Diagrammatic expansions for positive spectral functions

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Violations of positivity

- Photo-emission and photo-absorption spectra represent probability distributions and are positive semi-definite

- A general diagrammatic expansion for the self-energy or the density response function will not preserve this positivity

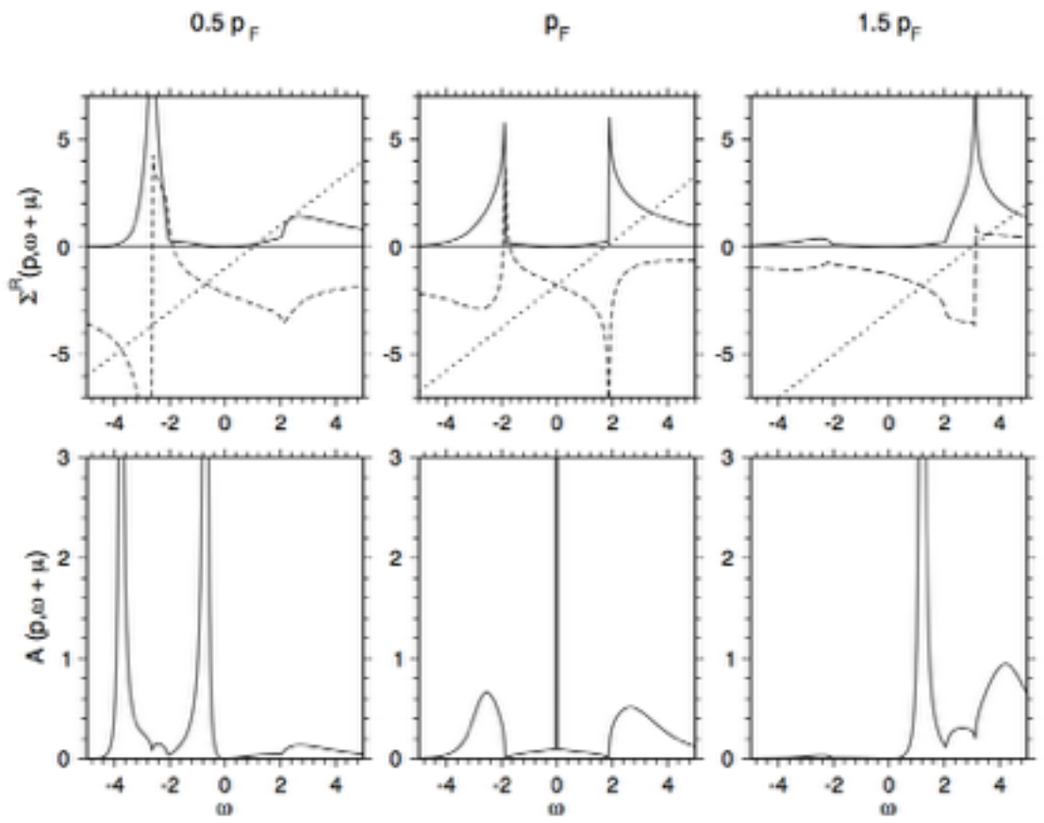
  1) happens in the simplest diagrammatic expansions beyond GW and RPA
  2) leads to wrong analytic properties
  3) prevents doing self-consistent calculations

- This feature has prevented progress in the study of vertex corrections which describe new physical processes

Why is the vertex interesting?

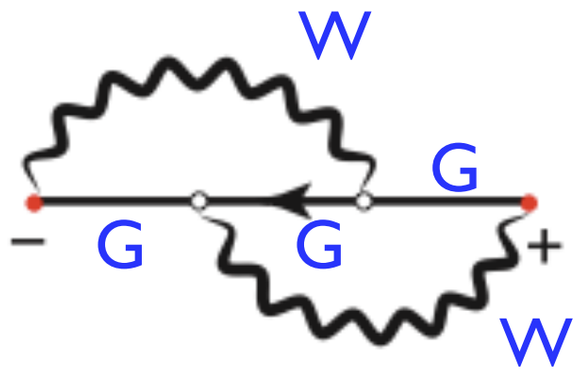
- To study plasmon satellites
- To investigate cancellations between dressing and vertex corrections
- To see if vertex corrections improve the bandwidth of the simple metals
- To look at plasmon life-times
- To make frequency-dependent xc-kernels in TDDFT
- …..

Spectral function for the self-energy and Green's function in non-sc GW



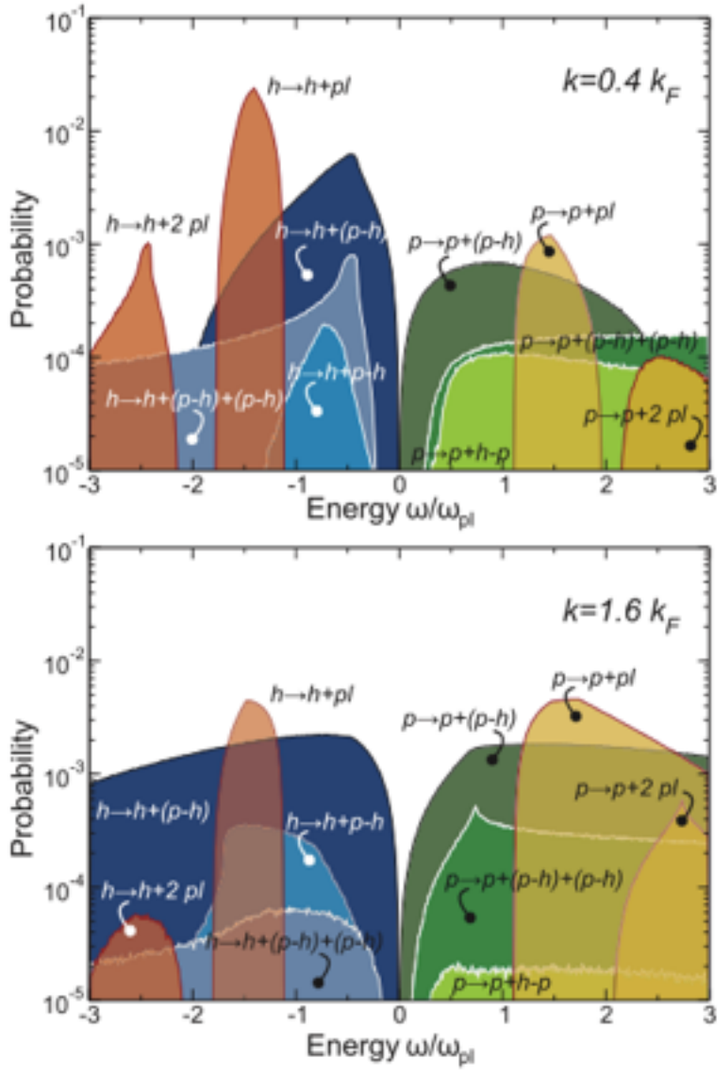
Vertex

GGGWW-approximation



The lowest order vertex contains new processes

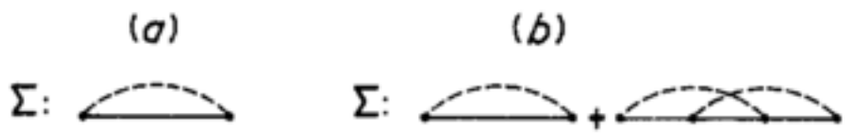
- generation of 2 plasmons
- generation of plasmon + (p-h) pair
- generation of 2 (p-h) pairs



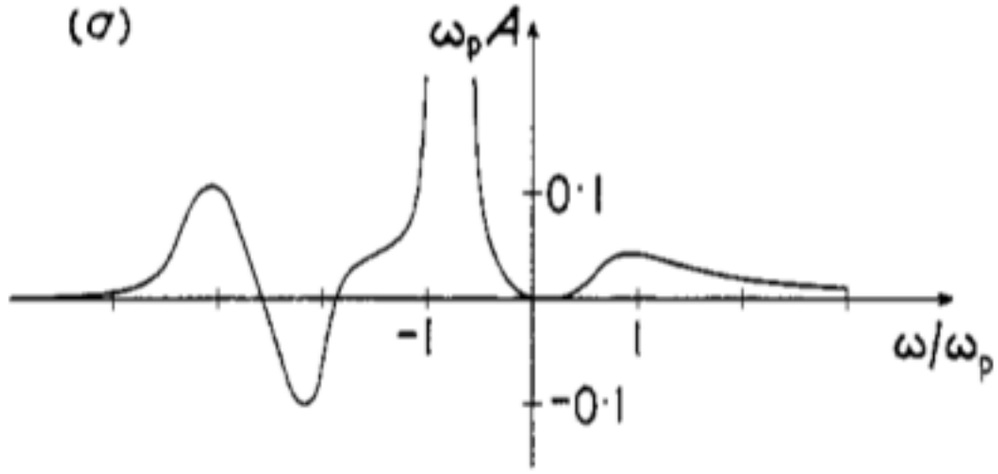
Y.Pavlyukh, A.Rubio, J.Berakdar,
Phys.Rev.B87,205124 (2013)

Spectral function with vertex is not positive

The vertex was already studied by Minnhagen in 1974 for the electron gas and found to be non-positive



P.Minnhagen, J.Phys.C: Solid State Phys. 7, 3013 (1974)



-wrong analytic properties
-prevents self-consistent calculations

How can we solve this problem?

Propagators

The basic building blocks of Feynman diagrams are the particle and hole propagators

$$G^{>}(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2) = -i\langle \hat{\psi}(\mathbf{x}_1 t_1)\hat{\psi}^{\dagger}(\mathbf{x}_2 t_2)\rangle \quad \Rightarrow \quad \text{particle propagator}$$

$$G^{<}(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2) = i\langle \hat{\psi}^{\dagger}(\mathbf{x}_2 t_2)\hat{\psi}(\mathbf{x}_1 t_1)\rangle \quad \Rightarrow \quad \text{hole propagator}$$

In frequency space they have the structure

$$iG^{>}(\mathbf{x}_1, \mathbf{x}_2; \omega) = 2\pi \sum_s g_s(\mathbf{x}_1) g_s^*(\mathbf{x}_2) \delta(\omega - [E_{N+1,s} - E_{N,0}]) \quad \Rightarrow \quad \text{addition energies}$$

$$-iG^{<}(\mathbf{x}_1, \mathbf{x}_2; \omega) = 2\pi \sum_s f_s(\mathbf{x}_2) f_s^*(\mathbf{x}_1) \delta(\omega - [E_{N,0} - E_{N-1,s}]) \quad \Rightarrow \quad \text{removal energies}$$

As integral kernels they are positive semi-definite (PSD) operators

$$\langle \varphi | -i\hat{G}^{<}(\omega) | \varphi \rangle \geq 0 \qquad \langle \varphi | i\hat{G}^{>}(\omega) | \varphi \rangle \geq 0$$

$$\hat{\mathcal{A}}(\omega) = i\left[\hat{G}^{>}(\omega) - \hat{G}^{<}(\omega)\right]$$

spectral function

The particle and hole propagators are related to the particle and hole components of the self-energy as

$$G^{\lessgtr}(\omega) = G^{\mathrm{R}}(\omega)\Sigma_{\mathrm{c}}^{\lessgtr}(\omega)G^{\mathrm{A}}(\omega)$$

where the retarded and advanced Green's functions are given by

$$G^{\mathrm{R/A}}(\omega) = i\int \frac{d\omega'}{2\pi}\frac{G^{>}(\omega) - G^{<}(\omega)}{\omega - \omega' \pm i\eta}$$

