Optical nonlinear processes in semiconductors: an ab-initio description

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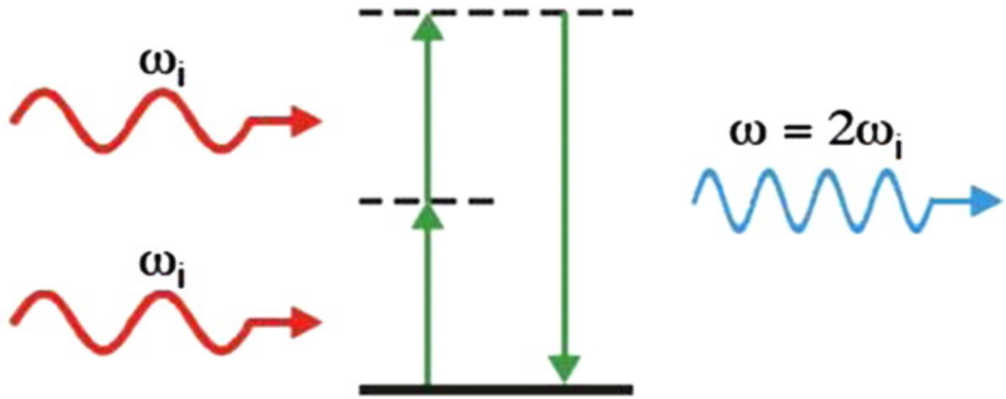
Outline

1. Introduction: definition and general features
2. SHG in condensed matter
    - Screening, Excitons and Local fields
    - GaAs
3. Nonlinear processes with static fields
    - Theoretical description
    - Electronic and ionic part
    - EFISH

Second harmonic generation



Amplitude

$$\chi^{(3)}E^{3} << \chi^{(2)}E^{2} << \chi^{(1)}E$$

First nonlinear term

but…

Symmetry

Centro-symmetric materials $\chi^{(2)} = 0$

in the dipole approximation
(Long wavelength limit)

Interest for Second Harmonic Generation:
in condensed matter

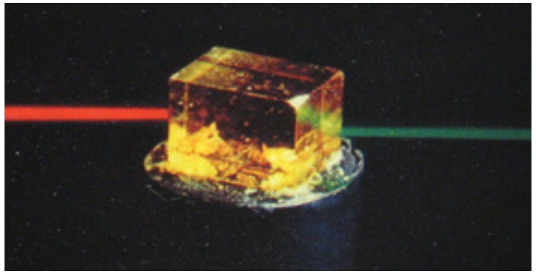
- Probe for materials :

  Sensitivity to local symmetries and selection rules for electronic transitions in $\chi^{(2)}$ gives access to states with different symmetries, compared to linear optics

  $\Rightarrow$ { Surfaces, Thin films, Interfaces, Nanowires

- Development and characterisation of new materials

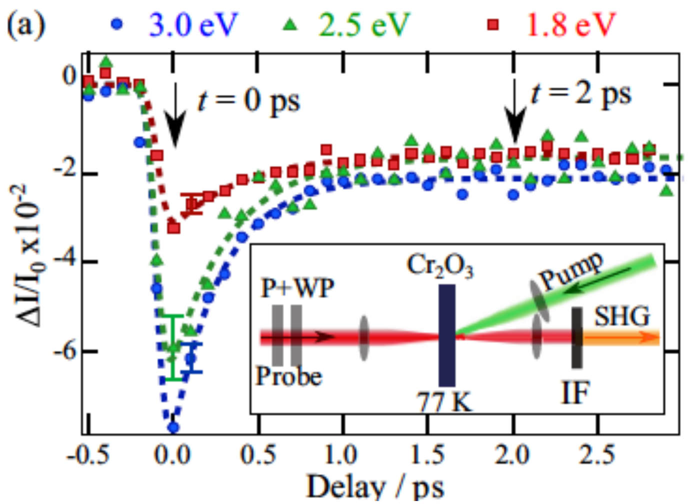
  New optical devices



Interest for Second Harmonic Generation:
in ultrafast processes

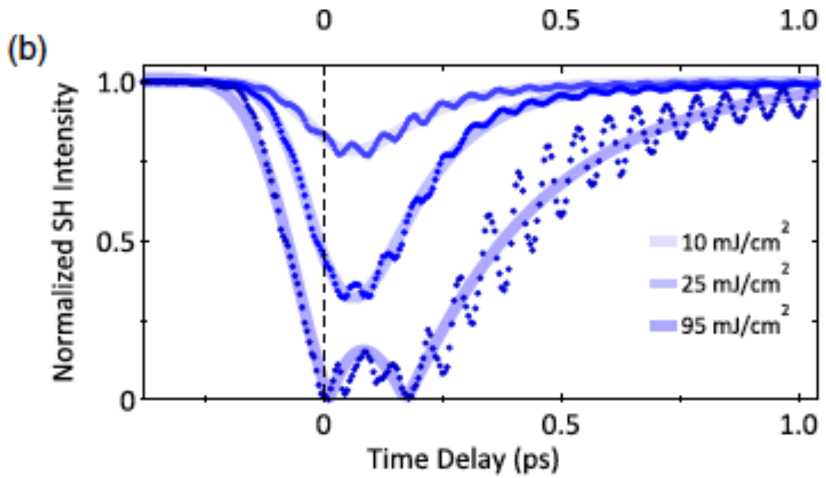
• Ultrafast probe for materials : time-resolved SHG

Ultrafast Demagnetization in Cr_2O_3



V. G. Sala *et al*, Phys. Rev. B **94** 014430 (2016)

Ultrafast Reversal of the Ferroelectric Polarization in $LiNbO_3$



R. Mankowsky *et al*,
Phys. Rev. Lett. **118** 197601 (2017)

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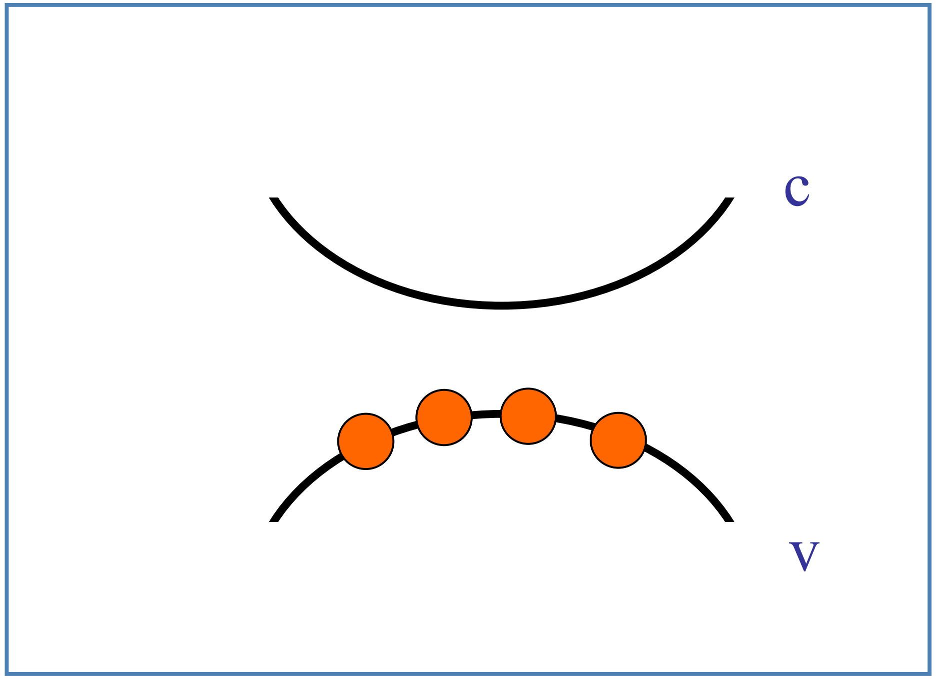
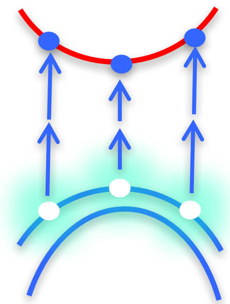
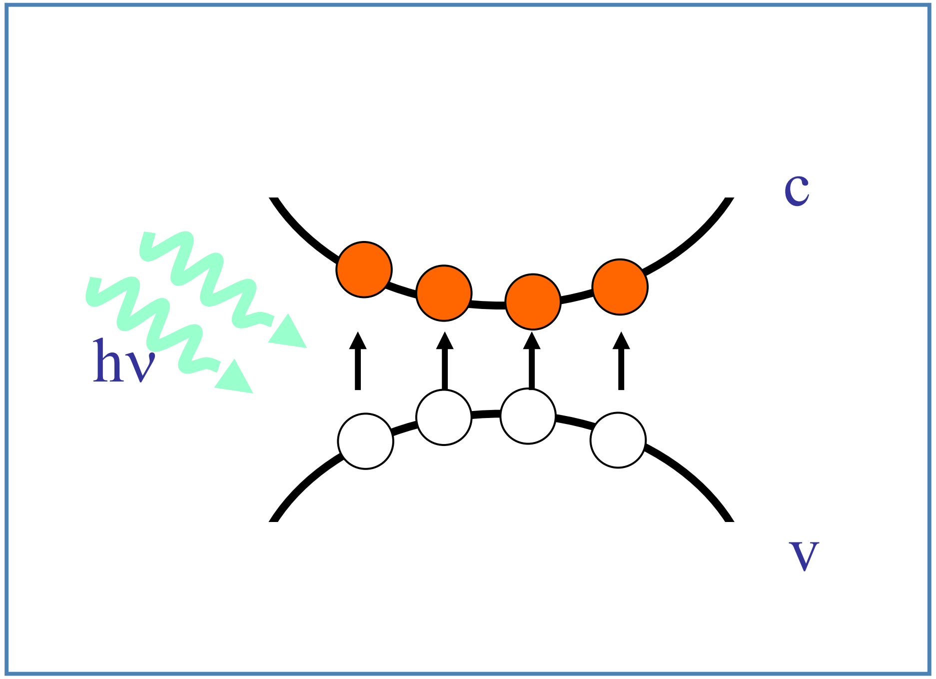
Band theory



