Optical properties of interacting electronic systems: many-body versus time-dependent density-functional approach

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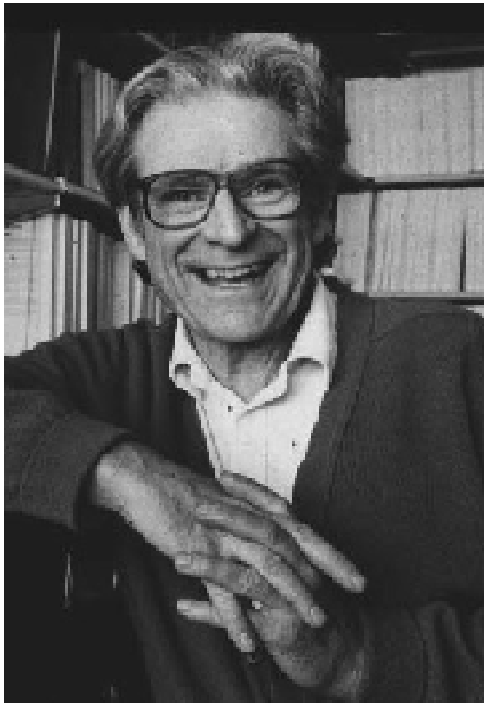
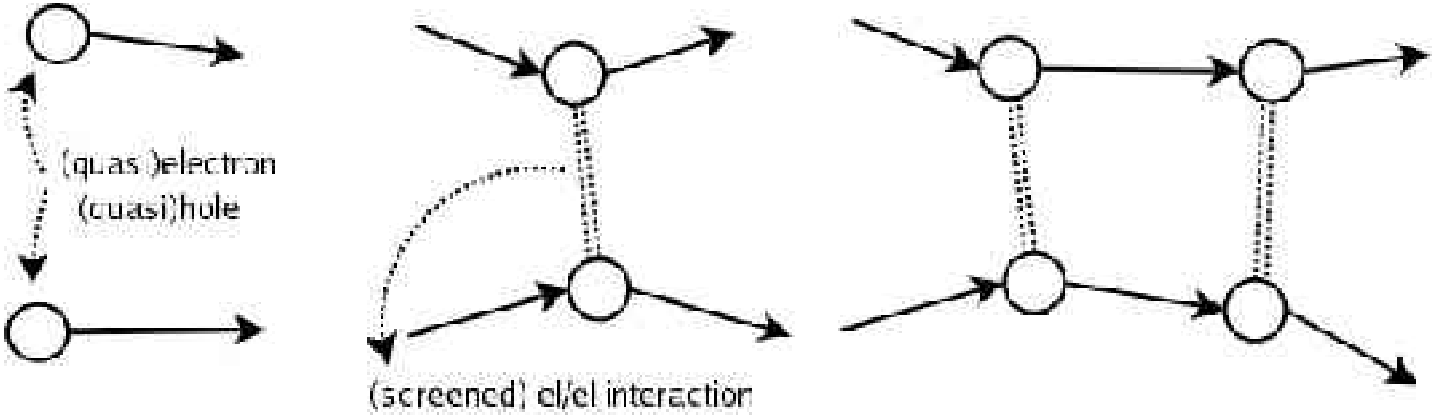
H.A. Bethe



E.E. Salpeter

The Bethe Salpeter equation (BSE): an equation for bound-state problems

[E.E. Salpeter and H.A. Bethe, Phys. Rev. **84**, 1232 (1951)]



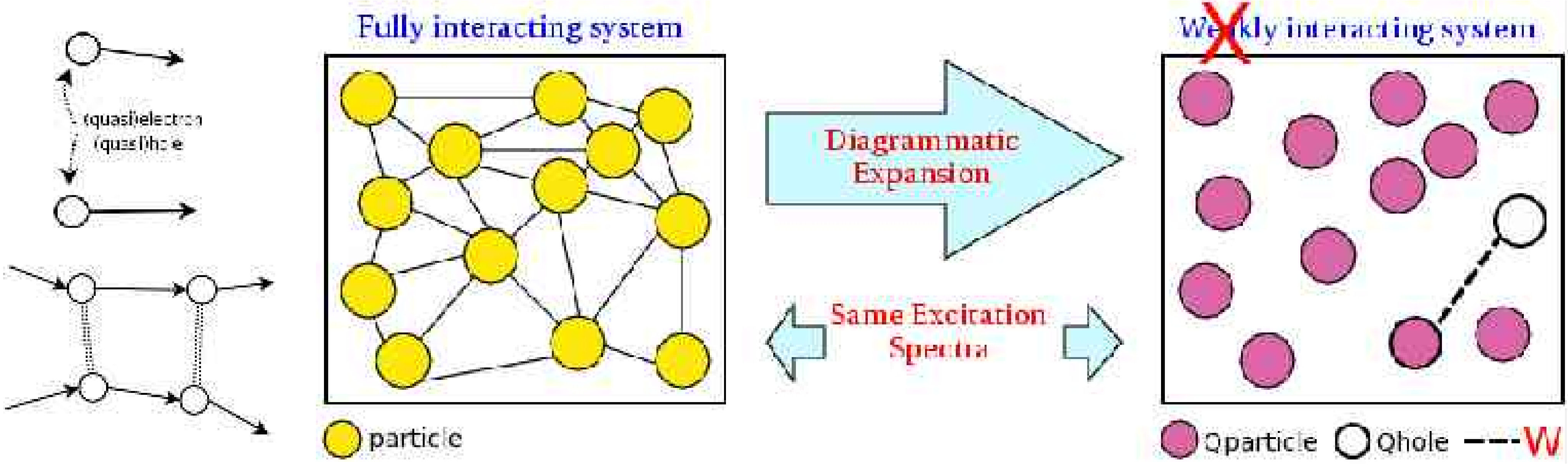
Part 1: Particles, quasiparticles and excitons: a brief introduction to BSE, Dyson equation and Time Dependent DFT.