Since $G^{\mathrm{A}}(\omega) = [G^{\mathrm{R}}(\omega)]^{\dagger}$

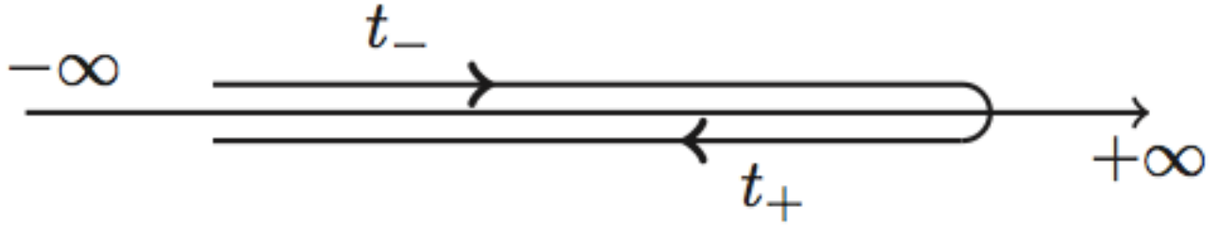
$\mp iG^{\lessgtr}(\omega)$ PSD $\iff$ $\mp i\Sigma_{\mathrm{c}}^{\lessgtr}(\omega)$ PSD

Another useful relation is

$$2\,\mathrm{Im}\Sigma^{\mathrm{R}}(\omega) = i\left[\Sigma_{\mathrm{c}}^{>}(\omega) - \Sigma_{\mathrm{c}}^{<}(\omega)\right]$$

Diagrammatic expansion of the spectral function

A direct expansion of the spectral function can be carried out using the Keldysh technique



The different propagators are obtained by restricting times on different branches of the contour

particle and hole propagators

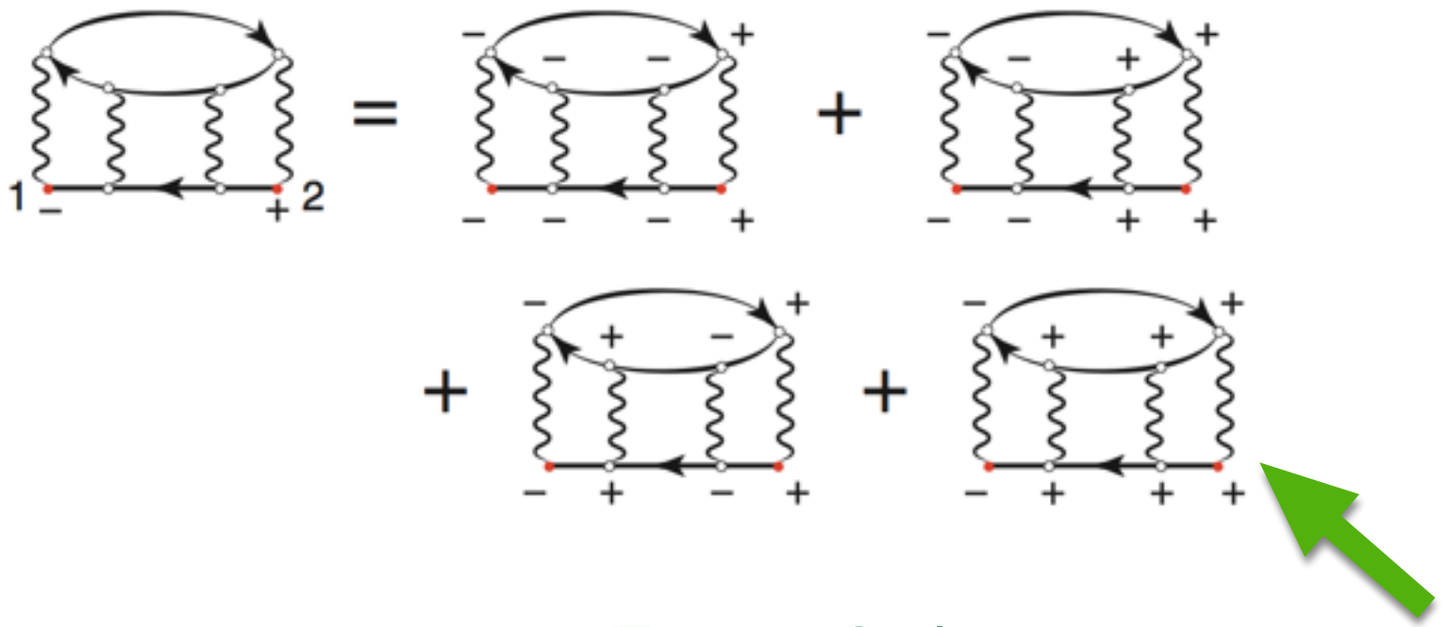
$$G^{-+}(t_1,t_2) = G^{<}(t_1,t_2)$$
$$G^{+-}(t_1,t_2) = G^{>}(t_1,t_2)$$

time- and anti-time ordered Green's functions

$$G^{--}(t_1,t_2) = \theta(t_1-t_2)G^{>}(t_1,t_2) + \theta(t_2-t_1)G^{<}(t_1,t_2) \rightarrow \text{time-ordered}$$
$$G^{++}(t_1,t_2) = \theta(t_2-t_1)G^{>}(t_1,t_2) + \theta(t_1-t_2)G^{<}(t_1,t_2) \rightarrow \text{anti-time ordered}$$

Then we can expand the spectral function for the hole scattering as, for example

$$\Sigma^<(1,2) =$$



The bare Coulomb interaction is instantaneous and only connects times on the same branch

$$v(1,2) = \delta(z_1, z_2) v(\mathbf{x}_1, \mathbf{x}_2)$$

Now we have a direct expansion of the spectral function, but how do we ensure that it is positive semi-definite (PSD) ?

We use the following expressions for the correlation part of the self-energy

$$\begin{aligned} i\Sigma_{\rm c}^>(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2) &= \langle \gamma_H(\mathbf{x}_1 t_1) \gamma_H^\dagger(\mathbf{x}_2 t_2) \rangle_{\rm irr} \\ -i\Sigma_{\rm c}^<(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2) &= \langle \gamma_H^\dagger(\mathbf{x}_2 t_2) \gamma_H(\mathbf{x}_1 t_1) \rangle_{\rm irr} \end{aligned}$$

where

$$\hat{\gamma}(\mathbf{x}_1) = \int d\mathbf{x}\, v(\mathbf{x}_1, \mathbf{x}) \hat{\psi}^\dagger(\mathbf{x}) \hat{\psi}(\mathbf{x}) \hat{\psi}(\mathbf{x}_1)$$

We can therefore write

$$-i\Sigma_{\rm c}^{<}(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2) = \langle \Psi_0 | \mathcal{U}(t_0, t_2)\gamma(\mathbf{x}_2)\mathcal{U}(t_2, t_0)\,\mathcal{U}(t_0, t_1)\gamma^{\dagger}(\mathbf{x}_1)\mathcal{U}(t_1, t_0)|\Psi_0\rangle_{\rm irr}$$

We connect the ground state using adiabatic switch-on to a non-interacting state

$$|\Psi_0\rangle = \mathcal{U}(t_0, \tau)|\Phi_0\rangle \quad (\tau \to +\infty)$$

to obtain

$$-i\Sigma_{\rm c}^{<}(\mathbf{x}_1 t_1, \mathbf{x}_2 t_2) = \langle \Phi_0 | \mathcal{U}(\tau, t_2)\gamma(\mathbf{x}_2)\mathcal{U}(t_2, \tau)\,\mathcal{U}(\tau, t_1)\gamma^{\dagger}(\mathbf{x}_1)\mathcal{U}(t_1, \tau)|\Phi_0\rangle_{\rm irr}$$

We insert a complete set of states here to establish a product structure

We take these states to be

$$|\chi_{\underline{pq}}^{(N)}\rangle = \hat{c}_{q_N}^{\dagger} \ldots \hat{c}_{q_1}^{\dagger}\,\hat{c}_{p_{N+1}} \ldots \hat{c}_{p_1}|\Phi_0\rangle$$

$$1 = \sum_{N=0}^{\infty} \frac{1}{N!(N+1)!} \sum_{\underline{pq}} |\chi_{\underline{pq}}^{(N)}\rangle\langle\chi_{\underline{pq}}^{(N)}|$$

p-labels run over occupied states and
q-labels run over unoccupied state

We then obtain

$$\Sigma_c^<(1,2) = i\left[\sum_{N=0}^{\infty}\frac{1}{(N+1)!N!}\sum_{\underline{pq}} S_{N,\underline{pq}}(2)S^*_{N,\underline{pq}}(1)\right]_{\rm irr}.$$

where

$$S^*_{\underline{pq}}(1) = \frac{\langle\Phi_0|T\left\{e^{-i\int_{-\tau}^{\tau}dt\hat{H}(t)}\hat{c}^\dagger_{p_1}(\tau^+)\dots\hat{c}^\dagger_{p_{N+1}}(\tau^+)\hat{c}_{q_1}(\tau)\dots\hat{c}_{q_N}(\tau)\gamma(\mathbf{x}_1t_1)\right\}|\Phi_0\rangle}{\langle\Phi_0|T\left\{e^{-i\int_{-\tau}^{\tau}dt\hat{H}(t)}\right\}|\Phi_0\rangle}$$

