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A direct approach to the calculation of many-body Green's functions: beyond quasiparticles

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A direct approach to the calculation of many-body Green' s functions: beyond quasiparticles

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Palaiseau Theoretical Spectroscopy Group & friends

A direct approach to the calculation of many-body Green' s functions: beyond quasiparticles

→ Many-body perturbation theory: GW

→ An alternative strategy

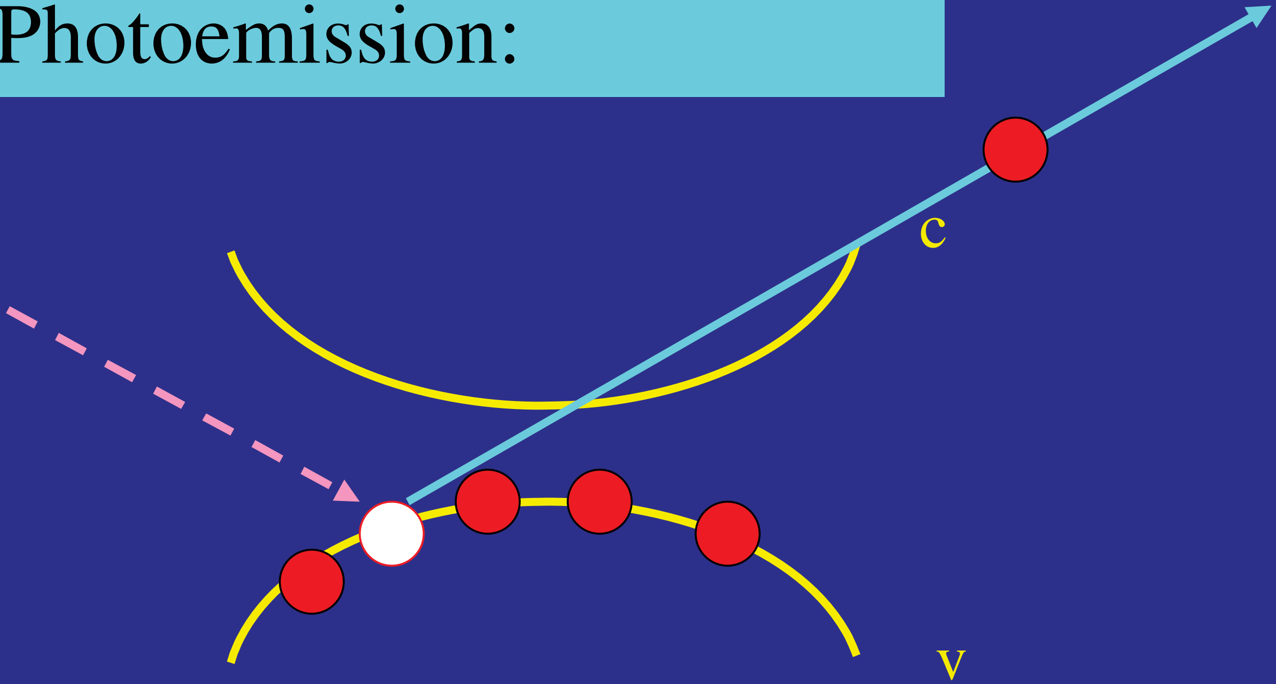
→ Insight from a simple model

→ An exponential solution for G

→ What's ongoing?

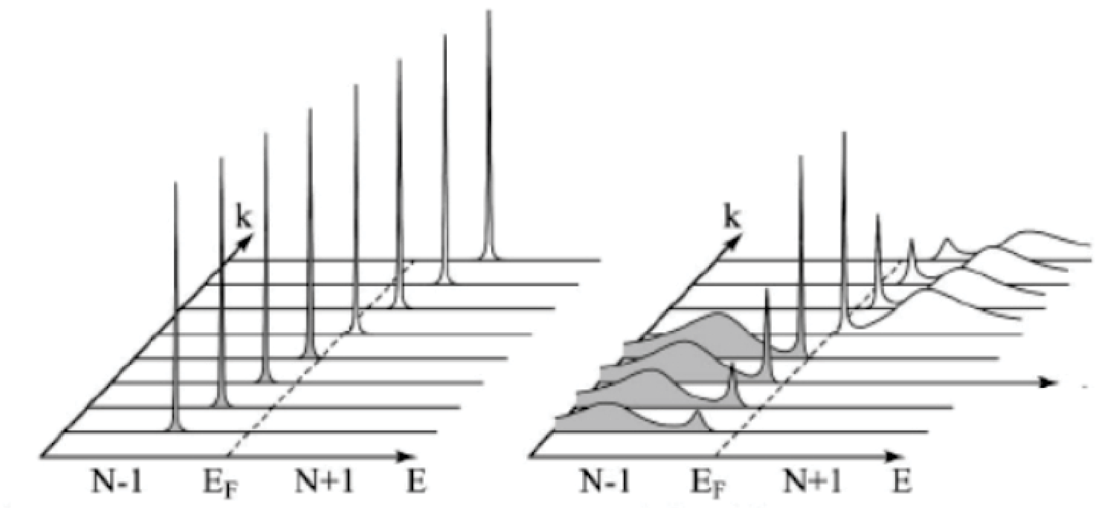
→ Outlook

Photoemission:



Hole - (N-1) (excited) electrons

$A(\omega) \sim \mathrm{Im}[\mathcal{G}(\omega)]$



From Damascelli et al., RMP 75, 473 (2003)

Coupling to other excitations!

Many-body perturbation theory

$$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 V_H \mathcal{G} + \mathcal{G}_0 \varphi \mathcal{G} + i\mathcal{G}_0 v_c \frac{\delta \mathcal{G}}{\delta \varphi}.$$

L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics*

$$\mathcal{G}(1,2) = -i \langle T[\psi(1)\psi^\dagger(2)] \rangle$$

$$1=(r_1,\sigma_1,t_1)$$

Many-body perturbation theory

$$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 V_H \mathcal{G} + \mathcal{G}_0 \varphi \mathcal{G} + i\mathcal{G}_0 v_c \frac{\delta \mathcal{G}}{\delta \varphi}.$$

L. P. Kadanoff and G. Baym, *Quantum Stat*

$\sim \mathcal{G}\mathcal{G} \rightarrow$ HF

Dyson equation: $\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 \Sigma \mathcal{G}$

$$\Sigma \sim i\, v_c \mathcal{G}$$

Many-body perturbation theory: GW

$$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 V_H \mathcal{G} + \mathcal{G}_0 \varphi \mathcal{G} + i\mathcal{G}_0 v_c \frac{\delta \mathcal{G}}{\delta \varphi}.$$

L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics*

1. Linearization $V_H[\varphi] = V_H^0 + v_c \chi \varphi$ .....

$$\begin{aligned}\mathcal{G}(t_1 t_2) &= \mathcal{G}_H(t_1 t_2) + \mathcal{G}_H(t_1 t_3)\bar{\varphi}(t_3)\mathcal{G}(t_3 t_2) \\ &+ i\mathcal{G}_H(t_1 t_3)\mathcal{W}(t_3 t_4)\frac{\delta \mathcal{G}(t_3 t_2)}{\delta \bar{\varphi}(t_4)},\end{aligned}$$

.......leads to screening: $\mathcal{W} = \varepsilon^{-1} v_c$

Many-body perturbation theory: GW

$$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 V_H \mathcal{G} + \mathcal{G}_0 \varphi \mathcal{G} + i\mathcal{G}_0 v_c \frac{\delta \mathcal{G}}{\delta \varphi}.$$

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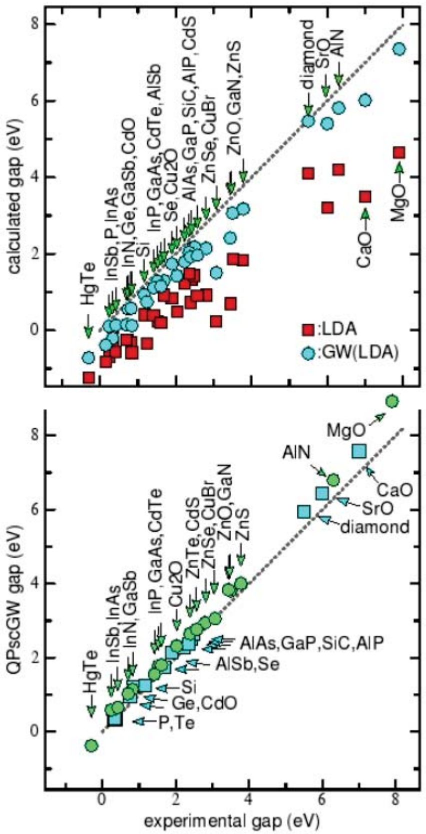
$\sim \mathcal{G}\mathcal{G}$

Lani et al., New J. Phys. 14, 013056 (2012)

$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 \Sigma \mathcal{G}$

$\rightarrow \Sigma \sim i\,\mathcal{W}\mathcal{G}$ "GW"

