Raman scattering through a quantum-critical point

Jim Freericks (Georgetown University)
Tom Devereaux (University of Waterloo)
Ralf Bulla (University of Augsburg)

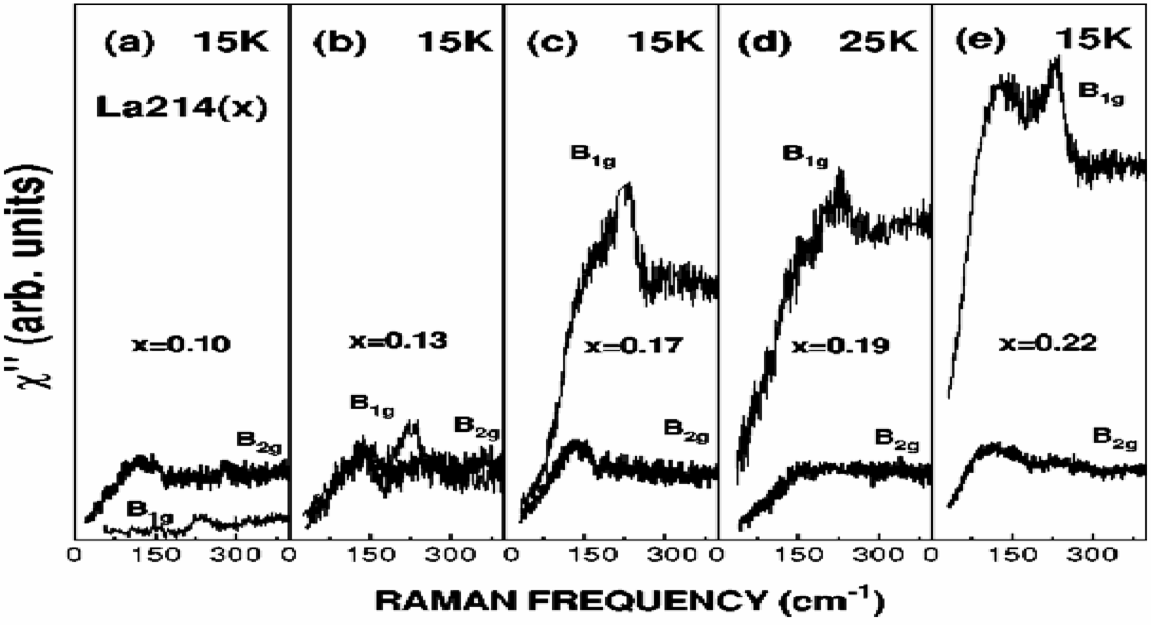
Funding: National Science Foundation (US)
National Science and Engineering Research Council (Canada)
Deutsche Forschungsgemeinshaft (Germany)
Thanks to: Paul Miller and Andrij Shvaika

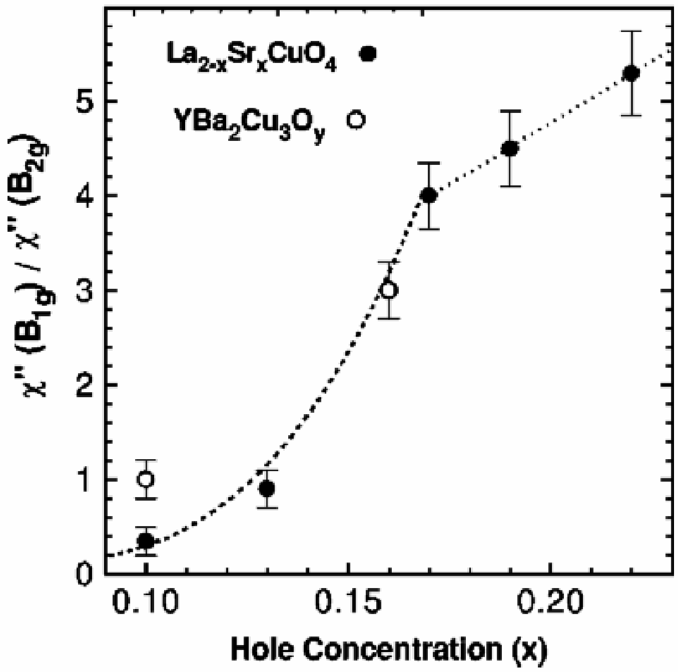
Raman scattering probes electronic excitations

- Inelastic scattering of light with electron-hole excitations of the correlated many-body system.
- Use of polarizers for the incoming 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}: $\gamma(k) \sim \sin(k_x a) \sin(k_y a)$

Review of Raman data for the cuprates

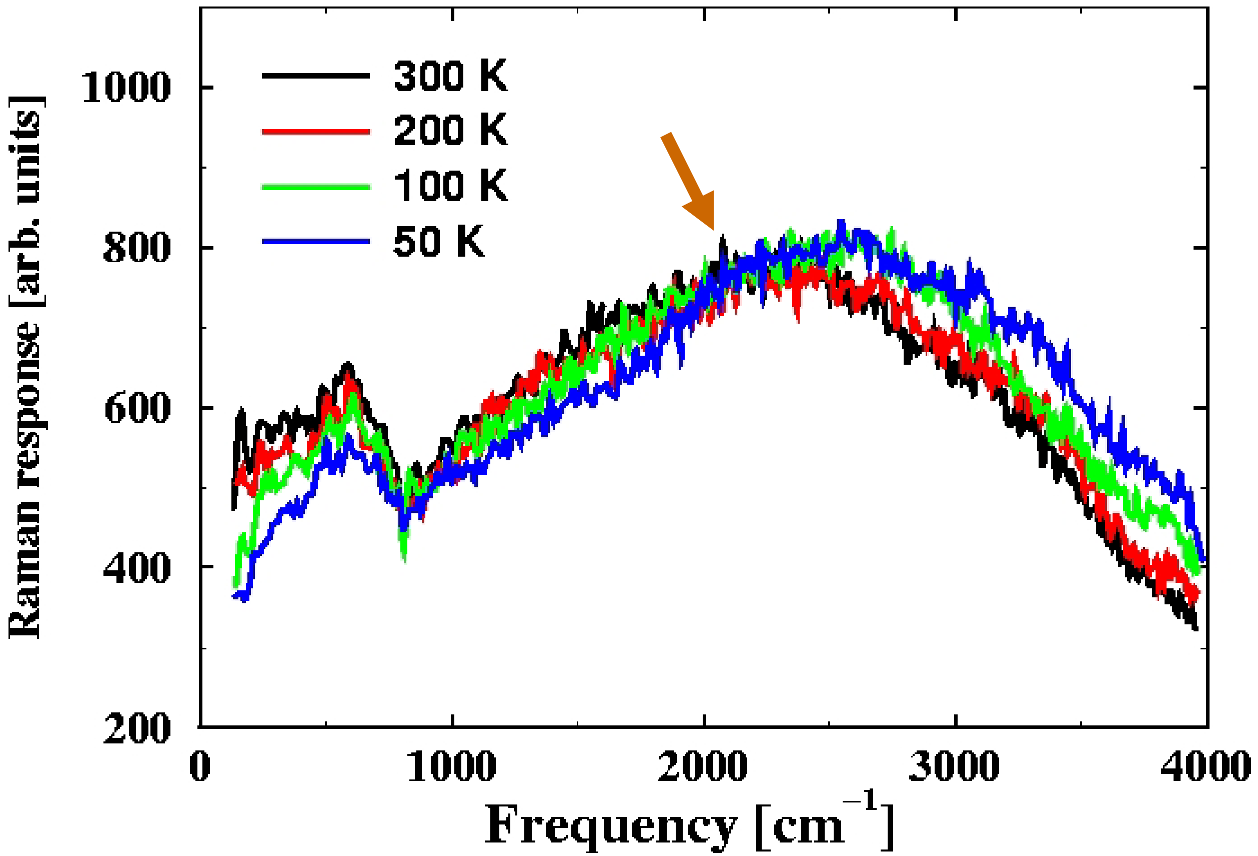
Normal state at low frequencies: J. G. Naeini *et al.*, PRB 1999





