NLO within WIEN2k

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TU Wien

Overview

- Introduction...
- Method...
- Program Modules...
- Examples...

Introduction

What is Nonlinear Optic ?

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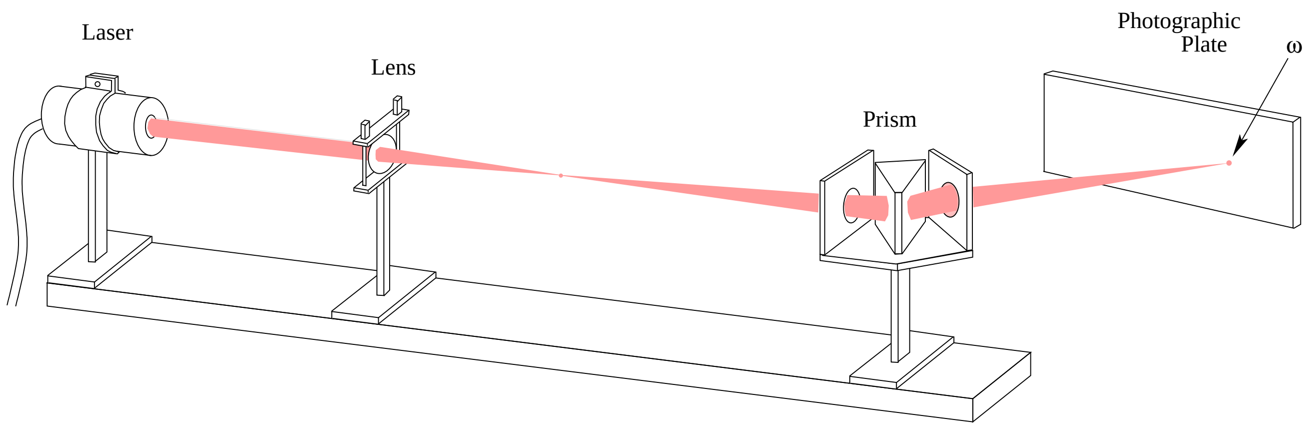
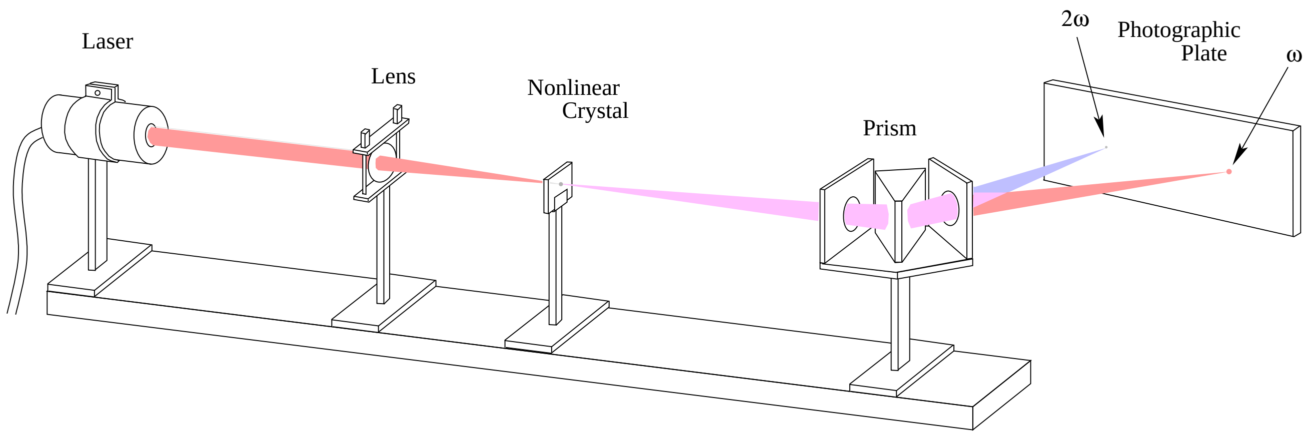


First NLO-Experiment by Franken (1961)

Introduction

What is Nonlinear Optic ?



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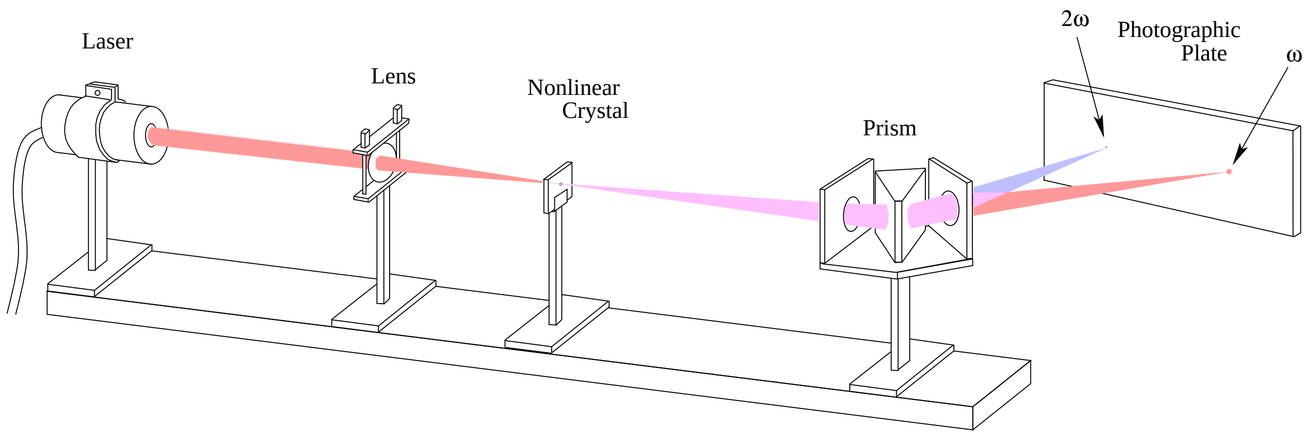

First NLO-Experiment by Franken (1961)

Second Harmonic Generation (SHG)

Introduction

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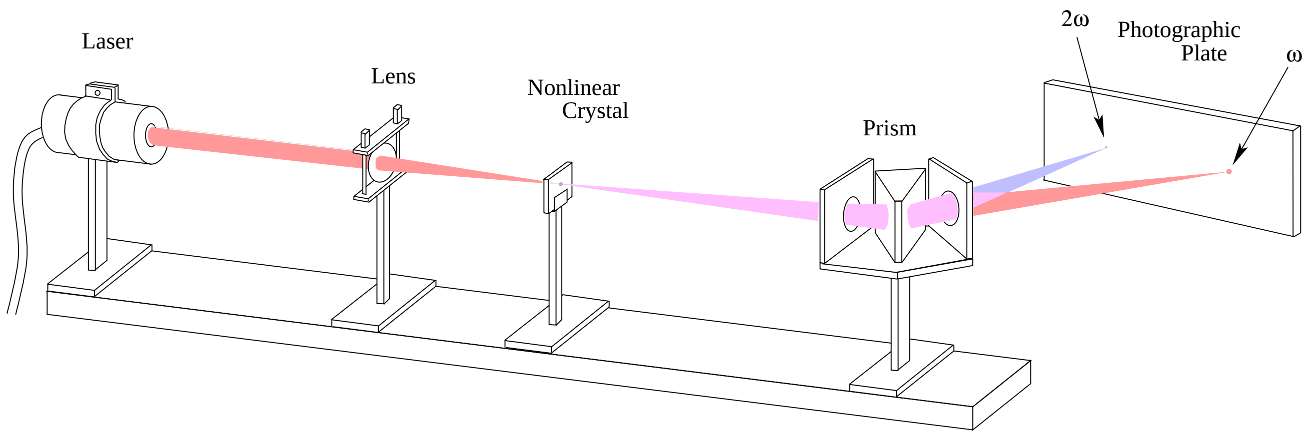

First NLO-Experiment by Franken (1961)

Second Harmonic Generation (SHG)

- 3 Photon Process: $\omega \quad + \quad \omega \quad \longrightarrow \quad 2\omega$
- High Light Intensity $\Rightarrow$ Focus
- Only in specific Crystals

Introduction

Which Crystals show SHG ?

Introduction

Which Crystals show SHG ?

- Bulk Material with Non Centrosymmetric Structure
- Every Surface is Non Centrosymmetric per Definition
- Typical NLO-Crystals used in Applications, i.e. SHG

Crystal	Formula	Class
BBO	β-BaB_2O_4	3
Lithiumniobat	$LiNbO_3$	$3\,3$
Quartz	α-SiO2	$3\,2$
KDP	KH_2PO_4	$\bar{4}\,2\,m$
Banana	$Ba_2NaNb_5O_{15}$	$2\,m\,m$
KTP	$KTiOPO_4$	$2\,m\,m$

Introduction

What is our Motivation ?

- Highly accurate DFT Program WIEN2k exists
- Understanding SHG on a microscopical scale

  $\Rightarrow$ Additional Package for SHG
- Experiments on the Computer
- Optimizing Crystals for SHG

Method

Source of the Nonlinearity

Method

Source of the Nonlinearity

- Interaction between Light and Matter

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- Incident Light $\Rightarrow$ Polarization of Media

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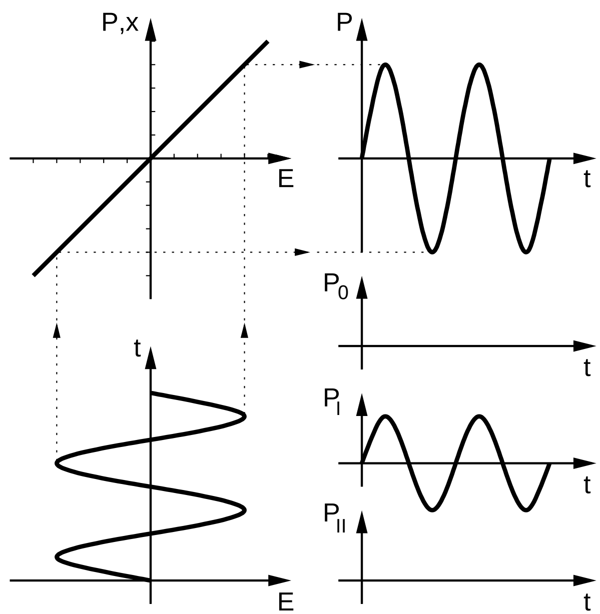

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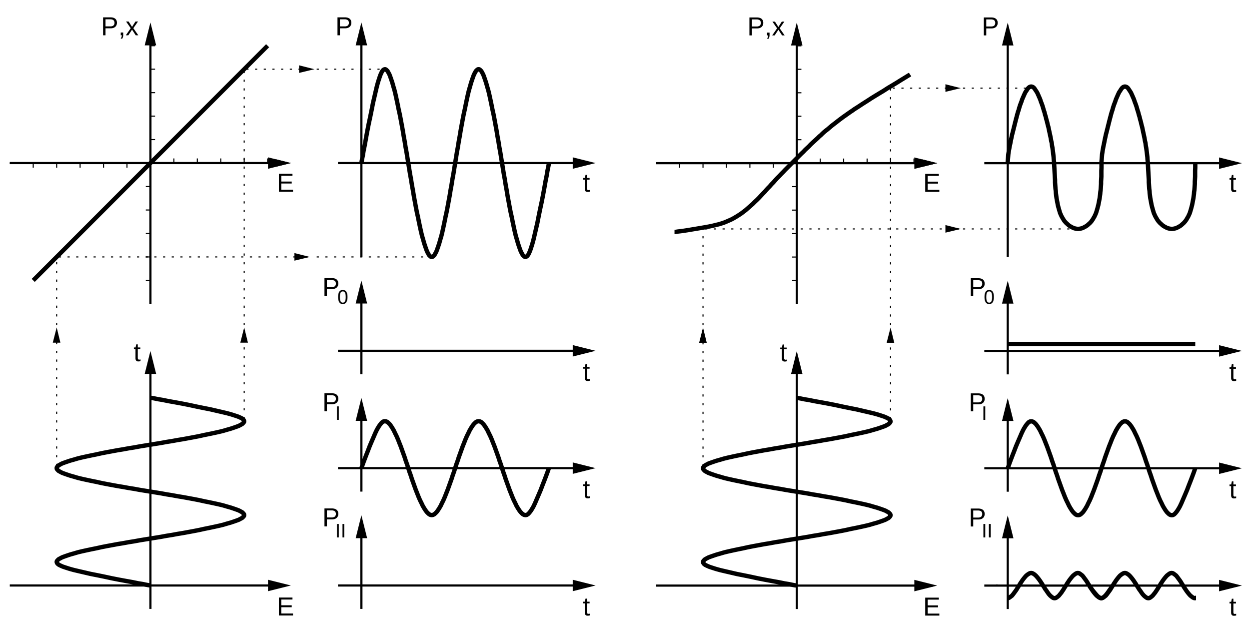

Method

Our Approach

Method

Our Approach

- Expansion into a Power Series

$$P^a = \underbrace{\sum_b \chi_I^{ab} E^b}_{LO} + \underbrace{\sum_b \sum_c \chi_{II}^{abc} E^b E^c + \cdots}_{NLO}$$

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- Perturbation Theory in Density Formalism

  (PRB **48**, 11705 (1993))

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- Perturbation Theory in Density Formalism (PRB **48**, 11705 (1993))
    - Formalism works only for Filled Bands

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  - Formalism works only for Filled Bands
  - Separate Susceptibility into several Parts

$$\chi_{II}^{abc}(-2\omega;\omega,\omega) = \underbrace{\chi_{ter}^{abc}(-2\omega;\omega,\omega)}_{interband} + \underbrace{\chi_{tra}^{abc}(-2\omega;\omega,\omega)}_{intraband}$$

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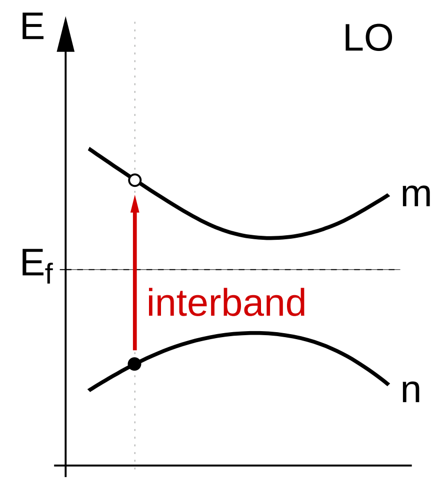

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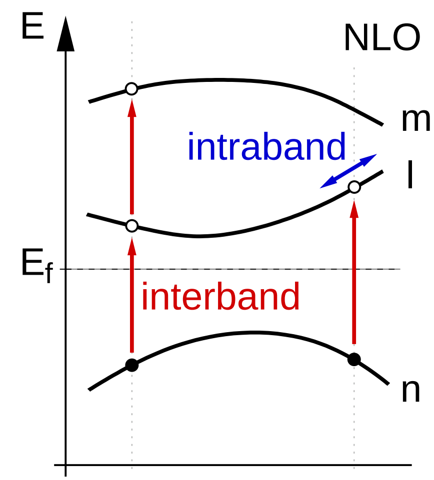