With optimized QP energies and wavefunctions

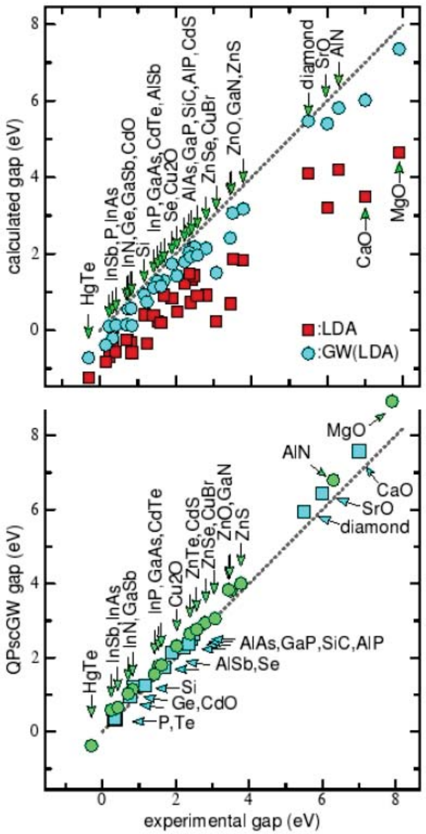


great bandstructure

Dyson equation →
no T=0 bandstructure problem.

van Schilfgaarde, Kotani, Faleev,
Phys. Rev. Lett. 96, 226402 (2006)

With optimized QP energies and wavefunctions

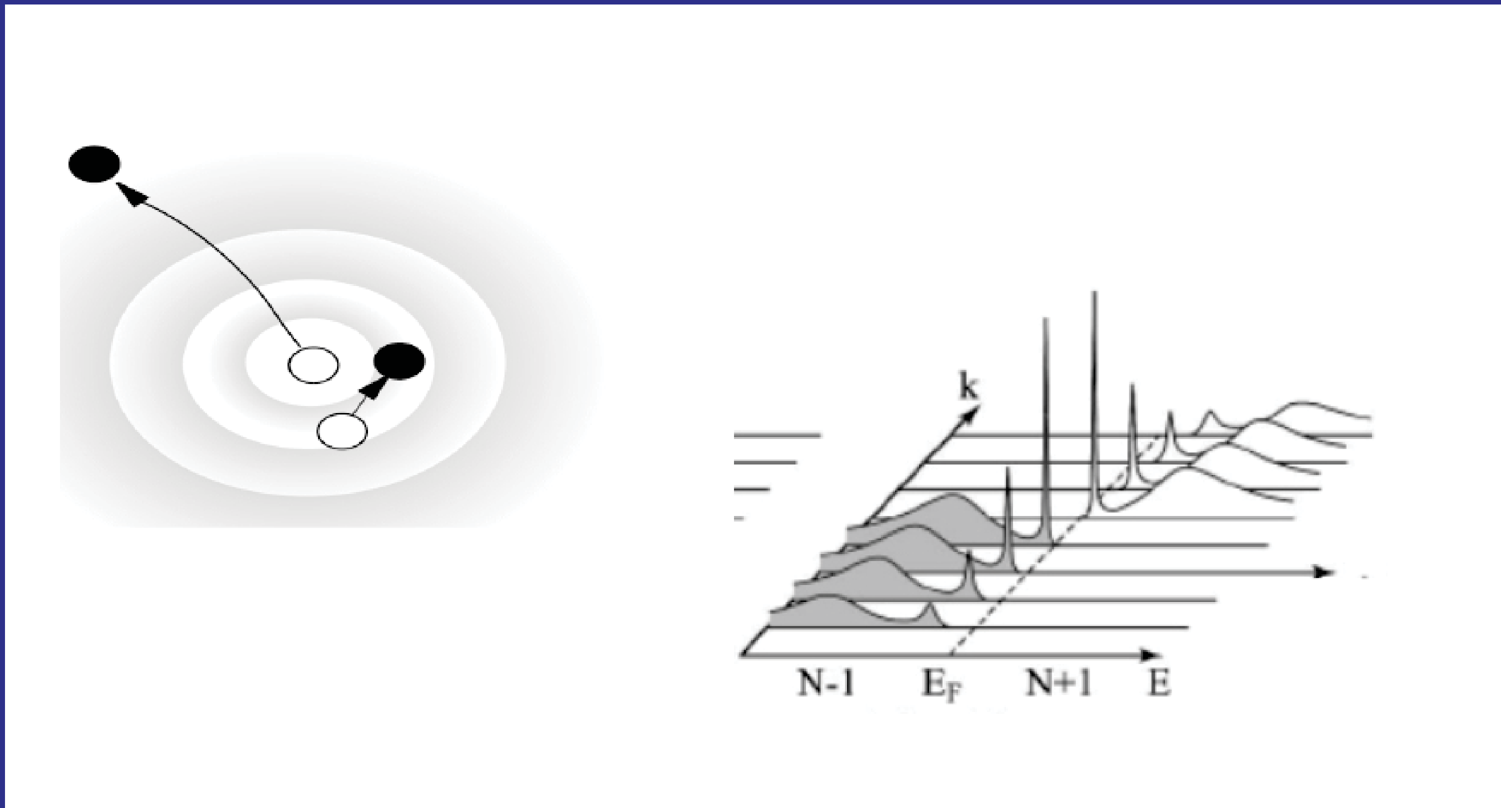


great bandstructure

What about the rest?

van Schilfgaarde, Kotani, Faleev,
Phys. Rev. Lett. 96, 226402 (2006)

GW: Electron-plasmon coupling

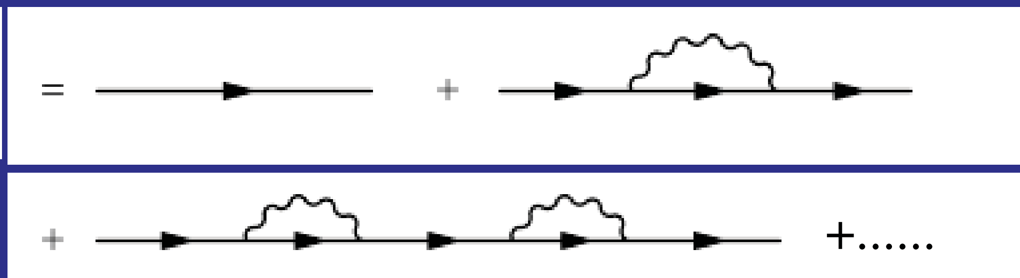


From Damascelli et al., RMP 75, 473 (2003)