Part 2: Bound excitons: *BSE* versus *Time-dependent Density-Functional Theory.*

Part 3: A *three-point vertex function* from TDDFT: lifetimes of a wide-gap insulator (LiF) beyond the GoWo approximation

Quasiparticles & Excitons

[F. Aryasetiawan, Rep. Prog. Phys. **61**, 237-312 (1998)
Onida, Reining, Rubio, Rev. Mod. Phys.**74**, 601 (2002)]



$$[\mathcal{H}_{KS} - V_{xc}(\mathbf{r})]\,\phi_\lambda(\mathbf{r}; E_\lambda) + \int d\mathbf{r}'\Sigma(\mathbf{r},\mathbf{r}'; E_\lambda)\,\phi_\lambda(\mathbf{r}'; E_\lambda) = E_\lambda\phi_\lambda(\mathbf{r}, E_\lambda)$$

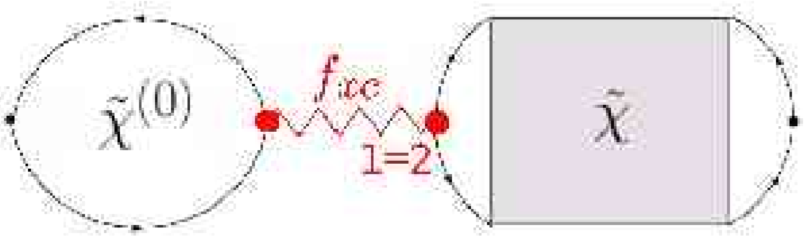
Dyson

$$\tilde{\chi}_{\mathbf{K}_1\mathbf{K}_2}(\omega) = i M^{-1}_{\mathbf{K}_1\mathbf{K}_2}(\omega) \qquad M_{\mathbf{K}_1\mathbf{K}_2}(\omega) = (\omega - \Omega_{\mathbf{K}_1})\,\delta_{\mathbf{K}_1\mathbf{K}_2} + W^{(RPA)}_{\mathbf{K}_1\mathbf{K}_2}$$

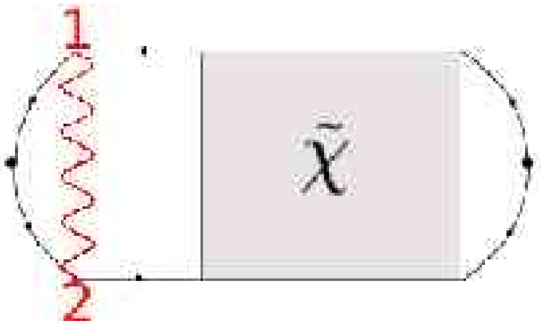
$$\mathbf{K}_1 \equiv \{c_1, v_1, \mathbf{k}_1\}$$

BSE

TDDFT



[E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52** (1984),
M. Petersilka, U. J. Gossmann, and E.K.U. Gross, Phys. Rev. Lett. **76** (1996)]



[AM, R. Del Sole, PRL**91**, 176402 (2003)]

Time Dependent DFT versus ...

$$\widehat{\chi}(\mathbf{q},\omega) = \widehat{\chi}^{(0)}(\mathbf{q},\omega) + \widehat{\chi}^{(0)}(\mathbf{q},\omega)\left[v_{\mathbf{q}} - \mathbf{f}_{xc}(\mathbf{q},\omega)\right]\widehat{\chi}(\mathbf{q},\omega)$$

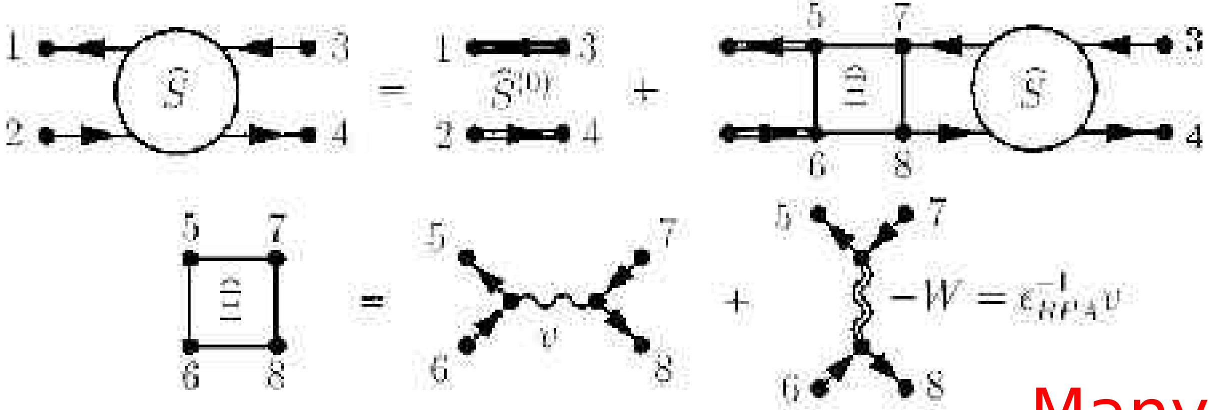
$$f_{xc}(\mathbf{r},t;\mathbf{r}'t') - \delta v_{xc}(\mathbf{r},t)/\delta\rho(\mathbf{r}',t')$$

$$\chi^{(0)}(\mathbf{r},\mathbf{r}';\omega) = \sum_{|ij\rangle}(f_i - f_j)\frac{\phi_i(\mathbf{r})\phi_j^*(\mathbf{r})\phi_i^*(\mathbf{r}')\phi_j(\mathbf{r}')}{\epsilon_i - \epsilon_j - \omega - i0^+}$$

Onida, Reining and Rubio, Rev. Mod. Phys. ***74****, 601 (2002)*

$$\widehat{\chi}(\mathbf{q},\omega) \propto \sum_{|ij\rangle|kl\rangle}\langle ij|\widehat{S}(\mathbf{q},\omega)|kl\rangle$$

$$\widehat{S}(\mathbf{q},\omega) = \widehat{S}^{(0)}(\mathbf{q},\omega) - \widehat{S}^{(0)}(\mathbf{q},\omega)\left[\widehat{V}(\mathbf{q}) - \widehat{W}(\mathbf{q})\right]\widehat{S}(\mathbf{q},\omega)$$



- xc-functional > vxc > fxc .

Sham Schluter, PRL **51**, 1888; Tokatly Pankratov PRL **86** 2078; Tokatly Stubner Pankratov PRB **65** 113107.

- EXX functional > vx > fx .

M. Stadele et al., PRL **79** 2089; Kim Gorling, PRL **89**, 096402;PRB **66** 035114.

- Long-range model.

Reining, Olevano, Rubio, Onida PRL **88**. 066404.

$$f_{xc} \sim \alpha/\mathbf{q}^2$$

- Sottile, Olevano, Reining, PRL **91**, 056402.

$$f_{xc} \approx W$$

- Density-based MBPT.

