Exciton-Phonon coupling in the finite temperature optical absorption of semiconductor

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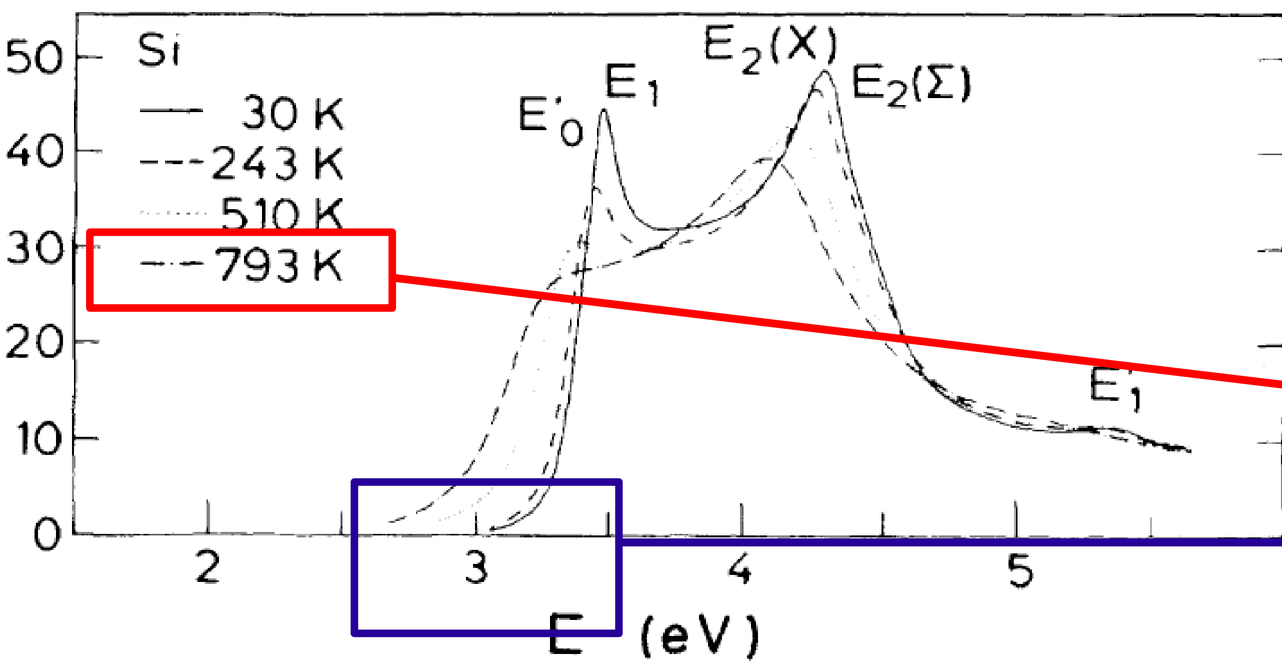
Do we need exciton-phonon coupling ?

PHYSICAL REVIEW B VOLUME 36, NUMBER 9 15 SEPTEMBER 1987-II

Temperature dependence of the dielectric function and interband critical points in silicon

P. Lautenschlager, M. Garriga, L. Viña,* and M. Cardona
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 30 April 1987)

The complex dielectric function $\epsilon(\omega)$ of Si was measured ellipsometrically in the 1.7–5.7-eV photon-energy range at temperatures between 30 and 820 K. The observed structures are analyzed by fitting the second-derivative spectrum $d^2\epsilon/d\omega^2$ with analytic critical-point line shapes. Results for the temperature dependence of the parameters of these critical points, labeled E_0', E_1, E_2, and E_1', are presented. The data show good agreement with microscopic calculations for the energy shift and the broadening of interband transitions with temperature based on the electron-phonon interaction. The character of the E_1 transitions in semiconductors is analyzed. We find that for Si and light III-V or II-VI compounds an excitonic line shape represents best the experimental data, whereas for Ge, α-Sn, and heavy III-V or II-VI compounds a two-dimensional critical point yields the best representation.



Debye Temperatures

Aluminium	426 K
Cadmium	186 K
Chromium	610 K
Copper	344.5 K
Gold	165 K
α-Iron	464 K
Lead	96 K
α-Manganese	476 K
Nickel	440 K
Platinum	240 K

Silicon	640 K
Silver	225 K
Tantalum	240 K
Tin (white)	195 K
Titanium	420 K
Tungsten	405 K
Zinc	300 K
Diamond	2200 K
Ice	192 K

wikipedia

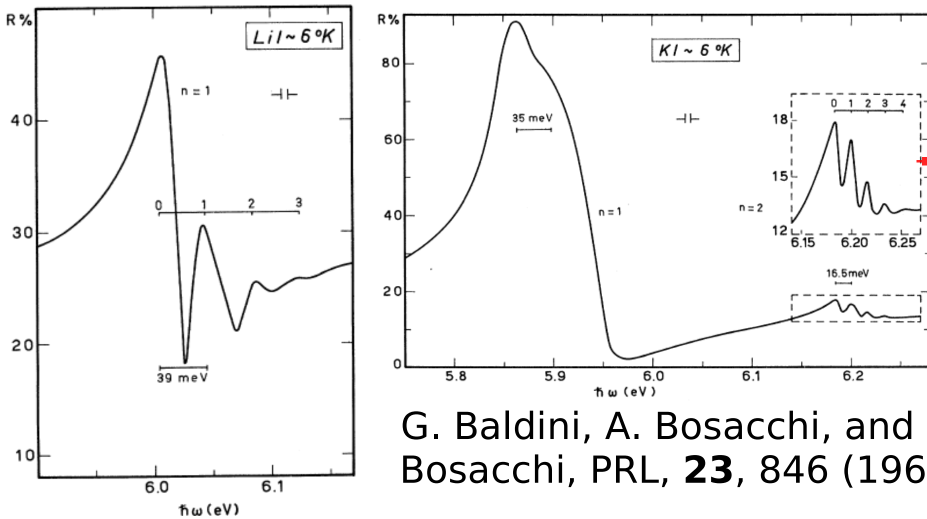
793 K = 68 meV
QP gap is 1200 meV

At the absorption treshold the QP lifetime is EXACTLY infinte

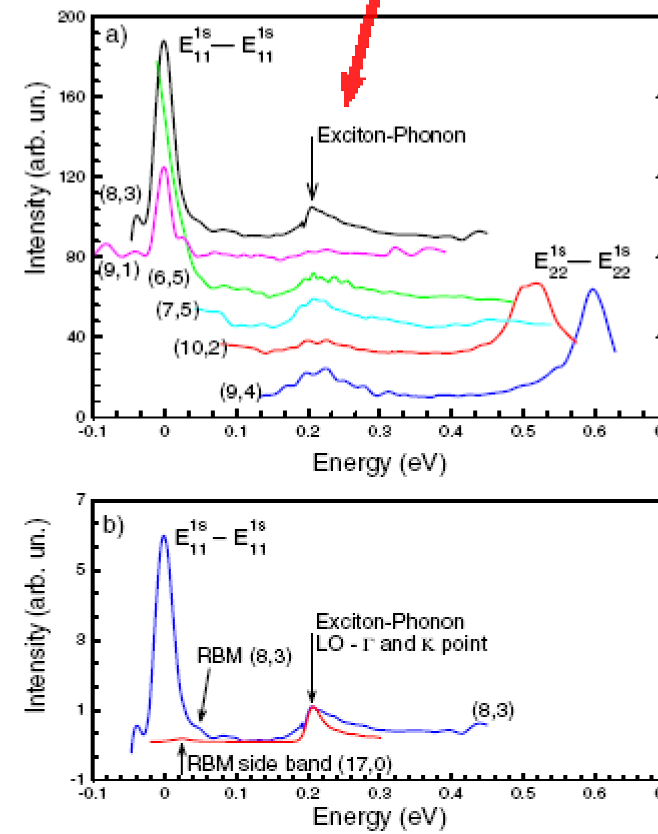
YES ! At least we do need phonons.

Are you sure we need exciton-phonon coupling ?

