GW approximation and linear response

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GW approximation and linear response

1. Introduction to the GW approximation

2. Formal derivation of the GW approximation

3. From GW to cumulant Green’s functions

4. Non-linear response cumulant approximation

5. Model results

6. Discussion: vertex corrections

7. Conclusions?

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$$O = \langle \hat{O} \rangle$$

$$O = \int ... \int dx_1 ... dx_N \, \Psi^*(x_1, ...x_N) \sum_{i,j,...} O(x_i, x_j, ...) \Psi(x_1, ...x_N)$$

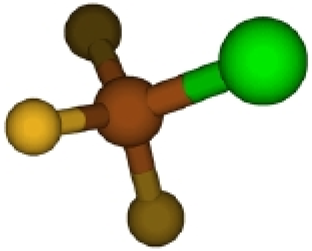
$$O = O[\Psi]$$

$$\Psi \longrightarrow O$$

$$G(x_1, x_1', t, t') = -i\langle N|T[\hat{\Psi}(x_1, t)\hat{\Psi}^\dagger(x_1', t')]|N\rangle$$

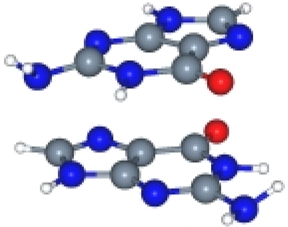
$\Psi(x_1, x_2, ..., x_N; t)$

CI, QMC



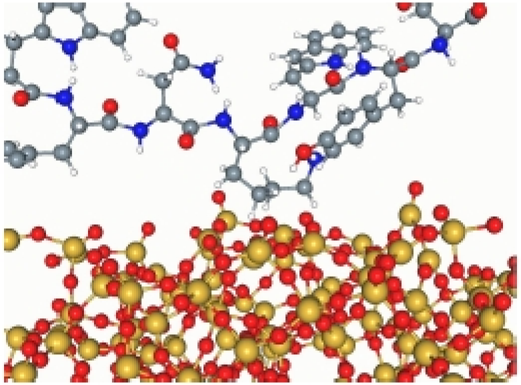
$G(x_1, x_1', t, t')$

Green's Functions



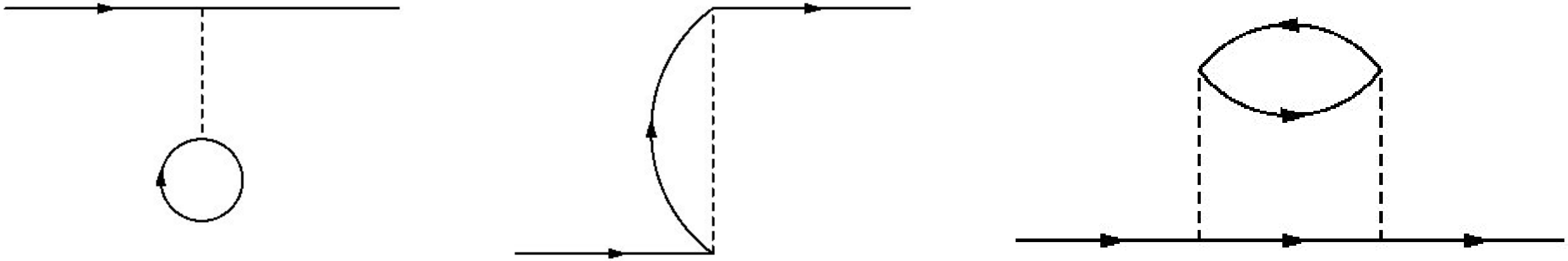
$n(\mathbf{r}; t)$

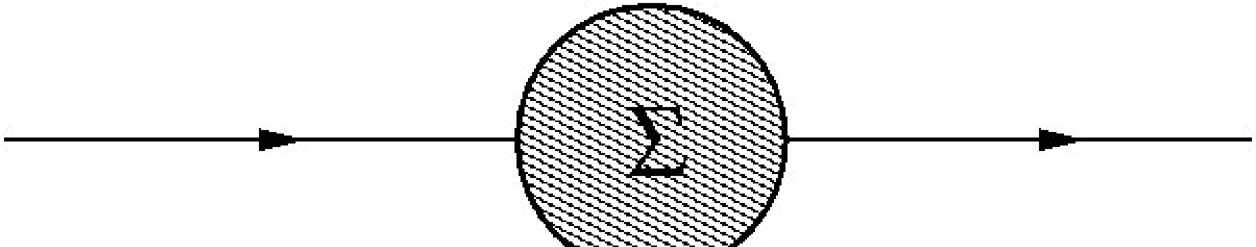
Density Functionals

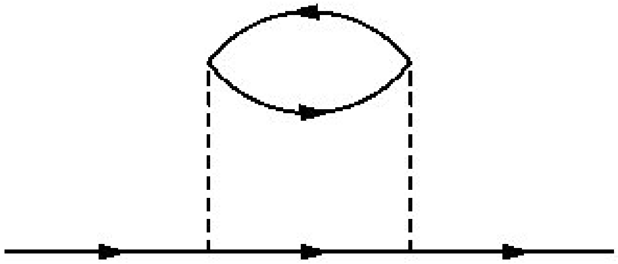


$$G(x_1, x_1', t, t') = -i\langle N|T[\hat{\Psi}(x_1, t)\hat{\Psi}^\dagger(x_1', t')]|N\rangle$$ **???**

Many things can happen to a particle that propagates in the middle of others......







+ ....

$\rightarrow \Sigma \sim i\,WG$ "GW"

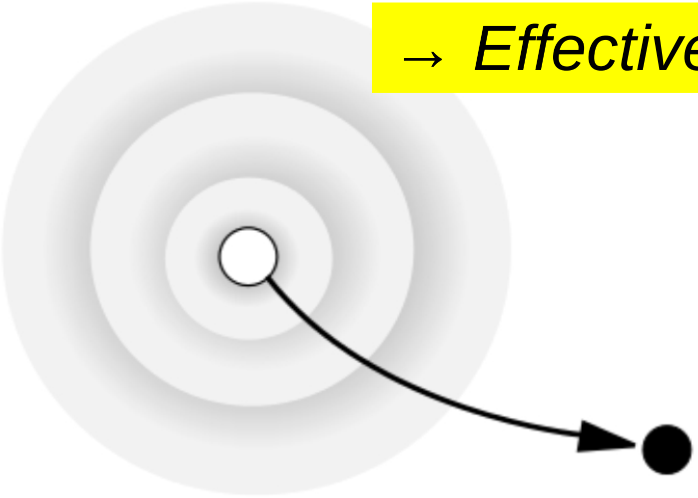
L. Hedin, Phys. Rev. 139:A796–823, 1965

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

GW

Hartree-Fock

→ *Effective* interaction brings in additional excitations



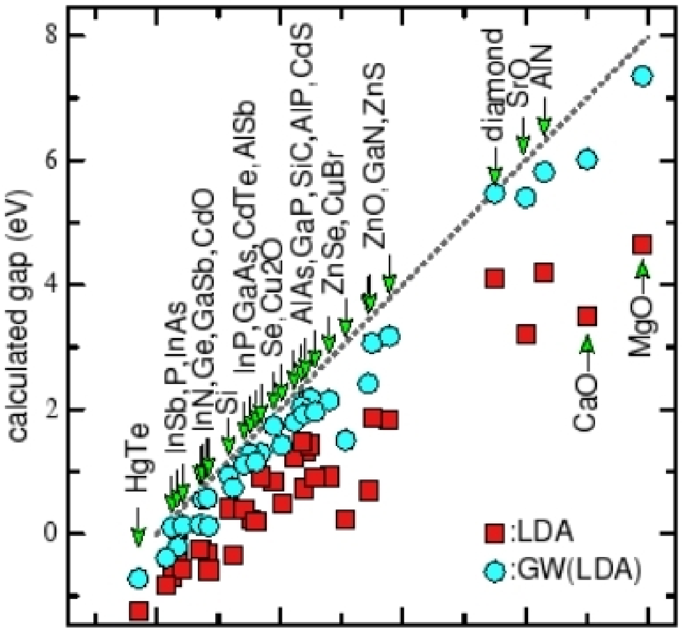
"physical!!!"

$$\rightarrow \Sigma \sim i\,WG \quad \text{"GW"}$$

L. Hedin (1965)

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

Usually good gaps and band structures in GW



Exp. Gap (eV)

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

Issues:

→ self-screening

(imagine to remove an electron: who contributes to W?)

W. Nelson, P. Bokes, P. Rinke, and R. W. Godby, Phys. Rev. A 75, 032505 (2007)

J. J. Fernandez, Phys. Rev. A 79, 052513 (2009)

P. Romaniello, S. Guyot, L. Reining, JCP 131, 154111 (2009)

Issues:

→ self-screening

→ strong correlation (degeneracy)

("the charge density" responds)

See, e.g., Romaniello, Bechstedt, Reining, PRB 85, 155131 (2012)

Issues:

→ self-screening

→ strong correlation (degeneracy)

→ linear response????

Issues:

→ self-screening

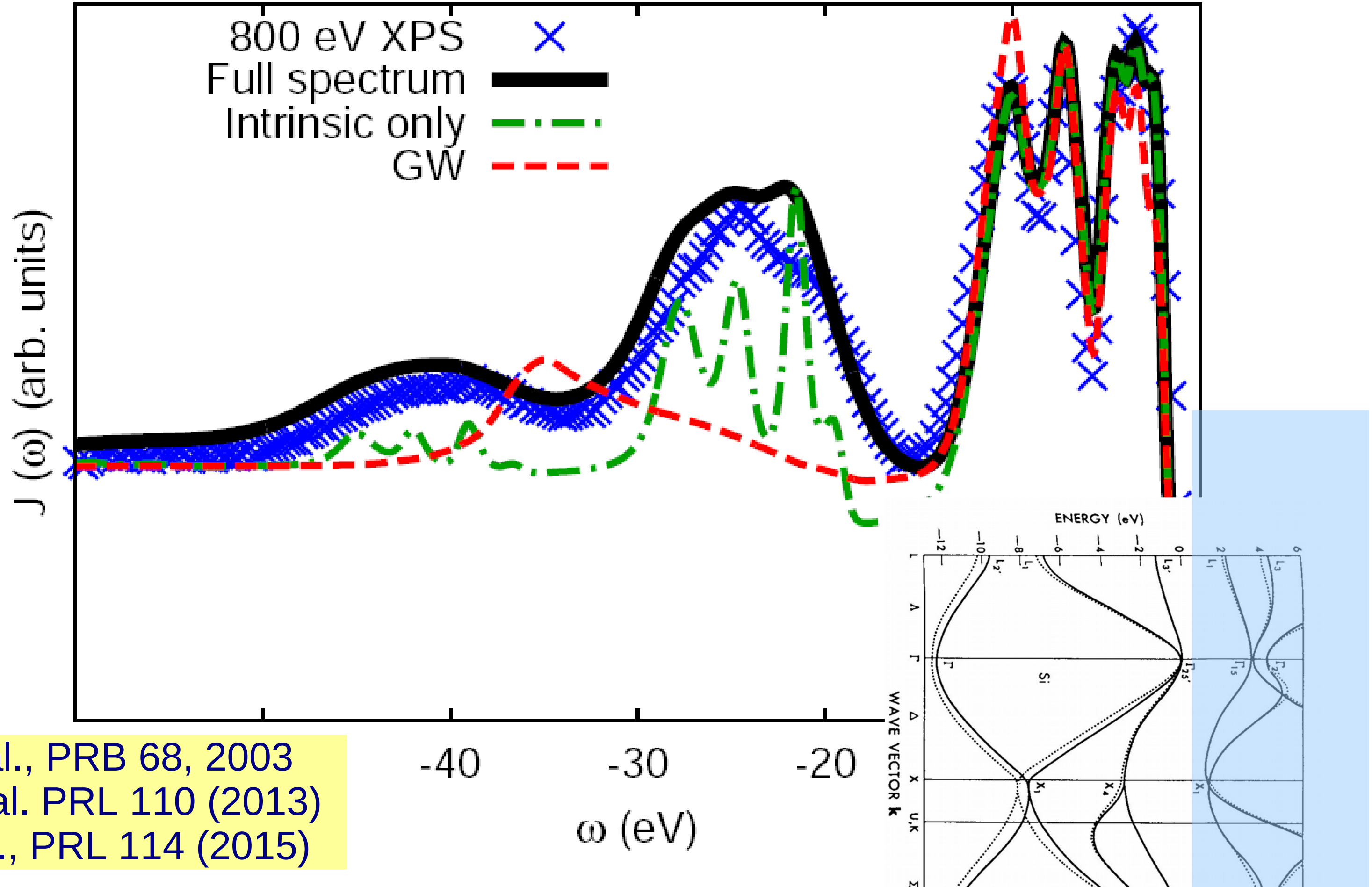
→ strong correlation (degeneracy)

→ linear response????