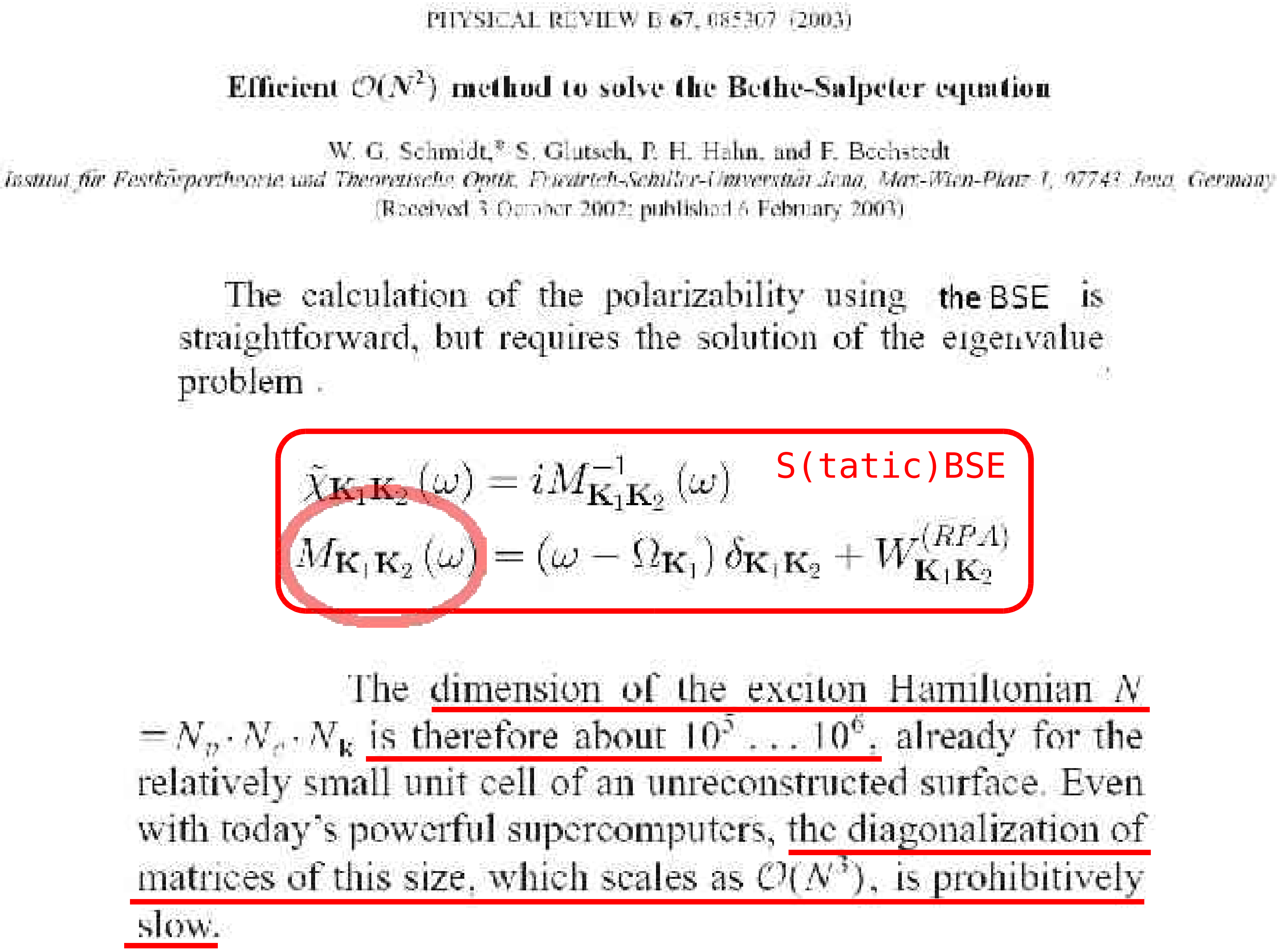
Bruneval, Olevano, Del Sole, Reining, PRL **94**, 186402.

- xc-functionals *a la'* TDDFT

von Barth Dahlen van Leeuwen Stefanucci cond-mat/0507604.

... Many-Body perturbation theory

The BSE: a O(NxN) problem



PHYSICAL REVIEW B **67**, 085307 (2003)

Efficient $\mathcal{O}(N^2)$ method to solve the Bethe-Salpeter equation

W. G. Schmidt,* S. Glutsch, P. H. Hahn, and F. Bechstedt

Institut für Festkörpertheorie und Theoretische Optik, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany

(Received 3 October 2002; published 6 February 2003)

The calculation of the polarizability using the BSE is straightforward, but requires the solution of the eigenvalue problem .

$$\tilde{\chi}_{\mathbf{K}_1\mathbf{K}_2}(\omega) = iM^{-1}_{\mathbf{K}_1\mathbf{K}_2}(\omega)$$

$$M_{\mathbf{K}_1\mathbf{K}_2}(\omega) = (\omega - \Omega_{\mathbf{K}_1})\,\delta_{\mathbf{K}_1\mathbf{K}_2} + W^{(RPA)}_{\mathbf{K}_1\mathbf{K}_2}$$

S(tatic)BSE

The dimension of the exciton Hamiltonian $N = N_v \cdot N_c \cdot N_{\mathbf{K}}$ is therefore about $10^5 \ldots 10^6$, already for the relatively small unit cell of an unreconstructed surface. Even with today's powerful supercomputers, the diagonalization of matrices of this size, which scales as $\mathcal{O}(N^3)$, is prohibitively slow.

Part 2: Bound excitons in Time-Dependent Density Functional Theory

- ***A many-body approach to the exchange-correlation kernel.***

- ***3D systems:Optical and Energy-Loss spectra of SiO2, LiF and diamond.***

A. Marini, R. Del Sole,and A. Rubio
Physical Review Letters ***91****, 256402 (2003)*

Many-Body approach to the Exchange-Correlation Kernel of TDDFT

$$\widehat{\chi}^{(0)}(\mathbf{q},\omega)+\sum_n \delta\widehat{\chi}^{(n)}(\mathbf{q},\omega) = \boxed{\widehat{\chi}^{BSE}(\mathbf{q},\omega) = \widehat{\chi}^{(0)}(\mathbf{q},\omega)+\widehat{\chi}^{(0)}(\mathbf{q},\omega)\left[v_{\mathbf{q}}+\mathbf{f}_{xc}(\mathbf{q},\omega)\right]\widehat{\chi}(\mathbf{q},\omega)}$$

