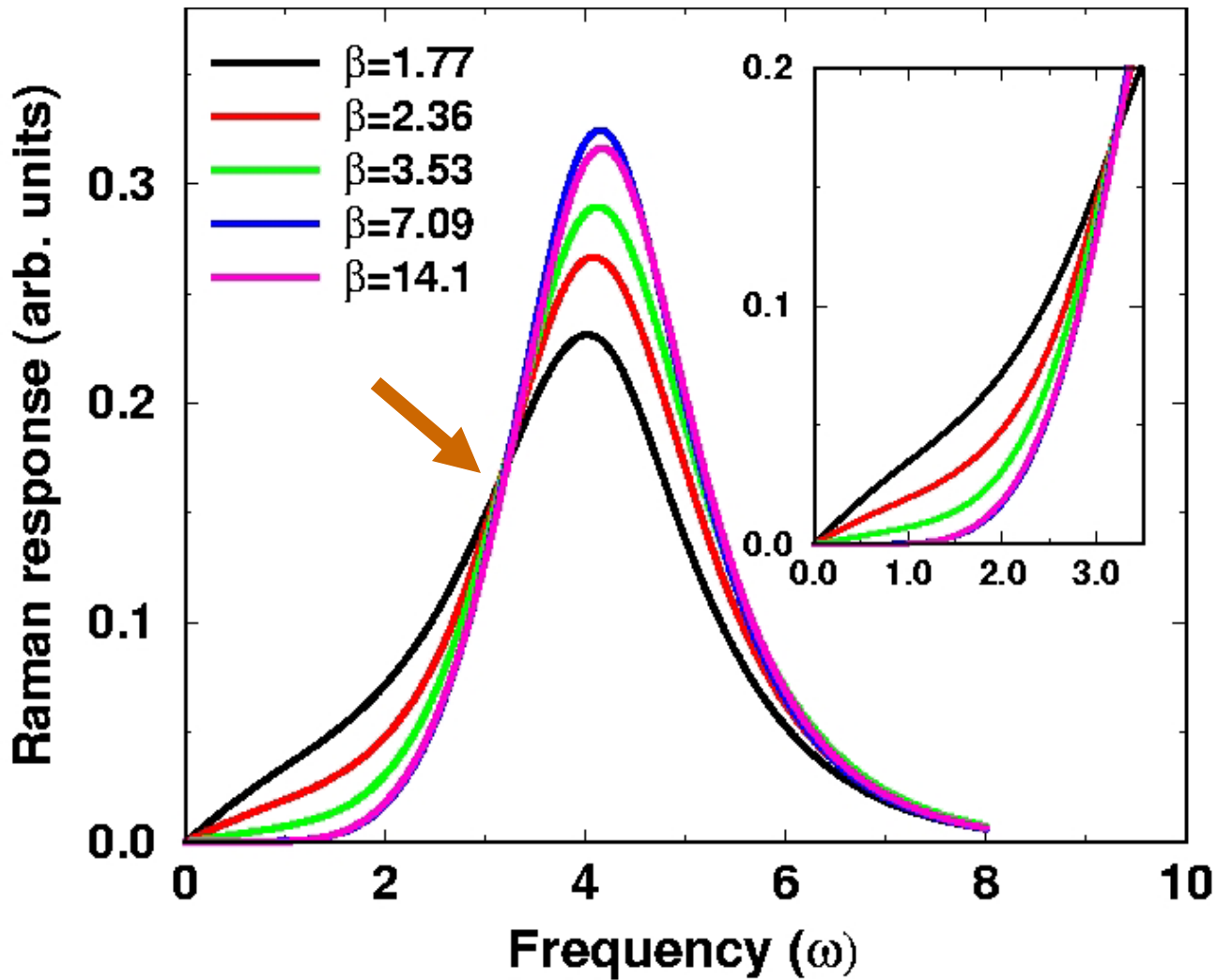
- Note the **charge transfer peak** as well as the **Fermi liquid peak** at low energy. As T goes to zero, the Fermi peak **sharpens** and **moves to lower energy**, as expected.
- **There is no low energy and low- T isosbestic point**, rather a high frequency isosbestic point seems to develop.

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.