→ satellites

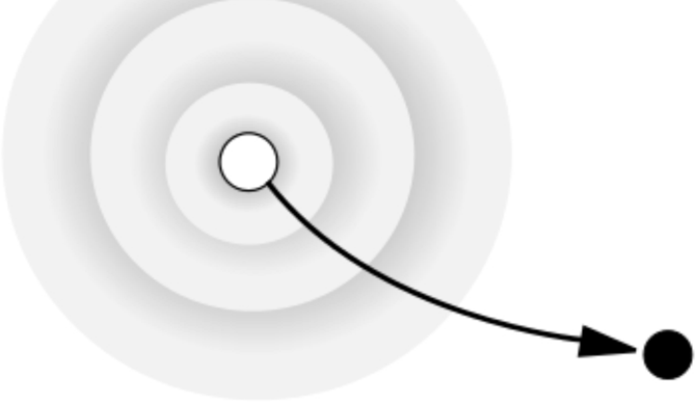
→ The one-body spectral function of silicon

M. Guzzo et al., PRL 107, 166401 (2011) in collab. with J. Kas and J. Rehr, M. Silly and F. Sirotti



Kheifets et al., PRB 68, 2003
Lischner et al. PRL 110 (2013)
Caruso et al., PRL 114 (2015)

→ *Effective* interaction brings in additional excitations



"physical!!!"

$\rightarrow \Sigma \sim i\, W G$ "GW"

L. Hedin (1965)

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

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Equation of motion of the one-body Green's funtion

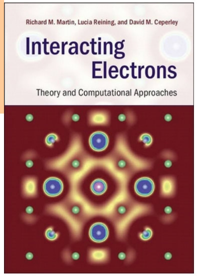
$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})\left\{[u(\bar{3}) + v_{Hu}(\bar{3})]G_u(\bar{3},2) + iv_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}\right\}$$

$$1 \equiv (\mathrm{r}_1, \sigma_1, t_1) \qquad f(\bar{1})g(\bar{1}) \equiv \int d1\, f(1)g(1) \qquad G_u \equiv G[u]$$

$$v_H(3) = -iv_c(3-\bar{5})G(\bar{5},\bar{5}^+)$$

J. Schwinger, PNAS. 37: 452 (1951)
L. P. Kadanoff and G. Baym, Quantum Statistical Mechanics, W. A. Benjamin, New York, 1964

R.M. Martin, L. Reining, D.M. Ceperley, “Interacting Electrons:Theory and Computational Approaches”, Cambridge (2016)



Giovanna Lani, Pina Romaniello, Arjan Berger, Matteo Guzzo, Adrian Stan, Lorenzo Sponza, Christine Giorgetti, Matteo Gatti, Walter Tarantino, Bernardo Mendoza, J. Sky Zhou, Marilena Tzavala, Stefano Di Sabatino, Pierluigi Cudazzo, John Rehr, Joshua Kas

Note: for DFT and RDMFT, see R. Fukuda et al., Progress of Theoretical Physics 92, 833 (1994)

Common approximations

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})\left\{[u(\bar{3}) + v_{Hu}(\bar{3})]G_u(\bar{3},2) + iv_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}\right\}$$

$$\frac{\delta G(3,2)}{\delta u(4)} = -G(3,\bar{5})\frac{\delta G^{-1}(\bar{5},\bar{6})}{\delta u(4)}G(\bar{6},2) \approx G(3,4)G(4,2)$$

$$G_u(1,2) \approx G^0(1,2) + G^0(1,\bar{3})\left\{[u(\bar{3}) + v_{Hu}(\bar{3})]G_u(\bar{3},2) + \underbrace{iv_c(\bar{3},\bar{4})G(\bar{3},\bar{4})}G(\bar{4},2)\right\}$$

Fock self-energy ($u \rightarrow 0$)

Common approximations

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})\left\{[u(\bar{3}) + v_{Hu}(\bar{3})]G_u(\bar{3},2) + iv_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}\right\}$$

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$$u_{\mathrm{cl}}(1) = u(1) + v_{Hu}(1)$$

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}$$

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u_{\mathrm{cl}}(\bar{5})}\frac{\delta u_{\mathrm{cl}}(\bar{5})}{\delta u(\bar{4}^+)}$$

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})W_u(\bar{3},\bar{5})\frac{\delta G_u(\bar{3},2)}{\delta u_{\mathrm{cl}}(\bar{5}^+)} \qquad G_u(\bar{3},\bar{5})G_u(\bar{5}^+,2)$$

GW approx. ($u \rightarrow 0$)

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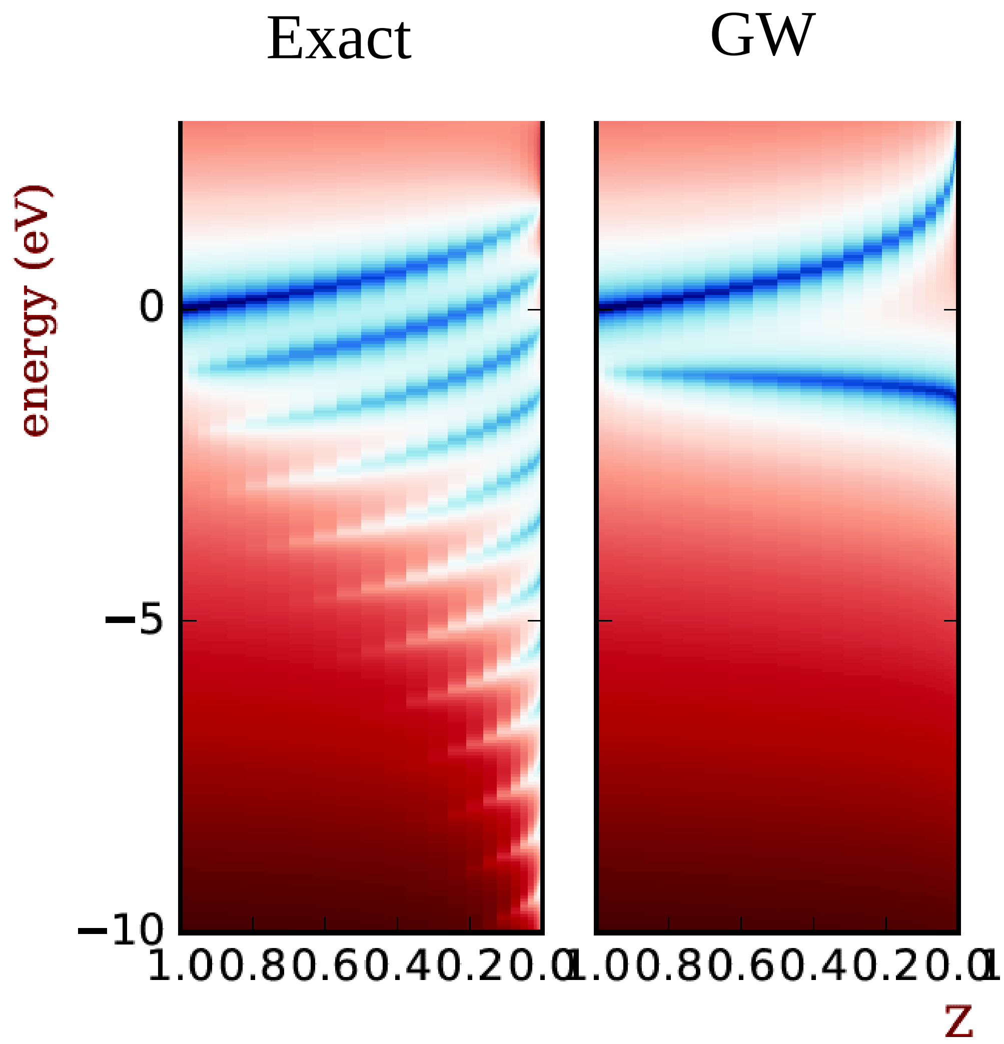
→ Electron-boson coupling

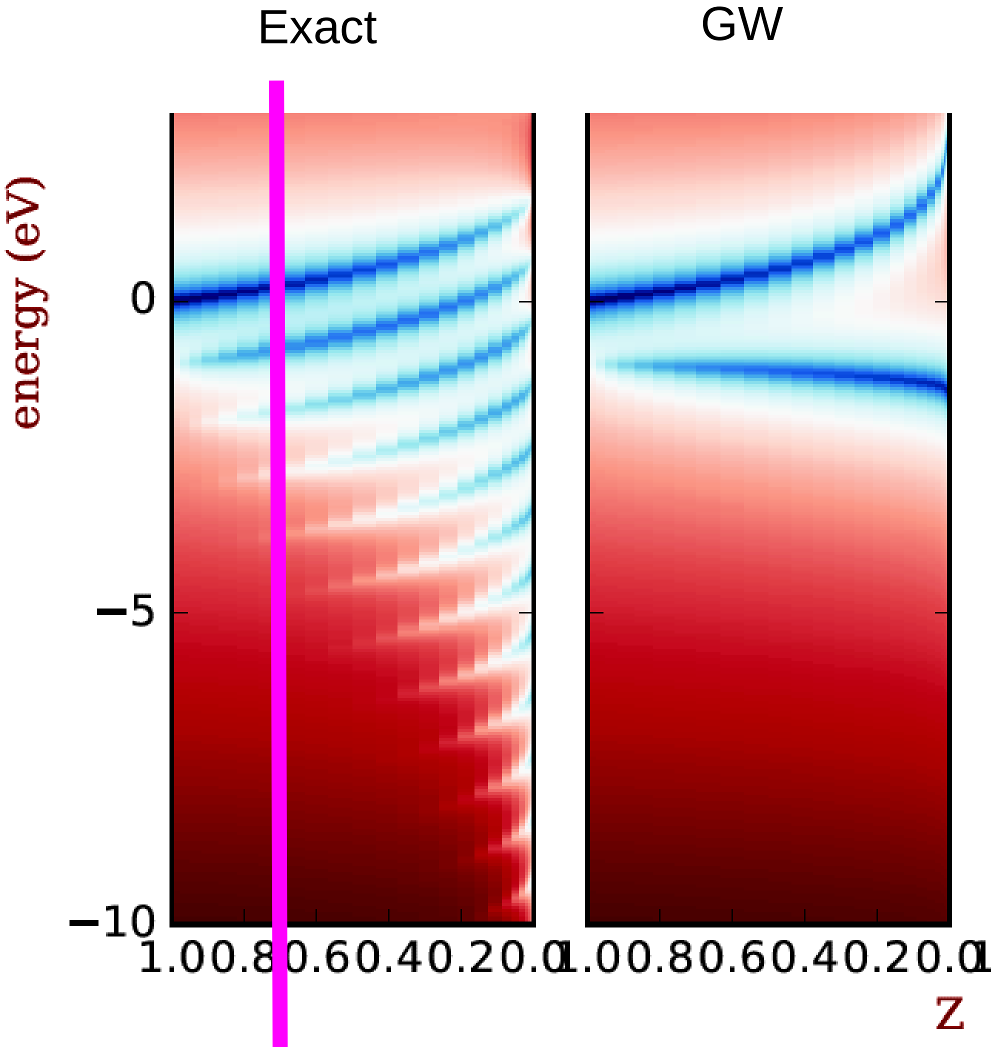
$$H = \epsilon_0 c^\dagger c + c c^\dagger g(a + a^\dagger) + \omega_0 a^\dagger a$$