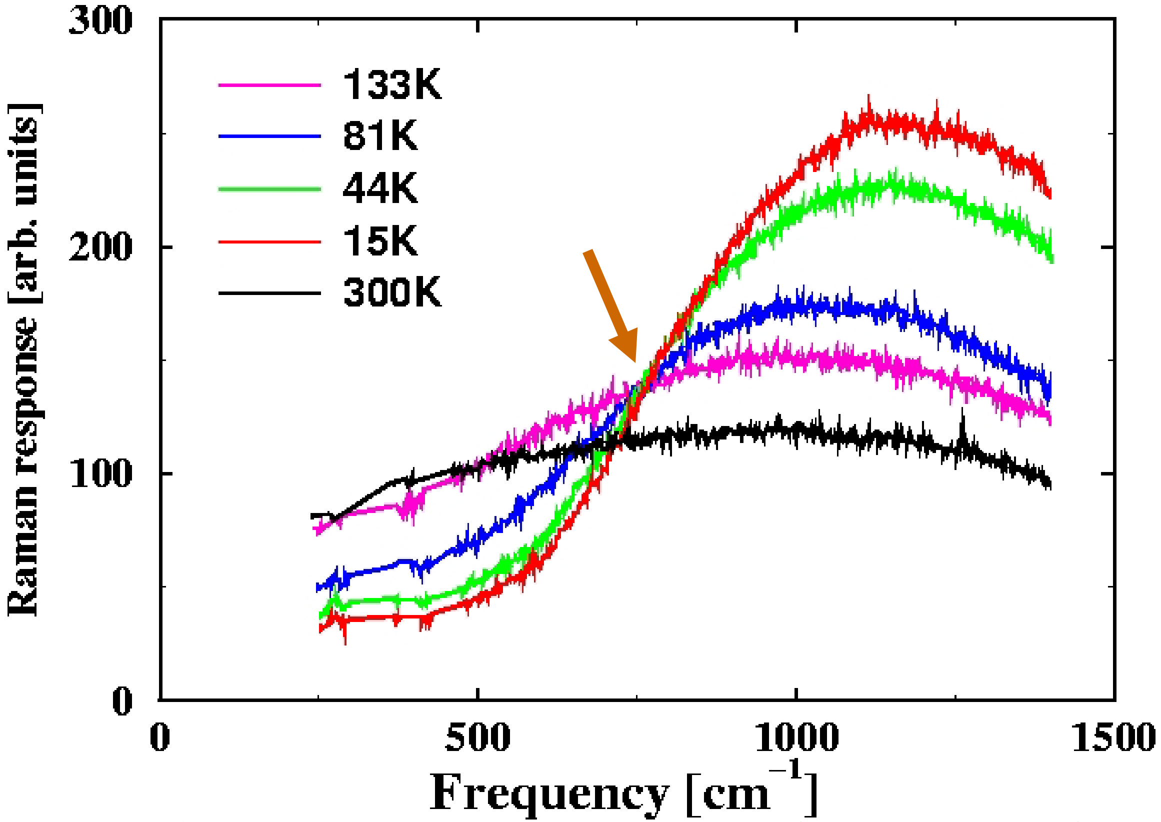
- B_{2g} - intensity largely **independent** of doping and temperature.
- B_{1g} - **loss** of low frequency spectral weight with underdoping.

