The Photoemission beamline: What can we do?

1. Bandstructure calculations: quasiparticles

2. The spectral function beyond bandstructure

3. Adding cross sections

4. More realistic transition probabilities

Photoemission Beamline

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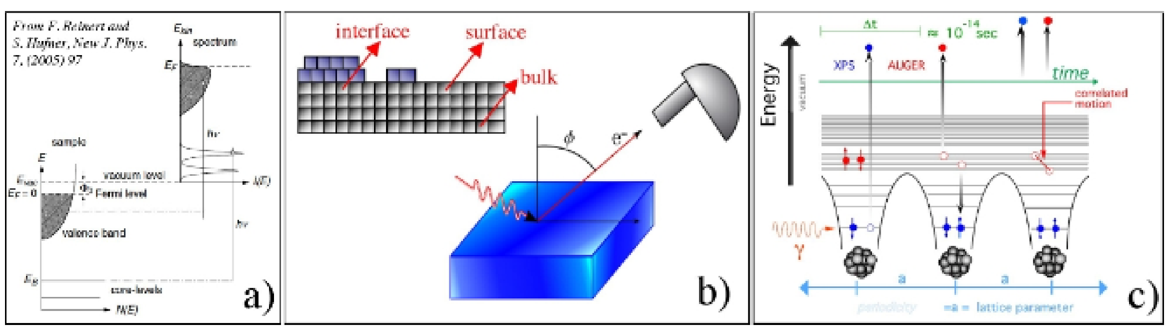
Photoemission Spectroscopy

Photoemission Spectroscopy (also known as Photoelectron Spectroscopy, PES) probes the energy levels of electrons, or more in general, the nature of chemical bonding and electron motion in a substance. PES is based on the *Photoelectric Effect*, which means that when light impinging on a surface is absorbed it induces the emission of electrons. Together with the related Auger spectroscopy, the PES technique is commonly referred as Electron Spectroscopy for Chemical Analysis (ESCA) and was pioneered by Swedish physicist Kai Siegbahn.



http://www.etsf.eu/beamlines/photoemission_spectroscopy

Photoemission Beamline

Beamline Coordinator

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What

- Reliable quasiparticle energies and band-gaps.
- Core and valence photoemission, angle resolved photoemission, thermal effects and electron-phonon coupling.
- Photoemission beyond the sudden approximation, lifetimes of electrons and holes, dependence of spectra on photon energy, spectral functions.
- Auger spectra.

Photoemission Beamline

Where

Metals, semiconductors, molecules, surfaces, nanosystems, including e.g. transition metals and their alloys, transition-metal oxides, graphite, etc.

How

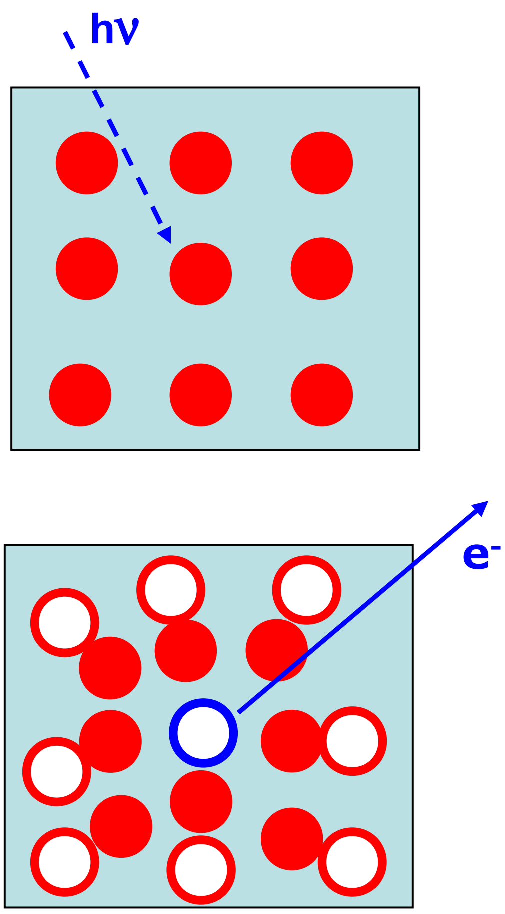
Density functional theory.

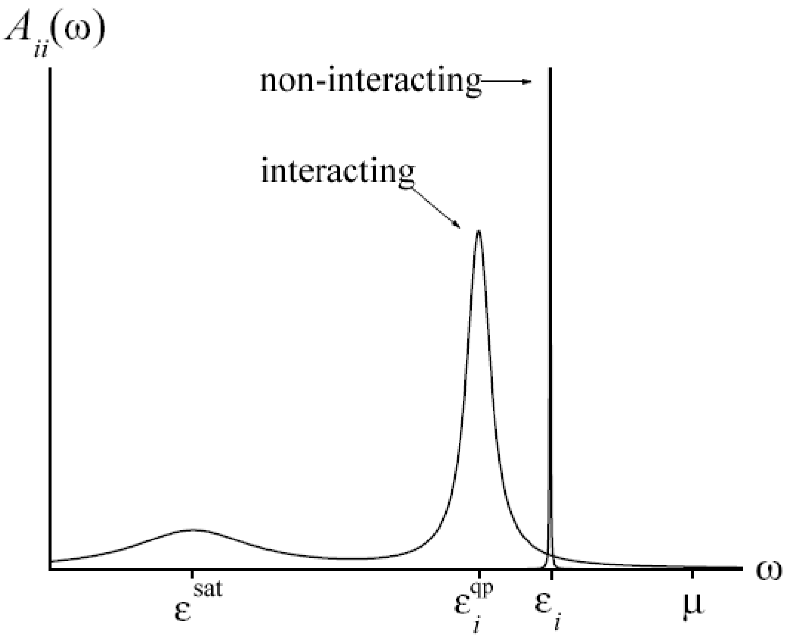
Many-body techniques: GW, T-matrix-approximation.

Codes

Mainly: abinit.org Yambo©

Photoemission

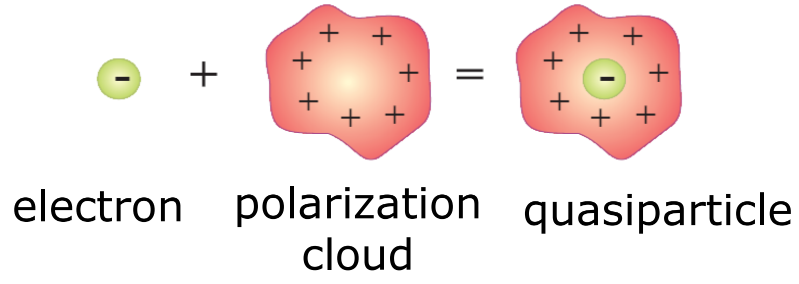




Additional charge

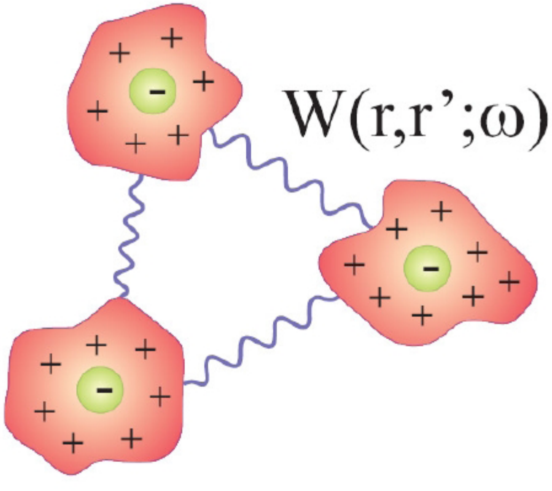
Relaxation – Screening - Correlation

Quasiparticles

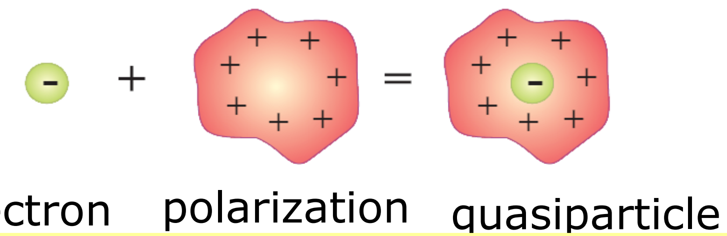


W screened Coulomb potential

$$W(r_1,r_2,\omega) = \varepsilon^{-1}(r_1,r_3,\omega)v(r_3,r_2)$$



Quasiparticles

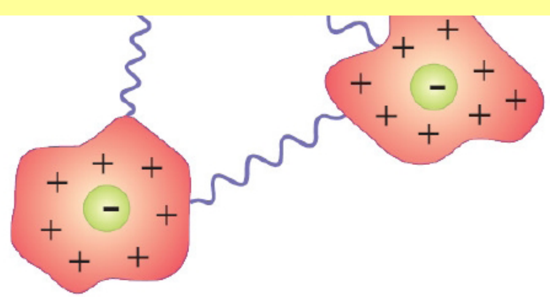


GW approximation:

dynamical screening
of the additional charge

W s

$$W(r_1,r_2,\omega) = \varepsilon^{-1}(r_1,r_3,\omega)v(r_3,r_2)$$



Standard G_0W_0 band structure

Kohn-Sham equation (DFT):

$$H_0(r)\varphi_{\mathrm{KS}}(r) + V_{xc}(r)\varphi_{\mathrm{KS}}(r) = \epsilon_{\mathrm{KS}}\varphi_{\mathrm{KS}}(r)$$

Quasiparticle equation (MBPT):

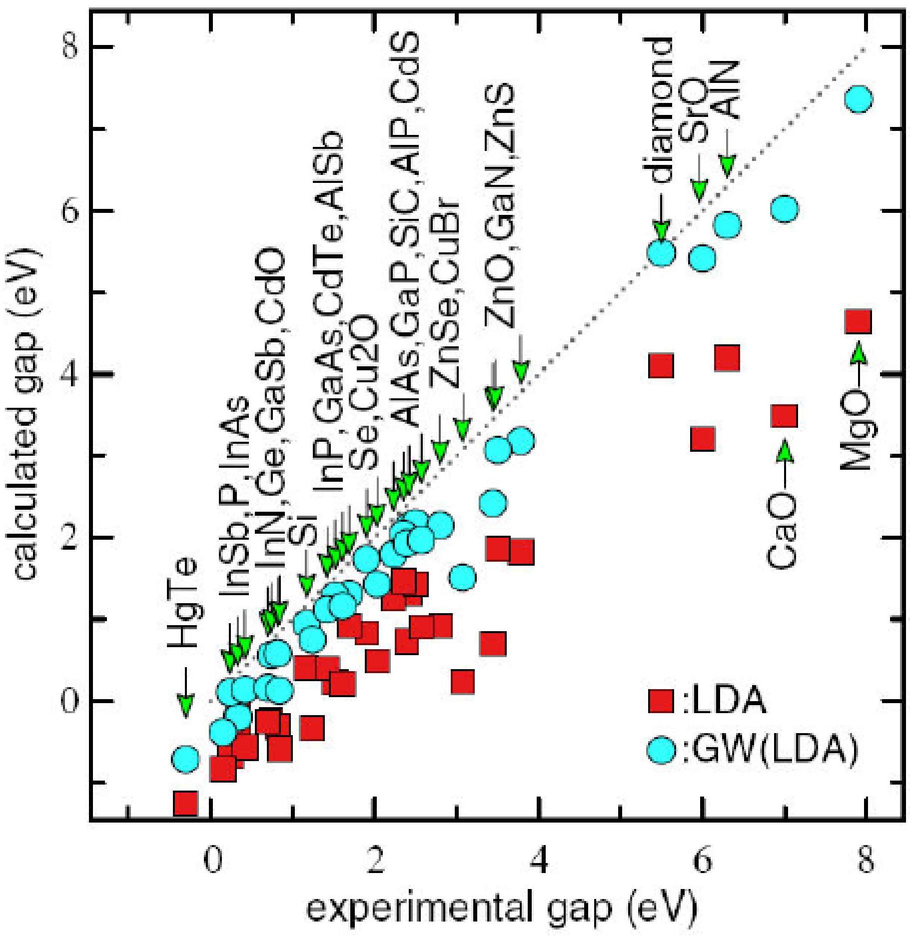
$$H_0(r)\phi_{\mathrm{QP}}(r) + \int dr'\ \Sigma(r, r', \omega = E_{\mathrm{QP}})\ \phi_{\mathrm{QP}}(r') = E_{\mathrm{QP}}\,\phi_{\mathrm{QP}}(r)$$