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Interband Contribution

$$\chi^{abc}_{ter}(-2\omega;\omega,\omega) = \frac{e^3}{8\pi^3\hbar^2}\int_{\mathrm{FBZ}} d^3k \sum_{nml} \frac{r^a_{nm}(r^b_{ml}r^c_{ln} + r^c_{ml}r^b_{ln})}{\omega_{ln} - \omega_{ml}} \times \left[\frac{2f_{nm}}{\omega_{mn} - 2\omega} + \frac{f_{ln}}{\omega_{ln} - \omega} + \frac{f_{ml}}{\omega_{ml} - \omega}\right]$$

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  $\Rightarrow$ Double Resonance (DR)

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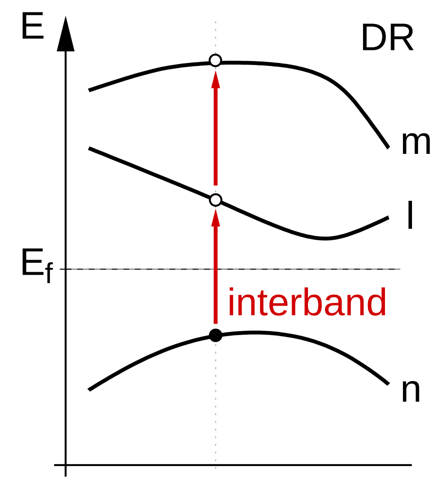

Intraband Contribution

$$
\begin{aligned}
\chi^{abc}_{tra}(-2\omega;\omega,\omega) \quad &= \quad i\,\frac{e^3}{8\pi^3\hbar^2}\int_{\mathrm{FBZ}} d^3k\,\sum_{nm} f_{nm}\Big[\frac{2r^a_{nm}(r^b_{mn;c}+r^c_{mn;b})}{\omega_{mn}(\omega_{mn}-2\omega)} \\
&+\frac{(r^a_{nm;c}r^b_{mn}+r^a_{nm;b}r^c_{mn})}{\omega_{mn}(\omega_{mn}-\omega)}-\frac{(r^b_{nm;a}r^c_{mn}+r^c_{nm;a}r^b_{mn})}{2\omega_{mn}(\omega_{mn}-\omega)} \\
&+\left(\frac{1}{\omega_{mn}-\omega}-\frac{4}{\omega_{mn}-2\omega}\right)\frac{r^a_{nm}}{\omega^2_{mn}}(r^b_{mn}\Delta^c_{mn}+r^c_{mn}\Delta^b_{mn})\Big]
\end{aligned}
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- Contribution of the mixed interband and intraband Processes

Intraband Contribution

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- Contribution of the mixed interband and intraband Processes
  - Modulation of the linear Response by the intraband Motion

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- Contribution of the mixed interband and intraband Processes
  - Modulation of the linear Response by the intraband Motion
  - Modification of the intraband Motion by the interband Polarization

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- $r^a_{nm;b}$ and Δ^b_{mn} are k-Point dependent

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  - Modulation of the linear Response by the intraband Motion
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- $r^a_{nm;b}$ and Δ^b_{mn} are k-Point dependent
- No Divergences are remaining

Method

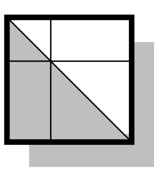
Algorithm

Method

Algorithm

- Starting with the Momentum Matrix Elements (MME)

$p_{nm}(\mathbf{k})$



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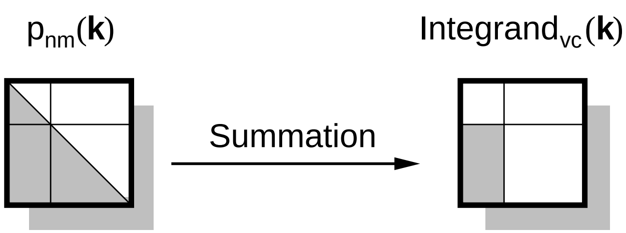
- Starting with the Momentum Matrix Elements (MME)
- Evaluate the Integrands

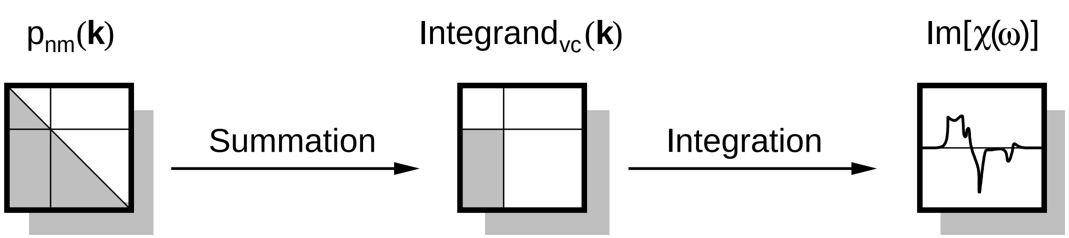


Method

Algorithm

- Starting with the Momentum Matrix Elements (MME)
- Evaluate the Integrands
- Integrate over the Energy Surface

  $\Rightarrow$ Imaginary Part of Susceptibility



Method

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- Starting with the Momentum Matrix Elements (MME)
- Evaluate the Integrands
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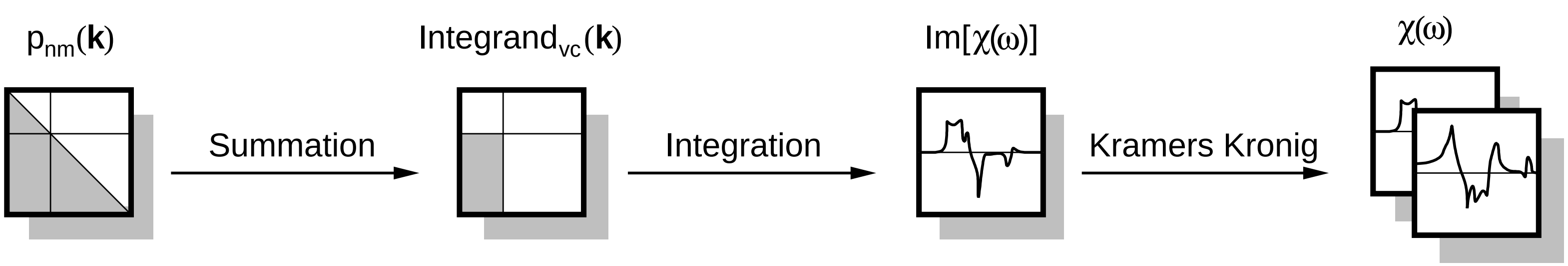
  ⇒ Imaginary Part of Susceptibility
- Apply the Kramers Kronig Relation

  ⇒ Real Part of Susceptibility



Method

Scissor Correction in SHG

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- Correction to the Band Gap Problem in DFT

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- Lift the unoccupied Bands by a constant Energy

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- Correction to the Band Gap Problem in DFT
- Lift the unoccupied Bands by a constant Energy
- Linear Response (LR)
  - Shift the Spectrum

    $\Rightarrow$ Absorption starts at Band Gap Energy

Method

Scissor Correction in SHG

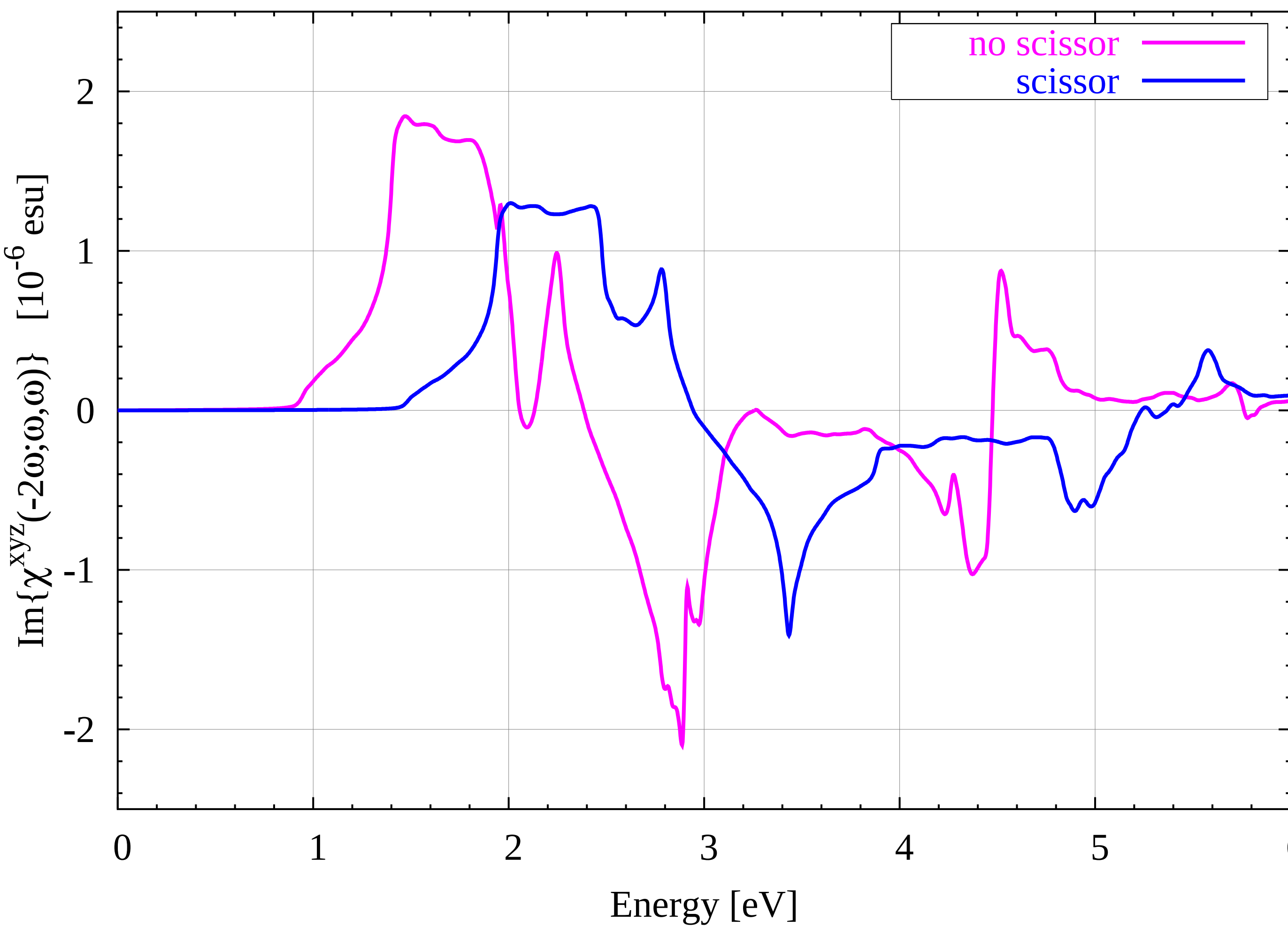
- Correction to the Band Gap Problem in DFT
- Lift the unoccupied Bands by a constant Energy
- Linear Response (LR)
  - Shift the Spectrum

    $\Rightarrow$ Absorption starts at Band Gap Energy
- SHG
  - Shift and Scale the Spectrum