YES ! Because it is possible to observe phonon sidebands

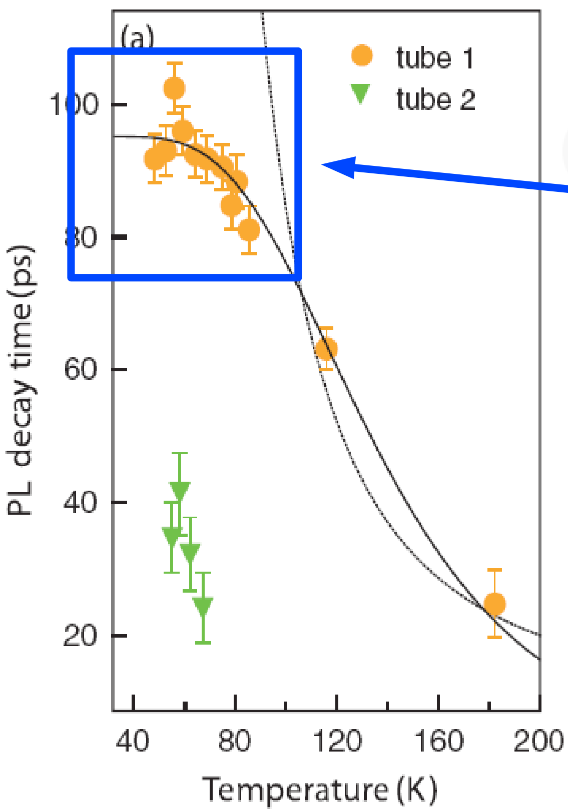


G. Baldini, A. Bosacchi, and B. Bosacchi, PRL, **23**, 846 (1969)

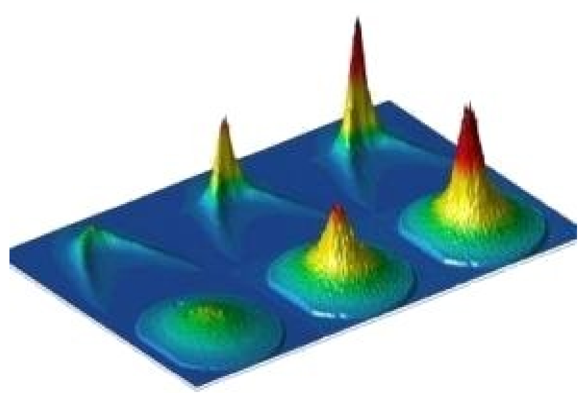


PRL 95, 247401 (2005)

YES ! Because phonons are responsible for the low-temperature saturation of the excitonic lifetime

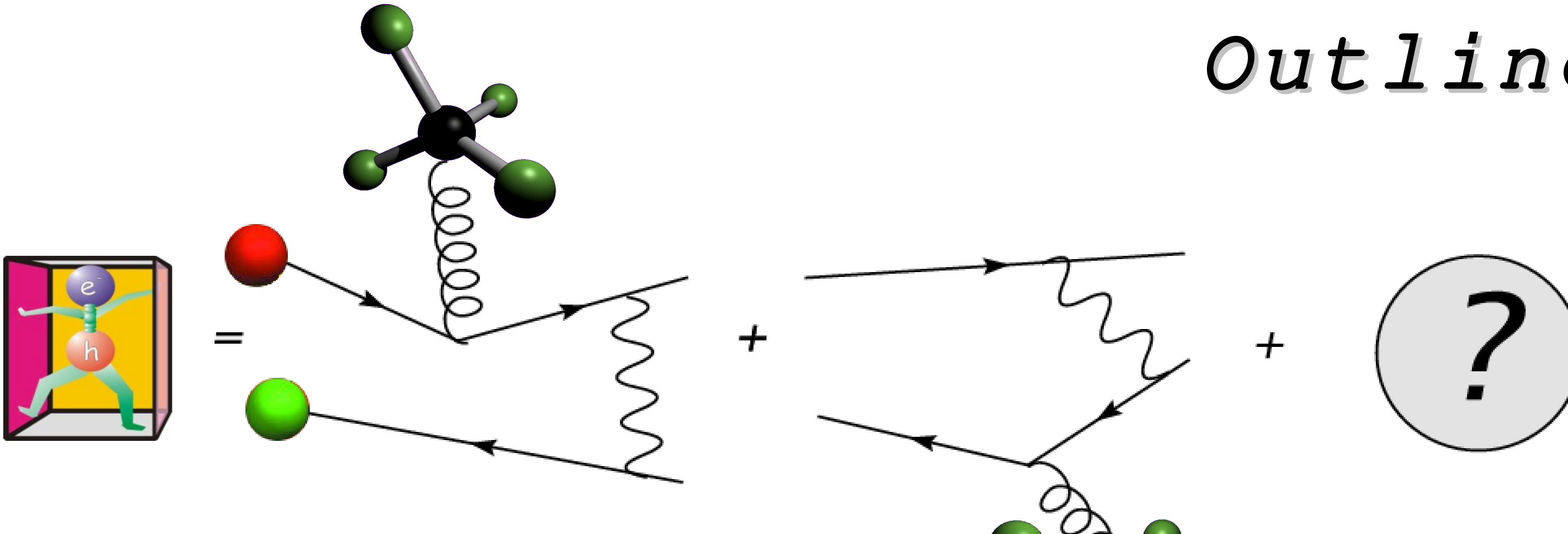


PRL 95, 197401 (2005)

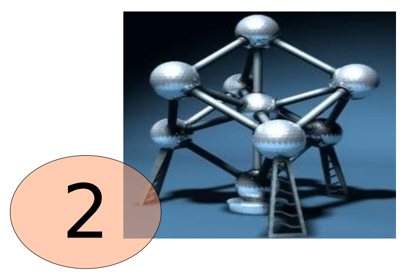


YES ! Because there are many different “flavours” of excitons: polaritons, polaronic excitons,...

Outline

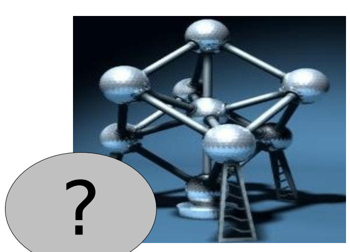


Polarons (I): Cardona-Allen approach



Polarons (II): Silicon and Diamond

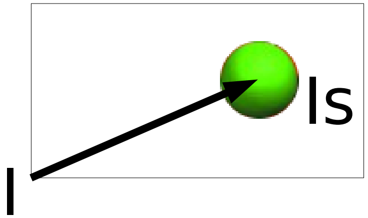
Finite temperature optics in the polaronic approximation



Dynamical BSE: the exciton phononic self-energy

Polarons
the Cardona-Allen approach

PRB **23**, 1495 (1981)



$$H = T + V_{SCF}\left(\{\mathbf{R}_{Is}\}\right)$$
$$\mathbf{R}_{Is} = \mathbf{R}_{Is} + \mathbf{u}_{Is}$$

$$\delta H = \delta H^{(1)} + \delta H^{(2)}$$
$$\delta H^{(1)} = \sum_{Is} \frac{\partial V_{SCF}}{\partial \mathbf{R}_{Is}} \mathbf{u}_{Is}$$
$$\delta H^{(2)} = \frac{1}{2} \sum_{IsJt} \frac{\partial^2 V_{SCF}}{\partial \mathbf{R}_{Is} \partial \mathbf{R}_{Jt}} \mathbf{u}_{Is} \mathbf{u}_{Jt}$$

Using standard 1st and 2nd order perturbation theory and the fact that $E_{n\mathbf{k}} = \langle n\mathbf{k}|H|n\mathbf{k}\rangle$

$$\delta E_{n\mathbf{k}} = \sum_{IsJt} \left[\frac{1}{2} \langle \frac{\partial^2 V_{SCF}}{\partial \mathbf{R}_{Is} \partial \mathbf{R}_{Jt}} \rangle + \sum_{m\mathbf{p}} \left(E_{n\mathbf{k}} - E_{m\mathbf{p}}\right)^{-1} \langle \frac{\partial V_{SCF}}{\partial \mathbf{R}_{Is}} |m\mathbf{p}\rangle \langle m\mathbf{p}| \frac{\partial V_{SCF}}{\partial \mathbf{R}_{Jt}} \rangle \right] \mathbf{u}_{Is} \mathbf{u}_{Jt}$$

Debye-Waller — Fan

Cardona-Allen further impose an *"acoustic sum rule"*

$$\delta E_{n\mathbf{k}}\left[\{\mathbf{u}_{Is} + \mathbf{v}\}\right] = \delta E_{n\mathbf{k}}\left[\{\mathbf{u}_{Is}\}\right]$$

$$\langle \frac{\partial^2 V_{SCF}}{\partial \mathbf{R}_{Is} \partial \mathbf{R}_{Jt}} \rangle = \sum_{m} \left(E_{n\mathbf{k}} - E_{m\mathbf{k}}\right)^{-1} F \left[\langle n\mathbf{k}| \frac{\partial V_{SCF}}{\partial \mathbf{R}_{Is}} |m\mathbf{p}\rangle \right]$$

Finally $\displaystyle \sum_{Is} \mathbf{u}_{Is} \langle n'\mathbf{k}+\mathbf{q}| \frac{\partial V_{SCF}}{\partial \mathbf{R}_{Is}} |n\mathbf{k}\rangle = \sum_{\mathbf{q}\lambda} g^{\mathbf{q}\lambda}_{n'n\mathbf{k}} \left(b^{\dagger}_{\mathbf{q}\lambda} + b_{\mathbf{q}\lambda}\right)$