Quasiparticle energies = 1st order perturbative corrections

$$E_{\mathrm{QP}} - \epsilon_{\mathrm{KS}} = \langle\varphi_{\mathrm{KS}}|\Sigma - V_{xc}|\varphi_{\mathrm{KS}}\rangle$$

See: M. Hybersten and S.G. Louie, PRB 34 (1986);
R.W. Godby, M Schlüter and L.J. Sham, PRB 37 (1988)

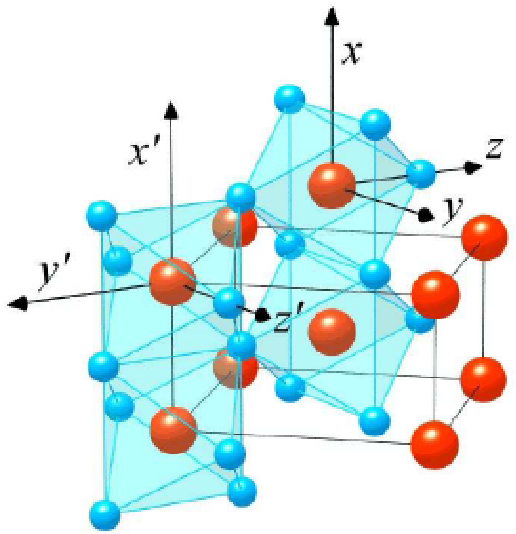
Standard G_0W_0 band structure

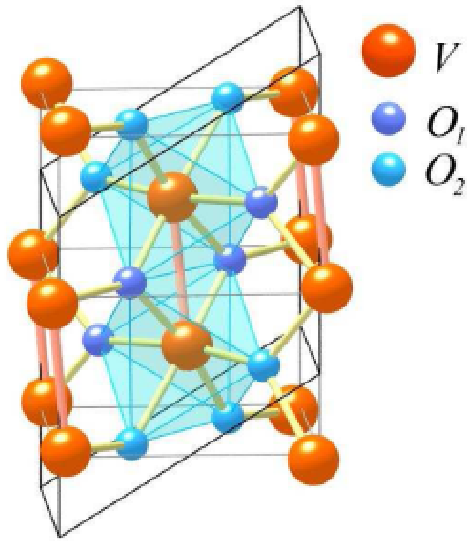


From: van Schilfgaarde *et al.*, PRL 96 (2006)

Is VO_2 strongly correlated ?

VO_2: double phase transition





T_c = 340 K
(Morin '59)

for $T > T_c$

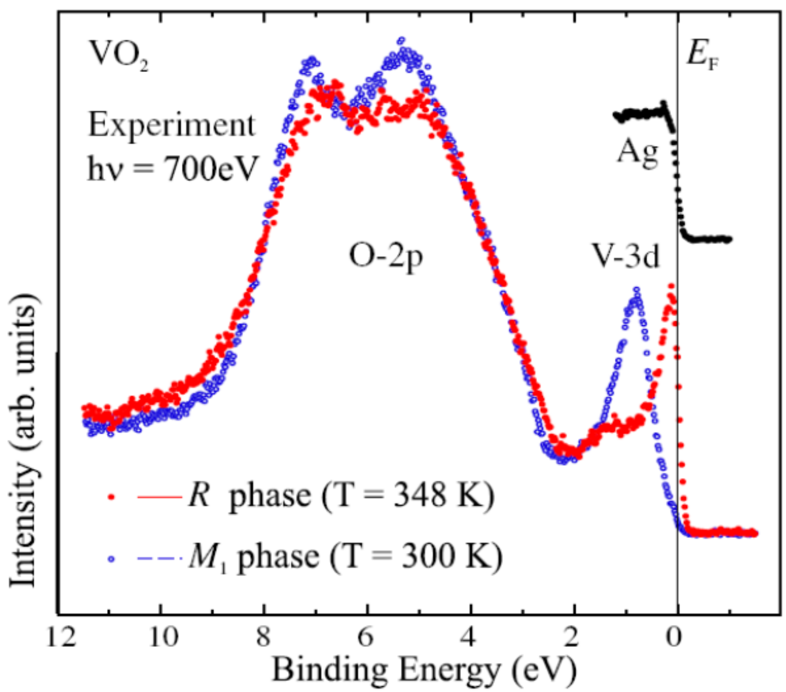
Rutile + Metal

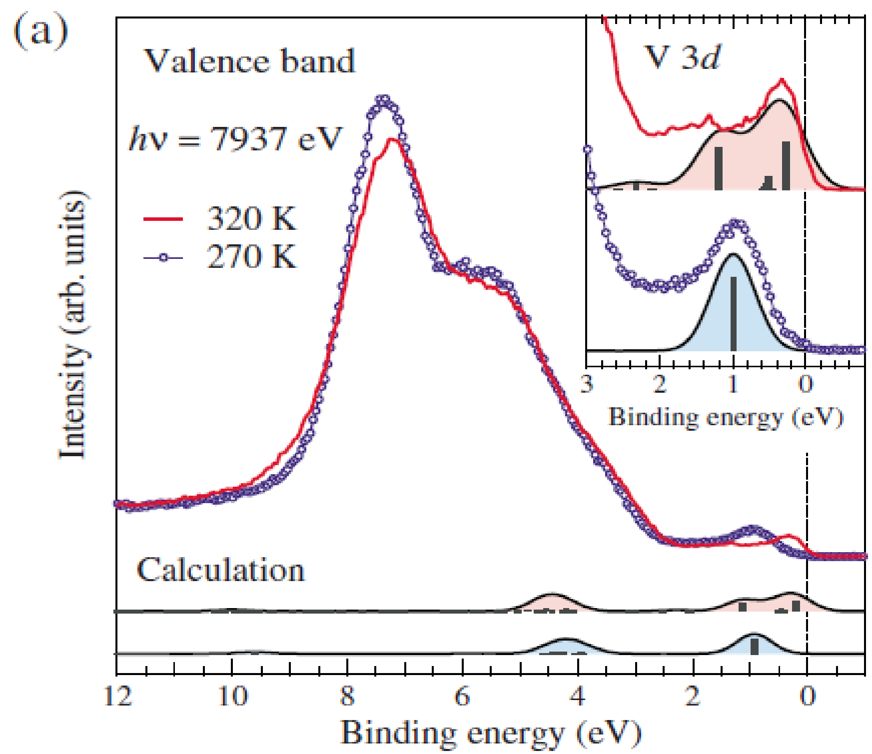
for $T < T_c$

Monoclinic + Insulator

Mechanism? Role of electronic correlation?

Photoemission spectra



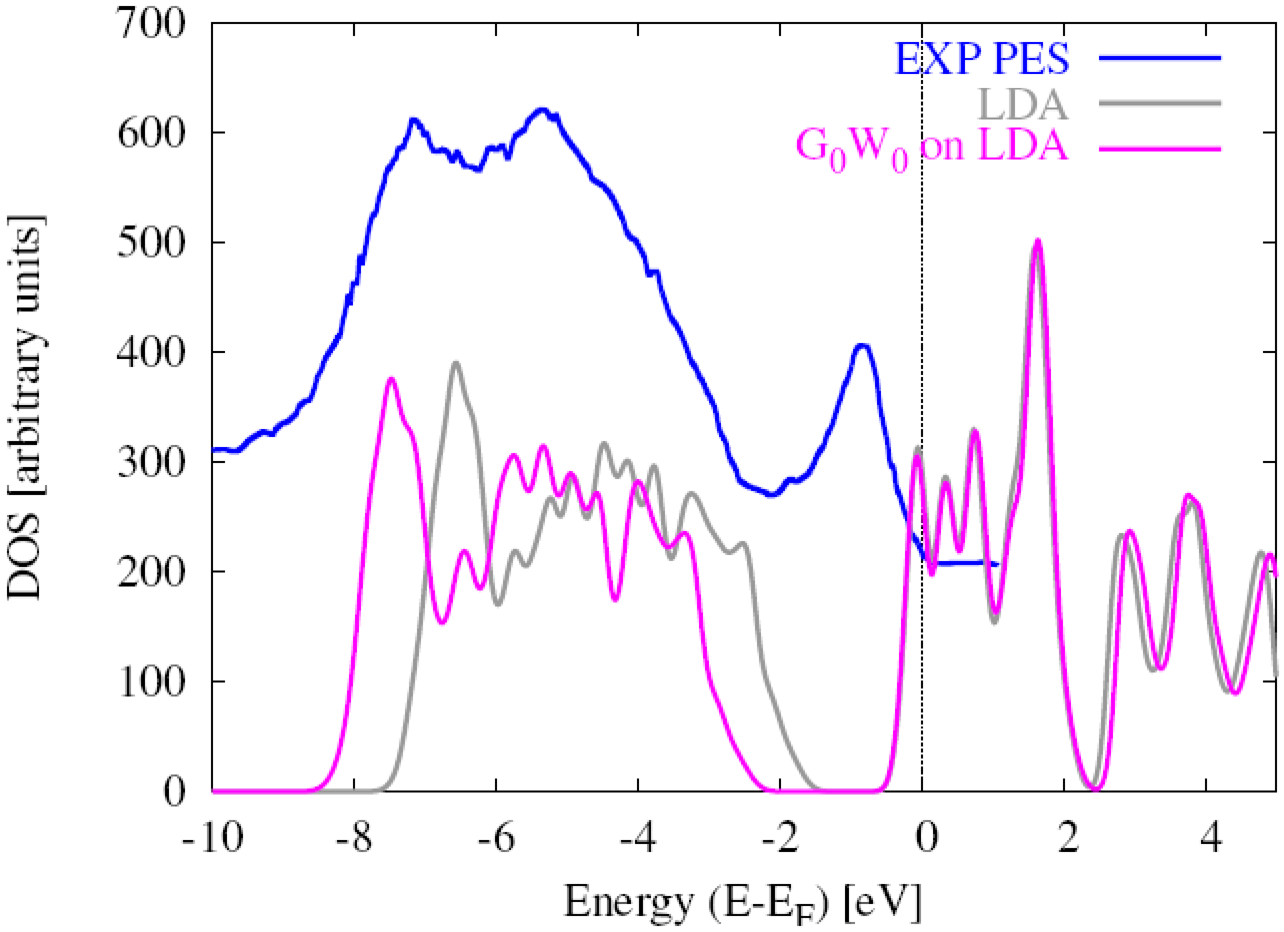


From: Koethe *et al*. PRL 97 (2006)

From: Eguchi *et al*. PRB 78 (2008)

Similar result in Suga *et al.,* New J. Phys. 11 (2009)

The insulator: standard G_0W_0



Beyond standard G_0W_0

Kohn-Sham equation (DFT):

$$H_0(r)\varphi_{\rm KS}(r) + V_{xc}(r)\varphi_{\rm KS}(r) = \epsilon_{\rm KS}\varphi_{\rm KS}(r)$$

Quasiparticle equation (MBPT):

$$H_0(r)\phi_{\rm QP}(r) + \int dr'\ \Sigma(r, r', \omega = E_{\rm QP})\ \phi_{\rm QP}(r') = E_{\rm QP}\,\phi_{\rm QP}(r)$$