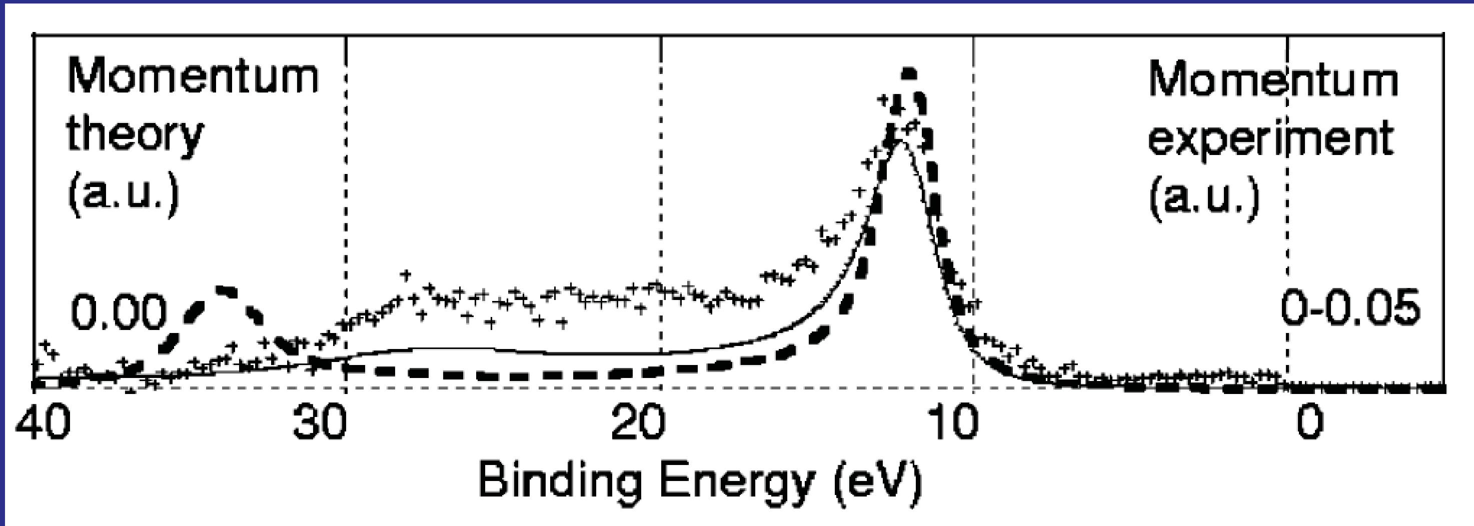
$\Sigma = iGW$

Hedin 1965

$\mathcal{G} =$



Valence satellites in "simple" silicon



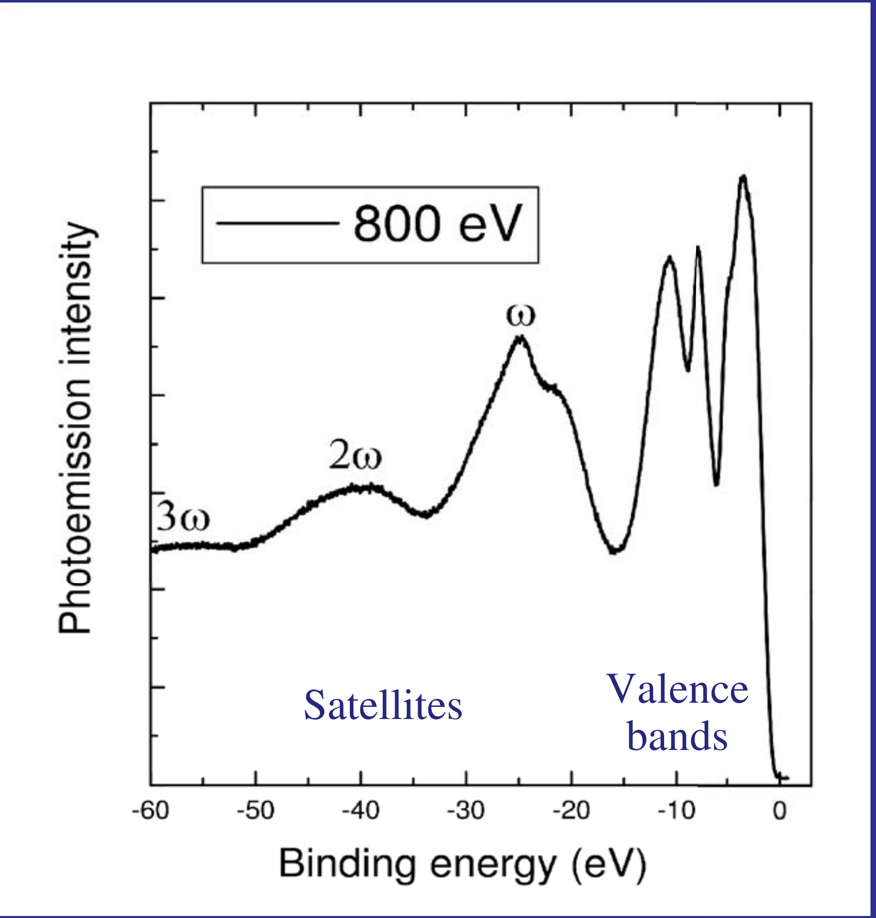
- - - Dashed: GW

Kheifets et al., PRB 68, 2003

GW: QP ok, satellite is completely wrong

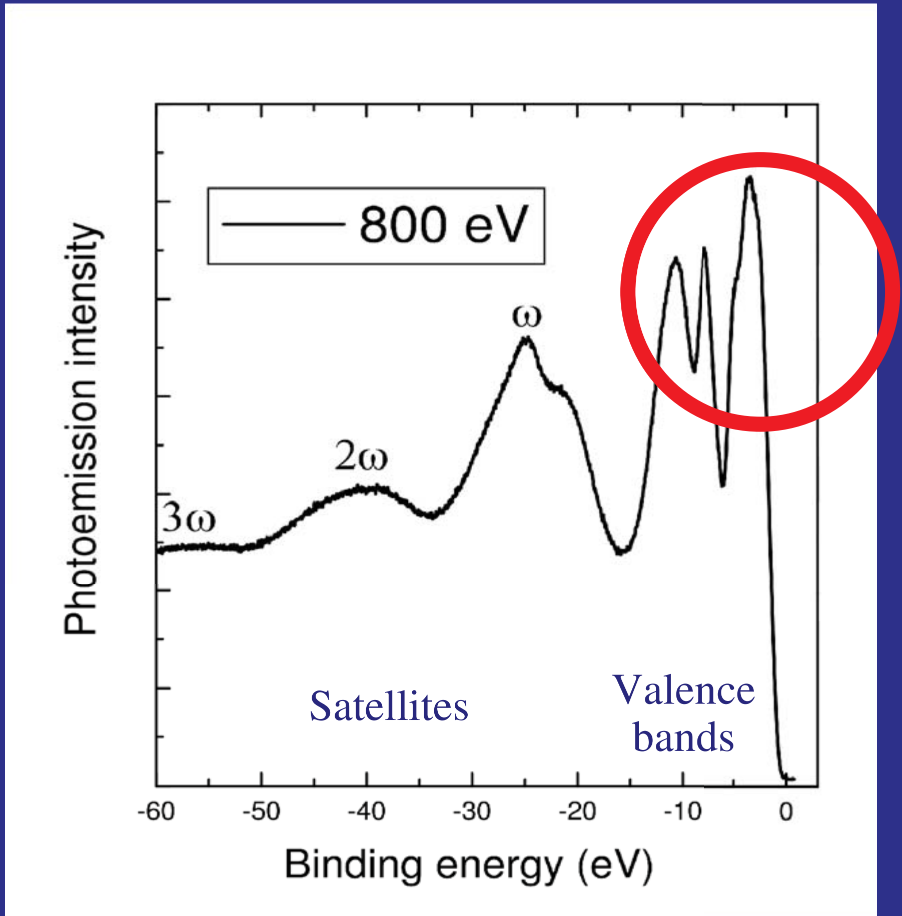
Experiment might also have limitations

XPS Spectrum of bulk Si



F. Sirotti and M. Silly, Synchrotron Soleil, France

XPS Spectrum of bulk Si



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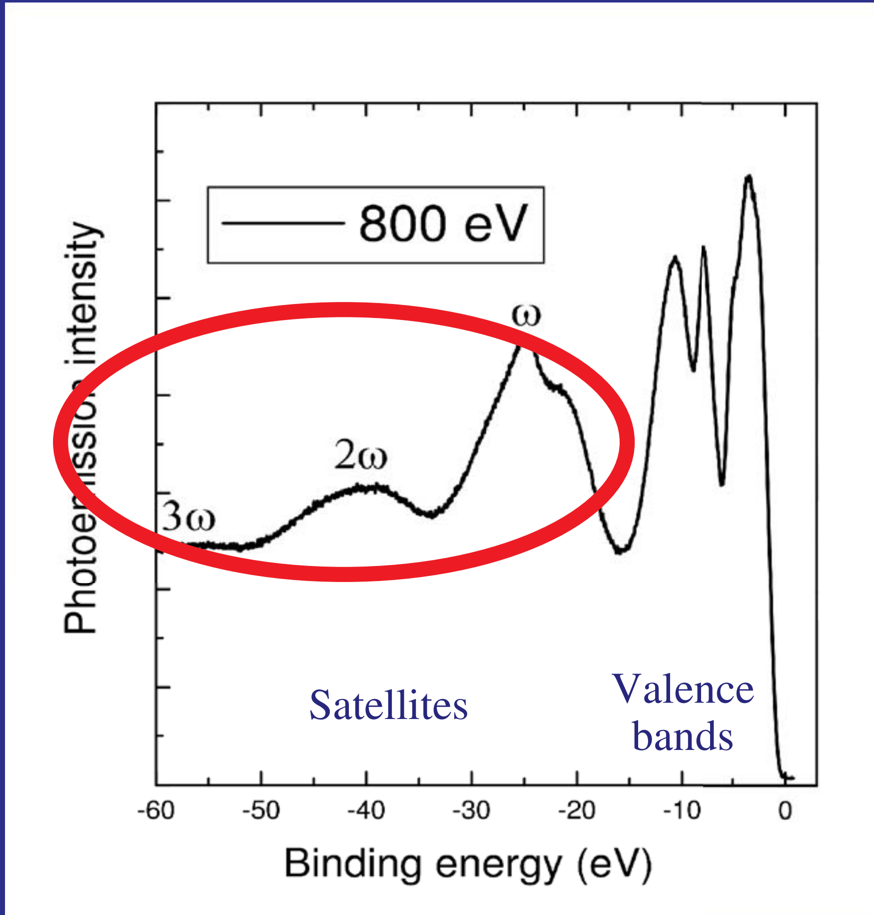
Quasiparticle bands

