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# Optical Properties of Be at High Temperatures from First Principles

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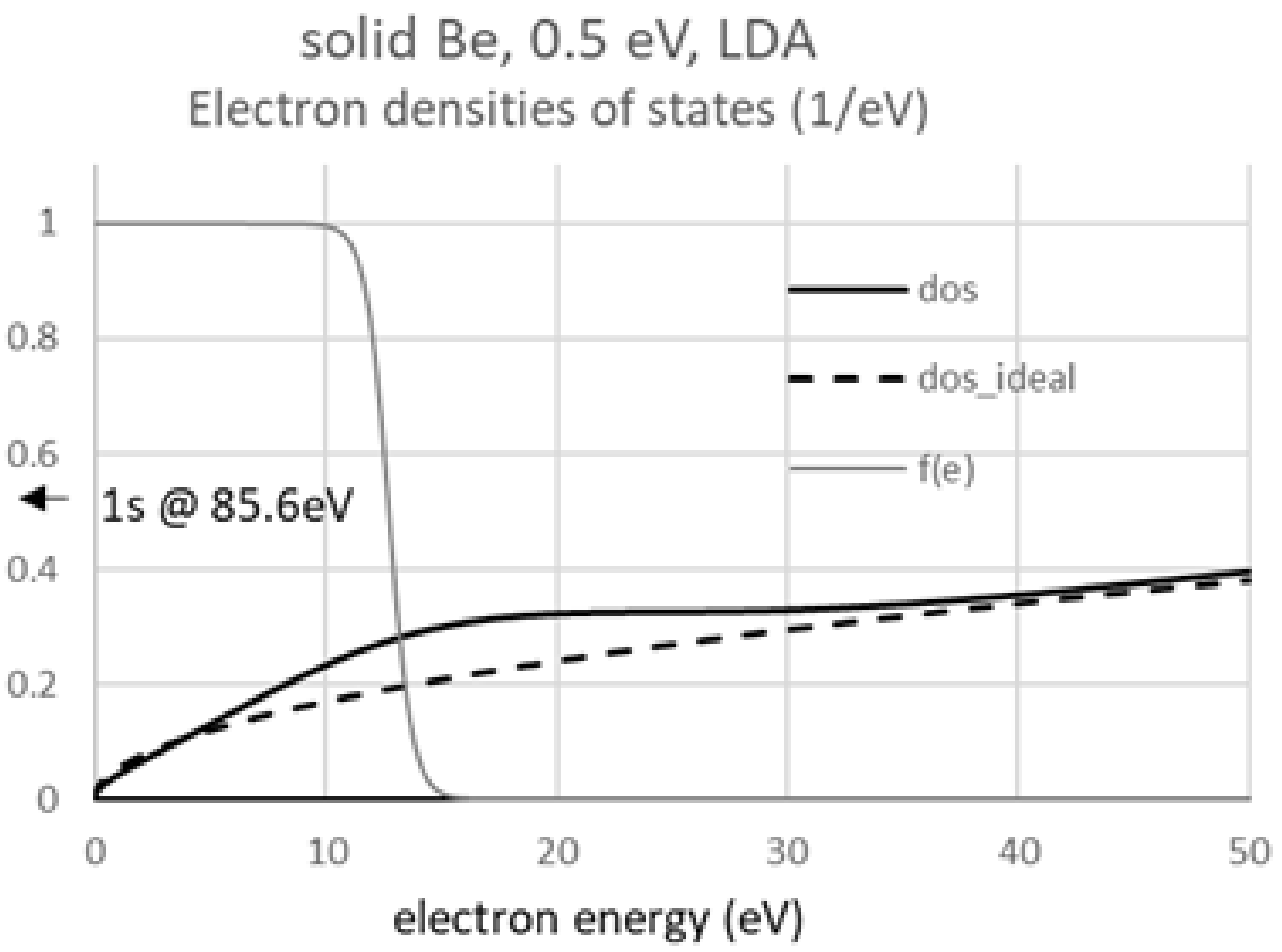
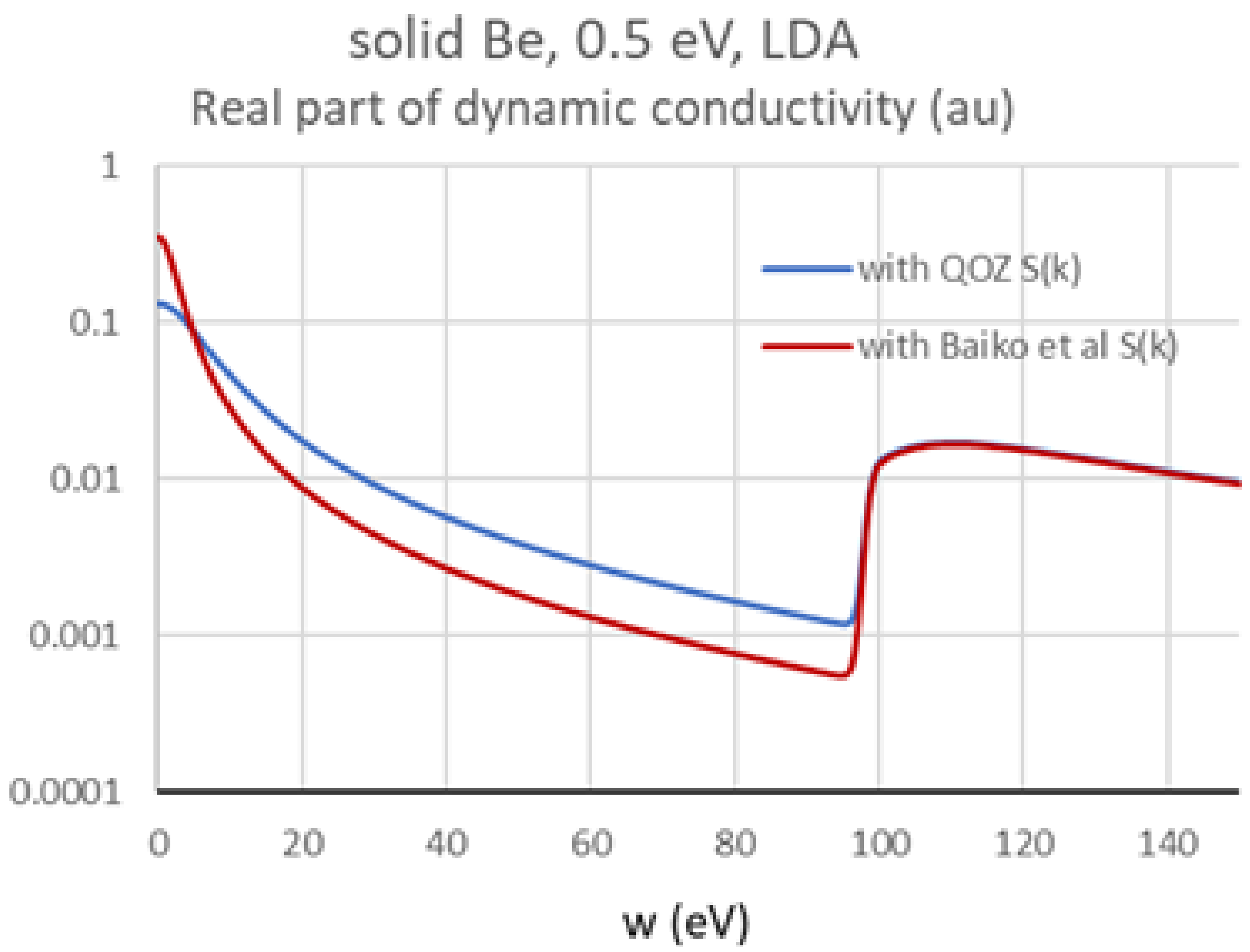
# Goals