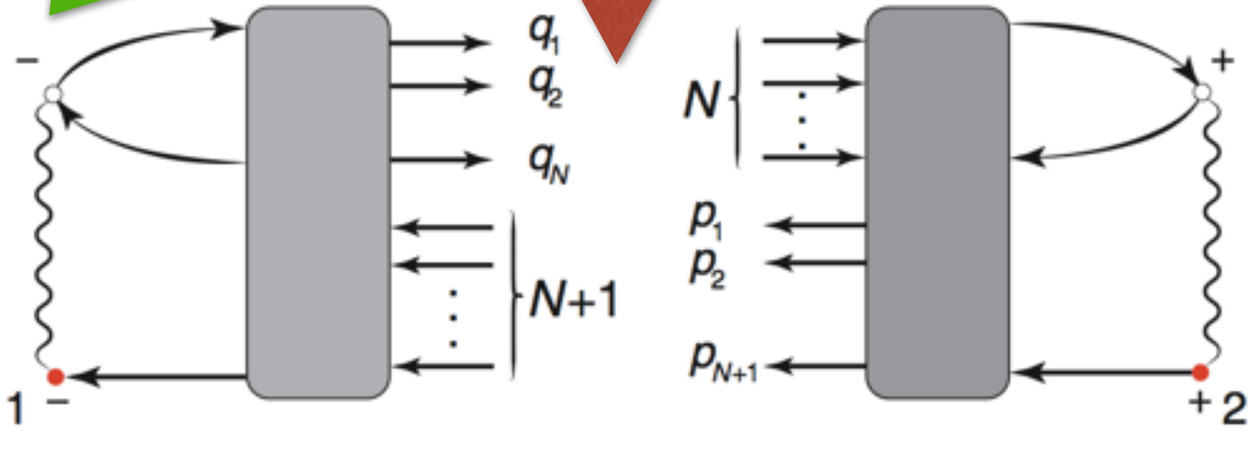
expanded in time-ordered Green's functions

$$S_{\underline{pq}}(2) = \frac{\langle\Phi_0|\bar{T}\left\{e^{i\int_{-\tau}^{\tau}dt\hat{H}(t)}\hat{\gamma}^\dagger(\mathbf{x}_2t_2)\hat{c}_{q_N}(\tau)\dots\hat{c}_{q_1}(\tau)\hat{c}^\dagger_{p_{N+1}}(\tau^+)\dots\hat{c}^\dagger_{p_1}(\tau^+)\right\}|\Phi_0\rangle}{\langle\Phi_0|\bar{T}\left\{e^{i\int_{-\tau}^{\tau}dt\hat{H}(t)}\right\}|\Phi_0\rangle}$$

expanded in anti-time-ordered Green's functions

latest time

time-ordered



anti-time-ordered

The in- and out-going lines of a half-diagram are particle or hole propagators

$$g^{--}_{q\mathbf{x}}(\tau,t_x) = g^{>}_{q\mathbf{x}}(\tau,t_x), \qquad g^{++}_{\mathbf{y}q}(t_y,\tau) = g^{>}_{\mathbf{y}q}(t_y,\tau),$$
$$g^{--}_{\mathbf{x}p}(t_x,\tau) = g^{<}_{\mathbf{x}p}(t_x,\tau), \qquad g^{++}_{p\mathbf{y}}(\tau,t_y) = g^{<}_{p\mathbf{y}}(\tau,t_y).$$

Bare propagators satisfy an idem-potency type relation

$$\sum_q g^{>}_{\mathbf{y}q}(t_y,\tau)g^{>}_{q\mathbf{x}}(\tau,t_x) = -ig^{>}_{\mathbf{yx}}(t_y,t_x),$$

$$\sum_p g^{<}_{\mathbf{x}p}(t_x,\tau)g^{<}_{p\mathbf{y}}(\tau,t_y) = ig^{<}_{\mathbf{xy}}(t_x,t_y).$$

In this way we can glue half-diagrams to obtain the usual self-energy diagrams

We still need to discard from the sum the reducible pieces, i.e. the diagrams that can be cut in two pieces by cutting a Green's function line. This leads to

$$-i\Sigma^{<}(1,2) = \sum_{N=1}^{\infty} \frac{1}{N!(N+1)!} \sum_{\underline{pq}} \tilde{S}_{N,\underline{pq}}(2)\tilde{S}^{*}_{N,\underline{pq}}(1)$$

irreducible half-diagrams

Taking Fourier transforms gives

$$-i\Sigma_c^<(1,2) = \sum_{N=1}^{\infty} \frac{1}{(N+1)!N!} \int \frac{d\omega}{2\pi} \frac{d\omega'}{2\pi} \times e^{-i\omega t_2 + i\omega' t_1} \sum_{\underline{pq}} \tilde{S}_{N,\underline{pq}}(\omega) \tilde{S}^*_{N,\underline{pq}}(\omega').$$

And therefore for this expression to depend on the time-difference we must have

$$\sum_{N=1}^{\infty} \frac{1}{(N+1)!N!} \sum_{\underline{pq}} \tilde{S}_{N,\underline{pq}}(\omega) \tilde{S}^*_{N,\underline{pq}}(\omega') = \mathcal{F}(\omega)\delta(\omega - \omega')$$

which shows that $\mathcal{F}(\omega) \geq 0$ and so

$$-i\Sigma^<(\omega) = \mathcal{F}(-\omega) \geq 0$$

We will rewrite the self-energy in more useful form for diagrammatic manipulations

$$\tilde{S}_{\underline{pq}} = \sum_{j\in I_N} \sum_{\substack{P\in\pi_{N+1}\\ Q\in\pi_N}} (-)^{P+Q} D^{(j)}_{\underline{pq}}$$

topologically equivalent half-diagrams

such that

$$-i\Sigma^{<}_{\rm c}(1,2) = \sum_{N=1}^{\infty} \sum_{j_1,j_2\in I_N} \sum_{\substack{P_1,P_2\in\pi_{N+1}\\ Q_1,Q_2\in\pi_N}} \frac{(-)^{P_1+Q_1+P_2+Q_2}}{(N+1)!N!} \sum_{\underline{pq}} D^{(j_2)}_{P_2(\underline{p})Q_2(\underline{q})}(2) D^{(j_1)*}_{P_1(\underline{p})Q_1(\underline{q})}(1)$$

Since

$$D^{(j_2)}_{P\circ P_2(\underline{p})Q\circ Q_2(\underline{q})}(2) D^{(j_1)*}_{P\circ P_1(\underline{p})Q\circ Q_1(\underline{q})}(1) = D^{(j_2)}_{P_2(\underline{p})Q_2(\underline{q})}(2) D^{(j_1)*}_{P_1(\underline{p})Q_1(\underline{q})}(1)$$

this can be rewritten as

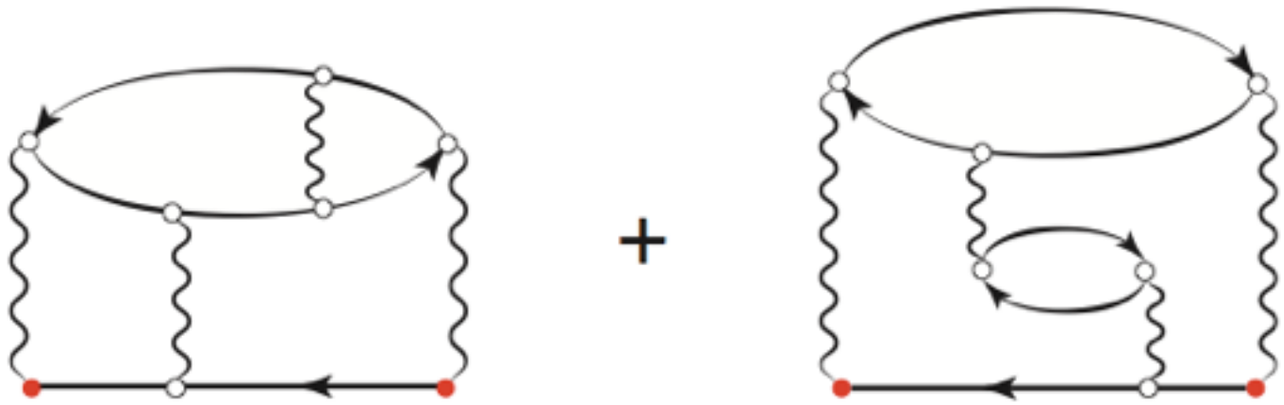
$$-i\Sigma^{<}_{\rm c}(1,2) = \sum_{N=1}^{\infty} \sum_{j_1,j_2\in I_N} \sum_{\substack{P_1\in\pi_{N+1}\\ Q_1\in\pi_N}} (-)^{P_1+Q_1} \sum_{\underline{pq}} D^{(j_2)}_{\underline{pq}}(2) D^{(j_1)*}_{P_1(\underline{p})Q_1(\underline{q})}(1)$$

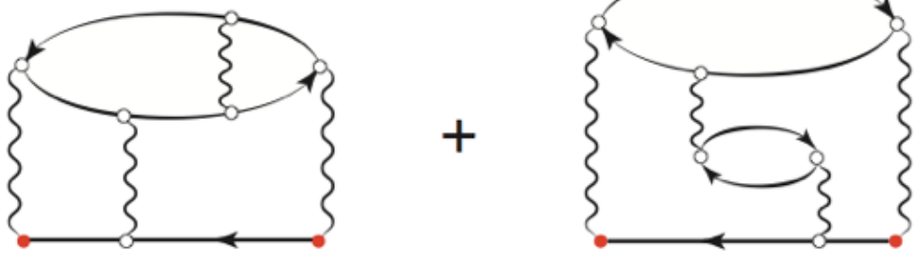
as there were N!(N+1)! relabelings with the same contribution

So far our derivation was exact, but for an approximate diagrammatic expression we have only a subset of the half-diagrams

The procedure is to judge whether such an approximation is PSD and, if not, extend it with a minimal set such that it is.

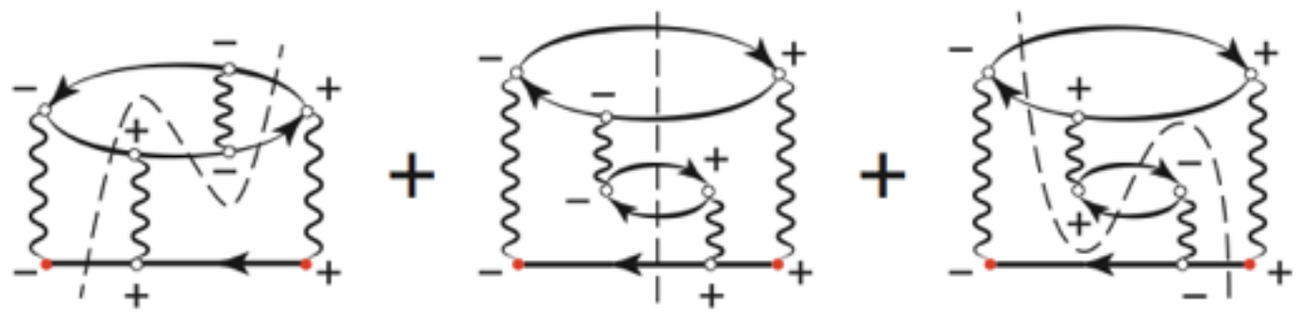
To illustrate the structure of such approximate terms we take the self-energy given by the two diagrams





$$\sum_{\underline{pq}} \Big[D^{(a)}_{p_1 p_2 p_3 q_1 q_2}(2) \Big(D^{(b)*}_{p_1 p_2 p_3 q_1 q_2}(1) - D^{(b)*}_{p_1 p_2 p_3 q_2 q_1}(1) \Big) - D^{(b)}_{p_1 p_2 p_3 q_1 q_2}(2) D^{(a)*}_{p_1 p_2 p_3 q_2 q_1}(1) \Big].$$

$$\Sigma_c^<(1,2) = i \sum_{N=1}^{\infty} \sum_{(j_1,j_2)\in\mathcal{I}_N} \sum_{\substack{P_1 \in \pi^{(j_1 j_2)}_{N+1,p} \\ Q_1 \in \pi^{(j_1 j_2)}_{N,q}}} (-)^{P_1+Q_1} \times \sum_{\underline{pq}} D^{(j_2)}_{\underline{pq}}(2) D^{(j_1)*}_{P_1(\underline{p})Q_1(\underline{q})}(1),$$



