Inelastic light scattering sum rules that relate to the potential energy of strongly correlated materials

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In collaboration with: Tom Devereaux, Andrij Shvaika, Oleg Vorobyov, Lance Cooper, and Ralf Bulla

History of the f-sum rule

- Discovered independently by **Thomas** and by **Kuhn** in 1925.
- In the original form, it showed that the **sum** of the oscillator strengths for the line spectra of a single electron **is equal to one**.
- The derivation is a simple consequence of the fact that ***the commutator of the momentum and position operators equals*** $i\hbar$.
- It was employed by Heisenberg in 1925 to **determine** the value of the commutator of momentum with position!
- In a system of many electrons, **it counts the total number of electrons**.

Application to correlated systems (superconductors)

Glover and Tinkham (1957) show that the **optical conductivity is suppressed** in a superconductor due to the coherent formation of electron pairs.

PHYSICAL REVIEW VOLUME 108, NUMBER 2 OCTOBER 15, 1957

Conductivity of Superconducting Films for Photon Energies between 0.3 and $40kT_c$*

R. E. GLOVER, III,† *University of California, Berkeley, California and University of North Carolina, Chapel Hill, North Carolina*

AND

M. TINKHAM, *University of California, Berkeley, California*

(Received May 17, 1957)

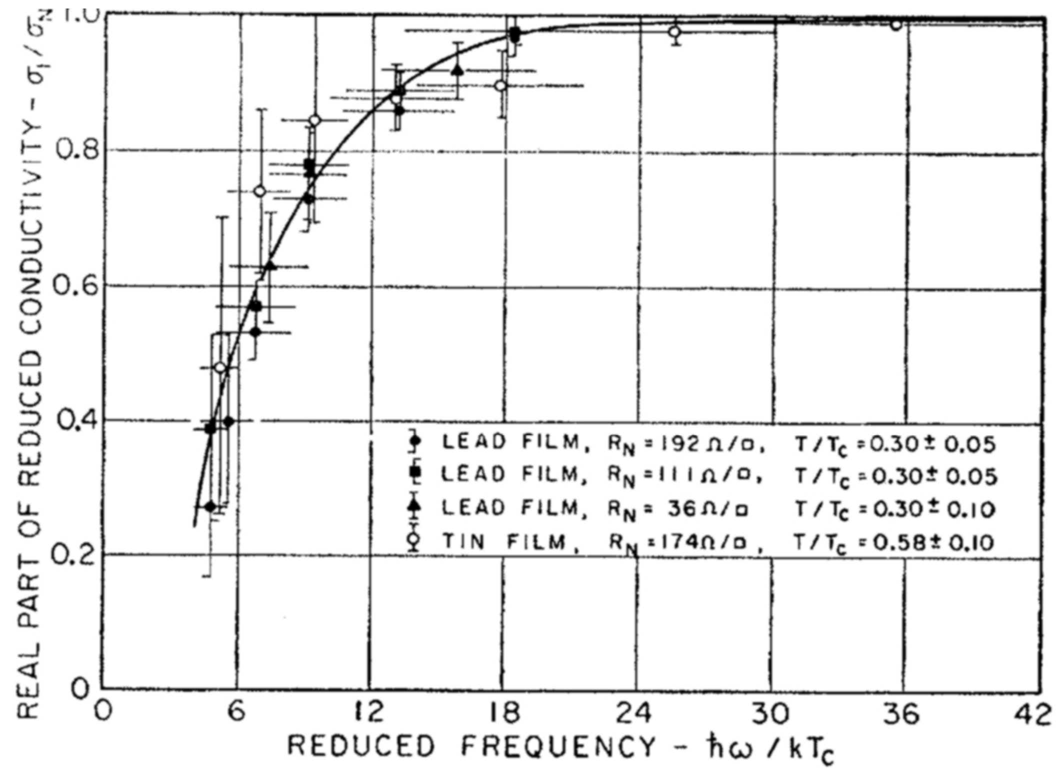


FIG. 6. Frequency dependence of σ_1/σ_N for several lead and tin films in the superconducting state.

Application to correlated systems (superconductors)

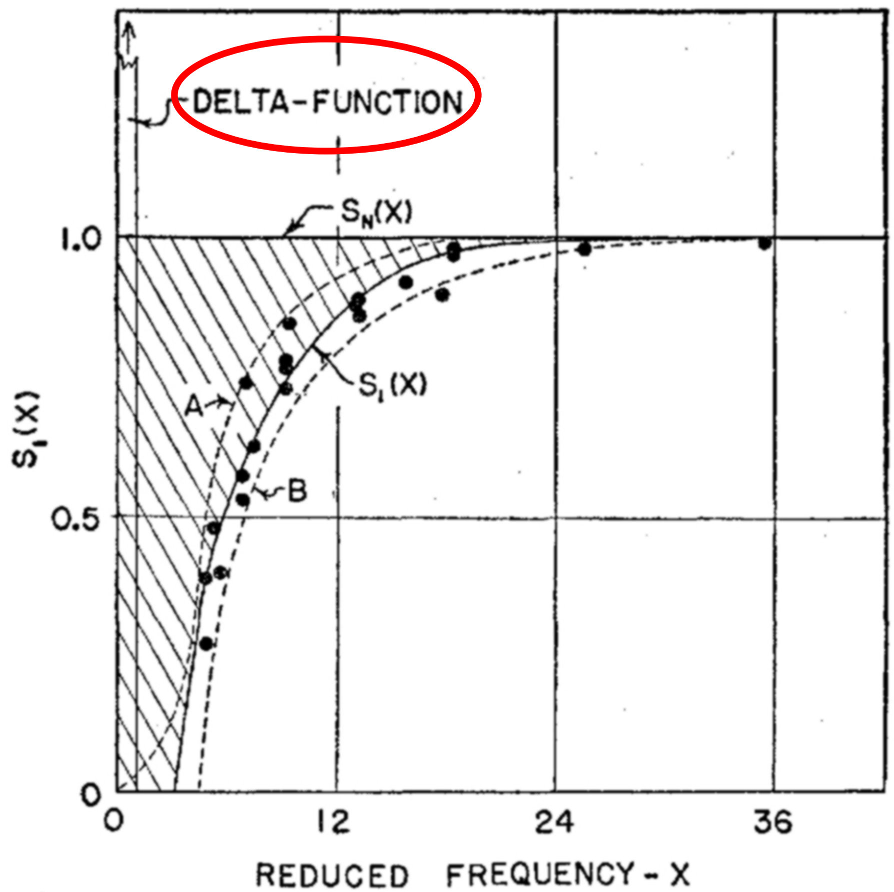
Conductivity of Superconducting Films:
A Sum Rule

Richard A. Ferrell, *University of Maryland, College Park, Maryland*

AND

Rolfe E. Glover, III, *University of North Carolina, Chapel Hill, North Carolina*

(Received December 23, 1957)



Ferrell and Glover (1957) show that the f-sum rule implies the **“lost” spectral weight is recovered in a zero frequency delta function**.

Low-energy sum rule relates to the kinetic energy

PHYSICAL REVIEW B VOLUME 16, NUMBER 6 15 SEPTEMBER 1977

Optical spectrum of a Hubbard chain

Pierre F. Maldague
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
(Received 18 April 1977)

Maldague shows that if we restrict the elastic light scattering to a single band, then the f-sum rule does not count the total electrons but rather **measures the average (correlated) kinetic energy**.

Hence the sum rule picks up a small **temperature dependence**.

Hence the integral of the optical conductivity (with a low-energy cutoff) can be employed to *measure the average kinetic energy* of a strongly correlated material.