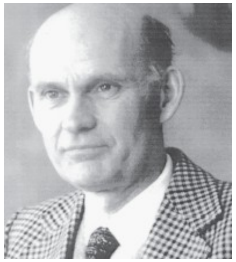
$$\delta E_{n\mathbf{k}} = \sum_{\mathbf{q}\lambda m} \left[\frac{|g^{\mathbf{q}\lambda}_{n'n\mathbf{k}}|^2}{E_{n\mathbf{k}} - E_{m\mathbf{k}+\mathbf{q}}} - \frac{\Lambda^{\mathbf{q}\lambda}_{n'n\mathbf{k}}}{E_{n\mathbf{k}} - E_{m\mathbf{k}}} \right] \left(2\langle N_{\mathbf{q}\lambda}\rangle + 1\right)$$

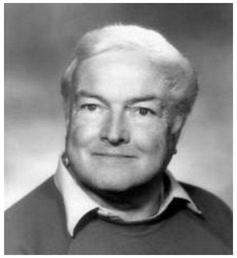
☺ Clear dependence on the temperature

☹ Polaron damping nelgected

Polarons, Hedin's equations, and Density Functional Perturbation Theory

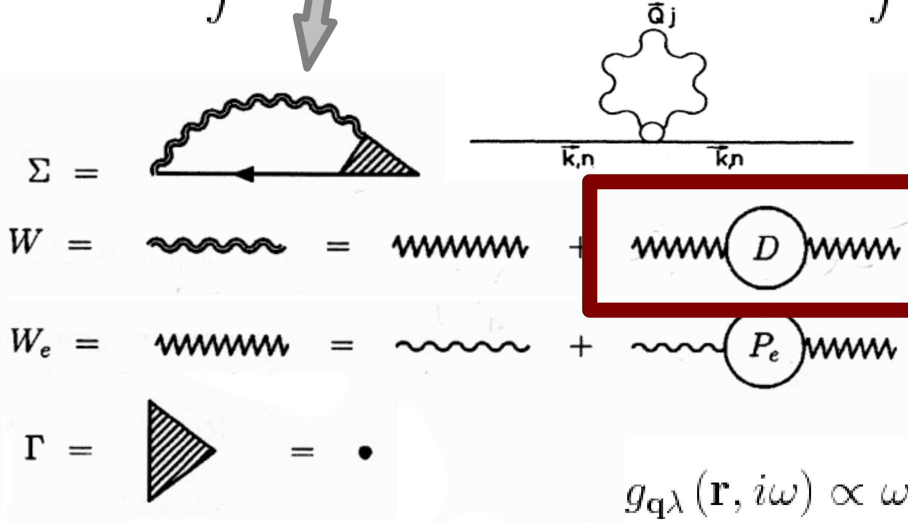
LH and SL, Solid. State Phys. **23**, 1 (1969); RvL, PRB **69**, 115110 (2004)





$$\delta E_{n\mathbf{k}} = \sum_{\mathbf{q}\lambda m} \left[\frac{|g^{\mathbf{q}\lambda}_{n'n\mathbf{k}}|^2}{E_{n\mathbf{k}} - E_{m\mathbf{k}+\mathbf{q}}} - \frac{\Lambda^{\mathbf{q}\lambda}_{n'n\mathbf{k}}}{E_{n\mathbf{k}} - E_{m\mathbf{k}}} \right] \left(2\langle N_{\mathbf{q}\lambda}\rangle + 1\right)$$

$$H_{MB} + \int d\mathbf{r}\rho\left(\mathbf{r}\right)\phi\left(\mathbf{r},t\right) \qquad H_{MB} + \int d\mathbf{r}\rho\left(\mathbf{r}\right)\phi\left(\mathbf{r}\right) - \int d\mathbf{R}N\left(\mathbf{R}\right)J\left(\mathbf{R},t\right)$$



Phononic contribution to the screened interaction

$$W_{ph}\left(\mathbf{r}_1,\mathbf{r}_2,i\omega\right) = \sum_{\mathbf{q}\lambda} \frac{2\omega_{\mathbf{q}\lambda}}{\omega^2 + \omega^2_{\mathbf{q}\lambda}} g_{\mathbf{q}\lambda}\left(\mathbf{r}_1,i\omega\right) g^*_{\mathbf{q}\lambda}\left(\mathbf{r}_2,i\omega\right)$$

$$g_{\mathbf{q}\lambda}\left(\mathbf{r},i\omega\right) \propto \omega^{-1/2}_{\mathbf{q}\lambda} \sum_{Is} \int d\mathbf{r}_1 \epsilon^{-1}_e\left(\mathbf{r},\mathbf{r}_1;i\omega\right) \epsilon\left(\mathbf{q}\lambda|s\right) \cdot \nabla V_{ion}\left(\mathbf{r}_1\right) e^{i\mathbf{q}\cdot(\mathbf{R}_I+\tau_s)}$$

SC field

- The SC field is frequency dependent
- There is no coupled equation for the phonon propagator
- There is no need of an explicit el-ph term in the Hamiltonian (Frohlich-like)

We can use DFPT to describe phonons as it is "almost" consistent with MB (DFPT uses a static test-electron dielectric function to screen the ionic potential)

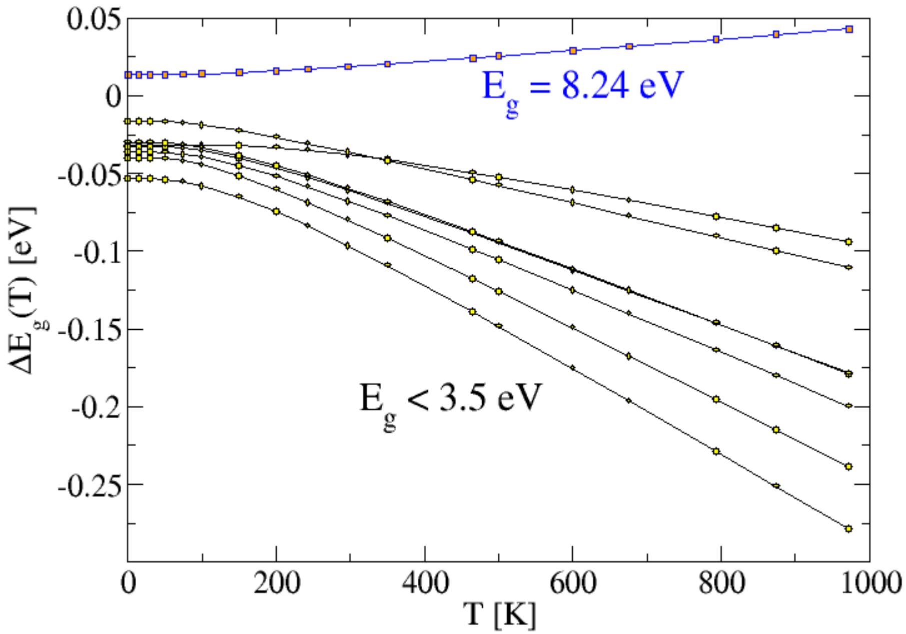
Polarons in silicon & diamond

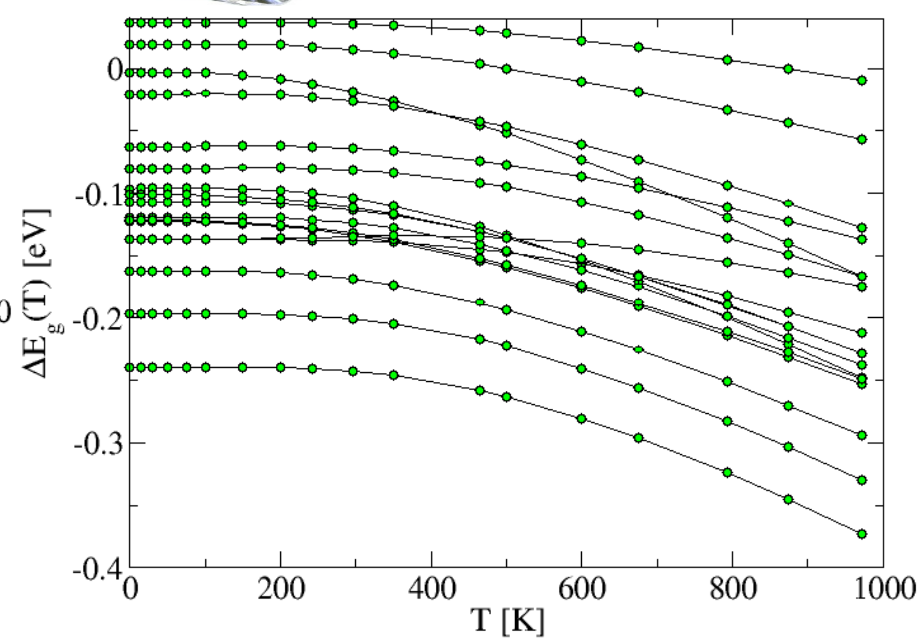
Using finite temperature diagrammatic tecniques the phononic GW self-energy is