$$A^h(\omega) = \sum_{n=0}^{\infty} \frac{\beta^n e^{-\beta}}{n!} \delta(\omega - \epsilon_0 - \beta\omega_0 - n\omega_0)$$

$$\beta = \frac{g^2}{\omega_0^2}$$



Exact
GW
energy (eV)
0
−5
−10
1.0
0.8
0.6
0.4
0.2
0.0
1.0
0.8
0.6
0.4
0.2
0.0
1
z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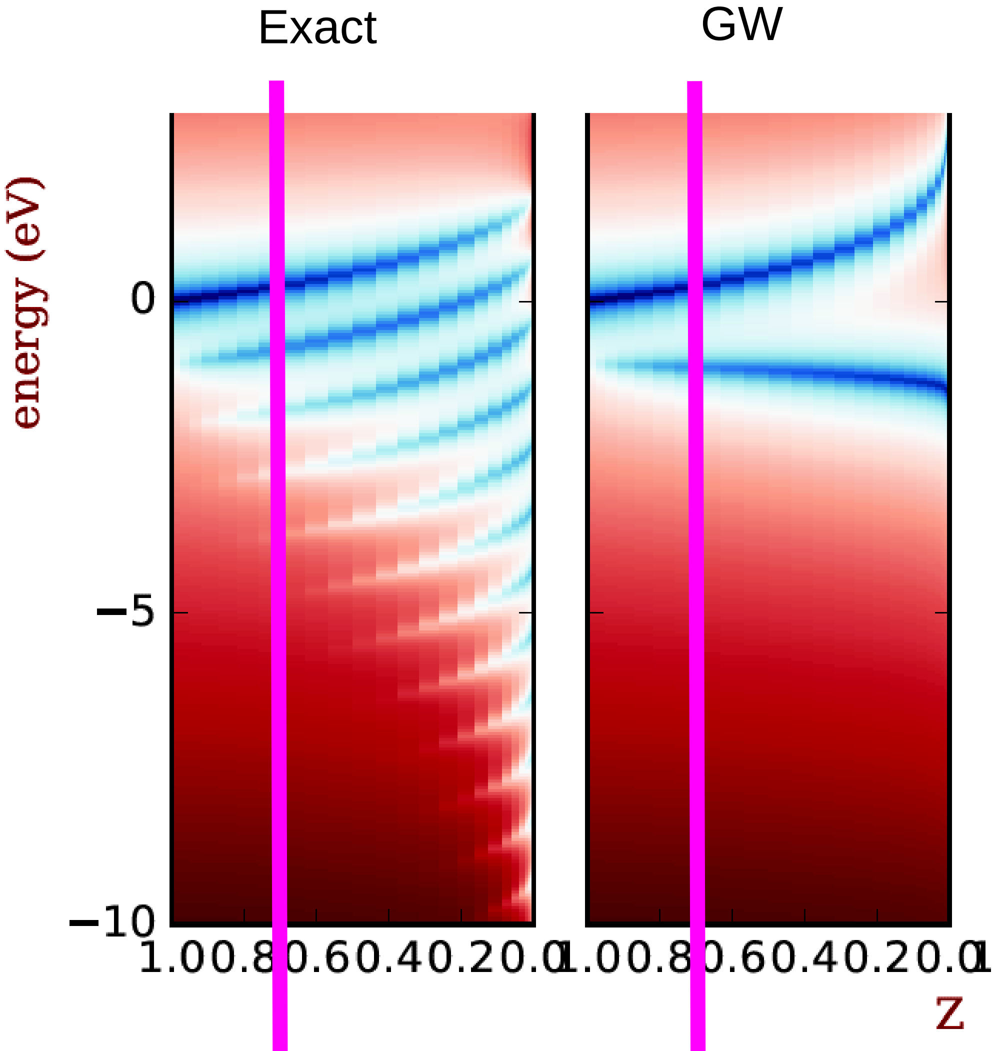
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0.0
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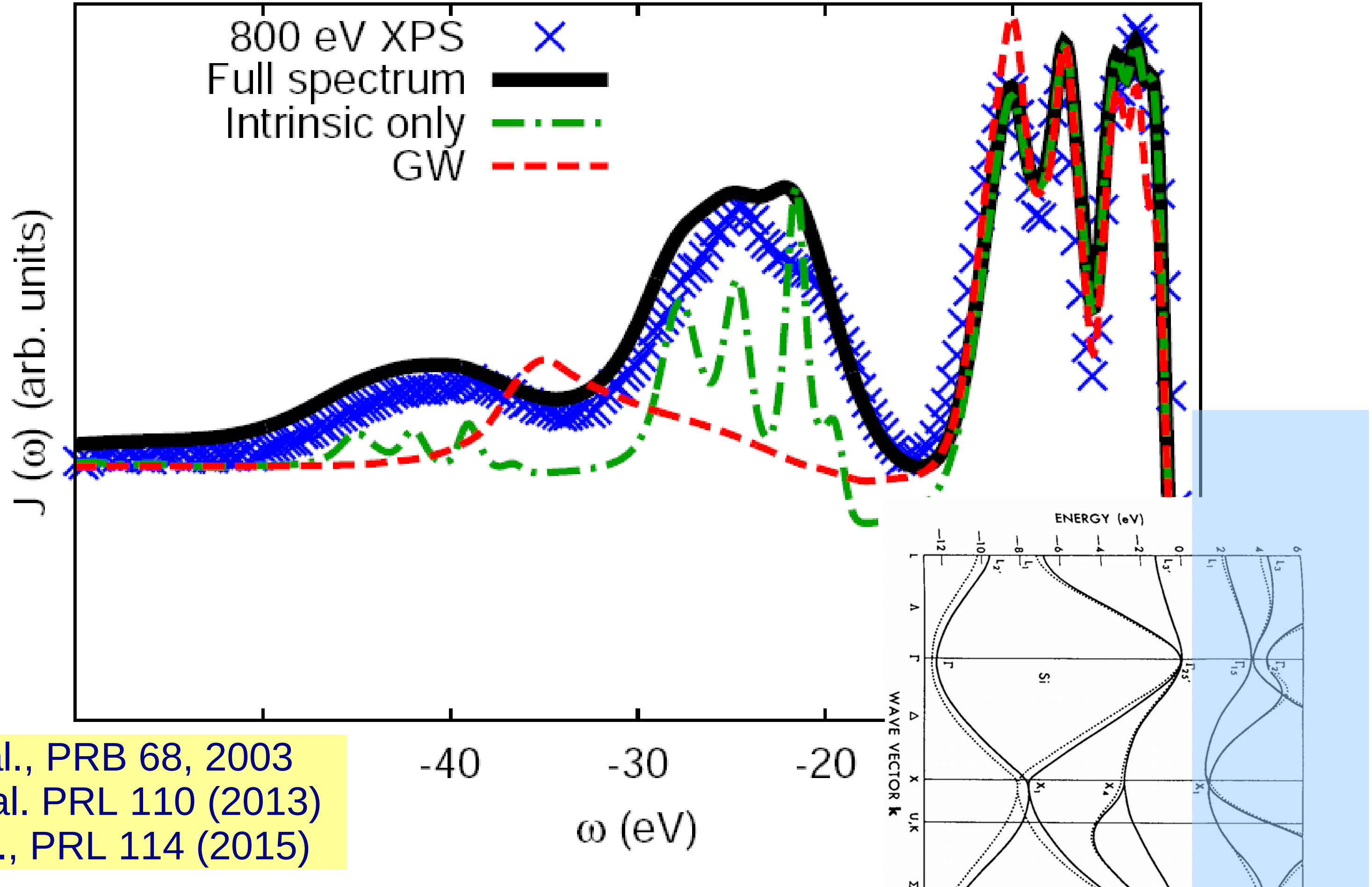
z

→ The one-body spectral function of silicon

M. Guzzo et al., PRL 107, 166401 (2011) in collab. with J. Kas and J. Rehr, M. Silly and F. Sirotti



Kheifets et al., PRB 68, 2003
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Caruso et al., PRL 114 (2015)

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}$$

Following derivation from:

Marilena Tzavala et al., "Nonlinear response in the cumulant expansion for core-level photoemission", Phys. Rev. Research 2, 033147 (2020)

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}$$

$$G_{\mathrm{cl}} = G^0 + G^0 u_{\mathrm{cl}} G_{\mathrm{cl}} \qquad\qquad G_{\mathrm{cl}} = G^H$$

$$G(12) = G^H(12) + G^H(1\bar{1})v(\bar{1}\bar{3})\frac{\delta G(\bar{1}2)}{\delta u(\bar{3}^+)}$$

→ slightly more compact notation

→ highlight corrections wrt Hartree

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\rm cl}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}$$

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$$G_{ij}(t_1t_2) = G^H_{ij}(t_1t_2) + G^H_{im}(t_1t_{\bar{1}})v_{mnkl}\frac{\delta G_{nj}(t_{\bar{1}}t_2)}{\delta u_{kl}(t^+_{\bar{1}})}$$

→ basis transformation: orbitals

→ repeated indices summed

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\text{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}$$

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$$G_{cc}(t_1 t_2) = G^H_{cc}(t_1 t_2) + iG^H_{cc}(t_1 \bar{t}_3)v_{cckl}\frac{\delta G_{cc}(\bar{t}_3 t_2)}{\delta u_{kl}(\bar{t}^+_3)}$$

→ isolated (core) orbital Approximation!
Neglect of overlap

$$G_u(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})v_c(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u(\bar{4}^+)}$$

$$G_{\mathrm{cl}} = G^0 + G^0 u_{\mathrm{cl}} G_{\mathrm{cl}} \qquad\qquad G_{\mathrm{cl}} = G^H$$

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$$G_{ij}(t_1t_2) = G^H_{ij}(t_1t_2) + G^H_{im}(t_1t_{\bar{1}})v_{mnkl}\frac{\delta G_{nj}(t_{\bar{1}}t_2)}{\delta u_{kl}(t^+_{\bar{1}})}$$

$$G_{cc}(t_1t_2) = G^H_{cc}(t_1t_2) + iG^H_{cc}(t_1\bar{t}_3)v_{cckl}\frac{\delta G_{cc}(\bar{t}_3t_2)}{\delta u_{kl}(\bar{t}^+_3)}$$

