Optical spectroscopy study on the charge dynamics of URu2Si2

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- **Some basics about optical spectrscopy**
  Optical constants, Kramers-Kronig transformation, inter- and intra-band transitions

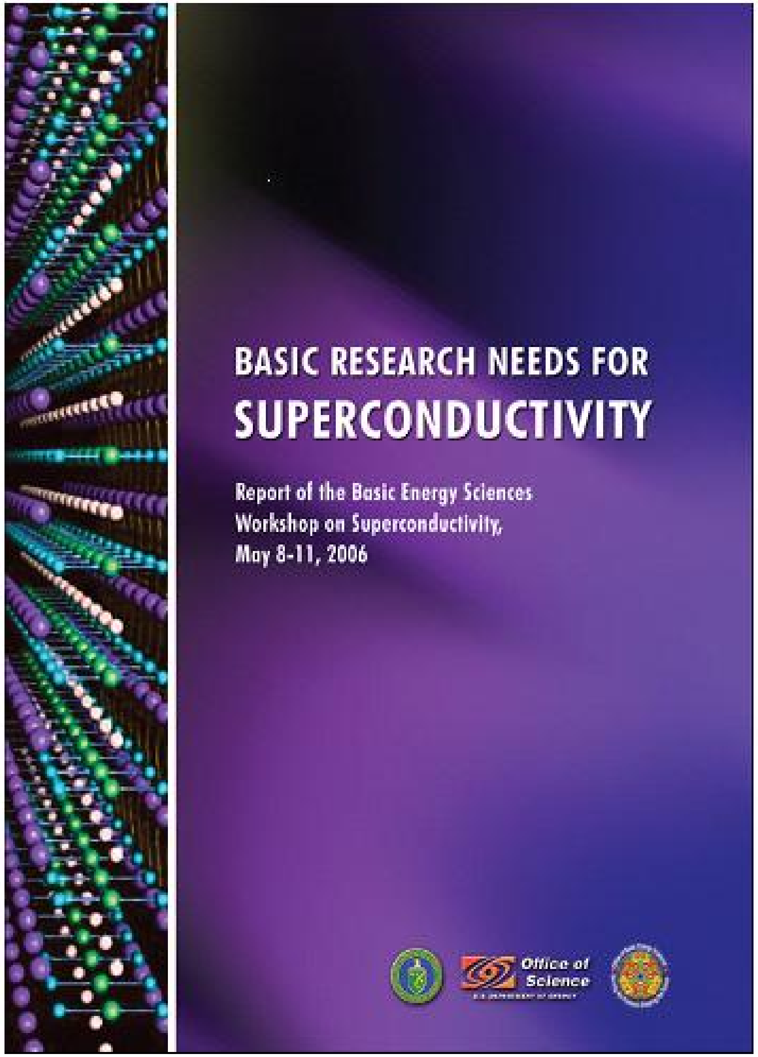
- **Example of application:**
  URu2Si2

Collaborators:

W. T. Guo, Z. G. Chen (optical measurements)

G. Luke (single crystals)

Some basics about optical spectroscopy technique



Emerging Experimental Techniques and Opportunities

- *Angle-Resolved Photoemission Spectroscopy (ARPES).* Wavelike quantum states of the electrons are defined in momentum space (*k*-space). ARPES allows direct determination of the complete momentum-space electronic structure, *A*(*k*, *E*), with remarkable energy and momentum resolution.

- *Spectroscopic Imaging-Scanning Tunneling Microscopy (SI-STM).* This is the complementary technique to ARPES that allows mapping of the energy-resolved quantum states in real space (*r*-space) with atomic resolution and yet over large sample areas.

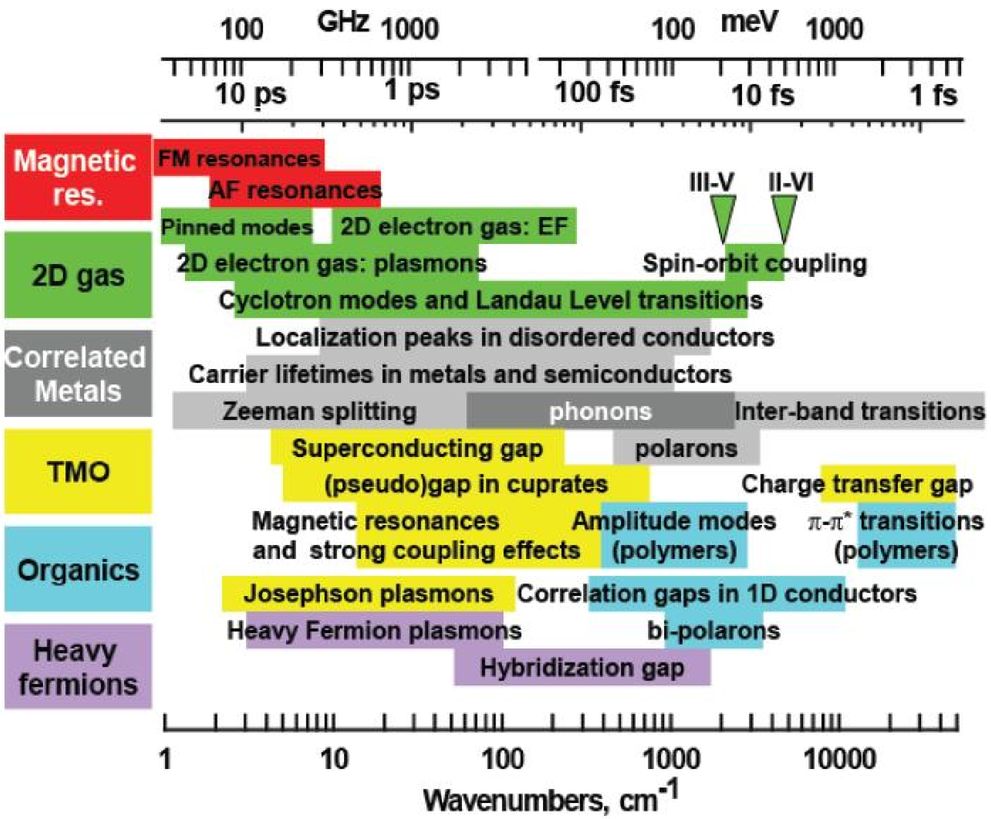
- *Microwave/terahertz/infrared/optical spectroscopies.* These probe the electronic excitations and charge dynamics in both the frequency and time domains. This information is the key to understanding the dynamical interactions of the electrons.

- *Resonant elastic and inelastic x-ray spectroscopy.* Resonant elastic and inelastic x-ray scattering can now reveal spin and charge density waves and superlattices with tiny modulation amplitudes. This information is critically important for understanding spatially periodic electronic states of matter.

- *Neutron Scattering (NS).* High-intensity NS for example, from the Spallation Neutron Source will allow precision measurements of both magnetic ground states and the complete spectrum of magnetic excitations in high-temperature and exotic superconductors.

- *NMR/NQR/μSR.* NMR measures spin dynamics, NQR measures the charge heterogeneity and dynamics, and μSR measures nanoscale variation in local magnetic field strength. These are essentially local spin/charge probes, but without imaging capabilities.

Optical spectroscopy of solids



Absorption mechanisms associated with various excitations and collective modes in solids → *optical experiments*

(i) measure both $R(\omega)$ and $T(\omega)$ for transparent materials

(ii) measure $R(\omega)$ or $T(\omega)$, then use Kramers-Kronig (KK) transformation

(iii) spectroscopic ellipsometry

(iv) THz time-domain spectroscopy…

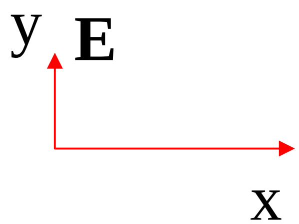
→ *complex optical constants*

D. N. Basov, Richard D. Averitt, Dirk van der Marel, Martin Dressel, Kristjan Haule
Rev. Mod. Phys., 2011

Units: 1 eV = 8065 cm^{-1} = 11400 K
1.24 eV=10000 cm^{-1}

Optical constants

Consider an electromagnetic wave in a medium



$$E_y = E_0 e^{i(qx - wt)} = E_0 e^{iw(x/v - t)} = E_0 e^{iw(\frac{nx}{c} - t)}$$

where $v \equiv w/q = c/n(w)$, $n(w)$: refractive index

If there exists absorption,

$$E_y = E_0 e^{-\frac{wKx}{c}} e^{iw(\frac{nx}{c} - t)}$$

K: attenuation factor

Intensity

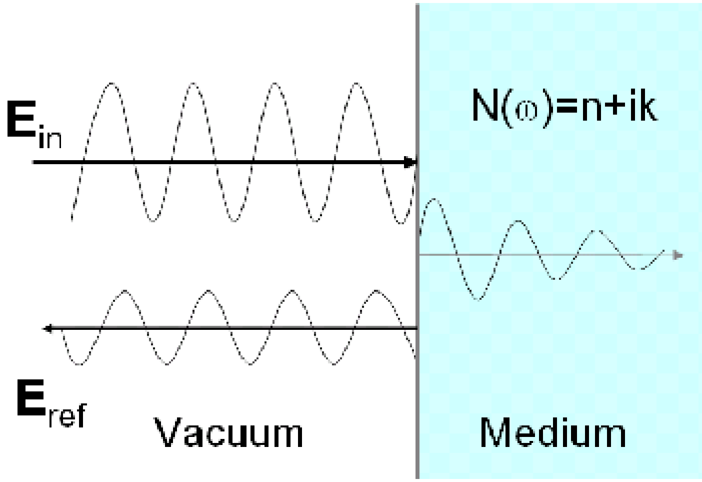
$$I \propto E_y^2 = E_0^2 e^{-\frac{2wKx}{c}}$$

Introducing a complex refractive index:

$$N(w) \equiv n(w) + iK(w)$$

$$E_y = E_0 e^{iw(\frac{N(w)x}{c} - t)}$$

Reflectivity



$$\frac{E_{ref}}{E_{in}} \equiv r = r(w)e^{iq(w)}$$

$$= \frac{n+iK-1}{n+iK+1} = \sqrt{\frac{(n-1)^2+K^2}{(n+1)^2+K^2}}e^{iq(w)}$$

$$R \propto |E_{ref}/E_{in}|^2 = |r(w)|^2 = \frac{(n-1)^2+K^2}{(n+1)^2+K^2}$$

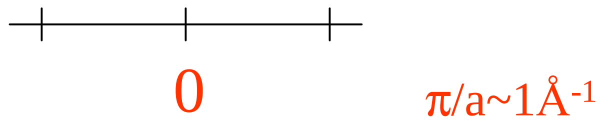
$$tgq = \frac{2K}{n^2+K^2-1}$$

$$n = \frac{1-R}{1+R-2R^{1/2}\cos q}$$

$$k = \frac{-2R^{1/2}\sin q}{1+R-2R^{1/2}\cos q}$$

If n, K are known, we can get R, θ; vice versa.

Dielectric function



$$D(q,w) \equiv e(q,w)E(q,w)$$

$$photon, q \to 0, e = e(w, q \to 0) = e(w)$$

Infrared
$q = 2\pi/\lambda \sim 10^{-4}$ Å^{-1}

$$\mathbf{Q} \sqrt{e(w)} = N(w)$$

$$\Rightarrow e(w) \equiv e_1(w) + ie_2(w) = (n(w) + iK(w))^2$$

$$e_1(w) = n^2(w) - K^2(w)$$
$$e_2(w) = 2n(w) \cdot K(w)$$

or

$$n = \frac{1}{\sqrt{2}} \sqrt{\sqrt{e_1^2(w) + e_2^2(w)} + e_1(w)}$$
$$k = \frac{1}{\sqrt{2}} \sqrt{\sqrt{e_1^2(w) + e_2^2(w)} - e_1(w)}$$

Conductivity

$$s = s_1(w) + s_2(w)$$

By electrodynamics, $e(w) = 1 + \frac{4pis(w)}{w}$

In a solid, considering the contribution from ions or from high energy electronic excitations

$$e(w) = e_\infty + \frac{4pis(w)}{w}$$

Now, we have several pairs of optical constants:

$$\begin{cases} n(\omega),\ K(\omega) \\ R(\omega),\ \theta\ (\omega) \\ \varepsilon_1(\omega),\ \varepsilon_2(\omega) \\ \sigma_1(\omega),\ \sigma_2(\omega) \end{cases}$$

Usually, $R(\omega)$ can be easily measured experimentally.