$$E_i = \varepsilon_i + \mathrm{Re}\Sigma(E_i)$$

XPS Spectrum of bulk Si



F. Sirotti and M. Silly, Synchrotron Soleil, France

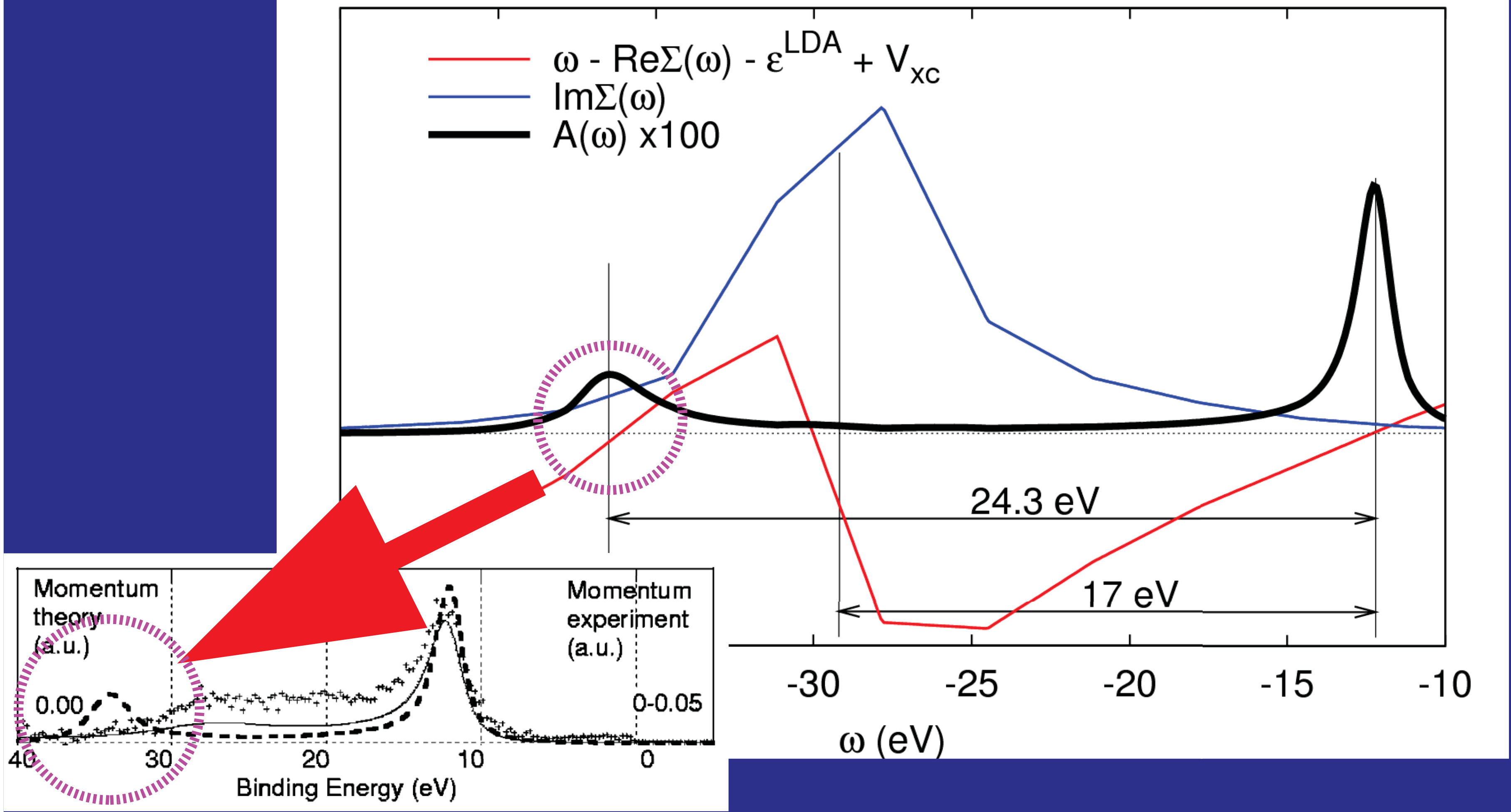
Plasmon satellites

$\mathrm{Im}\Sigma(\omega) \sim \mathrm{Im}W(\omega)$

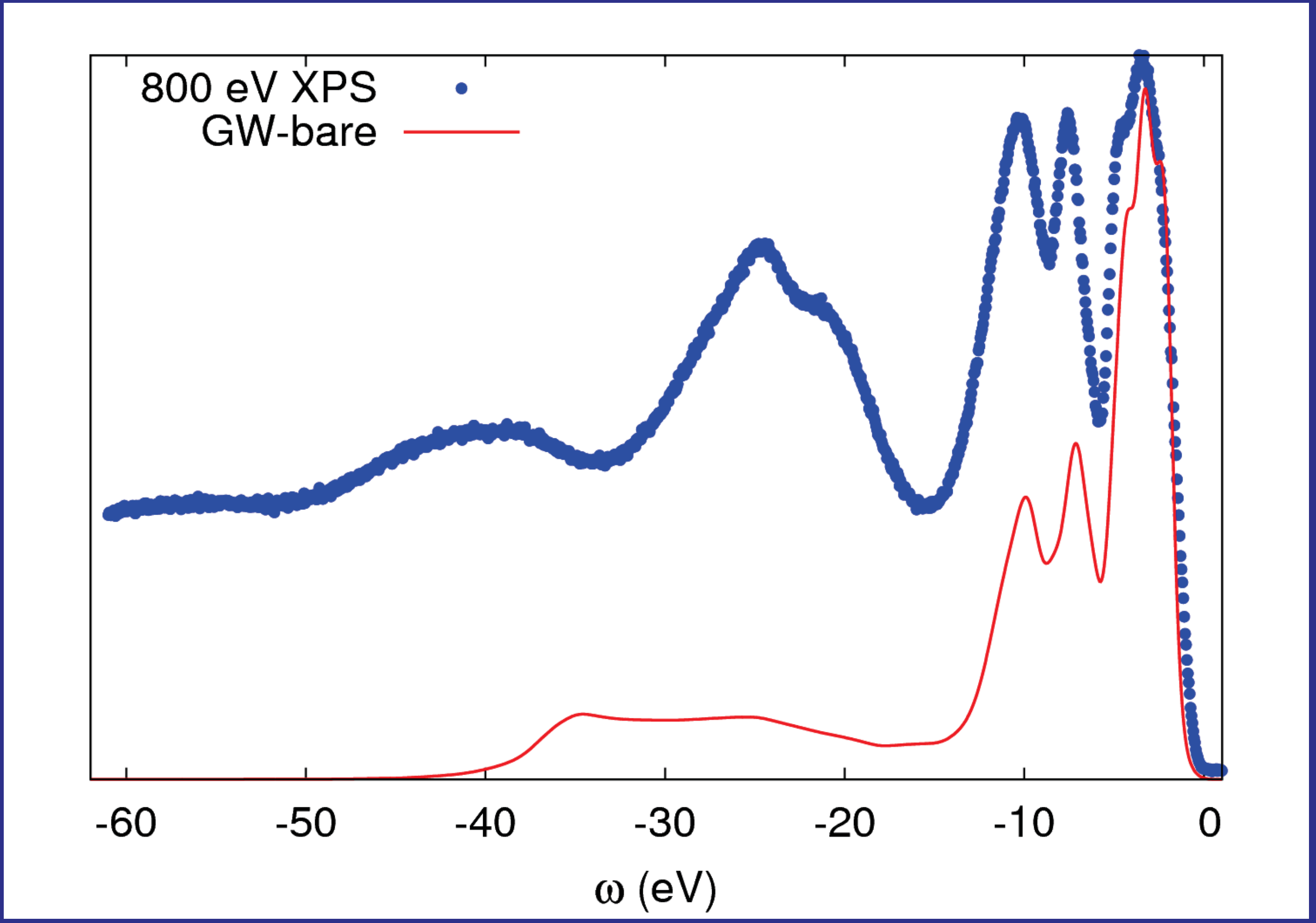
Peaked at 17 eV

Bottom valence: a plasmaron!

2 solutions for $E_i = \varepsilon_i + \mathrm{Re}\Sigma(E_i)$



Total G_0W_0 Spectral Function



G_0W_0 Spectral Function:

Plasmaron peaks stronger than plasmon peaks (Can mask plasmon contribution)

Artefact! Blomberg, Bergerse, Can. J. Phys. 50, 2286 (1972); Kus, Blomberg, Can. J. Phys. 51, 102 (1973)

No replicas within G_0W_0 (See e.g. for sodium Aryasetiawan et al., PRL 77, 1996)

G_0W_0 Spectral Function:

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GW pasmarons: suspicious!

An alternative strategy?

$$\mathcal{G} = \mathcal{G}_0 + \mathcal{G}_0 V_H \mathcal{G} + \mathcal{G}_0 \varphi \mathcal{G} + i\mathcal{G}_0 v_c \frac{\delta \mathcal{G}}{\delta \varphi}.$$

L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics*

1. Linearization $V_H = V_H^0 + v_c \chi \varphi$ .....

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Lani et al., New J. Phys. 14, 013056 (2012)

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Lani et al., New J. Phys. 14, 013056 (2012)

Solve differential equation!

Two problems:

* solve the set of differential equations

* pick the good solution!

→ Insight from a simple model

$$G = G_H + G_H \varphi \quad G \; + i\, G_H W \,\delta\; G/\delta\; \varphi$$

"1 point"

Lani et al., New J. Phys. 14, 013056 (2012)

Great for questions like: how to pick the good solution?