$N = 2$ 2 particle-hole pairs

$\mathcal{I}_N = \{(a,b),(b,a)\}$

$(j_1, j_2) = (b, a)$

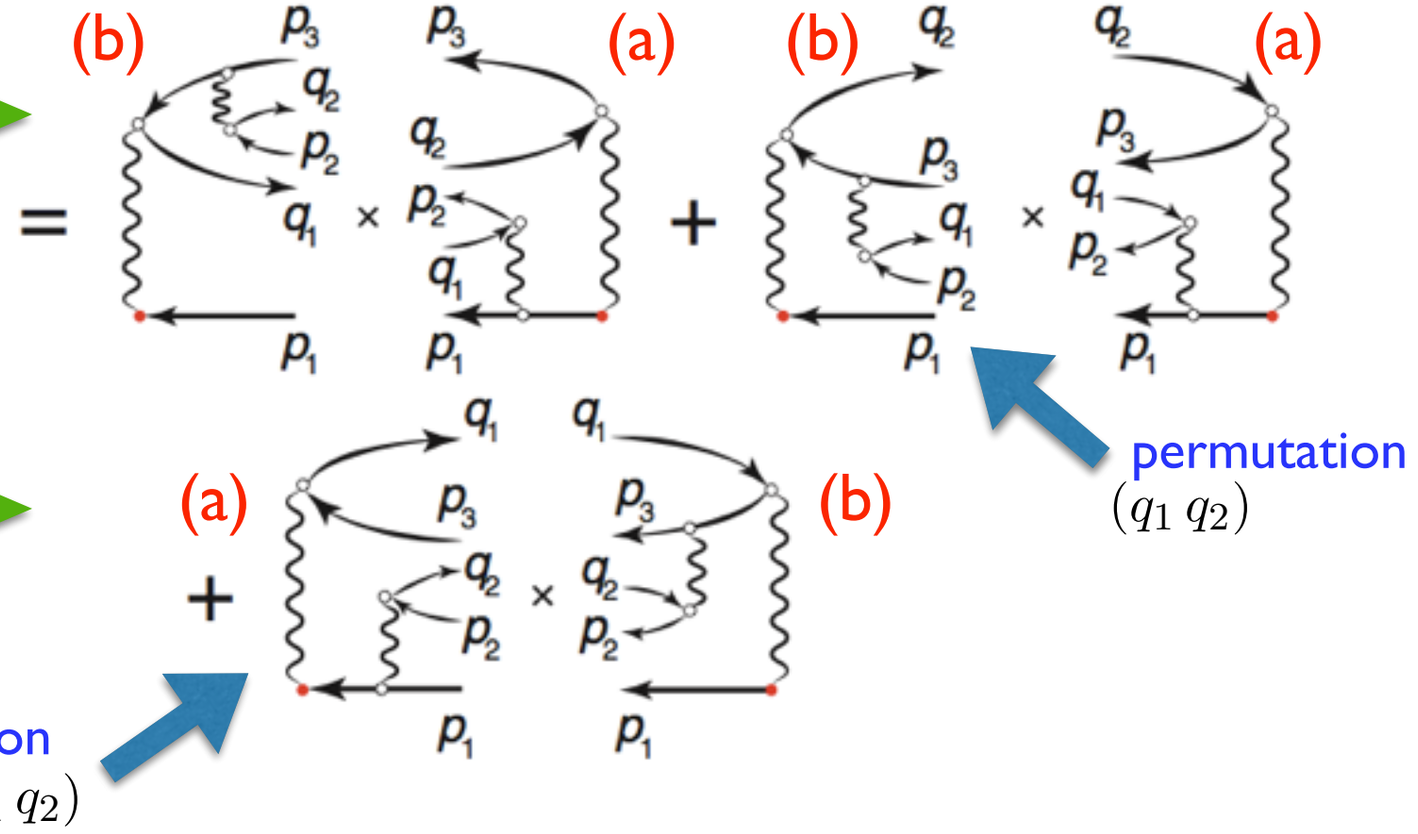
$\pi^{(b,a)}_{3,p} = \{1\}$

$\pi^{(b,a)}_{2,q} = \{1, (q_1\, q_2)\}$

$(j_1, j_2) = (a, b)$

$\pi^{(a,b)}_{3,p} = \{1\}$

$\pi^{(b,a)}_{2,q} = \{(q_1\, q_2)\}$



How to make this PSD?

We want to create a sum of products

$$\mathcal{I}_N = \{(a,b),(b,a)\}$$

$$\mathcal{I}_N \subset I_N \times I_N$$

$$I_N \times I_N = \{a,b\} \times \{a,b\} = \{(a,a),(a,b),(b,a),(b,b)\}$$

We take the smallest product set that contains our original set

$$\{1\} = \tilde{\pi}_{3,p} \supset \left\{\pi_{3,p}^{(b,a)}, \pi_{3,p}^{(a,b)}\right\} = \{1,1\} = \{1\}$$

$$\{1,(q_1\,q_2)\} = \tilde{\pi}_{2,q} \supset \left\{\pi_{2,q}^{(b,a)}, \pi_{2,q}^{(a,b)}\right\} = \{\{1,(q_1\,q_2)\},(q_1\,q_2)\} = \{1,(q_1\,q_2)\}$$

$$\tilde{\pi}_{N+1,p} \supset \bigcup_{(j_1,j_2)\in\mathcal{I}_N} \pi_{N+1,p}^{(j_1 j_2)},$$

$$\tilde{\pi}_{N,q} \supset \bigcup_{(j_1,j_2)\in\mathcal{I}_N} \pi_{N,q}^{(j_1 j_2)}.$$

Take the smallest subgroup of the permutation group such that

$$\Sigma^{<}_{c,\mathrm{PSD}}(1,2) = i \sum_{N=1}^{\infty} \sum_{j_1,j_2\in\tilde{I}_N} \sum_{\substack{P_1\in\tilde{\pi}_{N+1,p}\\ Q_1\in\tilde{\pi}_{N,q}}} (-)^{P_1+Q_1} \times \sum_{\underline{pq}} D^{(j_2)}_{\underline{pq}}(2) D^{(j_1)^*}_{P_1(\underline{p})Q_1(\underline{q})}(1)$$

The PSD property is more obvious when we write

$$-i\Sigma^{<}_{c,\mathrm{PSD}}(1,2) = \sum_{N=1}^{\infty} \sum_{j_1,j_2\in\tilde{I}_N} \sum_{\substack{P_1,P_2\in\tilde{\pi}_{N+1,p}\\ Q_1,Q_2\in\tilde{\pi}_{N,q}}} \frac{(-)^{P_1+Q_1+P_2+Q_2}}{d_{N+1,p}d_{N,q}} \sum_{\underline{pq}} D^{(j_2)}_{P_2(\underline{p})Q_2(\underline{q})}(2) D^{(j_1)*}_{P_1(\underline{p})Q_1(\underline{q})}(1)$$

particle-hole pairs

half-diagrams

permutations of half-diagram labels

labels of particle and hole states

$d_{N+1,p}$ and $d_{N,q}$ are the dimensions of the groups $\tilde{\pi}_{N+1,p}$ and $\tilde{\pi}_{N,q}$

Examples

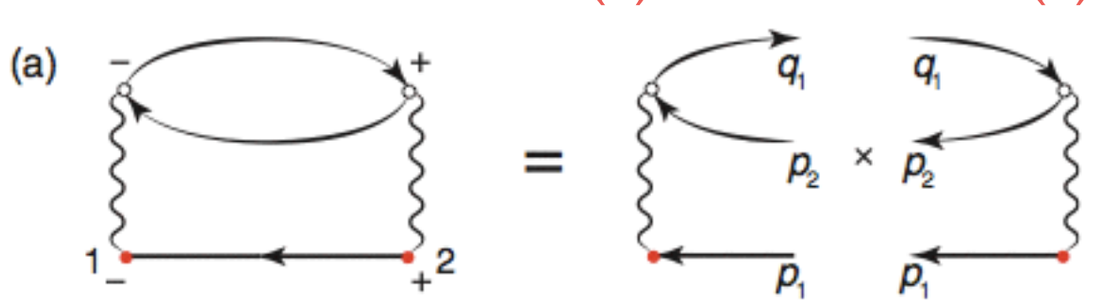
$N = 1$

$\mathcal{I}_2 = \{(a,a)\} = \{a\} \times \{a\}$

$\pi_{2,p}^{(a,a)} = \{1\}$

$\pi_{1,q}^{(a,a)} = \{1\}$

PSD



$N = 1$

$\mathcal{I}_2 = \{(a,a)\} = \{a\} \times \{a\}$

$\pi_{1,q}^{(a,a)} = \{1\}$

$\pi_{2,p}^{(a,a)} = \{(p_1\, p_2)\}$

$\{1\} = \tilde{\pi}_{1,q} \supset \left\{\pi_{1,q}^{(a,a)}\right\} = \{1\}$

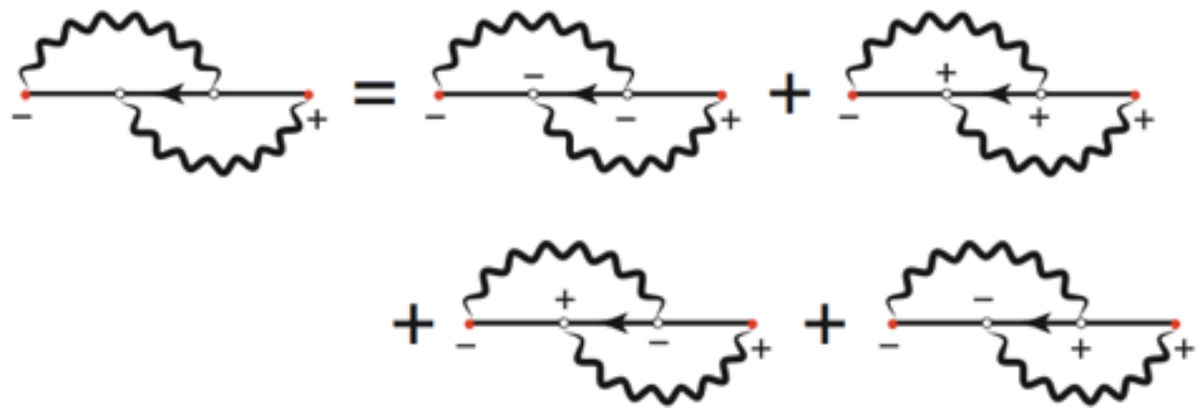
$\{1, (p_1\, p_2)\} = \tilde{\pi}_{2,p} \supset \left\{\pi_{2,p}^{(a,a)}\right\} = \{(p_1\, p_2)\}$