Ansatz: $G_{cc}(t_1t_2) = G^H_{cc}(t_1t_2)F(t_1t_2)$ → inspired by electron-boson solution

$$\frac{\delta G}{\delta U} = \frac{\delta G^H}{\delta U} F + G^H \frac{\delta F}{\delta U}$$

$$F(t_1 t_2) = 1 + i \frac{G^H_{cc}(t_1 \bar{t}_1)}{G^H_{cc}(t_1 t_2)} \times v_{cckl} \left[\frac{\delta G^H_{cc}(\bar{t}_1 t_2)}{\delta u_{kl}(\bar{t}_1^+)} F(\bar{t}_1 t_2) + G^H_{cc}(\bar{t}_1 t_2) \frac{\delta F(\bar{t}_1 t_2)}{\delta u_{kl}(\bar{t}_1^+)} \right]$$

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Supposing $G^H_{ck} = 0$ for $k \neq c$

with $W_c \equiv W_{cccc}$

$$F(t_1 t_2) = 1 + i \frac{G^H_{cc}(t_1 \bar{t}_1)}{G^H_{cc}(t_1 t_2)} \times \left[W_c(\bar{t}_1^+ \bar{t}_4; u) G^H_{cc}(\bar{t}_1 \bar{t}_4) G^H_{cc}(\bar{t}_4 t_2) F(\bar{t}_1 t_2) + v_{cckl} G^H_{cc}(\bar{t}_1, t_2) \frac{\delta F(\bar{t}_1 t_2)}{\delta u_{kl}(\bar{t}_1^+)} \right]$$

$$\frac{\delta G}{\delta U} = \frac{\delta G^H}{\delta U} F + G^H \frac{\delta F}{\delta U}$$

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Supposing $G^H_{ck} = 0$ for $k \neq c$

and using $W_c \equiv W_{cccc}$

$$W_{cccc}(t_1^+ t_4) = v_{cccc} \delta(t_4 t_1^+) + v_{cckl} v_{cck'l'} \frac{\delta n_{kl}(t_4)}{\delta u_{k'l'}(t_1^+)}$$

$$F(t_1 t_2) = 1 + i \frac{G^H_{cc}(t_1 \bar{t}_1)}{G^H_{cc}(t_1 t_2)} \times \left[W_c(\bar{t}_1^+ \bar{t}_4; u) G^H_{cc}(\bar{t}_1 \bar{t}_4) G^H_{cc}(\bar{t}_4 t_2) F(\bar{t}_1 t_2) + v_{cckl} G^H_{cc}(\bar{t}_1, t_2) \frac{\delta F(\bar{t}_1 t_2)}{\delta u_{kl}(\bar{t}_1^+)} \right]$$

$$
\begin{aligned}
F(t_1 t_2) = 1 + i\frac{G^H_{cc}(t_1 \bar{t}_1)}{G^H_{cc}(t_1 t_2)} \\
\times \Big[W_c(\bar{t}_1^{+} \bar{t}_4; u) G^H_{cc}(\bar{t}_1 \bar{t}_4) G^H_{cc}(\bar{t}_4 t_2) F(\bar{t}_1 t_2) \\
+ v_{cckl} G^H_{cc}(\bar{t}_1, t_2) \frac{\delta F(\bar{t}_1 t_2)}{\delta u_{kl}(\bar{t}_1^{+})} \Big]
\end{aligned}
$$

$$G^H_{cc}(t_1t_2) = i\exp(-i\varepsilon^0_c(t_1 - t_2) + i\int_{t_1}^{t_2} d\tau\, u^H_{cc}(\tau))\theta(t_2 - t_1)$$

$$F(t_1t_2) = 1 + i\frac{G^H_{cc}(t_1\bar{t}_1)}{G^H_{cc}(t_1t_2)} \times\Big[W_c(\bar{t}_1^+\bar{t}_4;u)G^H_{cc}(\bar{t}_1\bar{t}_4)G^H_{cc}(\bar{t}_4t_2)F(\bar{t}_1t_2) + v_{cckl}G^H_{cc}(\bar{t}_1,t_2)\frac{\delta F(\bar{t}_1t_2)}{\delta u_{kl}(\bar{t}_1^+)}\Big]$$

$$F(t_1t_2) = 1 + i\frac{G^H_{cc}(t_1\bar{t}_1)}{G^H_{cc}(t_1t_2)}$$

$$\times\Big[W_c(\bar{t}_1^+\bar{t}_4;u)G^H_{cc}(\bar{t}_1\bar{t}_4)G^H_{cc}(\bar{t}_4t_2)F(\bar{t}_1t_2)$$

$$+v_{cckl}G^H_{cc}(\bar{t}_1,t_2)\frac{\delta F(\bar{t}_1t_2)}{\delta u_{kl}(\bar{t}_1^+)}\Big]$$

$$G^H_{cc}(t_1t_2) = i\exp(-i\varepsilon_c^0(t_1-t_2)+i\int_{t_1}^{t_2}d\tau\, u^H_{cc}(\tau))\theta(t_2-t_1)$$

$$F(t_1t_2) = 1 - i\int_{t_1}^{t_2}d\bar{t}_1\int_{\bar{t}_1}^{t_2}d\tau\, W_c(\bar{t}_1^+\tau;u)F(\bar{t}_1t_2)$$

$$-v_{cckl}\int_{t_1}^{t_2}d\bar{t}_1\,\frac{\delta F(\bar{t}_1t_2)}{\delta u_{kl}(\bar{t}_1^+)}$$

$$F(t_1t_2) = 1 - i\int_{t_1}^{t_2} d\bar{t}_1 \int_{\bar{t}_1}^{t_2} d\tau\, W_c(\bar{t}_1^+\tau; u)F(\bar{t}_1t_2) - v_{cckl}\int_{t_1}^{t_2} d\bar{t}_1\, \frac{\delta F(\bar{t}_1t_2)}{\delta u_{kl}(\bar{t}_1^+)}$$

$$F(t_1t_2) = 1 - i\int_{t_1}^{t_2} d\bar{t}_1 \int_{\bar{t}_1}^{t_2} d\tau\, W_c(\bar{t}_1^+\tau; u)F(\bar{t}_1t_2) - v_{cckl}\int_{t_1}^{t_2} d\bar{t}_1\, \frac{\delta F(\bar{t}_1t_2)}{\delta u_{kl}(\bar{t}_1^+)}$$

$$F(t_1t_2) \equiv e^{C(t_1t_2)}$$

→ inspired by exponential cumulant solution

$$C(t_1t_2) = -i\int_{t_1}^{t_2} d\tau' \int_{\tau'}^{t_2} d\tau\, W_c(\tau'^+\tau; u) - v_{cckl}\int_{t_1}^{t_2} d\tau'\, \frac{\delta C(\tau' t_2)}{\delta u_{kl}(\tau'^+)}$$

$$C^0(t_1t_2) = -i\int_{t_1}^{t_2} d\tau' \int_{\tau'}^{t_2} d\tau W_c(\tau'^+\tau; u)$$

→ This is the linear response cumulant solution!!!

$$C^0(t_1 t_2) = -i \int_{t_1}^{t_2} d\tau' \int_{\tau'}^{t_2} d\tau W_c(\tau'^+ \tau; u)$$

In summary:

$$G_u(1,1') = G_{\mathrm{cl}}(1,1') + iG_{\mathrm{cl}}(1,\bar{2})W_u(\bar{2},\bar{3})\frac{\delta G_u(\bar{2},1')}{\delta u_{\mathrm{cl}}(\bar{3}^+)}$$

Can be solved exactly in linear response and for the case of isolated orbital

$$G(\tau) = G_{\mathrm{cl}}(\tau)\mathcal{F}(\tau) \qquad \mathcal{F}(t_1-t_2) = \exp\left[-i\int_{t_1}^{t_2} dt' \int_{t'}^{t_2} dt''\, \mathcal{W}(t't'')\right]$$

indep. of u_{cl}

$$G_u(1,1') = G_{\rm cl}(1,1') + iG_{\rm cl}(1,\bar{2})W_u(\bar{2},\bar{3})\frac{\delta G_u(\bar{2},1')}{\delta u_{\rm cl}(\bar{3}^+)}$$

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

indep. of $u_{\rm cl}$

$$G_u(1,1') = G_{\rm cl}(1,1') + iG_{\rm cl}(1,\bar{2})W_u(\bar{2},\bar{3})\frac{\delta G_u(\bar{2},1')}{\delta u_{\rm cl}(\bar{3}^+)}$$

Can be solved exactly in linear response and for the case of isolated orbital

$$G(\tau) = G_{\rm cl}(\tau)\mathcal{F}(\tau) \qquad \mathcal{F}(t_1 - t_2) = \exp\left[\cdots \right]$$

Functional of Σ_{GW}

indep. of $u_{\rm cl}$

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

$$+ \frac{\lambda}{\omega_p^2} \frac{1}{(\omega - \varepsilon^{QP} + \omega_p)^2 + \Gamma^2} +$$

$$+ \frac{1}{2}\left(\frac{\lambda}{\omega_p^2}\right)^2 \frac{1}{(\omega - \varepsilon^{QP} + 2\omega_p)^2 + \Gamma^2} +$$

$$\left. + \frac{1}{6}\left(\frac{\lambda}{\omega_p^2}\right)^3 \frac{1}{(\omega - \varepsilon^{QP} + 3\omega_p)^2 + \Gamma^2} + \ldots \right]$$