Experimental data for the cuprates



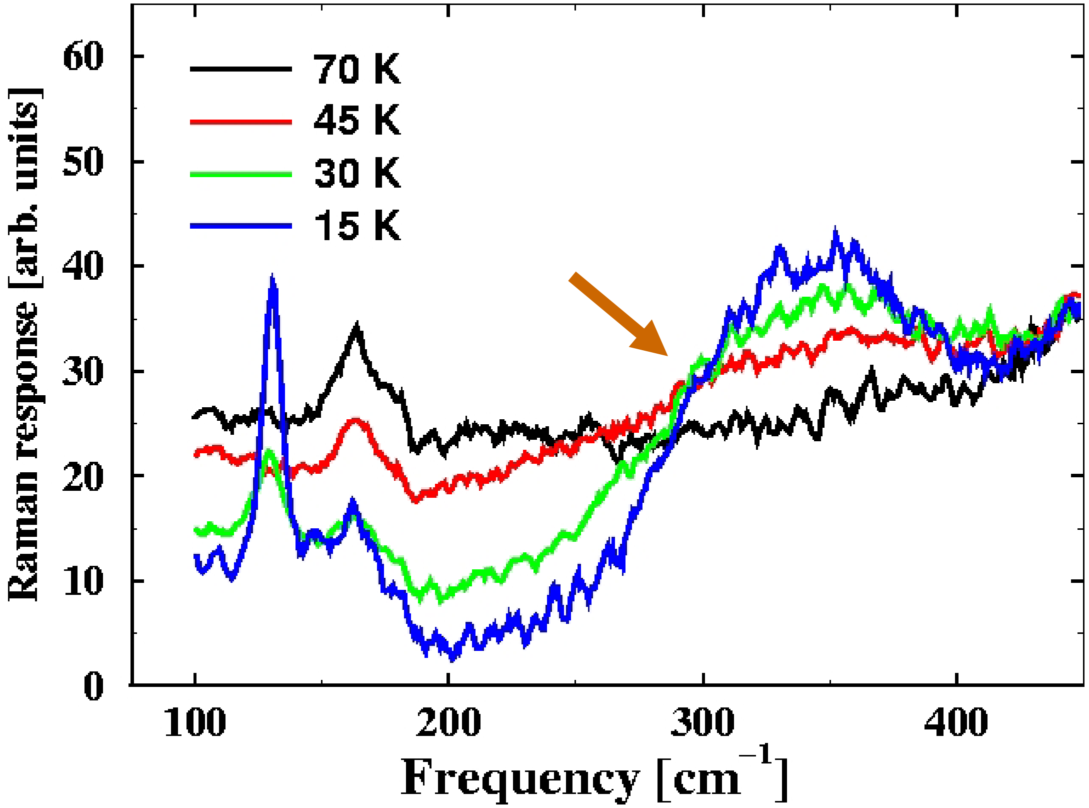
Data on underdoped Lanthanum Strontium Copper Oxide, from Chuck Irwin's group, shows the reduction of low-frequency spectral weight and the increase in the charge transfer peak, with an isosbestic point at about 2100 cm^{-1}.

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 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}.

Summary of Experimental Data

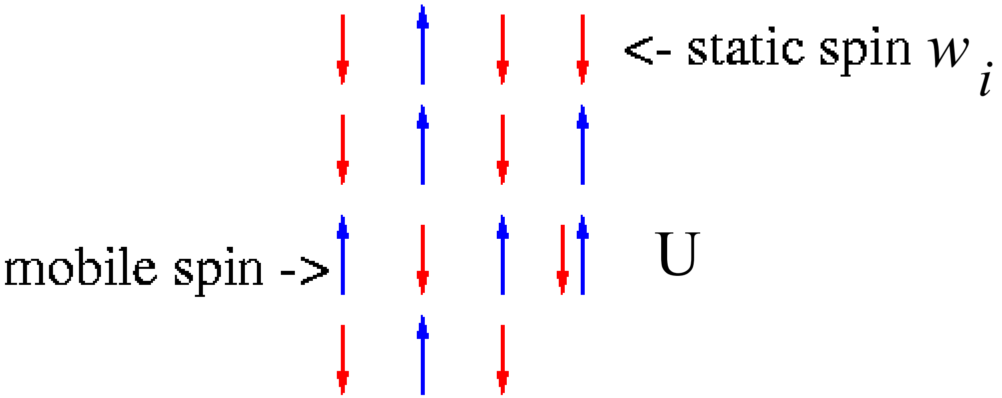
- Three characteristic behaviors are seen: (i) as T is lowered, there is a **redistribution of spectral weight** from low-frequency to high frequency; (ii) these regions are separated by an isosbestic point, where **the Raman response is independent** of T; (iii) the ratio of the twice spectral range where spectral weight is depleted to the onset temperature, where it first is reduced, is **much larger than 3.5** (typically 10-20).
- 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 Tc 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

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



<- static spin w_i

mobile spin ->

U

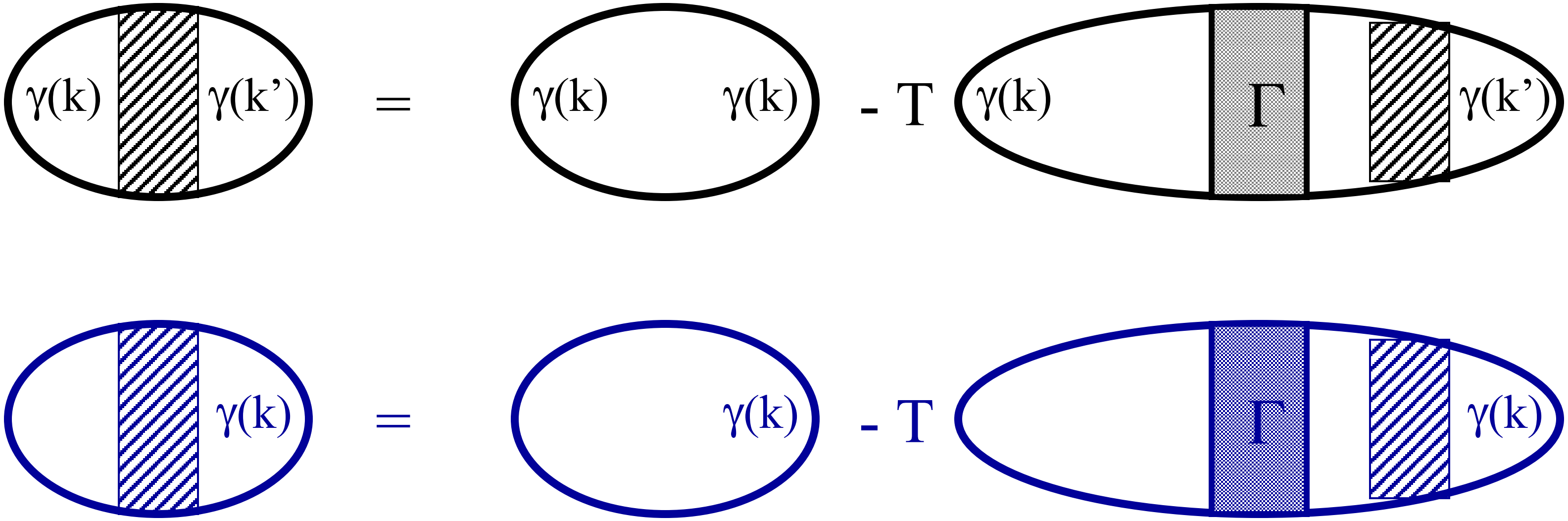
- **exactly solvable model** on a hypercubic lattice in infinite dimensions using dynamical mean field theory.
- possesses homogeneous, commensurate/incommensurate CDW phases, phase segregation, and **MI transitions**.
- ***Raman response can be constructed formally exactly***.

Formal Solution for 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



$\gamma(k) = -\,\varepsilon(k)$, 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 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.