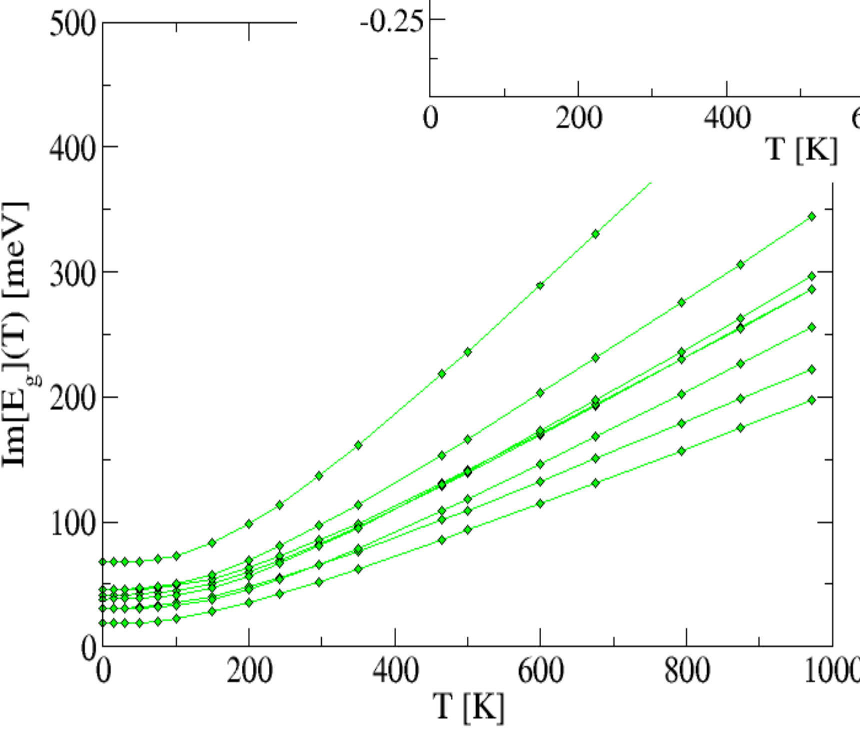
$$\Sigma^{FAN}_{n\mathbf{k}}(i\omega) = \sum_{\mathbf{q}\lambda m} |g^{\mathbf{q}\lambda}_{n'n\mathbf{k}}|^2 \left[\frac{\langle N_{\mathbf{q}\lambda}\rangle + 1 - f_{m\mathbf{k}-\mathbf{q}}}{i\omega - E_{m\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{q}\lambda}} + \frac{\langle N_{\mathbf{q}\lambda}\rangle + f_{m\mathbf{k}-\mathbf{q}}}{i\omega - E_{m\mathbf{k}+\mathbf{q}} + \omega_{\mathbf{q}\lambda}} \right]$$

☺ The FAN SE includes polaron dampings

☺ It reduces to the Cardona-Allen formulation in the "on mass-shell" approximation



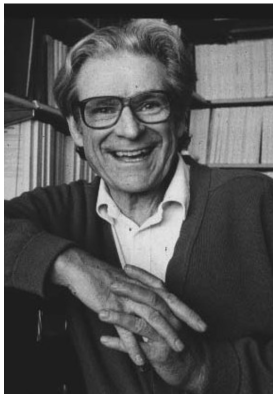


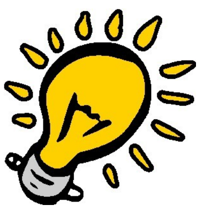


!!!

(...) the zero-point renormalization vibration amplitude is responsible for energy renormalizations up to 200 meV, of the order of accuracy claimed for state-pf-the-art ab initio calculations (...)

Excitons



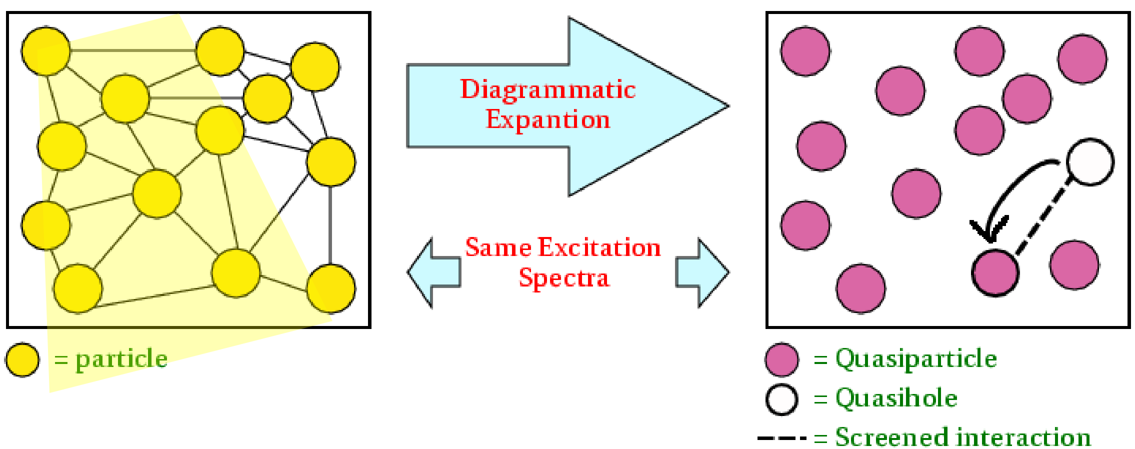


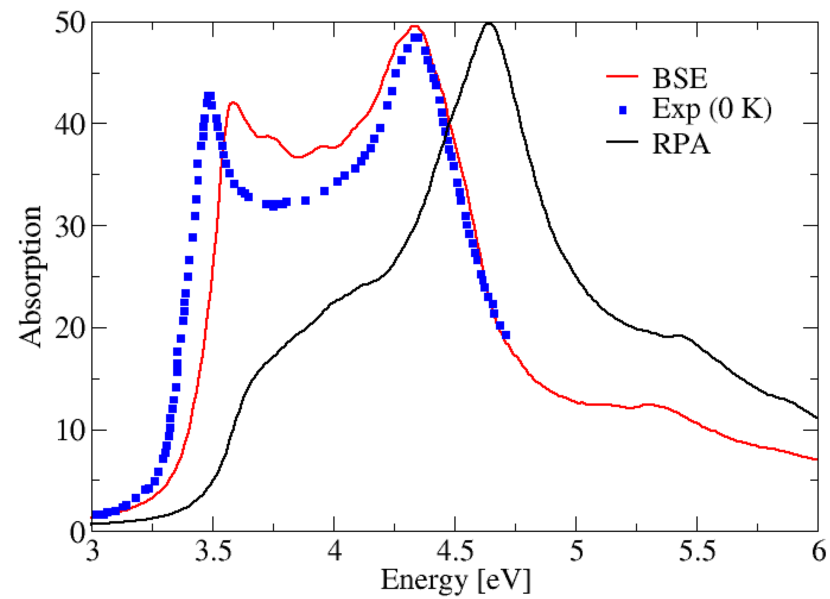
EXCITONS

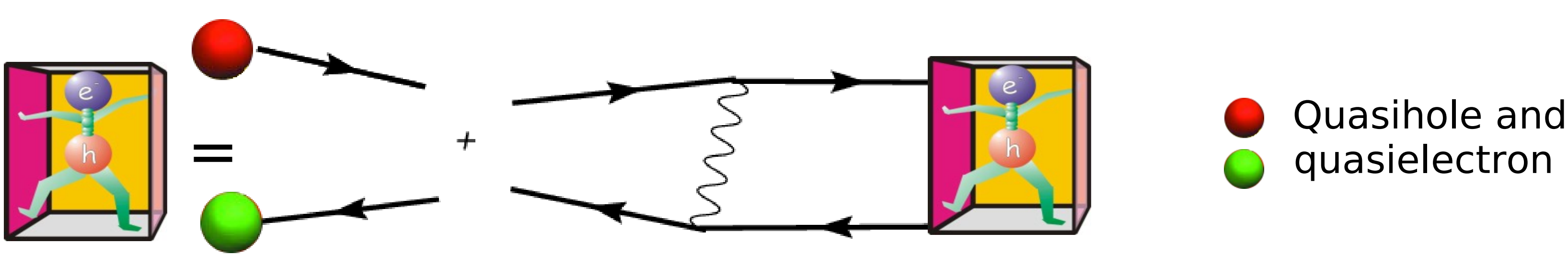
Memory Effects: AM, R. Del Sole, PRL, **91**, 176402 (2003).