→ Cumulant expansion in bosons

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, 19

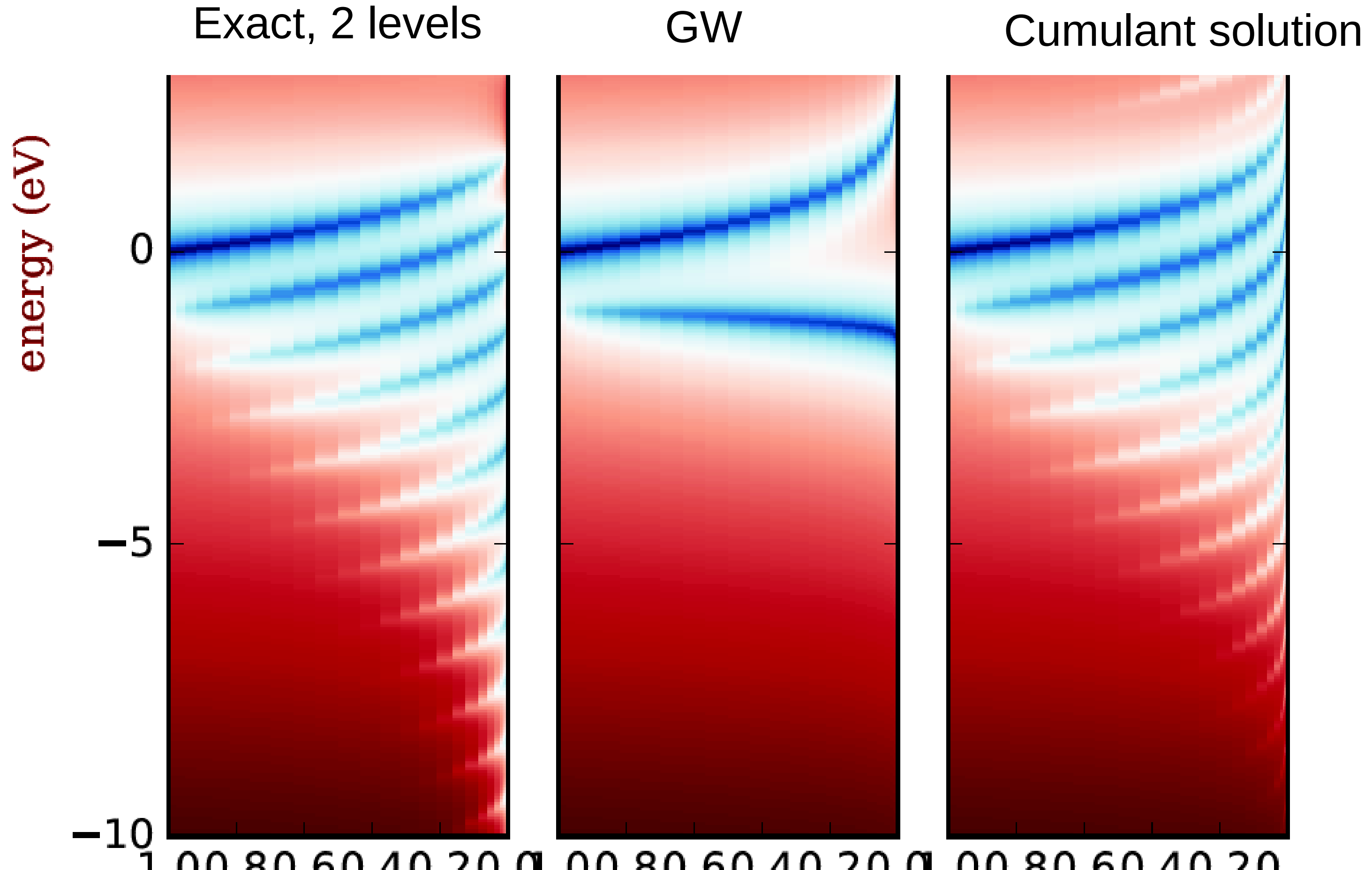
Silicon: Kheifets et al., PRB 68, 2003

In DMFT context: Casula, Rubtsov, Biermann, PRB 85, 035115 (2012)

Here:
→ the first in a series of approximations
→ link to GW
→ prescription for ingredients

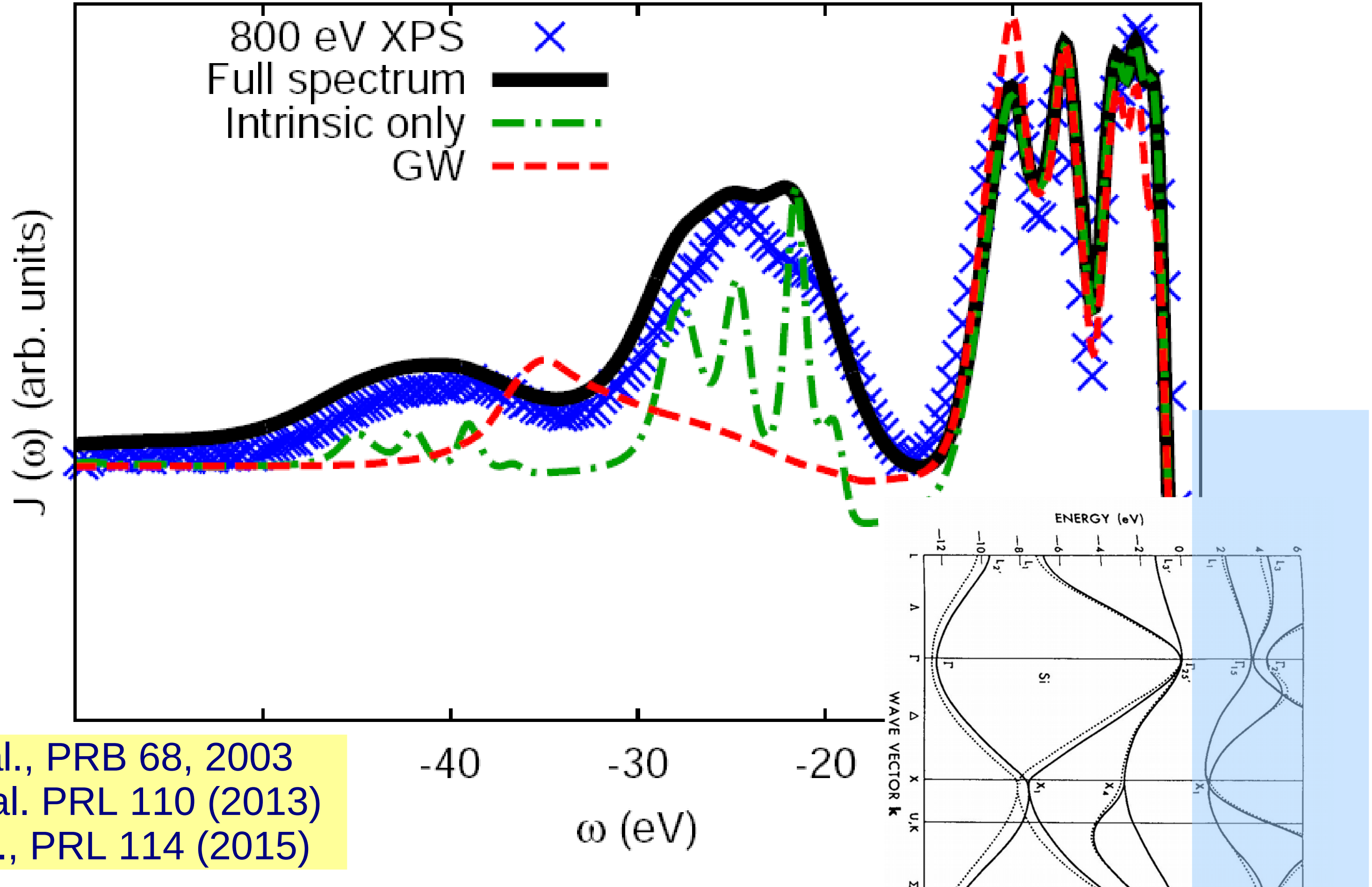
GW = bad solution of reasonable hamiltonian

Cumulant expansion = better solution, also for more than 1 level



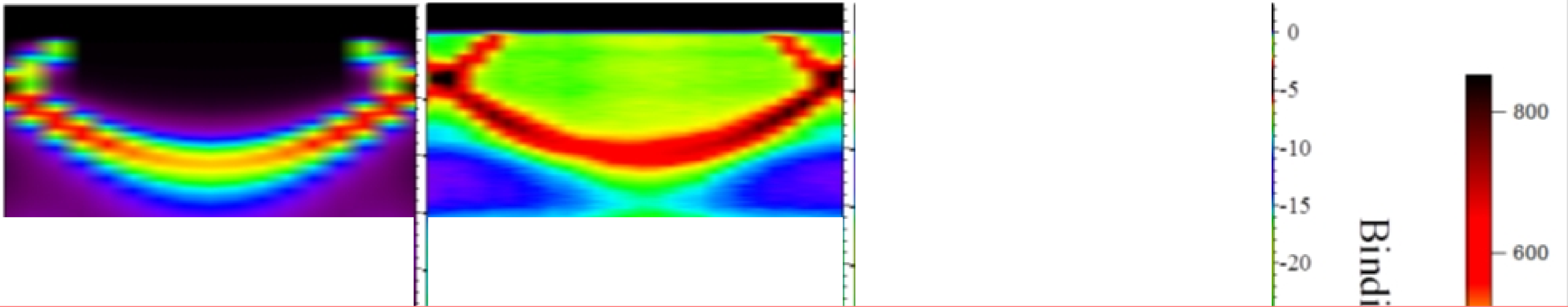
→ The one-body spectral function of silicon

M. Guzzo et al., PRL 107, 166401 (2011) in collab. with J. Kas and J. Rehr, M. Silly and F. Sirotti

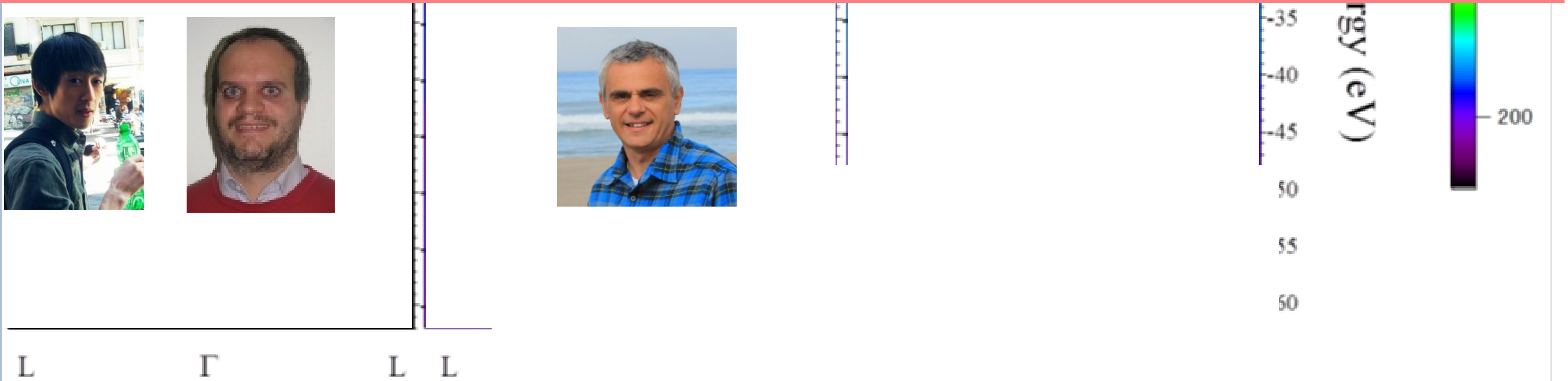


Kheifets et al., PRB 68, 2003
Lischner et al. PRL 110 (2013)
Caruso et al., PRL 114 (2015)

State-of-the-art: photoemission of bulk aluminum



Zhou, Reining, Nicolaou, Bendounan, Ruotsalainen,Vanzini, Kas, Rehr, Muntwiler, Strocov, Sirotti, Gatti, PNAS 117 (46), 28596 (2020)