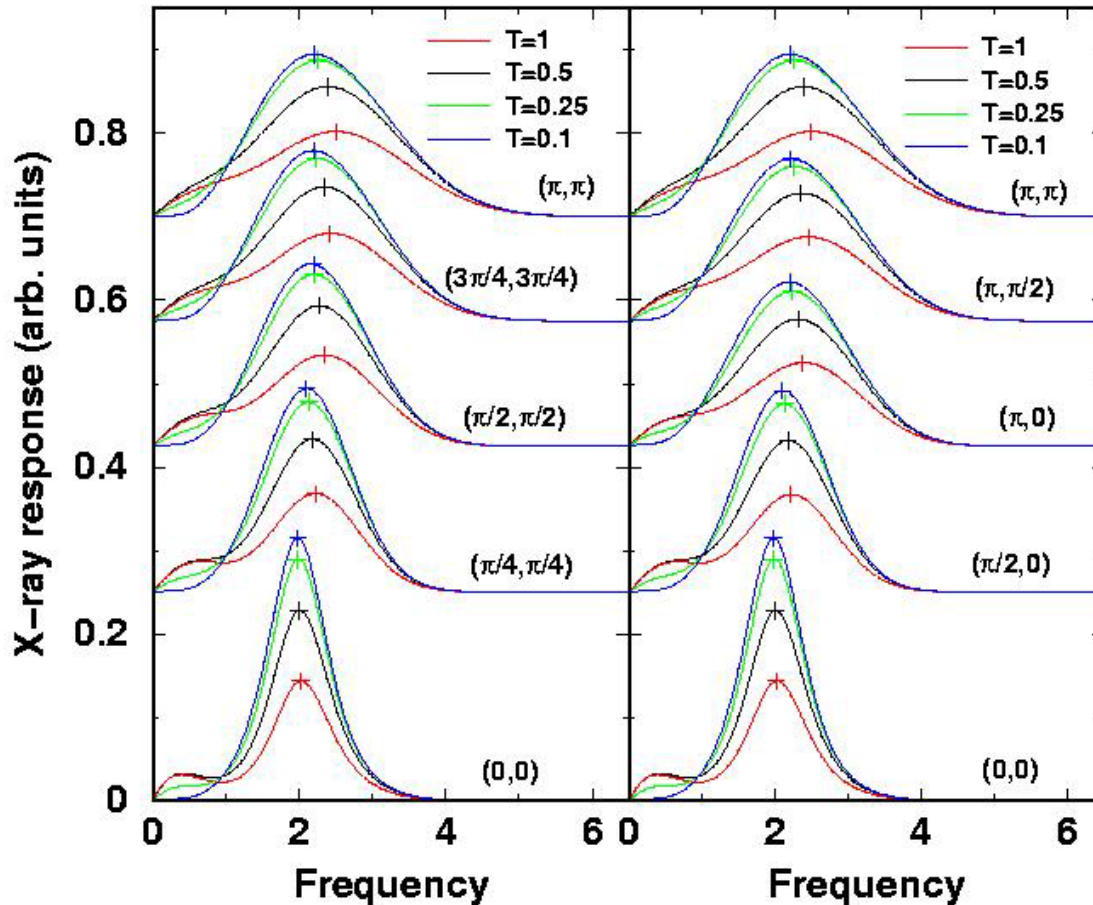
Summary Hubbard model

- 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.

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

zone diagonal

zone edge

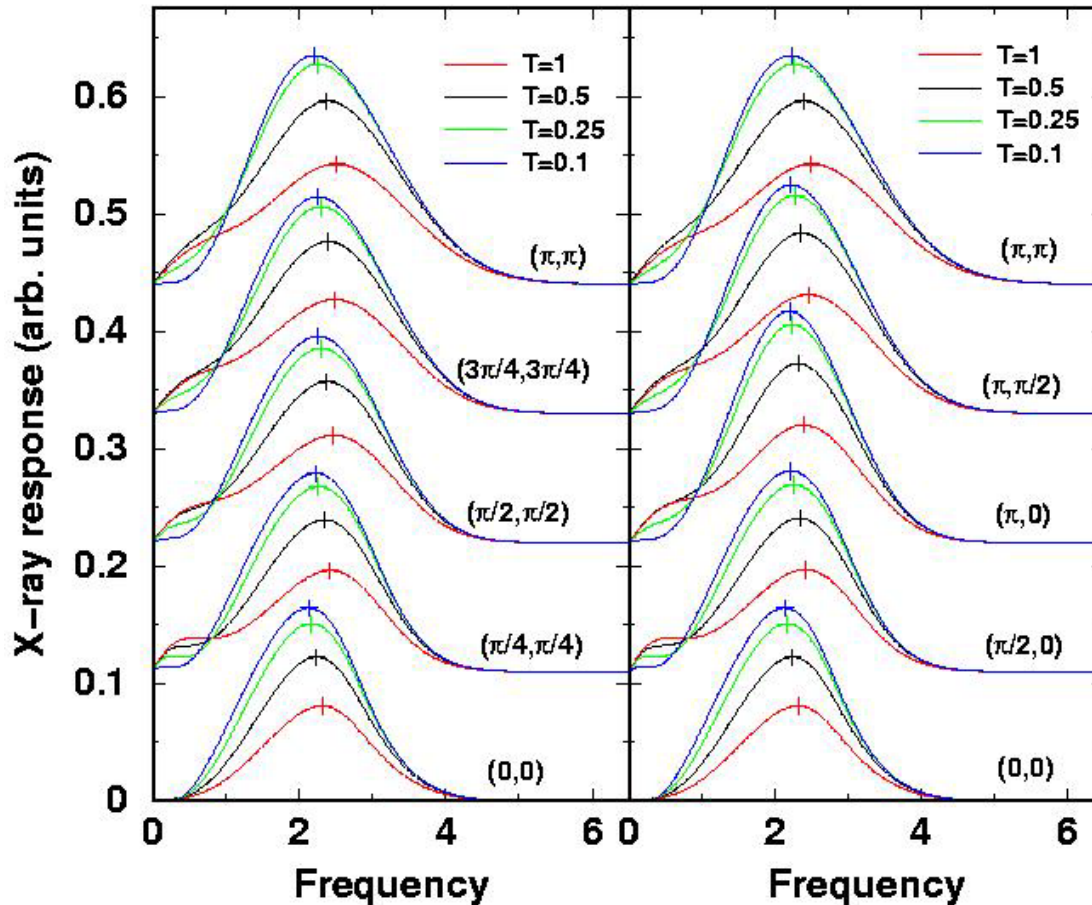


- Scattering of x-rays allows the photon to exchange **both momentum and energy** with the electron-hole excitations.
- We see a **broadening and dispersion** of the peaks, but the **same anomalous low-energy behavior and the isosbestic point**.

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

zone diagonal

zone edge



- 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 point recur for nonzero q .

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.