    $\Rightarrow$ Starts at half Band Gap Energy
  - Close Connection to DR

Method

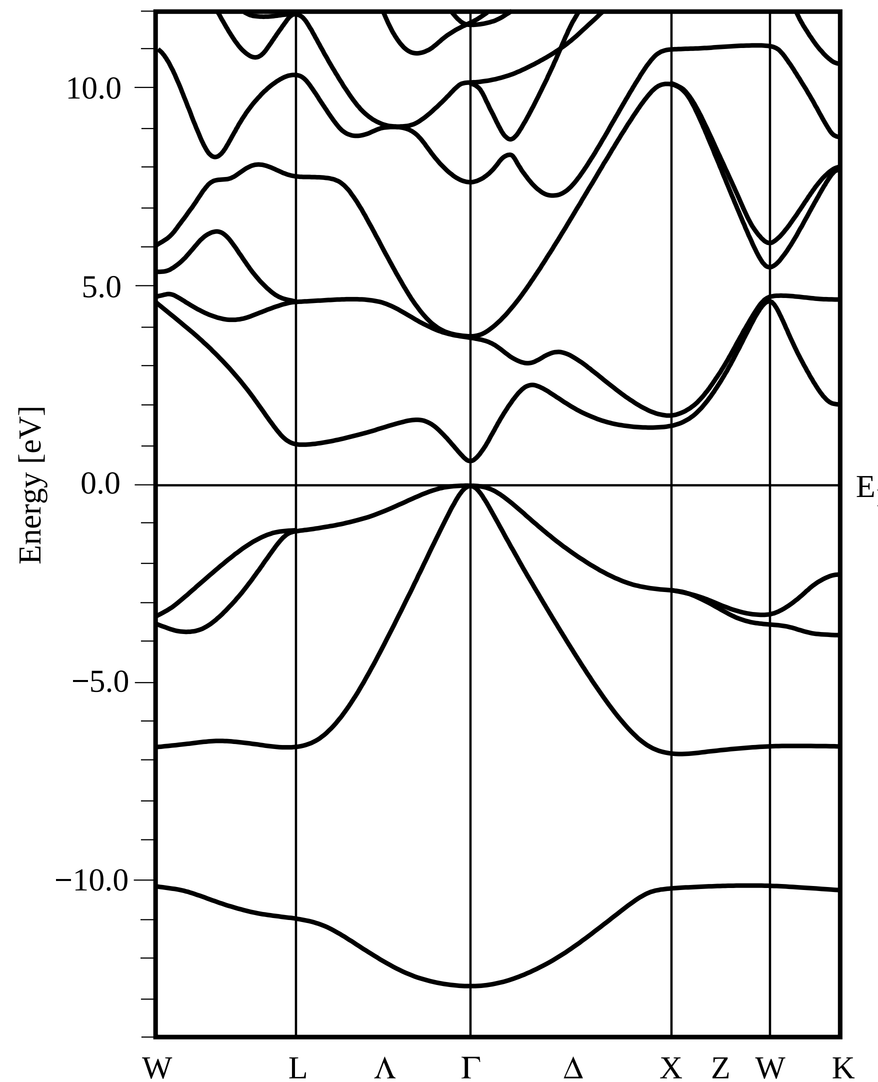
Scissor Correction in SHG



Method

Band Structure GaAs

- No Scissor Correction, GGA



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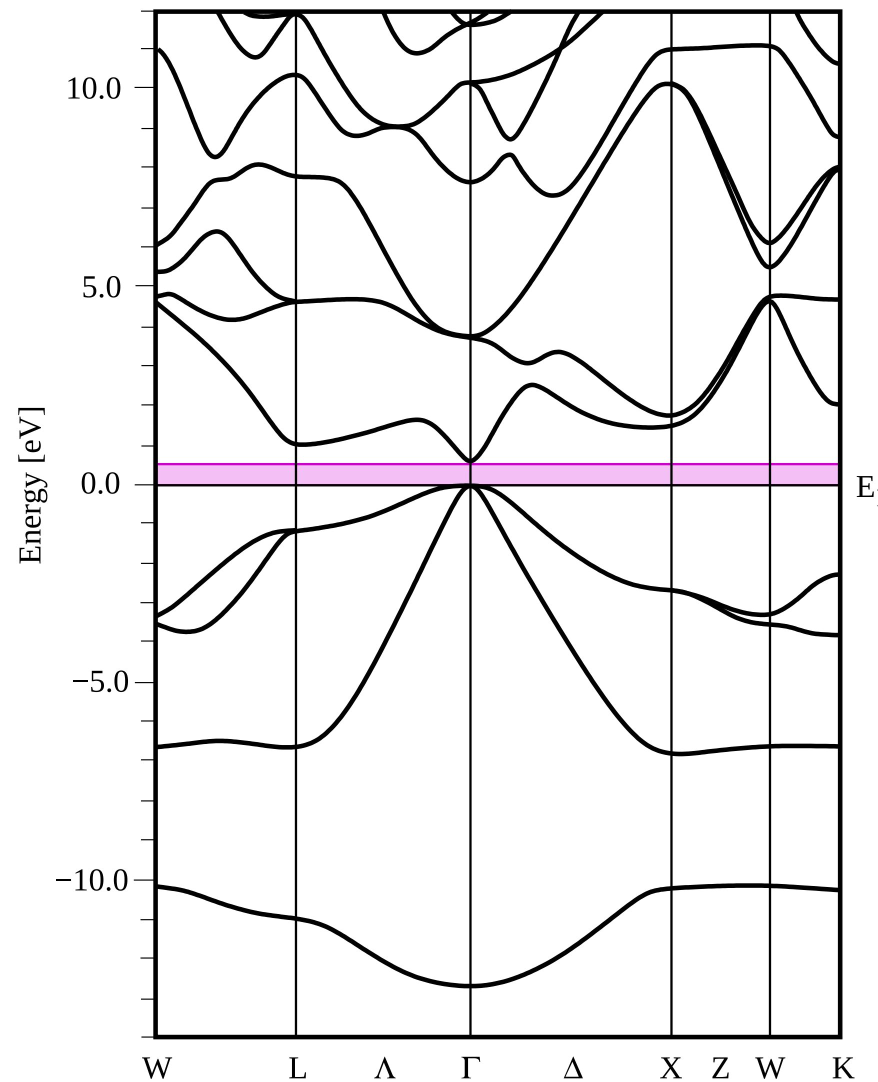
  $\Rightarrow$ GGA Band Gap 0.51 eV



Method

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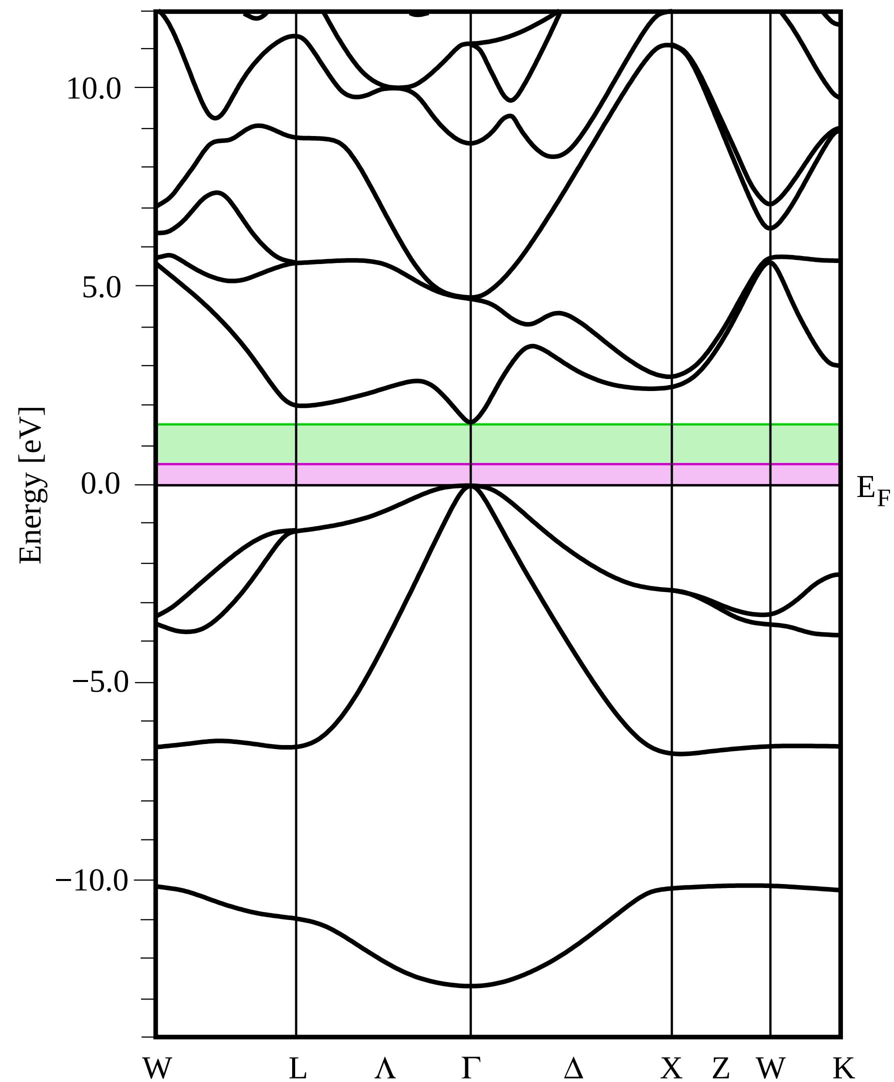
  $\Rightarrow$ corrected Band Gap 1.52 eV



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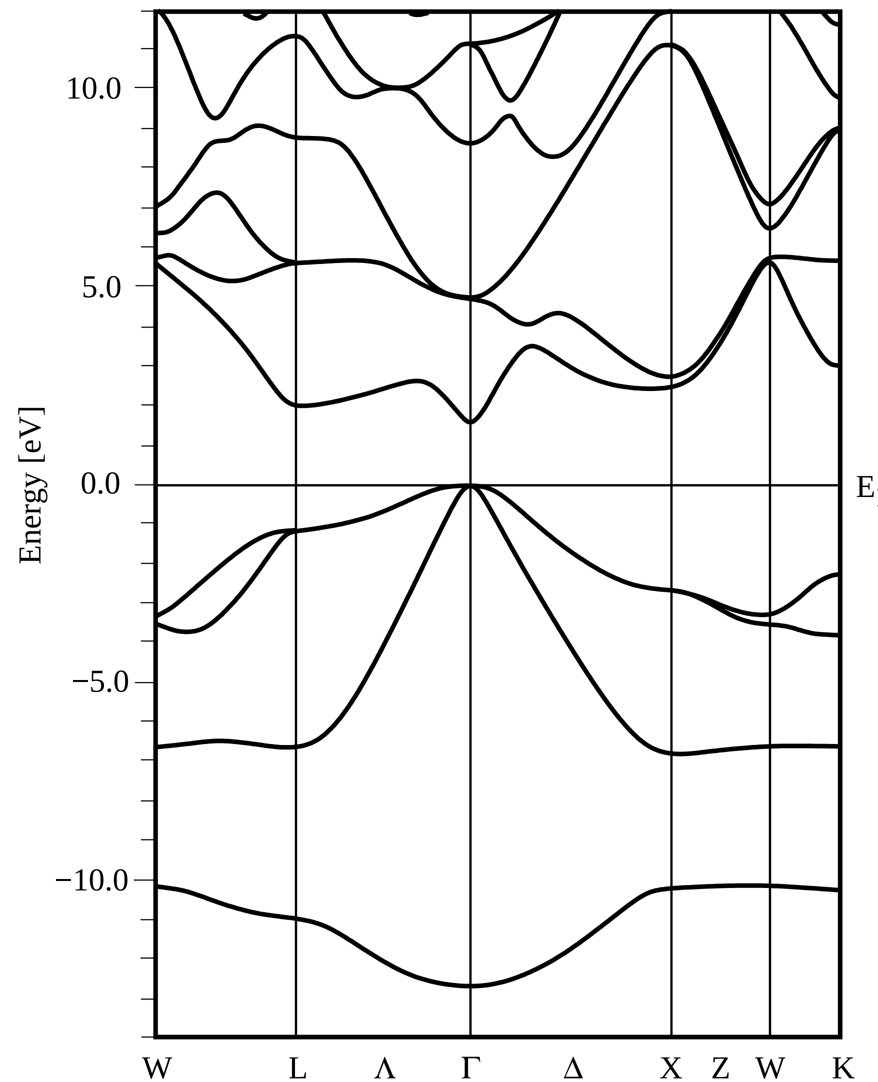
- Double Resonances at



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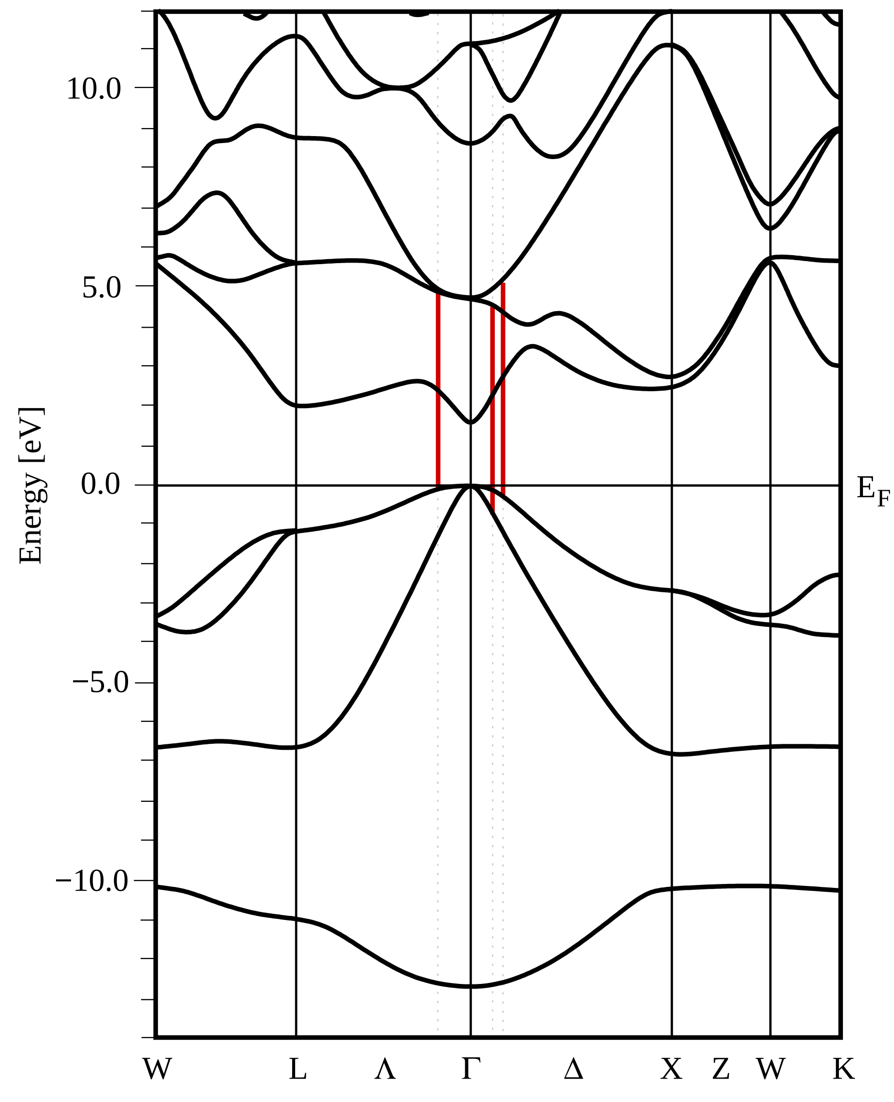
- With Scissor Correction 1.01 eV
  $\Rightarrow$ corrected Band Gap 1.52 eV
- Double Resonances at
  - 2.1-2.6 eV



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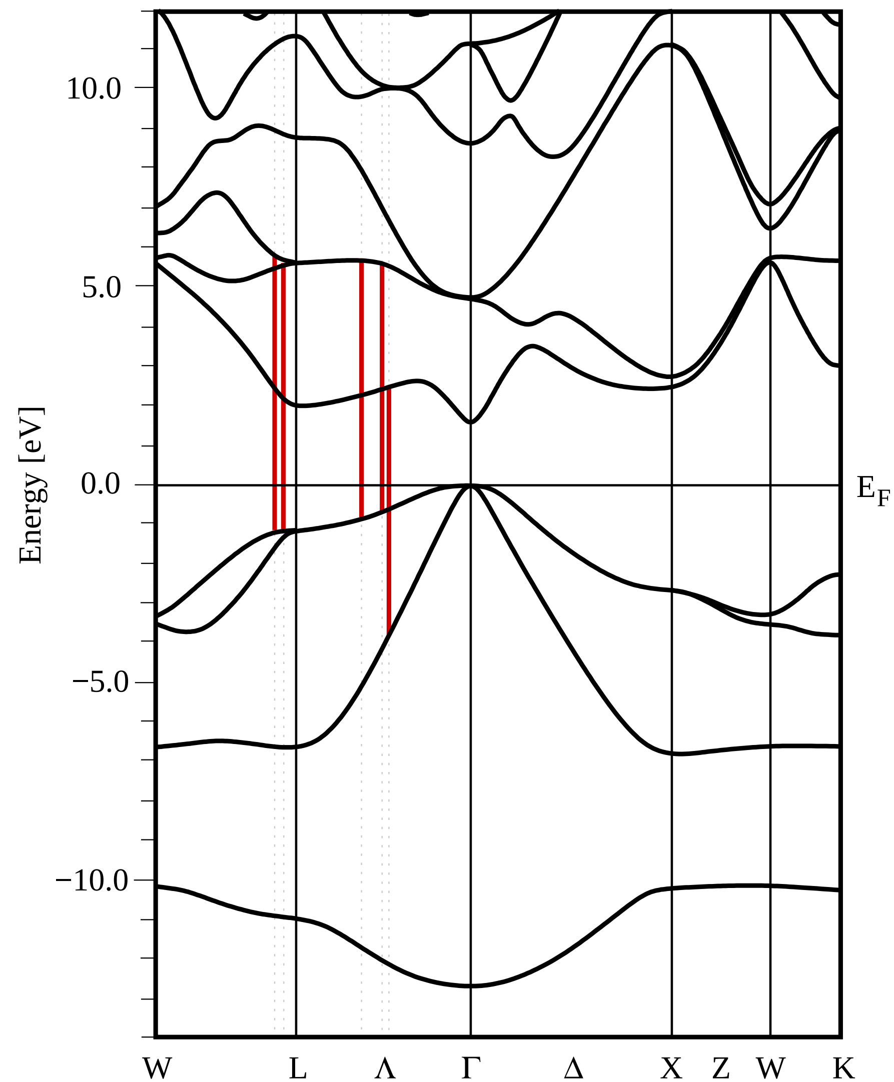
- Double Resonances at
  - 2.1-2.6 eV
  - 3.2-3.7 eV