Kramers-Kronig relation

-- the relation between the real and imaginary parts of a response function.

$$a_1(w) = \frac{2}{p} P \int_0^{+\infty} \frac{w' a_2(w')\mathrm{d}w'}{w'^2 - w^2}$$

$$a_2(w) = \frac{-2w}{p} P \int_0^{\infty} \frac{a_1(w')\mathrm{d}w'}{w'^2 - w^2}$$

For optical reflectance

$$r(w) = \sqrt{R(w)}e^{iq}$$

$$\Rightarrow \ln r(w) = (1/2)\ln R(w) + iq$$

$$q = \frac{w}{p} P \int_0^{\infty} \frac{\ln R(w')}{w^2 - w'^2} dw'$$

Low-ω extrapolations:

Insulator: R~ constant

Metal: Hagen-Rubens

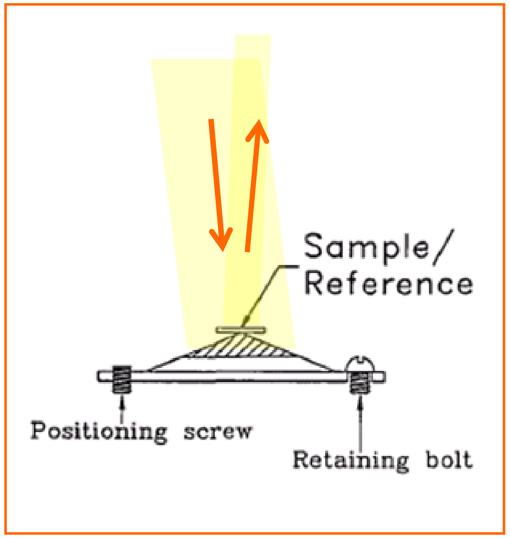
Superconductor: two-fluids model

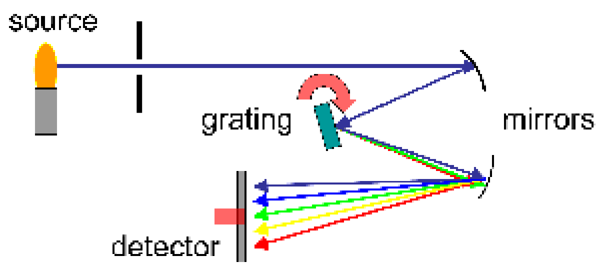
High-ω extrapolations:

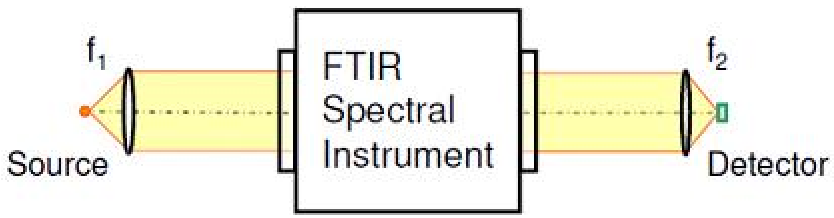
R~ ω^{-p} (p~0.5-1, for intermediate region)

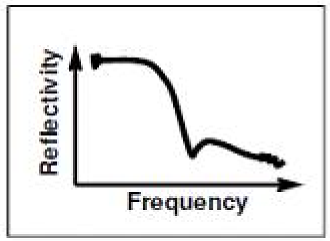
R~ ω^{-n} (n=4, above interband transition)

Reflectivity measurement









From R(ω) to $\sigma_1(\omega)$

$$R(\omega) \overset{KK}{\Rightarrow} \theta(\omega) \rightarrow \begin{cases} n(\omega), \kappa(\omega) \\ \varepsilon_1(\omega), \varepsilon_2(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{cases}$$

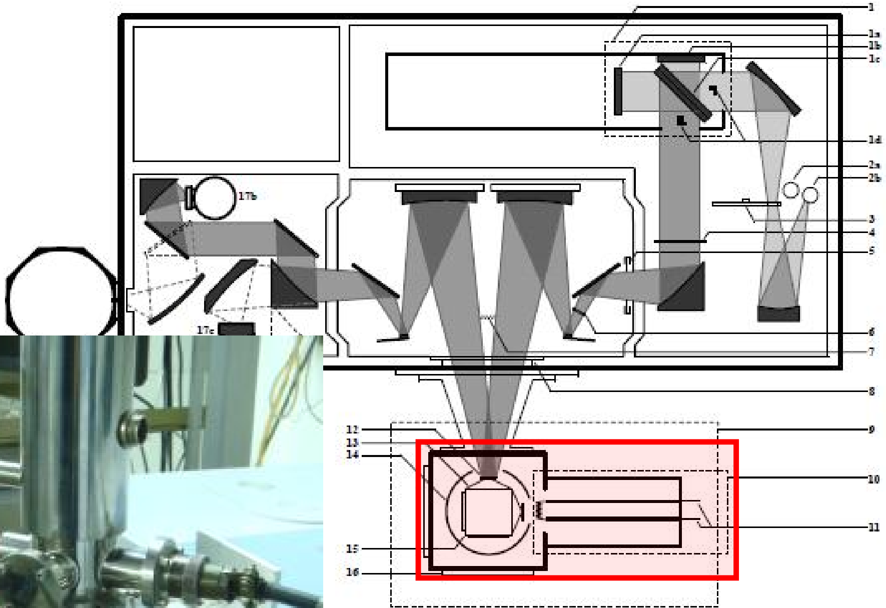
Si-beamsplitter for 113v

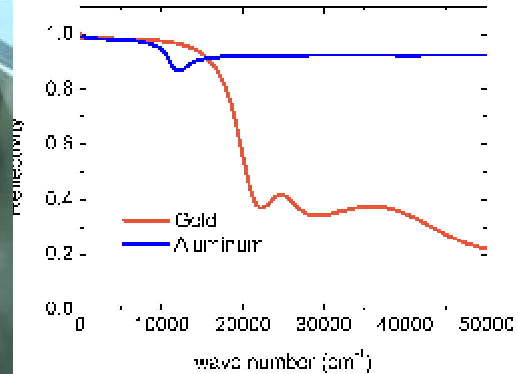
Bruker 113, 66v, 80v, and grating spectrometers

Energy range: 17 -50000 cm^{-1} (2 meV~6 eV)

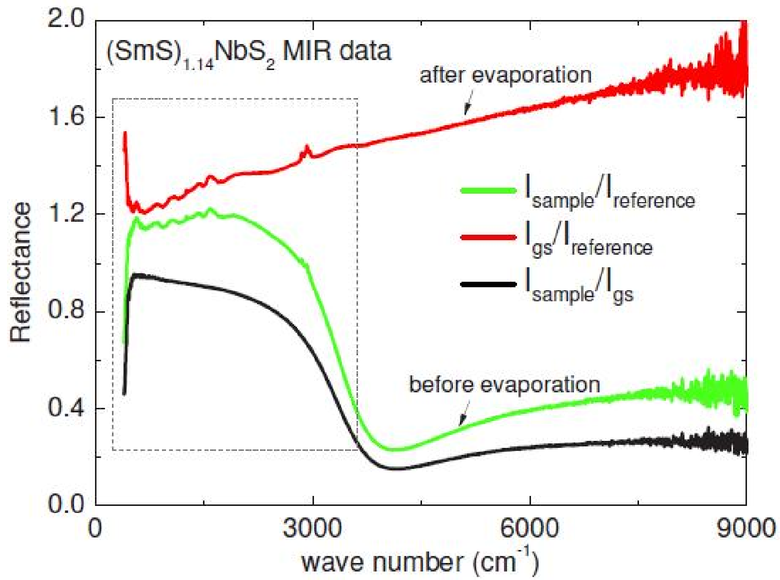
In-situ overcoating technique

FT-IR spectrometer



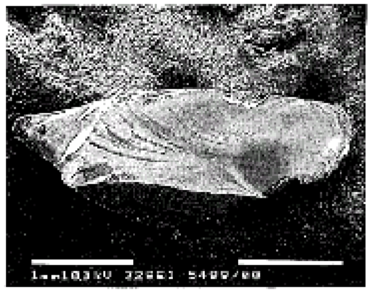


In situ evaporation



$$\left(\frac{R_s}{R_r}\right)\left(\frac{R_{gs}}{R_r}\right)^{-1} \equiv \frac{R_s}{R_{gs}}$$

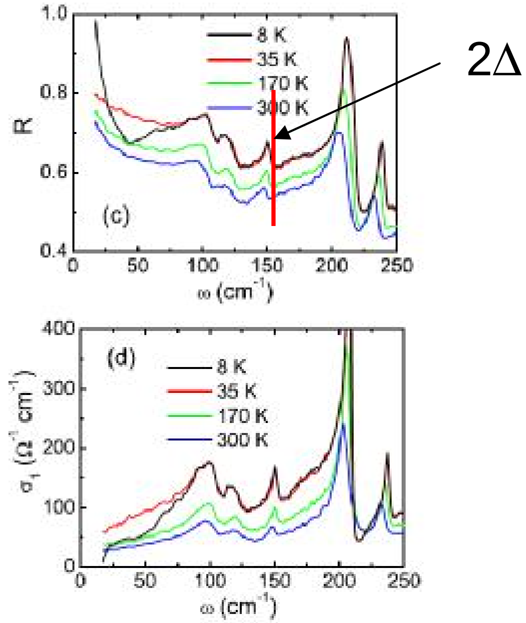
C. C. Homes *et al.*
Applied Optics 32,2976(1993)

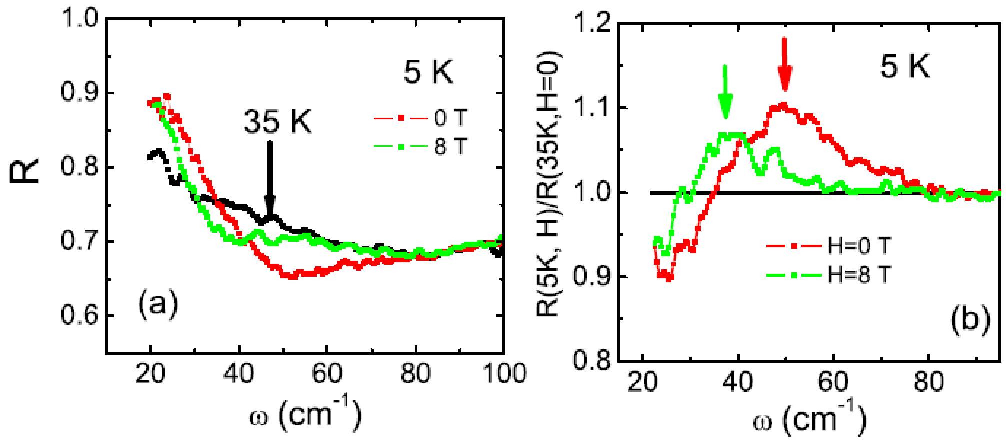


Optical measurement under magnetic field

(10 Tesla split coils from Cryomagnetic Inc.)