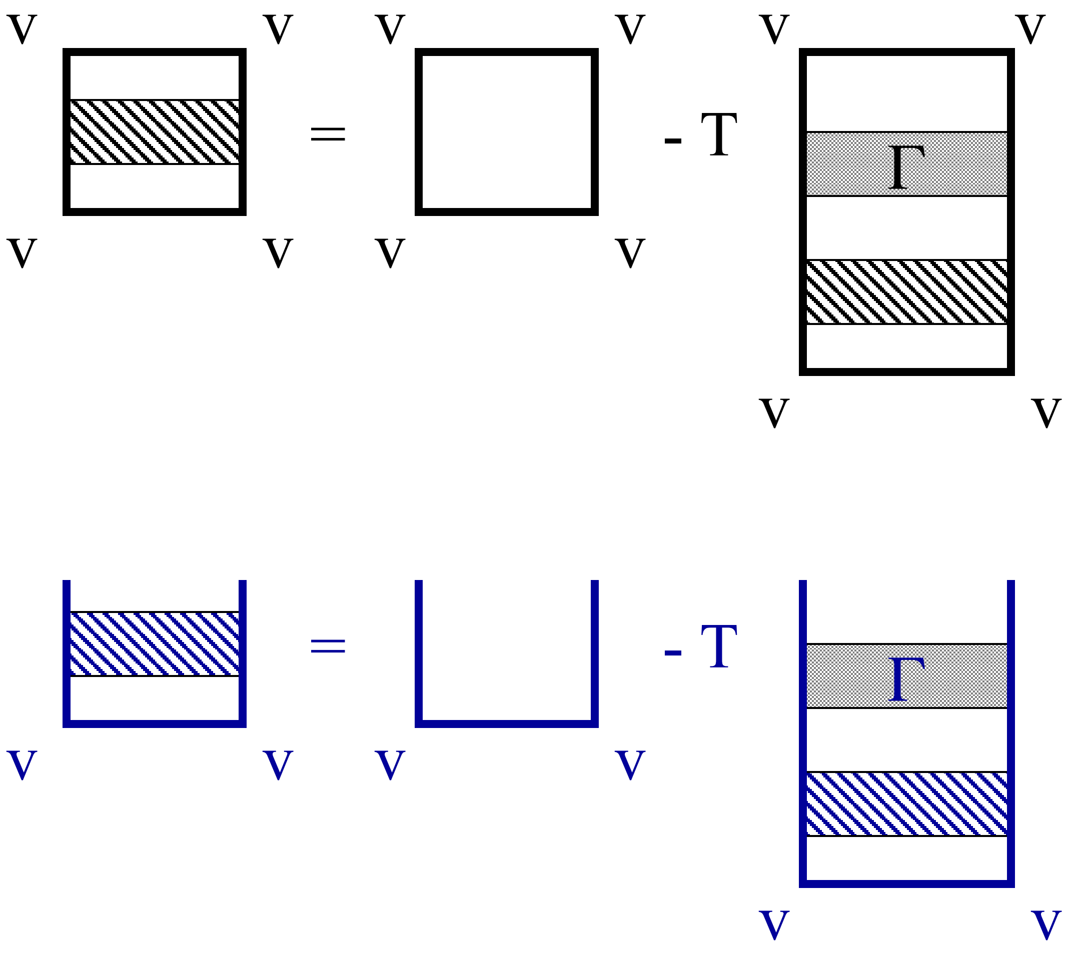
Formal Solution for Raman Scattering Response

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 **illustrate** schematic diagrams on the next page, but present no results for this process here.

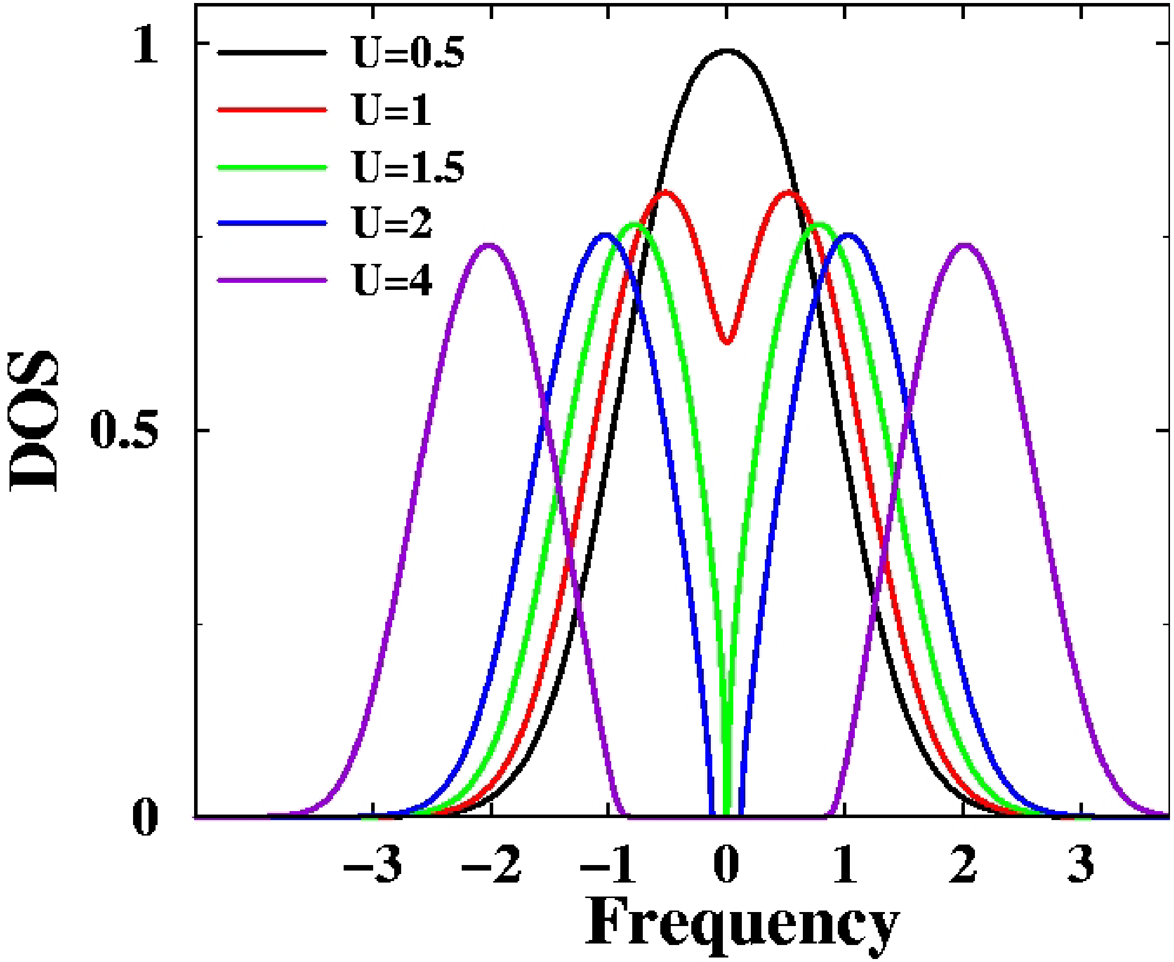
B_{2g} Raman scattering is purely resonant.