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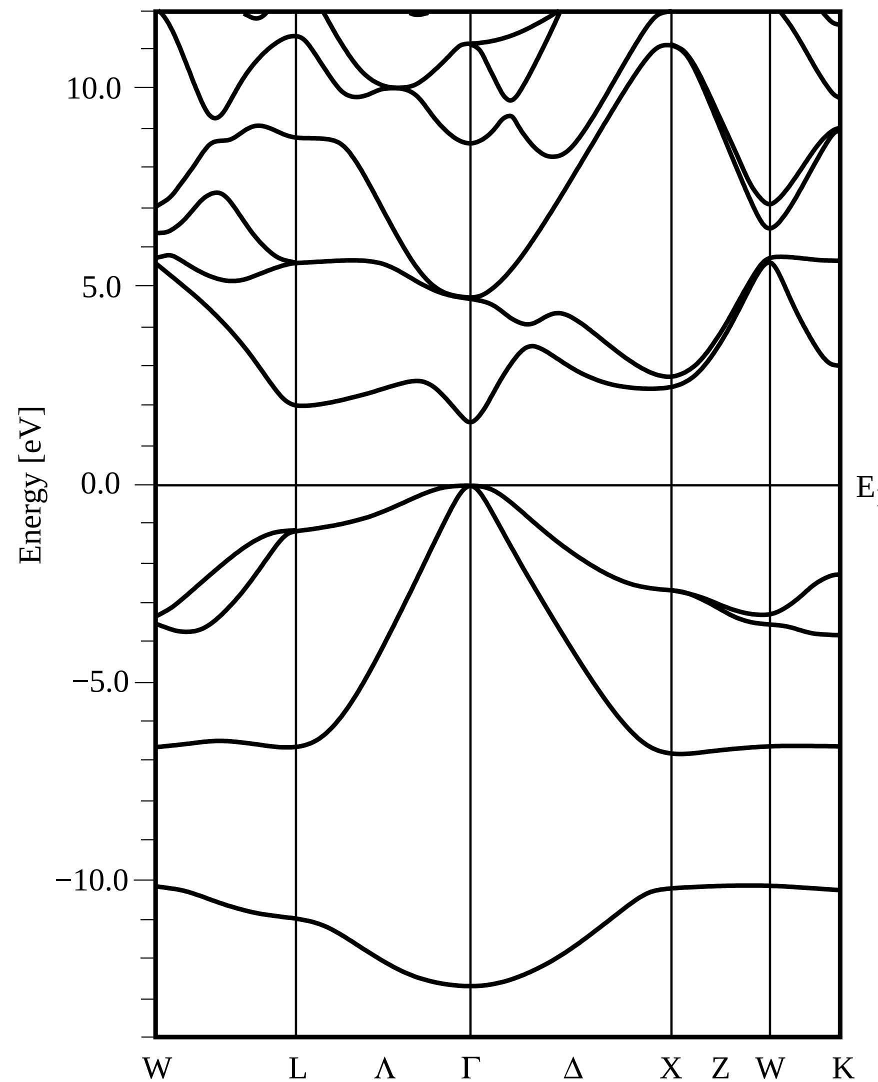
- Double Resonances at
  - 2.1-2.6 eV
  - 3.2-3.7 eV

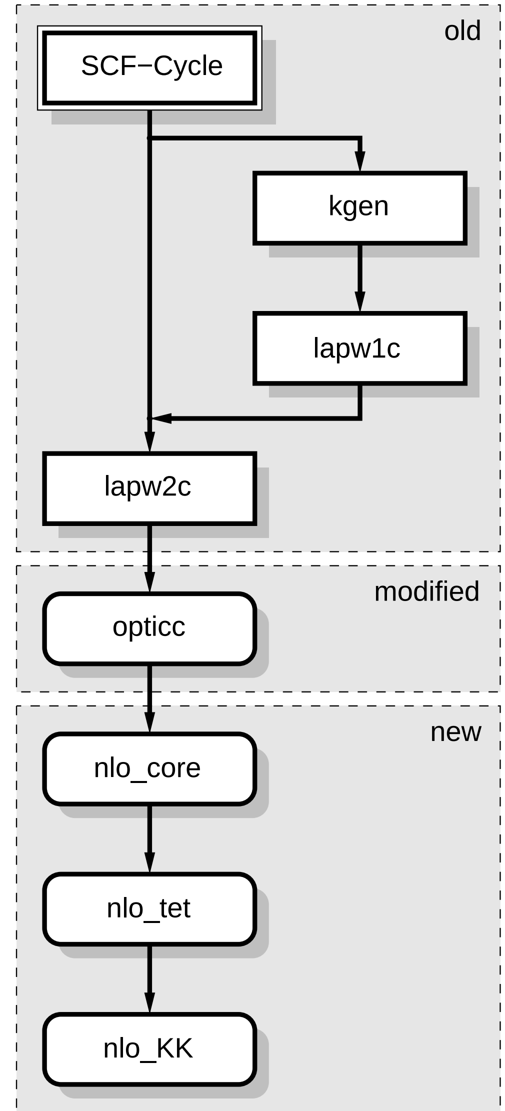
Note: This is only a 1d-Picture !



Program Modules

Program Flow

- Similar to the LO-Package
- Complex Program Version



Program Modules

Program Flow

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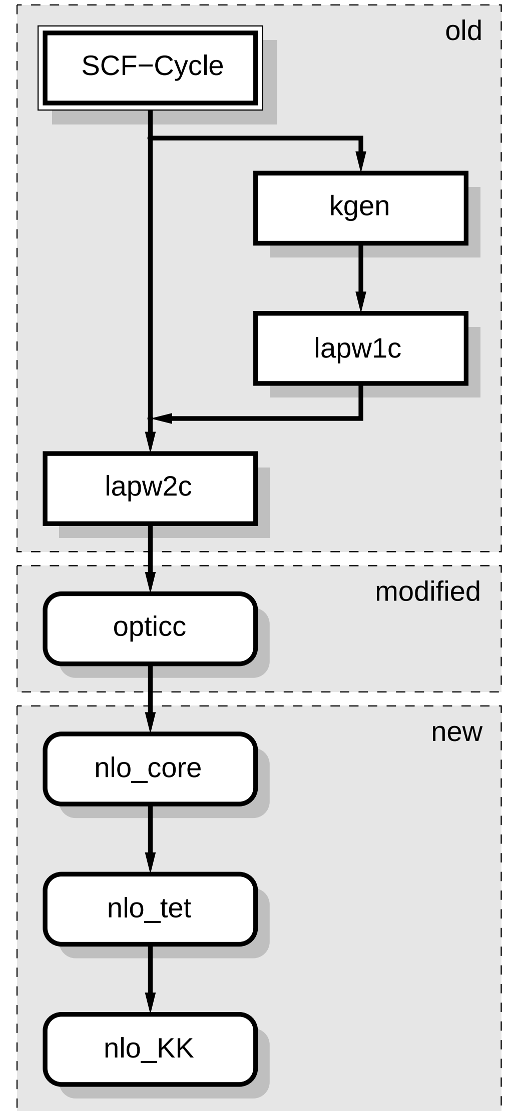
- Complex Program Version

Starting Point

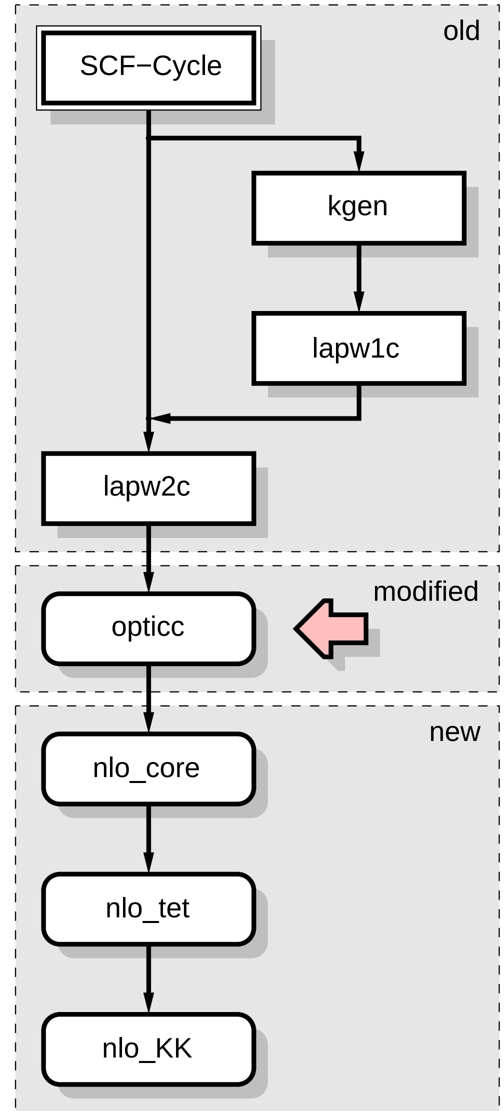
- Well Converged Potential
- Increase Number of k-Points
- Generate Eigenvectors and Eigenvalues
- Calculates the Fermi Energy



Program Modules

Module: opticc

- Compute MMEs
  - For each Band-Combination
  - For every k-Point
- Additional Working Mode
  - Output of pure MMEs
  - Output Symmetry Matrices

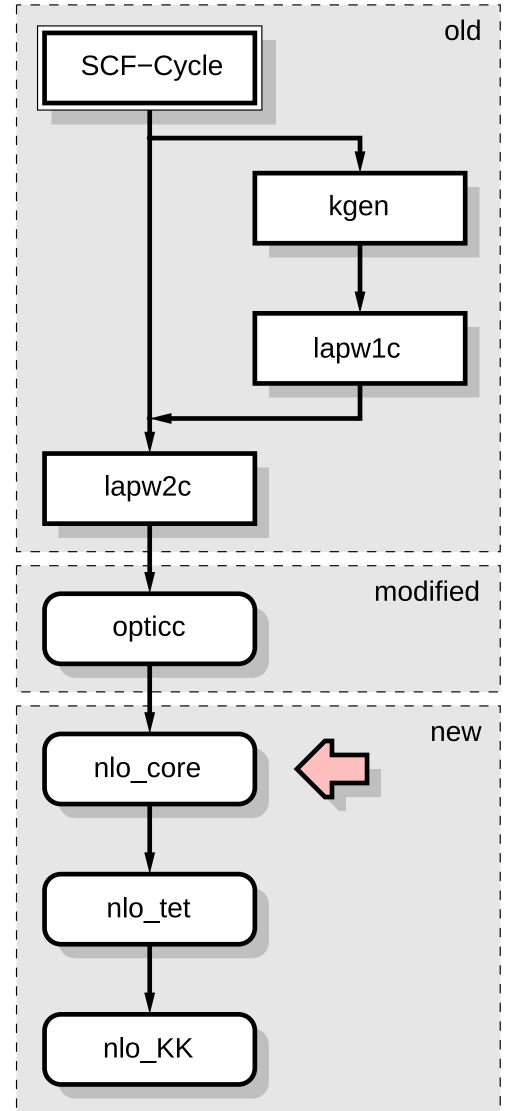


Program Modules

Module: nlo_core

- Evaluate the Integrands
- Object Oriented Structure

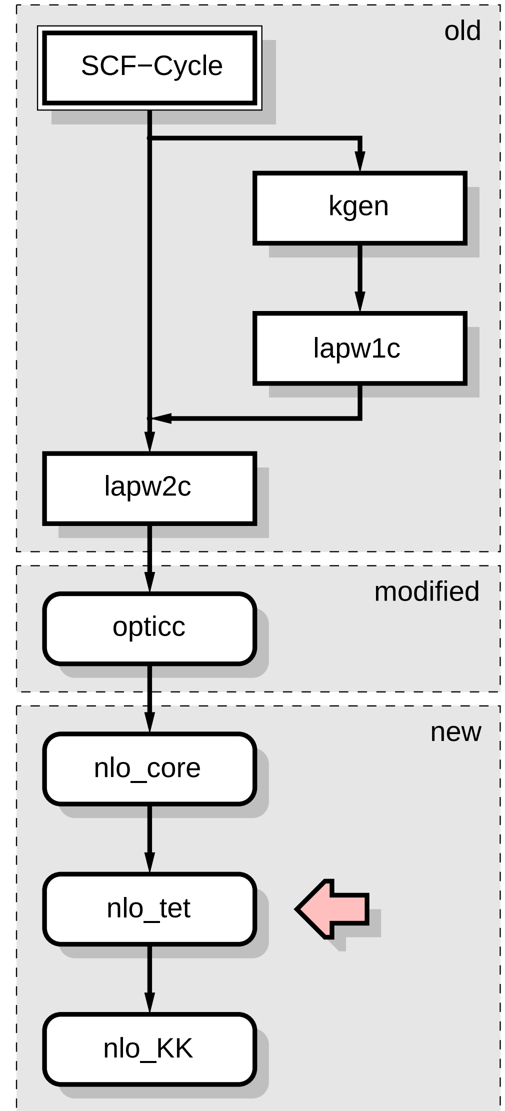
  $\Rightarrow$ easy add-on formulas
- Different Formulas Implemented:
  - SHG57 (PRB **57**, 3905 (1998))
  - SHG53 (PRB **53**, 10751 (1996))
  - OR61 (PRB **61**, 5337 (2000))
- Divergence Cutoff Parameter
- Scissor Correction



Program Modules

Module: nlo_tet

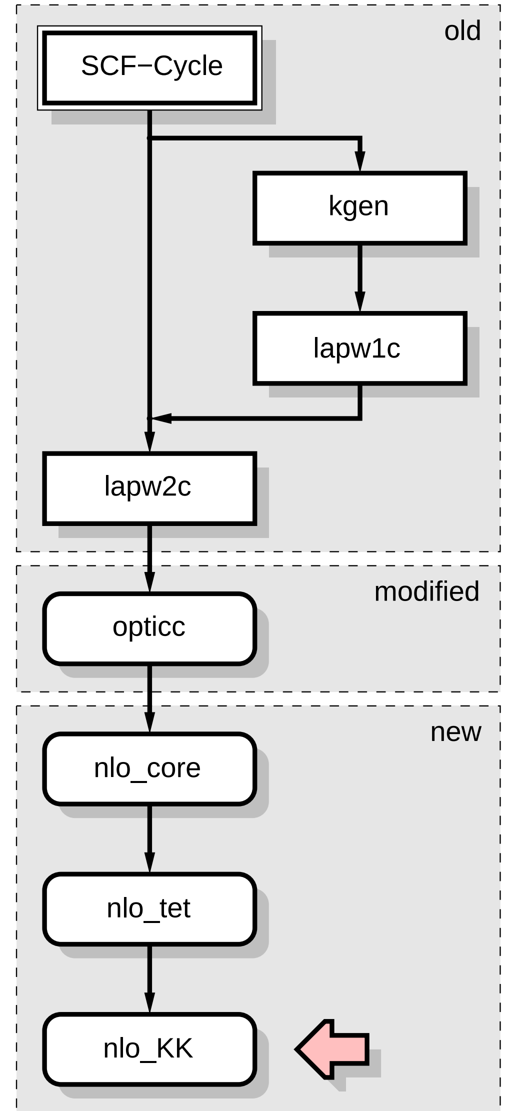
- Calculate the Imaginary Part of the Susceptibility by Integration
- Linearized Tetrahedron Method
- Independent from nlo_core
- Skip Tetrahedrons
- Interface to gnuplot