Application to high Tc superconductors

Superconductors that change color when they become superconducting

J.E. Hirsch
Department of Physics, University of California, San Diego, La Jolla, CA 92093-0319, USA

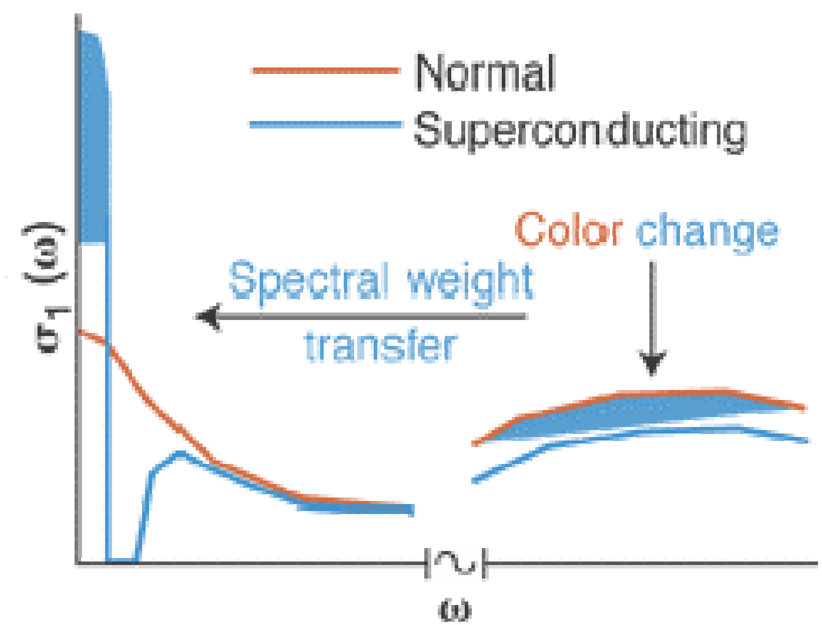
Received 10 August 1992

SCIENCE'S COMPASS

PERSPECTIVES: SUPERCONDUCTIVITY

The True Colors of Cuprates

J. E. Hirsch



Hirsch predicts that strong correlation effects in high Tc superconductors will **lead to shifts of spectral weight at high energies**, so that a simple application of the Ferrell-Glover sum rule will fail.

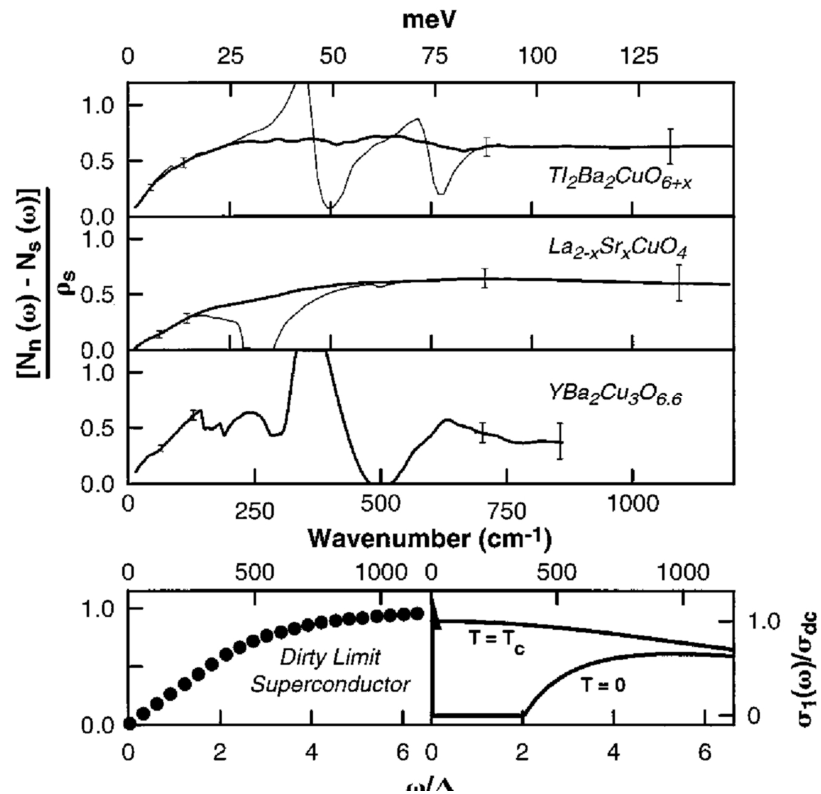
Experiments in high Tc superconductors (I)

Basov and collaborators were the first to see that the **Ferrell-Glover sum rule was not satisfied** at low energies in the high Tc compounds.

REPORTS

Sum Rules and Interlayer Conductivity of High-T_c Cuprates

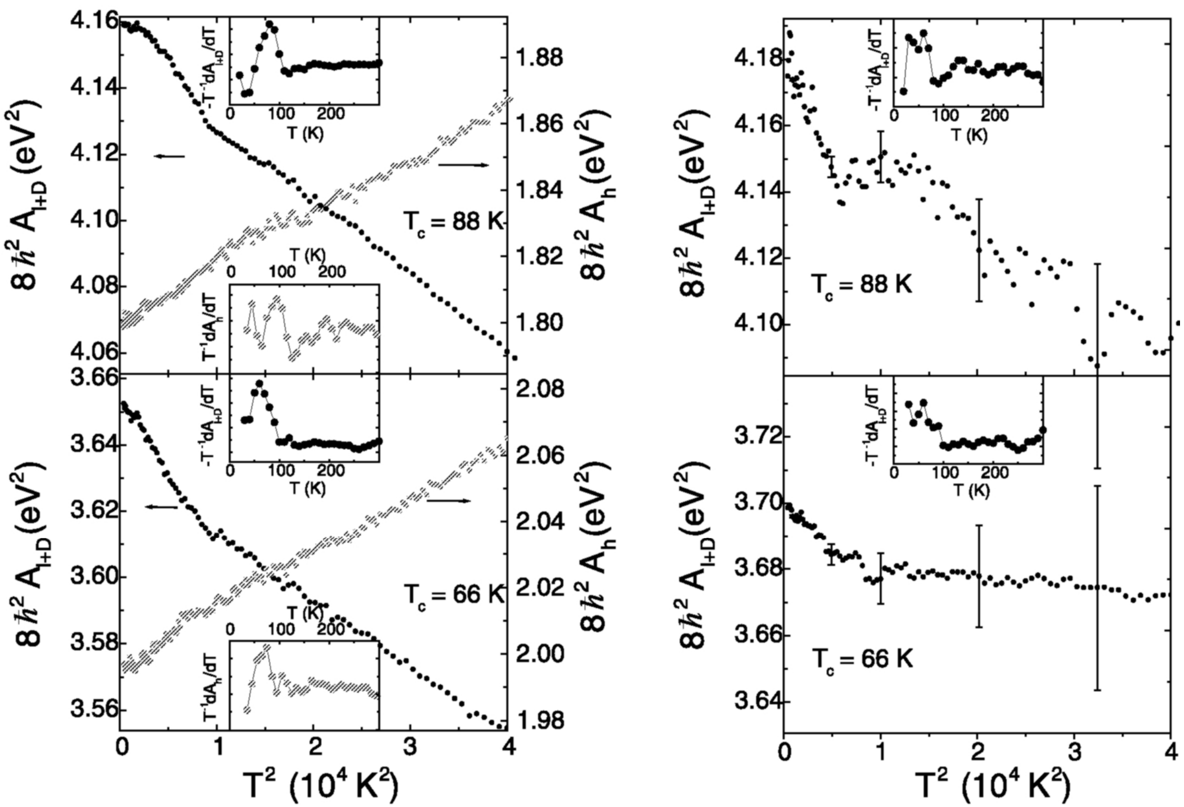
D. N. Basov, S. I. Woods, A. S. Katz, E. J. Singley, R. C. Dynes, M. Xu,* D. G. Hinks, C. C. Homes, M. Strongin



Experiments in high Tc superconductors (II)

Superconductivity-Induced Transfer of In-Plane Spectral Weight in $Bi_2Sr_2CaCu_2O_{8+\delta}$

H. J. A. Molegraaf,[1] C. Presura,[1] D. van der Marel,[1*] P. H. Kes,[2] M. Li[2]



Van der Marel and collaborators were able to show definitively this **high-energy spectral weight transfers** in the BSCCO compounds.