Band theory





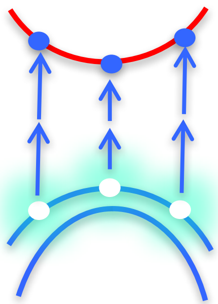
Independent particle approximation:

All the electrons make independent transitions

(IPA)

Fermi golden rule

Independent Particle Approximation



Second-order response : in "real life"

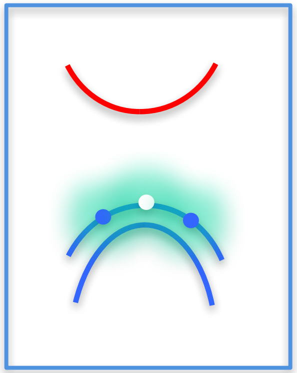
Independent Particle Approximation

$$\chi^{(2)}_{abc}(-2\omega,\omega,\omega)=\frac{-ie3}{\hbar^2 m^3\omega^3 V}\sum_{nml}\int d\vec{k}\,\frac{1}{E_m-E_n-2\omega-2i\eta}$$

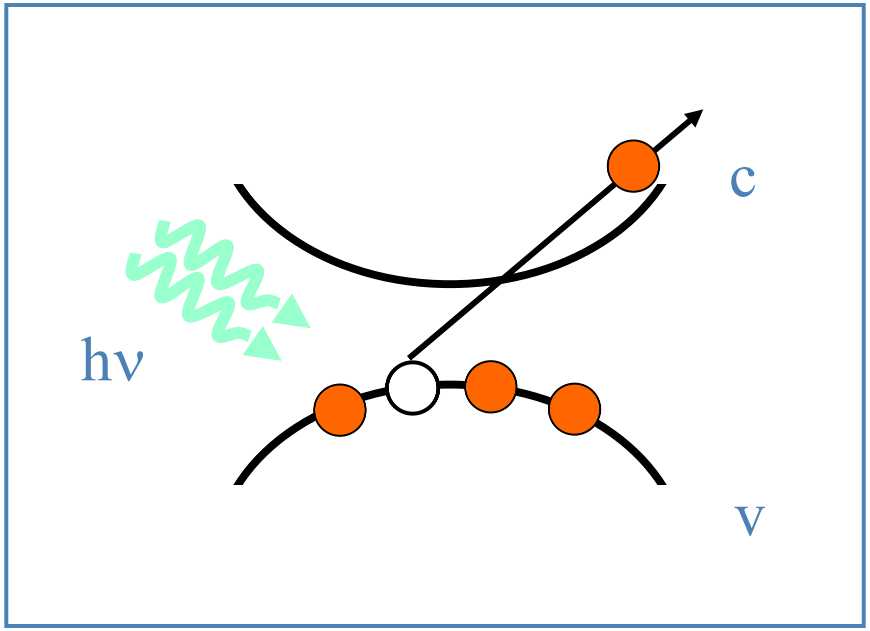
$$\times\left[f_{nl}(\vec{k})\frac{p^a_{nm}(\vec{k})\left\{p^b_{ml}(\vec{k})p^c_{\ln}(\vec{k})\right\}}{E_l-E_n-\omega-i\eta}+f_{ml}(\vec{k})\frac{p^a_{nm}(\vec{k})\left\{p^b_{ml}(\vec{k})p^c_{\ln}(\vec{k})\right\}}{E_m-E_l-\omega-i\eta}\right]$$

Dipole approximation (optical processes)