Quasiparticle energies = 1^{st} order perturbative corrections

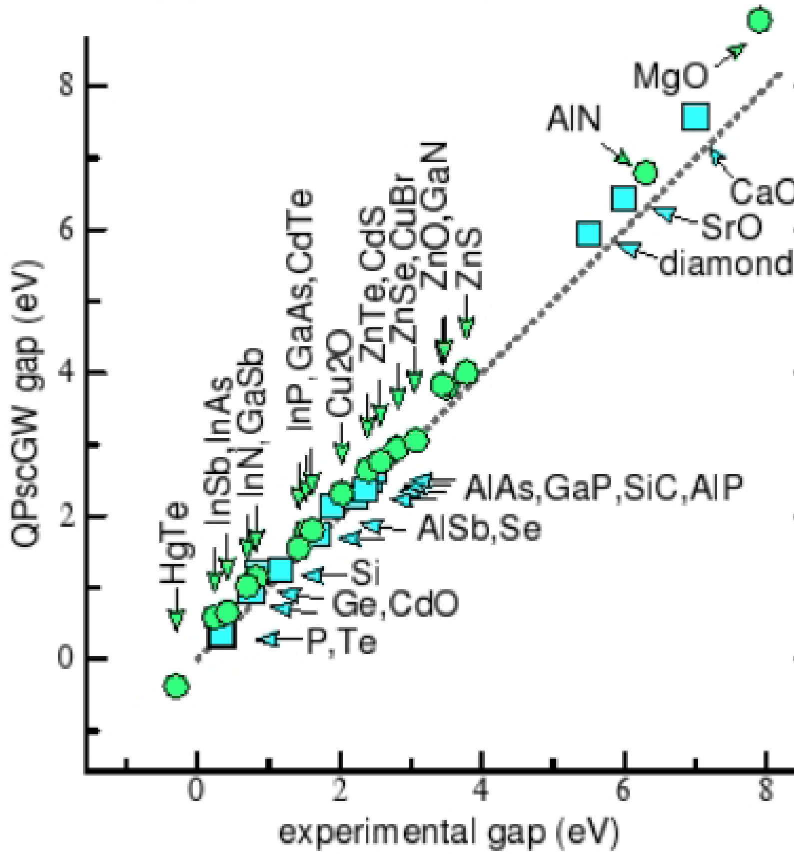
$$E_{\rm QP} - \epsilon_{\rm KS} = \langle\varphi_{\rm KS}|\Sigma - V_{xc}|\varphi_{\rm KS}\rangle$$

See: M. Hybersten and S.G. Louie, PRB 34 (1986);
R.W. Godby, M Schlüter and L.J. Sham, PRB 37 (1988)

Beyond standard G_0W_0

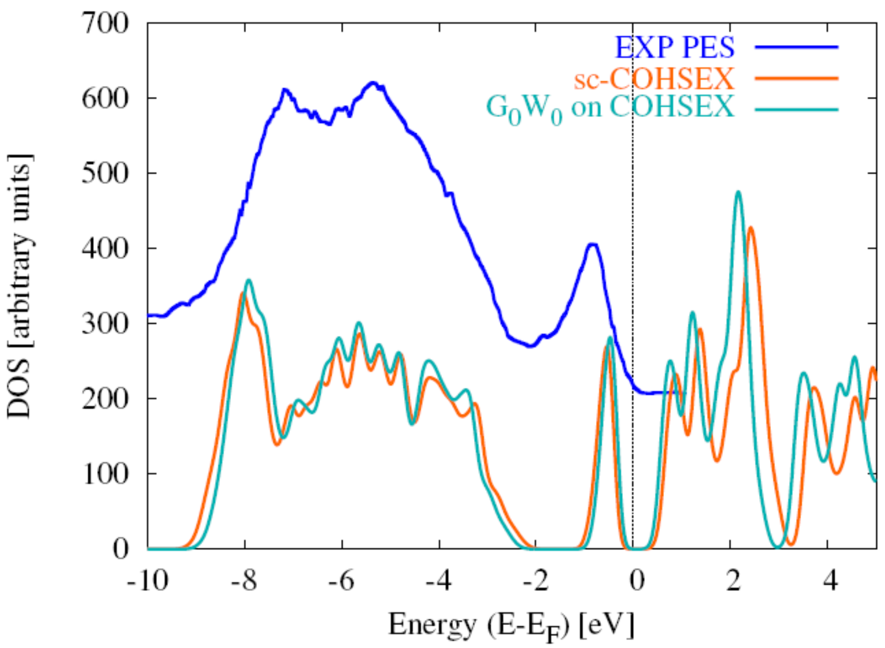
- DFT with EXX,… (e.g. Rinke *et al.* 2005)
- hybrid functionals (e.g. Fuchs *et al.* 2006)
- LDA+U (e.g. Jiang *et al.* 2009)
- effective quasiparticle Hamiltonians:
  - COHSEX approximation (Hedin 1965, Bruneval 2005)
  - GWscQP scheme (Faleev *et al.* 2004)
  - Löwdin procedure (Sakuma *et al.* 2009)

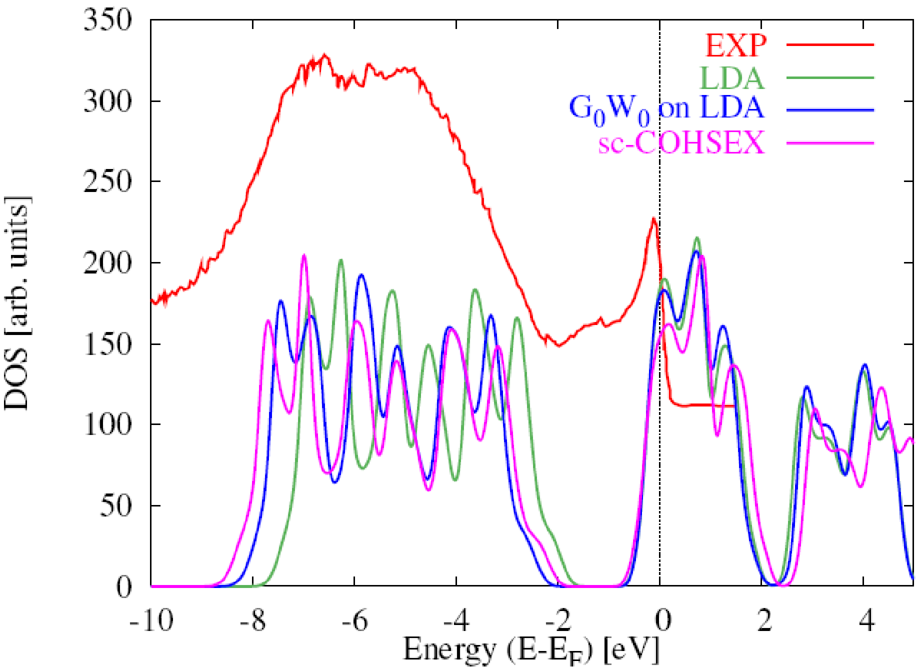
For VO_2 see: M. Gatti, F. Bruneval, V. Olevano, L. Reining, PRL 99 (2007);
R. Sakuma, T. Miyake, F. Aryasetiawan, PRB 78 (2008)



From: van Schilfgaarde *et al*., PRL 96 (2006)

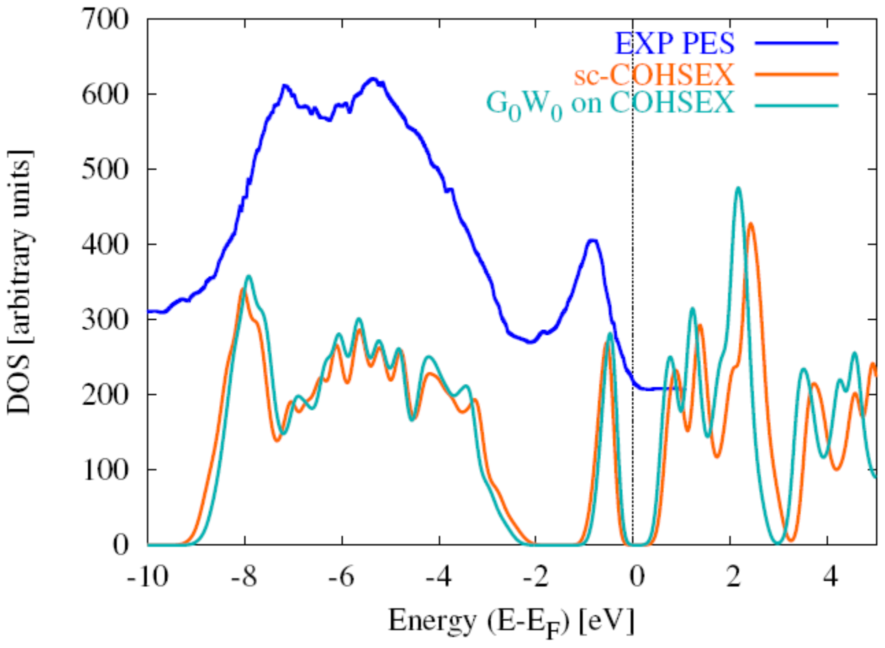
GW Quasiparticle DOS

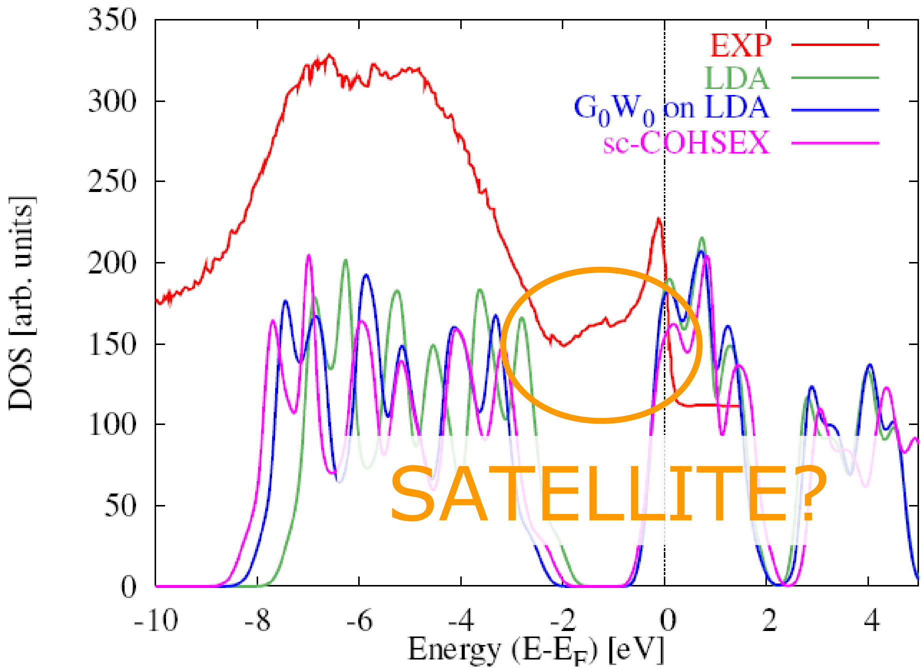




M. Gatti, F. Bruneval, V. Olevano, L. Reining, PRL 99 (2007)

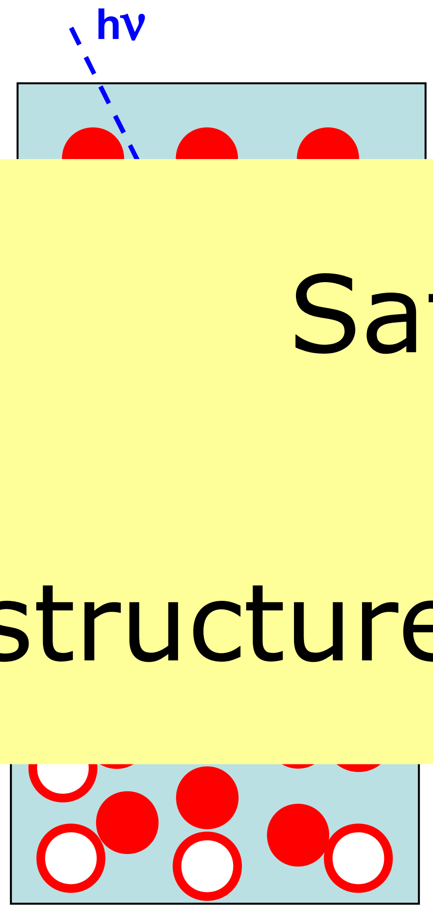
GW Quasiparticle DOS





M. Gatti, F. Bruneval, V. Olevano, L. Reining, PRL 99 (2007)

Photoemission



Satellites in GW:

structures in $W(\omega) = \varepsilon^{-1}(\omega)v$

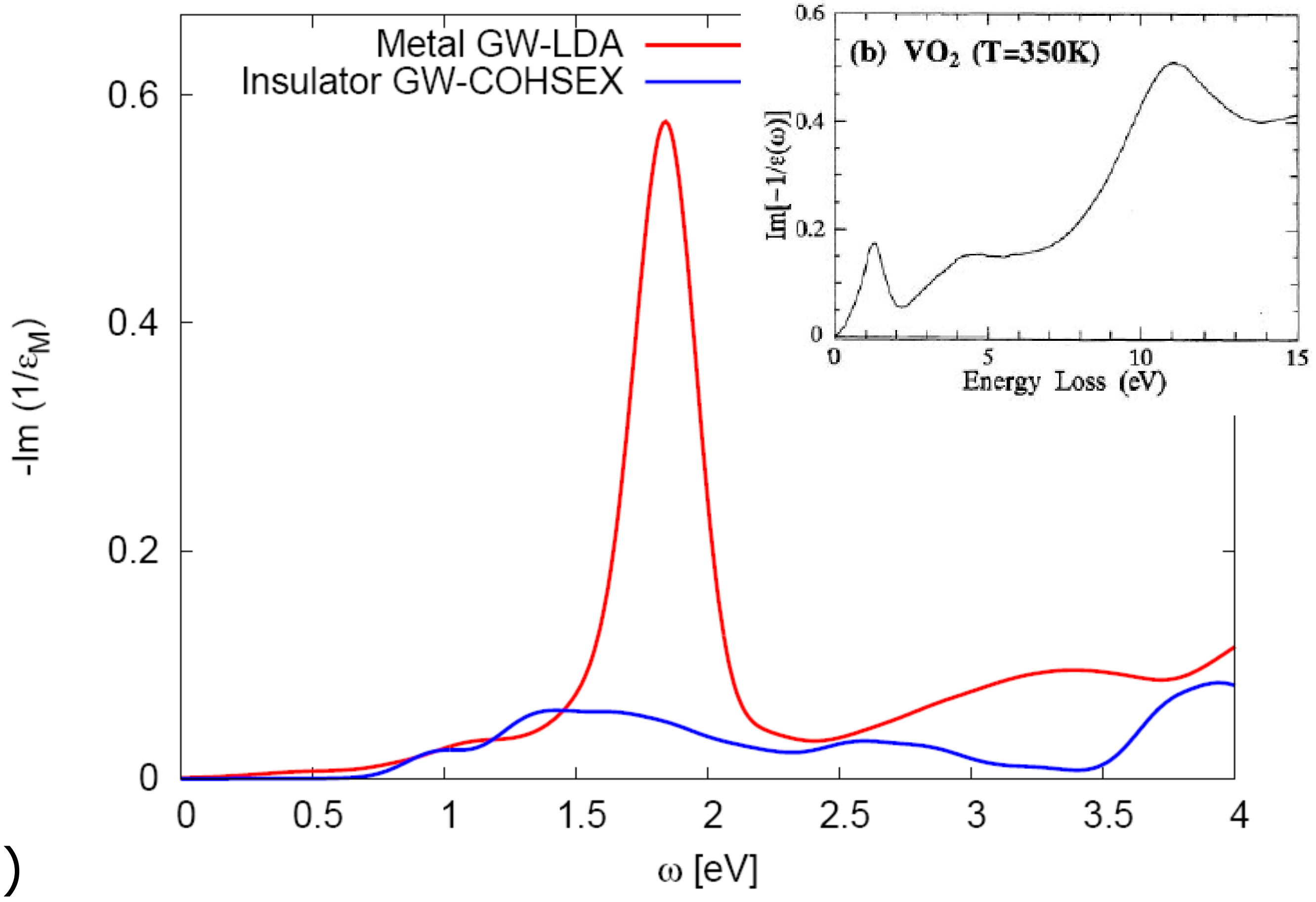
Quasiparticles and satellites

Additional charge

Relaxation – Screening - Correlation

VO_2: electron energy loss

$-\mathrm{Im}\,\varepsilon^{-1}(\omega)$

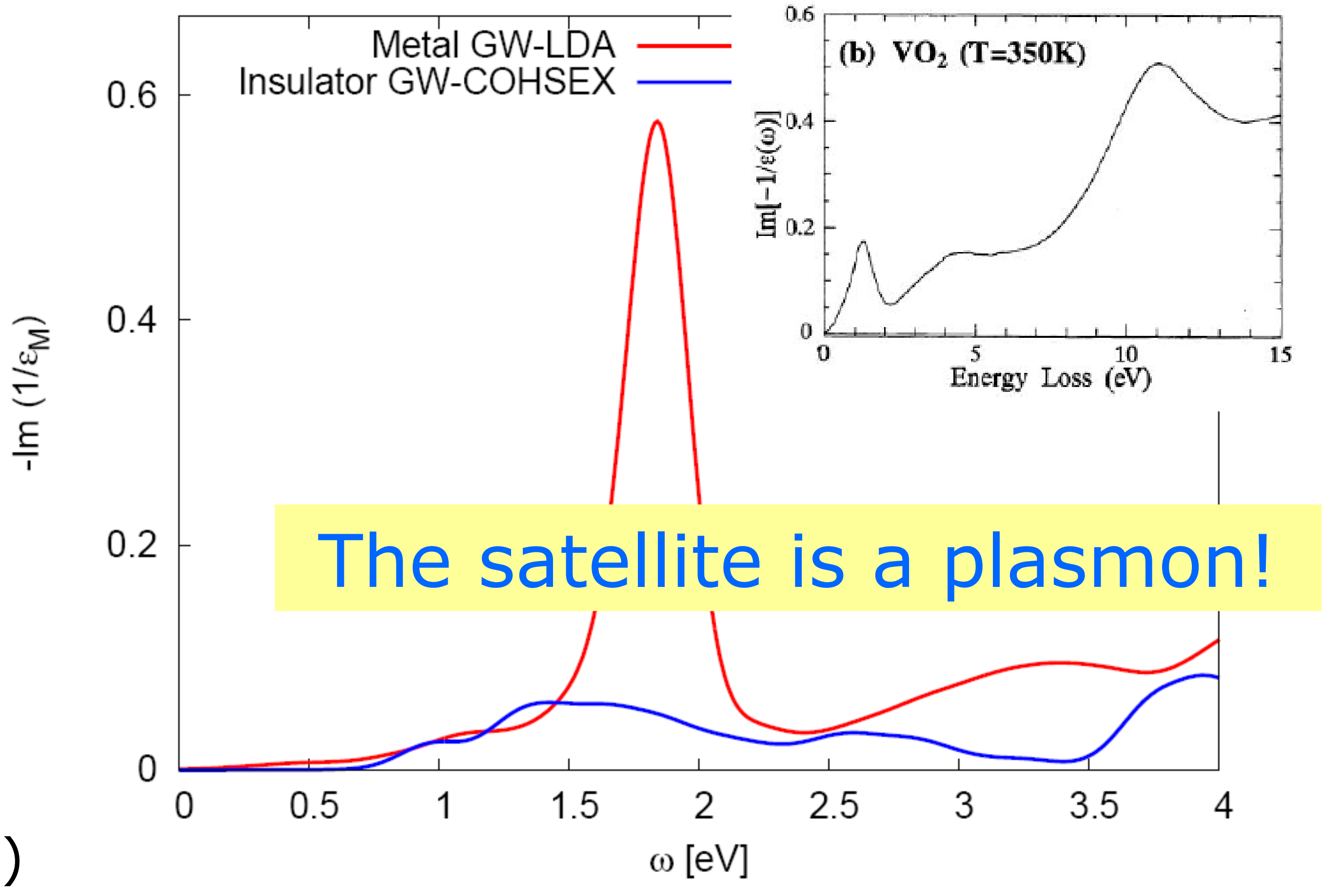


(small finite q)

Exp.: Abe *et al.* Jpn. J. Appl. Phys (1997)

VO_2: electron energy loss

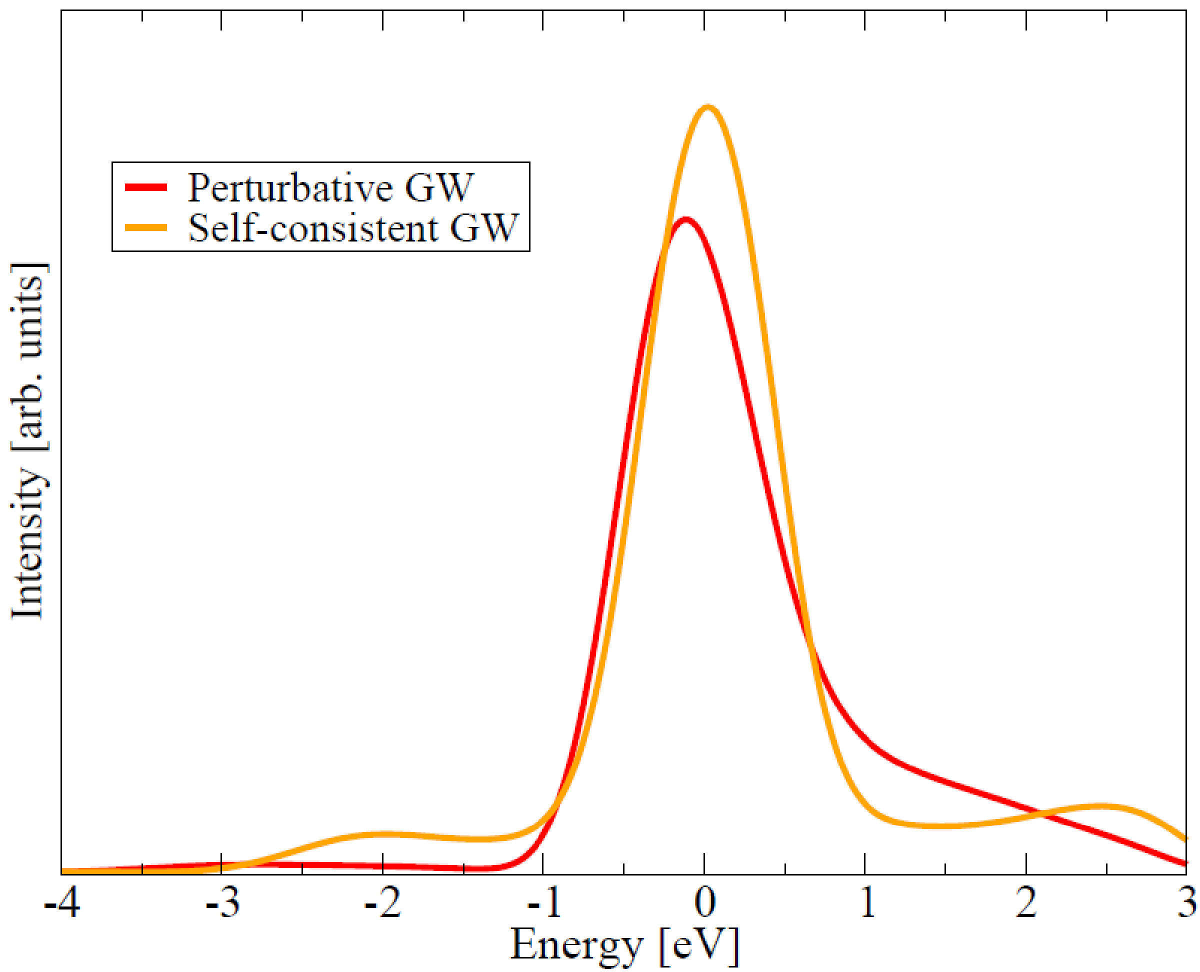
$-\mathrm{Im}\,\varepsilon^{-1}(\omega)$