The 2nd Born approximation is PSD

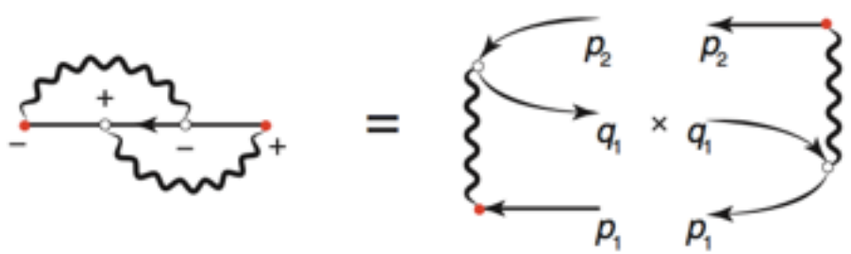
PSD

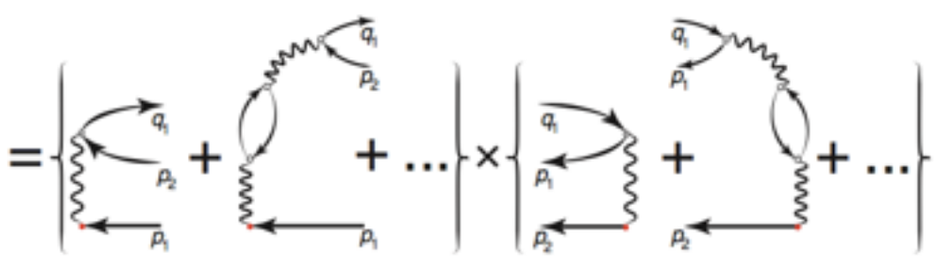
Making the vertex (GGGWW) positive

The spectral function of the vertex is given by

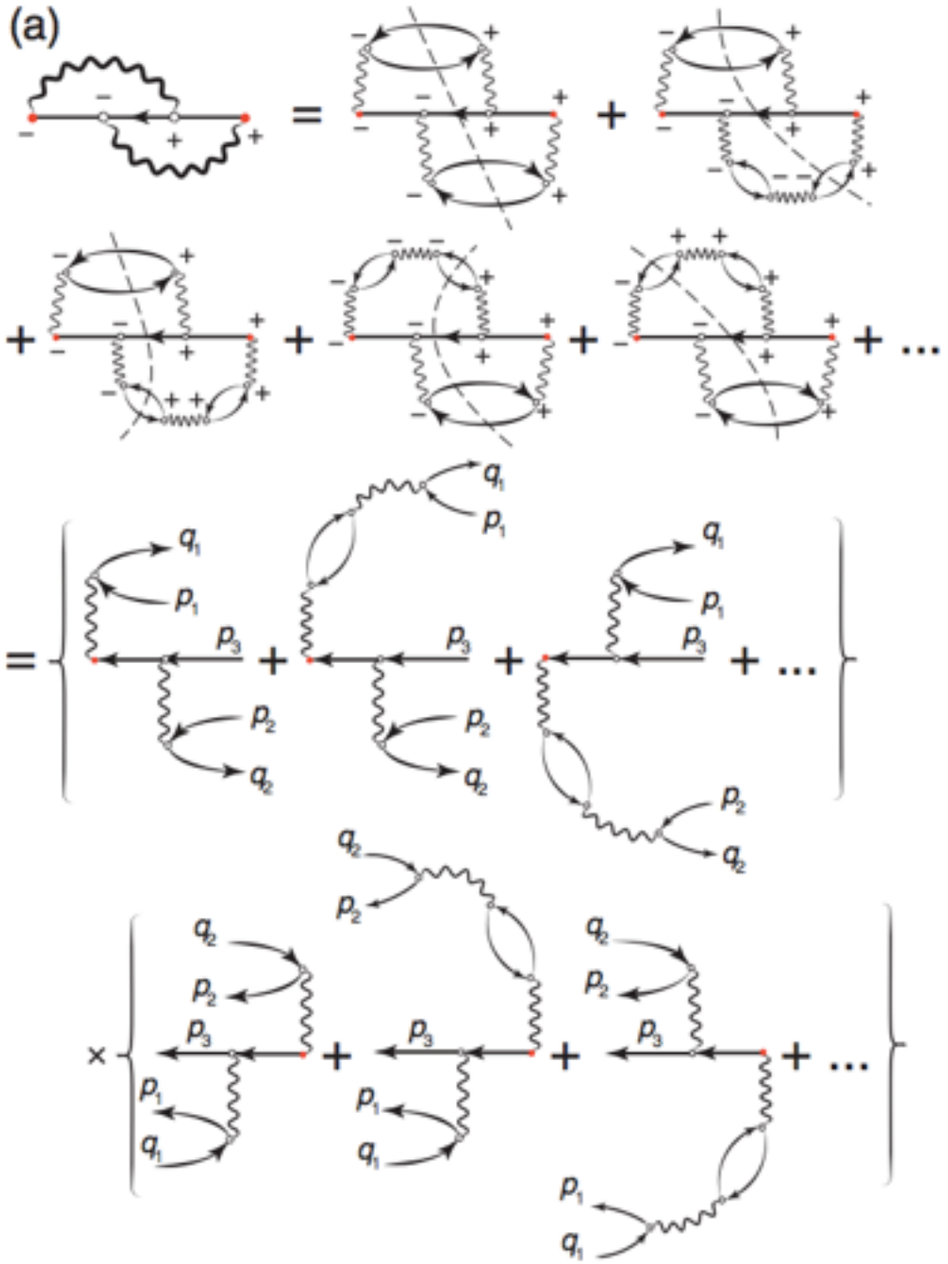


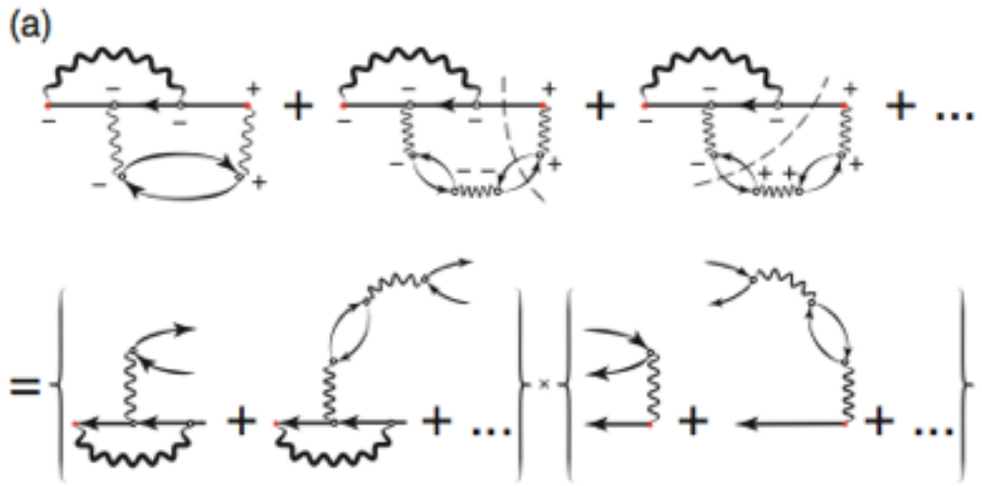
We can then write down all half-diagrams for this expressions. For example





.. and similarly for the other diagrams

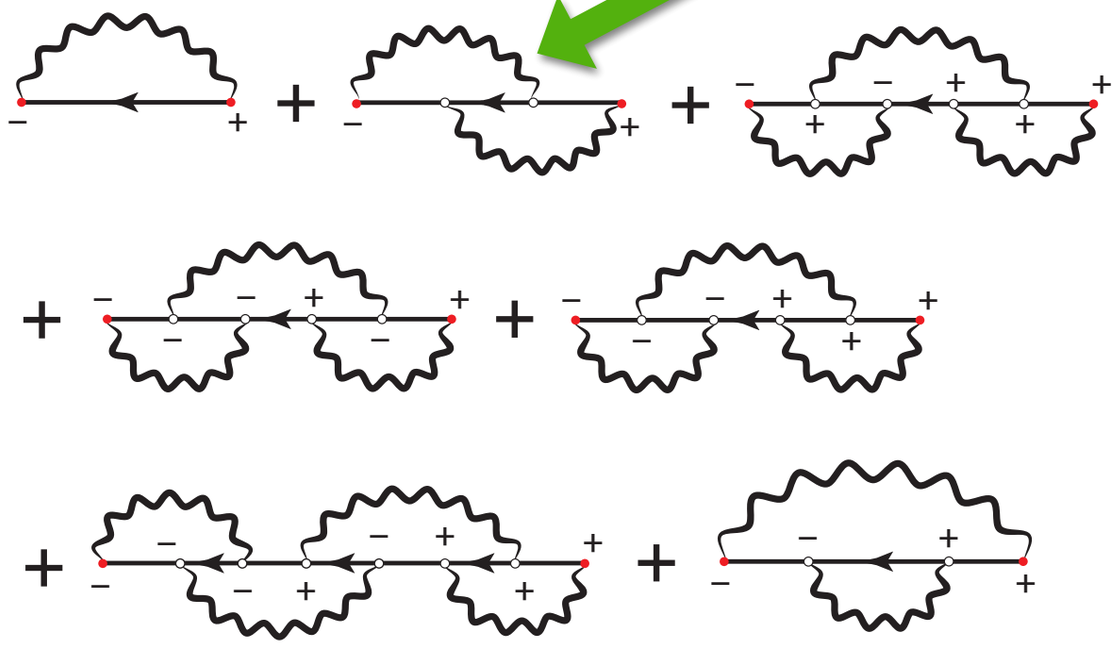

(a)
q_1
p_1
p_3
p_2
q_2


(a)

This gives the PSD vertex

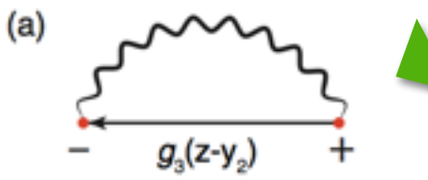
sum over internal +/- vertices

$$\Sigma^{<}_{\mathrm{PSD}} =$$

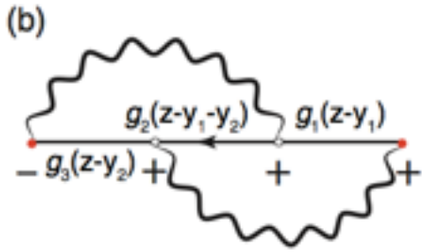


To make the vertex PSD we therefore need 3 partitions of a third order diagram and one partition of a fourth order diagram

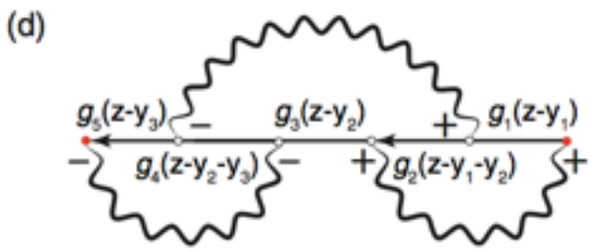
A simplified PSD vertex



GW



Two partitions of the vertex

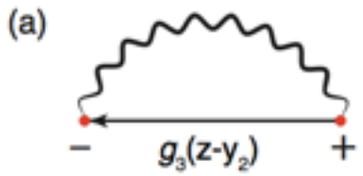


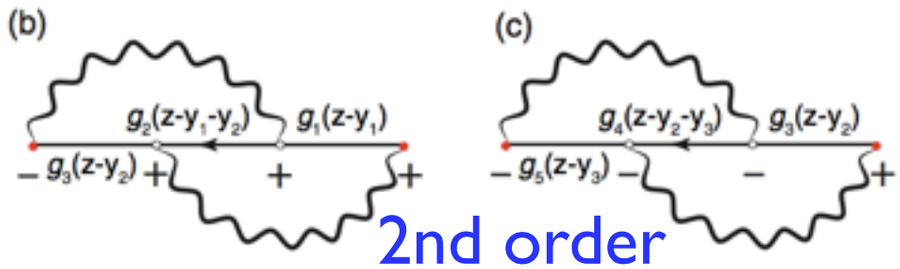
Addition diagram needed for PSD

To demonstrate the cutting rules we evaluate these diagrams for the electron gas using bare G's and W in plasmon pole approximation (the full-fledged vertex is in progress)