- Obtain an accurate and efficient method of calculating electrical conductivities of warm dense matter
- Benchmark and constrain average atom (AA) methods against density functional theory (DFT)
- Use results to design laboratory warm dense matter experiments
- Allow for the better understanding and interpretation of experimental results




- AA calculation of the Be density of states at 0.5 eV

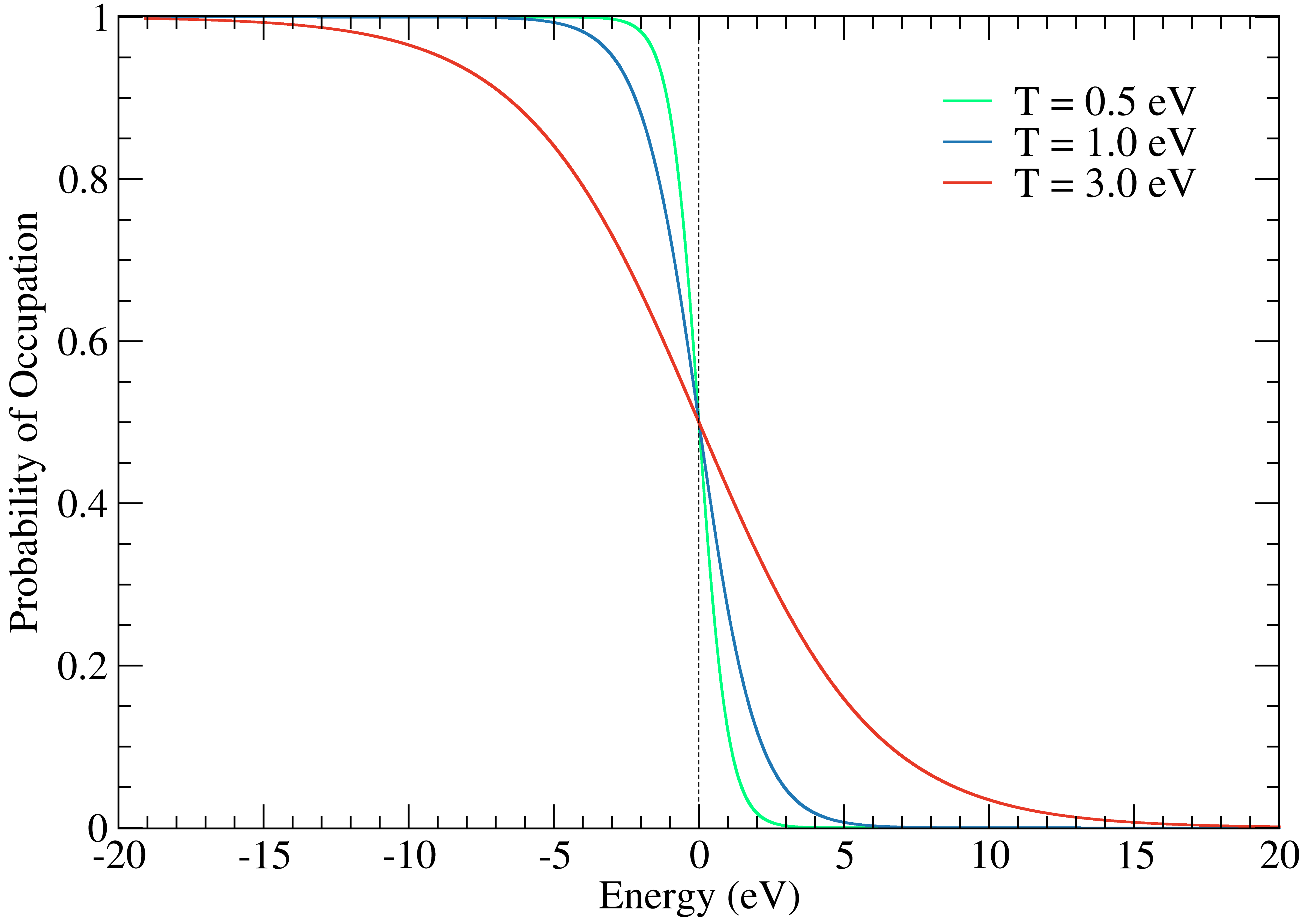



- AA calculation for the real part of the dynamic conductivity for Be

# Computational Methods

## Density Functional Theory

- Vienna Ab initio Simulation Package
- PBE and LDA functional
- PAW pseudopotential
- Computed electronic density of states, electronic band structure, and the macroscopic dielectric function
- Fermi smearing to determine occupancy
  - Set electron temperature