Screening

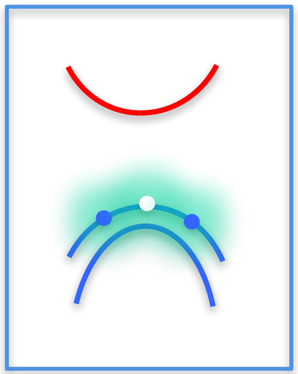


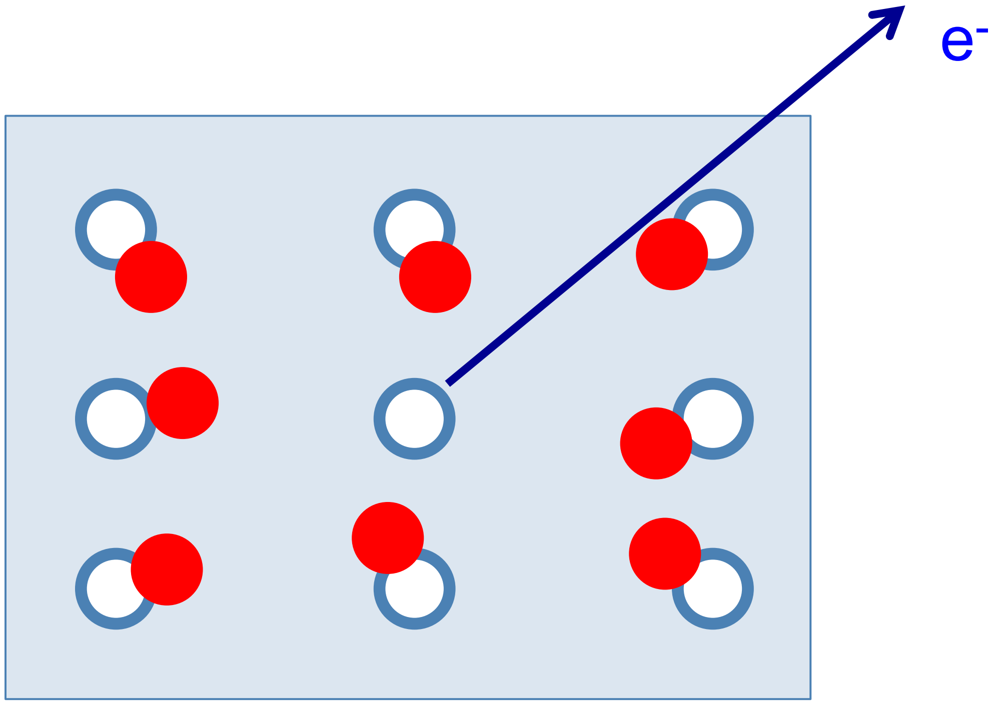
Creation of a hole



Hole + (N-1) electrons

Screening

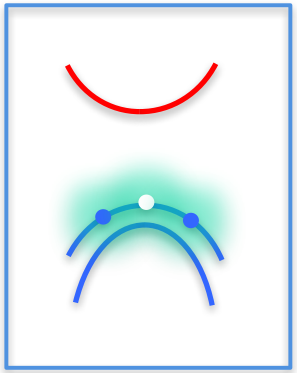


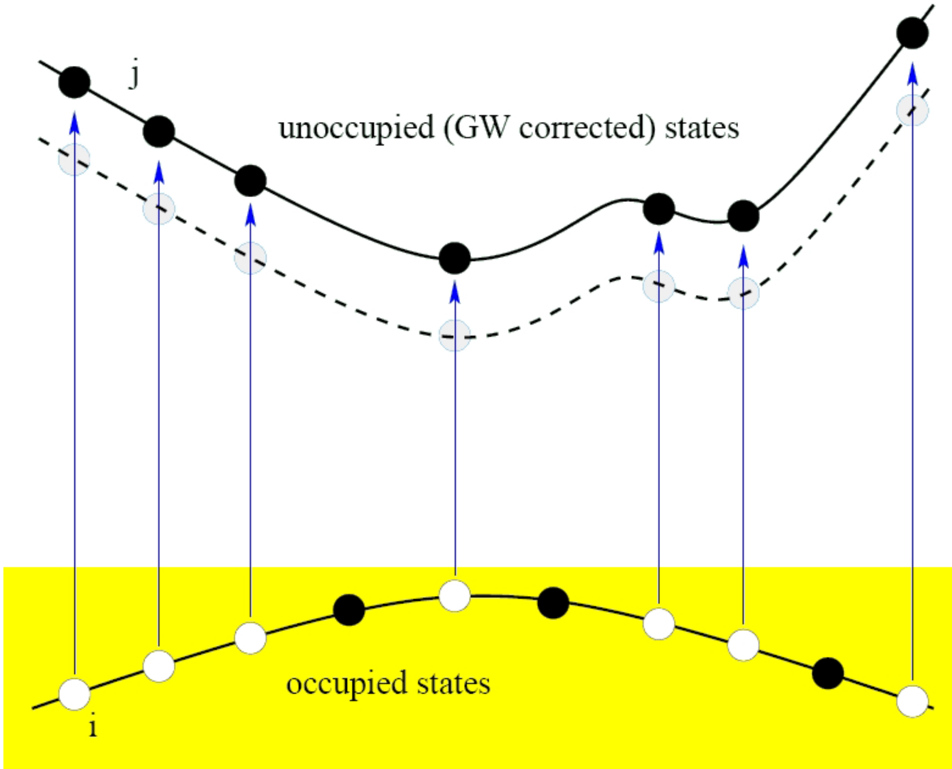


Hole + (N-1) electrons

reaction: polarisation, screening

Back to band structure

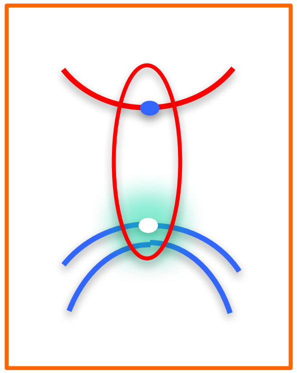




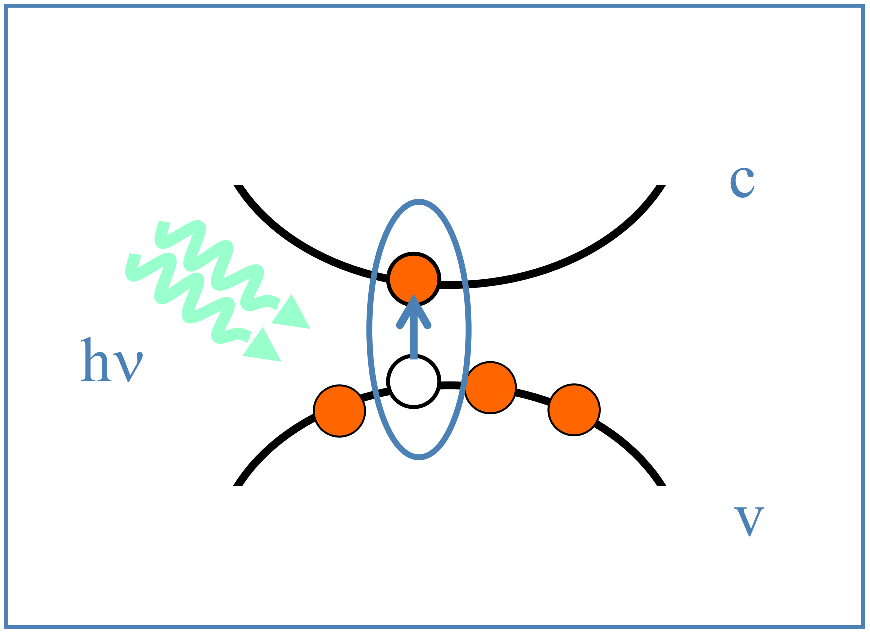
⇒Opening of the gap

GW or Scissor operator

Excitonic effects



Electron-hole interaction



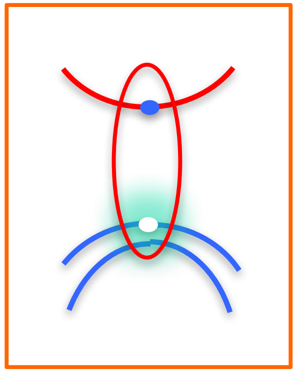
Bethe Salpeter Equation
(2-particles equation)

or

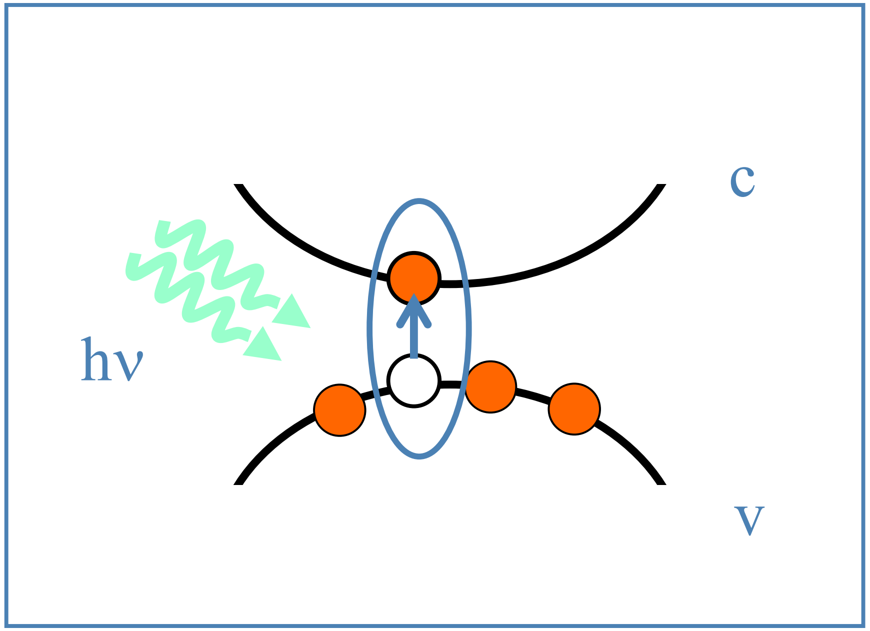
Time-Dependent Density-Functional Theory
(TDDFT)

1-particle picture → 2-particle picture

Excitonic effects



Electron-hole interaction



Time-Dependent Density-Functional Theory
(TDDFT)

1-particle picture → 2-particle picture

Towards the macroscopic polarization

External and total fields

$\mathbf{E}_{ext}(\omega) \longleftrightarrow \mathbf{E}_{tot}(\omega)$

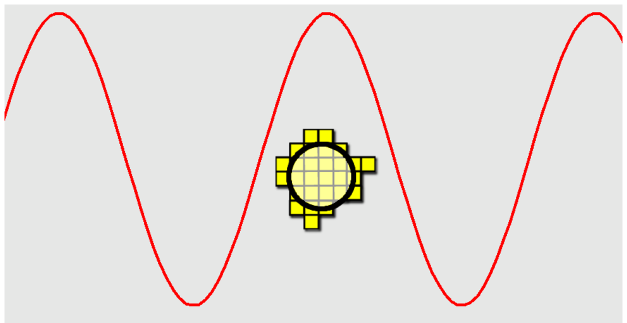
induced fields ⟶ large and irregular fluctuations over the atomic scale

$\mathbf{P}_M = \chi^{(2)} \mathbf{E}_{tot} \mathbf{E}_{tot}$

Macroscopic average

Average over distances :

- ✓ large compared to the cell diameter
- ✓ small compared to the wavelength of the external perturbation



The difference between the microscopic fields and the averaged (macroscopic) fields is called local fields.

TDDFT and response functions

Macroscopic susceptibility

Zinc-blende symmetry

$$\chi^{(2)}_{xyz}(2\mathbf{q},2\omega) = \frac{-i}{12 q_x q_y q_z}\left[\epsilon_M(2\mathbf{q},2\omega)\right]\left[\epsilon_M(\mathbf{q},\omega)\right]^2 \chi^{(2)}(2\mathbf{q},\mathbf{q},\mathbf{q},\omega,\omega)$$

ε_M : macroscopic dielectric function

$\chi^{(2)}$: second order response function

TDDFT and response functions

Response functions

Dyson equation

$$\chi^{(1)} = \chi_0^{(1)} + \chi_0^{(1)}(v + f_{xc})\chi^{(1)}$$

1st order

Coulomb potential

Non-interacting response function (Kohn-Sham)

Exchange-correlation kernel

$$f_{xc}(\mathbf{r}, t, \mathbf{r}', t') = \frac{\delta V_{xc}(\mathbf{r}, t)}{\delta \rho(\mathbf{r}', t')}$$

TDDFT and response functions

Response functions

Dyson equation
in momentum space

$$\chi^{(1)}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}',\omega) = \chi_0^{(1)}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}',\omega)$$
$$+\sum_{\mathbf{G}''\mathbf{G}'''} \chi_0^{(1)}(\mathbf{q}+\mathbf{G},\mathbf{q}+\mathbf{G}'',\omega)\left[v(\mathbf{q}+\mathbf{G}'',\mathbf{q}+\mathbf{G}''') + f_{xc}(\mathbf{q}+\mathbf{G}'',\mathbf{q}+\mathbf{G}''',\omega)\right]\chi^{(1)}(\mathbf{q}+\mathbf{G}''',\mathbf{q}+\mathbf{G}',\omega)$$

TDDFT and response functions

Response functions

Dyson equation

$$\left[1 - \chi_0^{(1)}(v + f_{xc})\right]\chi^{(2)} = \chi_0^{(2)}\left[1 + (v + f_{xc})\chi^{(1)}\right]^2 + \chi_0^{(1)} g_{xc}\chi^{(1)}\chi^{(1)}$$

2nd order

Coulomb potential

Non-interacting response function

Exchange-correlation kernel

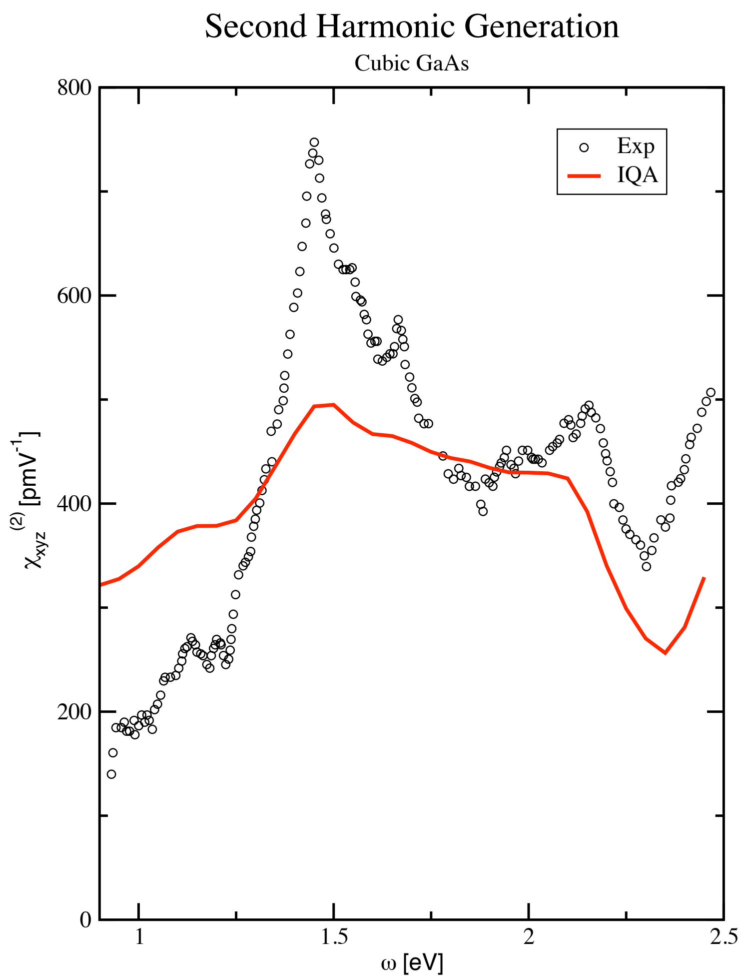
$$f_{xc}(\mathbf{r}, t, \mathbf{r}', t') = \frac{\delta V_{xc}(\mathbf{r}, t)}{\delta \rho(\mathbf{r}', t')}$$

$$g_{xc}(\mathbf{r}, t, \mathbf{r}', t', \mathbf{r}'', t'') = \frac{\delta^2 V_{xc}(\mathbf{r}, t)}{\delta \rho(\mathbf{r}', t')\delta \rho(\mathbf{r}'', t'')}$$

2light

See for instance: : E. K. U. Gross et al,
in Density Functional Theory II,
Topics in Current Chemistry, Vol 181, p81 (1996).