Relationship between the optical conductivity and nonresonant Raman scattering

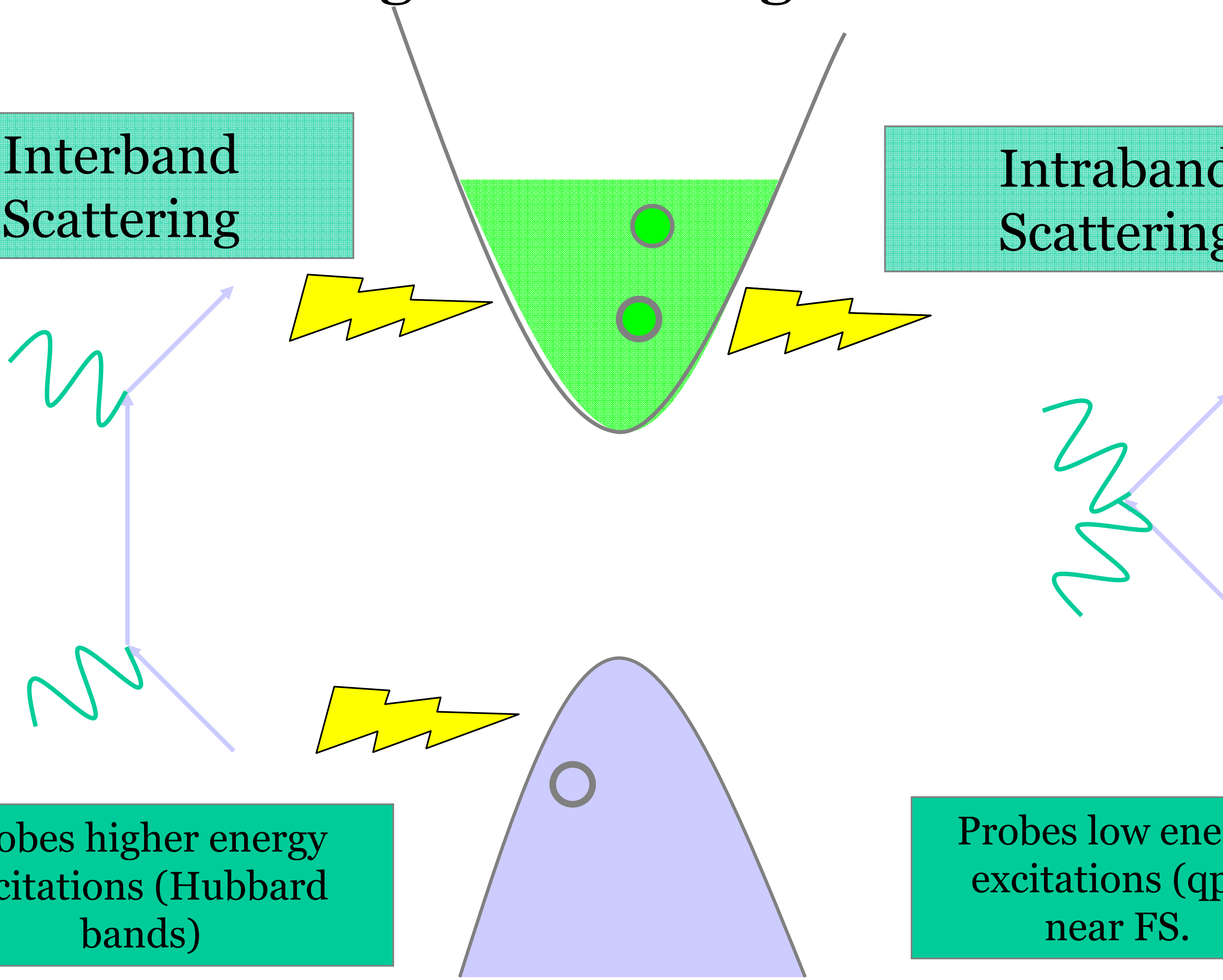
Shastry and Shraiman (1991) hypothesized that there was a **direct relationship** between the optical conductivity and the so-called nonresonant B_{1g} Raman scattering.

Freericks and Devereaux (2001) **proved** that the Raman scattering response function is **equal to the optical conductivity multiplied by the frequency** in dynamical mean field theory.

Inelastic light scattering of electrons

Interband Scattering

Intraband Scattering

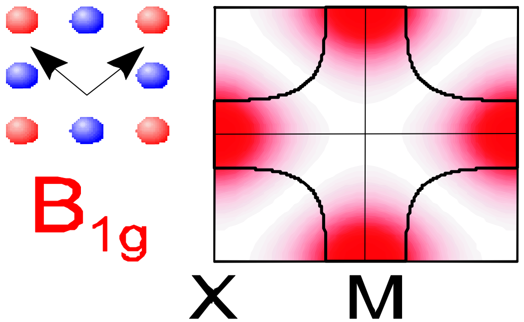


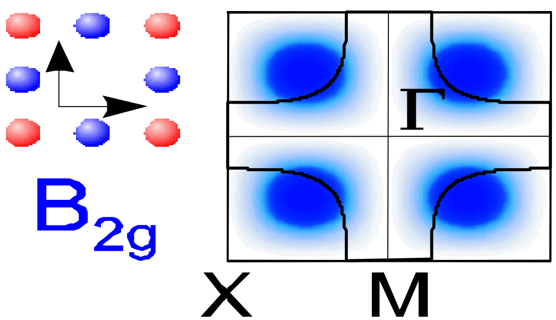
Probes higher energy excitations (Hubbard bands)

Probes low energy excitations (qps) near FS.

Polarization plays a key role in solids

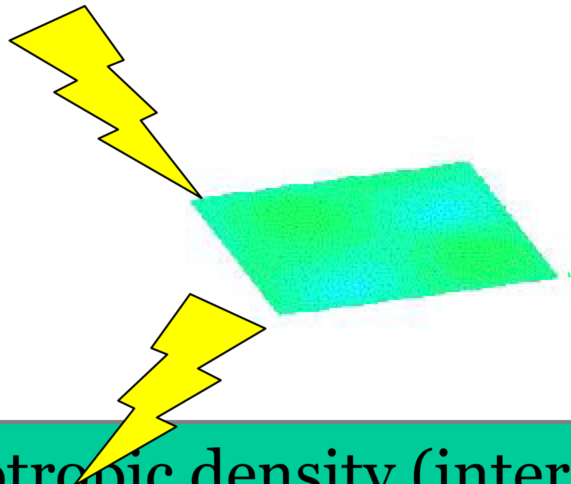
- Polarization orientations transform according to point group symmetry operations, and so do the excitations they create – described by the scattering vertex **γ(k)**.
- A_{1g}: $\gamma(k)$ ~ **isotropic**
- B_{1g}, B_{2g} : $\gamma(k)$ ~ **anisotropic**
- ***Project out excitations in different regions of the BZ.***