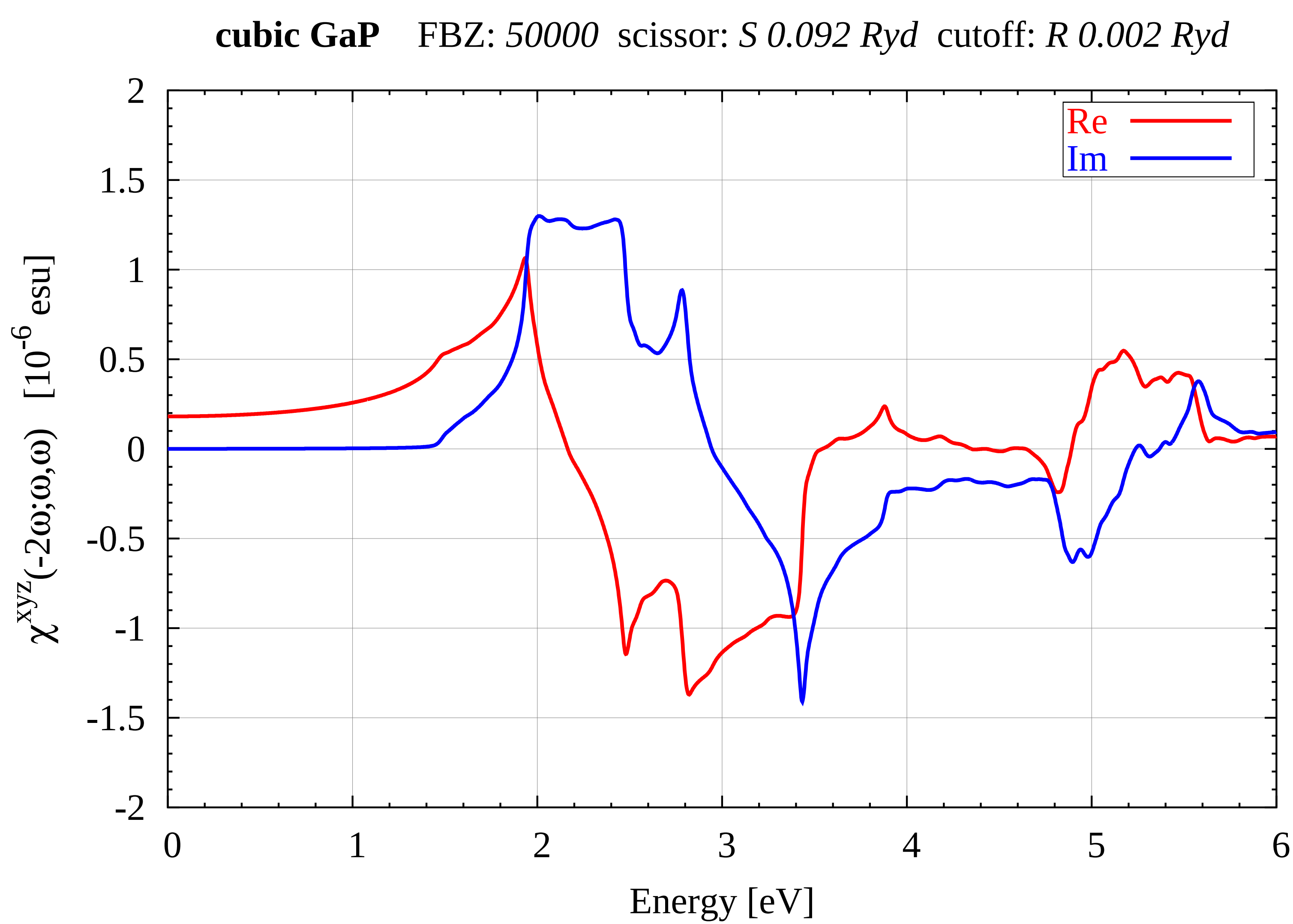
Program Modules

Module: nlo_KK

- Computes the Real Part of the Susceptibility with Kramers Kronig
- Integration by using Broadening
- Filter-Kernels
  - Lorentz
  - Gaussian
- Interface to gnuplot

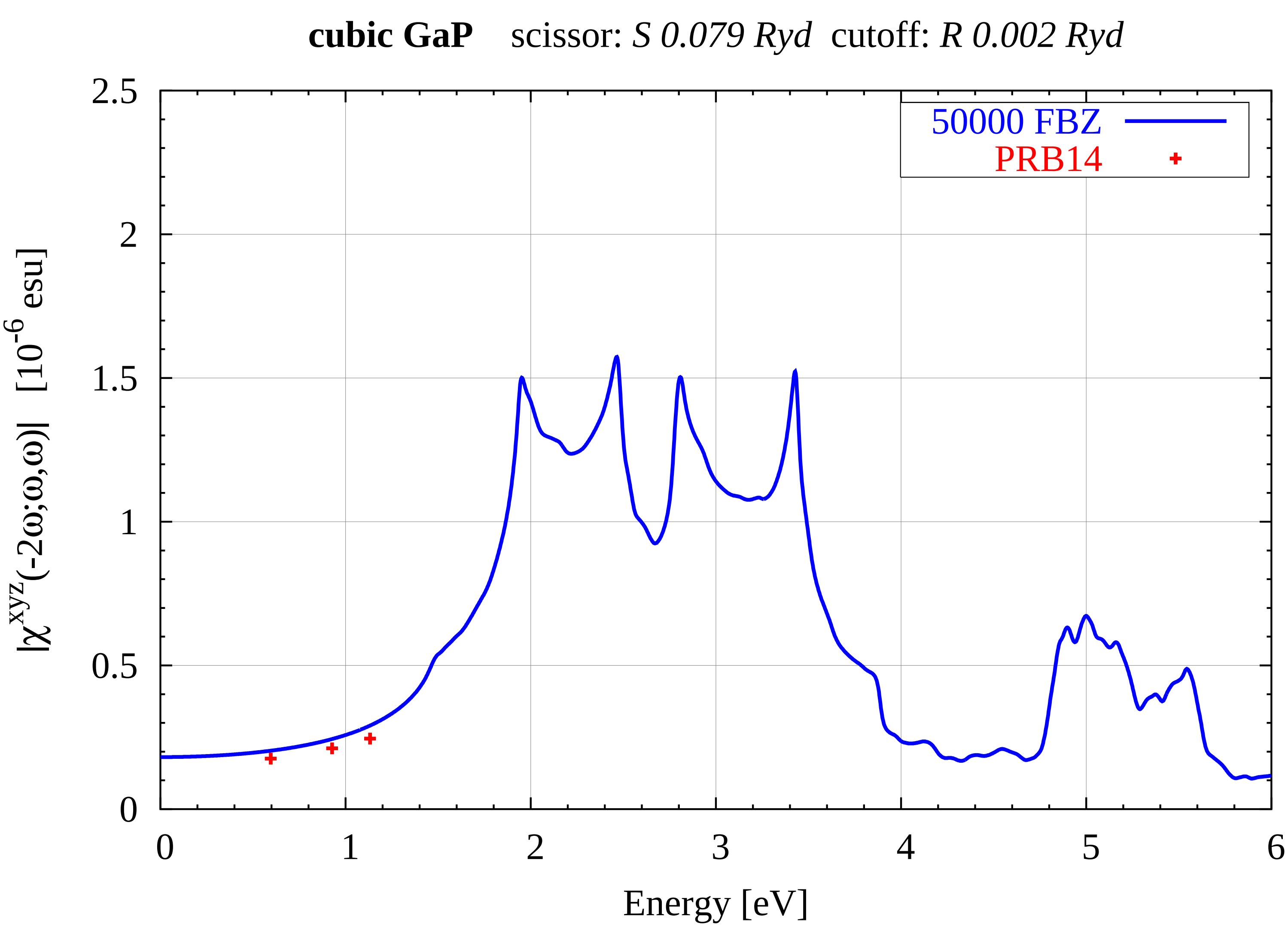


Examples



cubic GaP FBZ: *50000* scissor: *S 0.092 Ryd* cutoff: *R 0.002 Ryd*
Re
Im
$\chi^{xyz}(-2\omega;\omega,\omega)$ [10^{-6} esu]
2
1.5
1
0.5
0
-0.5
-1
-1.5
-2
0
1
2
3
4
5
6
Energy [eV]

Examples



Exp. Data (PRB **14**, 1693 (1976))

Examples

Static SHG

- Susceptibility at Zero Frequency $\chi_{II}(0)$
- Non Sensitive to the k-Point Density...
- Good Matches with Experiments (in $[pm/V]$)

Method	GaP	GaAs
LDA	165.4	857.3
LDA + scissors	78.7	176.9
GGA	142.6	521.5
GGA + scissors	75.6	167.4
Experiment*	74 ± 4	162 ± 10

* Exp. Data (PRL **20**, 272 (1972))

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Conclusion

Key Points in a NLO-Calculation

- Number of k-Points
- Upper Energy Limit in nlo_tet...
- Number of Conduction Bands
- Fine Tuning to Minimize the Effect of the DR
  - Density of the k-Mesh
  - Scissor Parameter
  - Divergence Cutoff Parameter
- Analysis of the remaining DR

Conclusion

Summery

- Bulk Materials
- Insulators and clean Semiconductors
- Separation into Inter- and Intraband Contribution
- Proper SHG in the Zero Frequency Limit
- Susceptibility shows correct Behavior at low Energies
- Results from the Literature reproducible

- No Metals
- Double Resonances

Conclusion

To Do

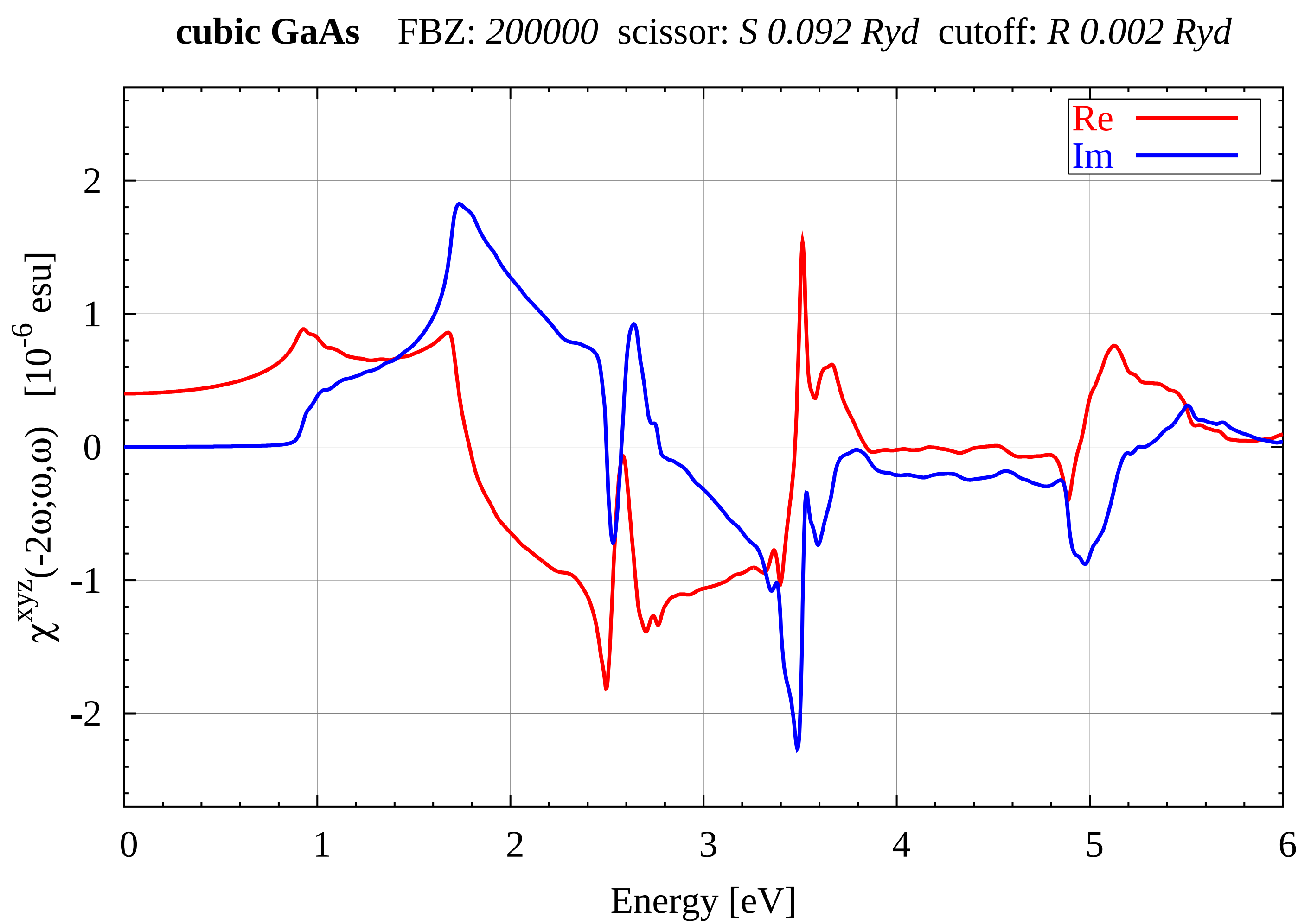
- Band Analysis Mode
- Sum Rule Checks (PRB **51**, 6925 (1995))
- Second Divergence Cutoff Parameter

  $\Rightarrow$ Controlling DR
- Additional Formulas in nlo_core
  - Static Limit $\chi_{II}(0)$
  - Third Harmonic Generation (THG), ...

Thank You !