Complex Materials: PRL **94**, 087404 (2005), PRL **96**, 126104 (2006), PRL **98**, 036807 (2007).

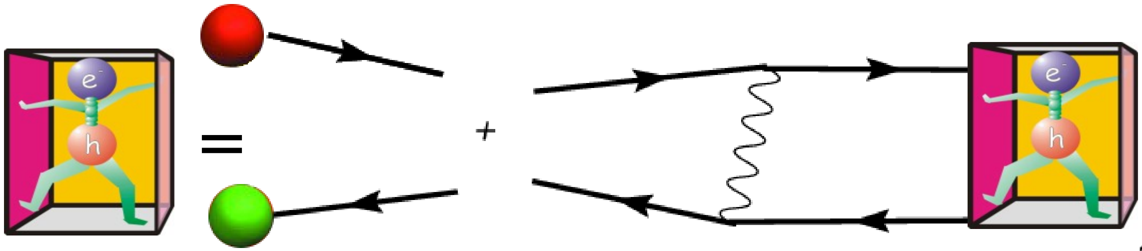
In TDDFT: AM, R. Del Sole, A. Rubio PRL **91**, 256402



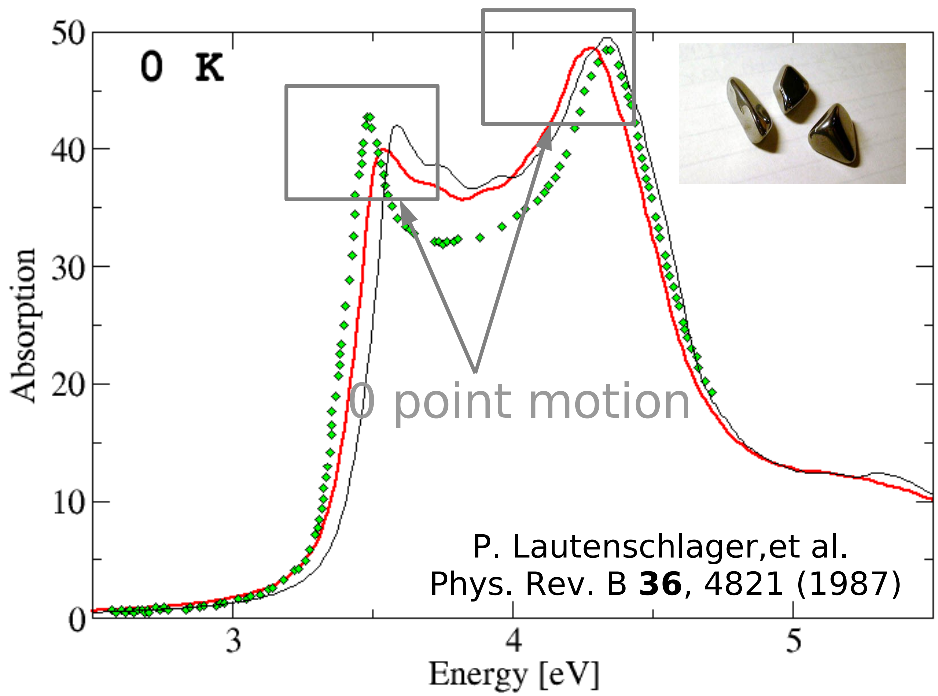


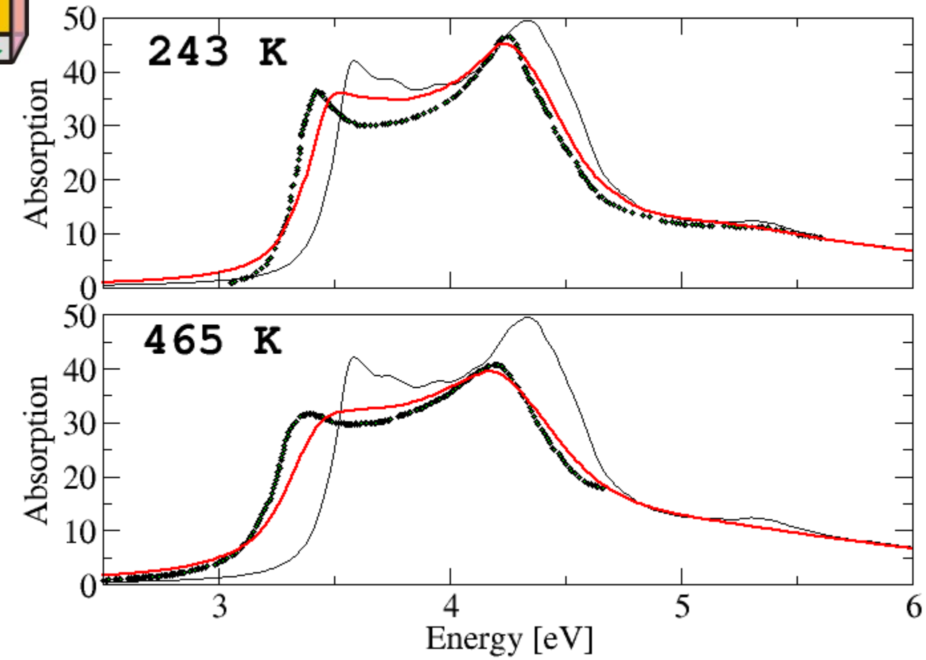


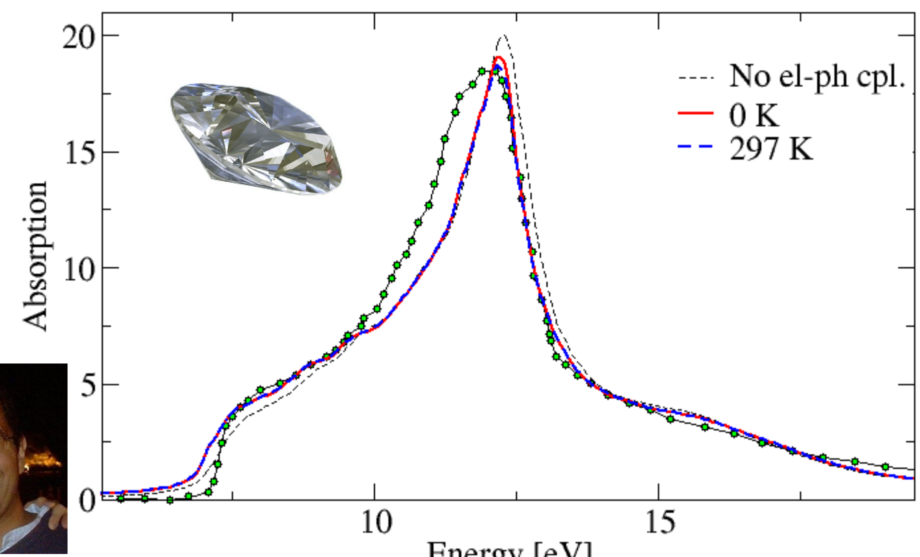
Finite T optics in the polaronic approach



Quasihole and quasielectron **polarons**







!!!

In any ab initio calculation of the optical spectra (RPA or BSE) the electron-hole damping is used as a parameter, fitted to get the best agreement with the experiment. In all the present calculations the numerical damping is 0.1 meV. The real, physical, damping is due to the polaronic widths.

Exciton-phonon coupling state-of-the-art

Showing results **1 - 25** of **892** total

Your Search
Full Text: exciton-phonon

Journals
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Phys. Rev. Lett. (138)
Phys. Rev. (33)
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Rev. Mod. Phys. (3)

Category
Featured in Physics News Update (1)
PRL Editors' Suggestion (1)

Year
1996 (45)
1999 (43)
2000 (39)
2004 (37)
1994 (37)
(Show All Years)

Bosonic Hamiltonians

B. Segall, G.F. Mahan PR **171**, 935 (1968)
A. Suna, PR **135**, A111 (1964)
Y. Toyozawa, J. Phys. Chem. Solids **25**, 59 (1964)

$$H = \sum_{\lambda\mathbf{K}} \epsilon_{\lambda\mathbf{K}} n^{exc}_{\lambda\mathbf{K}} + \sum_{\mathbf{q}} \omega_{\mathbf{q}} n^{ph}_{\mathbf{q}} + \sum_{\lambda\lambda'\mathbf{K}\mathbf{q}} V(\mathbf{q})_{\lambda\lambda'} c^{\dagger}_{\lambda\mathbf{K}+\mathbf{q}} c_{\lambda'\mathbf{K}} \left(a_{\mathbf{q}} + a^{\dagger}_{\mathbf{q}}\right)$$

???