(small finite q)

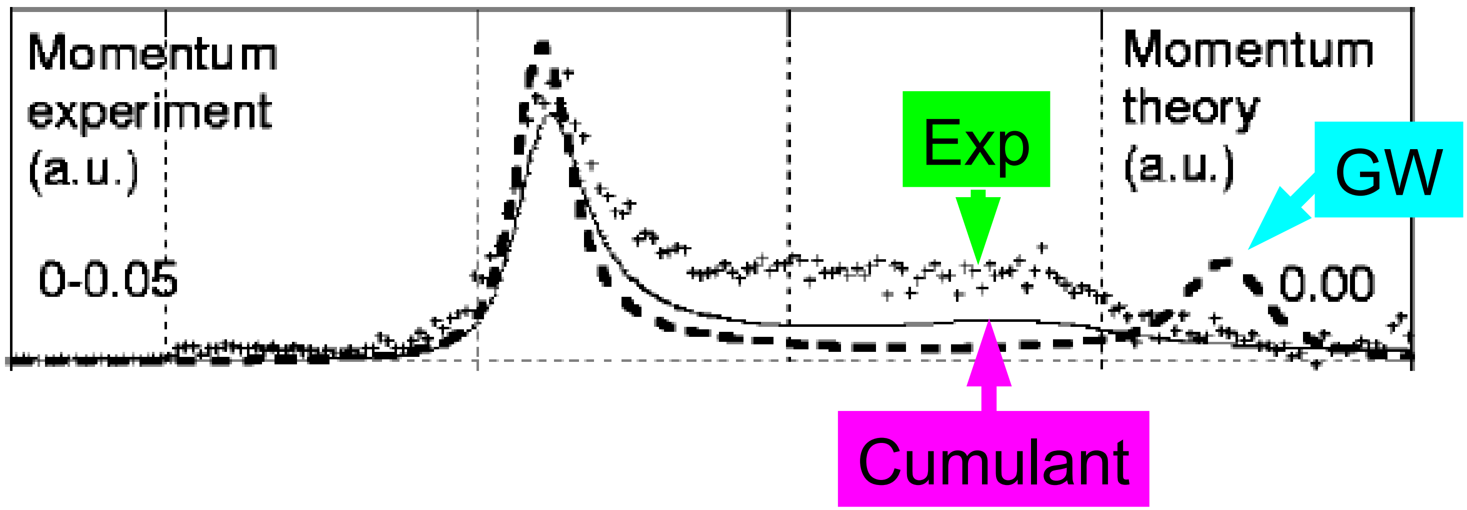
Exp.: Abe *et al.* Jpn. J. Appl. Phys (1997)

The metal: spectral function



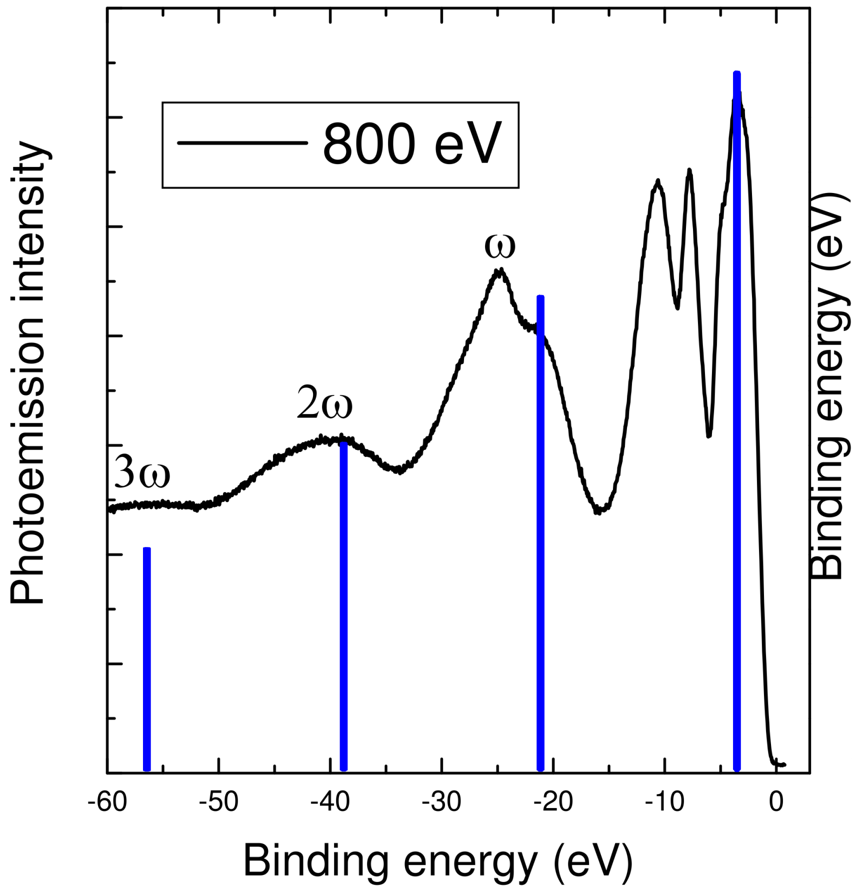
Beyond GW: cumulant expansion

Silicon



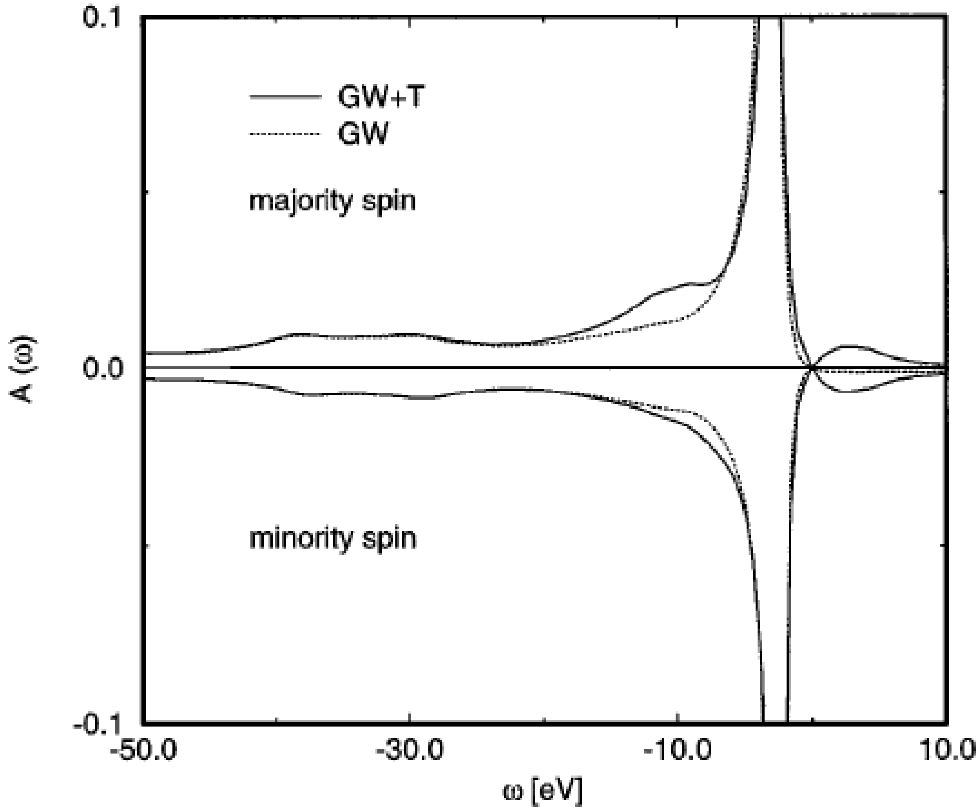
Kheifets, Sashin, Vos, Weigold, Aryasetiawan, PRB 68 (2003)

Beyond GW: cumulant expansion



Exp. F. Sirotti (Soleil) – Theo. M. Guzzo (Ecole Polytechnique) - 2010

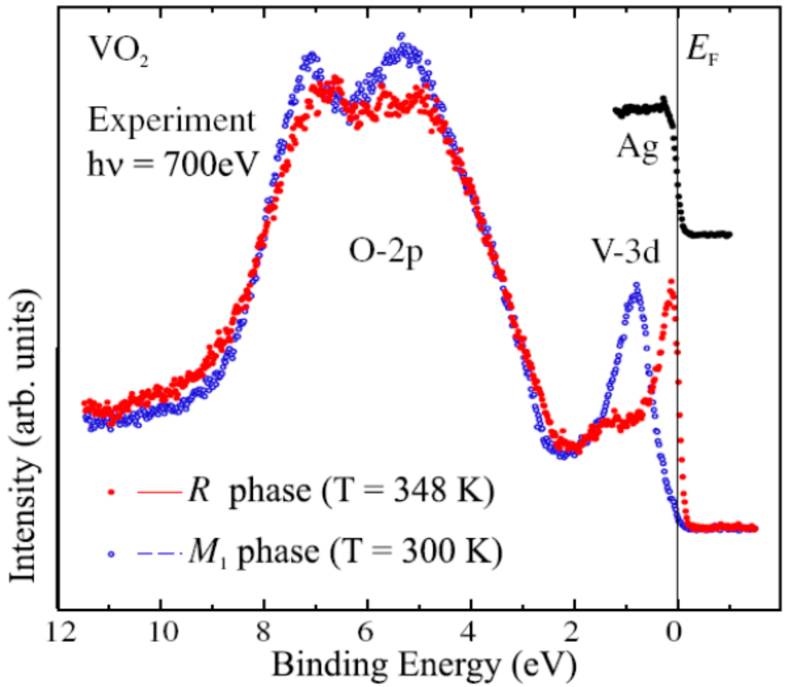
Beyond GW: T matrix



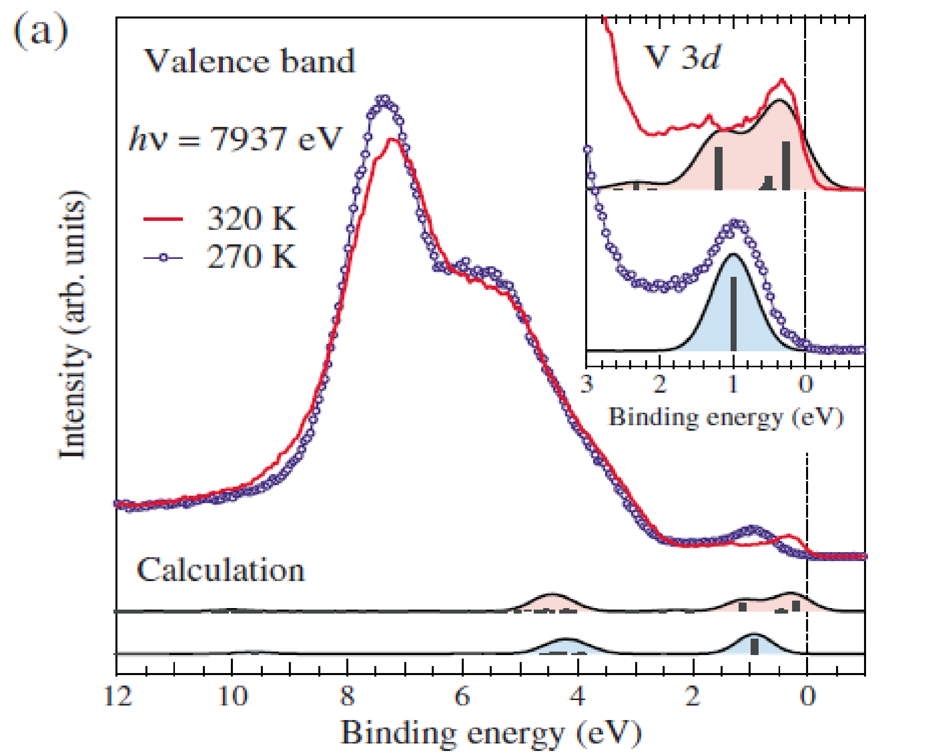
6 eV satellite in Ni: 2-hole bound state