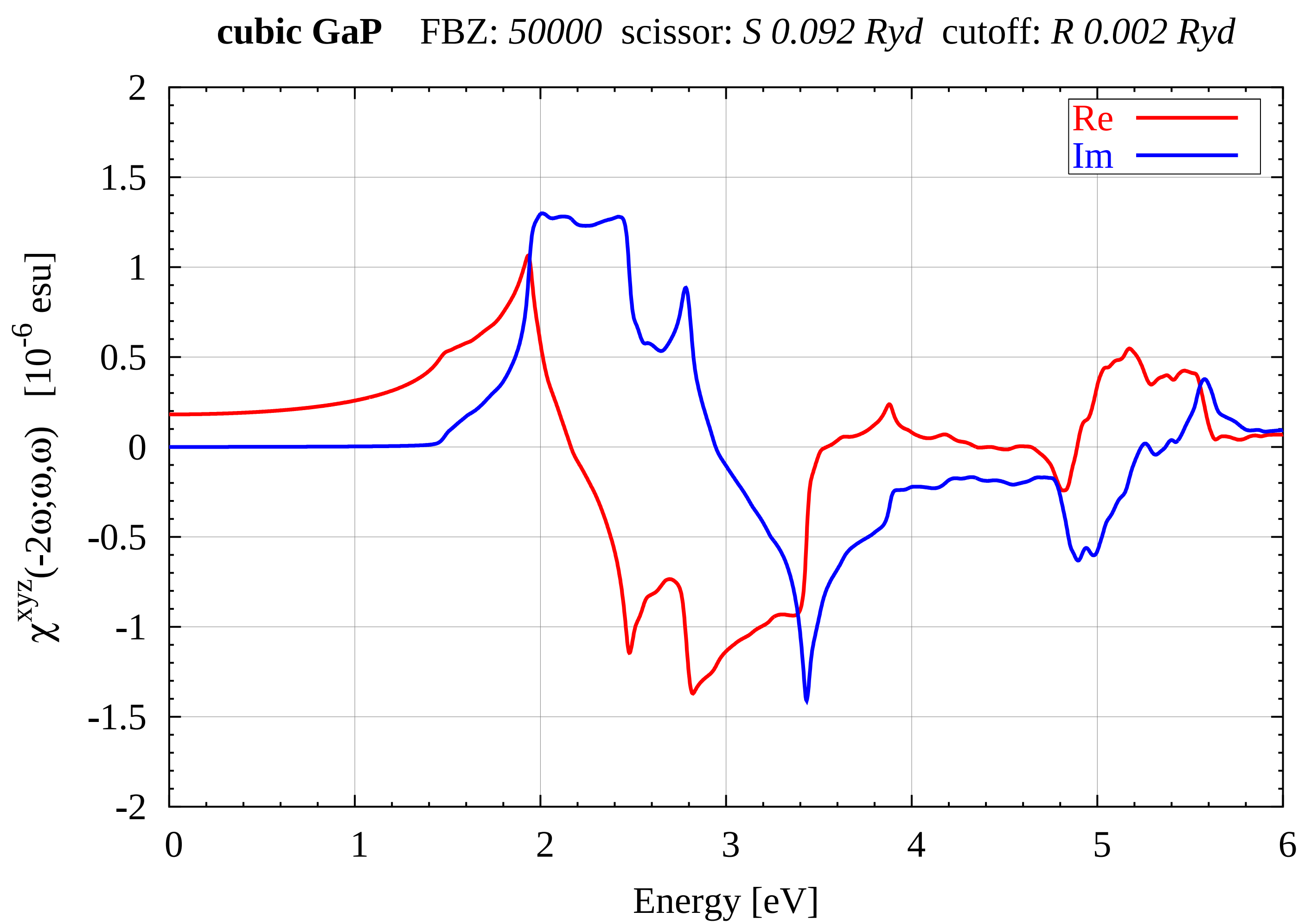
Examples

GaAs

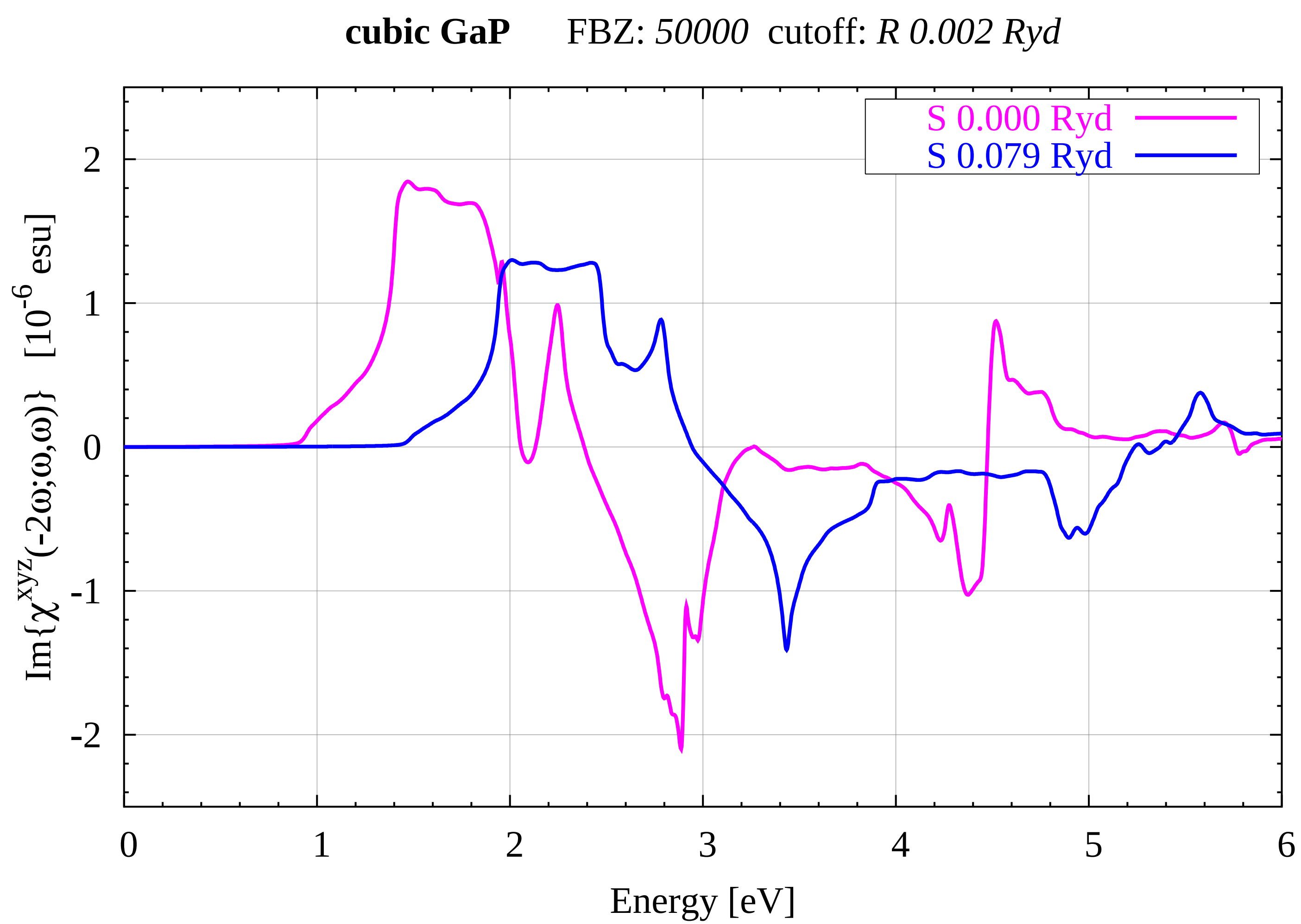


Examples

GaP

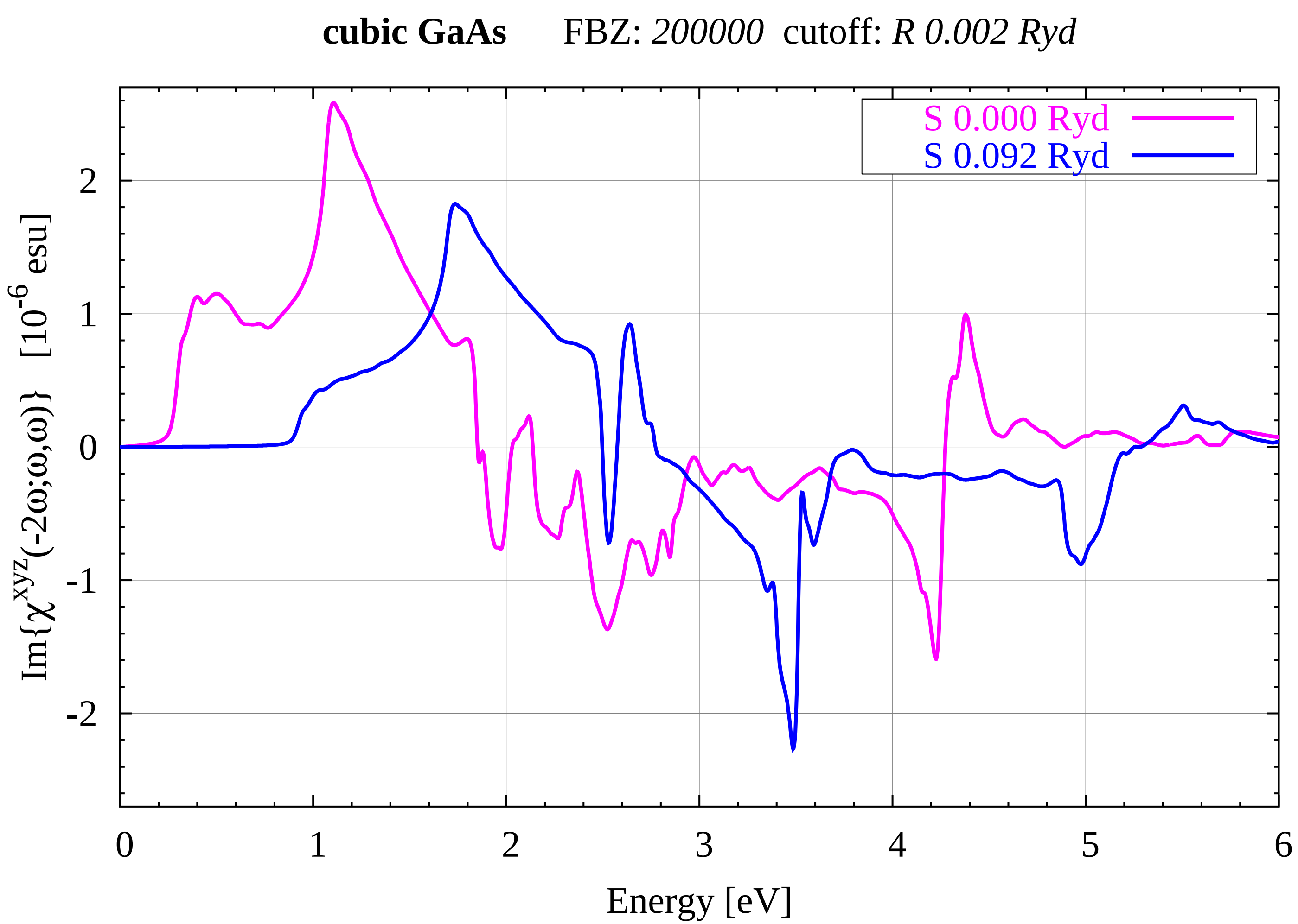


Scissor Correction



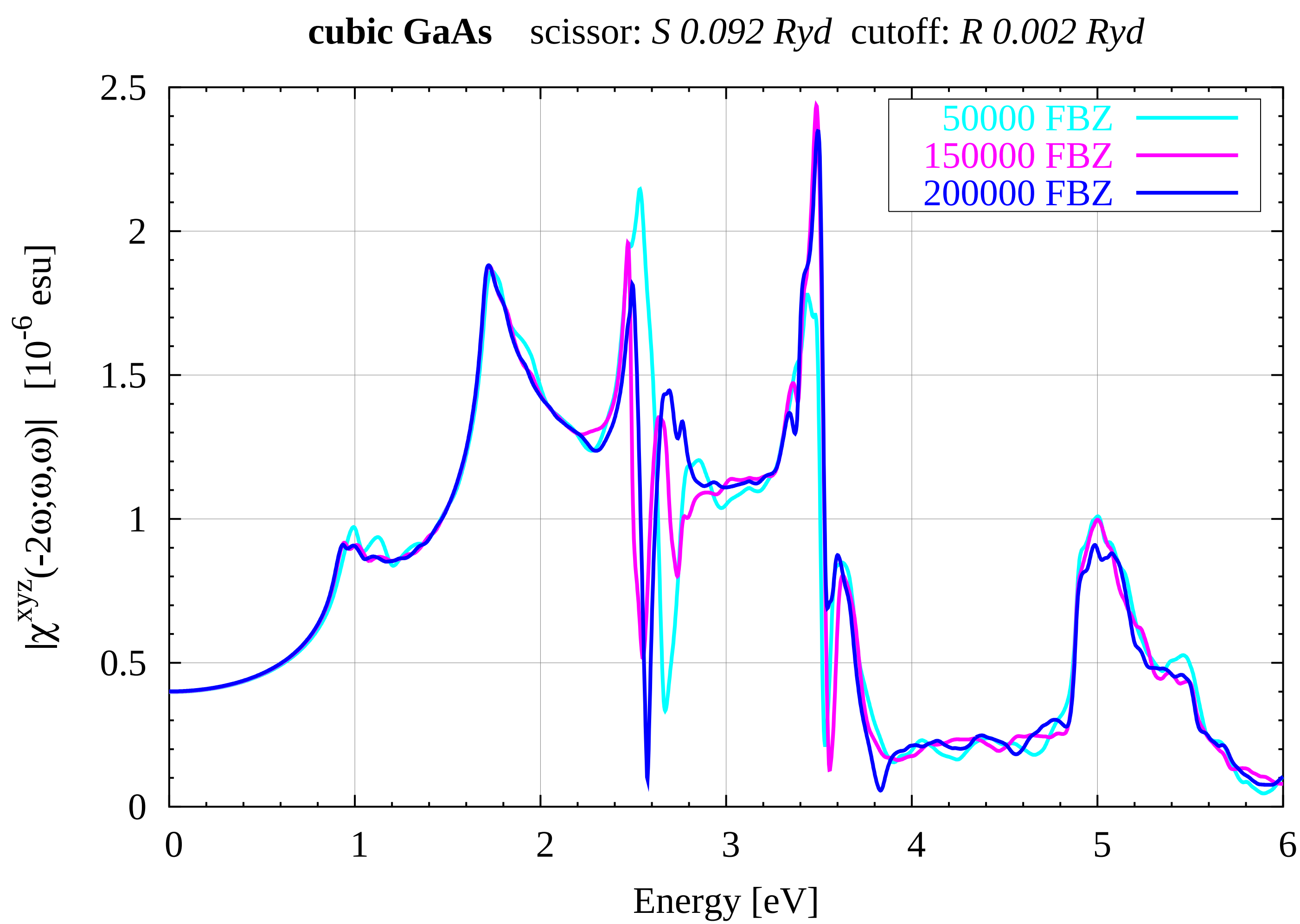
Examples

Scissor Correction



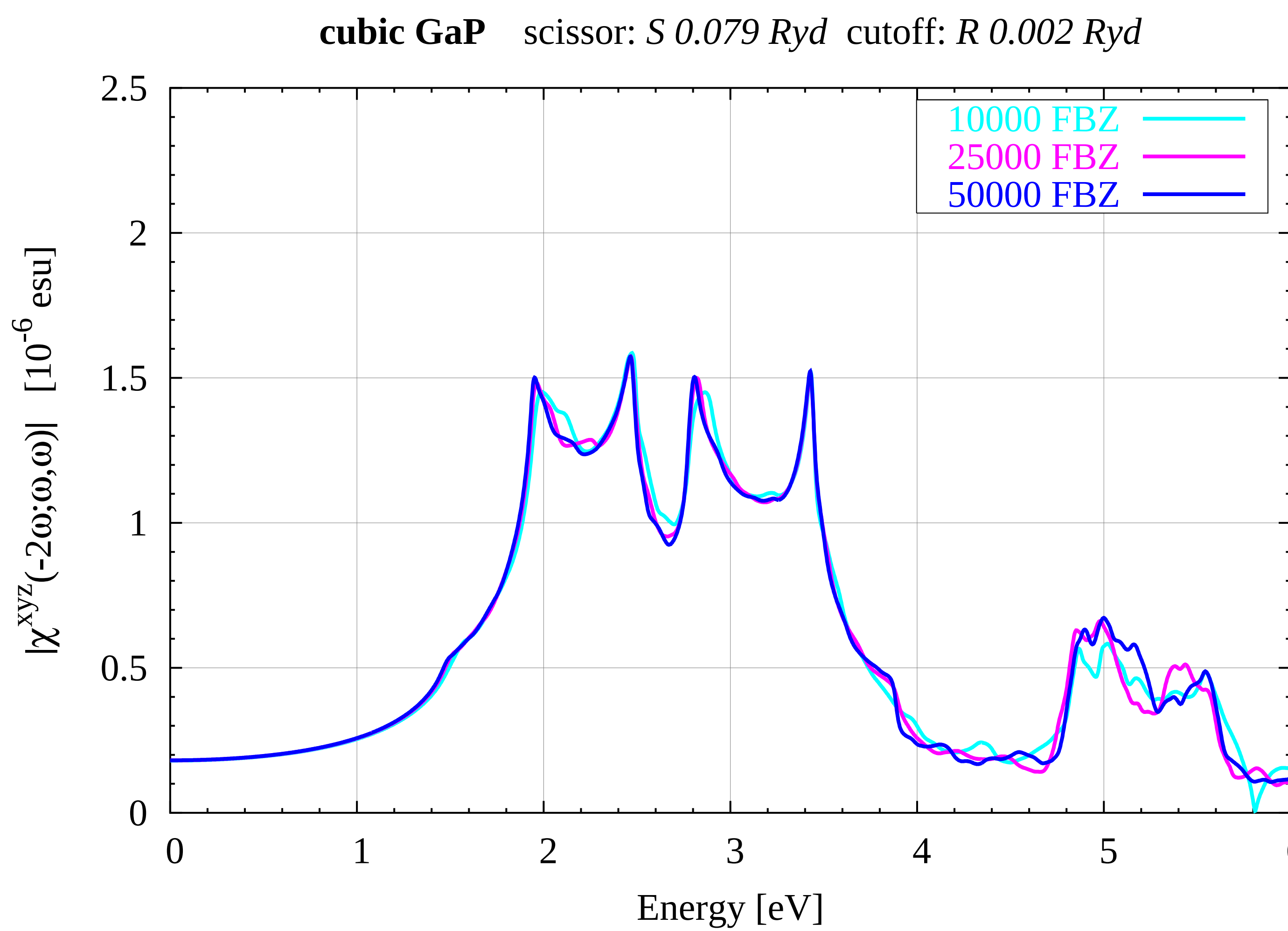