- Easy interpretation
- Impossible to do *Ab-Initio*: no idea how to evaluate the exc-ph coupling potential
- *"Interacting excitons should not be bosonized"* **M. Combescot**

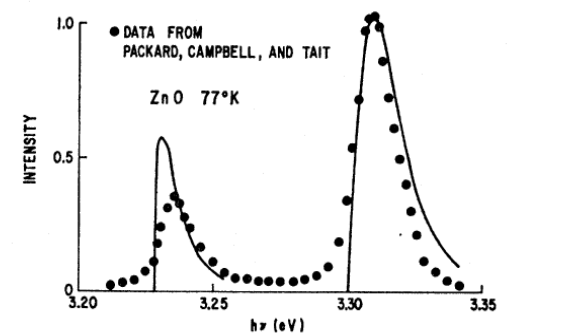


FIG. 9. Calculated one-LO- and two-LO-phonon-assisted fluorescence peaks for ZnO at 77°K compared with the spectrum observed by Packard *et al.*

$$H_{\text{el}} = \sum_{\alpha\mathbf{k}} \left(e_{\alpha\mathbf{k}} + \sum_{\mathbf{q}} n_{\alpha\mathbf{q}} V_{\mathbf{q}} \right) a^{\dagger}_{\alpha\mathbf{k}} a_{\alpha\mathbf{k}},$$

$$H_{\text{el-el}} = \sum_{\alpha\beta\mathbf{k}\mathbf{k}'\mathbf{q}} \tfrac{1}{2} V_{\mathbf{q}} a^{\dagger}_{\alpha\mathbf{k}} a^{\dagger}_{\beta\mathbf{k}'} a_{\beta\mathbf{k}'+\mathbf{q}} a_{\alpha\mathbf{k}-\mathbf{q}},$$

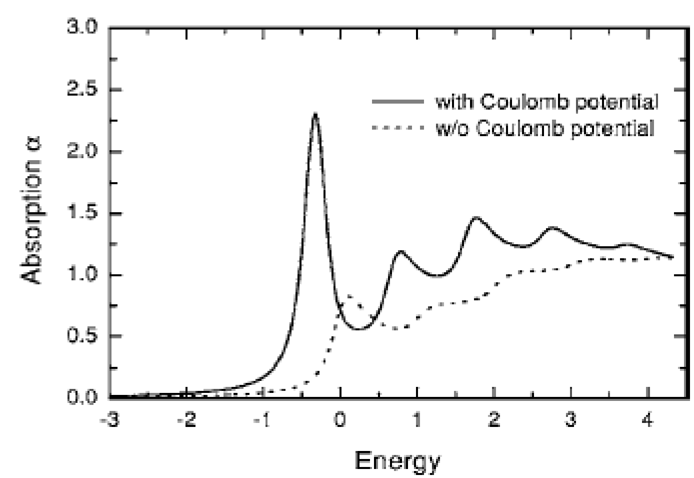
$$H_{\text{el-ph}} = \sum_{\alpha\mathbf{k}\mathbf{q}} \hbar\omega_{\mathbf{q}} g_{\alpha\mathbf{q}} a^{\dagger}_{\alpha\mathbf{k}-\mathbf{q}} a_{\alpha\mathbf{k}} (b_{-\mathbf{q}} + b^{\dagger}_{\mathbf{q}}),$$

$$H_{\text{ph}} = \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} \left(b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}} + \tfrac{1}{2} \right)$$

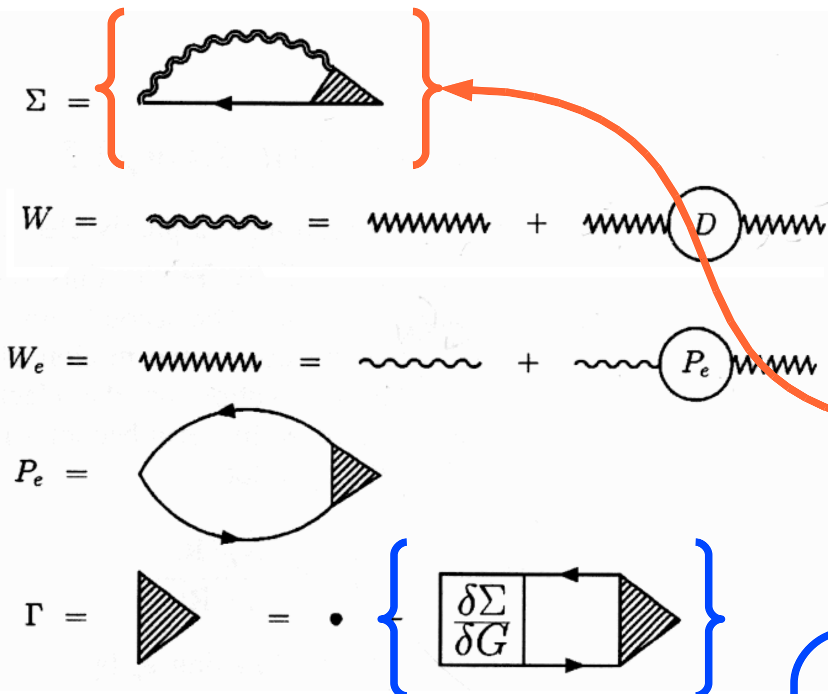
Frohlich Hamiltonians

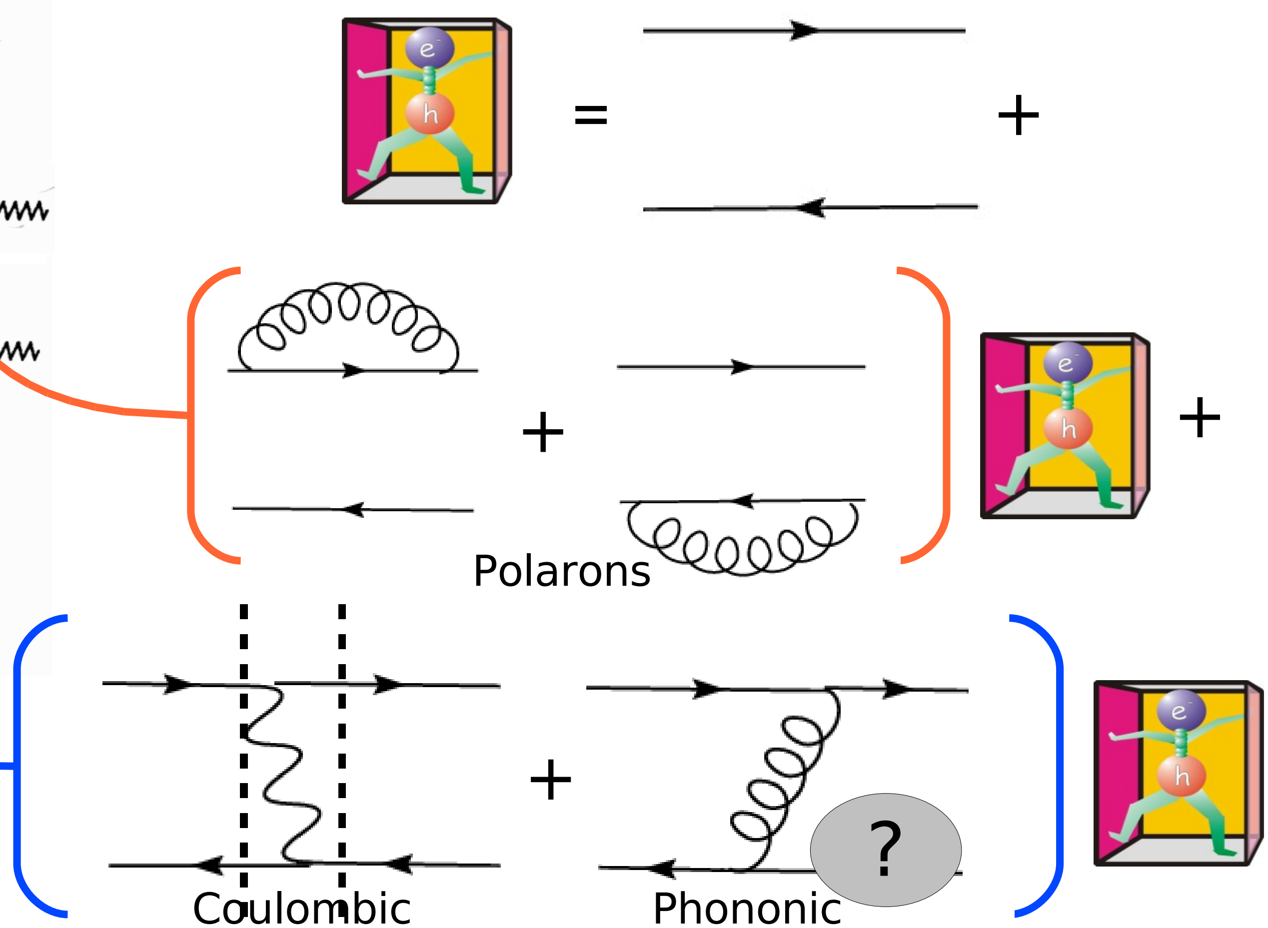
K. Hannewald and P. A. Bobbert, PRB **72**, 113202 (2005)