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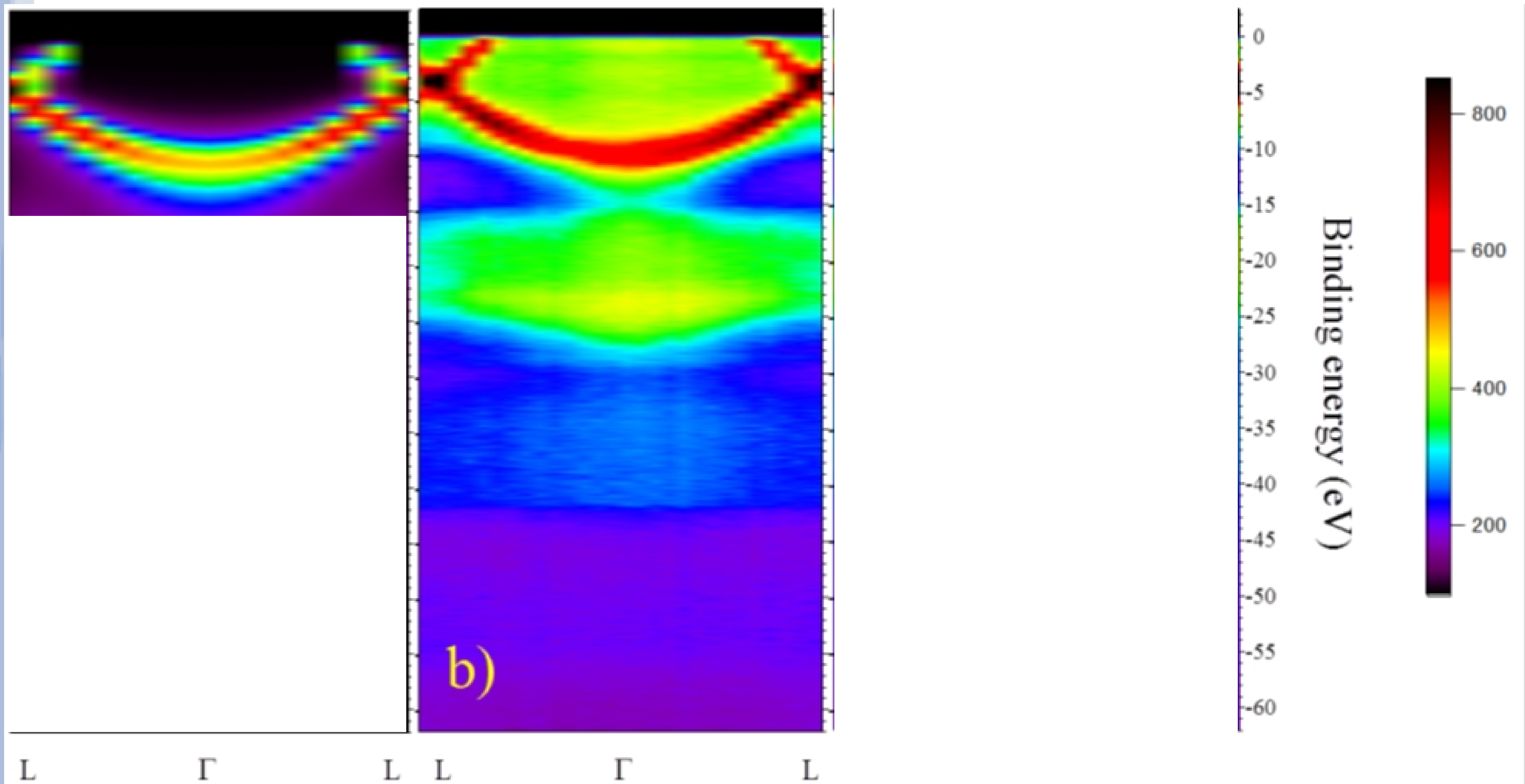

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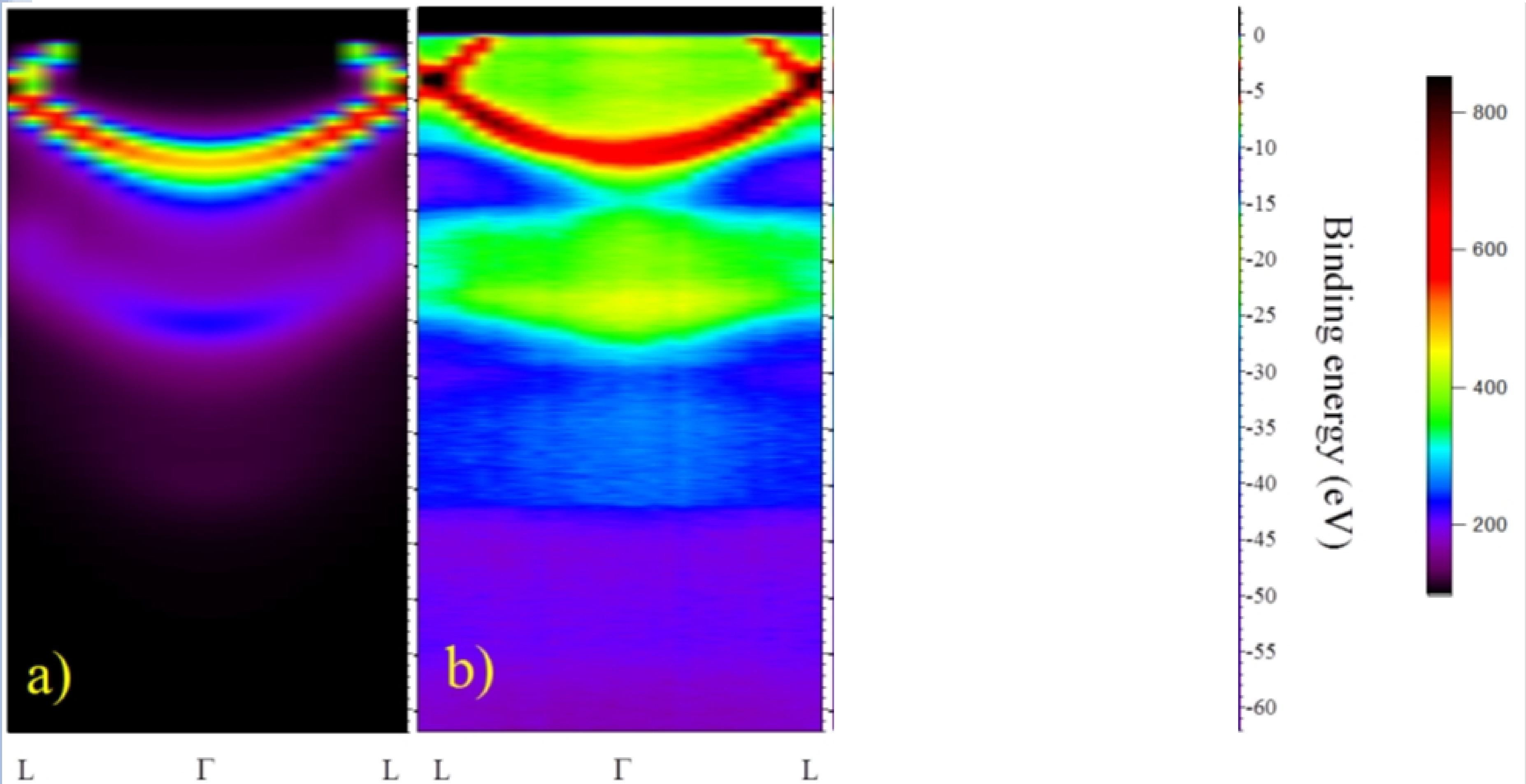
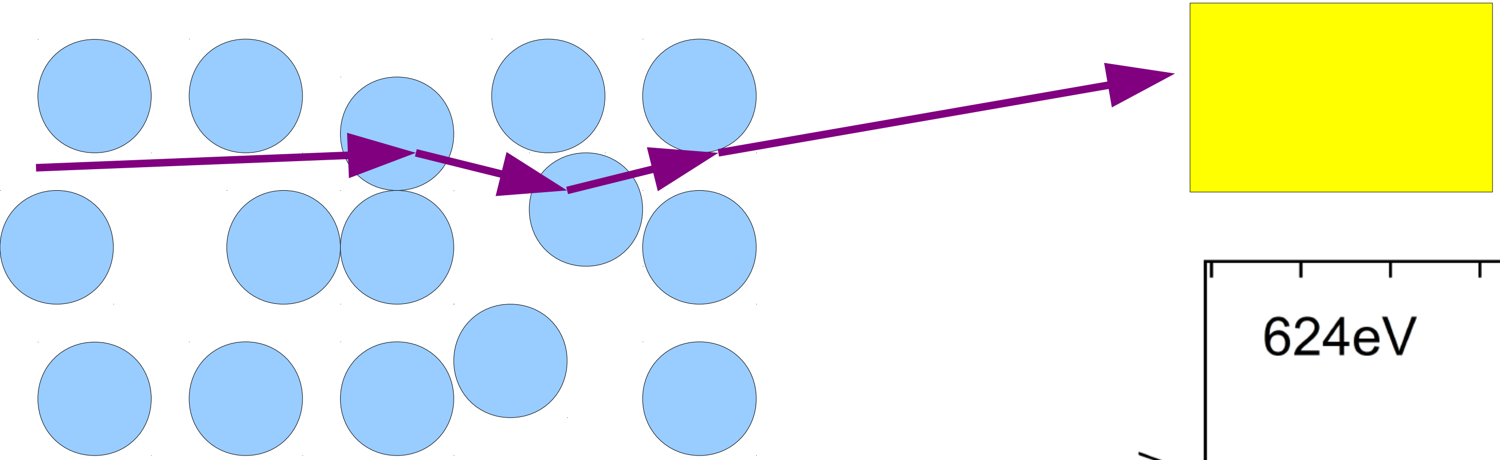
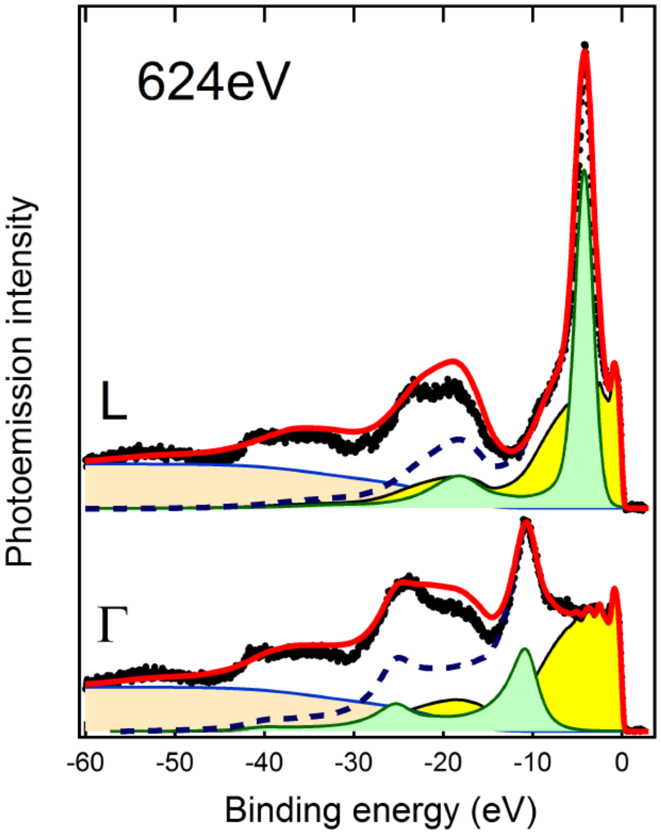






We measure at 35-50 K

In Bandstructure region:
Shevchik, PRB 16, 3428 (1977);
PRB.20, 3020 (1979).
J. Braun, et al.,PRB 88, 205409 (2013).
C. Sondergaard, et al.,PRB 63, 233102 (2001).
P. Hofmann, et al., PRB 66, 245422 (2002).