KxFe2-ySe2 Tc~30 K

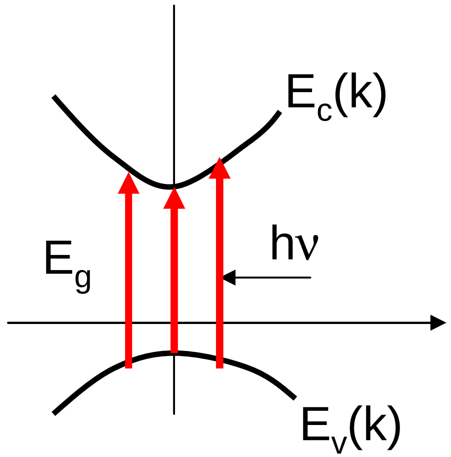




R. H. Yuan *et al.*, *Scientific Reports 2, 221 (2012)*

Interband transition

$$|v,k>, E_v(k)$$
$$|c,k>, E_c(k)$$



$$\hbar\omega = E_c(k) - E_v(k) \equiv E_{cv}(k)$$

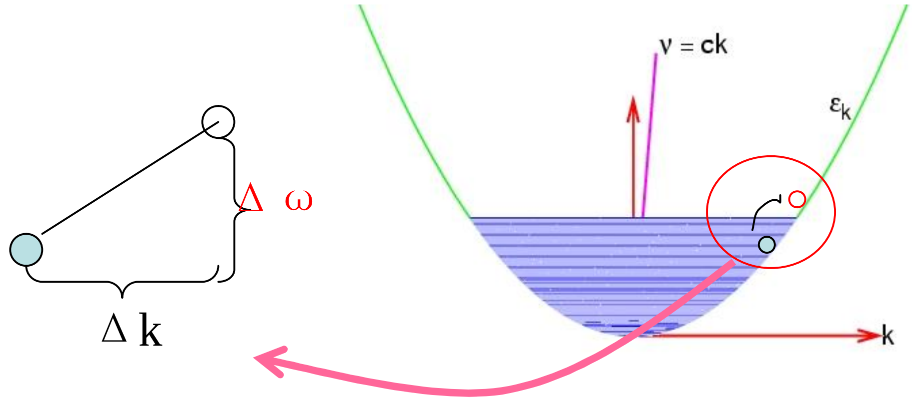
$$J(E) = \frac{V}{(2\pi)^3}\int \frac{ds}{|\nabla_k E_{cv}(k)|}$$

Kubo-Greenwood formula

$$\varepsilon_2(\omega) = \frac{8\pi^2 e^2}{m^2\omega^2} J(\hbar\omega)\,|\vec{p}_{vc}(\hbar\omega)|^2$$

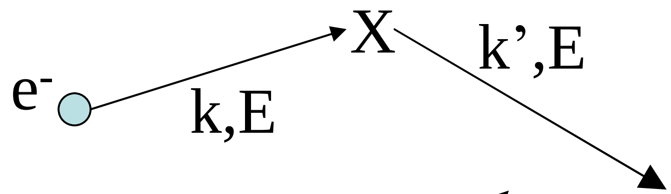
$$\sigma_1(\omega) = \frac{1}{4\pi}\omega\varepsilon_2(\omega)$$

Intraband transition

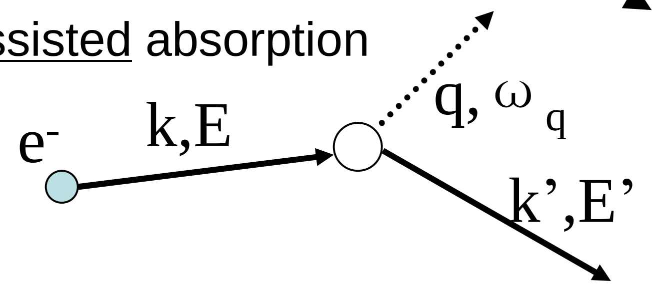


Infrared light cannot be absorbed directly by electron-hole excitation.

(a) Impurity-assisted absorption



(b) boson-assisted absorption



Holstein process, if phonons are involved.

Drude model

$$s(w)=\frac{s_0}{1-iwt}=\frac{w_D^2}{4p}\frac{1}{1/t-iw}$$

$$e(w)=e_\infty+\frac{4pi}{w}s(w)$$

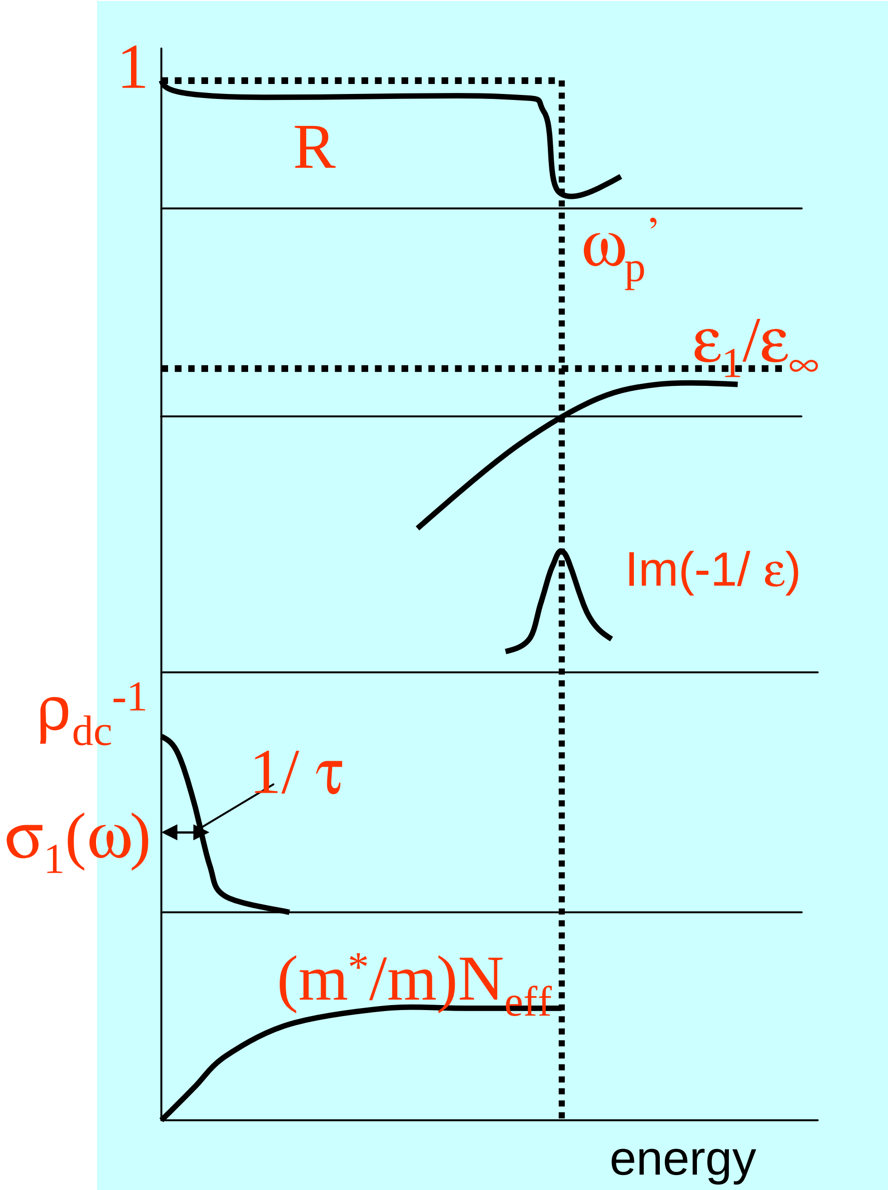
$$\Rightarrow e_1=e_\infty-\frac{w_p^2}{w^2+1/t^2}$$

$$e_2=\frac{4ps_1}{w}=\frac{w_p^2 t}{w}\frac{1}{1+w^2t^2}$$

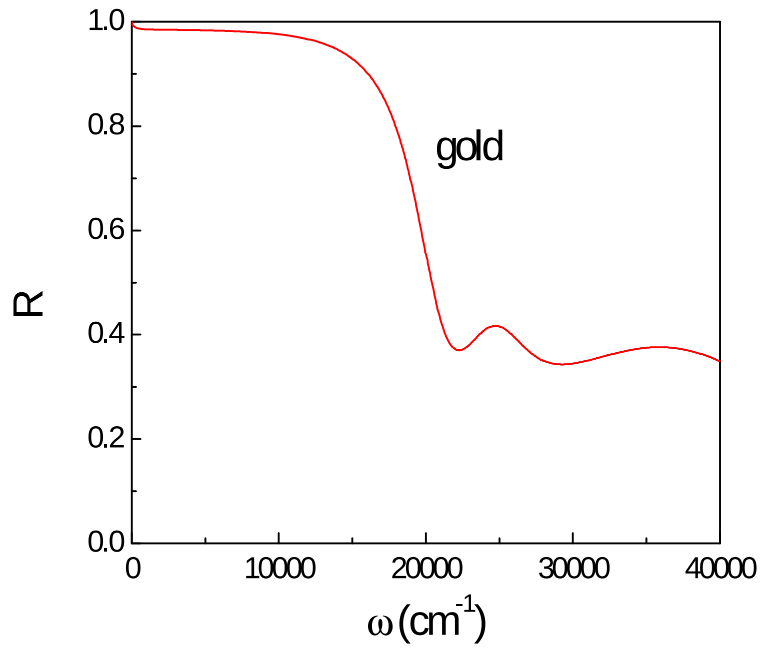
$$\mathrm{Im}\{\frac{-1}{e(w)}\}=\frac{w_p'^2 w/t}{(w^2-w_p'^2)^2+w^2t^{-2}}$$

$$w_p'=w_p/\sqrt{e_\infty}$$

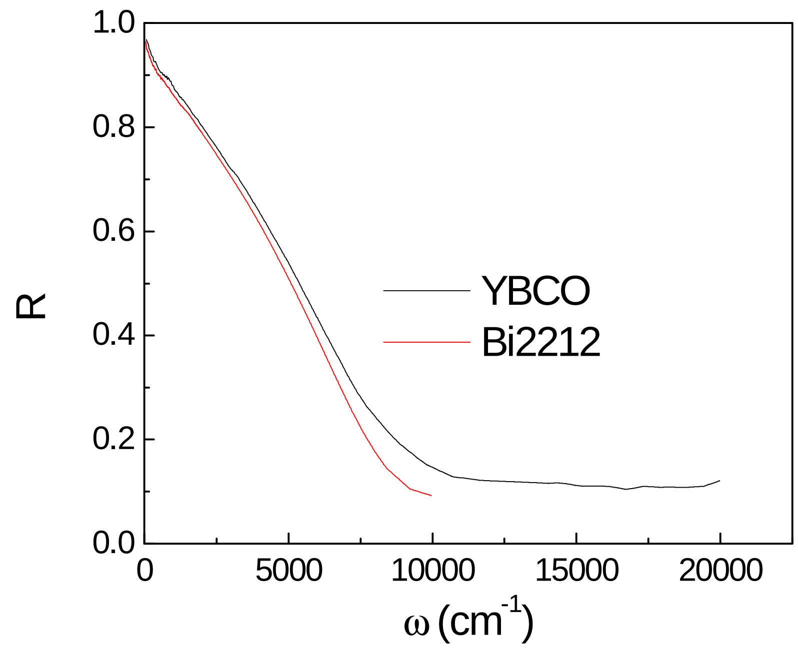
$$\int_0^\infty s_1(w)dw=\frac{w_p^2}{8}$$