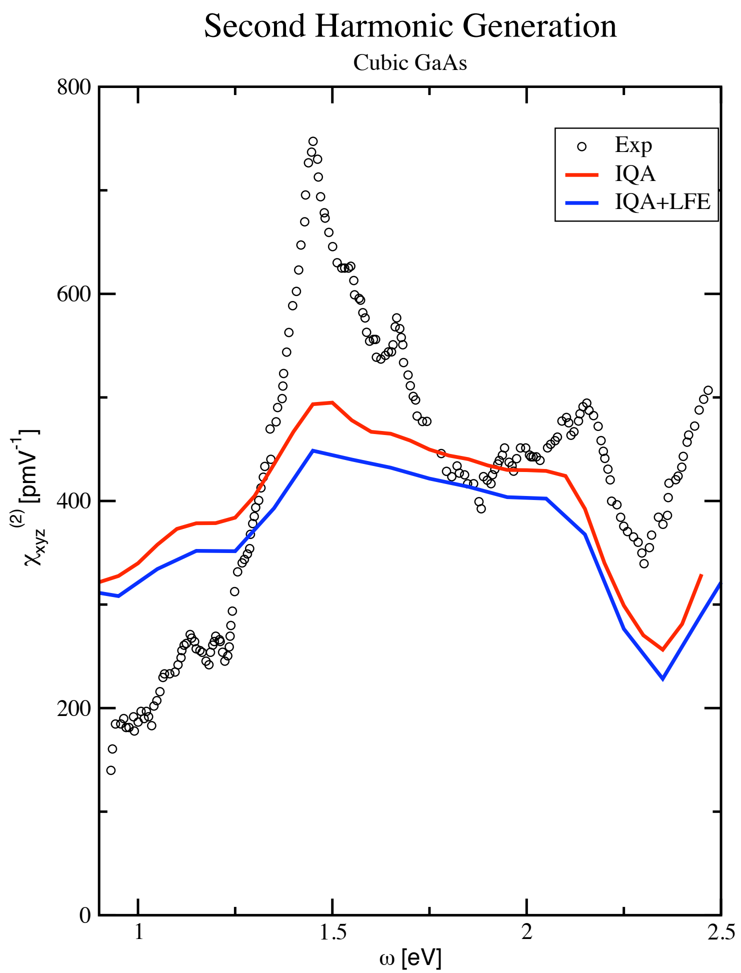
$\chi^{(2)}$ for GaAs



Screening (scissor)

Exp: S. Bergfeld and W. Daum, PRL (2003)

$\chi^{(2)}$ for GaAs

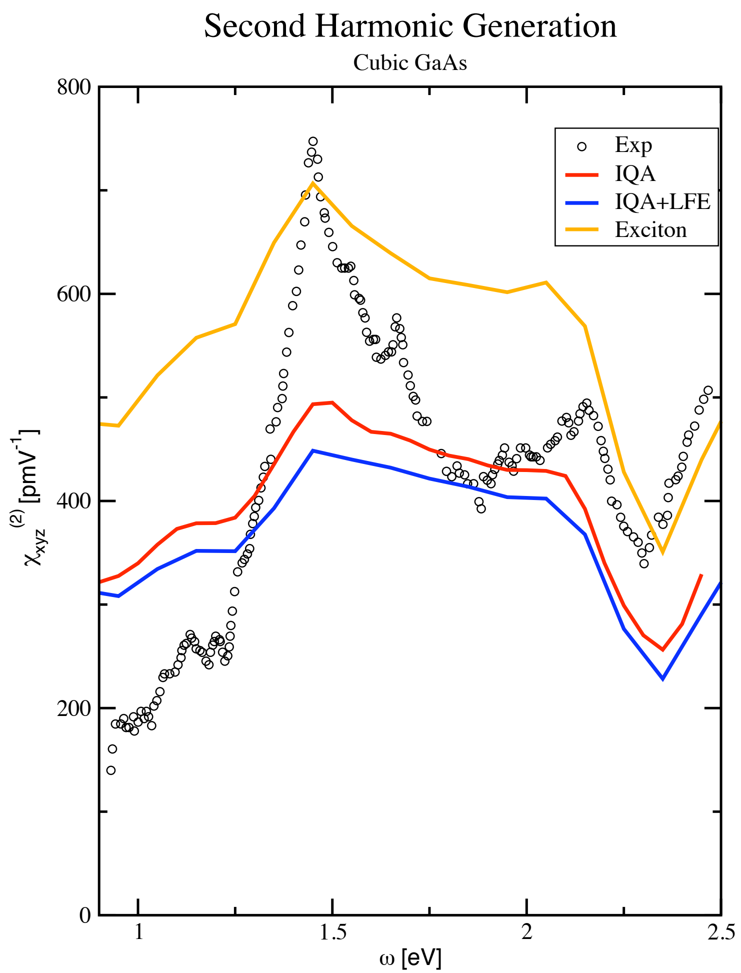


Screening (scissor)

Screening and local fields

Exp: S. Bergfeld and W. Daum, PRL (2003)

$\chi^{(2)}$ for GaAs



Exp: S. Bergfeld and W. Daum, PRL (2003)

Full calculation

Screening (scissor)

Screening and local fields

Exciton (Long range static kernel)

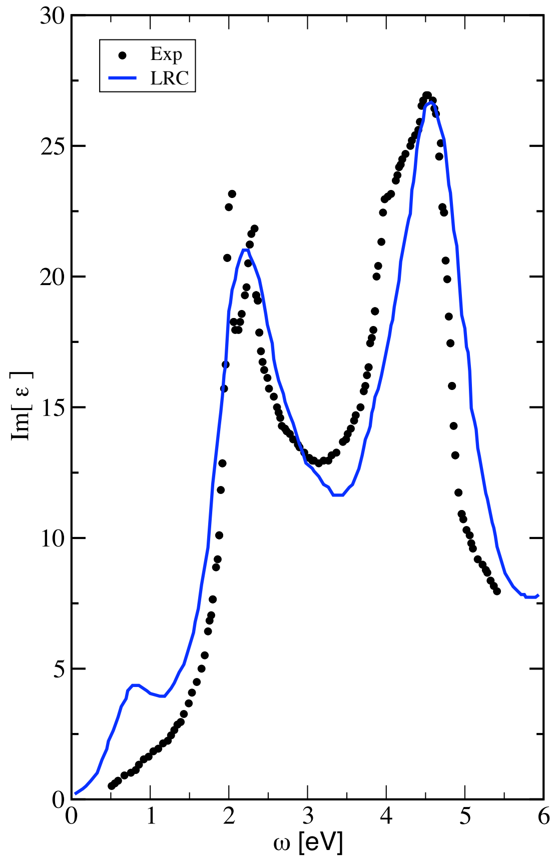
$$f_{xc} = \frac{\alpha}{q^2}$$

ε_M for GaAs

$$\chi^{(2)}_{xyz}(2\mathbf{q},2\omega) = \frac{-i}{12 q_x q_y q_z}\left[\epsilon_M(2\mathbf{q},2\omega)\right]\left[\epsilon_M(\mathbf{q},\omega)\right]^2 \chi^{(2)}(2\mathbf{q},\mathbf{q},\mathbf{q},\omega,\omega)$$

ε_M for GaAs

$$\chi^{(2)}_{xyz}(2\mathbf{q},2\omega) = \frac{-i}{12 q_x q_y q_z}\left[\epsilon_M(2\mathbf{q},2\omega)\right]\left[\epsilon_M(\mathbf{q},\omega)\right]^2 \chi^{(2)}(2\mathbf{q},\mathbf{q},\mathbf{q},\omega,\omega)$$

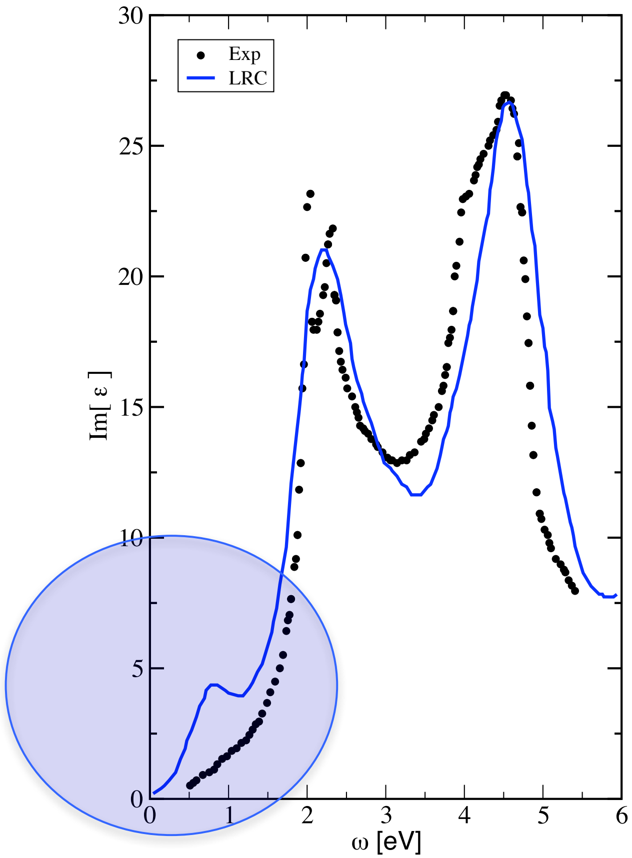


Linear dielectric function

- TDDFT (Long rang kernel)
- Similar results with BSE

ε_M for GaAs

$$\chi^{(2)}_{xyz}(2\mathbf{q}, 2\omega) = \frac{-i}{12 q_x q_y q_z} \left[\epsilon_M(2\mathbf{q}, 2\omega)\right] \left[\epsilon_M(\mathbf{q}, \omega)\right]^2 \chi^{(2)}(2\mathbf{q}, \mathbf{q}, \mathbf{q}, \omega, \omega)$$