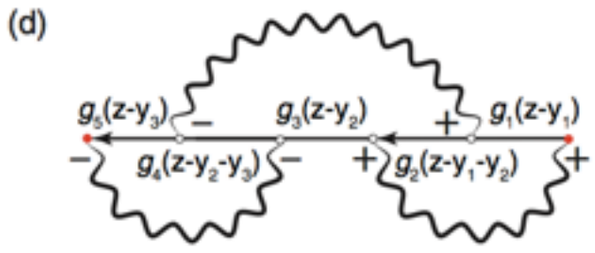
Numerical results: electron gas

1st order





2nd order

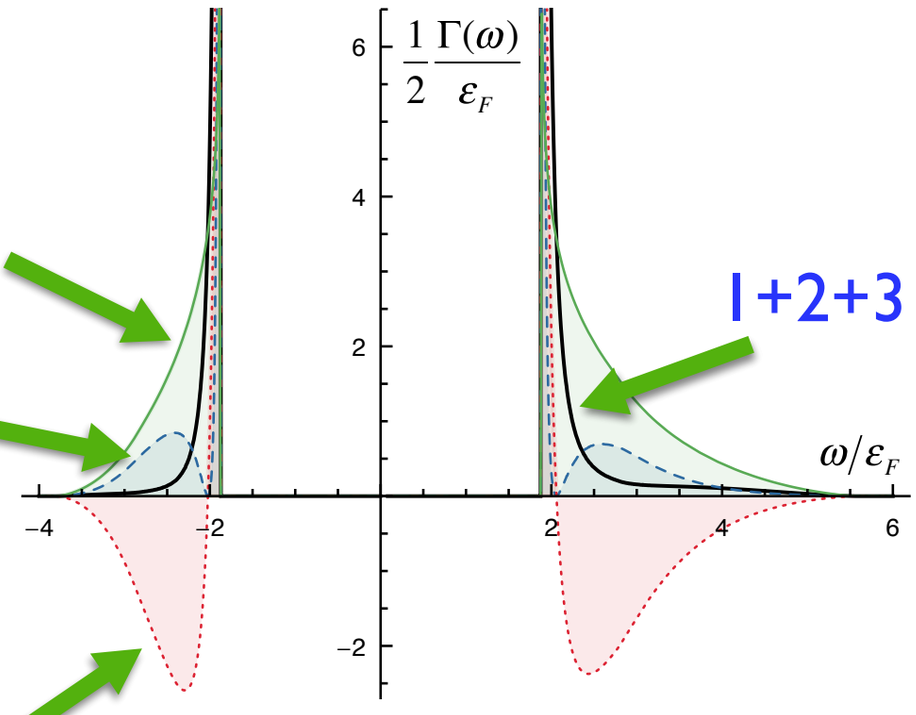


3rd order

$r_s = 4 \quad k = k_{\mathrm{F}}$

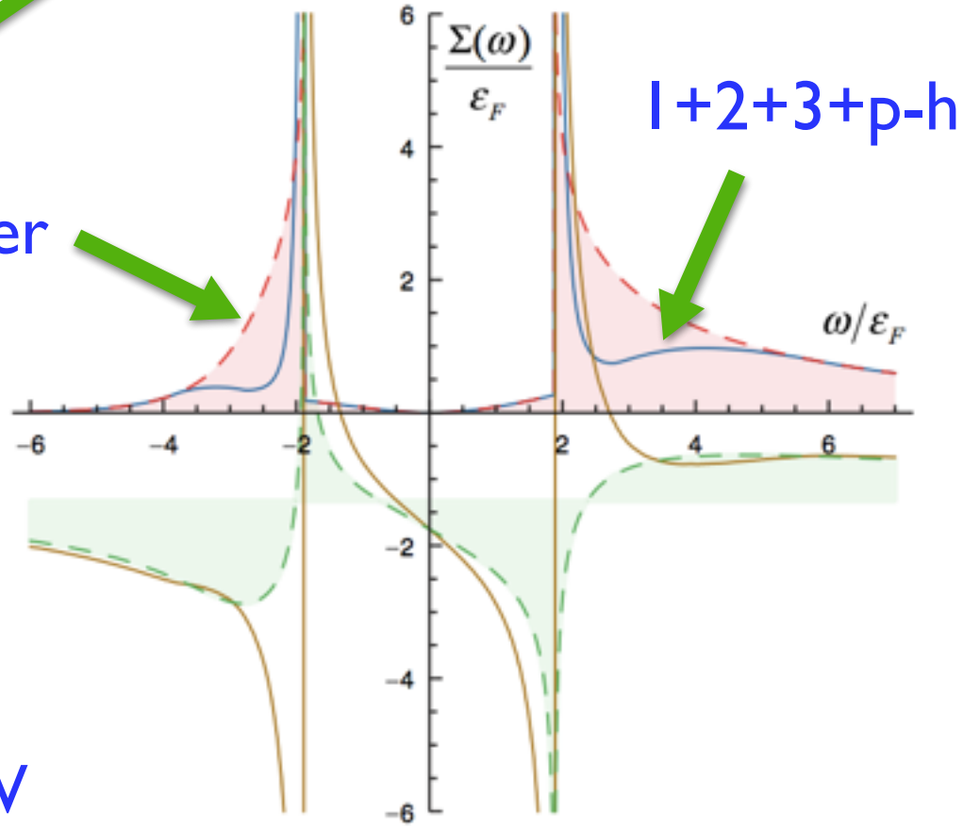
1st order

3rd order



1+2+3

2nd order



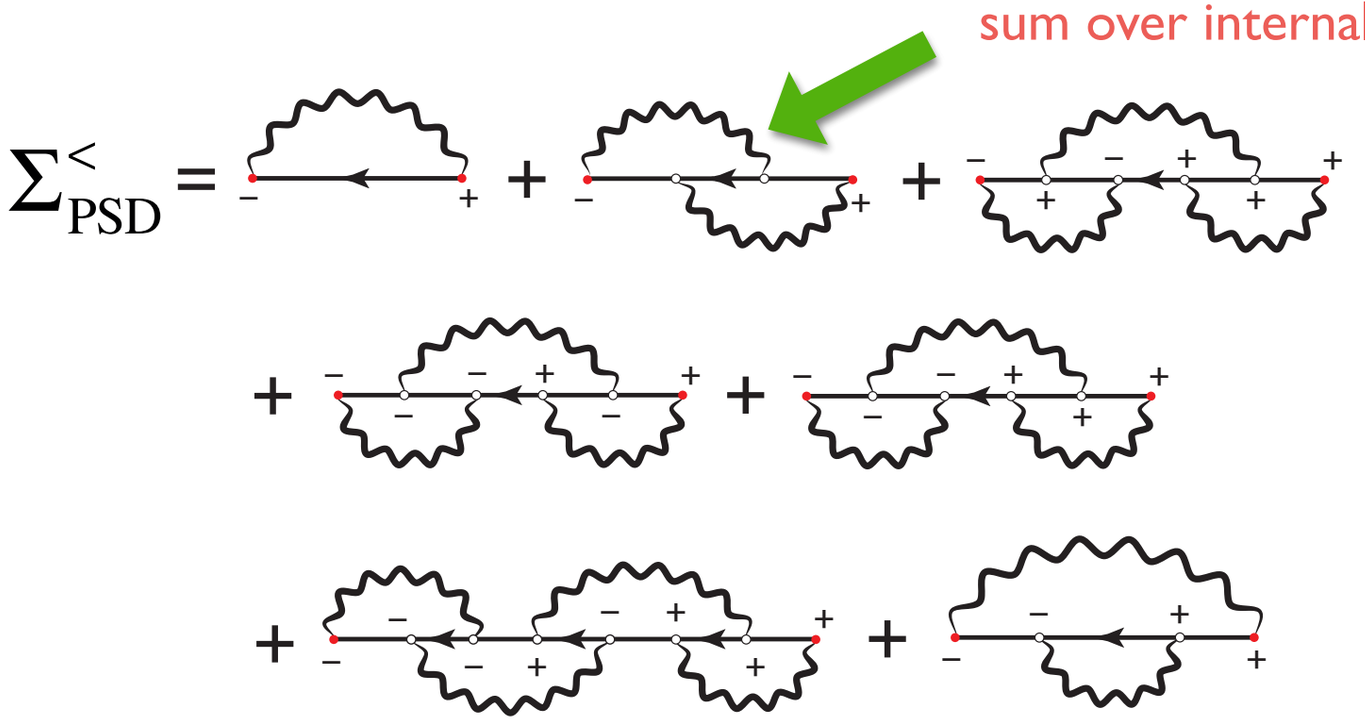
1st order

1+2+3+p-h

Here we also added
the p-h piece of GW

What about the full vertex beyond GW?