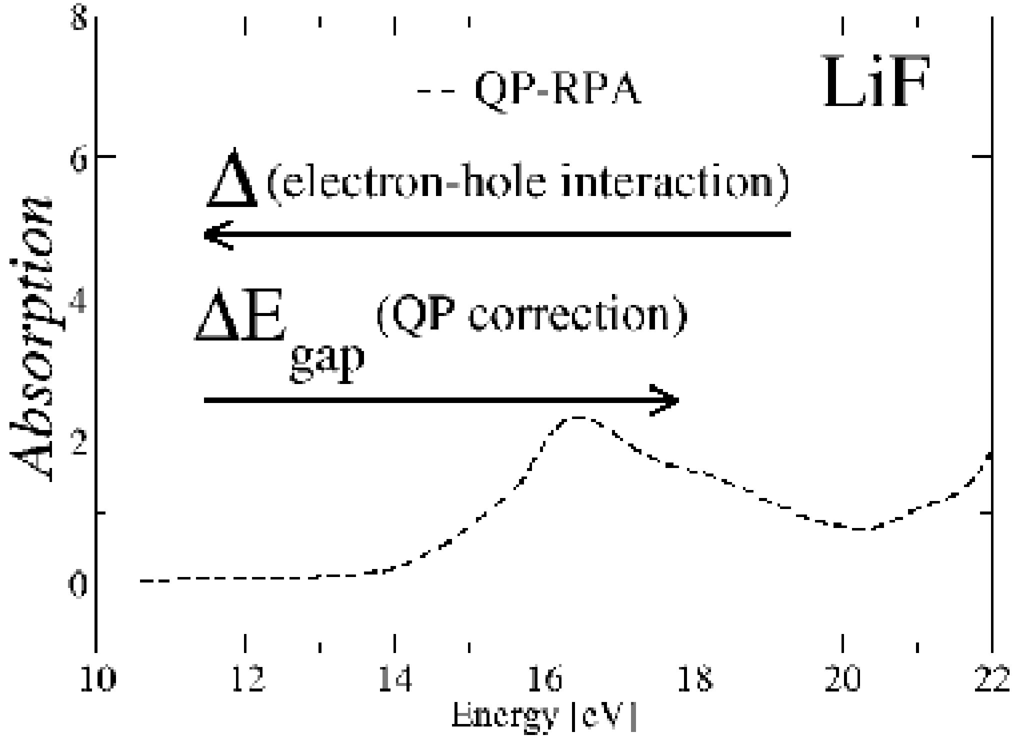
Hp. (1)

$$\mathbf{f}_{xc}(\mathbf{q},\omega) = \sum_n \mathbf{f}_{xc}^{(n)}(\mathbf{q},\omega)$$

$$\boxed{\widehat{\chi}_{DFT}^{(0)}(\mathbf{q},\omega) = \widehat{\chi}_{QP}^{(0)}(\mathbf{q},\omega)}$$

Hp. (2)

$$\boxed{\mathbf{f}_{xc}^{(n)}(\mathbf{q},\omega) = \left[\widehat{\chi}^{(0)}(\mathbf{q},\omega)\right]^{-1}\left[\delta\widehat{\chi}^{(n)}(\mathbf{q},\omega)\left(\widehat{\chi}^{(0)}(\mathbf{q},\omega)\right)^{-1} - \sum_{m=1,n-1}(-1)^m\,\delta\widehat{\chi}^{(m)}(\mathbf{q},\omega)\,\mathbf{f}_{xc}^{(n-m)}(\mathbf{q},\omega)\right]}$$



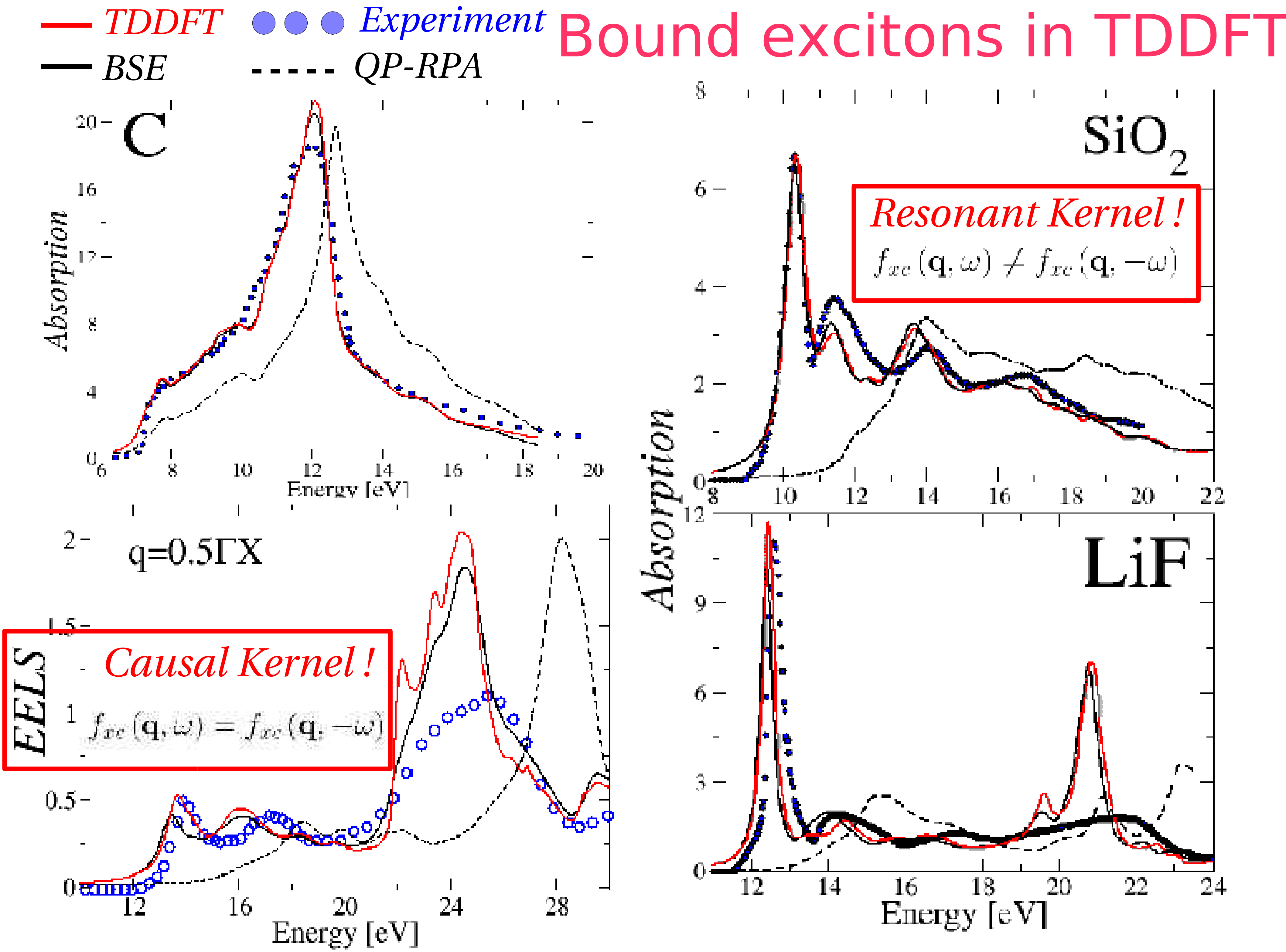
- The f_{xc} series *does not* converge unless the BS kernel diagonal is embodied in the IP polarization function, i.e.

$$\Delta = \langle ij|W(\mathbf{r},\mathbf{r}')|ij\rangle = 0$$

$$\widehat{\chi}^{(0)}(\mathbf{q},\omega) \longrightarrow \widehat{\chi}^{(0)}(\mathbf{q},\omega+\Delta)$$

Using this idea the higher order corrections to the first-order f_{xc} are not only well defined but numerically stable at all orders.