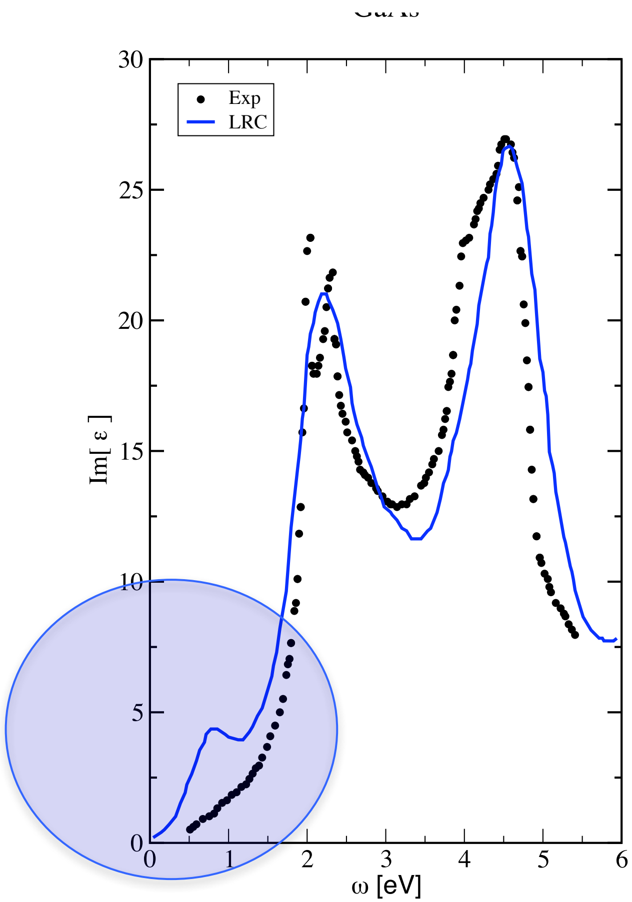


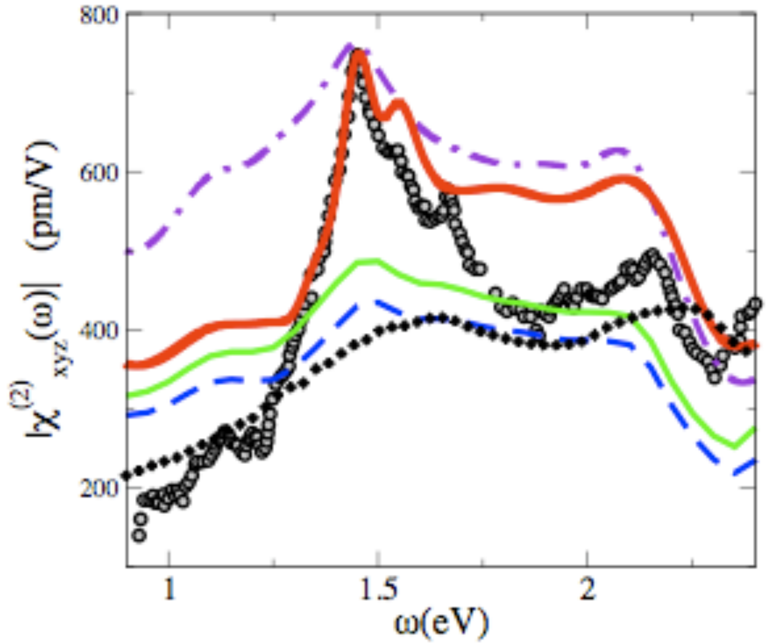
Linear dielectric function

- TDDFT (Long rang kernel)
- Similar results with BSE

In common : static approximation

$\chi^{(2)}$ and ε_M for GaAs





$\chi^{(2)}$ evaluated with the experimental dielectric functions

Good agreement with the experiment

The kernel should be improved in this region

(adiabatic approximation)

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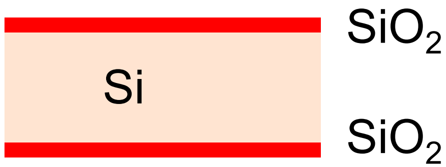
Why static fields?

Investigated charge carrier effects in silicon membranes using a femtosecond laser

WI Ndebeka, PH Neethling, CM Steenkamp, H Stafast, EG Rohwer

Frontiers in Optics, Optical Society of America, 2015



Second harmonic generation at the interface

EFISH : Electric Field Induced Second Harmonic

The time dependence of the resulting EFISH signal is a measure of the rate of trap site generation and population, by both electrons and holes being pumped across the interface

$$P_i(2\omega) = \sum_{jkl} \left[\chi^{(2)}_{ijk}(-2\omega; \omega, \omega) + 3\chi^{(3)}_{ijkl}(-2\omega; \omega, \omega, 0)\, \mathcal{E}_l \right] E_j(\omega)\, E_k(\omega)$$

Why static fields?

$$\chi^{(2)}_{ijk}(-2\omega;\omega,\omega)$$ → Non centro-symmetric materials, without charge accumulation

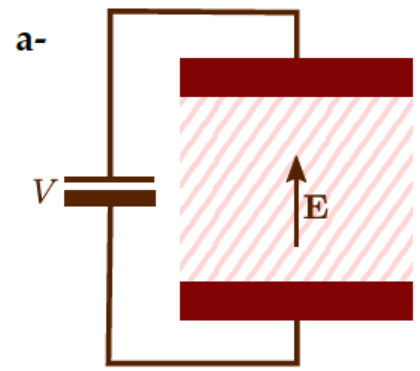
$$\chi^{(2)}_{ijk}(-2\omega;\omega,\omega) + 3\chi^{(3)}_{ijkl}(-2\omega;\omega,\omega,0)\ \mathcal{E}_l$$

Ex : Charge accumulation

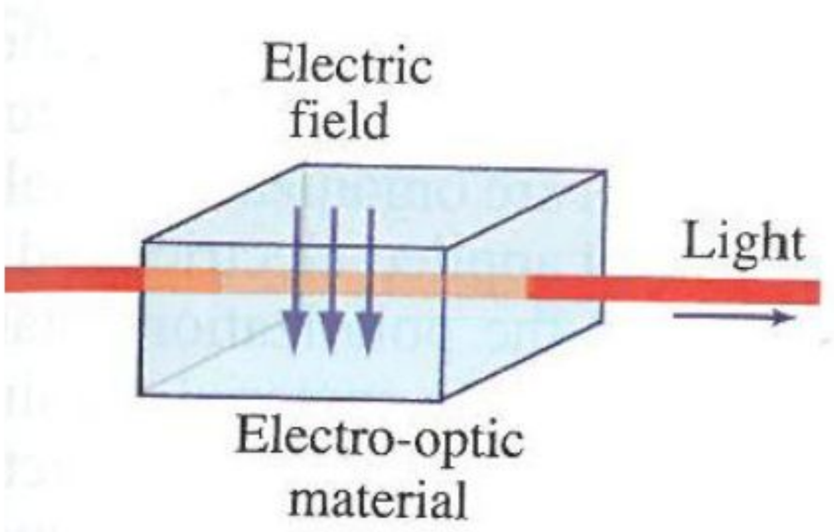
$$\chi^{(3)}_{ijkl}(-2\omega;\omega,\omega,0)\ \mathcal{E}_l$$ → Centro-symmetric materials, with an applied static field

Electro-optic effect : definition

Optical properties of a material can be modified by an applied electric field.

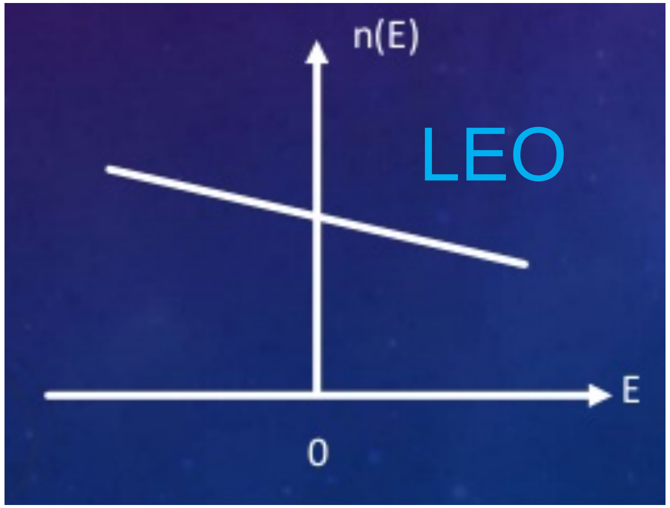


The refractive index changes as a function of the applied field

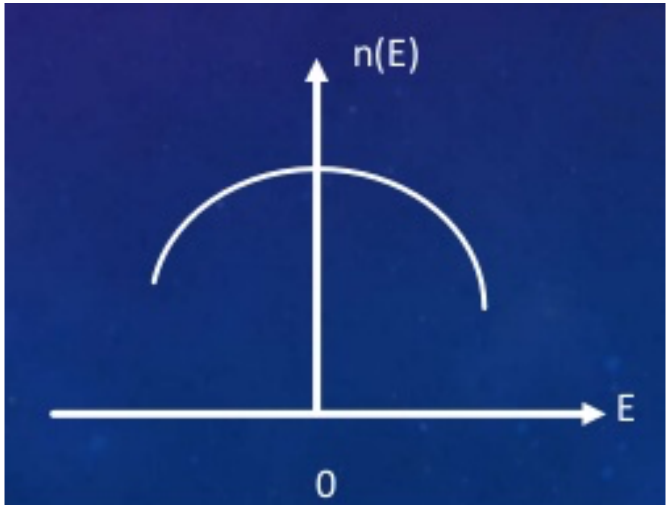


Electro-optic effect : definition

Linear electro-optic effect
(Pockels effect, 1893)