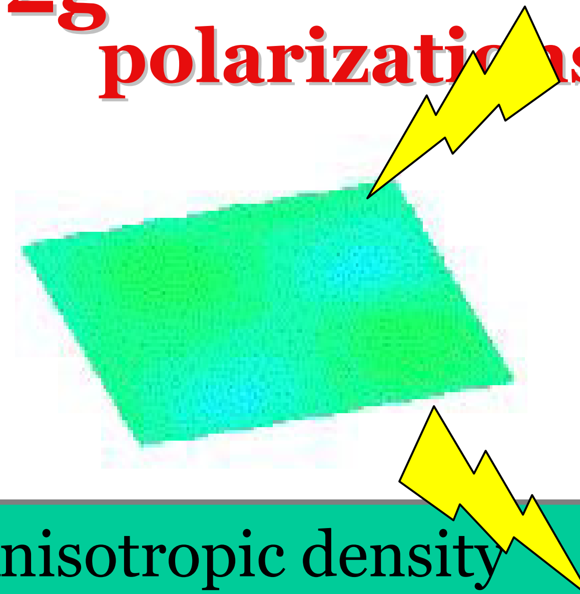
Polarizers determine the type of charge excitation

A_{1g} (parallel polarizations)



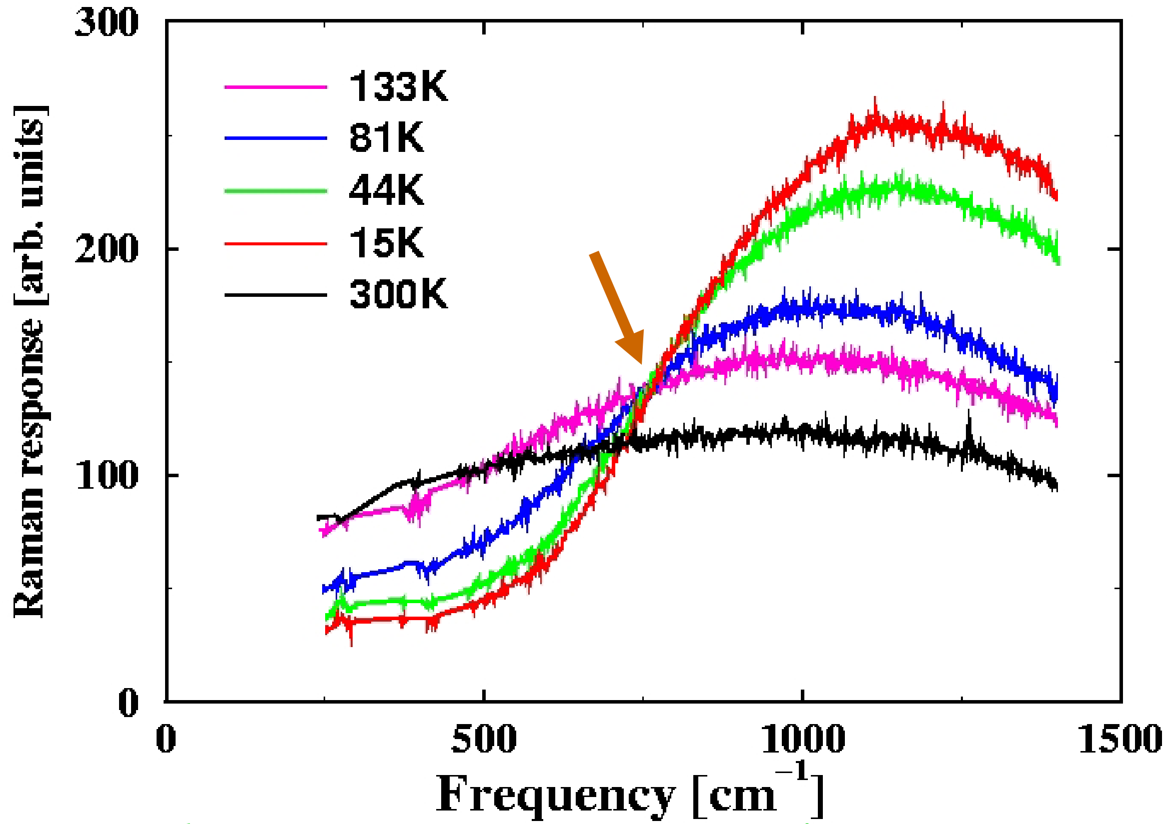
Isotropic density (intercell) fluctuations – couple to long-range Coulomb interactions ~ Im (1/ε)

B_{2g} (crossed polarizations)



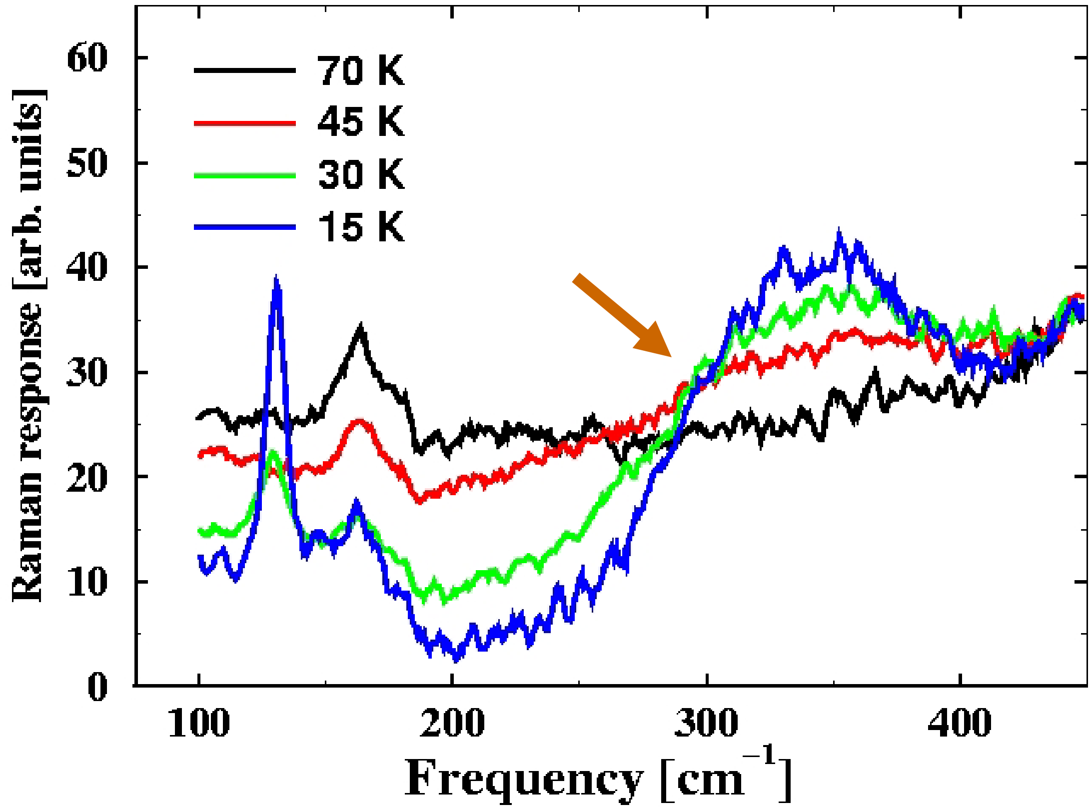
Anisotropic density (intracell) fluctuations – couple to short-range Coulomb interactions.