Bound excitons in TDDFT



Resonant Kernel !

$f_{xc}(\mathbf{q},\omega) \neq f_{xc}(\mathbf{q},-\omega)$

Causal Kernel !

$f_{xc}(\mathbf{q},\omega) = f_{xc}(\mathbf{q},-\omega)$

Schwäbisch Gmünd

Part 3: A Many-Body vertex function from TDDFT

- ***Two versus three-point vertex functions***

- ***A three-point vertex function from TDDFT***

- ***Excitonic effects on the lifetimes of wide gap insulators (LiF)***

A. Marini, and A. Rubio
Physical Review B ***70***, *081103(R) (2004)*

Polarization function, vertex & self-energy

$$\tau_{c\mathbf{k}}^{-1} \propto \sum_{\mathbf{G}_1,\mathbf{G}_2}\sum_{\mathbf{q},c'} \Omega_{c c'\mathbf{kq}}\left(\mathbf{G}_1,\mathbf{G}_2\right) Im\left[W^{TDDFT}_{\mathbf{G}_1\mathbf{G}_2}\left(\mathbf{q},\epsilon_{c\mathbf{k}}-\epsilon_{c'\mathbf{k}-\mathbf{q}}\right)\right]$$

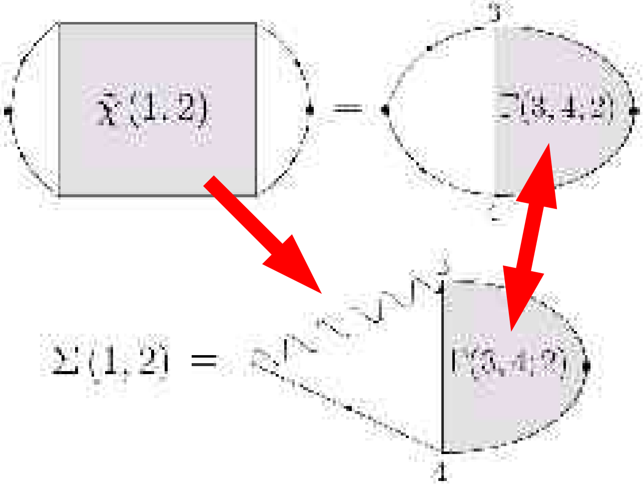
$$W^{TDDFT} = W \otimes \tilde{\Gamma}_{loc} = v\left[1-\left(v+f_{xc}\right)\chi_0\right]^{-1}$$

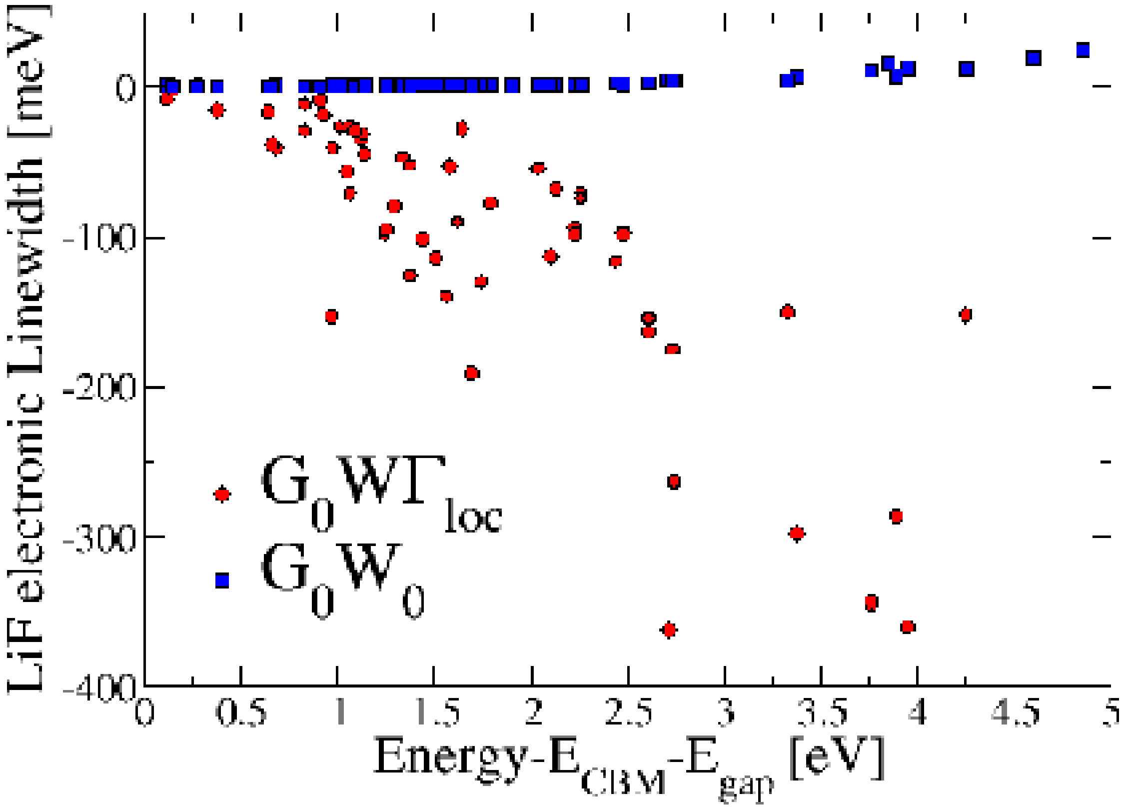
Small effect on the QPs using field corrections [PRL 62, 2718 (1989); PRB 49, 8024 (1994); PRB 56, 12832 (1997)]

$$\tilde{\chi}\left(1,2\right) = \chi_0\left(1,2\right) + \int d34\,\chi_0\left(1,3\right) f_{xc}\left(3,4\right)\tilde{\chi}\left(4,2\right)$$

$$\tilde{\Gamma}\left(1,2;3\right) = \delta\left(1,2\right)\tilde{\Gamma}_{loc}\left(1,3\right)$$

$$\tilde{\Gamma}_{loc}\left(1,2\right) = \left[\delta\left(1,2\right) - \int d3\, f_{xc}\left(1,3\right)\chi_0\left(3,2\right)\right]^{-1}$$