Springer, Aryasetiawan, Karlsson, PRL 80, 2389 (1998)

Photoemission spectra



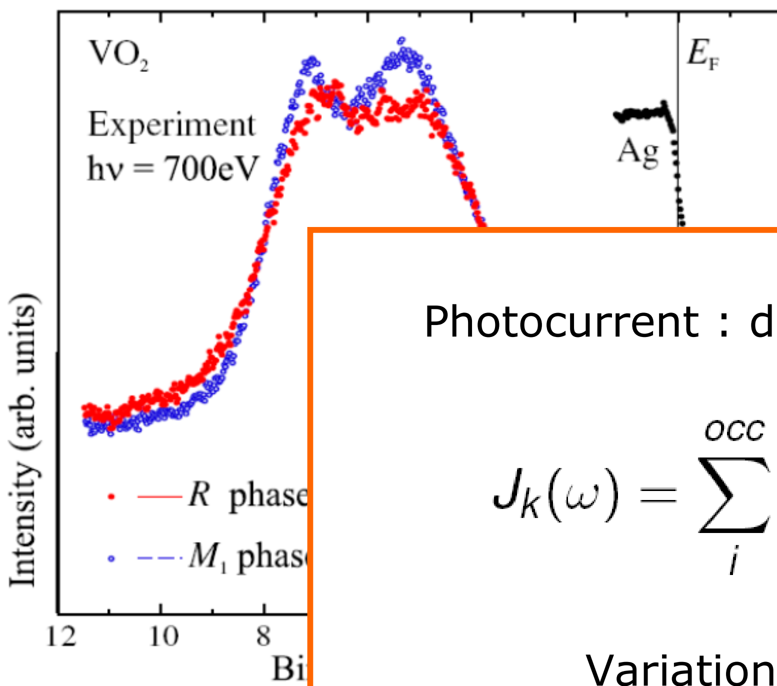
From: Koethe *et al*. PRL 97 (2006)

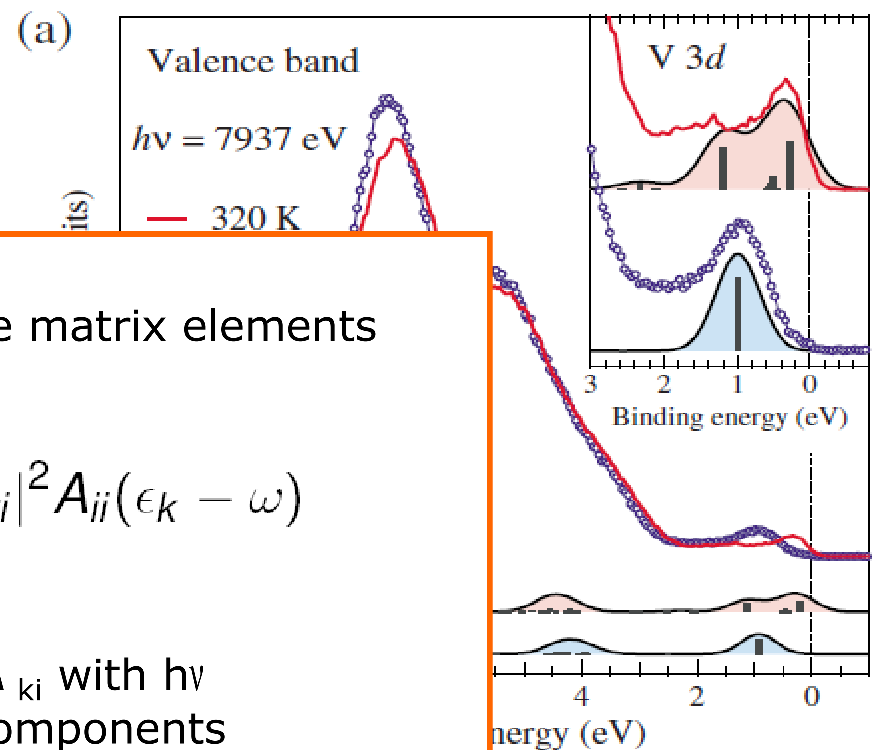


From: Eguchi *et al*. PRB 78 (2008)

Similar result in Suga *et al.,* New J. Phys. 11 (2009)

Photoemission spectra





Photocurrent : dipole matrix elements

$$J_k(\omega) = \sum_i^{occ} |\Delta_{ki}|^2 A_{ii}(\epsilon_k - \omega)$$

Variation of Δ_{ki} with hν
tunes s,p,d components

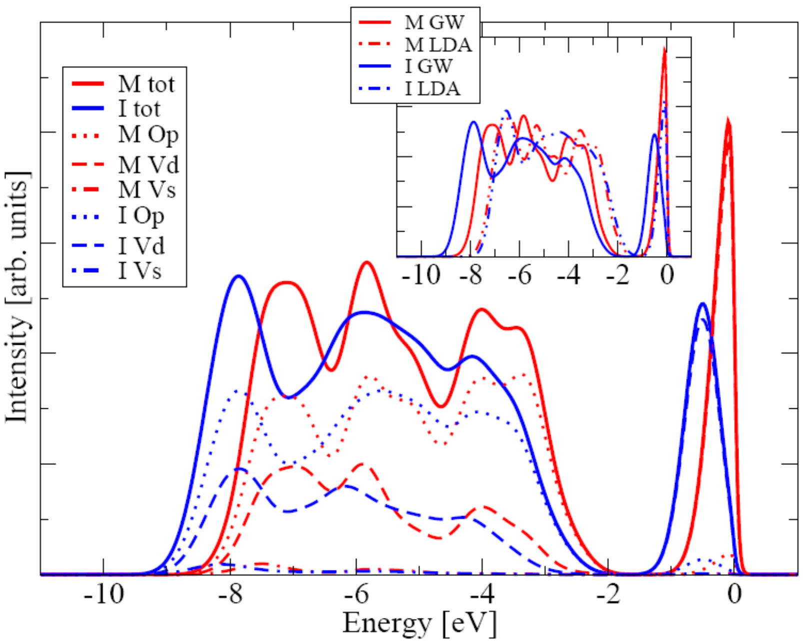
From: Koethe *et al*. PRL 97 (2006)

From: Eguchi *et al*. PRB 78 (2008)

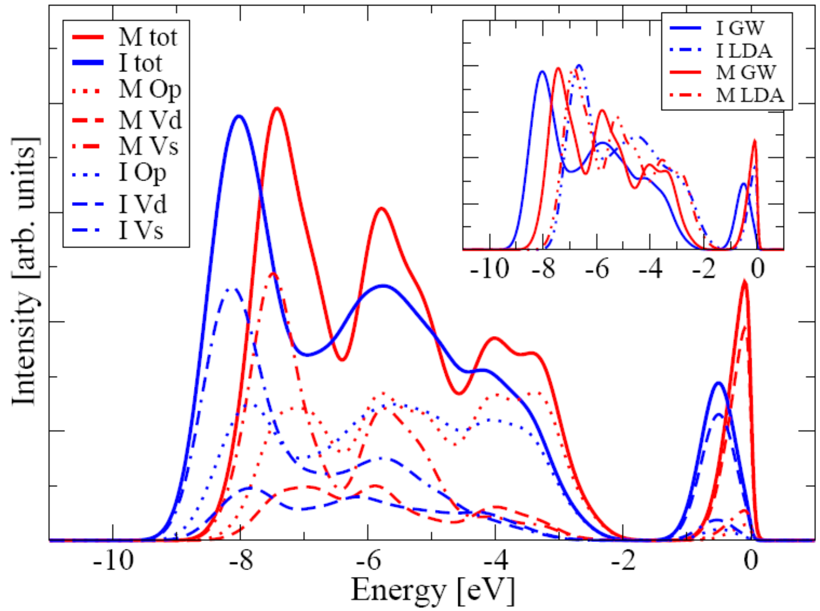
Similar result in Suga *et al.,* New J. Phys. 11 (2009)