Ex.: Does a self-consistent GW calculation always converge

* to the same result, for any starting point?

* to the physical solution, if there is more than one?

$G = G_H + G_H \varphi \quad G \; - G_H W \, \delta \; G/\delta \; \varphi$ "exact"

$G = G_H \qquad\qquad - G_H (W \, G) G$ GW

Quadratic equation: 2 solutions, G_1 and G_2!

Which one will we find?

$$G = G_H \; / (1 + G_H W \, G)$$

Solution 1:

→ iterate $G^{n+1} = G_H \; / (1 + G_H W \, G^n)$

→ continuous fraction. For all starting points:

Converges to G_1 with $\mathbf{G_1[W \rightarrow 0] = G_H}$

Determined by G[W → 0] = G_H

Benchmarking

1-point Hedin's equations

$$y_u(x) = y_0 + y_0 x y_u(x) + y_0 \Sigma_u y_u(x)$$

$$\Sigma_u = -u y_u(x) \Gamma_u$$

$$\Gamma_u = 1 + \frac{d\Sigma_u}{dy_u} \Gamma_u y_u^2(x)$$

- *Iterative* self-consistent scheme ($\Sigma = -u y_0$):

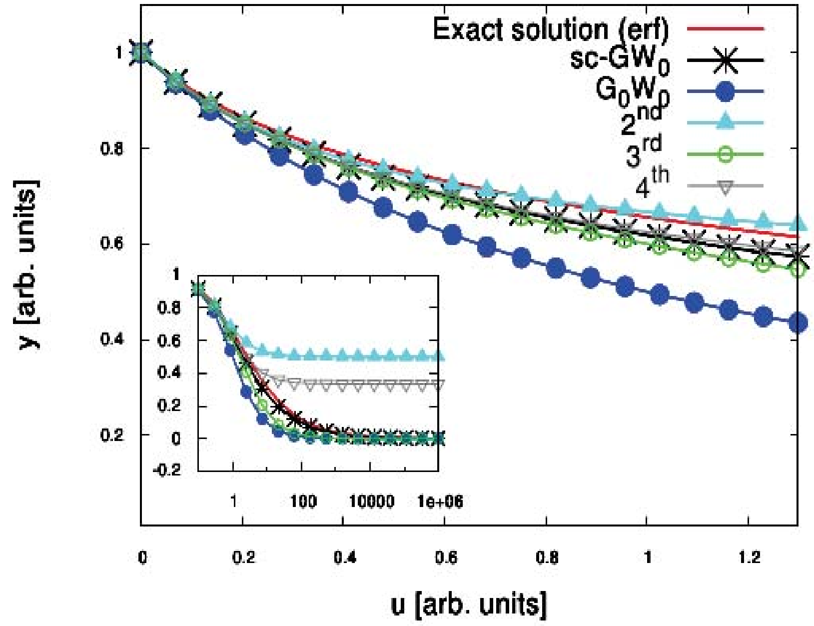
$$y^{(1)} = y_u^{G_0 W_0} = \frac{y_0}{1 + u y_0^2}$$

$$\cdots$$

$$y_u^{(4)} = y_0 \frac{1 + 3u y_0^2 + u^2 y_0^4}{1 + 4u y_0^2 + 3u^2 y_0^4}$$

- *Direct* self-consistent scheme ($\Sigma = -u y_u$):

$$y_u = \frac{\pm\sqrt{1 + 4u y_0^2} - 1}{2u y_0}$$



$G = G_H + G_H\varphi \quad G \; - G_H W \, \delta \; G/\delta \;\; \varphi$ "exact"

$G = G_H \qquad\qquad\qquad - G_H(W\,G)G$ GW

Quadratic equation: 2 solutions, G_1 and G_2!

Solution 1: iterate Dyson equation as usual
→ continuous fraction, well behaved $W \to 0$ limit.

Solution 2 ??? $G = 1/(WG) - 1(WG_H)$

→ iterate $G^{n+1} = 1/(WG^n) - 1(WG_H)$

→ continuous fraction. For all starting points:
Converges to G_2 with **$G_2[W \to 0]$ divergent!**

$G = G_H + G_H \varphi \quad G \; - G_H W \, \delta \; G/\delta \; \varphi$ "exact"

$G = G_H \qquad\qquad - G_H(W\, G)G$ GW

Quadratic equation: 2 solutions, G_1 and G_2!

Solution 1: iterate Dyson equation as usual
→ continuous fraction, well behaved W... mit.

Solution 2 ??? ...) – 1(WG$_H$)

→ iterate ... G) – 1(WG$_H$)

→ continuous fraction. For all starting points:
Converges to G_2 with **G_2[W → 0] divergent!**

The way you iterate!!!!

→ Self-consistency for GW_0 is a good thing

→ Expect that standard self-consistent GW_0 is ok

→ Expect more delicate situation beyond GW_0

→ 1-point model is marvellous playground:

* Benchmarks

* New approximations beyond GW

* insight : how to solve DE, how to pick solution.

More physics?

Solve linearized differential equation with times

G = G$_H$ + G$_H$φ G + i G$_H$W δ G/δ φ

$$\mathcal{G}(t_1t_2) = \mathcal{G}_H(t_1t_2) + \mathcal{G}_H(t_1t_3)\bar{\varphi}(t_3)\mathcal{G}(t_3t_2) + i\mathcal{G}_H(t_1t_3)\mathcal{W}(t_3t_4)\frac{\delta\mathcal{G}(t_3t_2)}{\delta\bar{\varphi}(t_4)},$$

Solve linearized differential equation with times

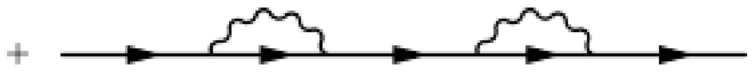
$G = G_H + G_H \varphi \;\; G + i\, G_H W \,\delta\; G/\delta\; \varphi$

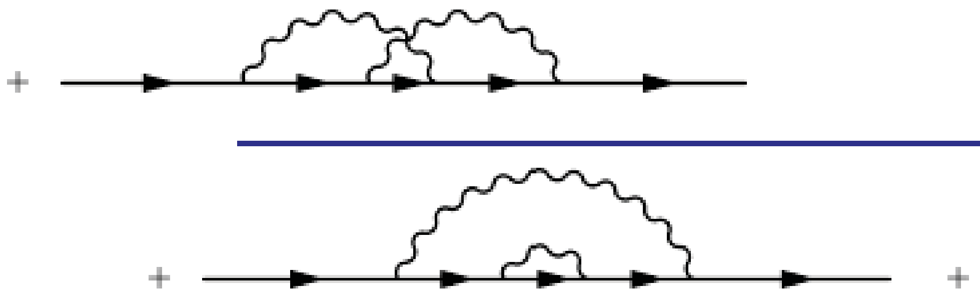
$$\mathcal{G}(t_1 t_2) = \mathcal{G}_H(t_1 t_2) + \mathcal{G}_H(t_1 t_3)\bar{\varphi}(t_3)\mathcal{G}(t_3 t_2) + i\mathcal{G}_H(t_1 t_3)\mathcal{W}(t_3 t_4)\frac{\delta \mathcal{G}(t_3 t_2)}{\delta \bar{\varphi}(t_4)},$$

cf U(ω) !!!

$$\mathcal{G}(t_1 t_2) = \mathcal{G}_\Delta(\tau) e^{i\Delta^{QP}\tau} e^{i\int_{t_1}^{t_2} dt' \left[\bar{\varphi}(t') - \int_{t'}^{t_2} dt'' \mathcal{W}(t' t'')\right]}$$

$\mathcal{G} =$

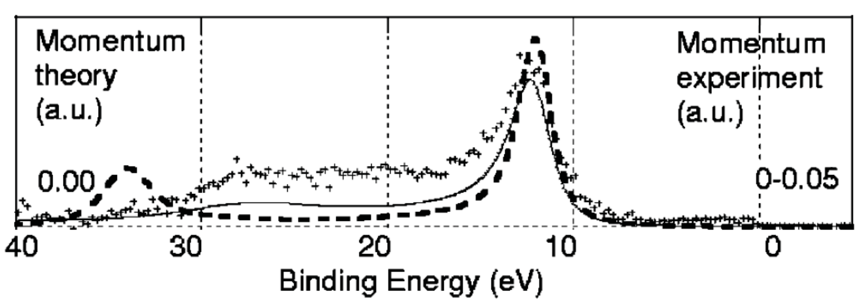




$$A(\omega) = \frac{\Gamma}{\pi} e^{-\frac{\lambda}{\omega_p^2}} \left[\frac{1}{(\omega - \varepsilon^{QP})^2 + \Gamma^2} + \right.$$
$$+ \frac{\lambda}{\omega_p^2} \frac{1}{(\omega - \varepsilon^{QP} + \omega_p)^2 + \Gamma^2} +$$
$$+ \frac{1}{2} \left(\frac{\lambda}{\omega_p^2} \right)^2 \frac{1}{(\omega - \varepsilon^{QP} + 2\omega_p)^2 + \Gamma^2} +$$
$$\left. + \frac{1}{6} \left(\frac{\lambda}{\omega_p^2} \right)^3 \frac{1}{(\omega - \varepsilon^{QP} + 3\omega_p)^2 + \Gamma^2} + \dots \right]$$