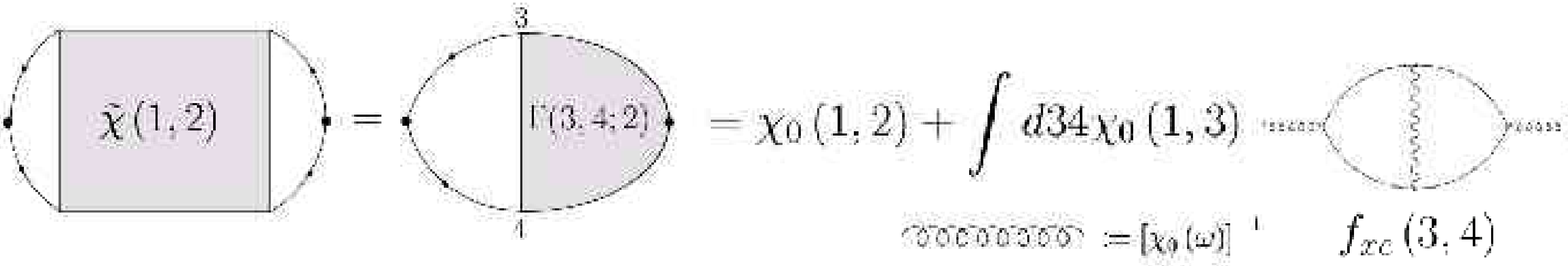




$$f_{xc}\left(\mathbf{r},\mathbf{r}';\omega\right) \approx -\frac{\alpha\left(\omega\right)}{\left|\mathbf{r}-\mathbf{r}'\right|}$$

$$-\left[1-\alpha\left(\omega\right)\right] > 1$$

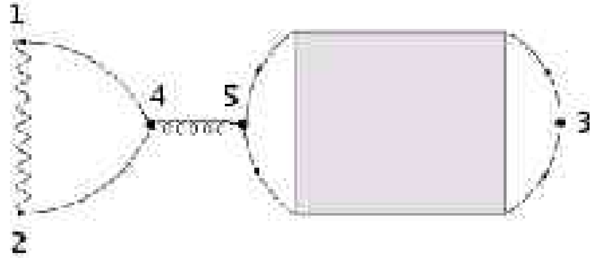
The 3-point vertex function

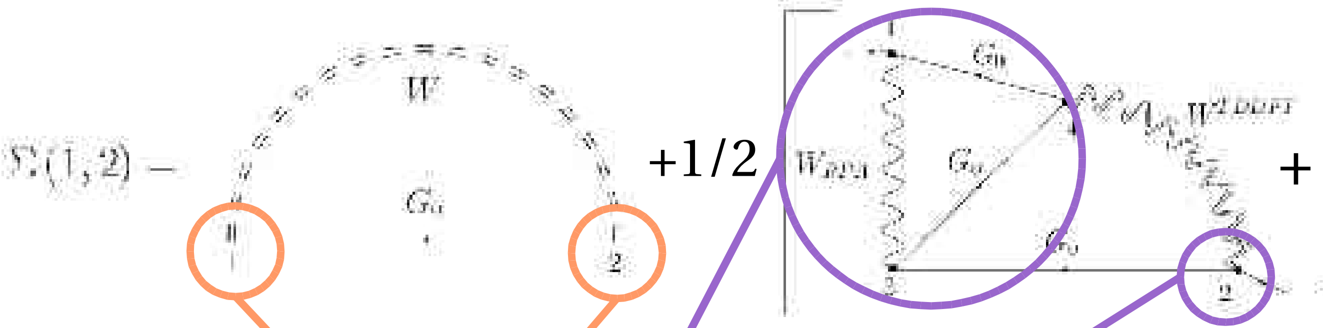


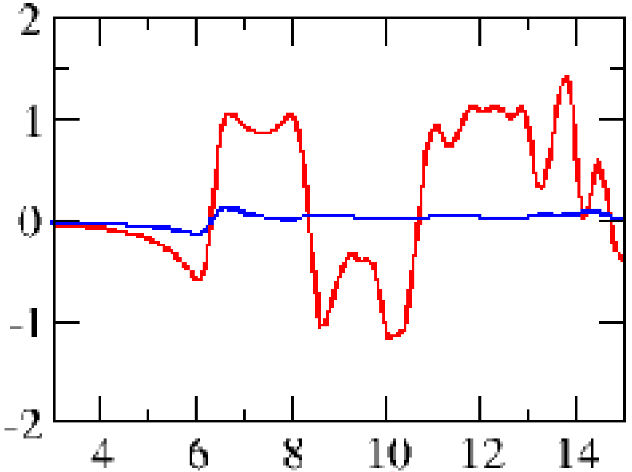
$$= \chi_0(1,2) + \int d34\, \chi_0(1,3) \cdots \tilde{\chi}(4,2)$$

$:= [\chi_0(\omega)]^{-1}$ $\quad f_{xc}(3,4)$

$$\widetilde{\Gamma}^{(1)}_{TDDFT}(1,2;3) \equiv \delta(1,2)\,\delta(2,3) - iW_0(1,2) \int d4\, G_0(1,4)\, G_0(4,2)\, \widetilde{\Gamma}_{loc}(4,3)$$





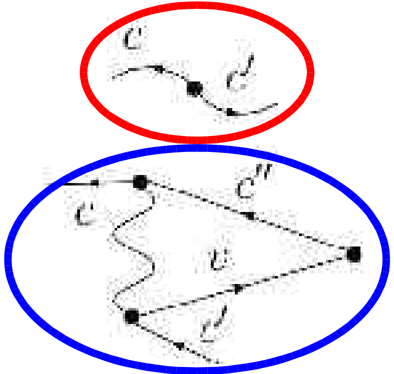


$$\tau^{-1}_{c\mathbf{k}} = \tau^{-1}_{c\mathbf{k},G_0W} + \Delta\tau^{-1}_{c\mathbf{k}}$$

$$\tau^{-1}_{c\mathbf{k},G_0W} \propto -\sum_{\mathbf{q},c'} \Omega_{cc'\mathbf{kq}} Im \left[W\left(\mathbf{q}, \epsilon_{c\mathbf{k}} - \epsilon_{c'\mathbf{k-q}}\right)\right]$$

$$\Delta\tau^{-1}_{c\mathbf{k}} \propto -\sum_{\mathbf{q},c'} \Gamma_{cc'\mathbf{kq}} Im \left[W^{TDDFT}\left(\mathbf{q}, \epsilon_{c\mathbf{k}} - \epsilon_{c'\mathbf{k-q}}\right)\right]$$