Measured spectrum contains also:

→ ~ cross sections

→ ~ background

→ extrinsic+interference

Measured spectrum contains also:

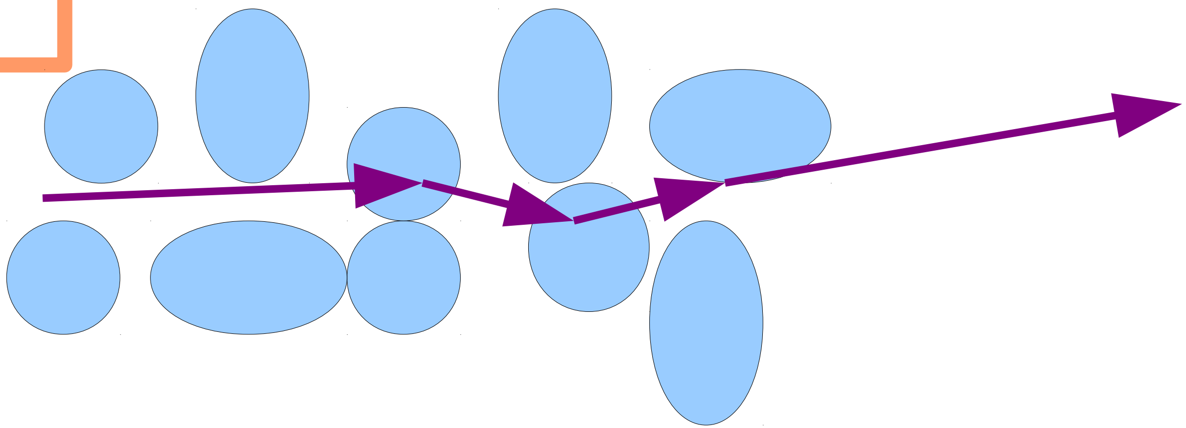
→ ~ cross sections

→ ~ background

→ extrinsic+interference

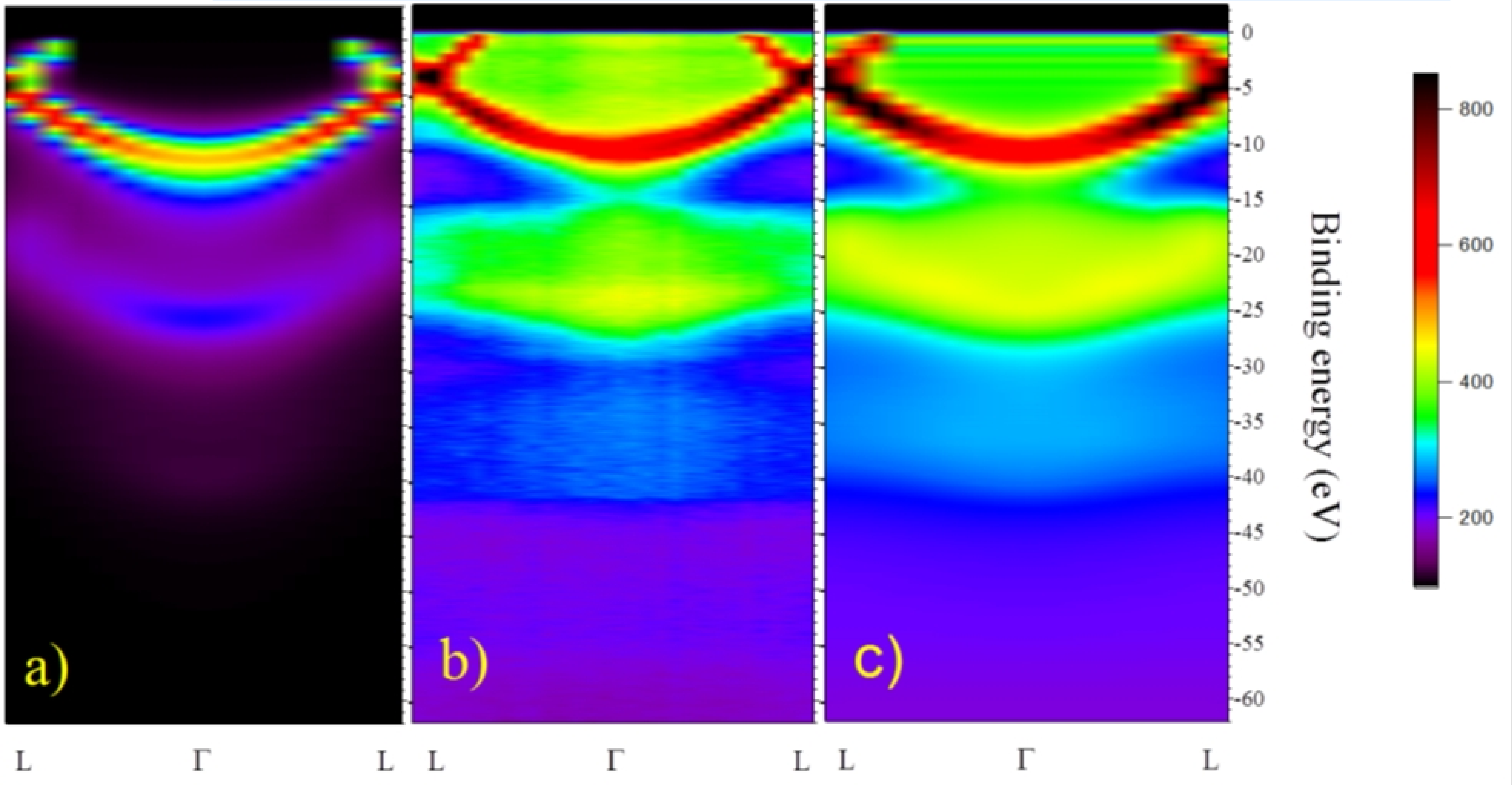
Z ~ 0.75
for both intrinsic
and ext./interf.

Scattering of outgoing photoelectron
→ enhancement of satellites



W. Bardyszewski and L. Hedin, Physica Scripta 32, 439 (1985)
L. Hedin, J. Michiels, J. Inglesfield, PRB 58, 15565 (1998).

State-of-the-art: photoemission of bulk aluminum



Zhou, Reining, Nicolaou, Bendounan, Ruotsalainen,Vanzini, Kas, Rehr, Muntwiler, Strocov, Sirotti, Gatti, PNAS 117 (46), 28596 (2020)

Issues:

→ self-screening

→ strong correlation (degeneracy)

→ **linear response????**

→ satellites

GW approximation and linear response

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Isolated orbital:

Marilena Tzavala et al., “Nonlinear response in the cumulant expansion for core-level photoemission”, Phys. Rev. Research 2, 033147 (2020)

$$C(t_1 t_2) = -i \int_{t_1}^{t_2} d\tau' \int_{\tau'}^{t_2} d\tau \, W_c(\tau'^{+} \tau; u) - v_{cckl} \int_{t_1}^{t_2} d\tau' \frac{\delta C(\tau' t_2)}{\delta u_{kl}(\tau'^{+})}.$$

$$C^0(t_1 t_2) = -i \int_{t_1}^{t_2} d\tau' \int_{\tau'}^{t_2} d\tau W_c(\tau'^{+} \tau; u)$$

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$$C^0(t_1 t_2) = -i \int_{t_1}^{t_2} d\tau' \int_{\tau'}^{t_2} d\tau W_c(\tau'^{+}\tau; u)$$

Iterate!!!

$$C^1(t_1 t_2) = i v_{cckl} \int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \int_{t_1}^{\tau'} d\tau'' \frac{\delta W_c(\tau' \tau; u)}{\delta u_{kl}(\tau''^+)}$$

cf G. D. Mahan, Phys. Rev. B 25, 5021 (1982)

$$C^1(t_1 t_2) = i v_{cckl} \int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \int_{t_1}^{\tau'} d\tau'' \frac{\delta W_c(\tau' \tau; u)}{\delta u_{kl}(\tau''^{+})}$$

cf G. D. Mahan, Phys. Rev. B 25, 5021 (1982)

$$\begin{aligned} C(t_1 t_2) = {} & -i \int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \, W_c(\tau, \tau') \\ & + i \int_{t_1}^{t_2} d\tau \sum_{m=1}^{\infty} v_{cck_1 l_1} \cdots v_{cck_m l_m} \\ & \times \frac{(-1)^{(m+1)}}{(m+1)!} \int_{t_1}^{\tau} d\tau' \cdots \int_{t_1}^{\tau} d\tau_m \\ & \times \frac{\delta^m W_c(\tau, \tau_m)}{\delta u_{k_1 l_1}(\tau') \delta u_{k_2 l_2}(\tau_1) \cdots \delta u_{k_m l_m}(\tau_{m-1})} \end{aligned}$$

$$C^1(t_1t_2) = iv_{cckl}\int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \int_{t_1}^{\tau'} d\tau'' \frac{\delta W_c(\tau'\tau;u)}{\delta u_{kl}(\tau''^+)}$$

$$C(t_1t_2) = -i\int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \, W_c(\tau,\tau')$$

$$+ i\int_{t_1}^{t_2} d\tau \sum_{m=1}^{\infty} v_{cck_1l_1}\cdots v_{cck_ml_m}$$

$$\times \frac{(-1)^{(m+1)}}{(m+1)!}\int_{t_1}^{\tau} d\tau' \cdots \int_{t_1}^{\tau} d\tau_m$$

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cf G. D. Mahan, Phys. Rev. B 25, 5021 (1982)

→ is it necessary to go beyond C^1?

$$C^1(t_1t_2) = iv_{cckl}\int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \int_{t_1}^{\tau'} d\tau'' \frac{\delta W_c(\tau'\tau;u)}{\delta u_{kl}(\tau''^{+})}$$

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cf G. D. Mahan, Phys. Rev. B 25, 5021 (1982)

→ is it necessary to go beyond C^1?

→ how to use this expression?

$$C^1(t_1t_2) = iv_{cckl}\int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \int_{t_1}^{\tau'} d\tau'' \frac{\delta W_c(\tau'\tau;u)}{\delta u_{kl}(\tau''^+)}$$

$$C(t_1t_2) = -i\int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \, W_c(\tau,\tau') + i\int_{t_1}^{t_2} d\tau \sum_{m=1}^{\infty} v_{cck_1l_1}\cdots v_{cck_ml_m} \times \frac{(-1)^{(m+1)}}{(m+1)!}\int_{t_1}^{\tau} d\tau' \cdots \int_{t_1}^{\tau} d\tau_m \times \frac{\delta^m W_c(\tau,\tau_m)}{\delta u_{k_1l_1}(\tau')\delta u_{k_2l_2}(\tau_1)\cdots\delta u_{k_ml_m}(\tau_{m-1})}$$

cf G. D. Mahan, Phys. Rev. B 25, 5021 (1982)

→ is it necessary to go beyond C^1?

→ how to use this expression?

$$W_c(t_1^+t_4) \equiv W_{cccc}(t_1^+t_4) = v_{cccc}\delta(t_4t_1^+) + v_{cckl}v_{cck'l'}\frac{\delta n_{kl}(t_4)}{\delta u_{k'l'}(t_1^+)}.$$

would suggest
to integrate density response to all orders.

$$C^1(t_1 t_2) = i v_{cckl} \int_{t_1}^{t_2} d\tau \int_{t_1}^{\tau} d\tau' \int_{t_1}^{\tau'} d\tau'' \frac{\delta W_c(\tau'\tau; u)}{\delta u_{kl}(\tau''^{+})}$$

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→ how to use this expression?

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would suggest
to integrate density response to all orders.

BUT we are not causal – and used that!

Impose correct analytic properties by extending exact lowest order relation to all orders:

$$C_{\mathrm{TDR}}(t_1 t_2) = -i v_{cccc}(t_2 - t_1) + \int_0^\infty \frac{d\omega}{\pi} \frac{v_{ccij} \mathrm{Re}[\Delta n_{ij}(\omega)]}{\omega} \times [e^{-i\omega(t_2 - t_1)} + i\omega(t_2 - t_1) - 1]$$

To lowest order we have:

$$C^0(t_1 t_2) = -i v_{cccc}(t_2 - t_1) + \int_0^\infty \frac{d\omega}{\pi} \frac{v_{ccij} \mathrm{Re}\, \Delta n^0_{ij}(\omega)}{\omega} f(\omega, t_2 - t_1)$$

$$f(\omega, t) \equiv (e^{-i\omega t} + i\omega t - 1)$$

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$$f(\omega, t) \equiv (e^{-i\omega t} + i\omega t - 1)$$

How good is this?????

When would you expect to see non-linear effects?