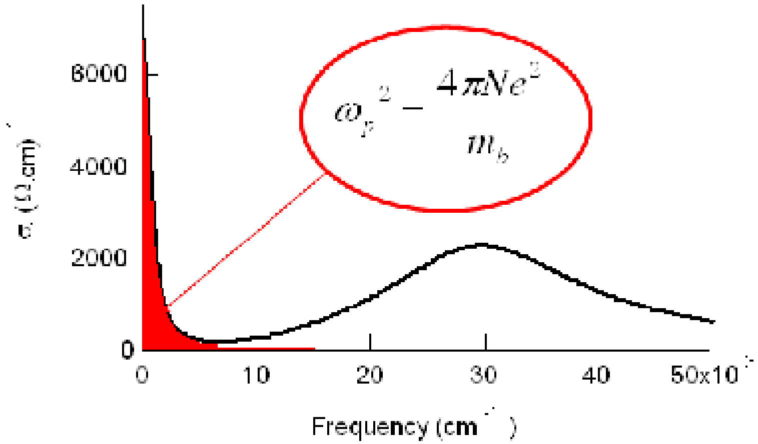
Simple metal



High-T_c cuprates



Simple metal

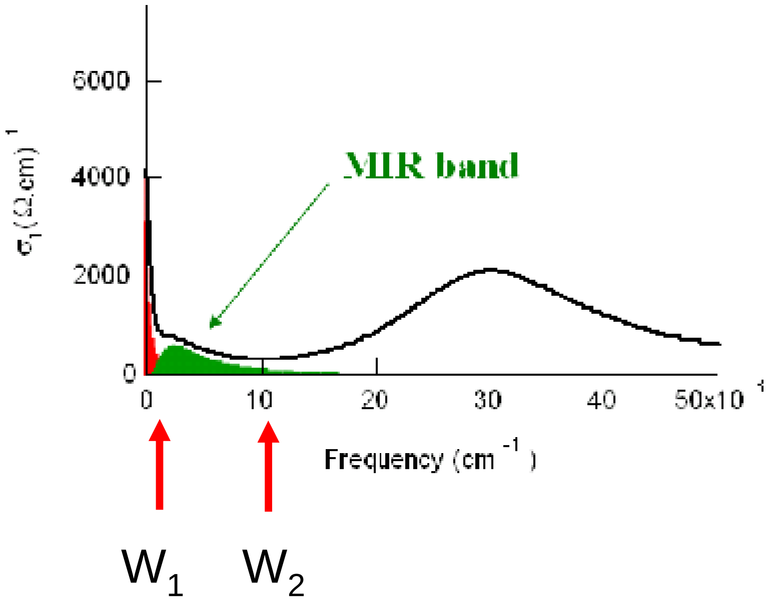


Correlated metal

$$\omega_p^{Drude\,2} = \frac{4\pi Ne^2}{m^*}$$

$$\omega_p^{Tot\,2} = \omega_p^{Drude\,2} + \omega_p^{MIR\,2} = \frac{4\pi Ne^2}{m_b}$$

$$m^*/m_b = \frac{\omega_p^{Tot\,2}}{\omega_p^{Drude\,2}}$$



or

$$\frac{m^*}{m_B} = \frac{\int_0^{W2} s_1(w, T > T_{co})\,dw}{\int_0^{W1} s_1(w, T << T_{co})\,dw}$$

for heavy fermions

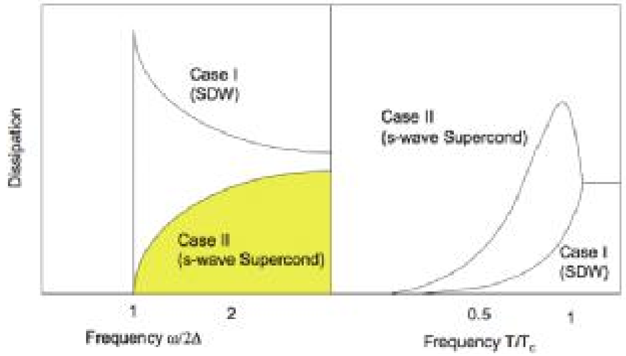
Symmetry broken state

Superconductor vs density wave state

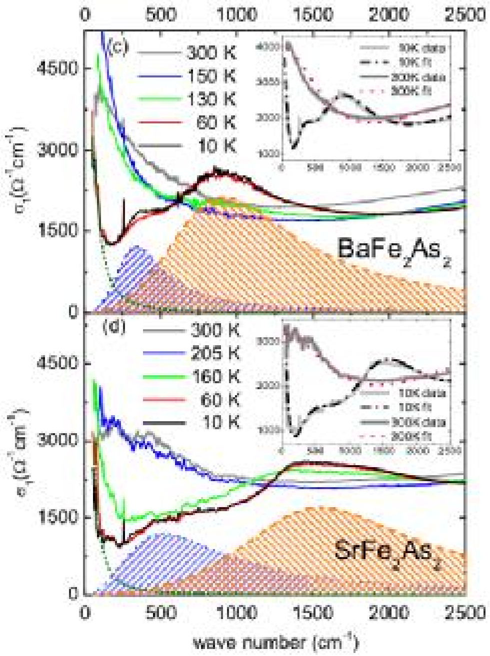
$$\frac{\sigma_1(\omega,T)}{\sigma_n} = \frac{2}{\hbar\omega}\int_{\Delta}^{\infty}\frac{[f(E)-f(E+\hbar\omega)](E^2\pm\Delta^2+\hbar\omega E)}{(E^2-\Delta^2)^{1/2}[(E+\hbar\omega)^2-\Delta^2]^{1/2}}dE$$
$$-\frac{1}{\hbar\omega}\int_{\Delta-\hbar\omega}^{-\Delta}\frac{[1-2f(E+\hbar\omega)](E^2\pm\Delta^2+\hbar\omega E)}{(E^2-\Delta^2)^{1/2}[(E+\hbar\omega)^2-\Delta^2]^{1/2}}dE,$$

- for case I

+ for case II



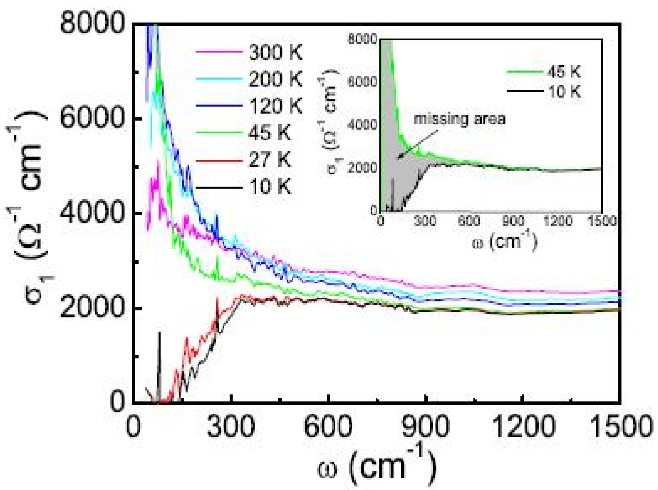
AFe_2As_2 (A=Ba, Sr), SDW gap



W.Z.Hu et al.

PRL (08)

$Ba_{0.6}K_{0.4}Fe_2As_2$, SC gap



G. Li et al. PRL (08)

URu_2Si_2

REVIEWS OF MODERN PHYSICS, VOLUME 83, OCTOBER–DECEMBER 2011

Colloquium: Hidden order, superconductivity, and magnetism: The unsolved case of URu_2Si_2

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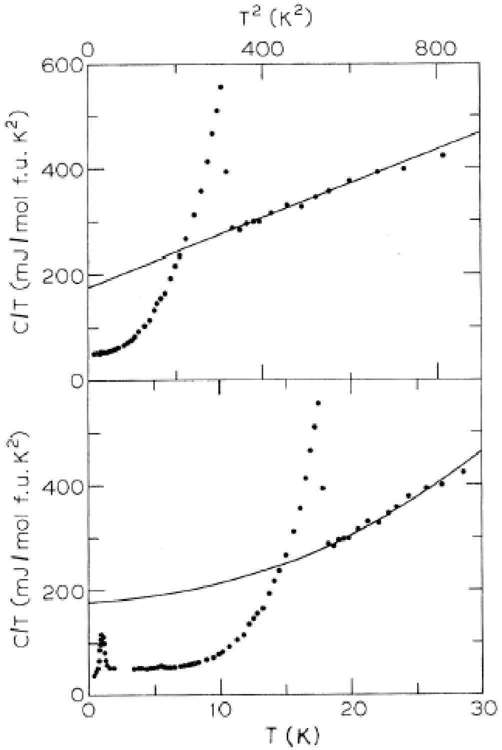
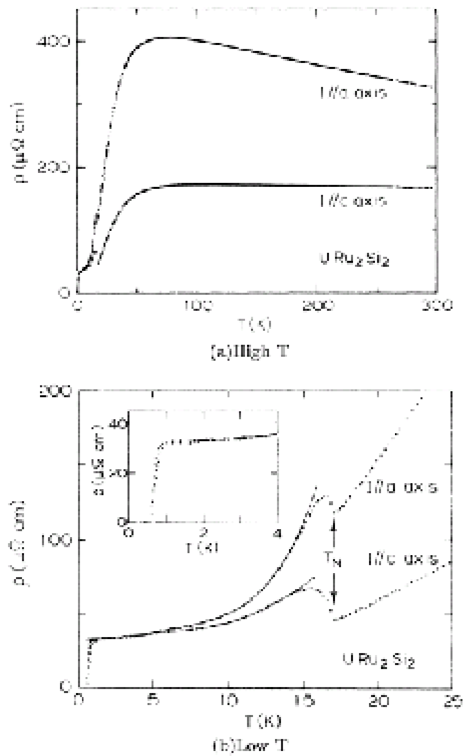
(published 16 November 2011)

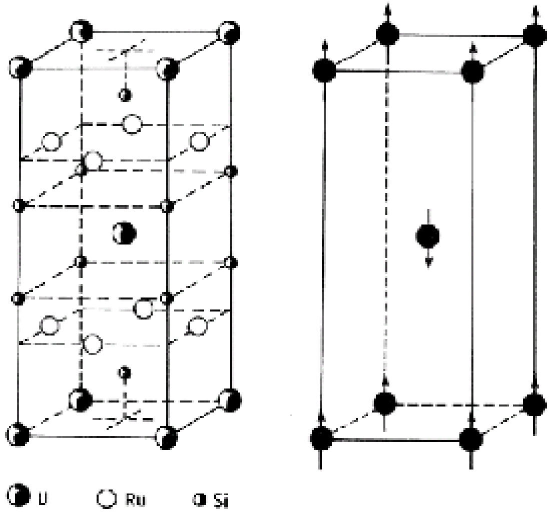


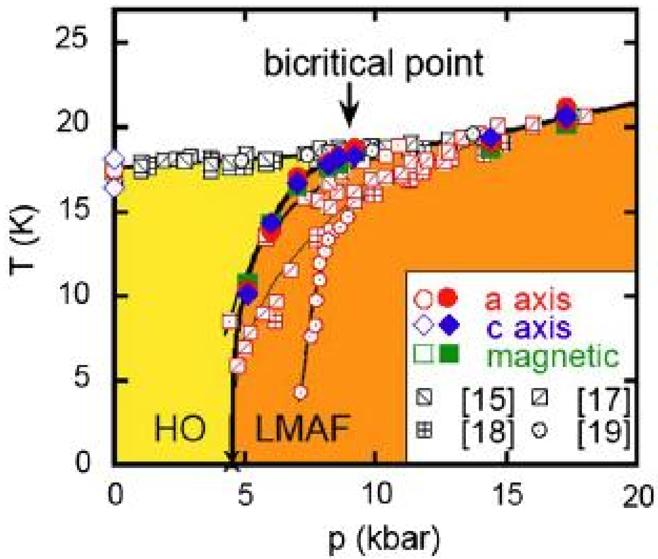


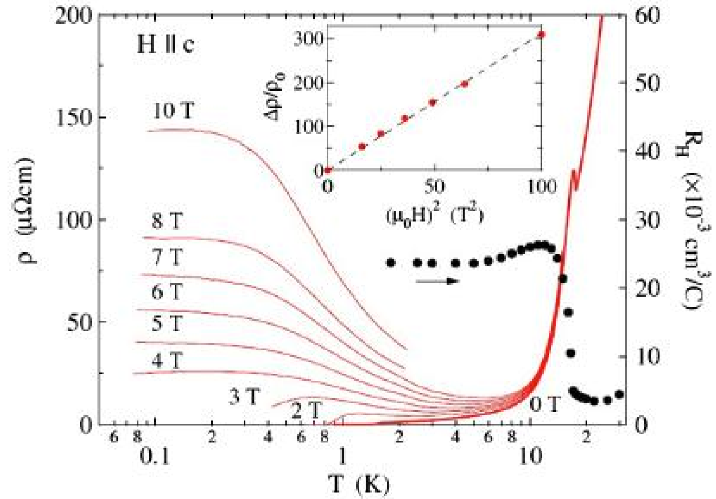
Palstra et al.