Diagrams for the B_{2g} Raman response



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

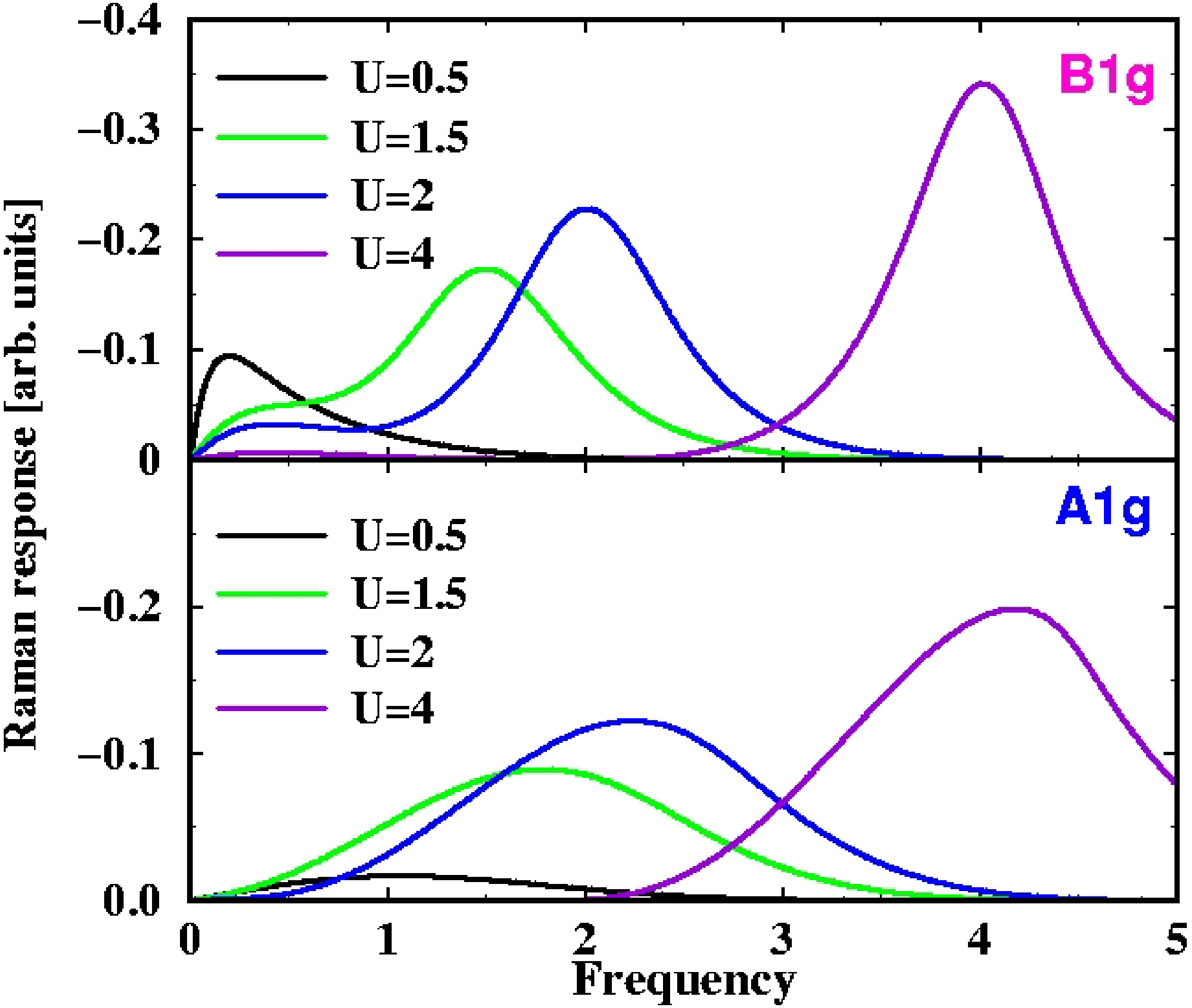
Metal-Insulator transition



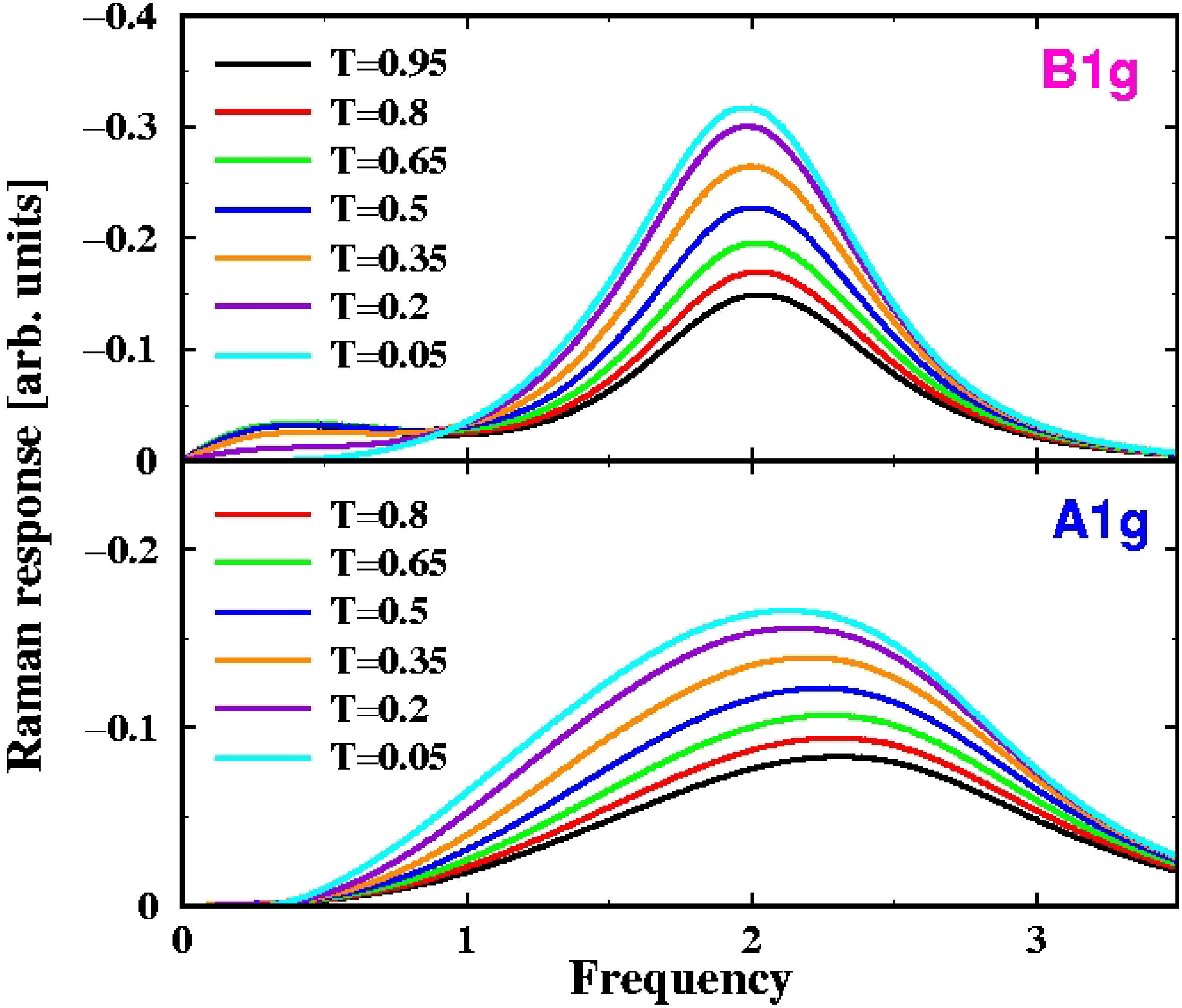
- **Correlation-induced** gap drives the single-particle DOS to zero at U=1.5
- Interacting DOS is **independent of T** in DMFT (Van Dongen, PRB, 1992)
- **Examine Raman response through the (T=0) quantum phase transition**.