## $G_0W_0$

- Computed electronic band structure and the frequency dependent self-energy

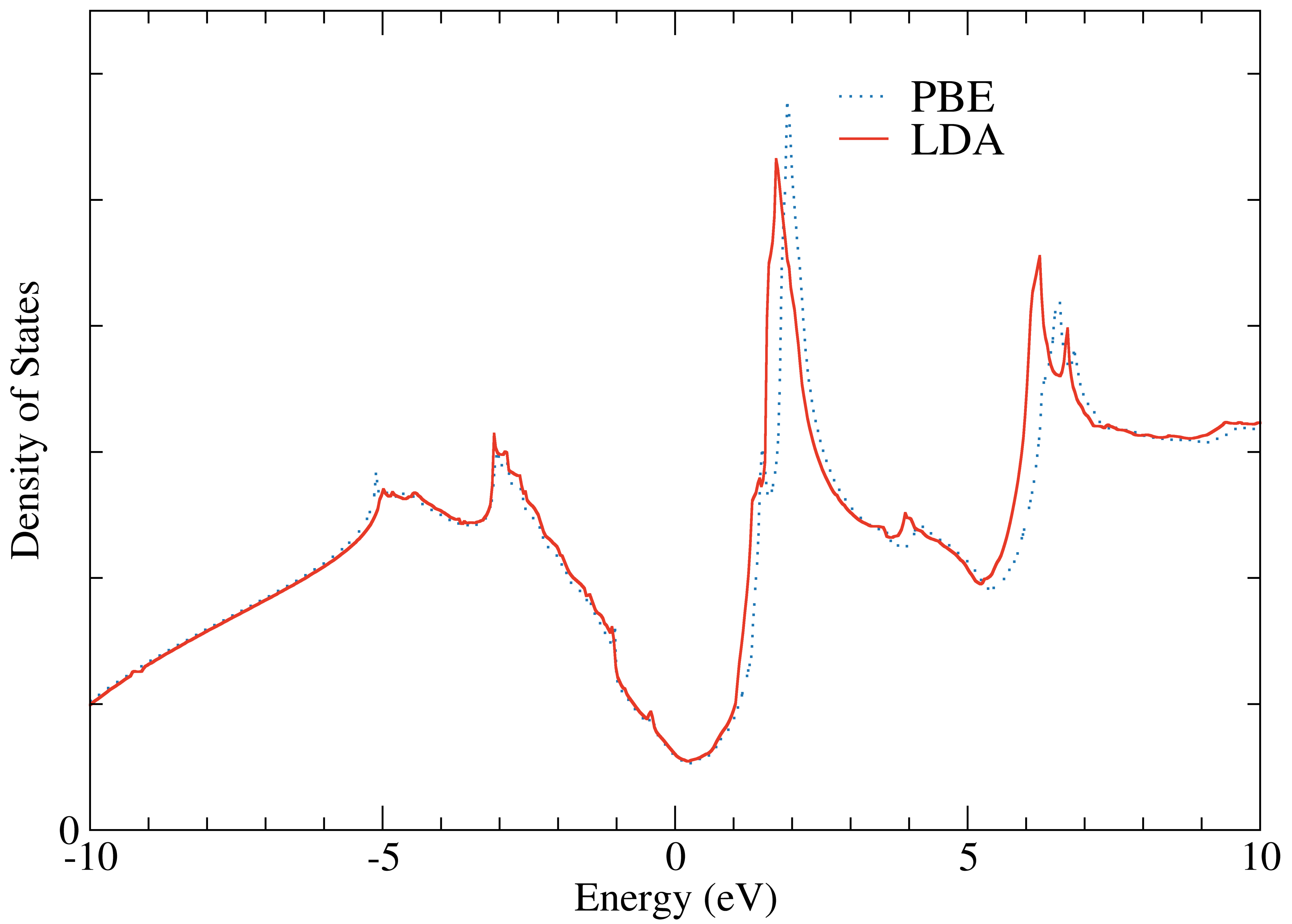
# DFT Results

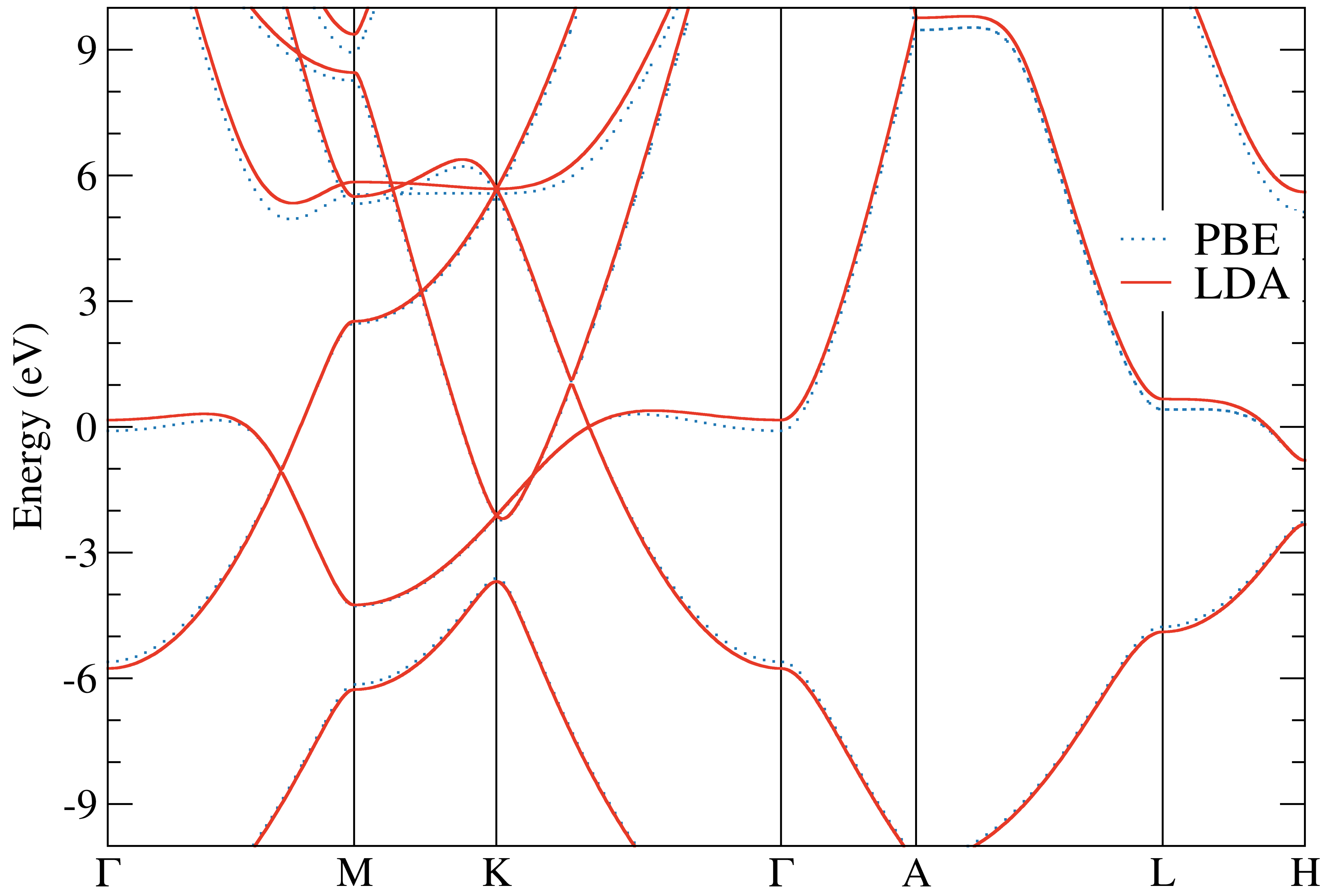
| | LDA | PBE | Experiment [1] |
|---|---|---|---|
| $a_0$ lattice constant (Å) | 2.231 | 2.264 | 2.287 |
| $c_0$ lattice constant (Å) | 3.525 | 3.574 | 3.583 |
| Cell Volume (Å$^3$) | 15.190 | 15.876 | 16.230 |

- LDA and PBE reproduce experimental results well
- PBE has better agreement overall
  - PBE volume has a 2% difference compared to experiment
  - LDA volume has a 6% difference compared to experiment

## Ground State Electronic Structure

- PBE and LDA: good agreement in both electronic density of states and electronic band structure
- LDA functional is used for the rest of the calculation because it allows for a better comparison to Average Atom