- ØItinerant AFM
- ØCDW/SDW
- ØU-moment AFM

C. Broholm et al





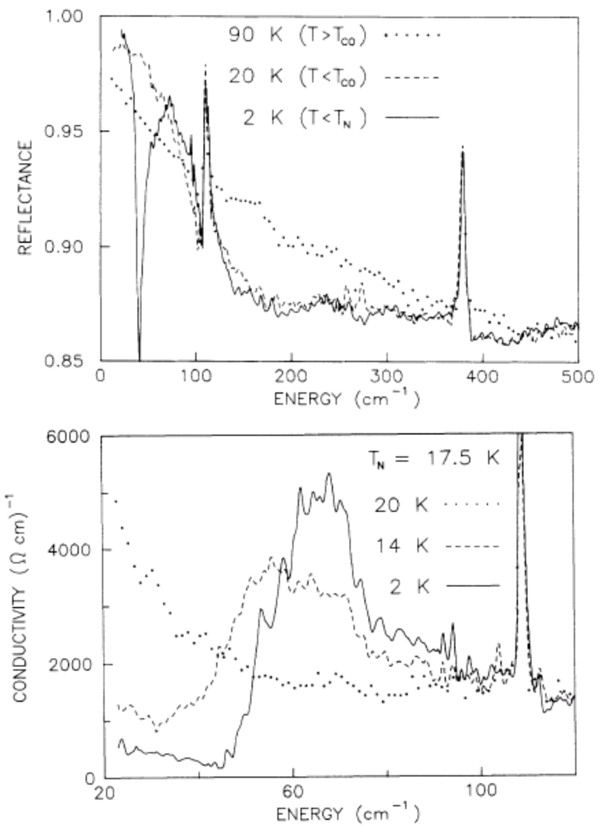


Kasahara et al.

Semimetal

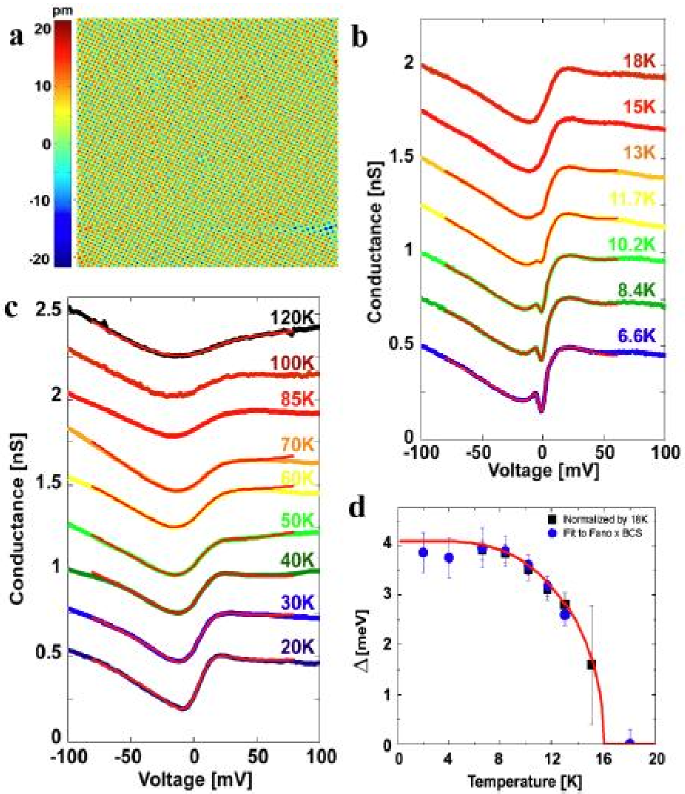
A number of experiments indicate gap opening below T_H

Optical spectroscopy



Bonn et al. PRL 1988

STM

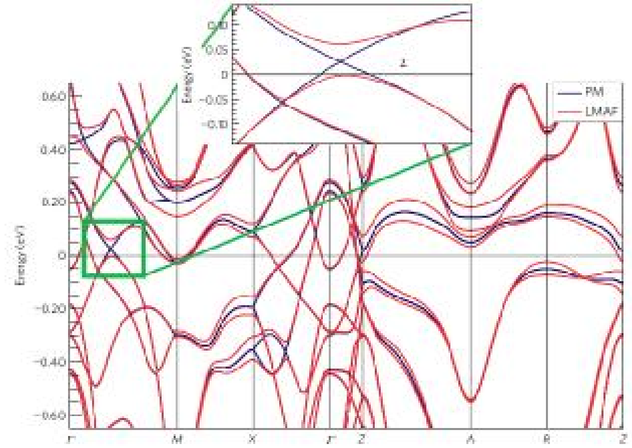


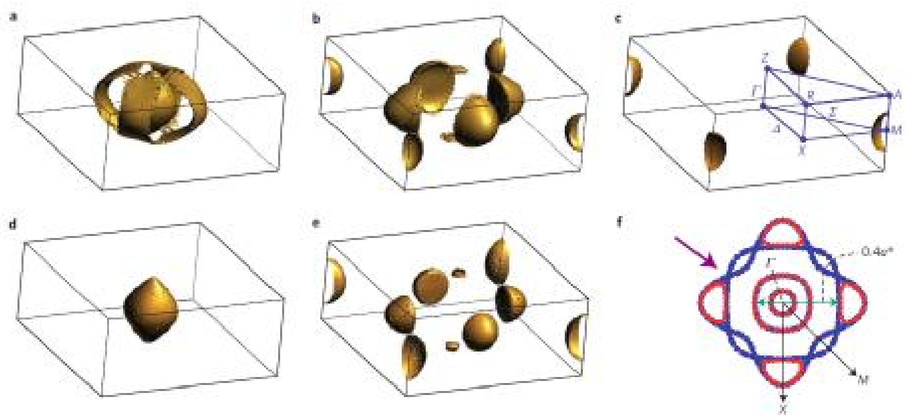
Aynajian et al, PNAS 2010

Hidden order in URu_2Si_2 originates from Fermi surface gapping induced by dynamic symmetry breaking

nature materials
2009

S. Elgazzar[1]*, J. Rusz[1], M. Amft[1], P. M. Oppeneer[1†] and J. A. Mydosh[2]

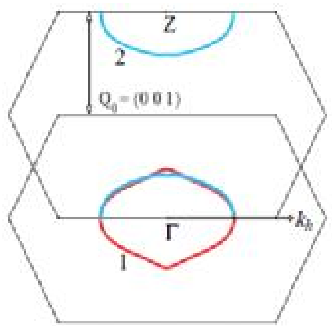


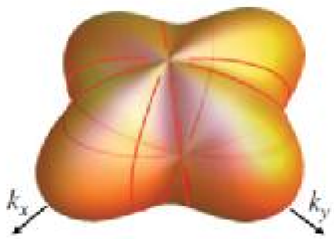


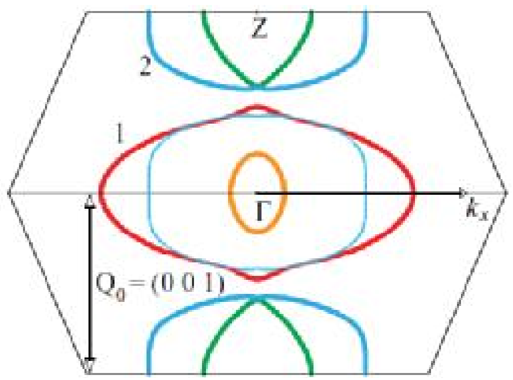
PHYSICAL REVIEW B **84**, 241102(R) (2011)

Spin and orbital hybridization at specifically nested Fermi surfaces in URu_2Si_2

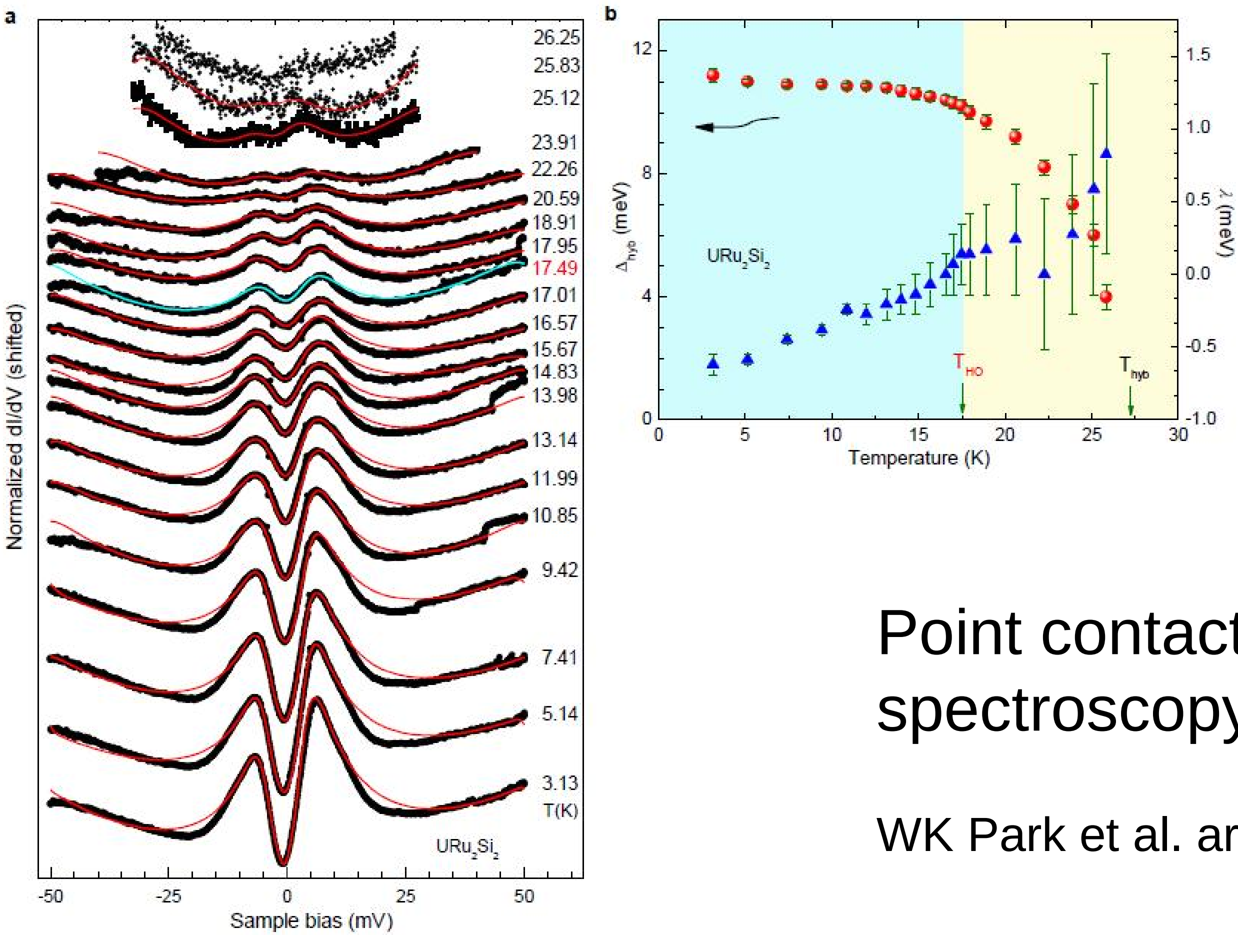
P. M. Oppeneer,[1] S. Elgazzar,[2] J. Rusz,[1] Q. Feng,[1] T. Durakiewicz,[3] and J. A. Mydosh[4]







However, some other experiments indicate gap formation up to a higher T~25 – 30 K



Point contact tunneling spectroscopy

WK Park et al. arXiv:1110.5541

PHYSICAL REVIEW B **84**, 161101(R) (2011)

Evidence of a hidden-order pseudogap state in URu_2Si_2 using ultrafast optical spectroscopy