Exponential solution: ↔ cumulant expansion

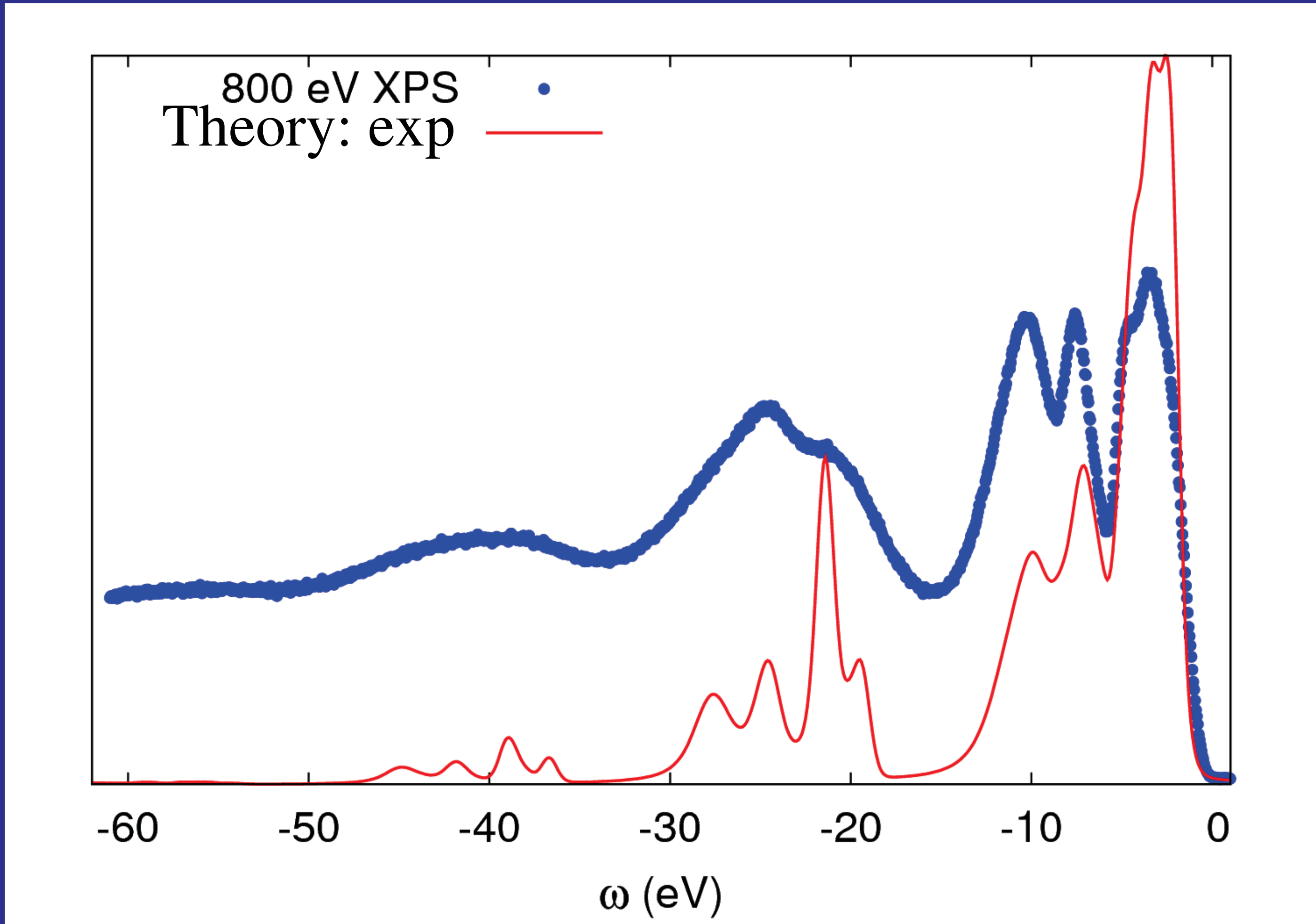
L. Hedin, Physica Scripta **21**, 477 (1980), ISSN 0031-8949.
L. Hedin, J. Phys.: Condens. Matter **11**, R489 (1999).
P. Nozieres and C. De Dominicis, Physical Review **178**, 1097 (1969), ISSN 0031-899X.
D. Langreth, Physical Review B **1**, 471 (1970).
Sodium: Aryasetiawan et al., PRL 77, 1996)



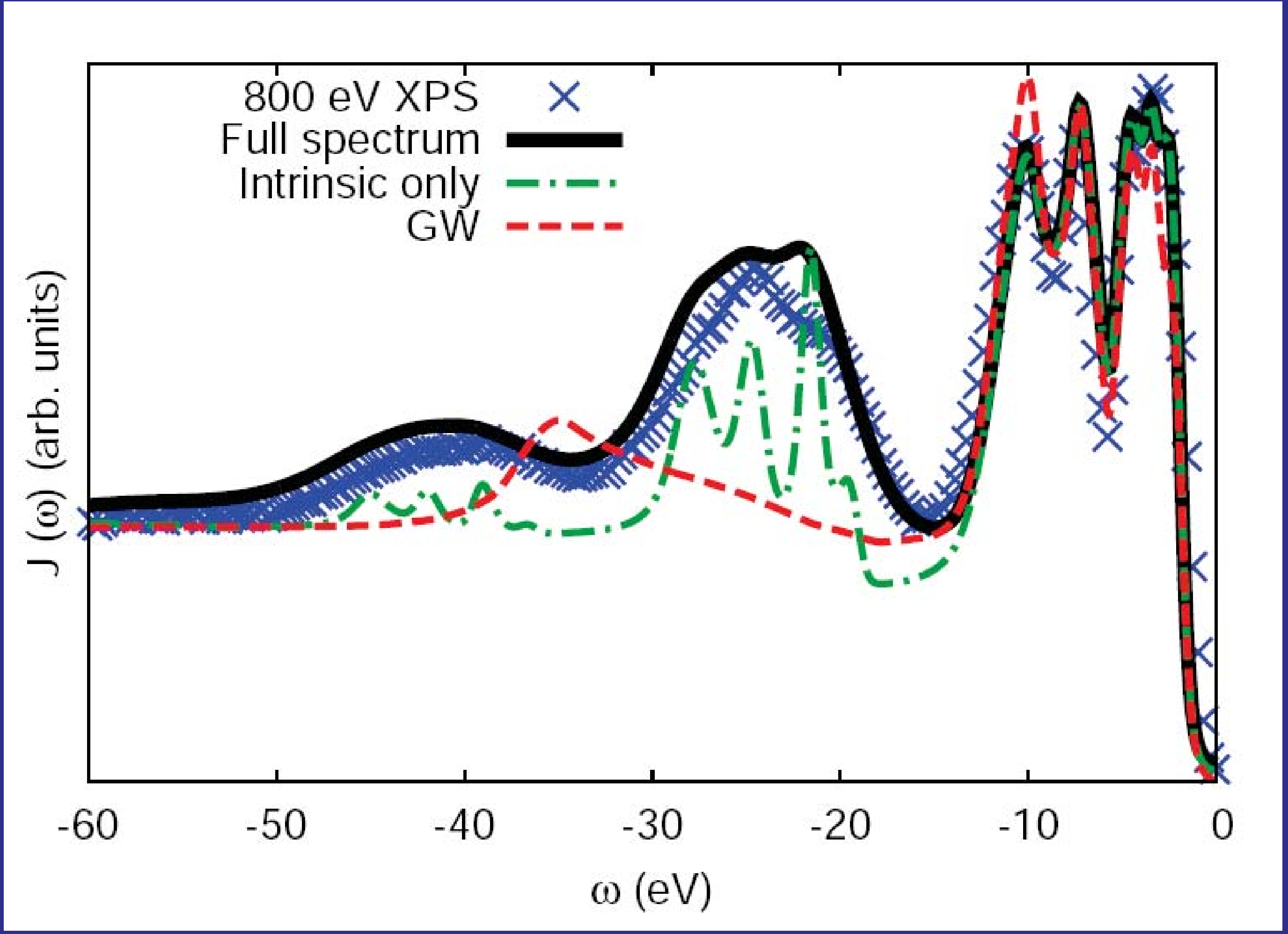
Kheifets et al., PRB 68, 2003
Silicon

Here: one possible simple approximation

Spectral Function from exponential:



Spectrum, exponential versus GW and experiment:

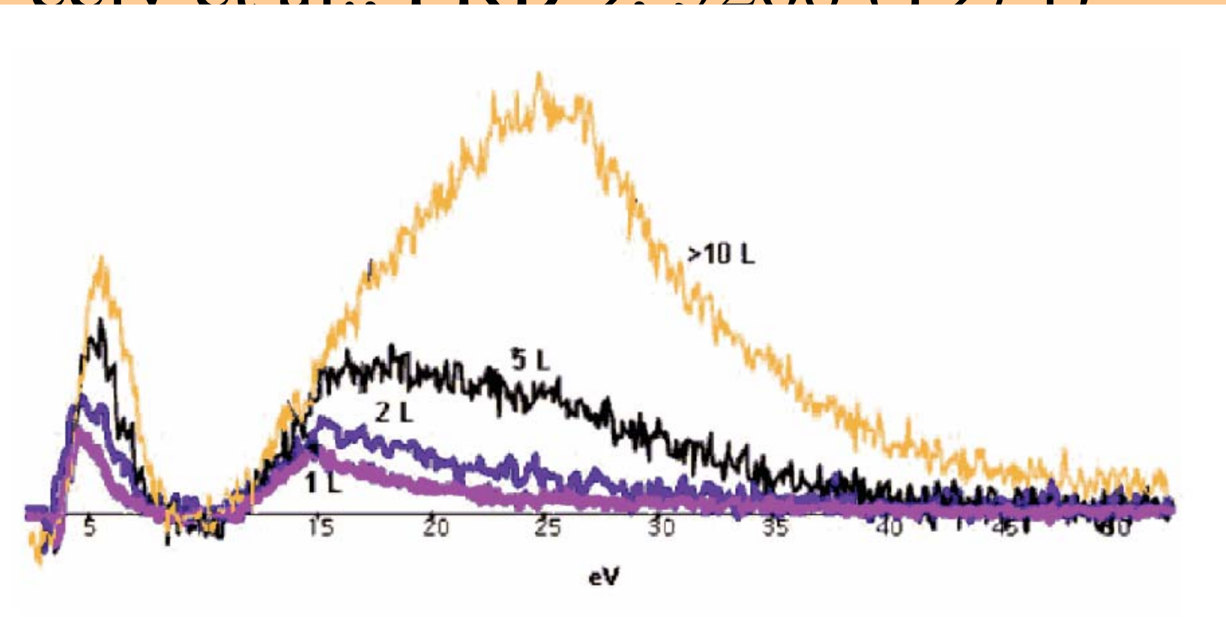


M. Guzzo et al., PRL 107, 166401 (2011)

Graphite

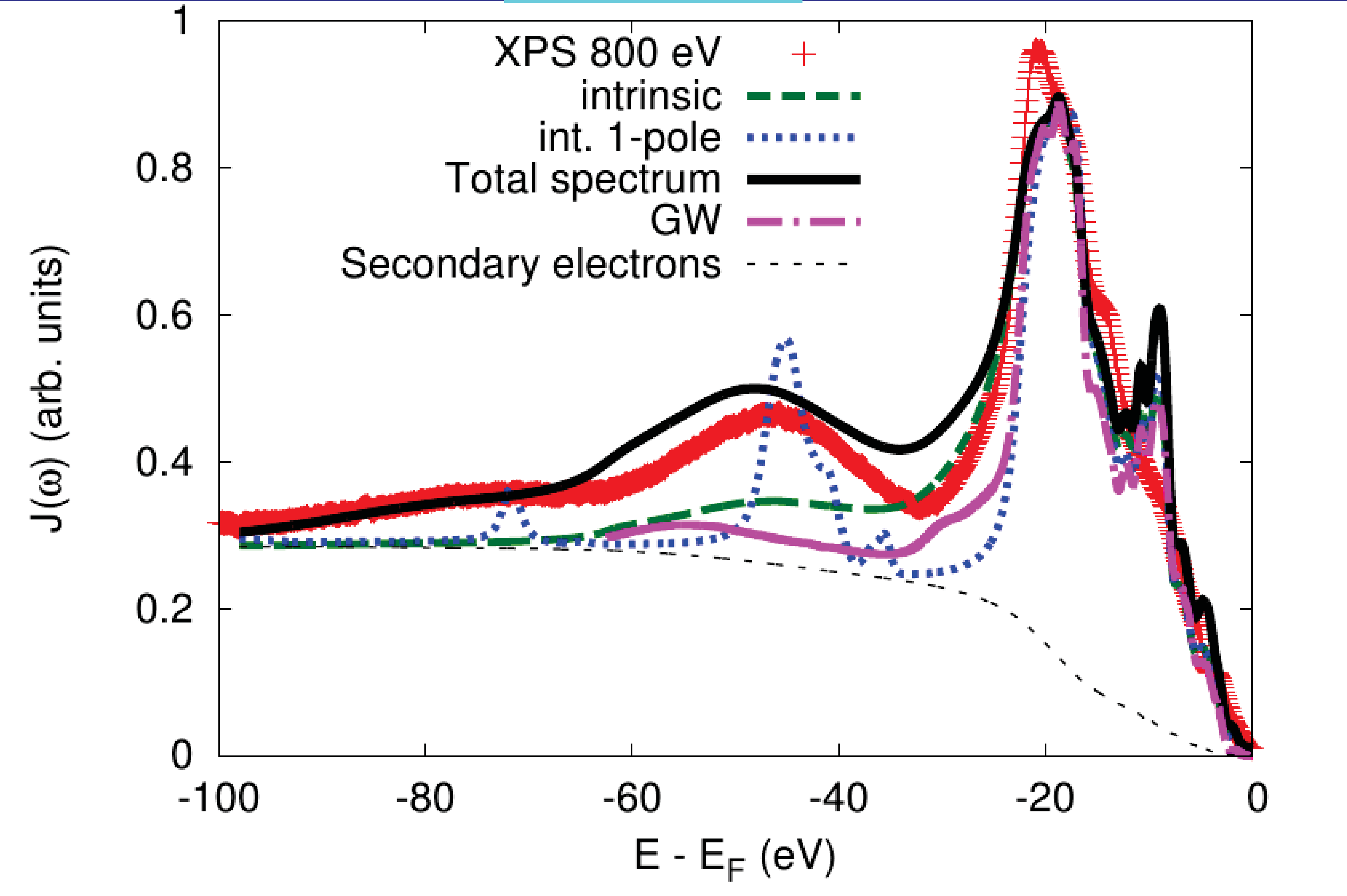
Satellites?

See e.g. Vos et al., PRB 63, 033108 (2001)
Sattler et al., PRB 63, 155204 (2001)
McFeely et al., PRB 9, 5268 (1974)