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



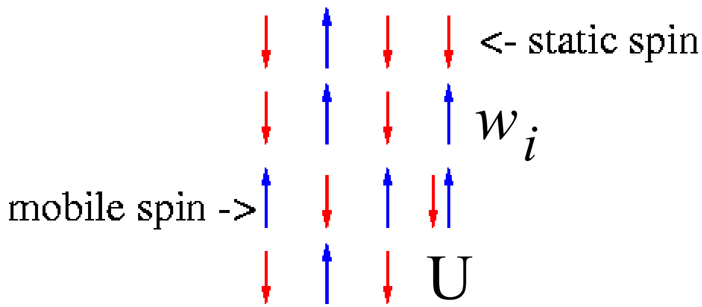
- *Nyhus et al, 1995 and 1997* Raman scattering on **SmB_6**. Note the appearance of the **isosbestic point** near 300 cm^{-1}.
- Below 30K, there is an **increase** in low frequency spectral weight in a narrow peak at about 130 cm^{-1}.

Summary of Experimental Data (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.

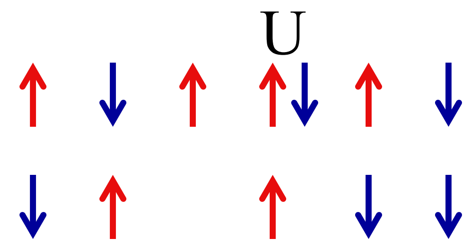
Spinless Falicov-Kimball Model

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



Hubbard Model

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



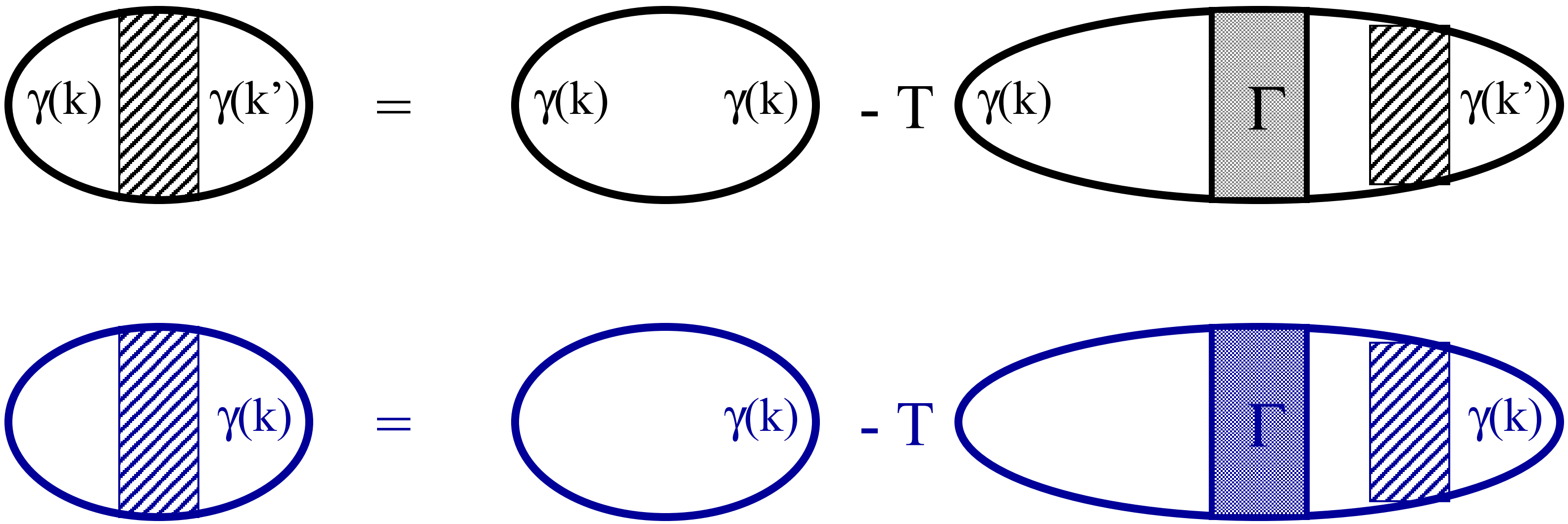
Both electrons are now **mobile**

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

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

•***Inelastic light scattering can be constructed formally exactly***.

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. (FK model only)

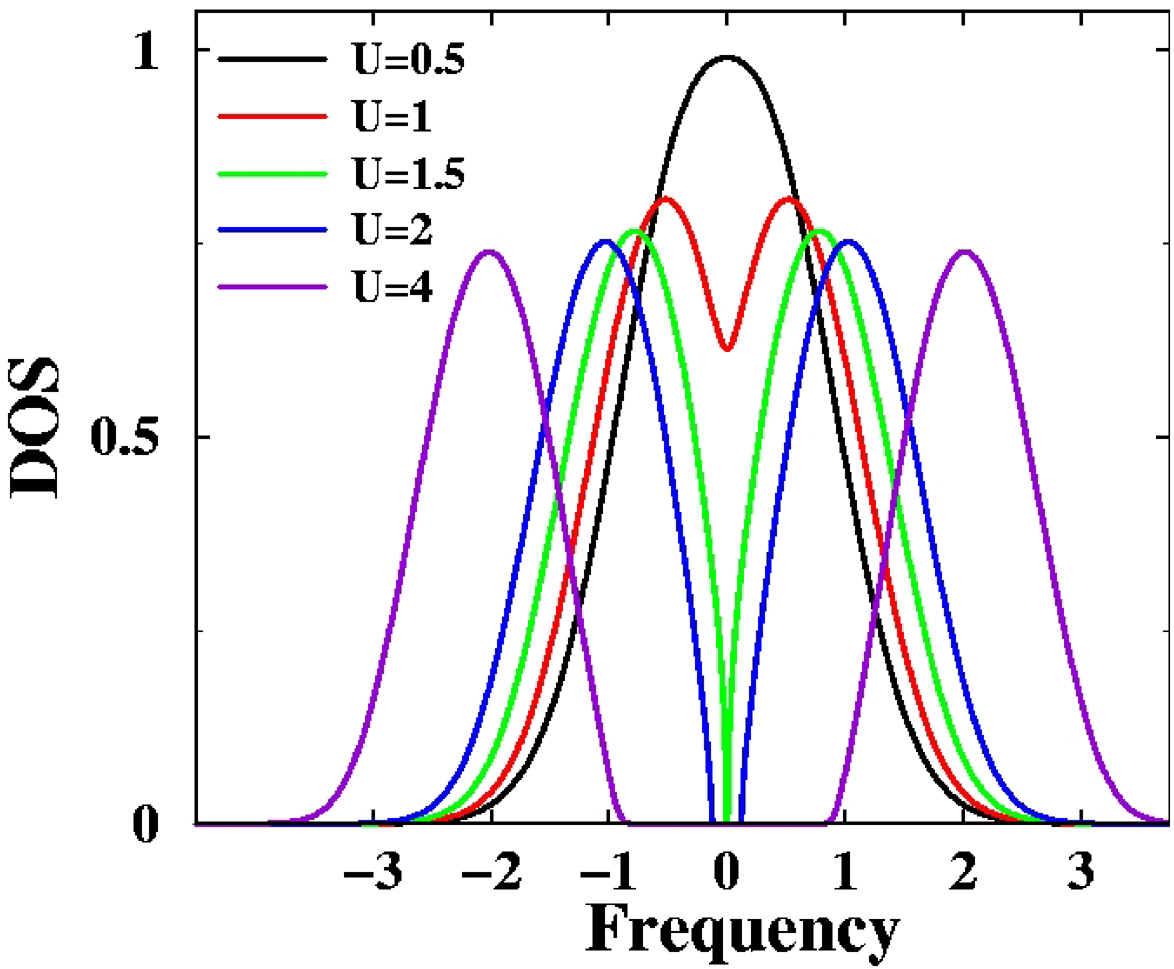
Formal Solution for the Light Scattering Response

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).
- This Raman (**q=0**) response is **identical** to that of the optical conductivity multiplied by one factor of frequency (Shastry and Shraiman, PRL, 1990).