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.



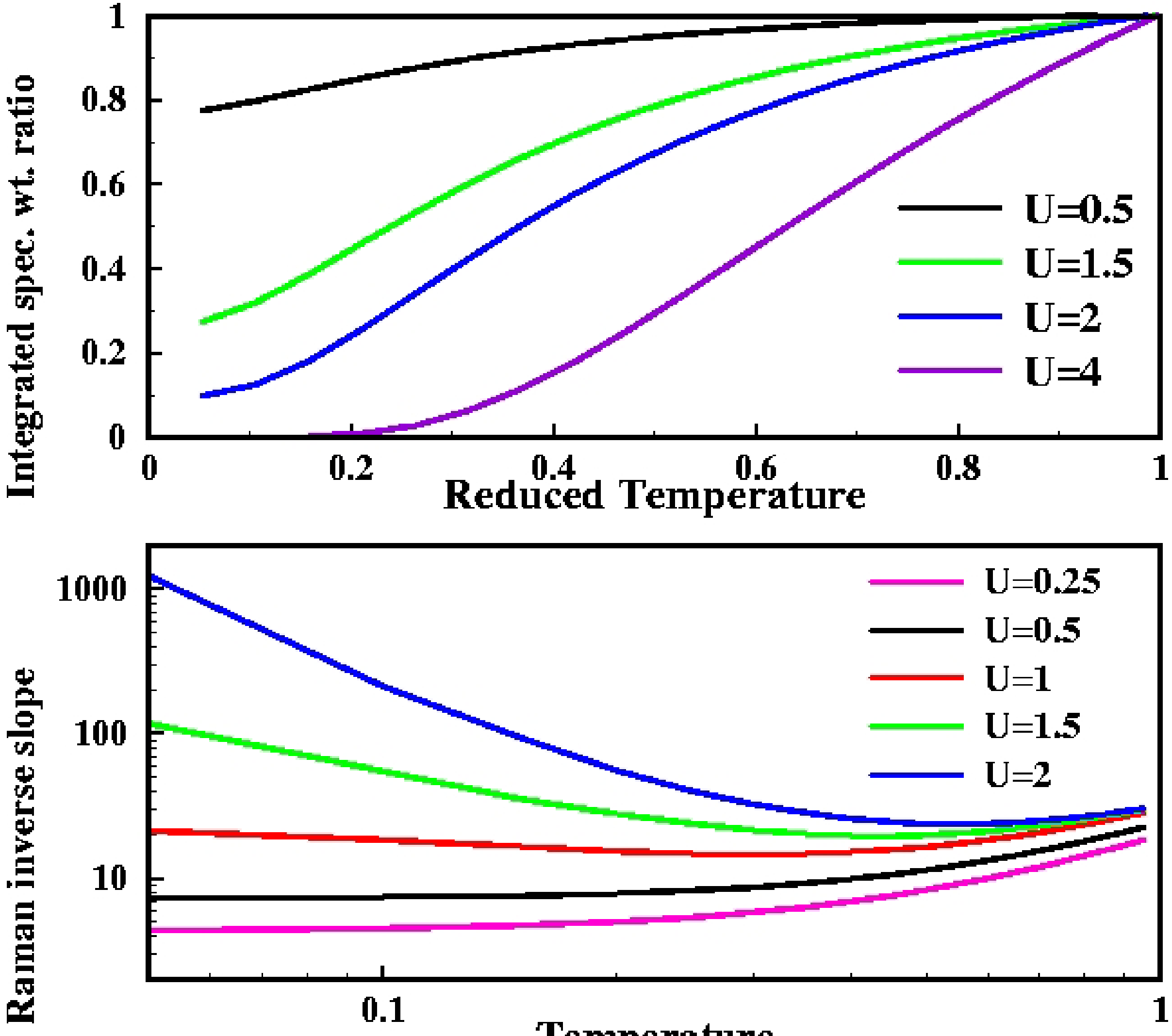
Nonresonant Raman Response (U=2)



- 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}).

Spectral weight and inverse Raman slope

- The B_{1g} Raman response is **sharply depleted** at low-T.
- The inverse Raman slope changes from **nearly constant** uncorrelated metallic behavior to a **rising** pseudogap or insulating behavior as the correlations increase.