Quadratic electro-optic effect
(DC Kerr effect, 1875)



Discovered well before the laser

Can be described as a nonlinear process inside the material

Linear electro-optic effect (LEO) requires non-centrosymmetric materials

Electro-optic effect : definition

Linear electro-optic effect
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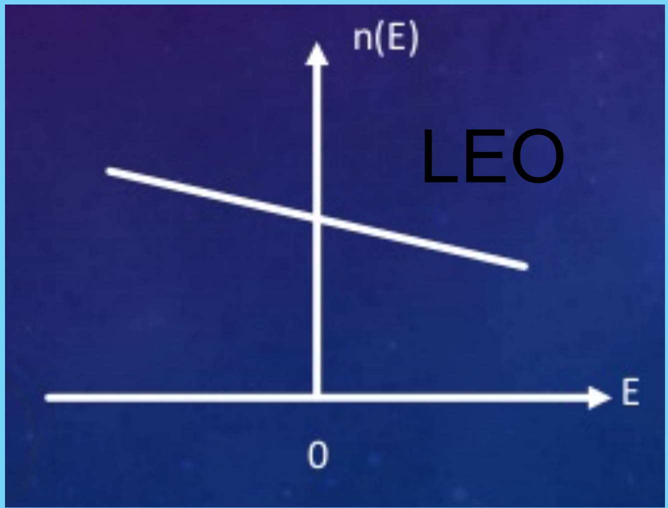

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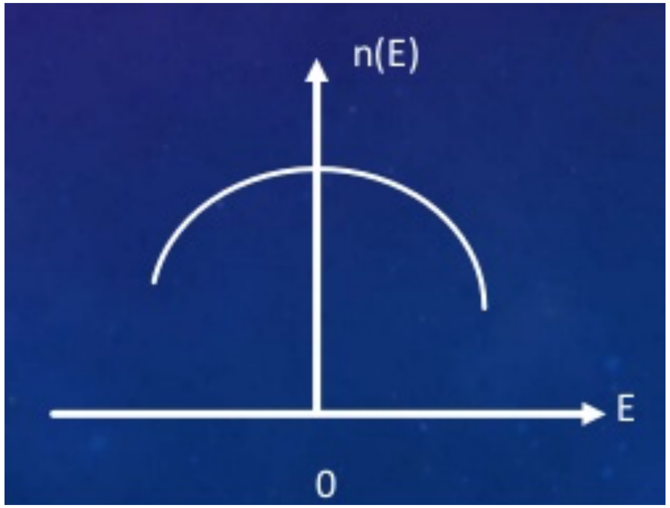

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Potential applications

- Towards high speed optical modulators : Giant electro-optic effect in Si/SiGe coupled quantum wells
- Strained Si photonic crystal waveguide: data processing in all-silicon components
- Quasiperfect phase matching conditions in homogeneous crystals based on controlled birefrengence
- nondestructive and noninvasive probe of surfaces and interfaces in semiconductors

Theoretical description

Linear electro-optic effect

$$\tilde{\epsilon}_{ij} = \epsilon_{ij} + \sum_k 8\pi\,\chi^{(2)}_{ijk}(\omega)\,\mathcal{E}_k$$

Second order susceptibility

$$\chi^{(2)}_{ijk}(\omega) = \lim_{\omega_2 \to 0} \chi^{(2)}_{ijk}(-\omega - \omega_2; \omega, \omega_2)$$

Perturbation theory

« Dressed » Hamiltonian

$$\tilde{H} = H_0 + \mathcal{E}.\mathbf{r}$$

$\tilde{\epsilon}_{ij}$ obtained from the eigenstates of $\tilde{H}$

Non perturbative in $\mathcal{E}$
DC-Kerr effect included

$\mathcal{E}$: Static field

Theoretical description

Linear electro-optic effect

$$\tilde{\epsilon}_{ij} = \epsilon_{ij} + \sum_k 8\pi\, \chi^{(2)}_{ijk}(\omega)\, \mathcal{E}_k$$

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DC-Kerr effect included

$\mathcal{E}$: Static field

Theoretical description

Previous work:

- Mainly performed for SHG
- ω-dependent susceptibility: Independent Particle Approximation [1]
- Static susceptibility : based on the Berry phase (ferroelectric oxides) [2,3]

[1] J. L. P. Hughes and J. E. Sipe, Phys. Rev. B 53, 10751 (1996)
[2] M. Veithen, X. Gonze and P. Ghosez, Phys. Rev. B 71, 125107 (2005)
[3] M. Veithen, PhD thesis, Univesité de Liège, Belgium (2005)

Theoretical description

$$\mathbf{P}^{(2)}(\omega) = 2\,\chi^{(2)}(-\omega;\omega,0)\,\mathbf{E}(\omega)\,\mathcal{E}$$

LEO susceptibility tensor

Modified dielectric tensor

$$\tilde{\varepsilon}_{ij}(\omega) = \varepsilon_{ij}(\omega) + \sum_k 8\pi\ \chi^{(2)}_{ijk}(-\omega;\omega,0)\mathcal{E}_k$$

Impermeability tensor

$$\tilde{\eta}_{ij}(\omega) = \eta_{ij}(\omega) + \sum_k r_{ijk}(\omega)\,\mathcal{E}_k$$

$\eta = \varepsilon^{-1}$

Electro-optic coefficient

$$\chi^{(2)}_{ijk}(-\omega;\omega,0) = -\frac{1}{8\pi}\, n_i^2(\omega) n_j^2(\omega) r_{ijk}(\omega)$$

Theoretical description

General expression for $\chi^{(2)}$ $(\mathbf{q}_1,\omega_1)$ $(\mathbf{q}_2,\omega_2)$

$$\chi_0^{(2)}(\mathbf{q},\mathbf{q}_1,\mathbf{q}_2,\omega_1,\omega_2) = \frac{2}{V}\sum_{n,m,p}\sum_{\mathbf{k}} \frac{\langle\phi_{n,\mathbf{k}}|e^{-i\mathbf{q}\mathbf{r}}|\phi_{m,\mathbf{k}+\mathbf{q}}\rangle\langle\phi_{m,\mathbf{k}+\mathbf{q}}|e^{i\mathbf{q}_1\mathbf{r}_1}|\phi_{p,\mathbf{k}+\mathbf{q}_2}\rangle\langle\phi_{p,\mathbf{k}+\mathbf{q}_2}|e^{i\mathbf{q}_2\mathbf{r}_2}|\phi_{n,\mathbf{k}}\rangle}{E_{n,\mathbf{k}} - E_{m,\mathbf{k}+\mathbf{q}} + \omega_1 + \omega_2 + 2i\eta}$$
$$\times\left(\frac{f_{n,\mathbf{k}} - f_{p,\mathbf{k}+\mathbf{q}_2}}{E_{n,\mathbf{k}} - E_{p,\mathbf{k}+\mathbf{q}_2} + \omega_2 + i\eta} + \frac{f_{m,\mathbf{k}+\mathbf{q}} - f_{p,\mathbf{k}+\mathbf{q}_2}}{E_{p,\mathbf{k}+\mathbf{q}_2} - E_{m,\mathbf{k}+\mathbf{q}} + \omega_1 + i\eta}\right) + [(\mathbf{q}_1,\omega_1) \leftrightarrow (\mathbf{q}_2,\omega_2)]$$

$\omega_2 \to 0$ $\mathbf{q}_2 \to 0$ Special care for the limit: correct ordering

(1) $\mathbf{q}_2 \to 0$

(2) $\omega_2 \to 0$

Brut force numerical evaluation can be problematic

→ k.p perturbation theory

Theoretical description

$$\chi_0^{(2)}(\hat{\mathbf{q}},\hat{\mathbf{q}}_1,\hat{\mathbf{q}}_2,\omega,0)=\frac{1}{V}\sum_{\mathbf{k}}\sum_{n,m,p}\sigma_{n,m,p}\,\hat{\mathbf{r}}_{nm,\mathbf{k}}(\hat{\mathbf{q}})\Bigg[\hat{\mathbf{r}}_{mp,\mathbf{k}}(\hat{\mathbf{q}}_1)\,\hat{\mathbf{r}}_{pn,\mathbf{k}}(\hat{\mathbf{q}}_2)\Bigg(-\frac{f_{np}}{(E_{nm,\mathbf{k}}+\tilde{\omega})E_{np,\mathbf{k}}}$$
$$-\frac{f_{mp}}{(E_{nm,\mathbf{k}}+\tilde{\omega})(E_{pm,\mathbf{k}}+\tilde{\omega})}+\frac{1}{2}\frac{f_{nm}\,E_{np,\mathbf{k}}}{(E_{nm,\mathbf{k}}+\tilde{\omega})(E_{nm,\mathbf{k}})^2}-\frac{1}{2}\frac{f_{nm}\,E_{pm,\mathbf{k}}}{(E_{nm,\mathbf{k}}+\tilde{\omega})^2E_{nm,\mathbf{k}}}$$
$$+\frac{1}{2}\frac{f_{mp}\,E_{np,\mathbf{k}}}{(E_{pm,\mathbf{k}}+\tilde{\omega})(E_{pm,\mathbf{k}})^2}+\frac{1}{2}\frac{f_{mp}\,E_{nm,\mathbf{k}}}{(E_{pm,\mathbf{k}}+\tilde{\omega})^2E_{pm,\mathbf{k}}}+\frac{1}{2}\frac{f_{np}\,(E_{pm,\mathbf{k}}+E_{nm,\mathbf{k}})}{(E_{np,\mathbf{k}}+\tilde{\omega})(E_{np,\mathbf{k}})^2}\Bigg)$$
$$+\hat{\mathbf{r}}_{pn,\mathbf{k}}(\hat{\mathbf{q}}_1)\,\hat{\mathbf{r}}_{mp,\mathbf{k}}(\hat{\mathbf{q}}_2)\Bigg(-\frac{f_{np}}{(E_{nm,\mathbf{k}}+\tilde{\omega})(E_{np,\mathbf{k}}+\tilde{\omega})}-\frac{f_{mp}}{(E_{nm,\mathbf{k}}+\tilde{\omega})E_{pm,\mathbf{k}}}$$
$$+\frac{1}{2}\frac{f_{np}\,E_{pm,\mathbf{k}}}{(E_{np,\mathbf{k}}+\tilde{\omega})(E_{np,\mathbf{k}})^2}-\frac{1}{2}\frac{f_{nm}\,E_{pm,\mathbf{k}}}{(E_{nm,\mathbf{k}}+\tilde{\omega})(E_{nm,\mathbf{k}})^2}+\frac{1}{2}\frac{f_{nm}\,E_{np,\mathbf{k}}}{(E_{nm,\mathbf{k}}+\tilde{\omega})^2E_{nm,\mathbf{k}}}$$
$$+\frac{1}{2}\frac{f_{np}\,E_{nm,\mathbf{k}}}{(E_{np,\mathbf{k}}+\tilde{\omega})^2E_{np,\mathbf{k}}}+\frac{1}{2}\frac{f_{mp}\,(E_{np,\mathbf{k}}+E_{nm,\mathbf{k}})}{(E_{pm,\mathbf{k}}+\tilde{\omega})(E_{pm,\mathbf{k}})^2}\Bigg)\Bigg]$$