Identification of the peak: QP DOS

XPS



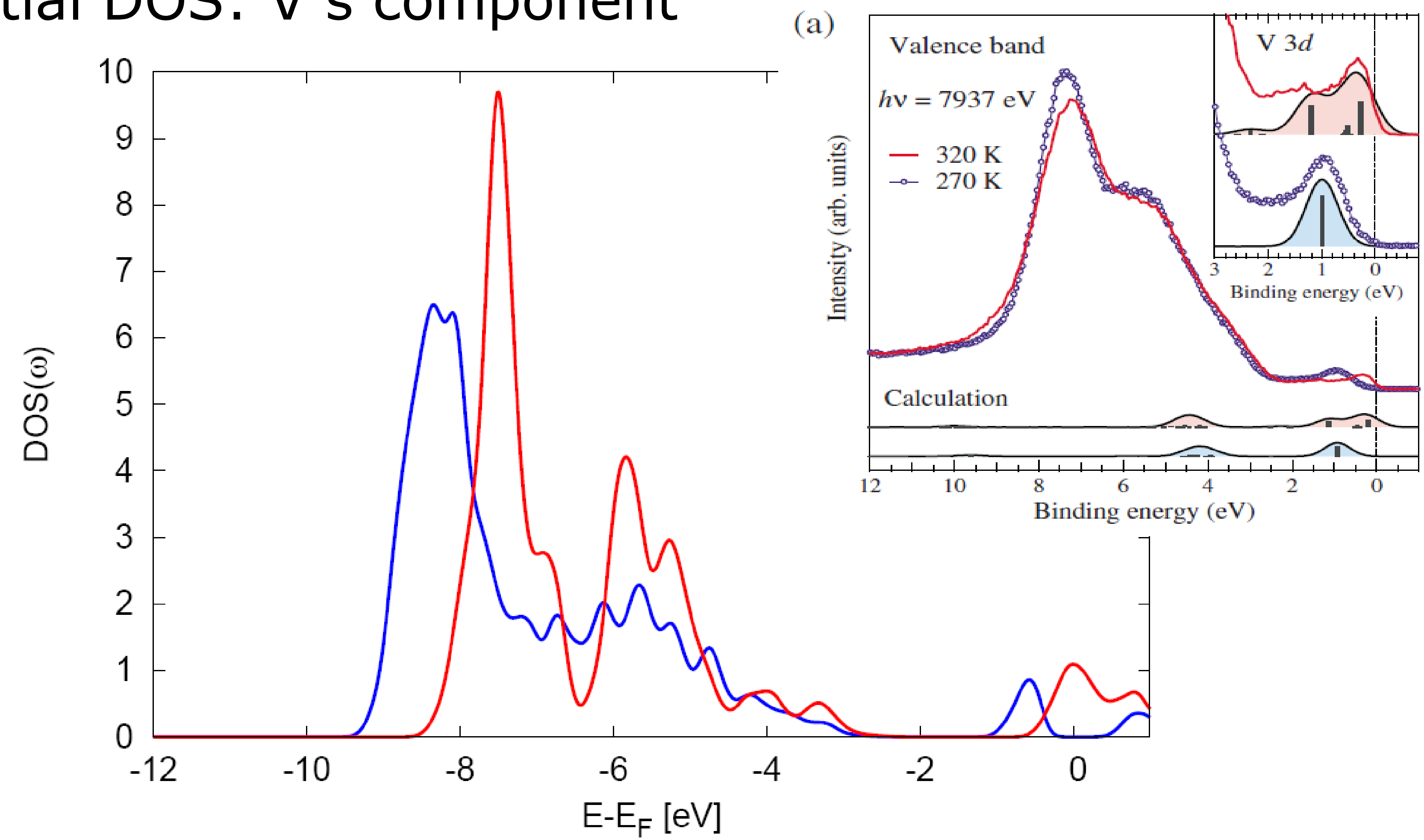
HAXPES



GW QP DOS weighted with cross sections
from Scofield and Yeh-Lindau

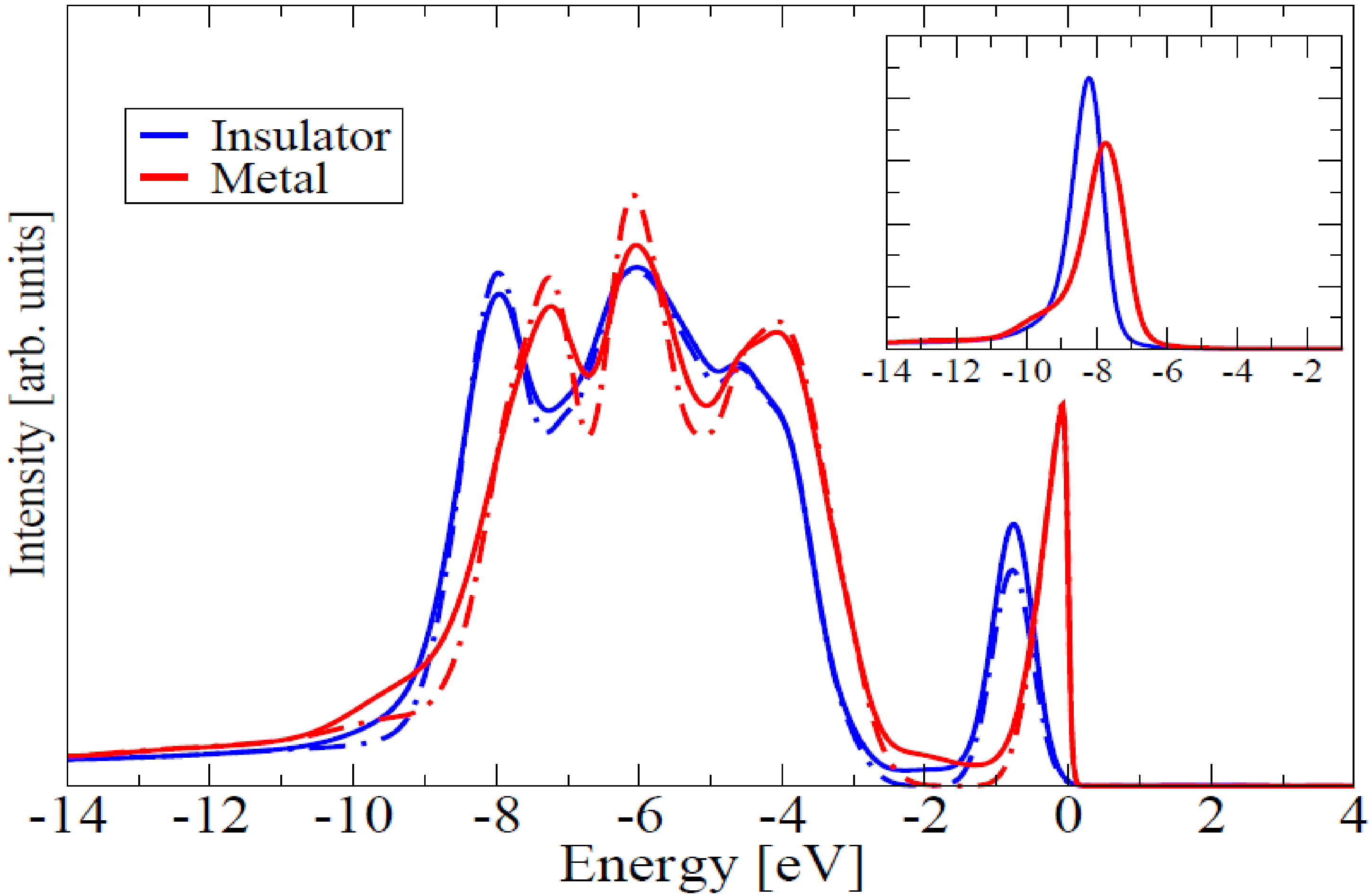
Identification of the peak: QP DOS

Partial DOS: V s component

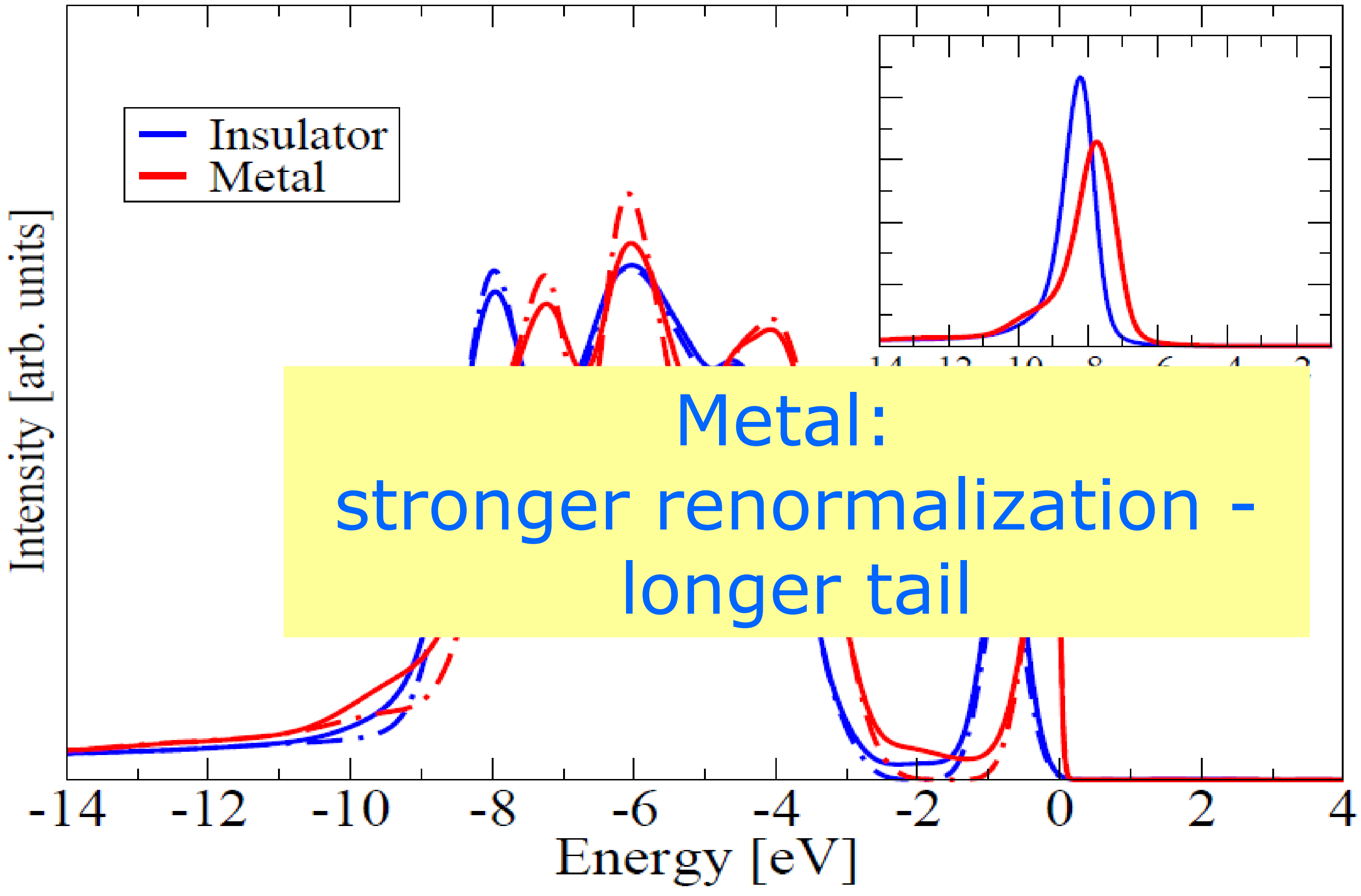


See also: E. Papalazarou, M. Gatti, et al., PRB 80 (2009).