Examples

k-Point Dependency...



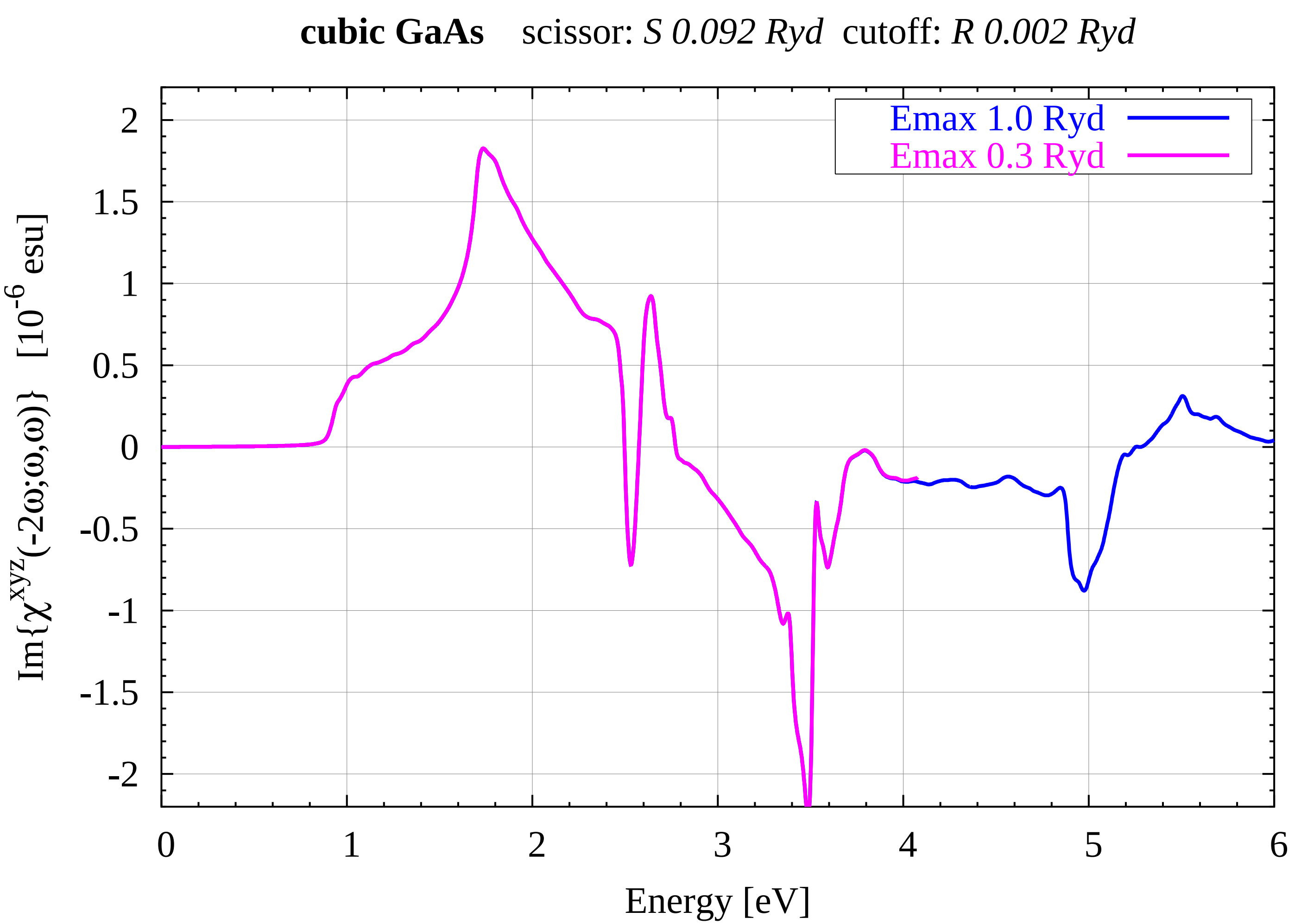
Examples

k-Point Dependency...



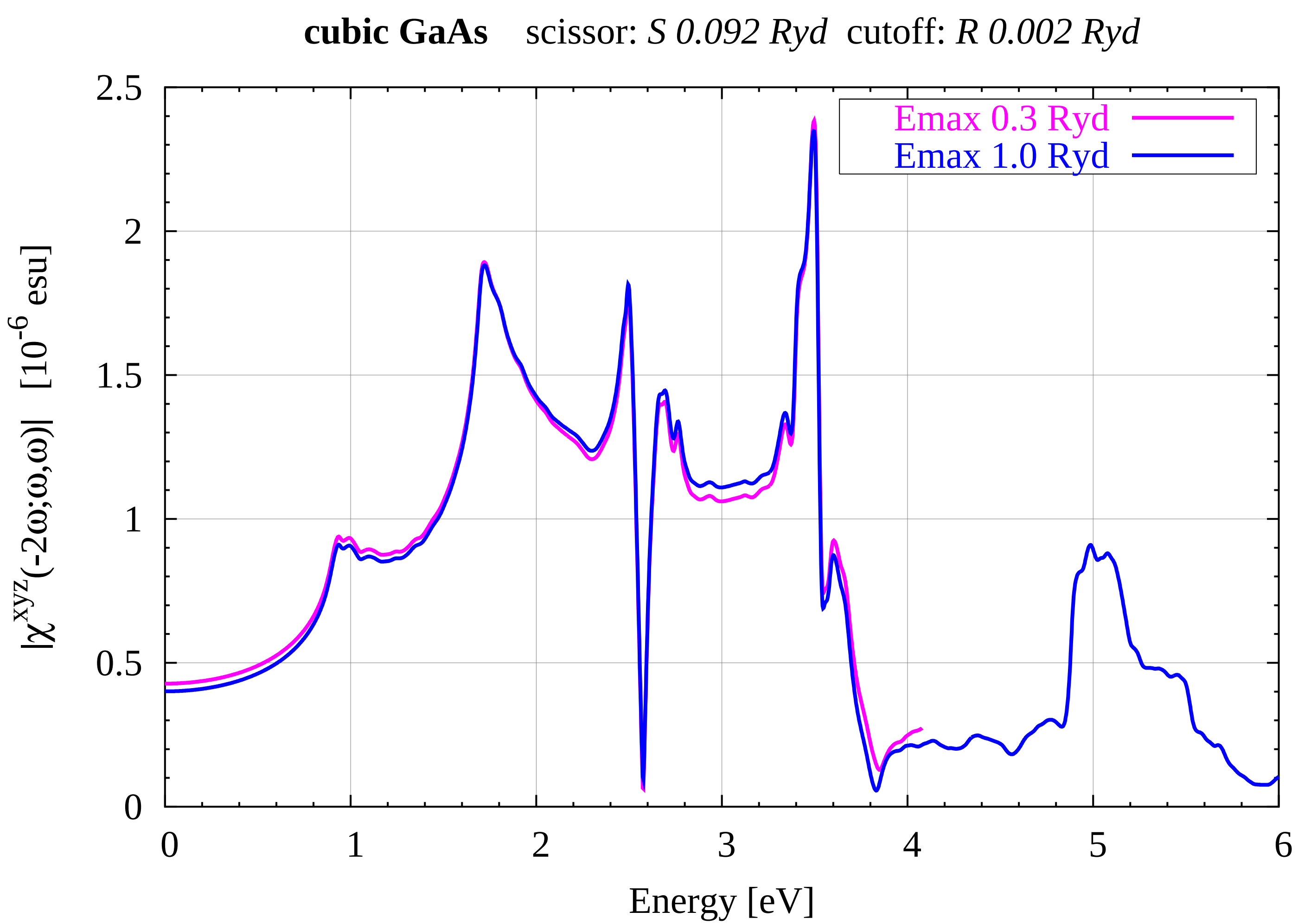
Examples

Energy Range and Kramers Kronig I...



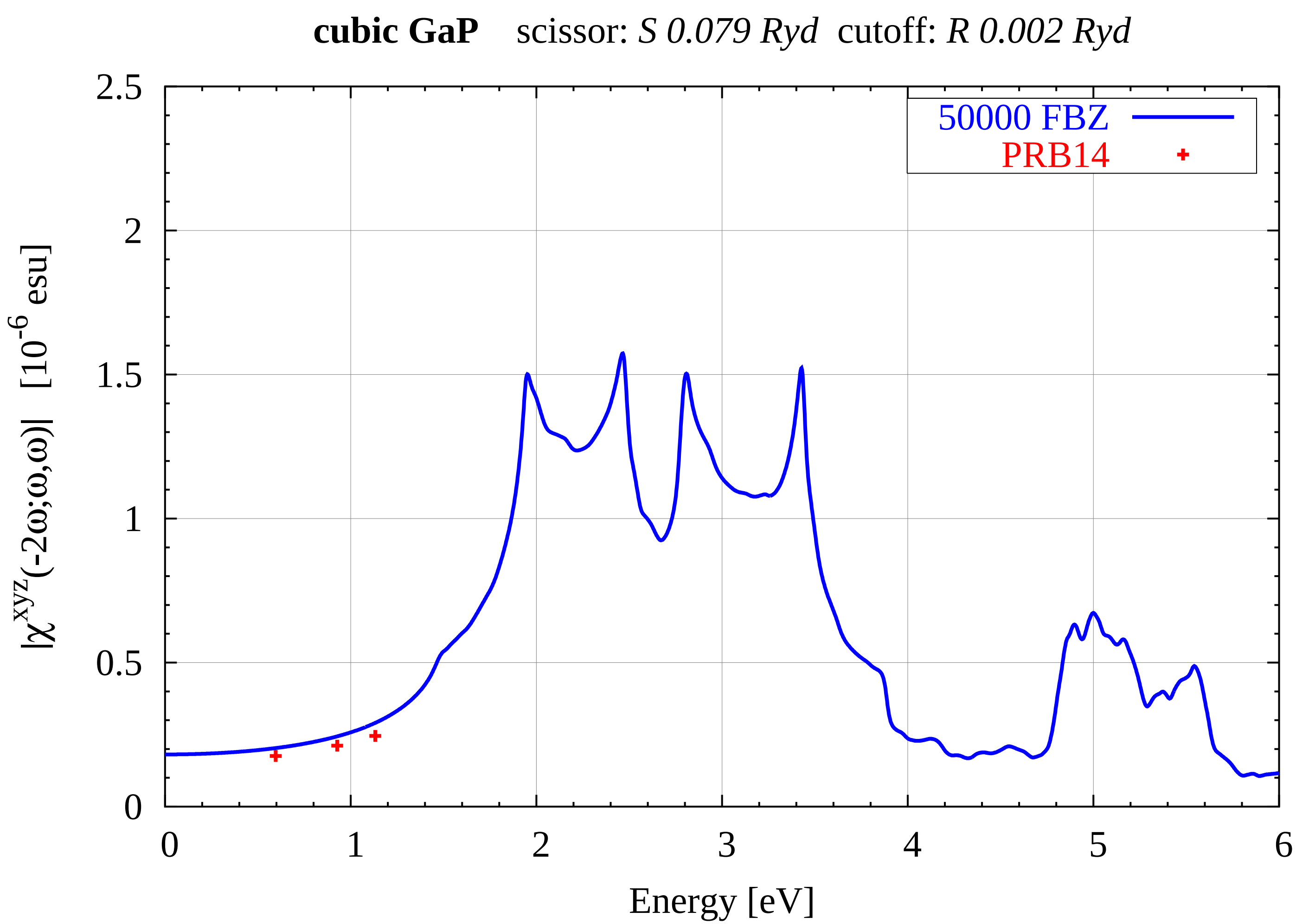
Examples

Energy Range and Kramers Kronig II...