Interband transitions

Intraband transitions

Theoretical description

Present status of the calculation:

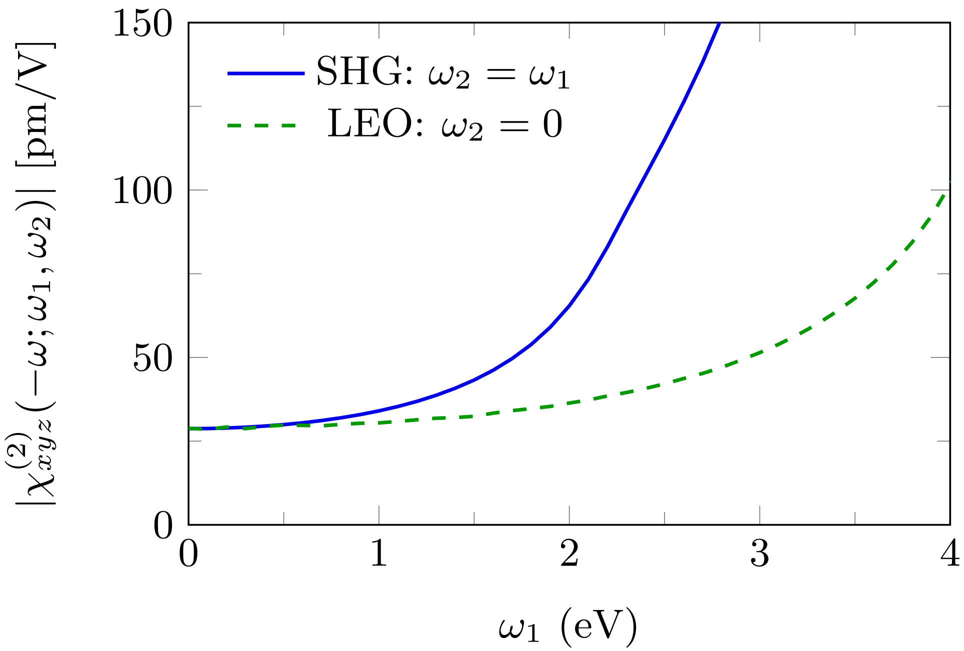
- No local field included (restricted to weakly inhomogeneous materials)
- Screening: Scissor operator (→ additional terms)
- Inclusion of the excitonic effect: long-range kernel

Scalar
Dyson-equation

$$\chi^{(2)}(\mathbf{q},\mathbf{q}_1,\mathbf{q}_2,\omega_1,\omega_2) = \chi_0^{(2)}(\mathbf{q},\mathbf{q}_1,\mathbf{q}_2,\omega_1,\omega_2)\left[1+\frac{\alpha}{q_1^2}\chi^{(1)}(\mathbf{q}_1,\omega_1)\right]\left[1+\frac{\alpha}{q_2^2}\chi^{(1)}(\mathbf{q}_2,\omega_2)\right] + \chi_0^{(1)}(\mathbf{q},\omega)\frac{\alpha}{q^2}\chi^{(2)}(\mathbf{q},\mathbf{q}_1,\mathbf{q}_2,\omega_1,\omega_2)$$

Preliminary results

Electro-optic effect vs Second harmonic generation



3C-SiC

$$\lim_{\omega\to 0} \chi^{(2)}_{\mathrm{SHG}}(-2\omega;\omega,\omega) = \lim_{\omega\to 0} \chi^{(2)}_{\mathrm{LEO}}(-\omega;\omega,0)$$

Electronic and ionic contributions

A surprising experimental result !

GaAs

In the transparent region

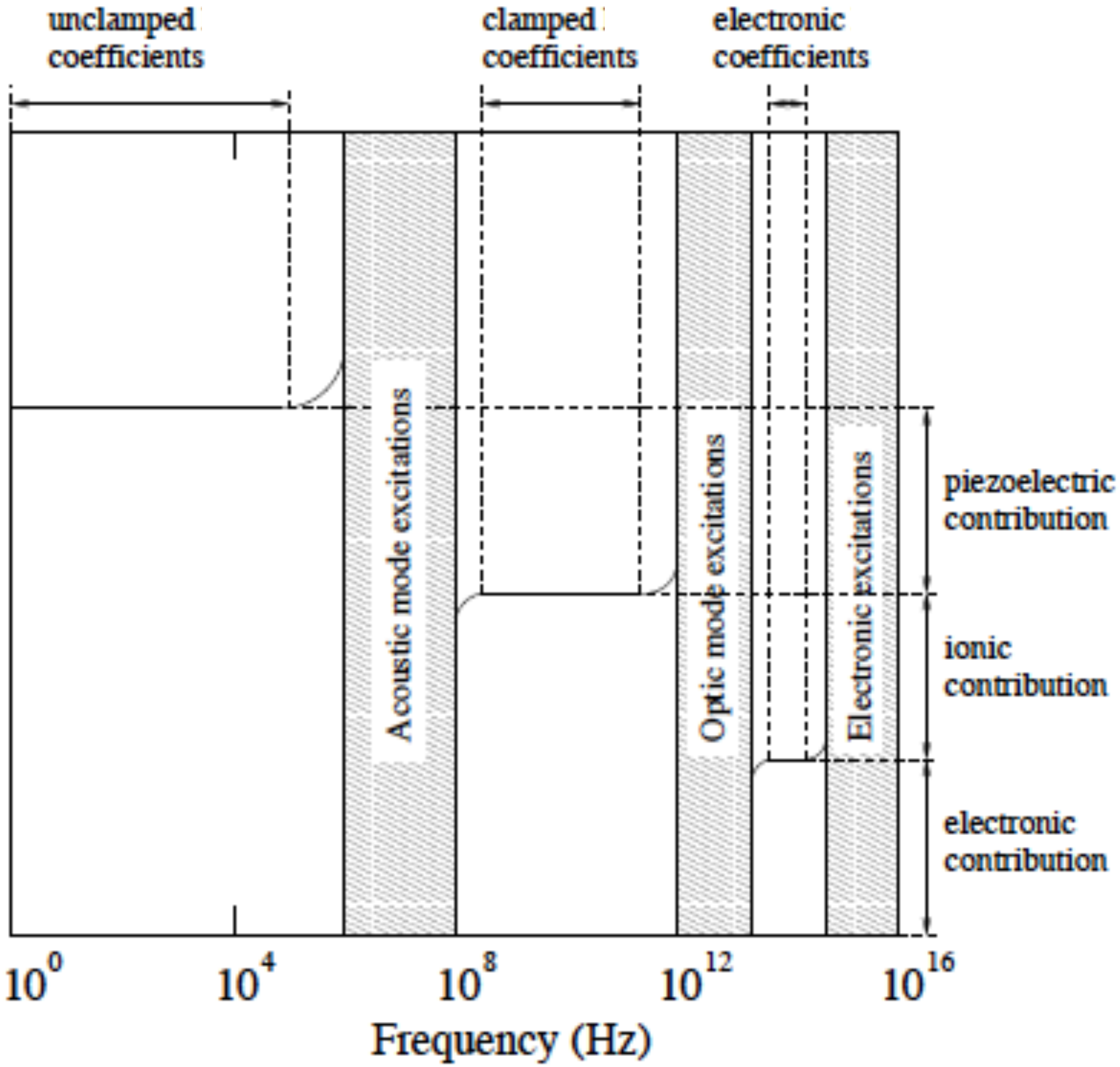
SHG $|\chi^{(2)}_{xyz}(-2\omega;\omega,\omega)| = 200 \text{ pm/V}$

LEO $|\chi^{(2)}_{xyz}(-\omega;\omega,0)| = 100 \text{ pm/V}$

$$\lim_{\omega\to 0} \chi^{(2)}_{\mathrm{SHG}}(-2\omega;\omega,\omega) = \lim_{\omega\to 0} \chi^{(2)}_{\mathrm{LEO}}(-\omega;\omega,0)$$

Expected !!!

Electronic and ionic contributions



10^{-4} eV 1eV

Electronic and ionic contributions

3 contributions:

- electronic
- ionic
- piezzoelectric

Electronic contribution: ω >1eV

direct transition from valence electrons

Ionic contribution: $\omega < 10^{-3}$ eV

ionic lattice displacements

Piezzo contribution:

modification of the shape of the unit cell due to electric forces

Electronic + ionic → clamped coefficient

Electronic + ionic + piezzo→ unclamped coefficient

The relative amplitude of these contributions depends on the frequency range and on the material

Electronic and ionic contributions

A ~~surprising~~ experimental result !