$$\frac{\Omega_{cc'}}{\Gamma_{cc'}} =$$



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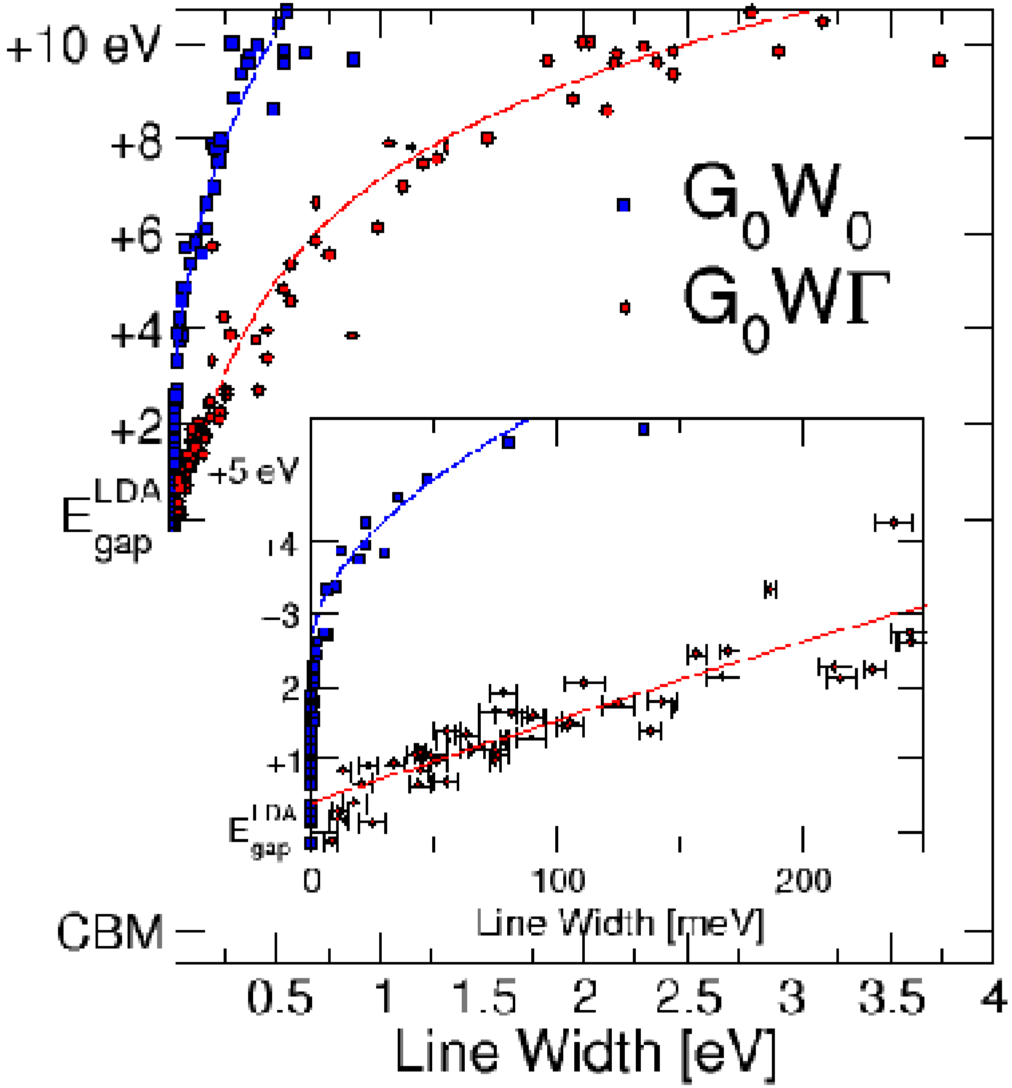
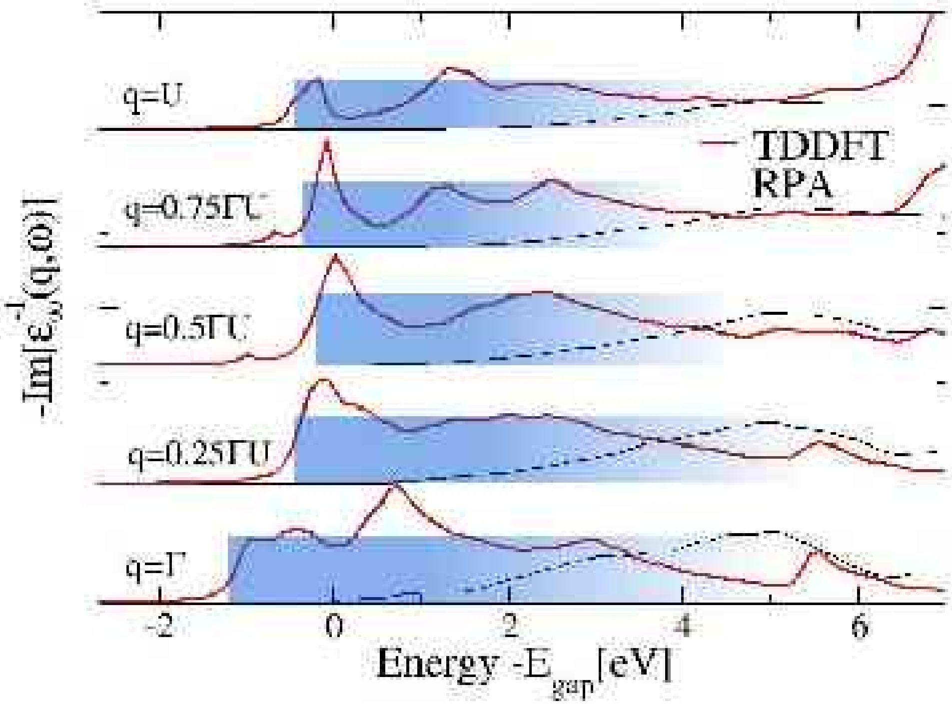
Electronic linewidths of LiF: excitonic effects





$$\tau^{-1}_{G_0W} \sim \sum_{c'\mathbf{q}} \Omega_{cc'\mathbf{kq}} Im\left[W\left(\epsilon_{c\mathbf{k}} - \epsilon_{c'\mathbf{k}-\mathbf{q}}\right)\right]$$