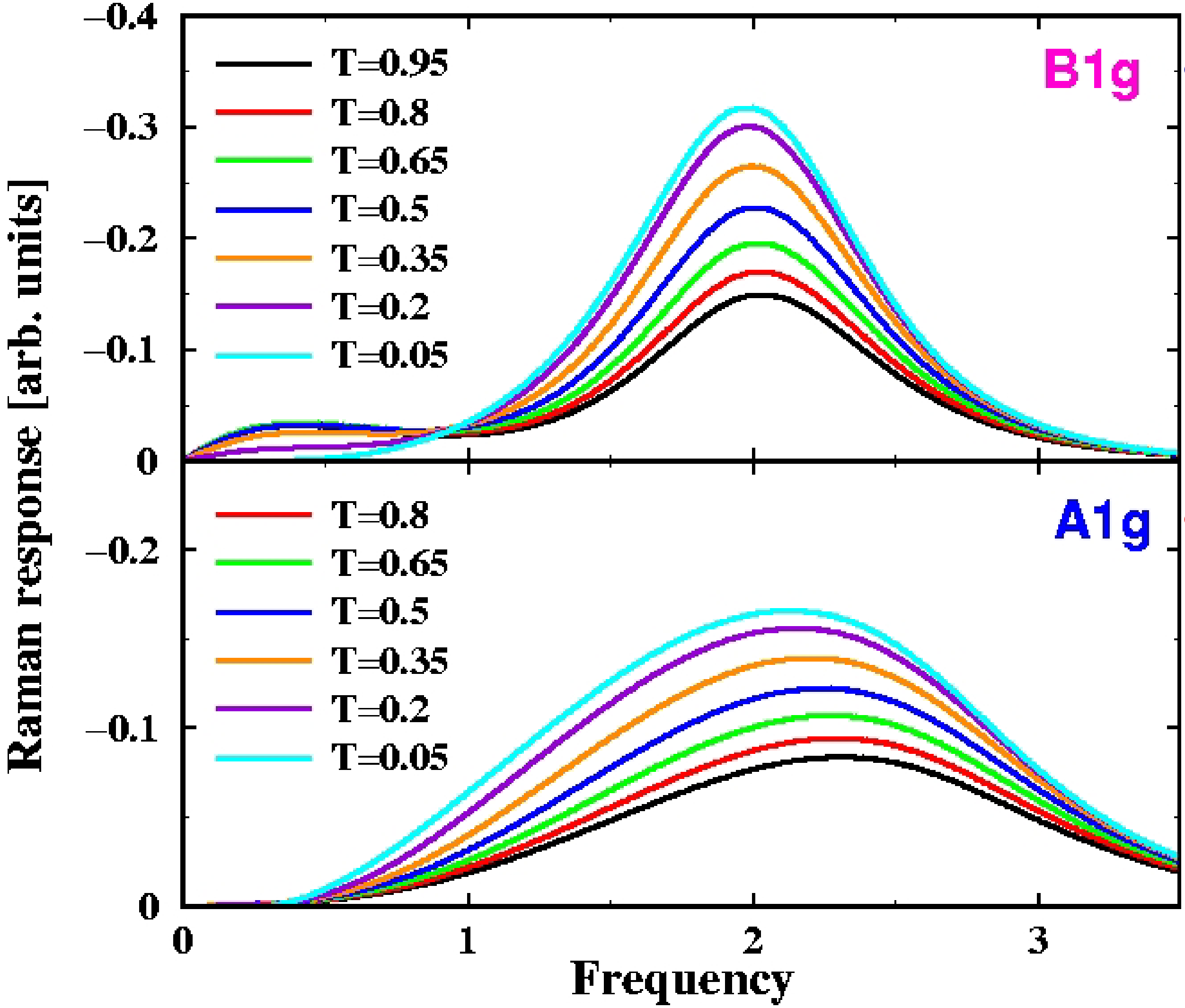
The nonresonant B_{1g} Raman response is closely related to the optical conductivity.

FK model Metal-Insulator transition (NFL)



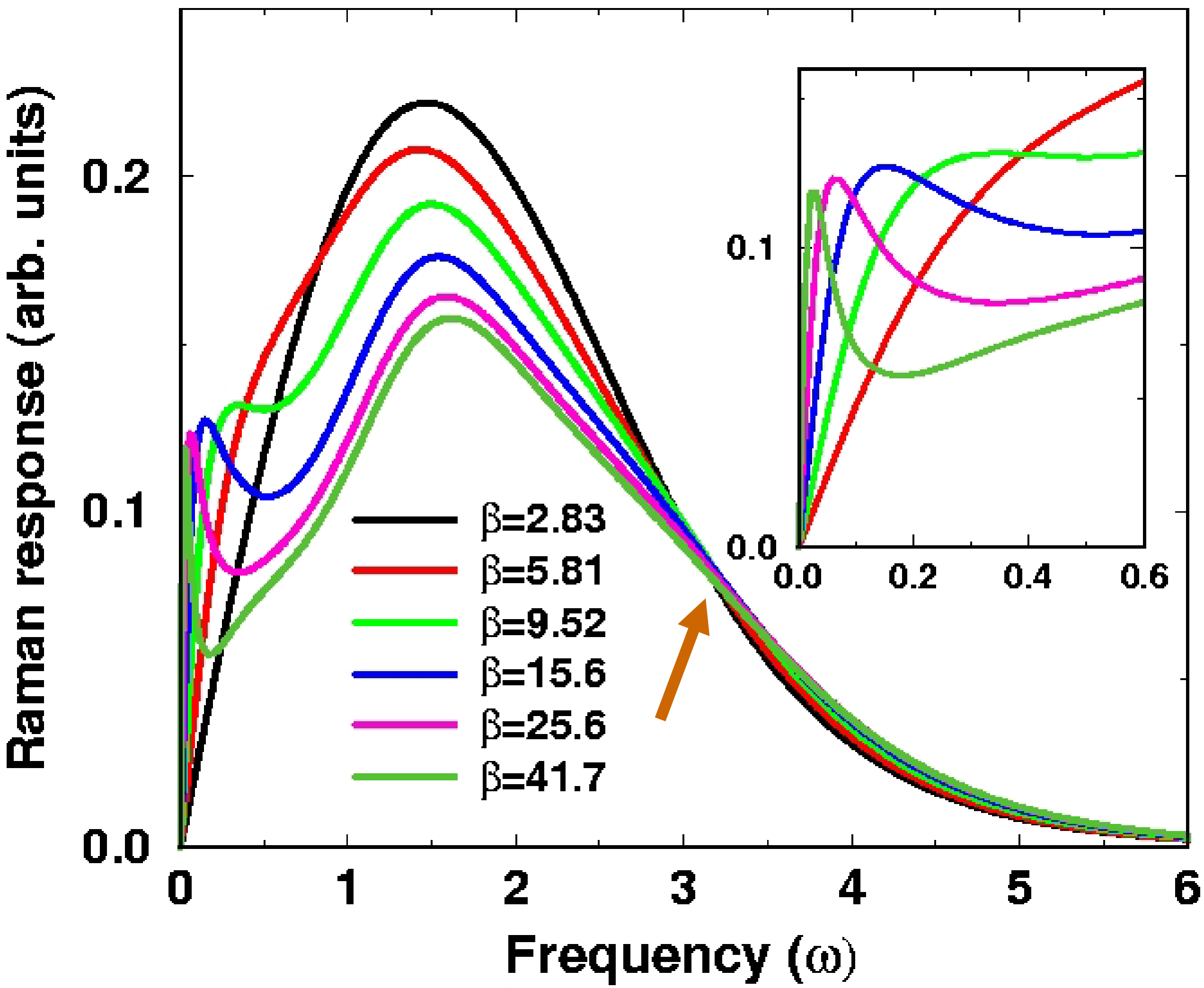
- **Correlation-induced** gap drives the single-particle DOS to zero at ω=0 for U>√2
- Interacting DOS is **independent of T** in DMFT (Van Dongen, PRB, 1992)
- ***Examine Raman response through the (T=0) quantum phase transition.***