Beyond QP: GW spectral function



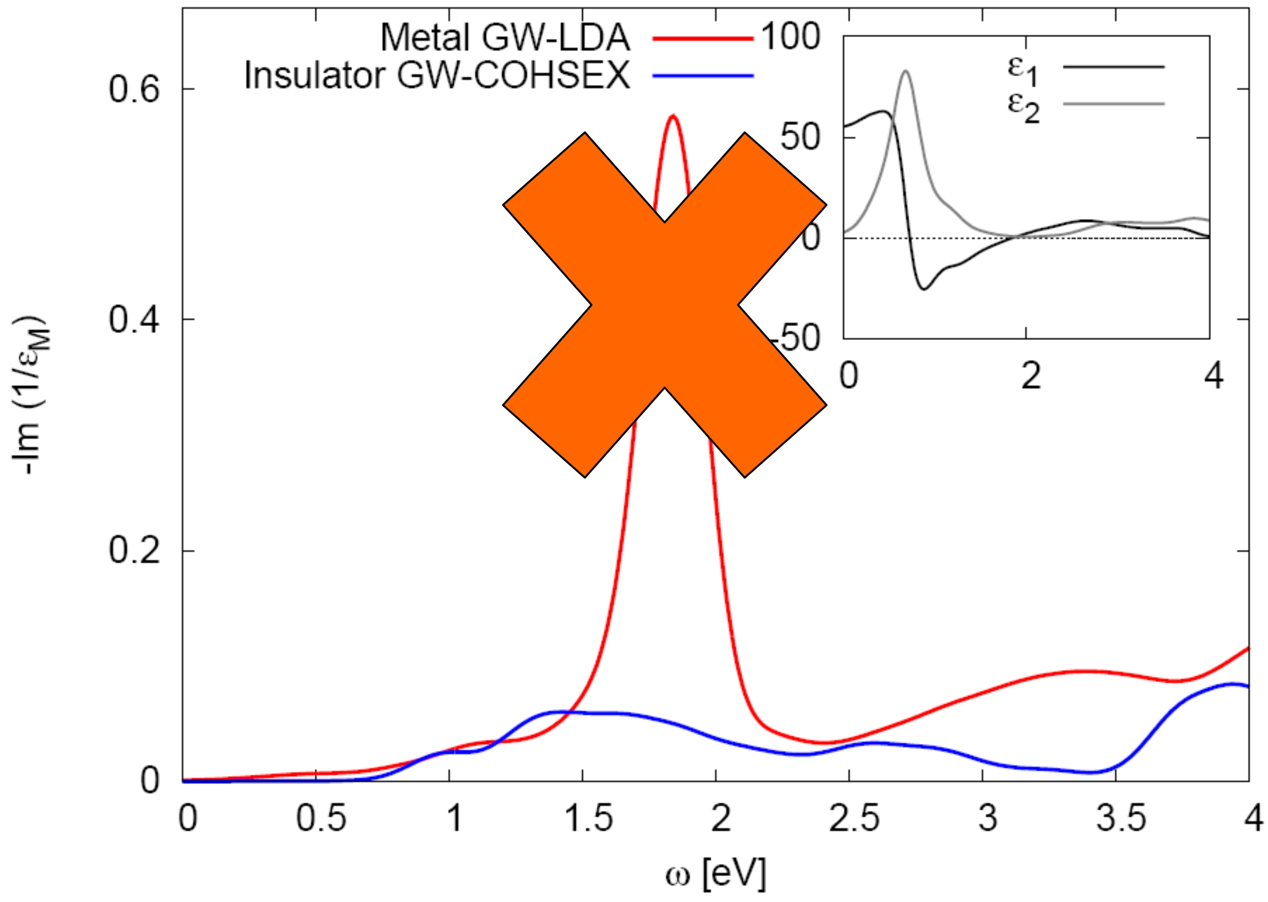
Beyond QP: GW spectral function



Metal:
stronger renormalization -
longer tail

VO_2: electron energy loss

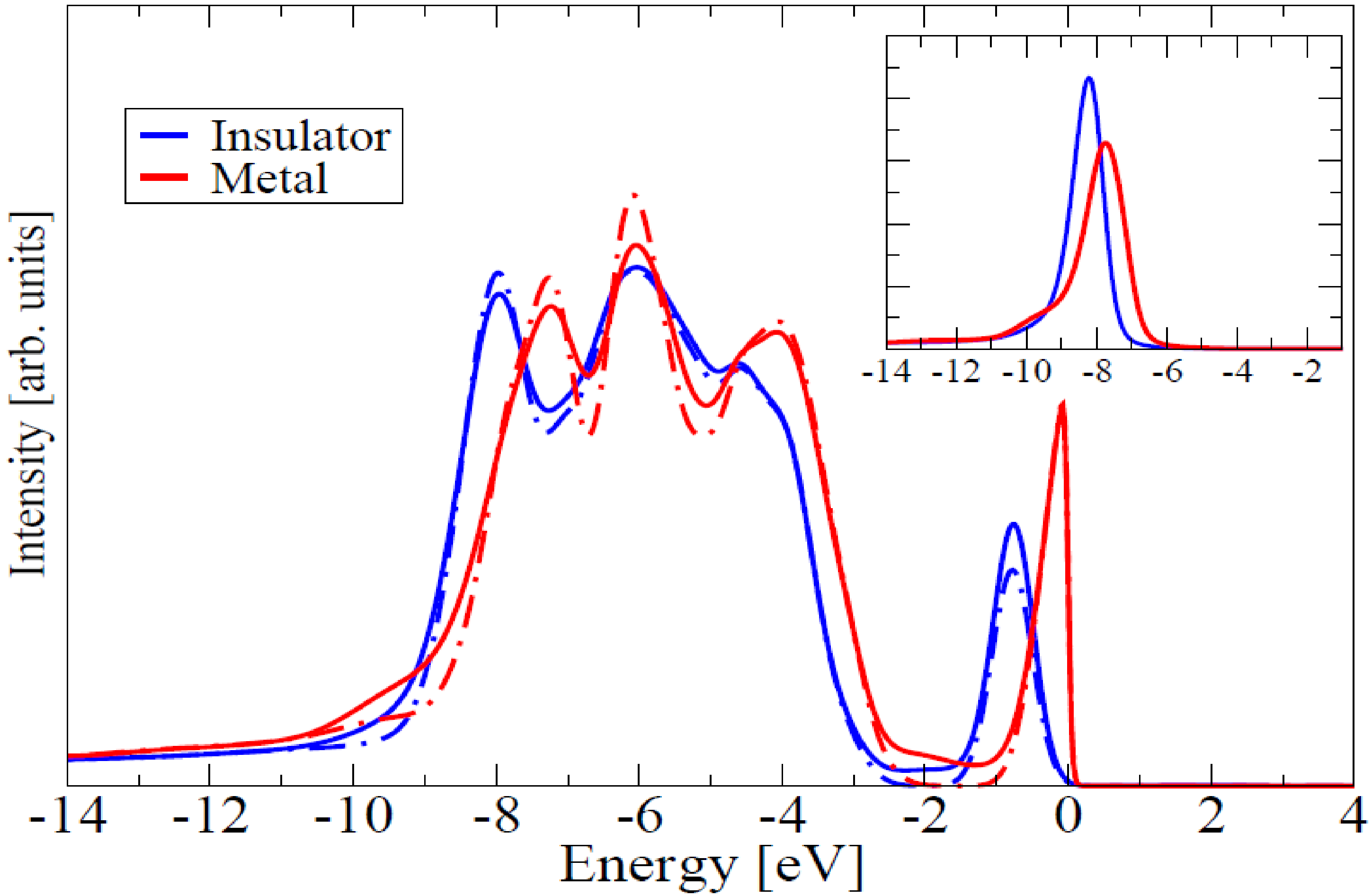
$-\mathrm{Im}\,\varepsilon^{-1}(\omega)$



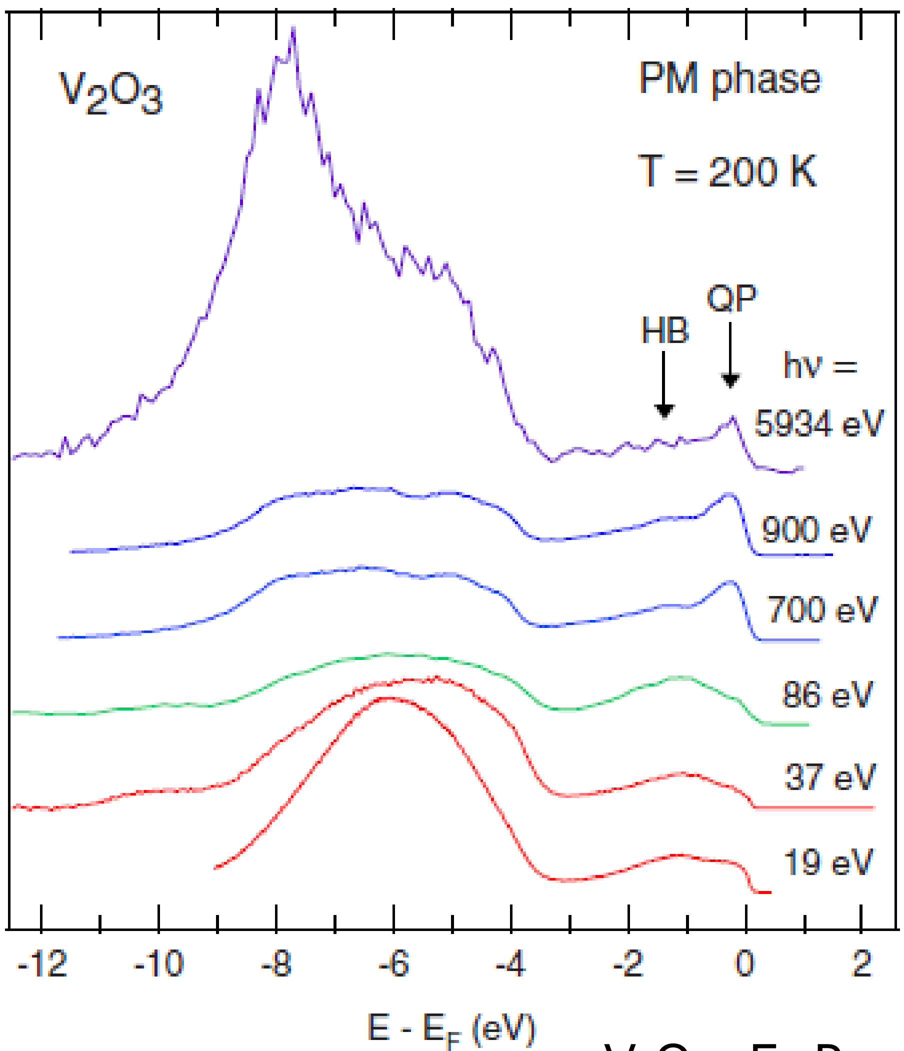
(small finite q)

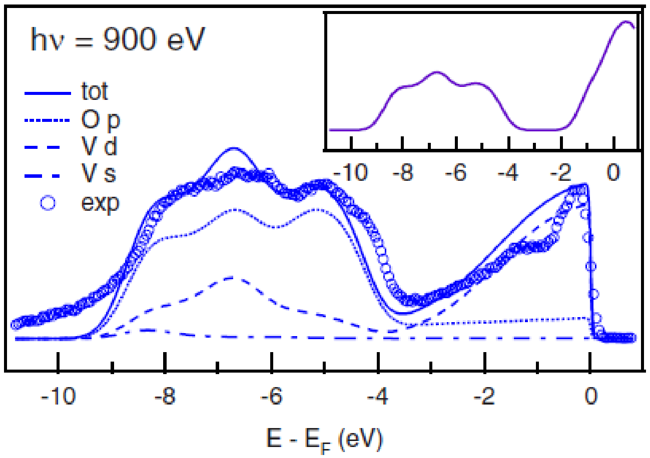
see also Exp.: Abe *et al.* Jpn. J. Appl. Phys (1997)

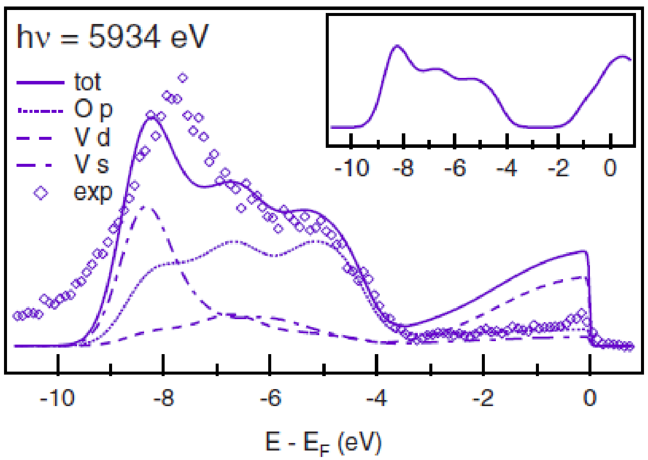
Beyond QP: GW spectral function



More on cross sections





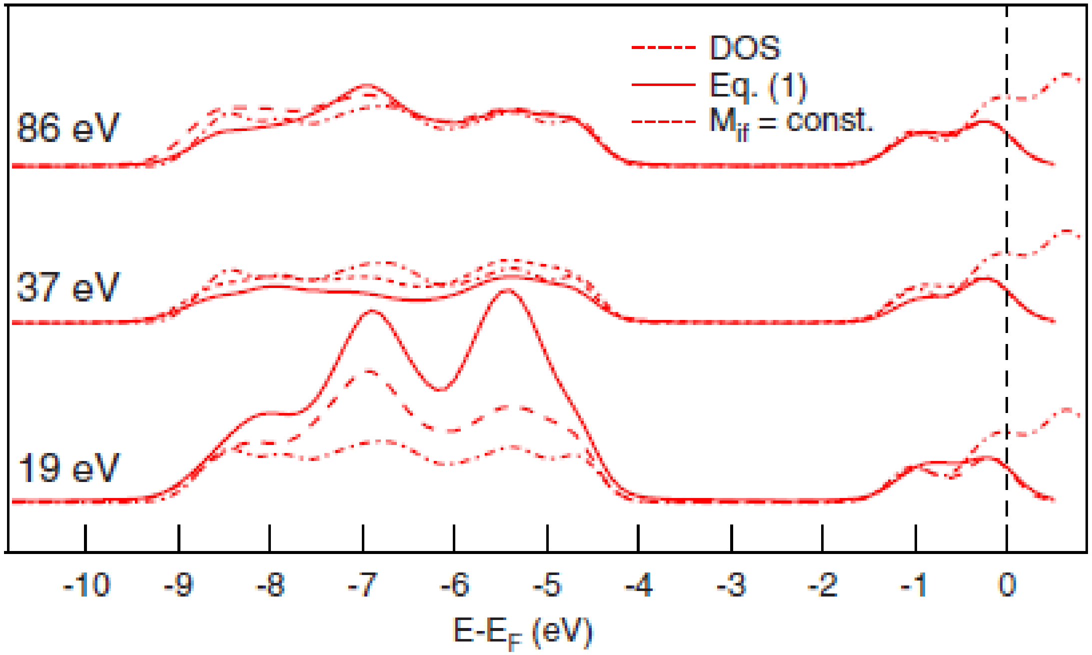


V_2O_3: E. Papalazarou et al, PRB 80 (2009)

More on cross sections

$$J_E(\omega) = \sum_{if} |M_{if}|^2 \delta(\epsilon_f - \epsilon_i - E)\, \delta(\epsilon_f - \omega).$$

E = photon energy; ω = photoelectron energy; ε_i, ε_f = initial, final states



E. Papalazarou *et al*, PRB 80 (2009)

The Photoemission beamline: What can we do?

1. Bandstructure calculations: quasiparticles

2. The spectral function beyond bandstructure

3. Adding cross sections

4. More realistic transition probabilities

See: http://www.etsf.eu/beamlines/photoemission_spectroscopy