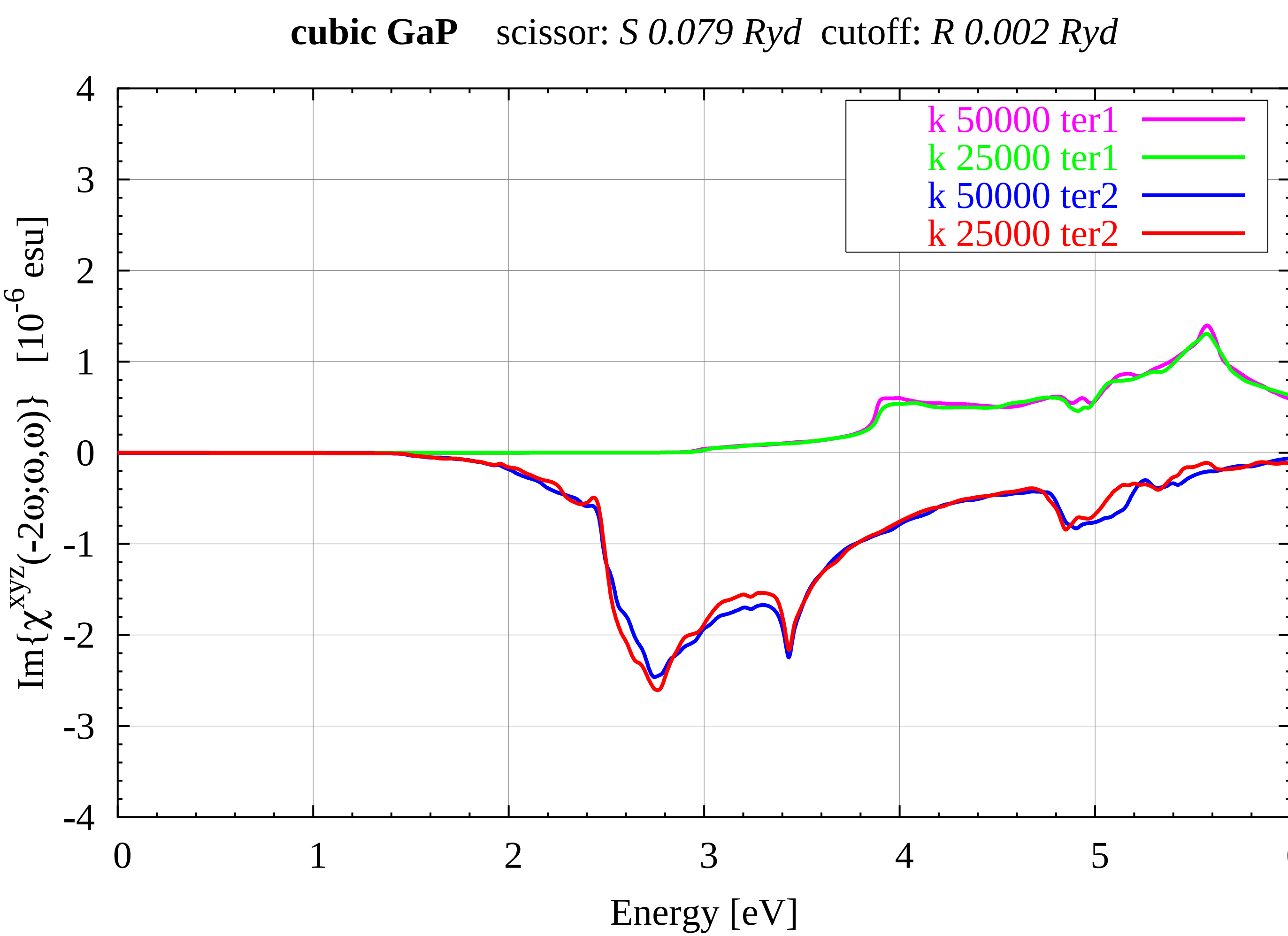
Examples

GaP



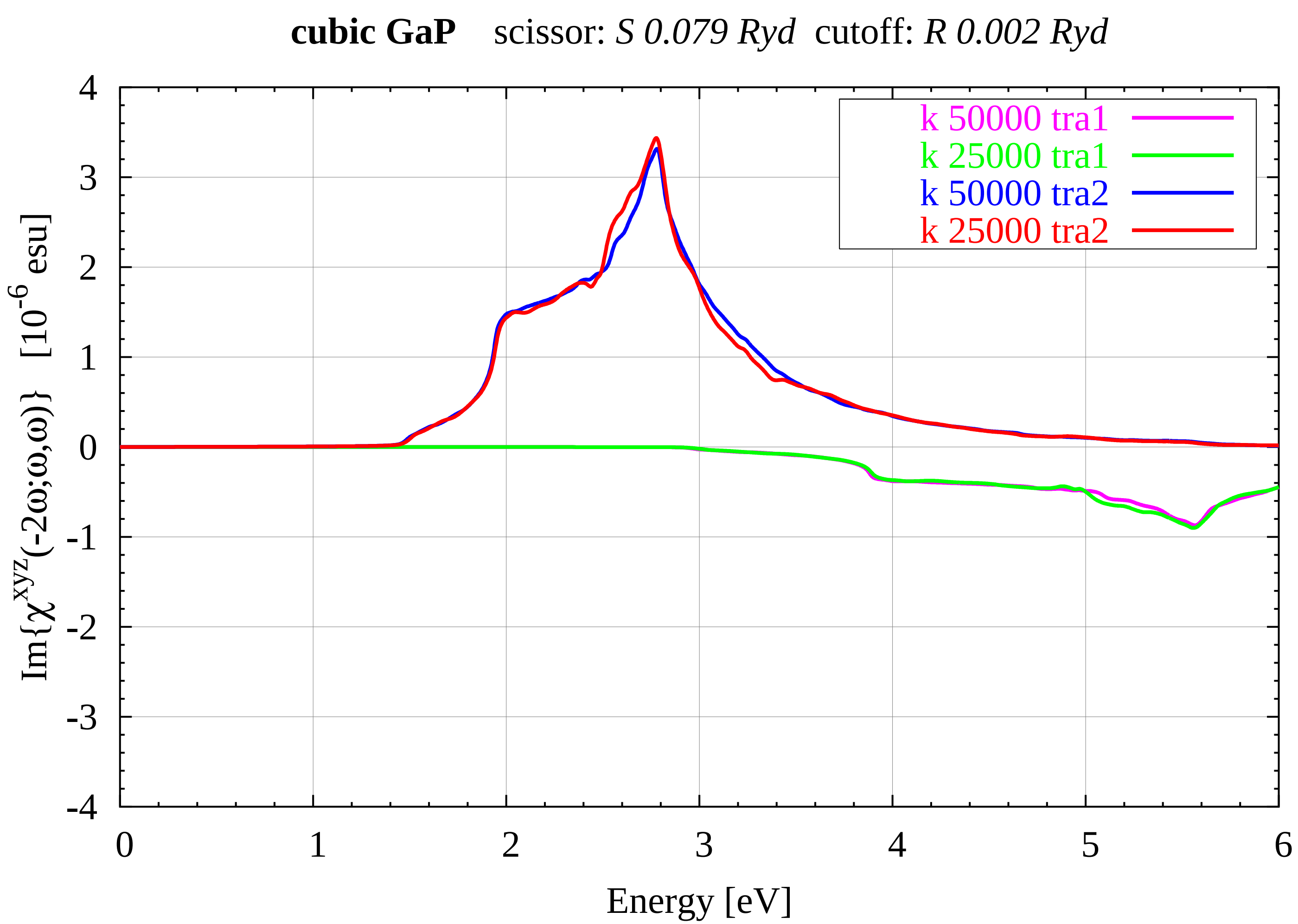
Exp. Data (PRB **14**, 1693 (1976))

Double Resonances



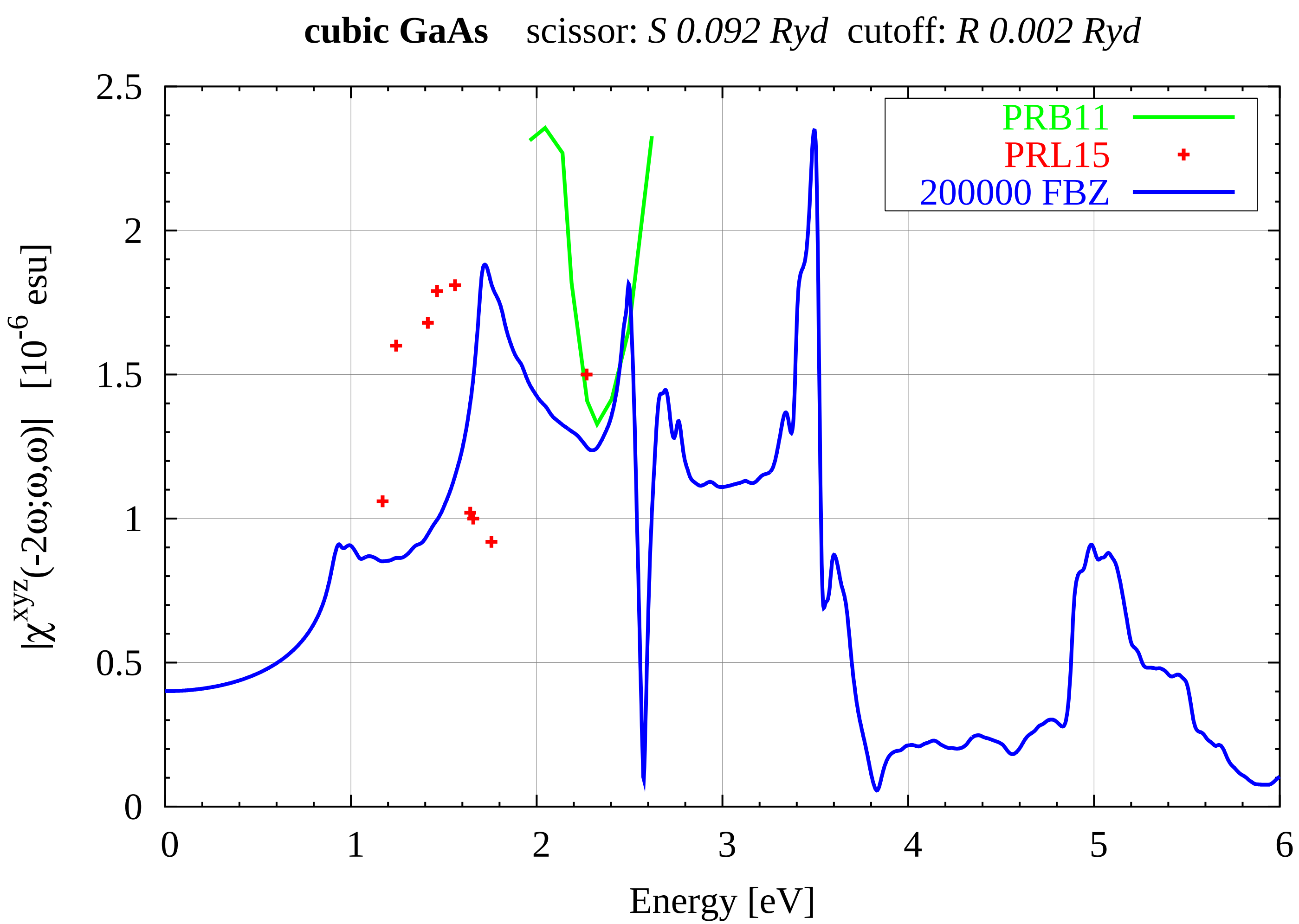
Examples

Double Resonances



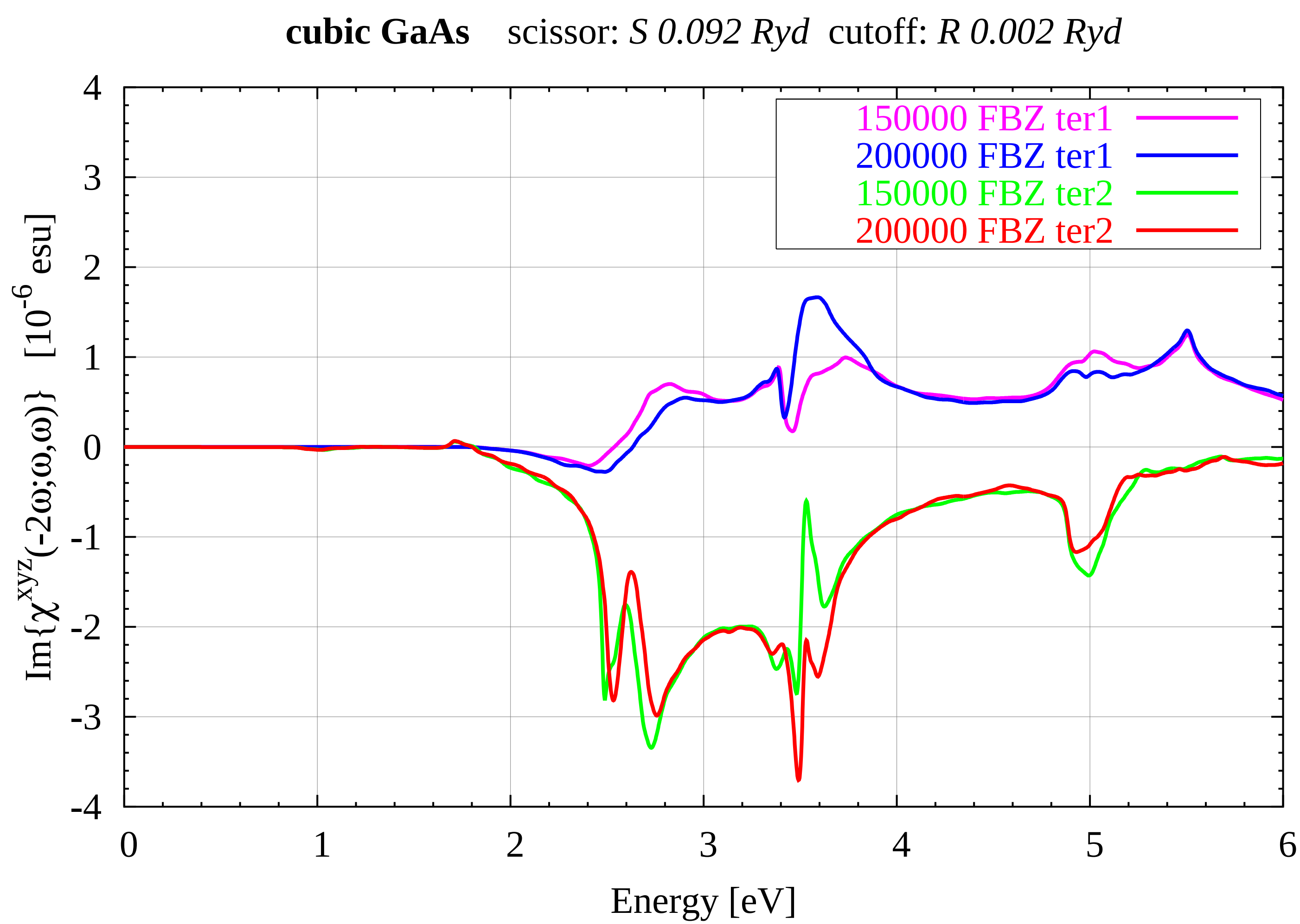
Examples

GaAs



Exp. Data (PRL **15**, 415 (1965)) and (PRB **11**, 3867 (1975))

Double Resonances



Examples

Double Resonances

cubic GaAs scissor: *S 0.092 Ryd* cutoff: *R 0.002 Ryd*