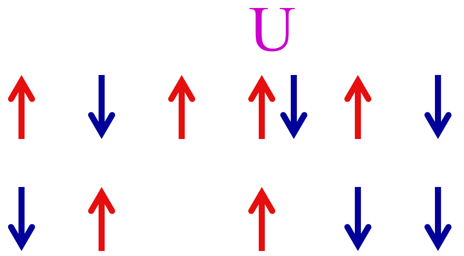


Summary (Falicov-Kimball model)

- 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

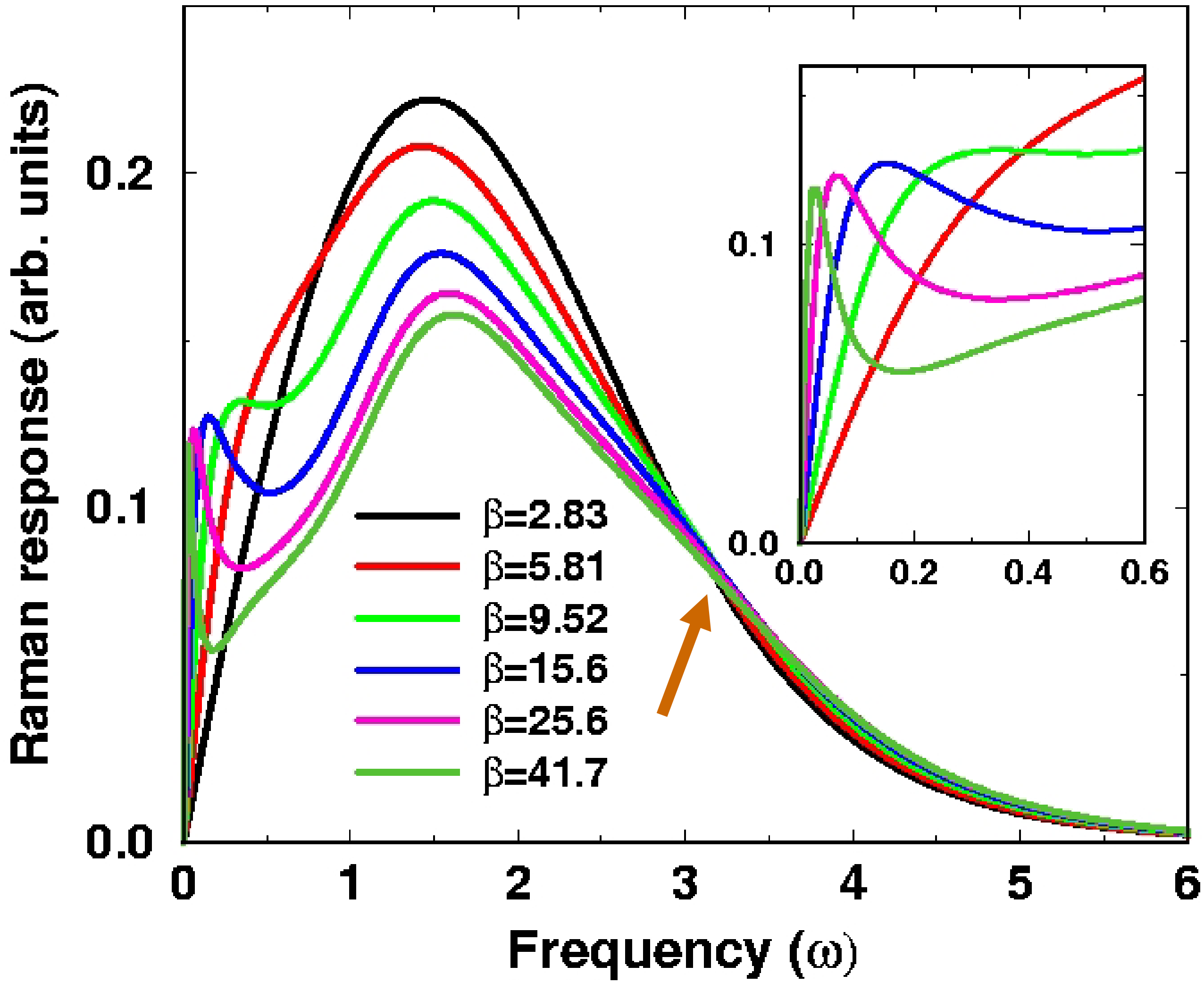
$$H = -\frac{t}{2\sqrt{d}} \sum c^{*}_{i\sigma} c_{j\sigma} + U \sum n_{i\uparrow} n_{i\downarrow}$$