Nonresonant Raman Response (U=2)



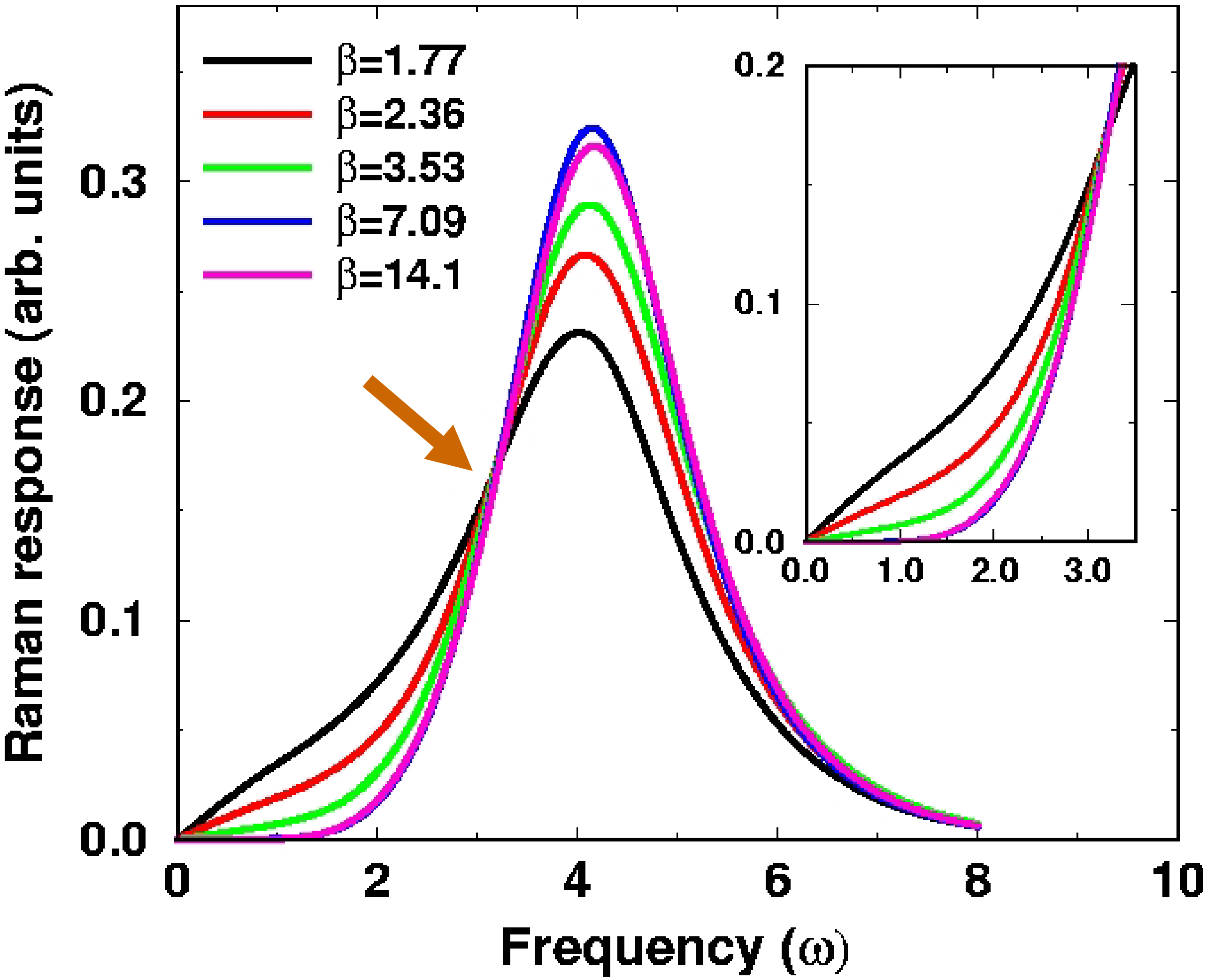
- The low-frequency B_{1g} response develops at a low temperature over a **wide frequency range** of O(1).
- An **isosbestic point** divides where spectral weight increases or decreases as T is lowered (B_{1g}).

Nonresonant B_{1g} Raman (Hubbard metal)



- 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.

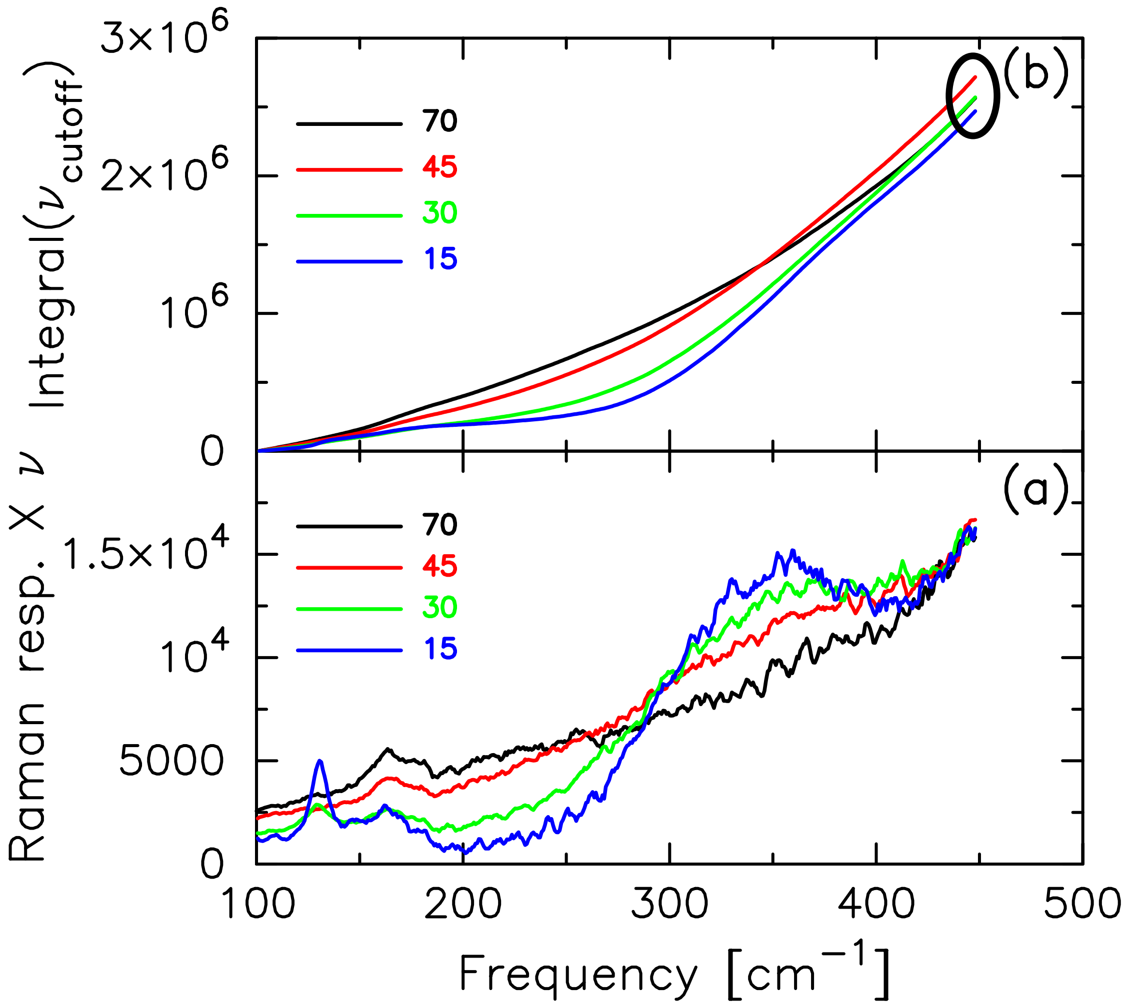
Nonresonant B_{1g} Raman (Hubbard insulator)