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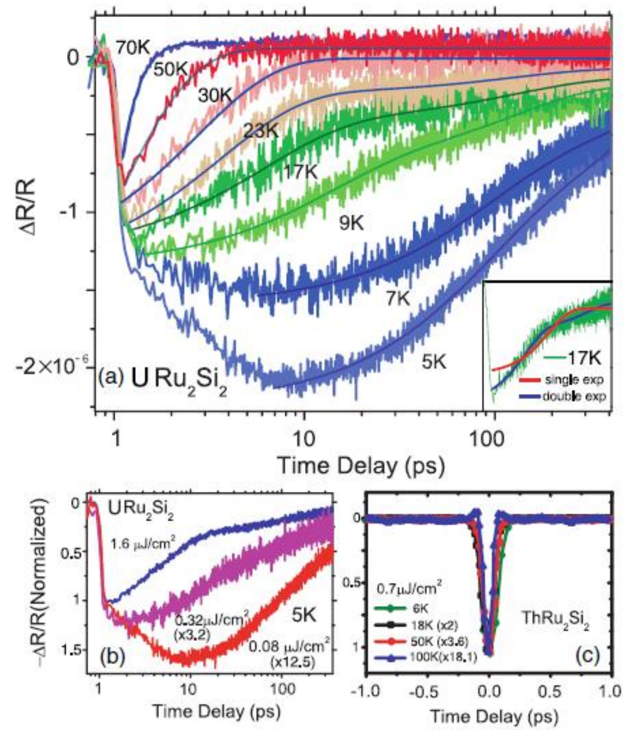

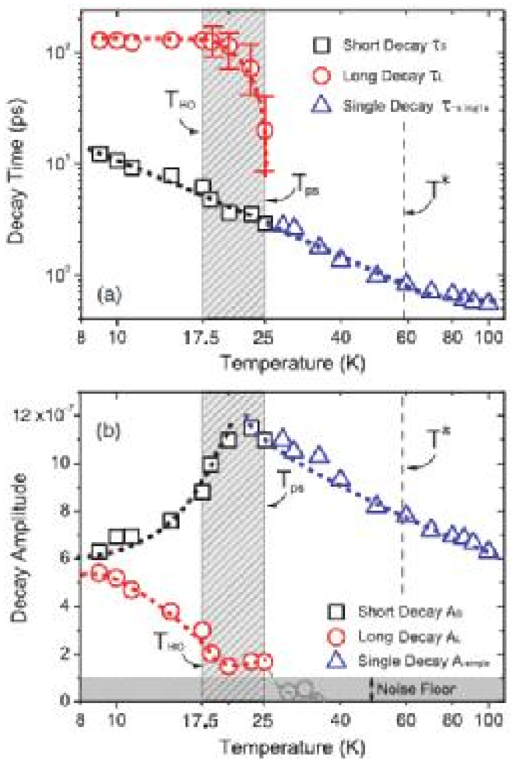

A gap would impedes the relaxiation of excited quasiparticles.

$\Delta R(t)/R = A_0 + A_S \exp(-t/\tau_S) + A_L \exp(-t/\tau_L)$, where τ_S and τ_L are the short and long decay times, respectively.

PHYSICAL REVIEW B **84**, 214410 (2011)

Hidden-order pseudogap in URu_2Si_2

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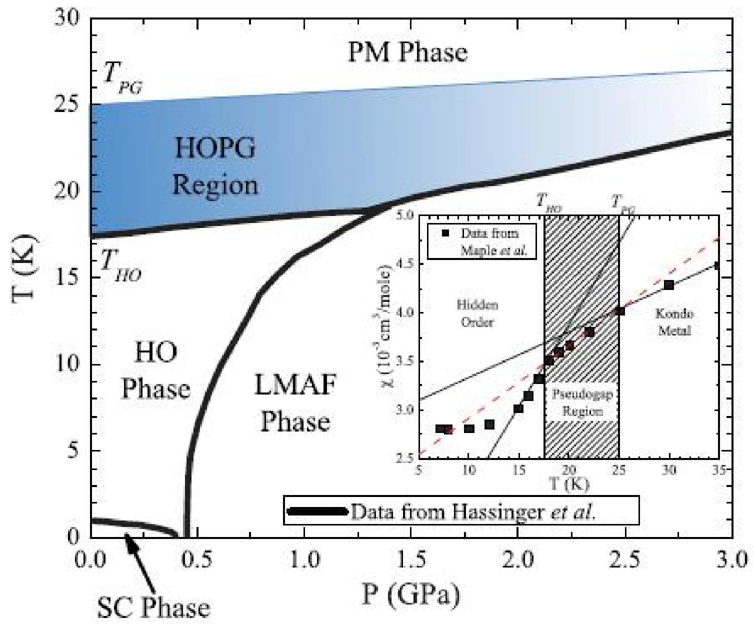
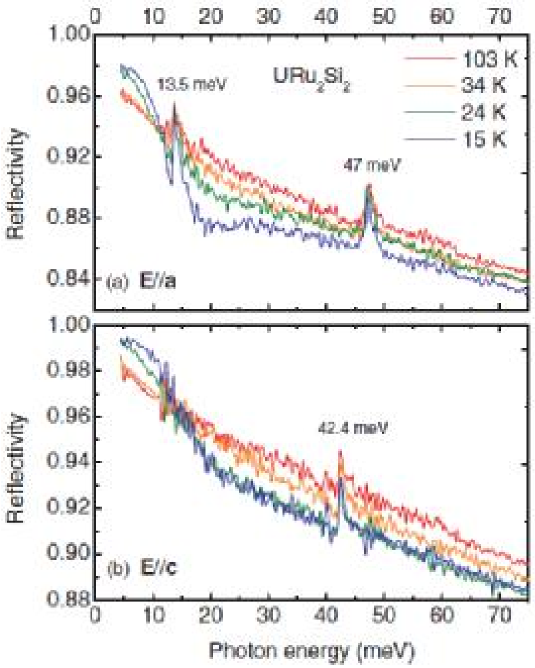
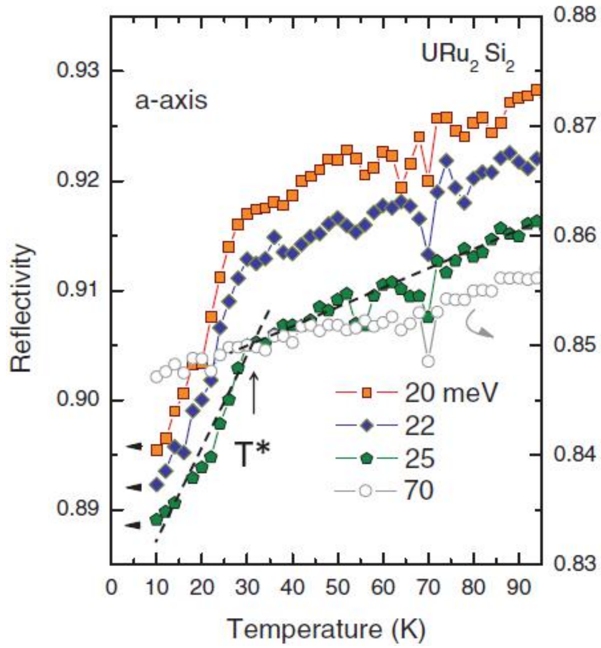
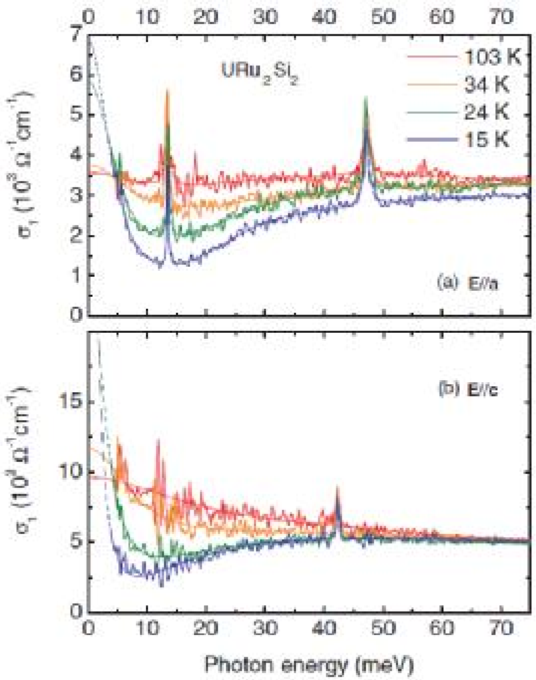
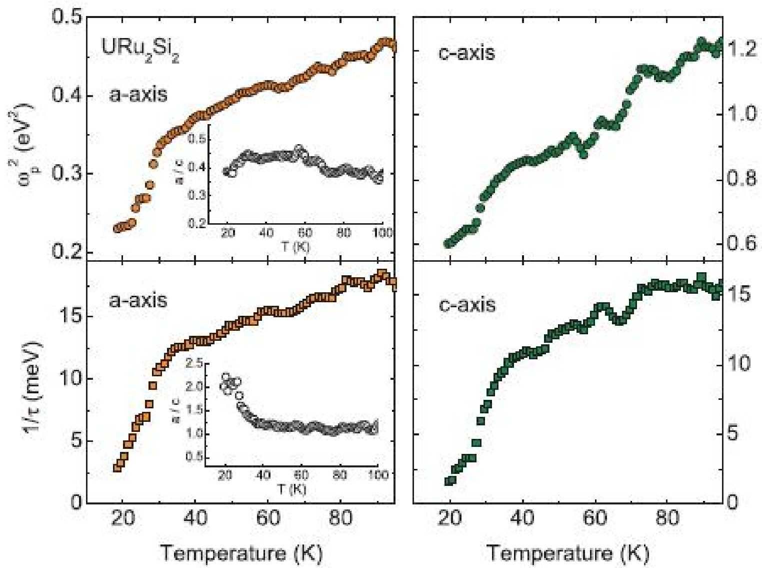
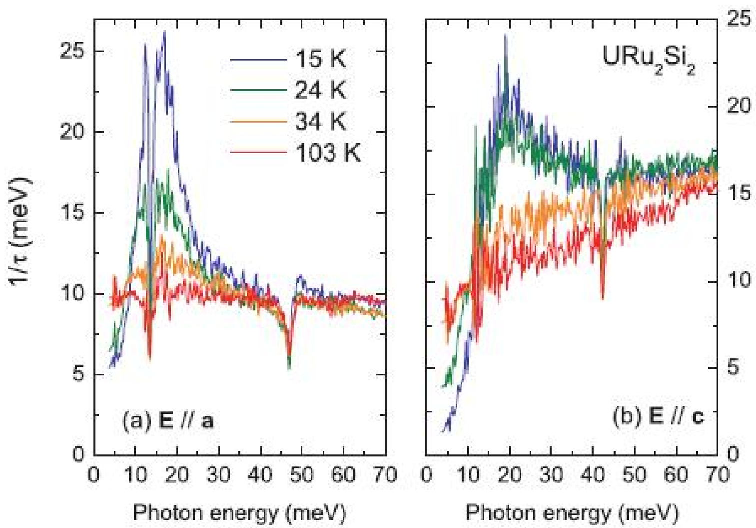