Both electrons are now **mobile**

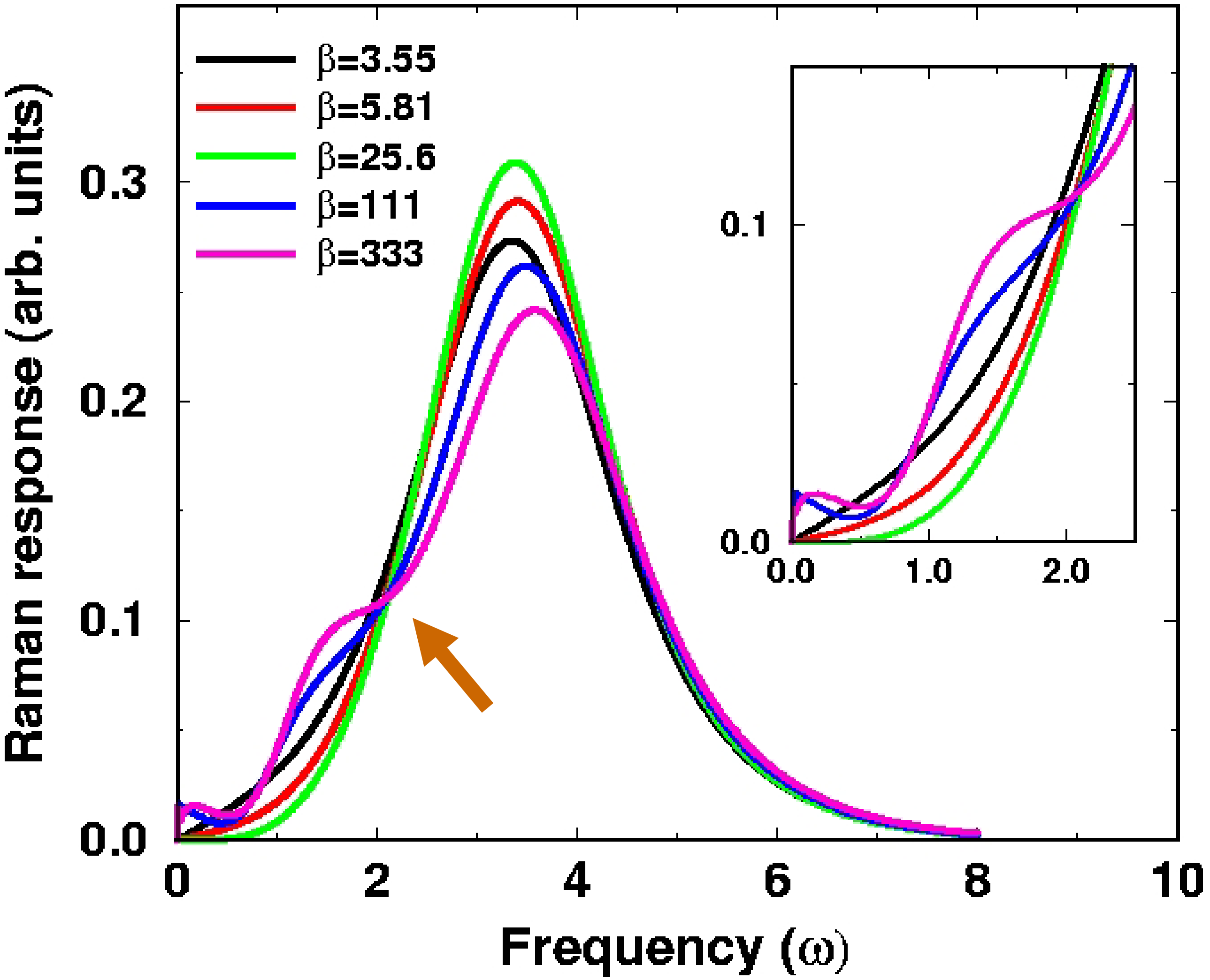
- **Exactly solvable** model on a hypercubic lattice in infinite dimensions using dynamical mean field theory (but requires NRG calculations to extract real frequency information).
- The irreducible charge vertex is problematic to calculate because it possesses too large a dynamic range for the max-ent 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)



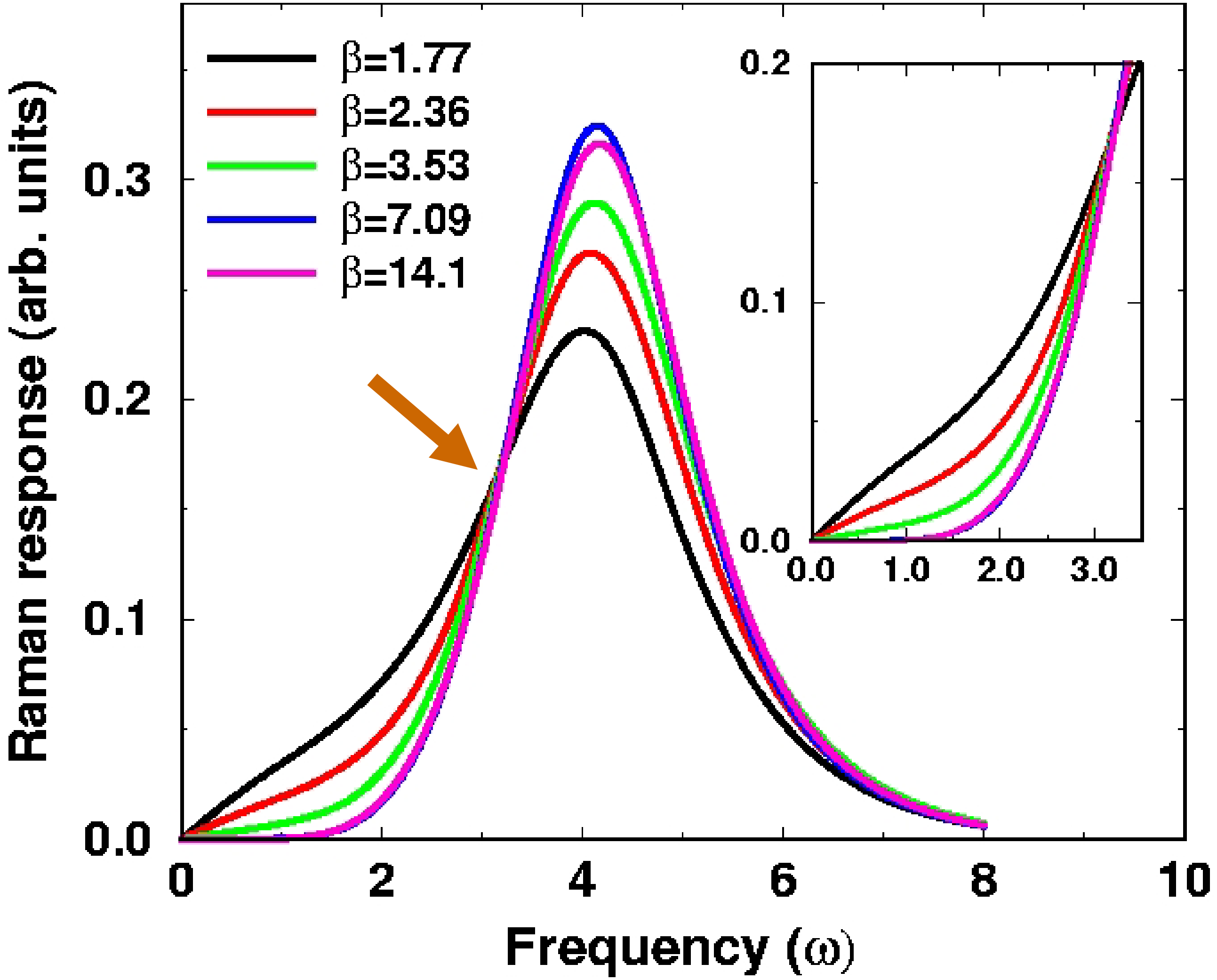
- Note the **charge transfer peak** as well as the **Fermi liquid peak** at low energy. As T goes to zero, the Fermi peak **sharpens** and **moves to lower energy**, as expected.
- There is **no low energy and low-T isosbestic point**, rather a high frequency isosbestic point seems to develop.

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-dependant.

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.
- Illustrated how **resonant** Raman scattering can also be solved in the Falicov-Kimball model (but presented no results).