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→ when the boson is not fixed

$$H = \epsilon_0 c^\dagger c + c c^\dagger g(a + a^\dagger) + \omega_0 a^\dagger a$$

When would you expect to see non-linear effects?

→ when the boson is not fixed

→ when a change in the potential changes the reponse

$$G_u(1,1') = G_{\text{cl}}(1,1') + iG_{\text{cl}}(1,\bar{2})W_u(\bar{2},\bar{3})\frac{\delta G_u(\bar{2},1')}{\delta u_{\text{cl}}(\bar{3}^+)}$$

When would you expect to see non-linear effects?

→ when the boson is not fixed

→ when a change in the density changes the reponse

→ when the charge induced in linear response changes the response of the system

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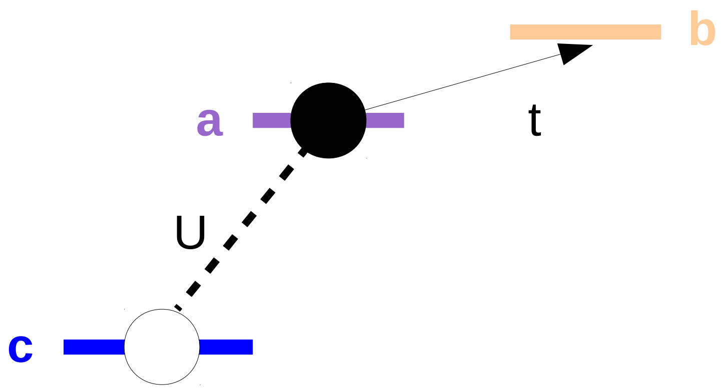
$$\hat{H} = \epsilon_0 \hat{c}^\dagger \hat{c} + \epsilon_a^0 \hat{n}_a + \epsilon_b^0 \hat{n}_b - U \hat{n}_h \hat{n}_a - t(\hat{c}_a^\dagger \hat{c}_b + \hat{c}_b^\dagger \hat{c}_a)$$

Isolated orbital:

Marilena Tzavala et al., “Nonlinear response in the cumulant expansion for core-level photoemission”, Phys. Rev. Research 2, 033147 (2020)



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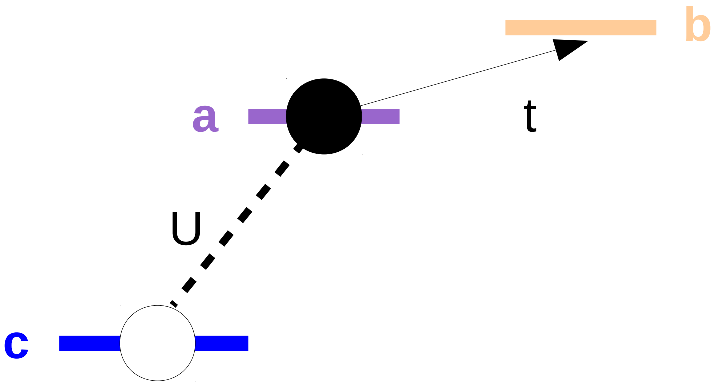
“→ when the charge induced in linear response changes the response of the system”



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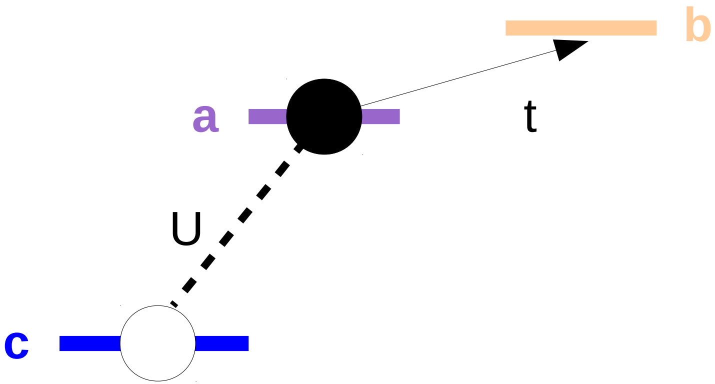

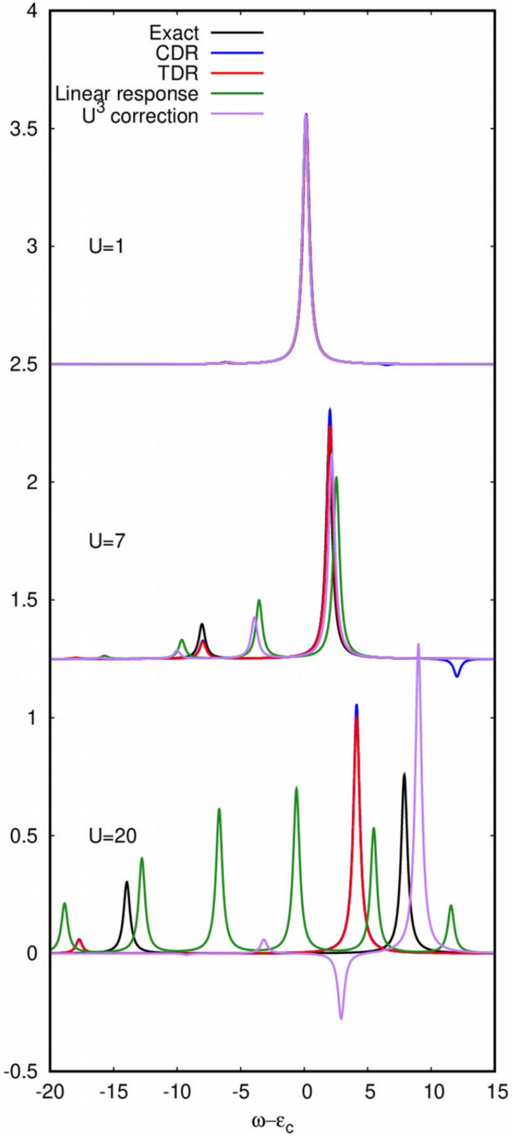

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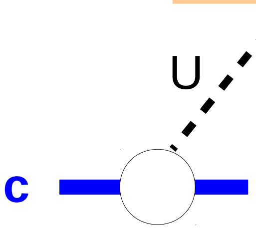
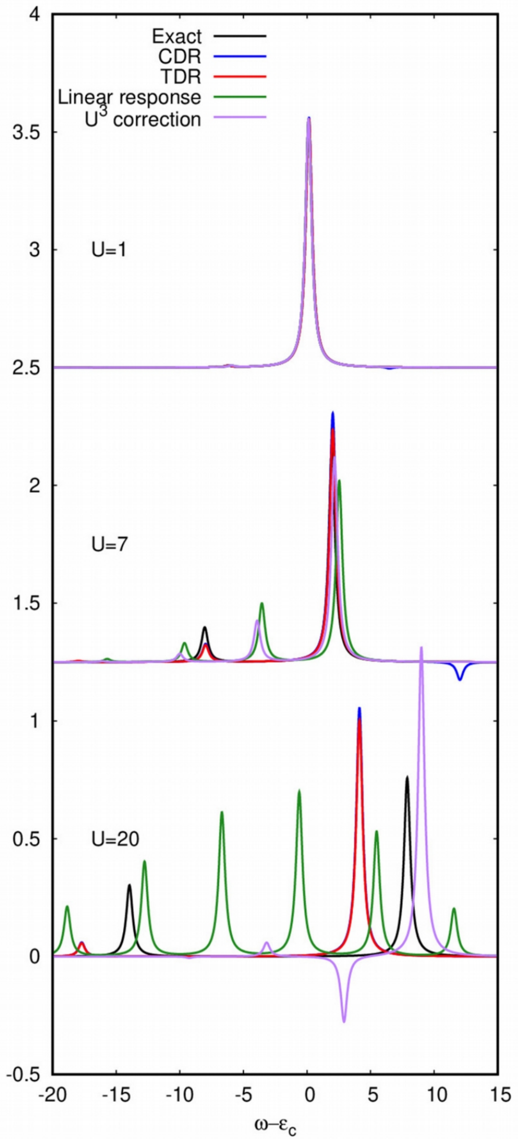
Isolated orbital:

Marilena Tzavala ... n
for core-level pho
Phys. Rev. Resear

- → for small system, LR cumulant meaningless
- → (except for environment)
- → beyond LR “knows” this
- → no longer simple electron-boson
- → our approx promising for moderate coupling
- → TDDFT can be used





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Go beyond in Dyson equation ?

$$G(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})W_u(\bar{3},\bar{4})\frac{\delta G_u(\bar{3},2)}{\delta u_{\mathrm{cl}}(\bar{4}^+)};$$

$$G(1,2) = G^0(1,2) + G^0(1,\bar{3})u_{\mathrm{cl}}(\bar{3})G_u(\bar{3},2) + iG^0(1,\bar{3})W_u(\bar{3},\bar{4})G(\bar{3},\bar{5})\left(-\frac{\delta G_u^{-1}(\bar{5},\bar{6})}{\delta u_{\mathrm{cl}}(\bar{4}^+)}\right)G(\bar{6},2)$$

G^0 Σ G

Dyson equation: approximate Σ

Hedin's equations
L. Hedin, “New method for calculating the one-particle Green’s function with application to the electron-gas problem,” Phys. Rev. 139:A796–823, 1965

One should expect that in principle this should yield non-linear response

$$\frac{\delta G^{-1}}{\delta u_{\text{cl}}} \longrightarrow \frac{\delta \Sigma^{GW}}{\delta u_{\text{cl}}} = i\frac{\delta G}{\delta u_{\text{cl}}}W + iG\frac{\delta W}{\delta u_{\text{cl}}}$$

$$\frac{\delta G^{-1}}{\delta u_{\mathrm{cl}}} \longrightarrow \frac{\delta \Sigma^{GW}}{\delta u_{\mathrm{cl}}} = i\frac{\delta G}{\delta u_{\mathrm{cl}}}W + iG\frac{\delta W}{\delta u_{\mathrm{cl}}}$$

$$\approx iGGW + GWGGGW$$

$$\frac{\delta W}{\delta u_{\mathrm{cl}}} = -W\frac{\delta W^{-1}}{\delta u_{\mathrm{cl}}}W$$

$$W^{-1} = v_c^{-1} - P \approx v_c^{-1} + iGG$$

$$\frac{\delta G^{-1}}{\delta u_{\mathrm{cl}}} \longrightarrow \frac{\delta \Sigma^{GW}}{\delta u_{\mathrm{cl}}} = i\frac{\delta G}{\delta u_{\mathrm{cl}}}W + iG\frac{\delta W}{\delta u_{\mathrm{cl}}}$$

$$\approx iGGW + GWGGGW$$

This is approx. contained in the LR cumulant

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See also A. Schindlmayr and R. W. Godby, Phys. Rev. Lett. 80, 1702 (1998)

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This is approx. contained in the LR cumulant

This is a higher order vertex correction

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Would have to sum somehow higher orders

$$W^{-1} = v_c^{-1} - P \approx v_c^{-1} + iGG$$

GW approximation and linear response

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3. From GW to cumulant Green’s functions

4. Non-linear response cumulant approximation

5. Model results

6. Discussion: vertex corrections

7. Conclusions?

GW approximation and linear response

→ GW and cumulant: ~ effective e-boson ham

→ GW bad solution of that ham

→ sometimes ham bad

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→ GW and cumulant: ~ effective e-boson ham

→ GW bad solution of that ham

→ sometimes ham bad

→ for small system, LR cumulant meaningless

→ (except for environment)

→ beyond LR "knows" this

→ no longer simple electron-boson

→ our approx promising for moderate coupling

→ TDDFT can be used