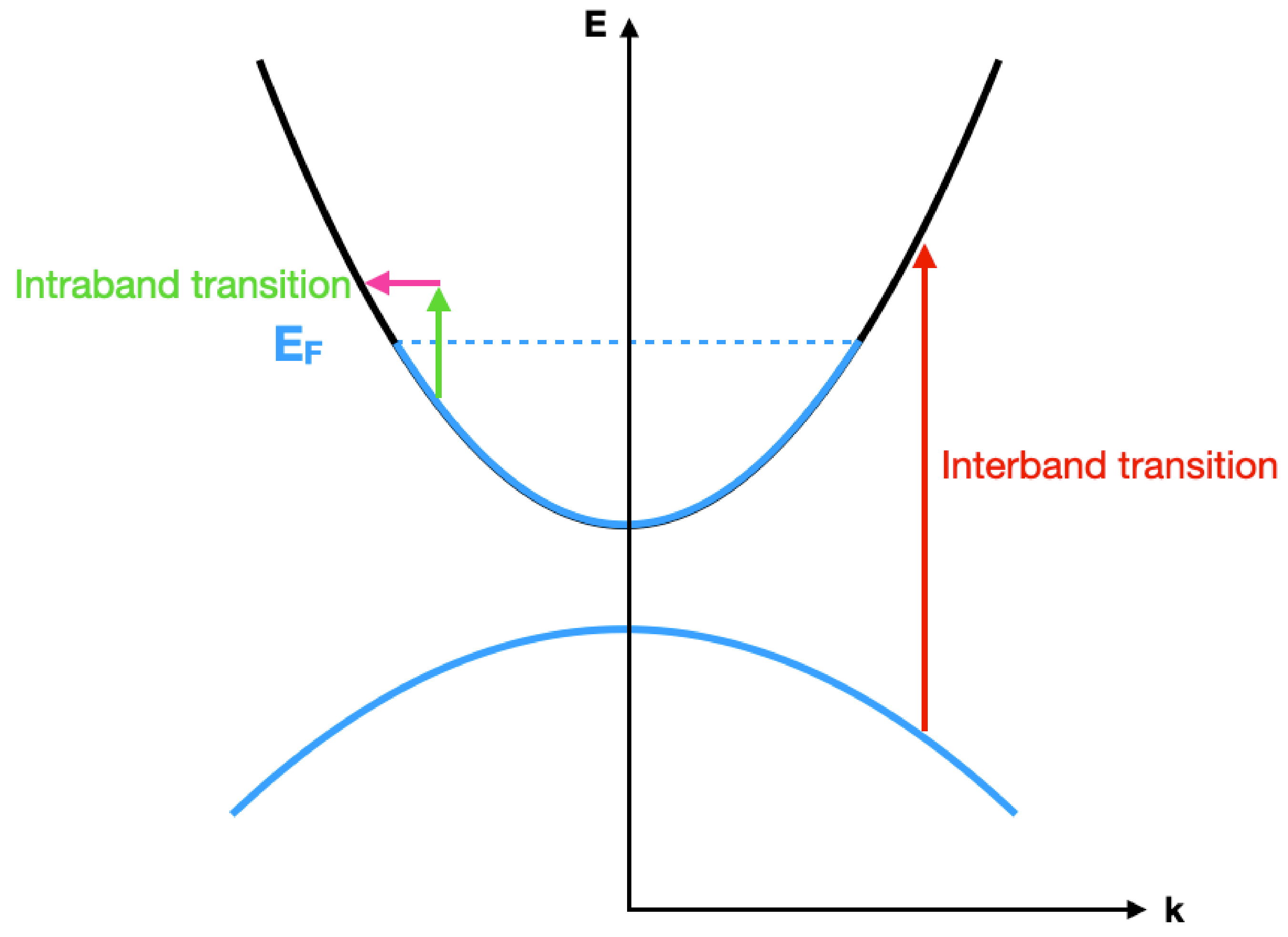
# DFT Optics

## Absorption in Beryllium

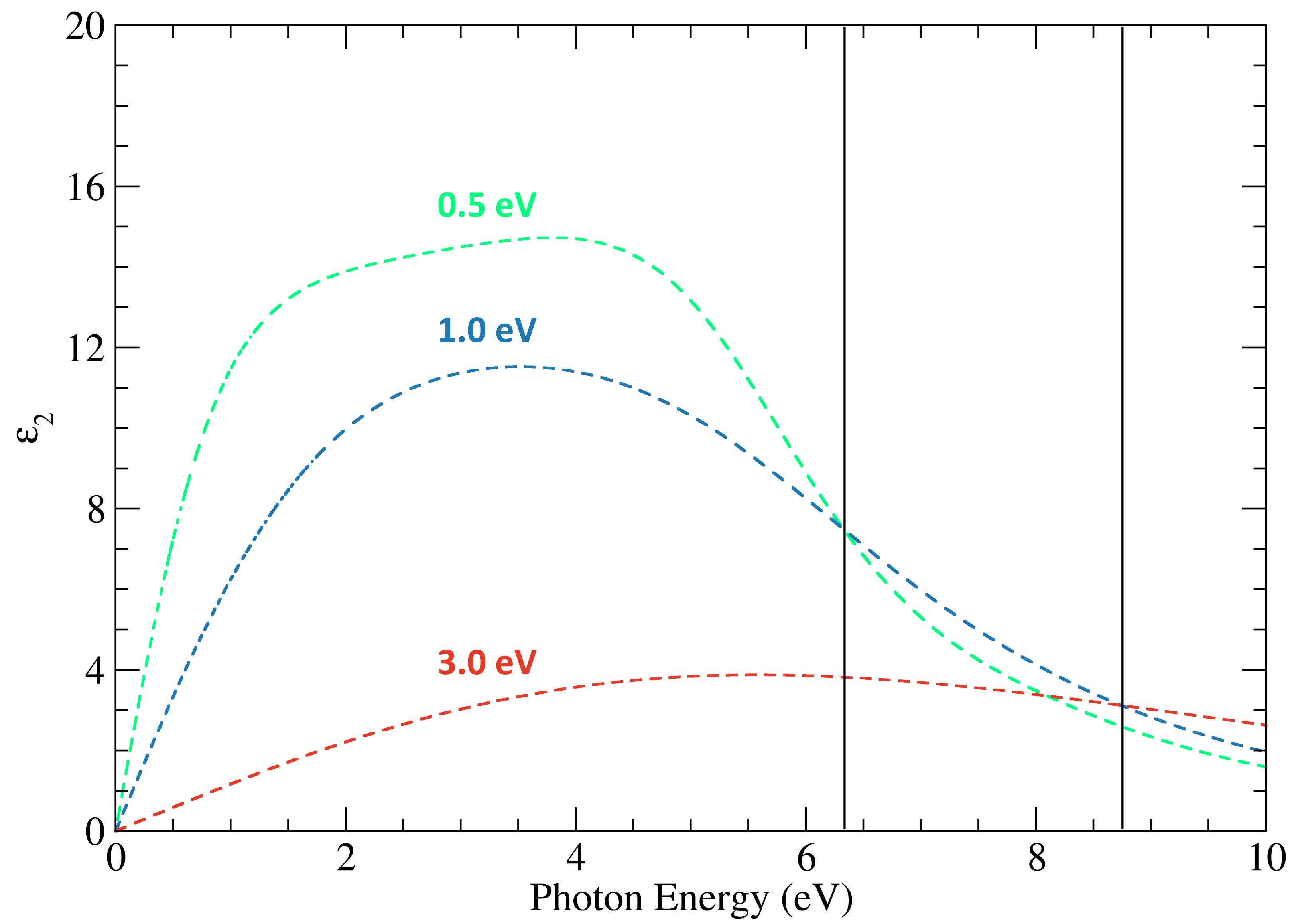
- Low T: stronger absorption at low energy and weaker absorption at high energy
- High T: weaker absorption at low energy and stronger absorption at high energy