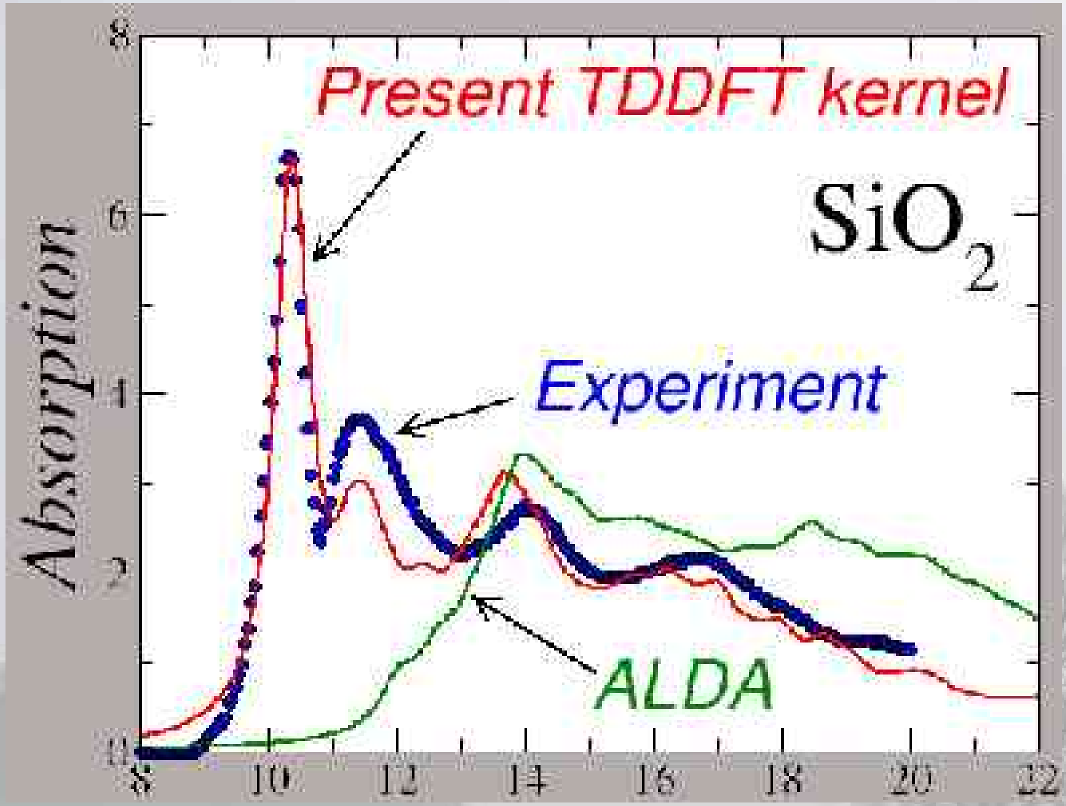
$$\tau^{-1}_{i,G_0W} \sim \int dE_f\, \overline{\Omega}_{if}\, \overline{\rho}\left(E_f\right) Im\left[W\left(E_f - E_i\right)\right]$$

$$\boxed{\tau^{-1}_{i,G_0W} \approx C_0\left(E_f - E_{CBM} - E_{gap}\right)}$$

Similar energy dependence has been observed in highly correlated materials [Phys. Today **54** (1), 29 (2001)] or layered materials (graphite) [PRL **87**, 246405 (2001)]

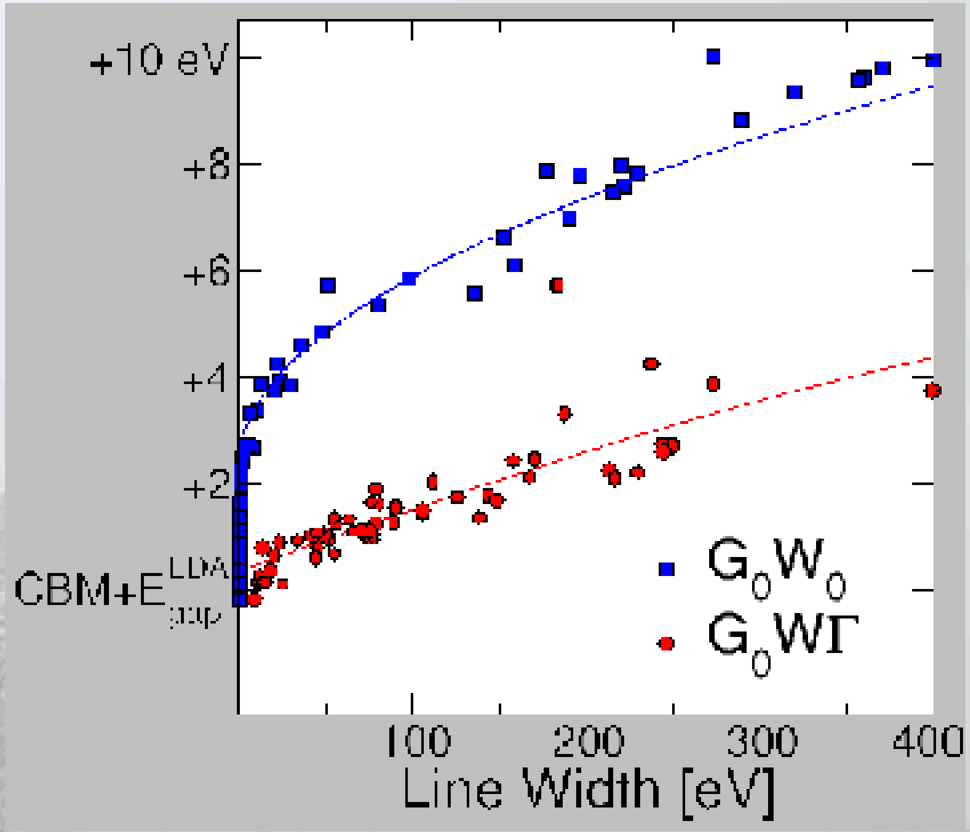
AM, and A. Rubio, Physical Review B ***70****, 081103(R) (2004)*

Conclusions



*AM, R. Del Sole, A. Rubio PRL**91**, 256402 (2003)*

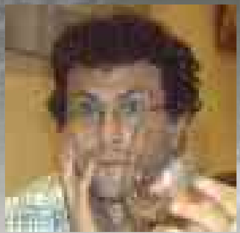
Strongly bound and resonant excitons in the optical spectra of wide-gap insulators can be described within TDDFT using a first-order, fxc kernel based on MBPT.

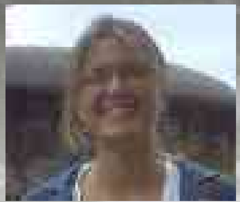


*AM, and A. Rubio, PRB**70**, 081103(R) (2004)*

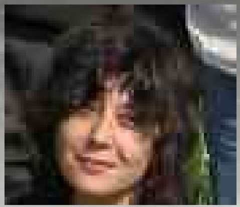
Anomalous linewidths energy dependence in LiF, explained in terms of strong excitonic effects in the self-energy vertex function.


Lucia Reining


Daniele Varsano


Myrta Gruening


Ludger Wirtz


Olivia Pulci