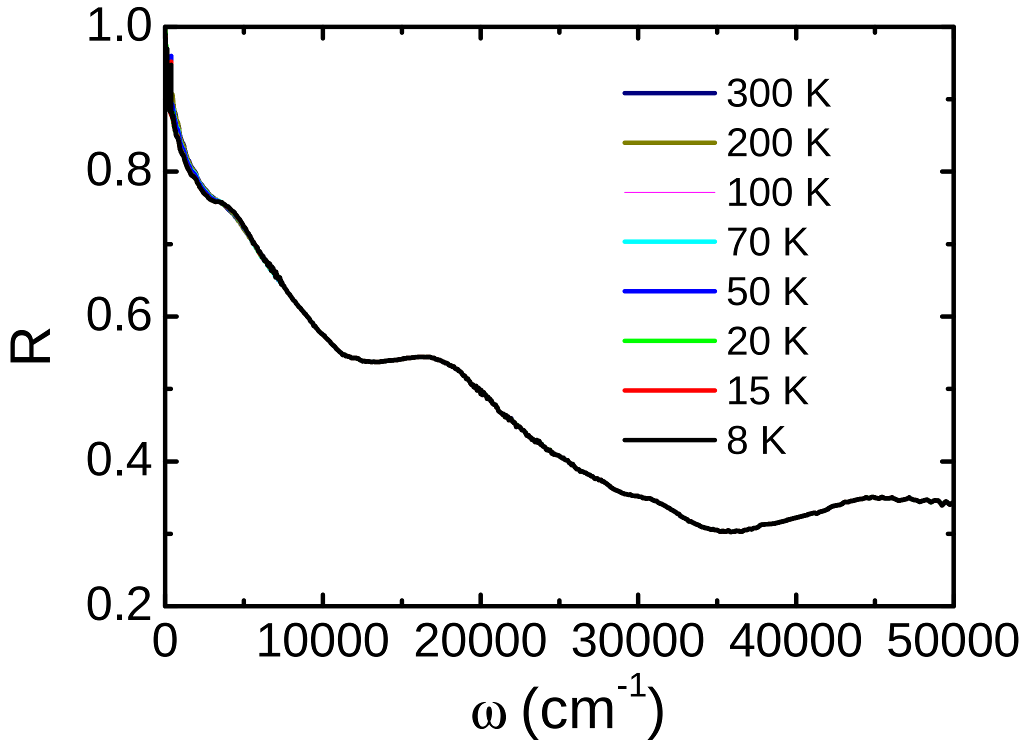


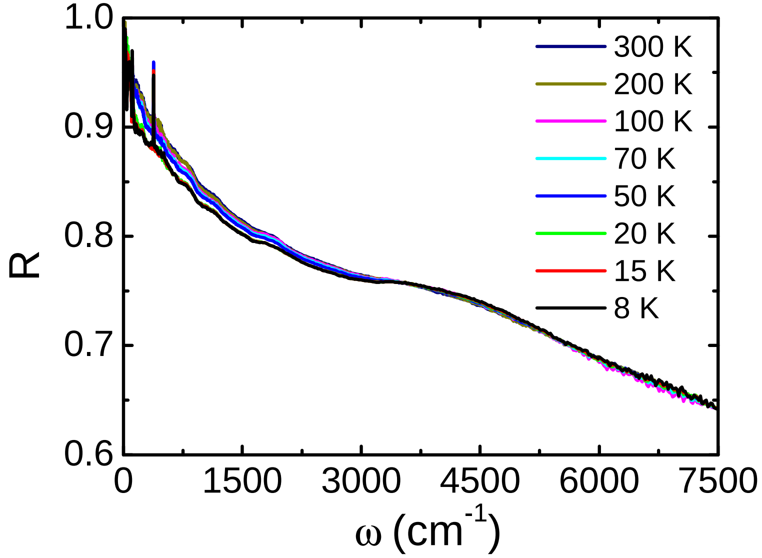


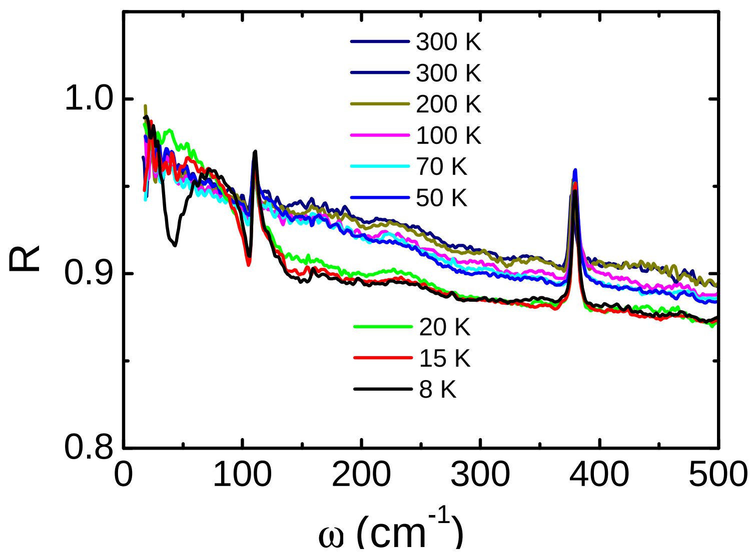


J. Levallois et al PRB 2011

Reflectance

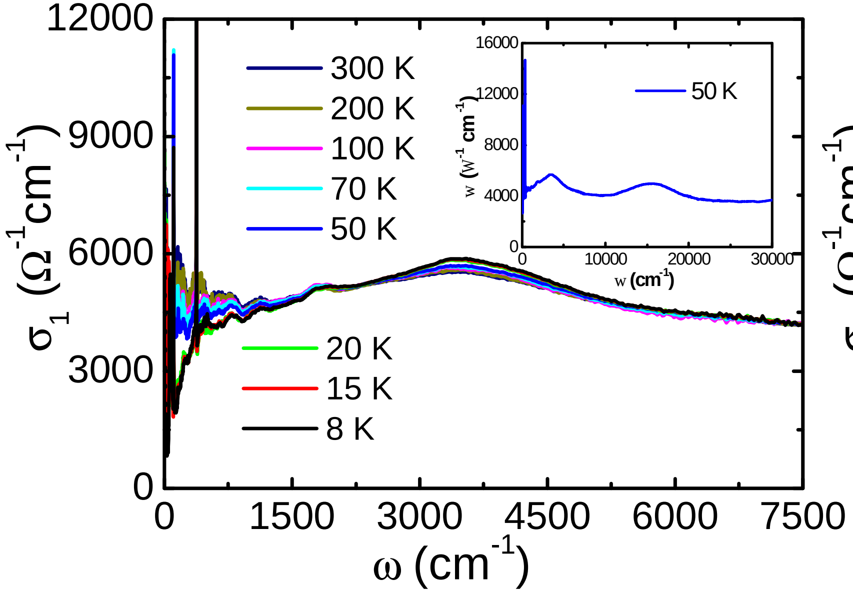


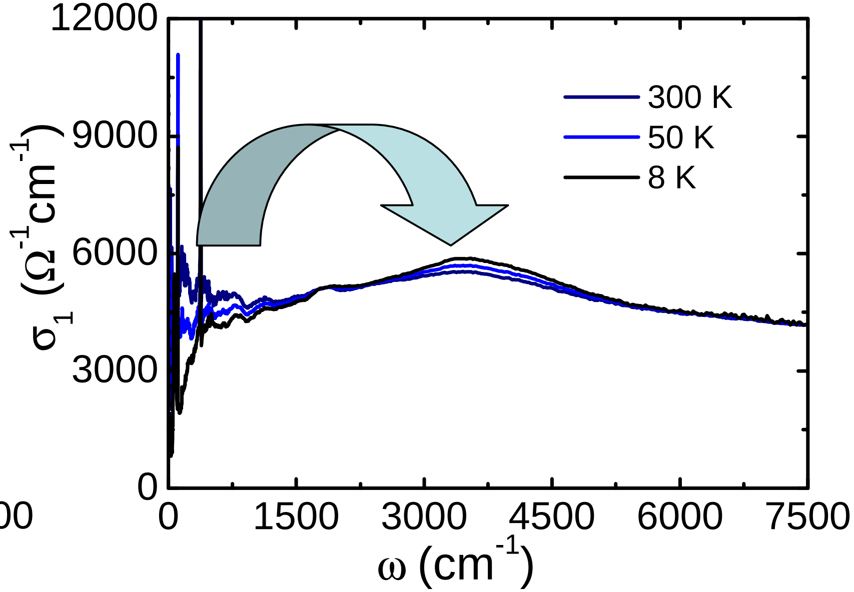




Samples from G. Luke

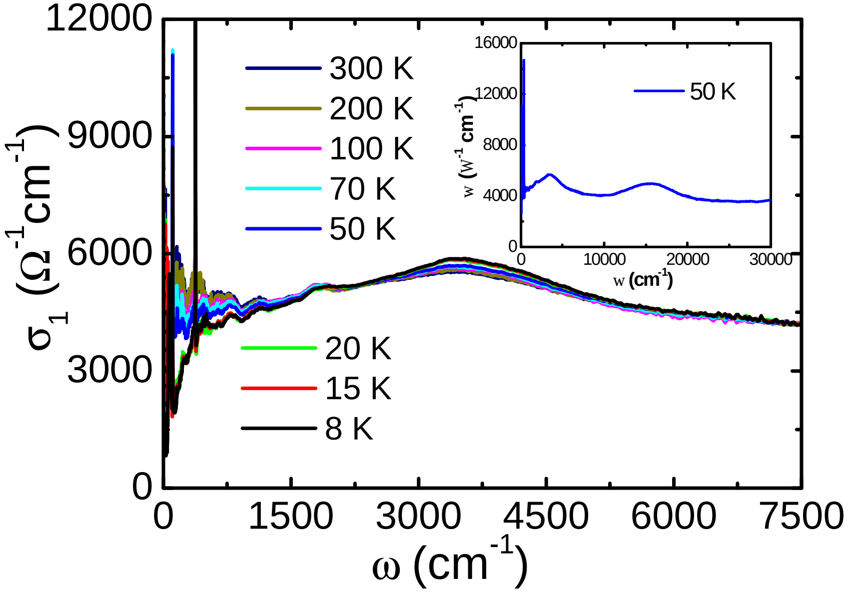
Optical conductivity

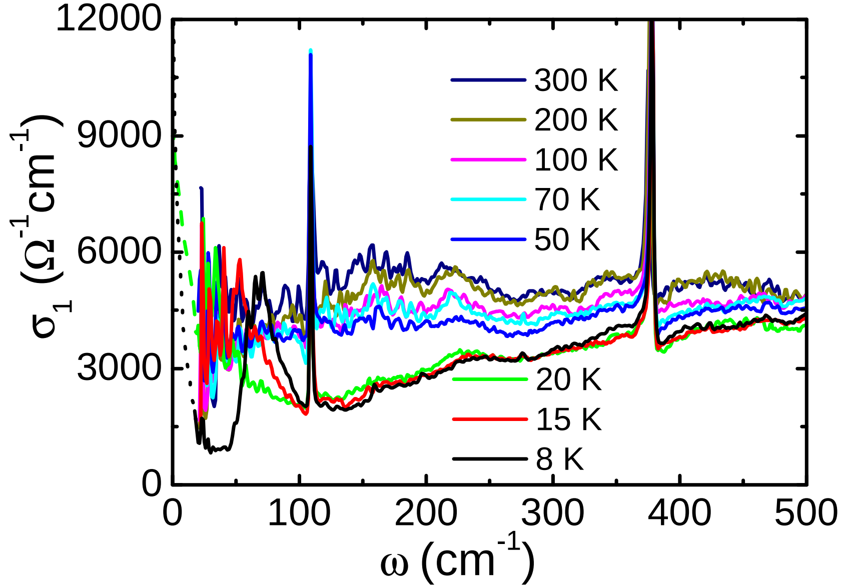




ØT>50 K, $\sigma_1(\omega)$ shows a non-metallic behavior. The Drude component is completely absent. Actually, $\sigma_1(\omega)$ shows a decreasing tendency with decreasing ω. Clearly, there are no well defined quasiparticles above 50 K.

ØWith T from 300 K to 50 K, the spectral weight is transferred from the low-ω regime (below 2000 cm-1) to higher ω region (centered at about 4000 cm-1).

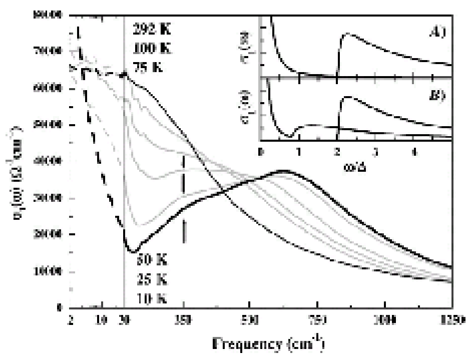




Ø T<< T_{co}, roughly below 20 K, there appears an abrupt spectral weight suppression below 400 cm-1. ⇨ formation of hybridization gap.

Ø A small part of the suppressed spectral weight was transferred to the low-ω side, leading to a narrow Drude component, while the majority of the suppressed spectral weight was still transferred to the high ω side centered near 4000 cm-1.