- Widely used
- Phonon overscreening [RvL, PRB **69**, 115110 (2004)]



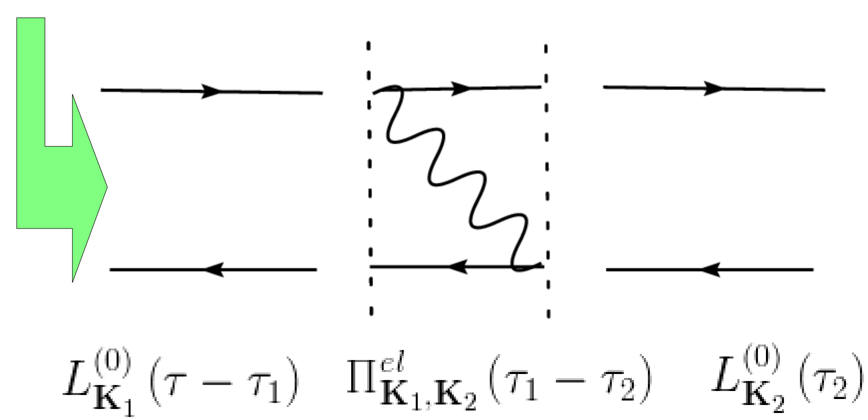
The dynamical BSE (DBSE)





A correct derivation of the BSE from the generalized Hedin's equations yields an indirect (polaronic mediated) and a direct exciton-phonon coupling

The dynamical BSE [AM, R. Del Sole, PRL **91**, 176402 (2003)] was introduced to sum the frequency dependent Coulombic interaction



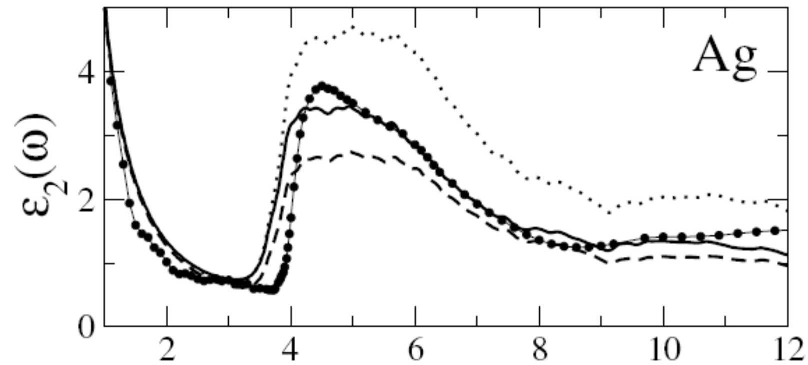
$$\mathbf{K} \equiv (n_e, n_h, \mathbf{k})$$

A phononic kernel in the DBSE

electronic DBSE

$$\tilde{L}_{\mathbf{K}_1,\mathbf{K}_2}(i\omega) = \tilde{L}^{(0)}_{\mathbf{K}_1}(i\omega)\left[\delta_{12} + \sum_{\mathbf{K}_3} W_{\mathbf{K}_1,\mathbf{K}_3}\tilde{L}_{\mathbf{K}_3,\mathbf{K}_2}(i\omega)\right]$$

$$\tilde{L}_{\mathbf{K}_1,\mathbf{K}_2}(i\omega) = \tilde{L}^{(0)}_{\mathbf{K}_1}(i\omega)\left[\delta_{12} + \sum_{\mathbf{K}_3} \Pi^{el}_{\mathbf{K}_1,\mathbf{K}_3}(i\omega)\,\tilde{L}_{\mathbf{K}_3,\mathbf{K}_2}(i\omega)\right]$$

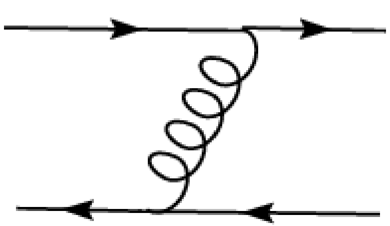


AM, R. Del Sole, PRL **91**, 176402 (2003)

phononic DBSE

$$\tilde{L}_{\mathbf{K}_1,\mathbf{K}_2}(i\omega) = \tilde{L}^{(0)}_{\mathbf{K}_1}(i\omega)\left[\delta_{12} + \sum_{\mathbf{K}_3}\left(\Pi^{el}_{\mathbf{K}_1,\mathbf{K}_3}(i\omega) + \Pi^{ph}_{\mathbf{K}_1,\mathbf{K}_3}(i\omega)\right)\tilde{L}_{\mathbf{K}_3,\mathbf{K}_2}(i\omega)\right]$$

+ -



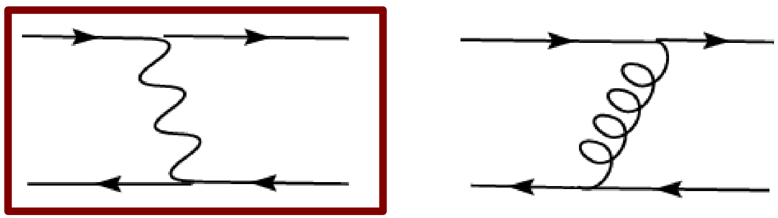
$$\Pi^{ph}_{\mathbf{K}_1,\mathbf{K}_2}(i\omega) = -\sum_{\lambda} g^{\mathbf{q}\lambda}_{c_2c_1\mathbf{k}_1}\left(g^{\mathbf{q}\lambda}_{v_2v_1\mathbf{k}_1}\right)^* \sum_I \left[\frac{1+\langle N_{\mathbf{q}\lambda}\rangle}{i\omega+\Delta_I-\omega_{\mathbf{q}\lambda}} + \frac{\langle N_{\mathbf{q}\lambda}\rangle}{i\omega+\Delta_I+\omega_{\mathbf{q}\lambda}}\right]$$

$$\Delta_1 = \epsilon_{v_2\mathbf{k}_1-\mathbf{q}} - \epsilon_{c_1\mathbf{k}_1} \qquad \Delta_2 = \epsilon_{v_1\mathbf{k}_1} - \epsilon_{c_2\mathbf{k}_1-\mathbf{q}}$$

The phononic kernel enhances the screening (like in the polaronic SE) reducing the electron-hole interaction

The phonon frequencies in the DBSE kernel are responsible for the phonon sidebands (an additional contribution stem from the polaronic self-energy diagrams)

A phonic self-energy for the exciton



$$\tilde{L}(i\omega) = \tilde{L}^{(0)}(i\omega)\left[1 - \tilde{L}^{(0)}(i\omega)\left(W + \Pi^{ph}(i\omega)\right)\right]^{-1}$$

$$\tilde{L}^{e}(i\omega) = \tilde{L}^{(0)}(i\omega)\left[1 - \tilde{L}^{(0)}(i\omega)\,W\right]^{-1}$$

Similarly to the quasiparticle case the phononic and electronic parts can be summed separately

$$\tilde{L}(i\omega) = \tilde{L}^{(0)}(i\omega)\left[1 - \tilde{L}^{(0)}(i\omega)\,W\right]^{-1}\left[1 - \tilde{L}^{(0)}(i\omega)\left[1 - \tilde{L}^{(0)}(i\omega)\,W\right]^{-1}\Pi^{ph}(i\omega)\right]^{-1}$$

$$\tilde{L}(i\omega) = \tilde{L}^{e}(i\omega)\left[1 - \tilde{L}^{e}(i\omega)\,\Pi^{ph}(i\omega)\right]^{-1}$$

If we rotate in the excitonic basis $|I\rangle = \sum_{\mathbf{K}} \langle \mathbf{K}|I\rangle$ $\quad \langle I|\tilde{L}^{e}(i\omega)|J\rangle = -\delta_{IJ}(i\omega - E_I)^{-1}$

$$\tilde{L}(i\omega) = -\left[\delta_{IJ}(i\omega - E_I) - \langle I|\Pi^{ph}(i\omega)|J\rangle\right]^{-1}$$