GaAs

In the transparent region

SHG $|\chi^{(2)}_{xyz}(-2\omega;\omega,\omega)|=200 \text{ pm/V}$ Electronic Contribution @1eV

LEO $|\chi^{(2)}_{xyz}(-\omega;\omega,0)|=100 \text{ pm/V}$ Electronic+ionic Contributions @1eV

Faust-Henry coefficient: $$C^{FH} = \frac{\chi^{(2)ionic}}{\chi^{(2)electronic}}$$

Extracted from Raman scattering efficiency of TO and LO phonon modes

$$C^{FH} = -0,51$$

Measured at ω=1eV

Electronic and ionic contributions

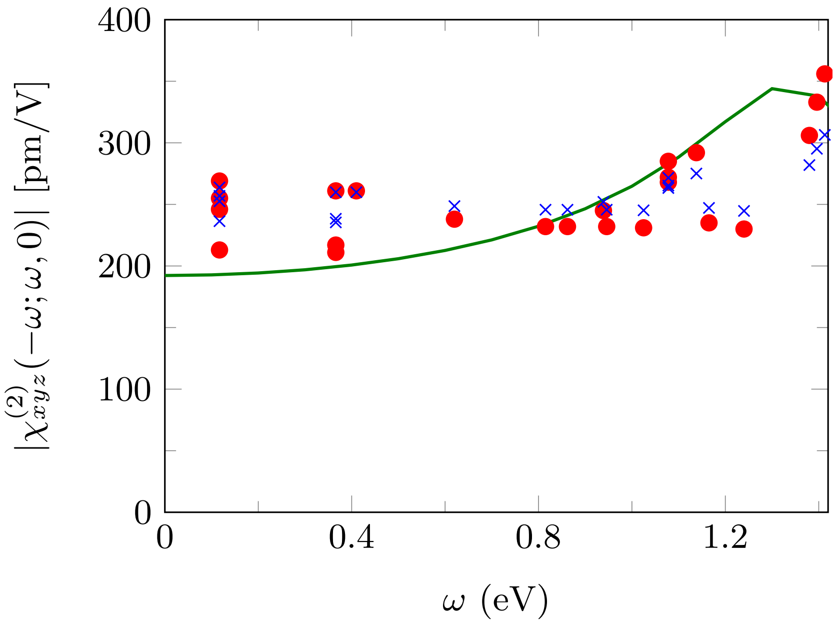
GaAs Electronic contribution

— This work

• and X : Extracted from [1]

•: C^{FH} constant in [0 , 1.5eV]

x : $\chi^{(2)ionic}$ constant [0 , 1.5 eV]



S. Adachi, *GaAs and Related Materials: Bulk Semiconducting and Superlattice Properties* (World Scientific, Teaneck, NJ, 1994).

Electronic and ionic contributions

Cubic SiC: zinc-blende symmetry

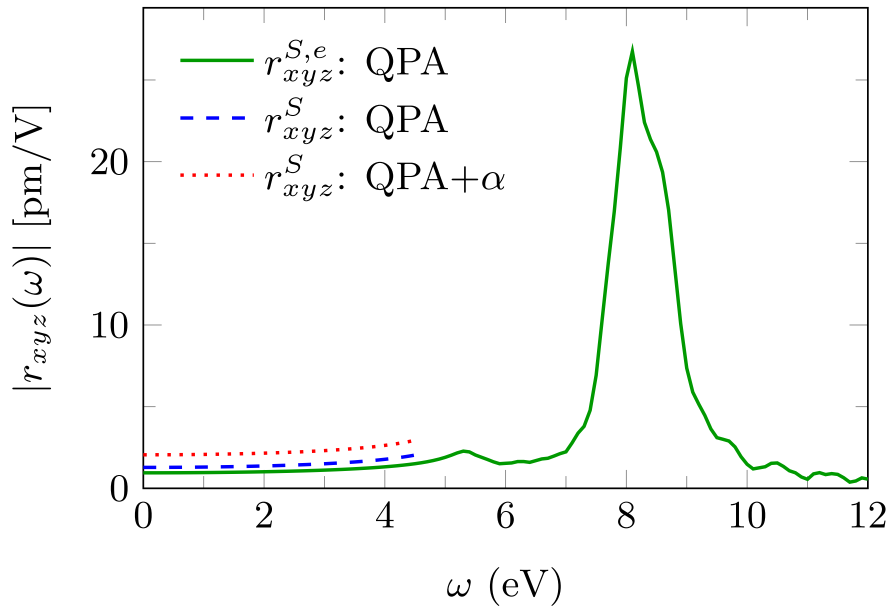
Electro-optic coefficient

$$r_{ijk}(\omega) = 8\pi \frac{\chi^{(2)}_{ijk}(-\omega;\omega,0)}{n_i^2(\omega)n_j^2(\omega)}$$

Electronic contribution

Clamped coefficient (e+i)

Clamped coefficient (α-kernel)



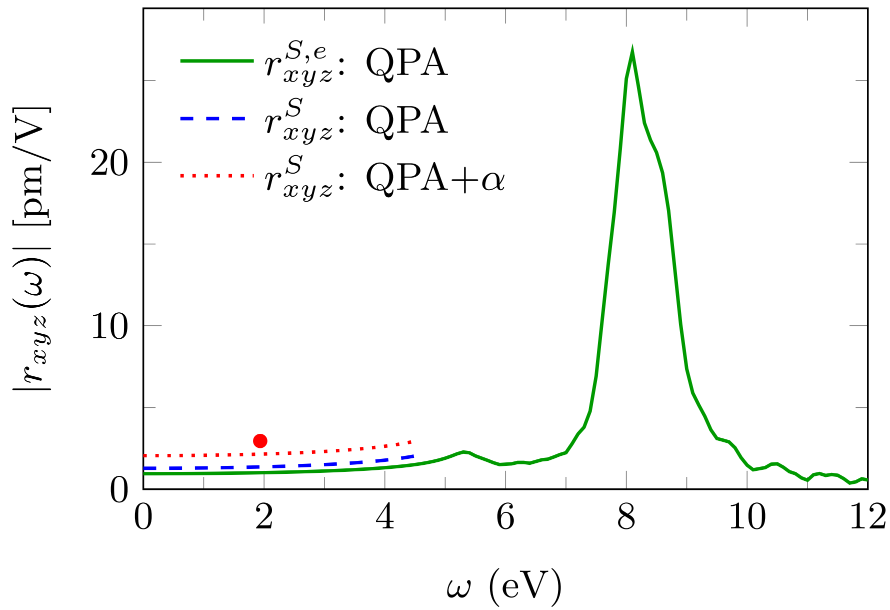
C^{FH}=+0.35

Electronic and ionic contributions

Cubic SiC: zinc-blende symmetry

Electro-optic coefficient

$$r_{ijk}(\omega) = 8\pi \frac{\chi^{(2)}_{ijk}(-\omega;\omega,0)}{n_i^2(\omega)n_j^2(\omega)}$$



Electronic contribution

Clamped coefficient (e+i)

Clamped coefficient (α-kernel)

C^{FH}=+0.35

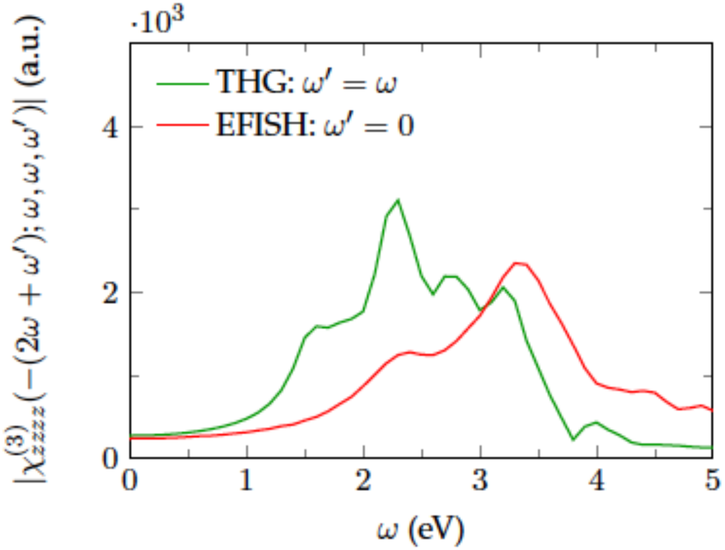
	α (a.u.)	$\chi^{(2)}_{xyz}$ (pm/V)	r^S_{41} (pm/V)
Expt.			2.7 ± 0.5
QPA	0	24.8	1.37[1]
QPA+α_{static}	0.3	39.1	2.15[1]
QPA+α	0.5	55.2	2.40[1]

G. L. Harris, *Properties of Silicon Carbide* (IET, Stevenage, 1995).

EFISH

Electronic part of the $\chi^{(3)}$

Cubic SiC: zinc-blende symmetry



Importance of the ionic contribution?

Nothing like the Faust-Henry coefficient is available

What's next?

* Electronic contributions for LEO and EFISH: done!

* Precise evaluation of the Faust-Henry coefficient for LEO : $C^{FH} = \frac{\chi^{(2)ionic}}{\chi^{(2)electronic}}$

Experimental values only for simple materials available

Theoretical values for ω=0 (poor agreement)

Frequency-dependent coefficient required

Material design for optical modulators

TeraHertz Generation

$$\chi^{(2)}_{ijk}(-\omega;\omega,0) = \chi^{(2)}_{kij}(0;-\omega,\omega)$$

* Importance of the ionic contribution for EFISH

Casting

Formalism and GaAs : E. Luppi and H. Hübener (PhD)
LSI, Ecole Polytechnique

LEO and EFISH : L. Prussel (PhD)
LSI, Ecole Polytechnique

Thank you for your attention