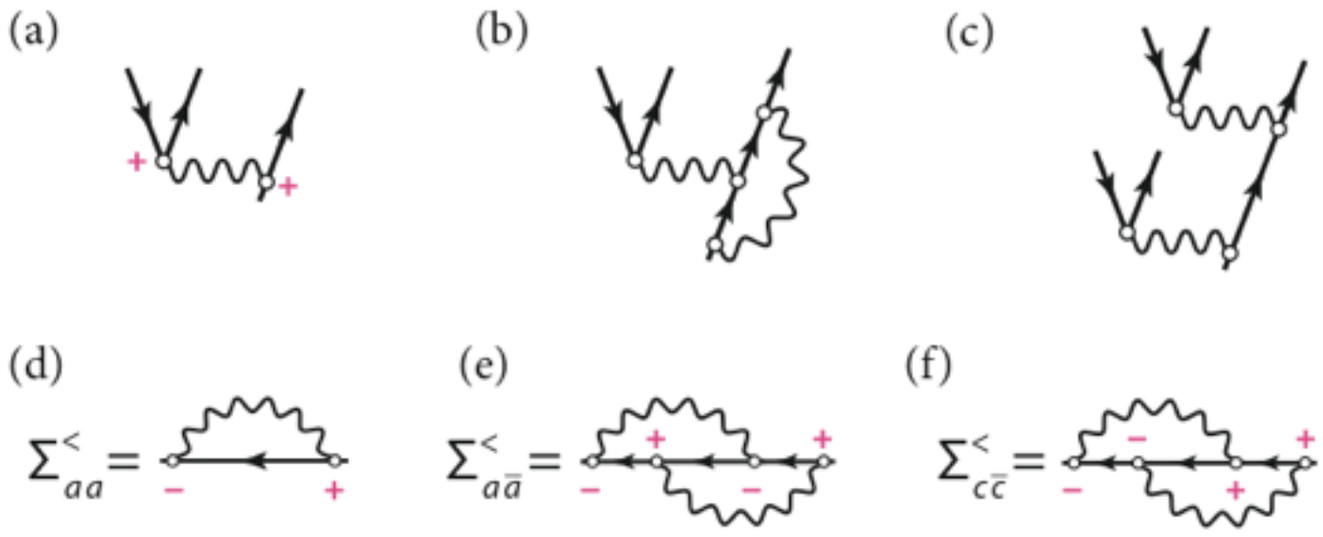
sum over internal +/- vertices

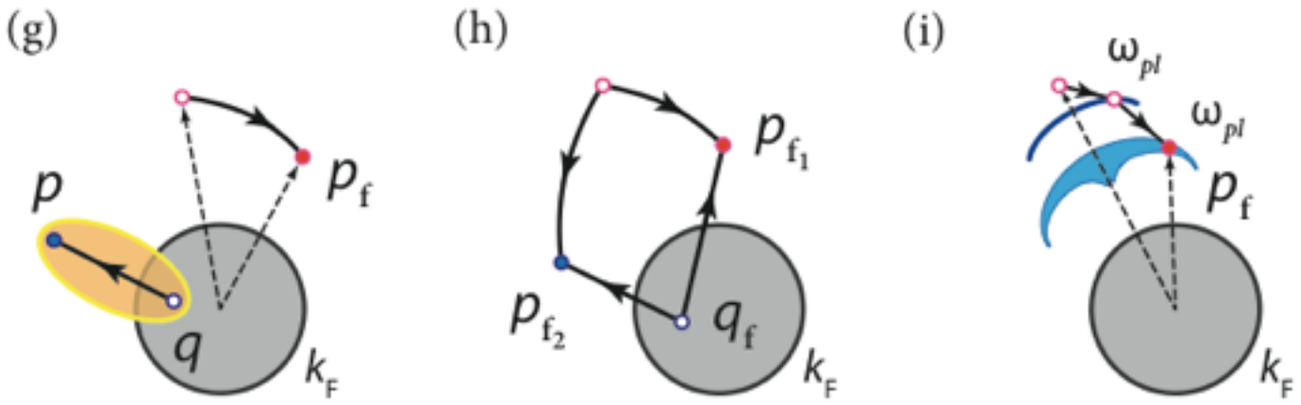


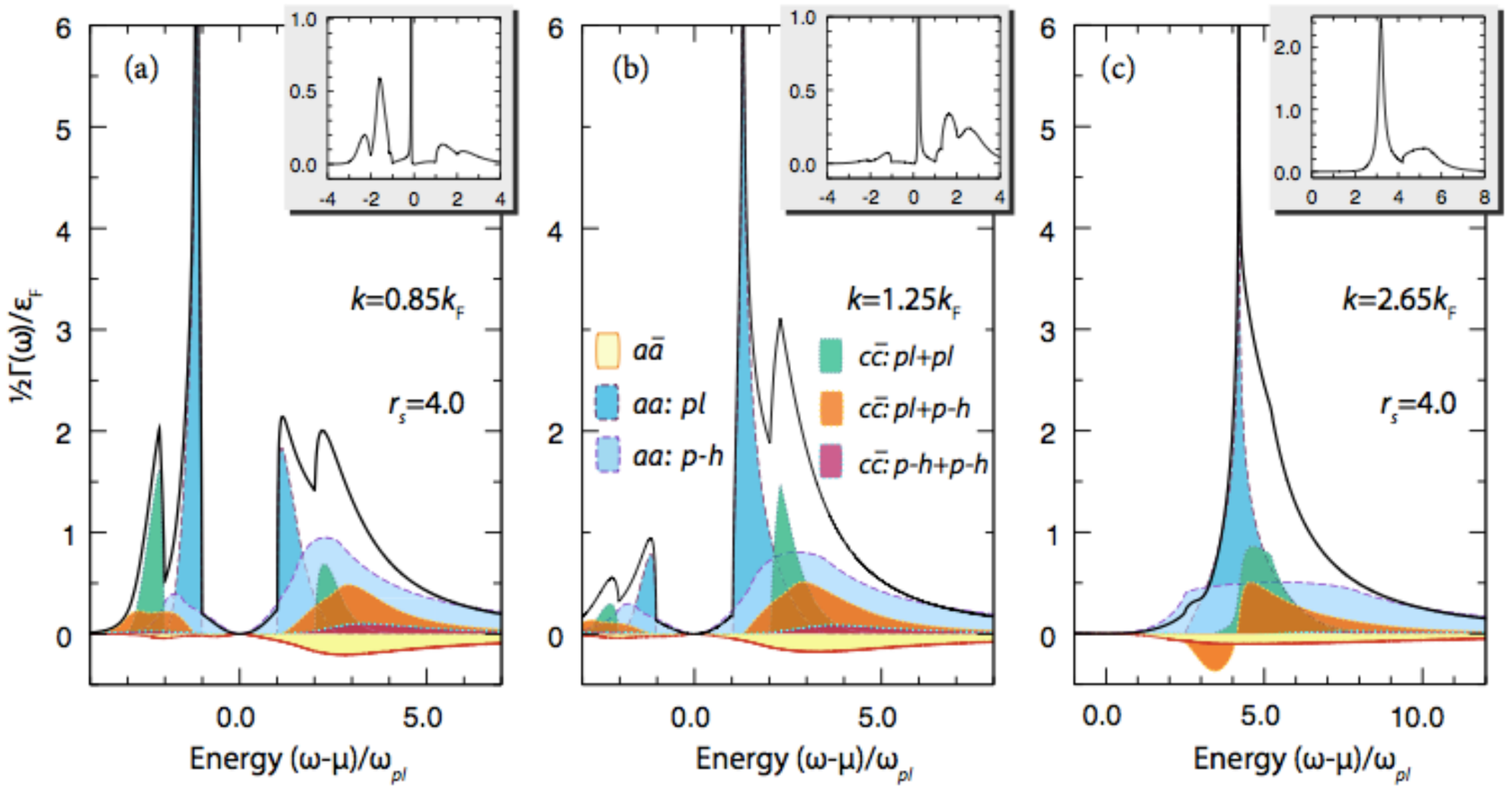
Even for the electron gas it is quite costly to calculate the higher order diagrams. We performed calculations with Green's functions from first iteration GW and found many cancellations..

Y.Pavlyukh, A.-M. Uimonen, G.Stefanucci, RvL,
Phys.Rev.Lett.117, 206402 (2016)

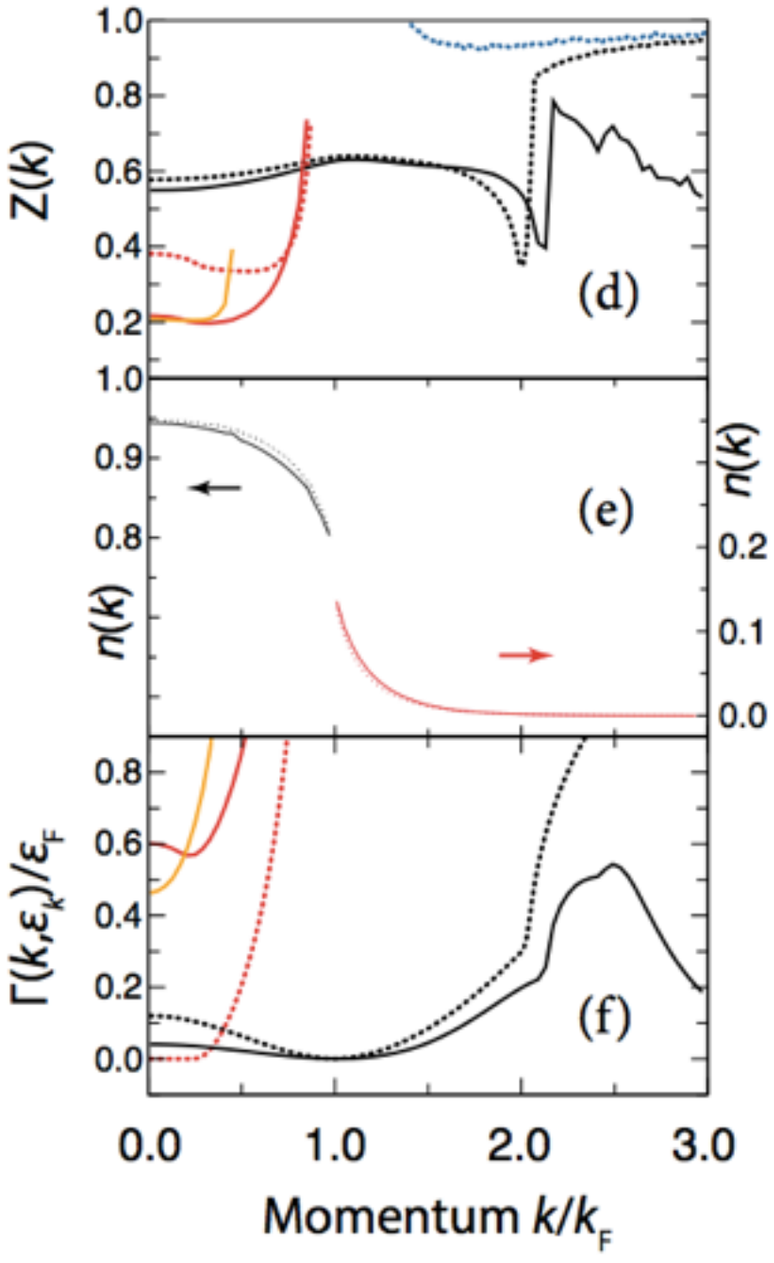
Dressing vs. vertices







Due to negative corrections around the chemical potential in the rate function, vertex corrections sharpen the quasi-particle peak as compared to G_0W_0