- 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.

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.



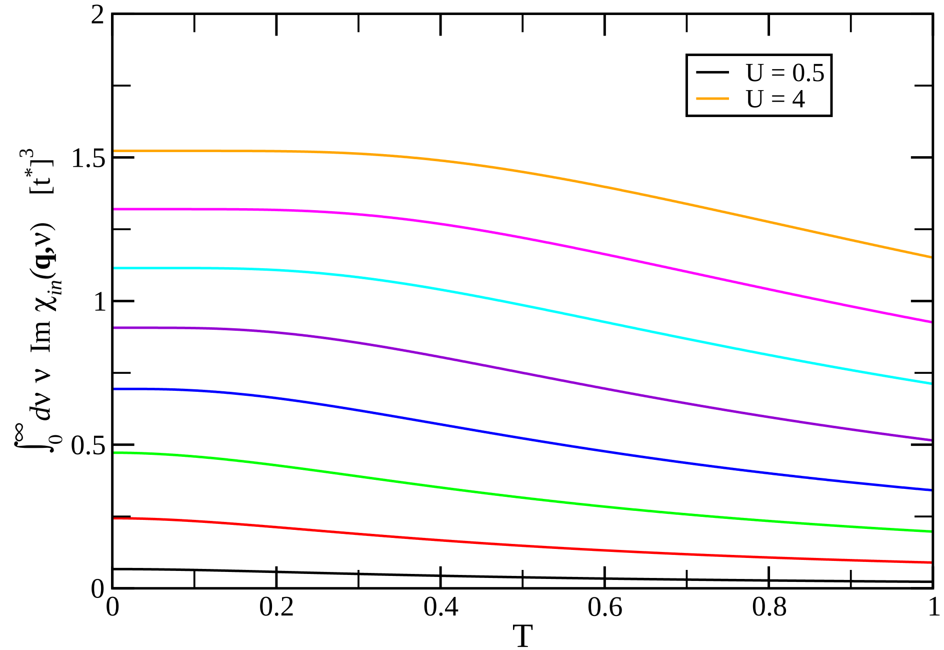
Theory for the sum rule

- A simple analysis of the nonresonant response function shows the **first moment** of the Raman signal is proportional to $\langle[\rho_{Raman},[H,\rho_{Raman}]]\rangle$, 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 commutator with the **potential energy**!

Hence Raman scattering can be employed to measure quantities *related to* the average potential energy of a strongly correlated material.

Results for the

The sum rule for the FK model is **almost constant** as a function of T at low T.



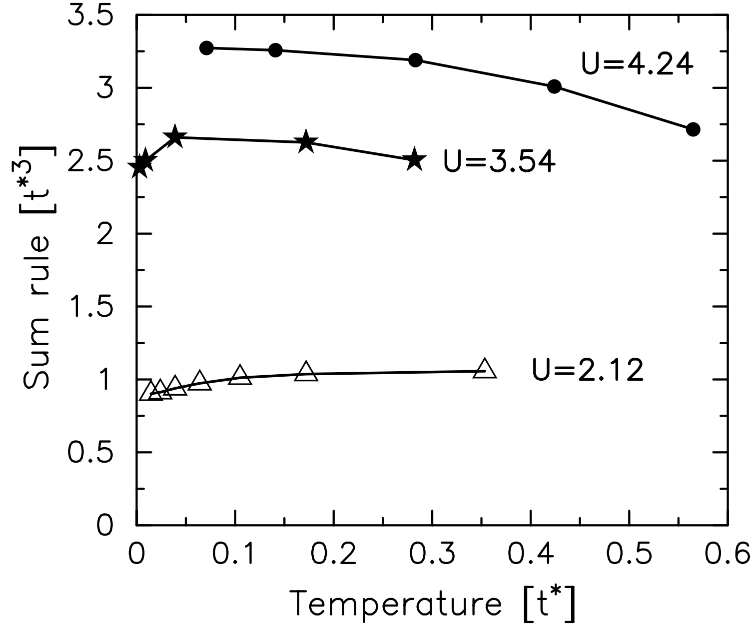
$$-\frac{\pi}{2Z}Tr\{e^{-\beta H}\frac{Ut^{*2}}{2dN}\sum_{i\delta\delta'}c_i^{*}c_{i+\delta+\delta'}e^{-iQ(\delta+\delta')}\ [w_i\ -w_{i+\delta}-w_{i+\delta'}+w_{i+\delta+\delta'}]\}$$

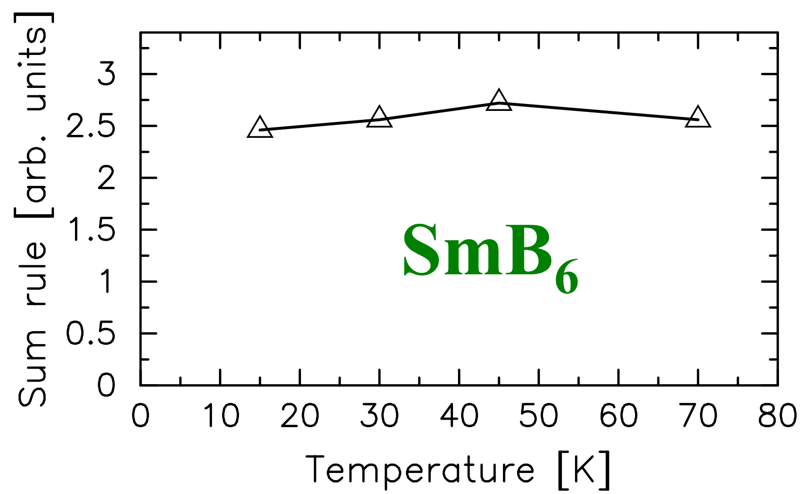
A_{1g} corresponds to **Q**=0; B_{1g} to **Q**=(π,0,π,0,…)

The potential explicitly energy enters when δ+δ‘=0

Results for the sum rule (Hubbard)

- For the Hubbard model, the sum rule appears to have a **low-T decline**.
- This behavior is similar to SmB_6.
- The sum rule is more complicated than the FK model, involving **spin-flip terms** in addition to potential energy and more complicated terms.





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.
- 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.