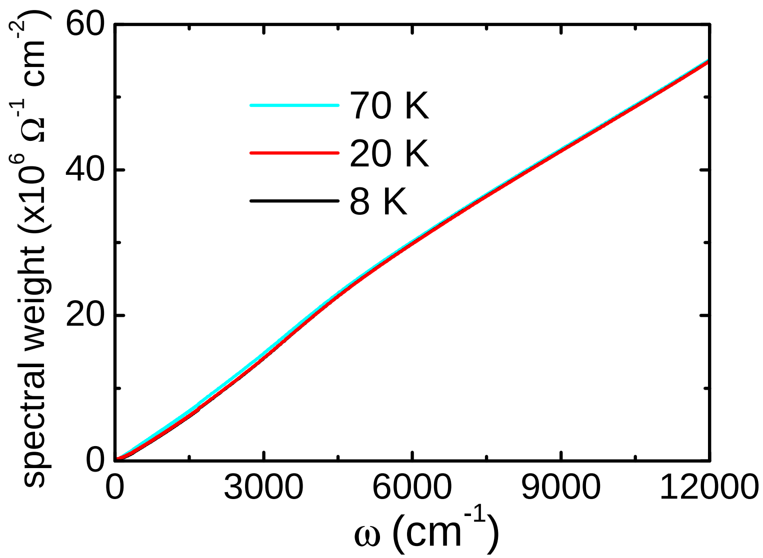
Thes energy scale of the spectral weight transfer is high compared to some 4f electron-based heavy fermion systems, e.g. CeCoIn5.

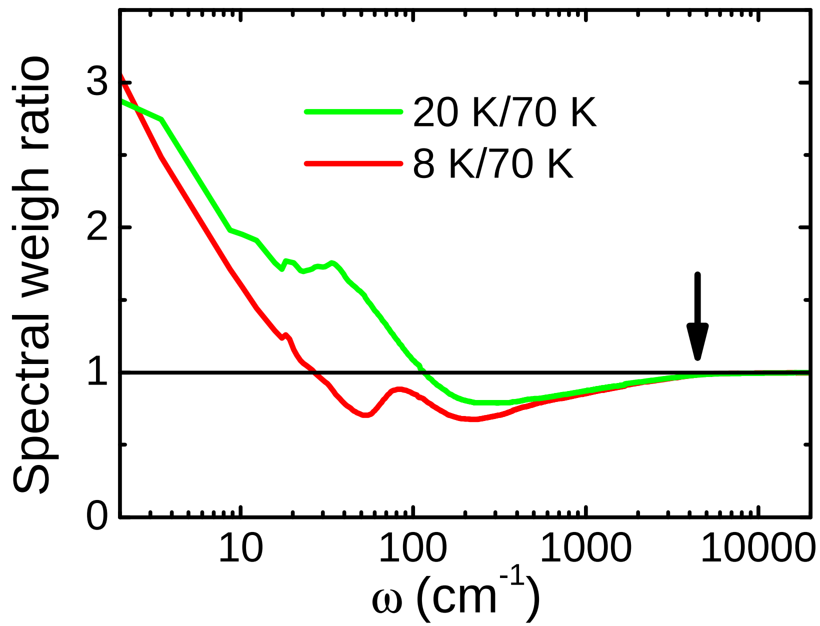


CeCoIn5

Singley, et al. PRB 2002

Spectral weight transfer





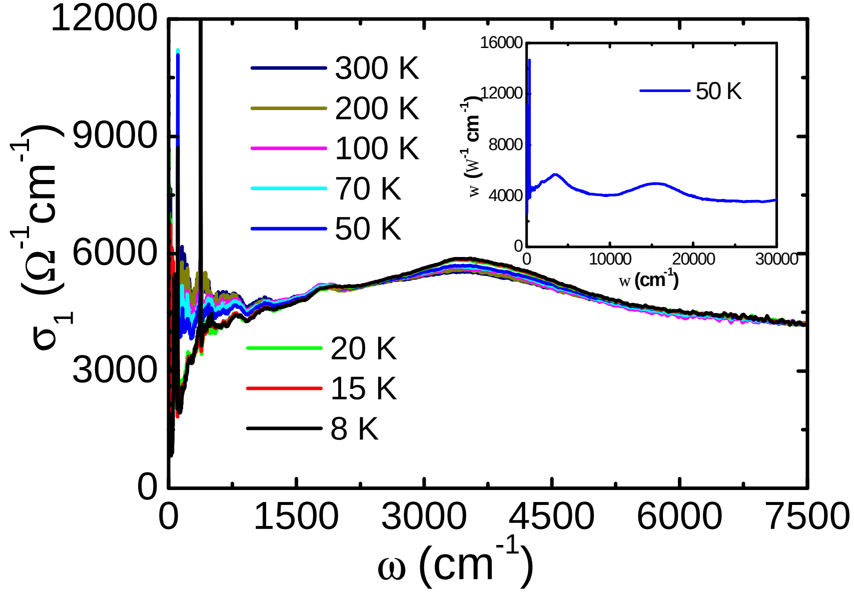
$$w_p^2 \propto \int_0^W s_1(w)dw$$

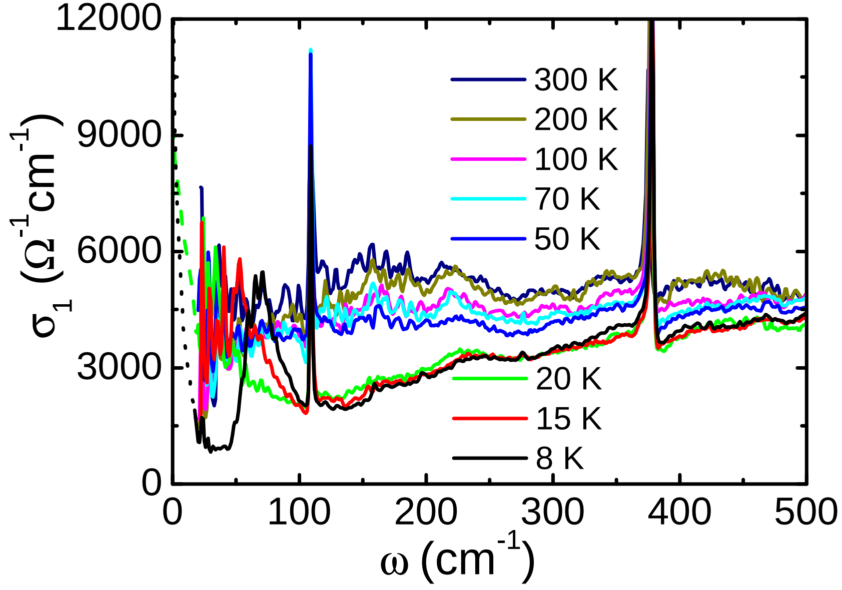
$$\begin{cases} w_p = 1.95\times10^4 cm^{-1} \\ w_p = 4.28\times10^3 cm^{-1} \\ w_p = 2.094\times10^3 cm^{-1} \end{cases}$$

W=2000 cm^{-1} for T=50 K and 300 K;

W=135 cm^{-1} for T=20 K

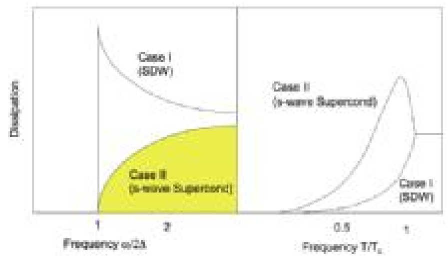
W=42 cm^{-1} for T=8 K





- **Ø**The formation of hybridization gap is associated with renormalization of heavy quasiparticle. The mass enhancement is roughly $m^*/m_B \sim 21$.

- **Ø**Below the T_H, we observe clearly the opening of a density-wave type energy gap: a large fraction of the coherent Drude spectral weight was removed and piled up just above the energy gap of 60 cm-1 ($2\Delta/k_BT_c \sim 5.7$). $n_{8K}/n_{20K} \sim 0.24$.



"Text book"-like density wave type energy gap below T_H.

Conclusions:

- Ø different from the point contact tunneling spectroscopy measurement, the Hybridization gap (~15 meV) is completely different from the density-wave type gap (2Δ~8 emV) in the hidden order state;

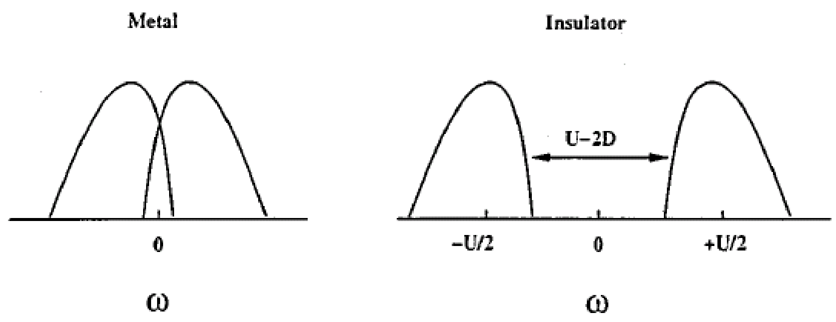
- Ø No other pseudogap is detected;

- Ø The formation of the hybridization gap is a crossover phenomenon, being associated with the formation of a narrow Drude component (or well-defined heavy quasiparticles);

- Ø The opening of the density wave gap below T_H results in the removal of a large fraction of Fermi surfaces (about 75%) and a rapid reduction of the scatterings.

Thank you!

Hubbard U physics: $\rho(\omega)$



$$\rho(\omega) \equiv -\frac{1}{\pi}\sum_{\mathbf{k}} \mathrm{Im} G(\mathbf{k}, \omega + i0^+)$$

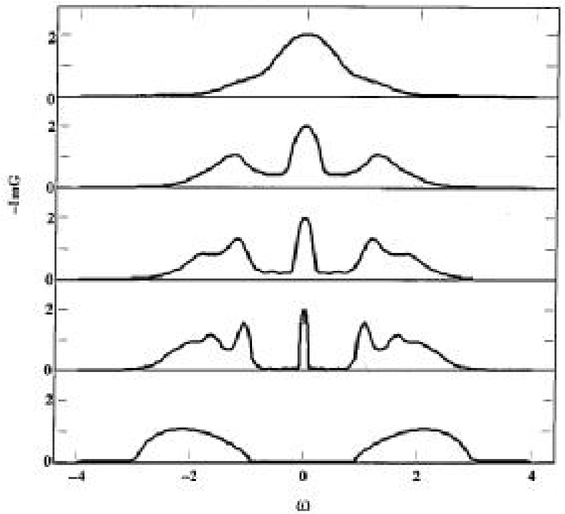
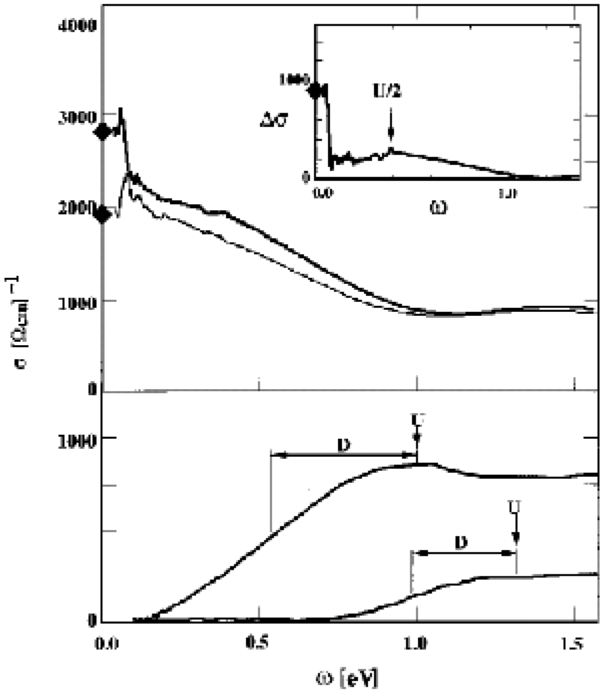


FIG. 30. Local spectral density $\pi D\rho(\omega)$ at T=0, for several values of U, obtained by the iterated perturbation theory approximation. The first four curves (from top to bottom, U/D =1,2,2.5,3) correspond to an increasingly correlated metal, while the bottom one (U/D=4) is an insulator.



V2O3

served. As T is lowered, there is an enhancement of the spectrum at intermediate frequencies of order 0.5 eV; more notably, a sharp low-frequency feature emerges that extends from 0 to 0.15 eV.

DFMT