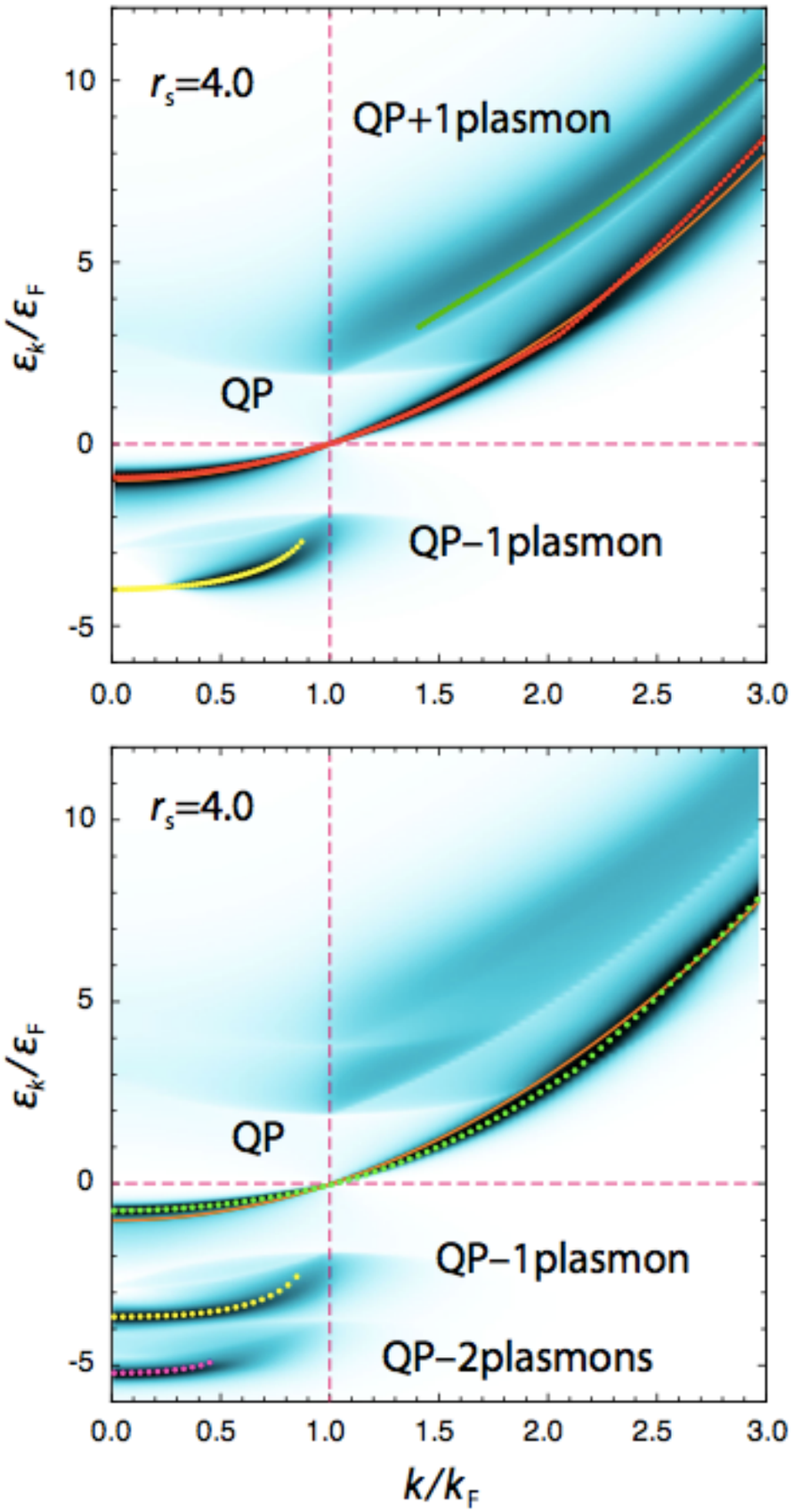


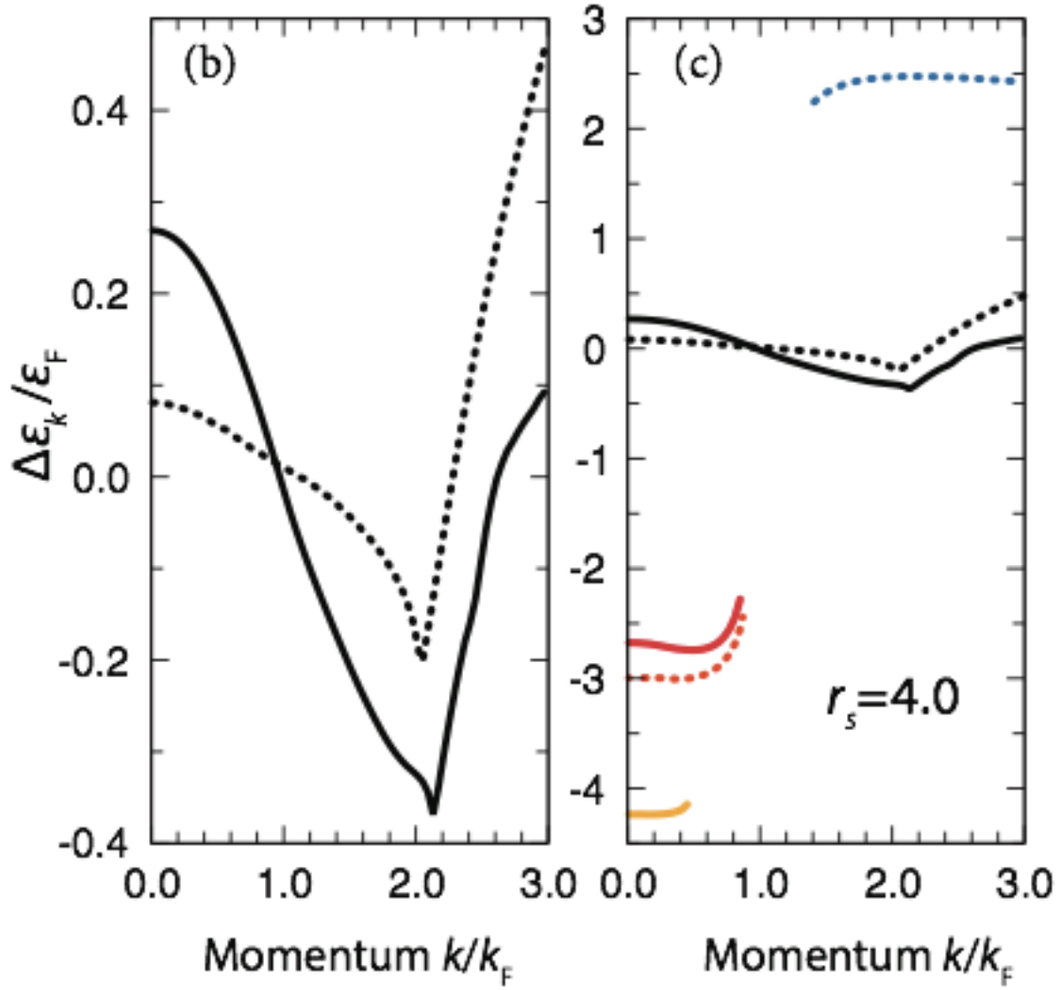
$$A(k,\omega) = Z^{(\alpha)}(k)\frac{1/\tau_k^{(\alpha)}}{(\omega-\omega_k^{(\alpha)})^2 + 1/(2\tau_k^{(\alpha)})^2}$$

$$Z^{(\alpha)}(k) = \left(1 - \frac{\partial}{\partial\omega}\,\mathrm{Re}\,\Sigma^{\mathrm{R}}(k,\omega)\Big|_{\omega=\omega_k^{(\alpha)}}\right)^{-1}$$

Vertex corrections do not change the momentum distribution and quasi-particle weight (Z-factor) much compared to G_0W_0 .The Z-factor is in good agreement with Quantum Monte Carlo results
(Holzmann et al. PRL 107, 110402 (2011))

Y.Pavlyukh, A.-M. Uimonen, G.Stefanucci, RvL, Phys.Rev.Lett.117, 206402 (2016)





Vertex corrections:

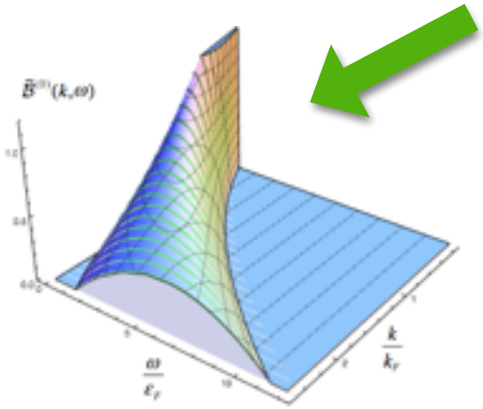
- Reduce the band width by 27 percent (sc GW increases by 20 percent
- Wash out the plasmon above the chemical potential
- Reduce the first plasmon energy

Polarisability beyond RPA

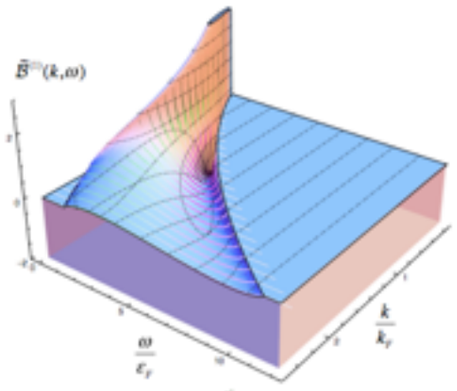
A.-M.Uimonen, G.Stefanucci, Y.Pavlyukh, RvL, PRB91,115104 (2015)

As a final remark we note that the same procedure can be done for the polarisability

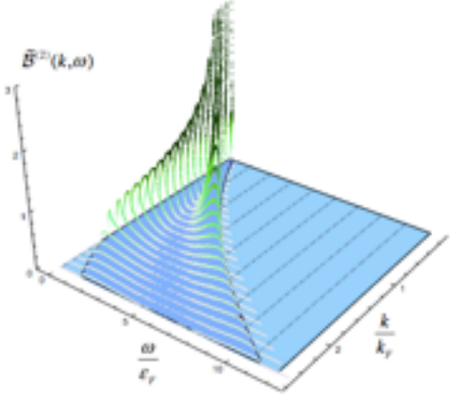
Lindhard function



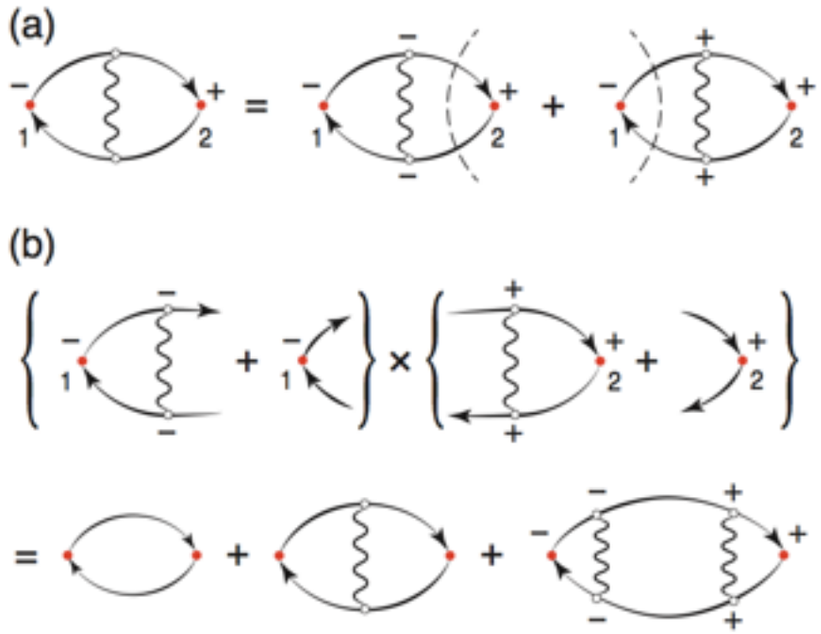
1



2



3



1 2 3

1+2 is not PSD

1+2+3 is PSD

Conclusions:

We developed a perturbation approach that guarantees the positivity of the spectral function

When applied to the homogeneous electron gas we found that there are cancellations between vertex corrections and dressing of the diagrams

We found that the simples PSD vertex beyond GW improves several features of the GW approach
(plasmon satellites, reduction bandwidth, reducing plasmon energy)

Things left out due to time:

- Cutting procedure with dressed Green's functions
- Relation between positivity and analyticity properties

D. Karlsson, RvL, Phys. Rev. B94, 125124 (2016)

Outlook

We are currently analysing in more detail some properties of the vertex corrections and the cancellations

It is possible to apply the PSD vertex we developed also for real systems and worked out the equations for doing Monte Carlo integrations

We also investigate in more detail the polarisability of the electron gas to investigate different approximations beyond RPA