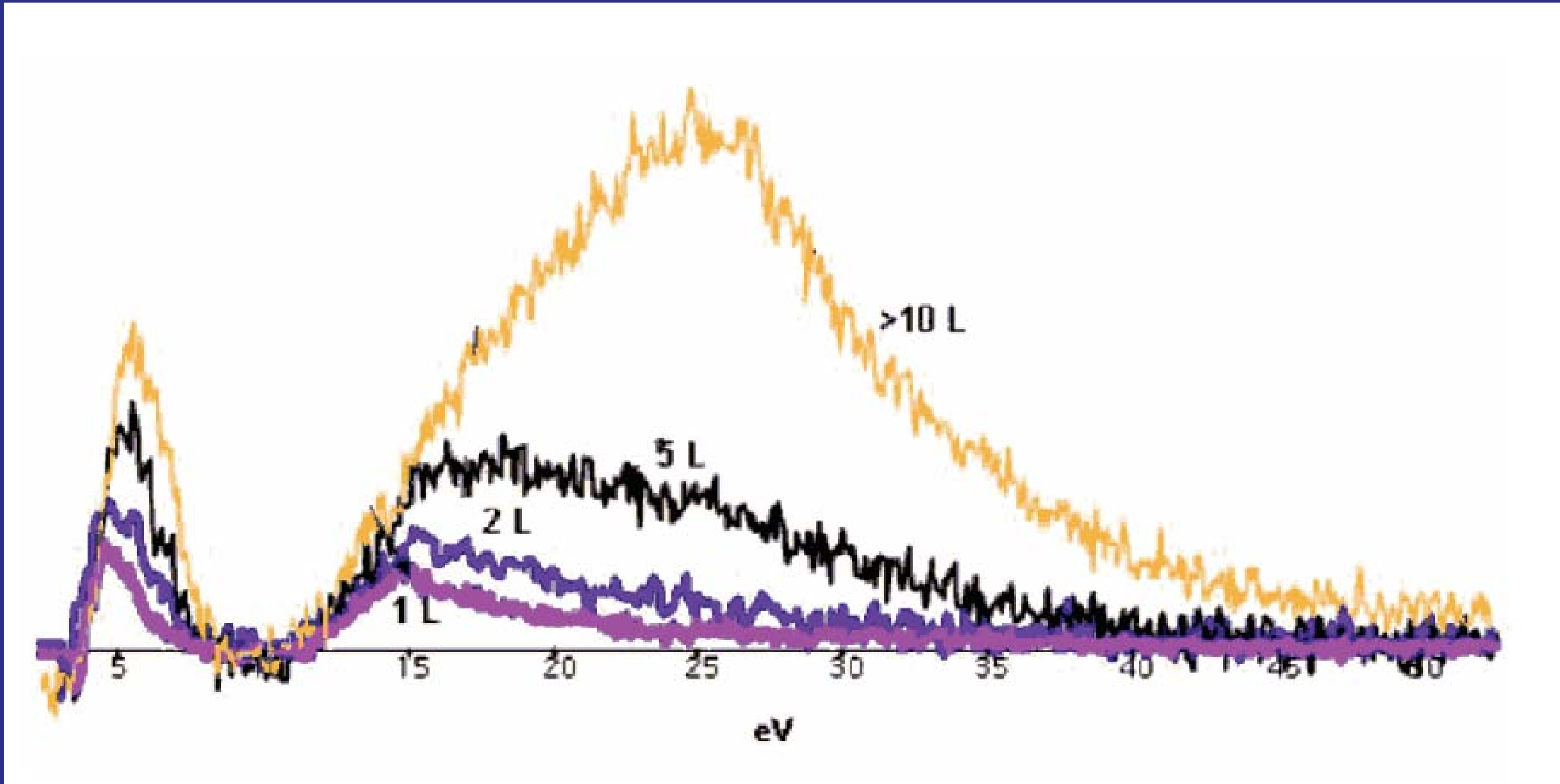


Exp: Eberlein et al., Phys. Rev. B 77, 233406 (2008)

Graphite

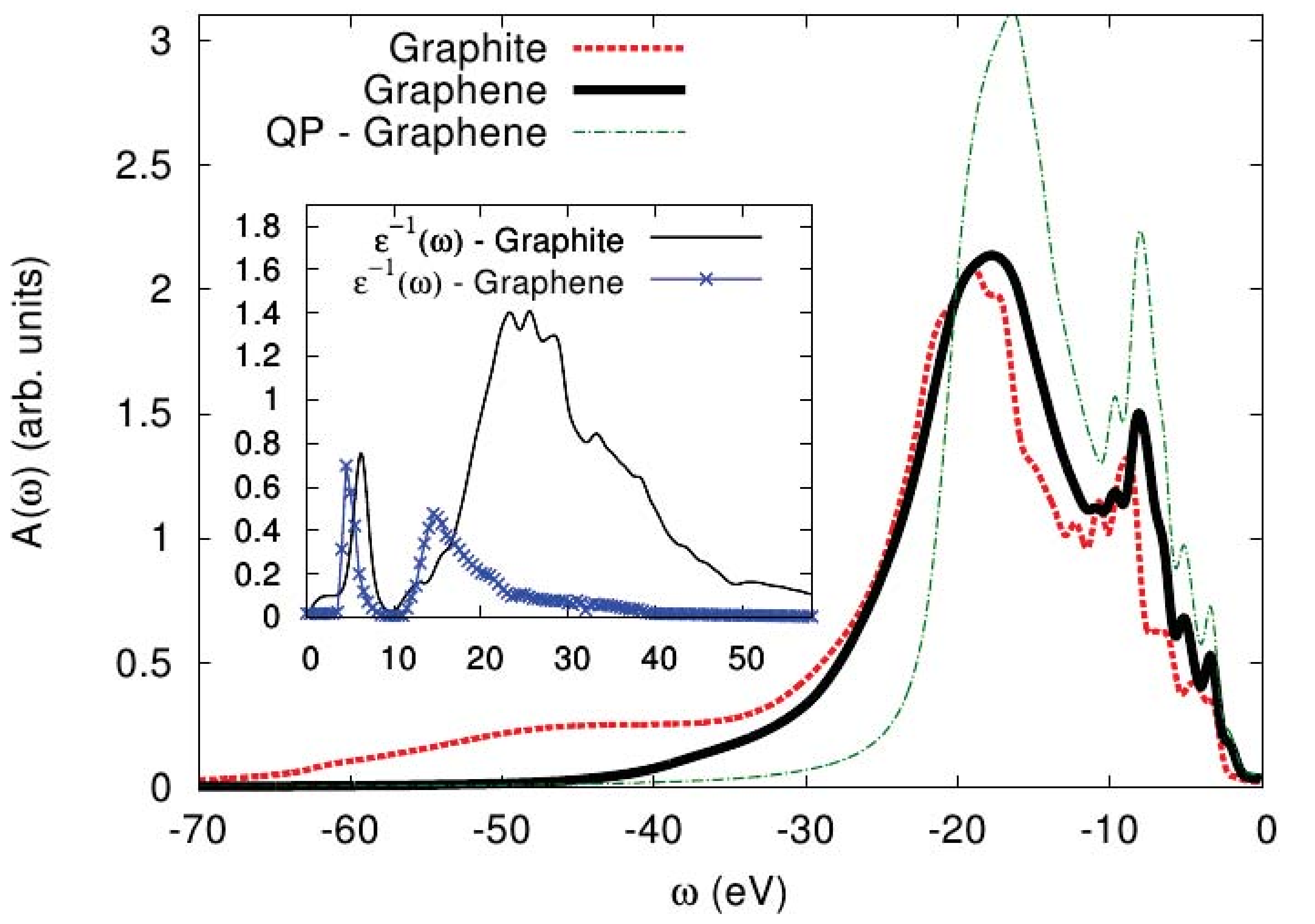


M. Guzzo et al., PhD thesis. Exp: SOLEIL TEMPO beamline.



Exp: Eberlein et al., Phys. Rev. B 77, 233406 (2008)

Graphite → Graphene



Exponential calculation, spectral function of graphite and graphene

Very good description of plasmon satellites

Can make predictions

There is more in life.........

→ What's ongoing ?

$$G(1,2;[\bar{\varphi}]) = G_H^0(1,2) + \int d3d5\, G_H^0(1,3)\bar{\varphi}(3)G(3,2;[\bar{\varphi}]) +$$

$$+ i \int d3d5\, G_H^0(1,3)W(3^+,5)\frac{\delta G(3,2;[\bar{\varphi}])}{\delta\bar{\varphi}(5)}$$

$$(1) \rightarrow (r_1, \sigma_1, t_1)$$

Solution of the (almost) full equations......

→ What's ongoing ?

→ We have: explicit $G(1,2,[\varphi\ ,W,\mathbf{q}])$

→ We have sum rules for q
* from the Differential eq.
* from the $W \to 0$ condition

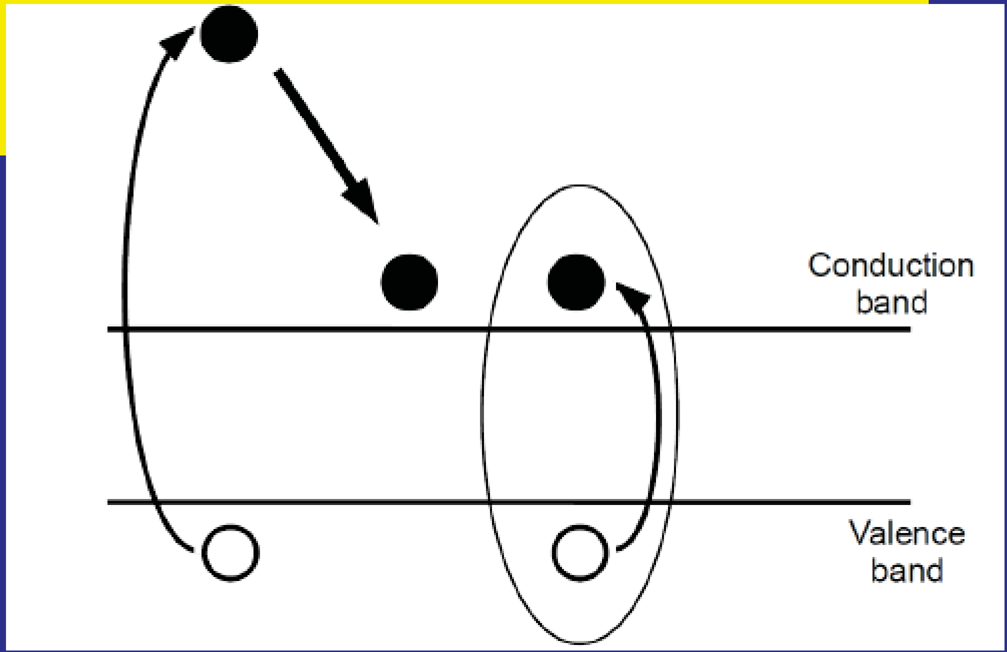
→ We have symmetry constraints
* from rules on fctl derivatives

Outlook

→ Full solution (ok ok, some approxs....)

→ coupling of other excitations

→ 2-particle G



Palaiseau Theoretical Spectroscopy Group & friends

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Francesco Sottile, Matteo Gatti, Christine Giorgetti, Lucia Reining

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U. Washington: John Rehr, Joshua Kas

Synchrotron SOLEIL: Fausto Sirotti, Matthieu Silly

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