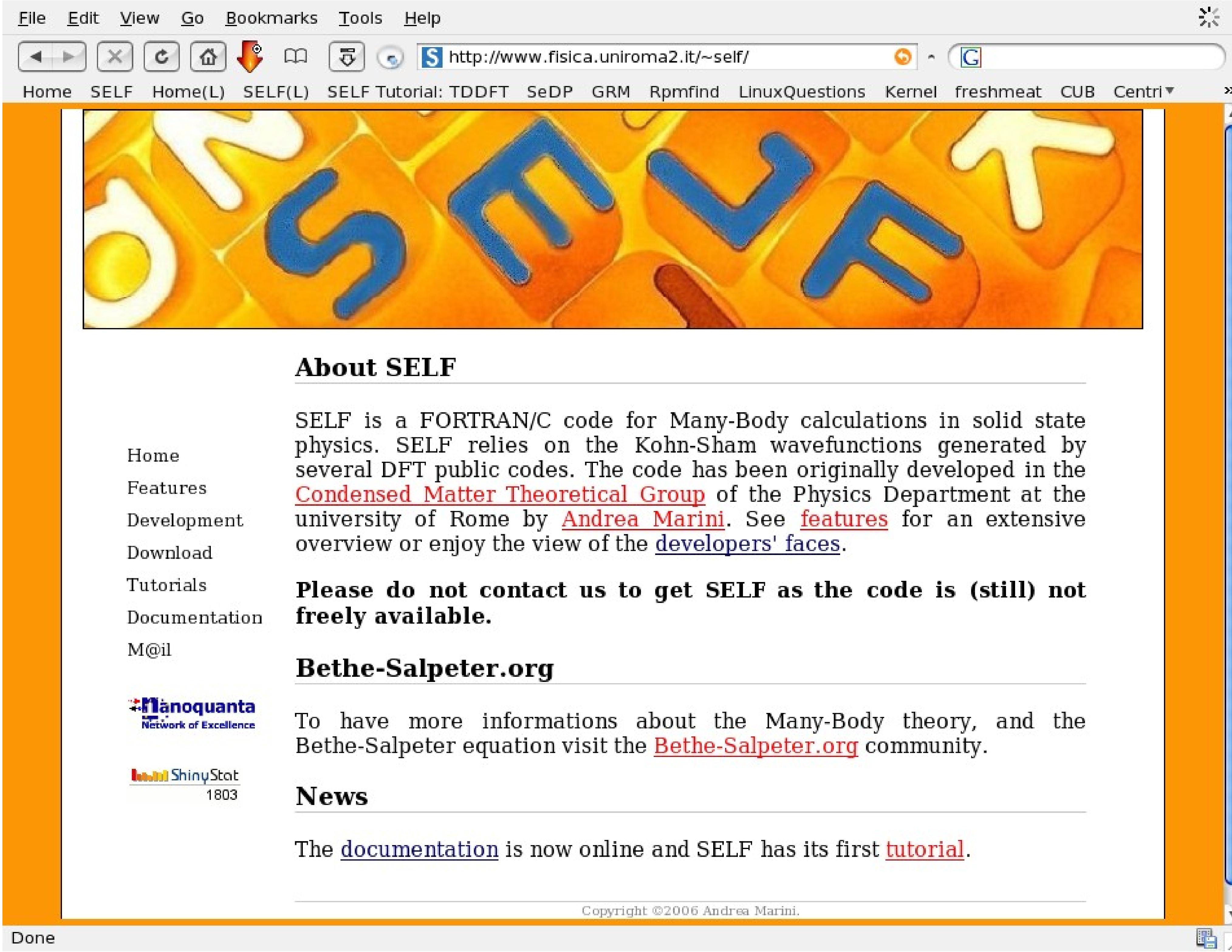
Excitonic Self-Energy

The excitonic self-energy implies all the physical consequences of the self-energy potential in any quasiparticle theory: **damping, phonon replicas, fictitious exciton-phonon coupling potential. No bosonic Hamiltonians are needed.**

Conclusions...

- The finite temperature optical properties in presence of weak electron-phonon coupling can be described with the standard BSE in the polaronic picture
- The excitonic damping and zero-point motion effect are in excellent agreement with the experiment, being small in systems (like diamond) with large Debye energy.
- In presence of STRONG electron-phonon coupling the dynamical extension of the BSE includes a phononic kernel that act to reduce the electron-hole interaction.
- The DBSE can be reduced in the static excitons basis to yield a phononic self-energy WITHOUT ANY drastic assumption on the Hamiltonian.

...and...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About SELF
SELF is a FORTRAN/C code for Many-Body calculations in solid state physics. SELF relies on the Kohn-Sham wavefunctions generated by several DFT public codes. The code has been originally developed in the Condensed Matter Theoretical Group of the Physics Department at the university of Rome by Andrea Marini. See features for an extensive overview or enjoy the view of the developers' faces.
Please do not contact us to get SELF as the code is (still) not freely available.
Bethe-Salpeter.org
To have more informations about the Many-Body theory, and the Bethe-Salpeter equation visit the Bethe-Salpeter.org community.
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The documentation is now online and SELF has its first tutorial.
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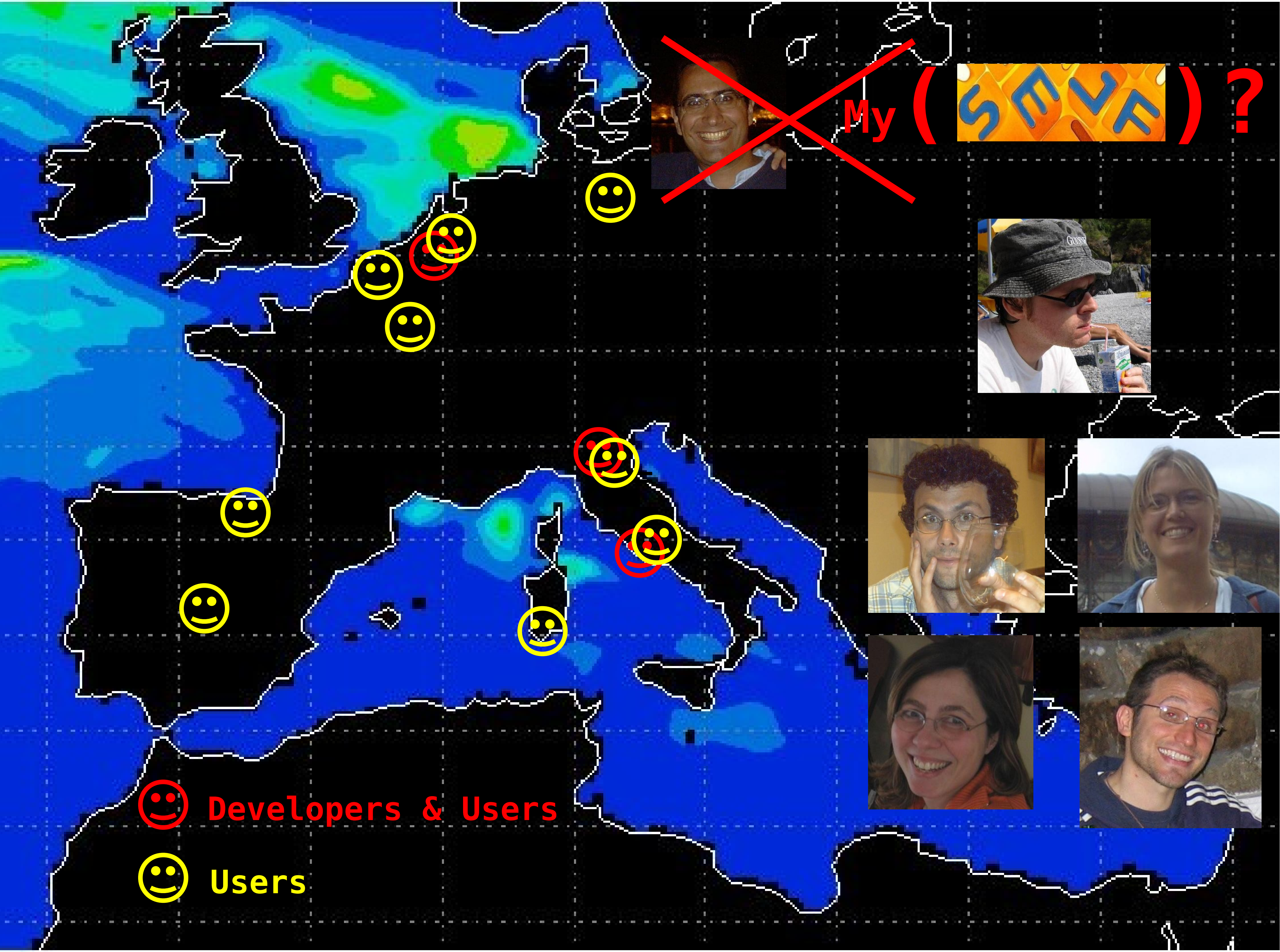
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Developers & Users
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"SELF, a shiny pot of fun and happiness"

[C. Hogan]

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