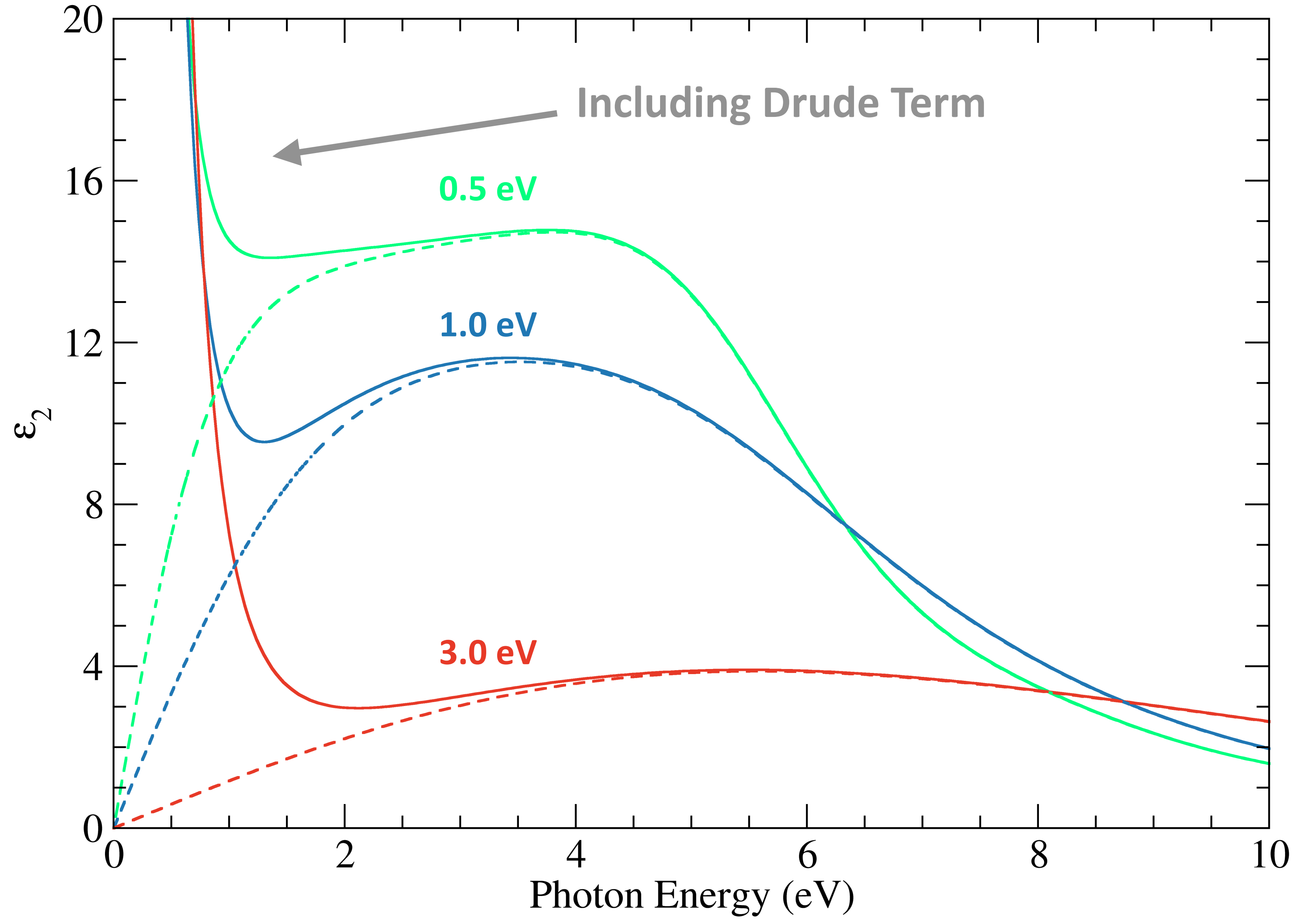
## Intraband Transitions

- Because beryllium is a metal, intraband transitions occur




- Use a Drude term in the density functional theory calculation to account for the intraband transitions
- Absorption at low energy very strong at all temperatures
- At 2.5 eV intraband transitions stop having an influence on the absorption spectrum



# Electron-Electron Relaxation Time

## *GW* Approach [2]

- Perform a *GW* calculation
  - *GW* takes the DFT electronic structure and calculates the corrections to the Kohn-Sham energies [3]
  - *G* is the electronic Green's function
  - *W* is the screened Coulomb interaction

1. Calculate the imaginary part of the self-energy to find the electron-electron term for scattering out of the state n**k**

$$\Gamma_{n\mathbf{k}}^{\text{out, (ee)}} = -2\text{Im}\Sigma(\varepsilon_{n\mathbf{k}})/\hbar$$

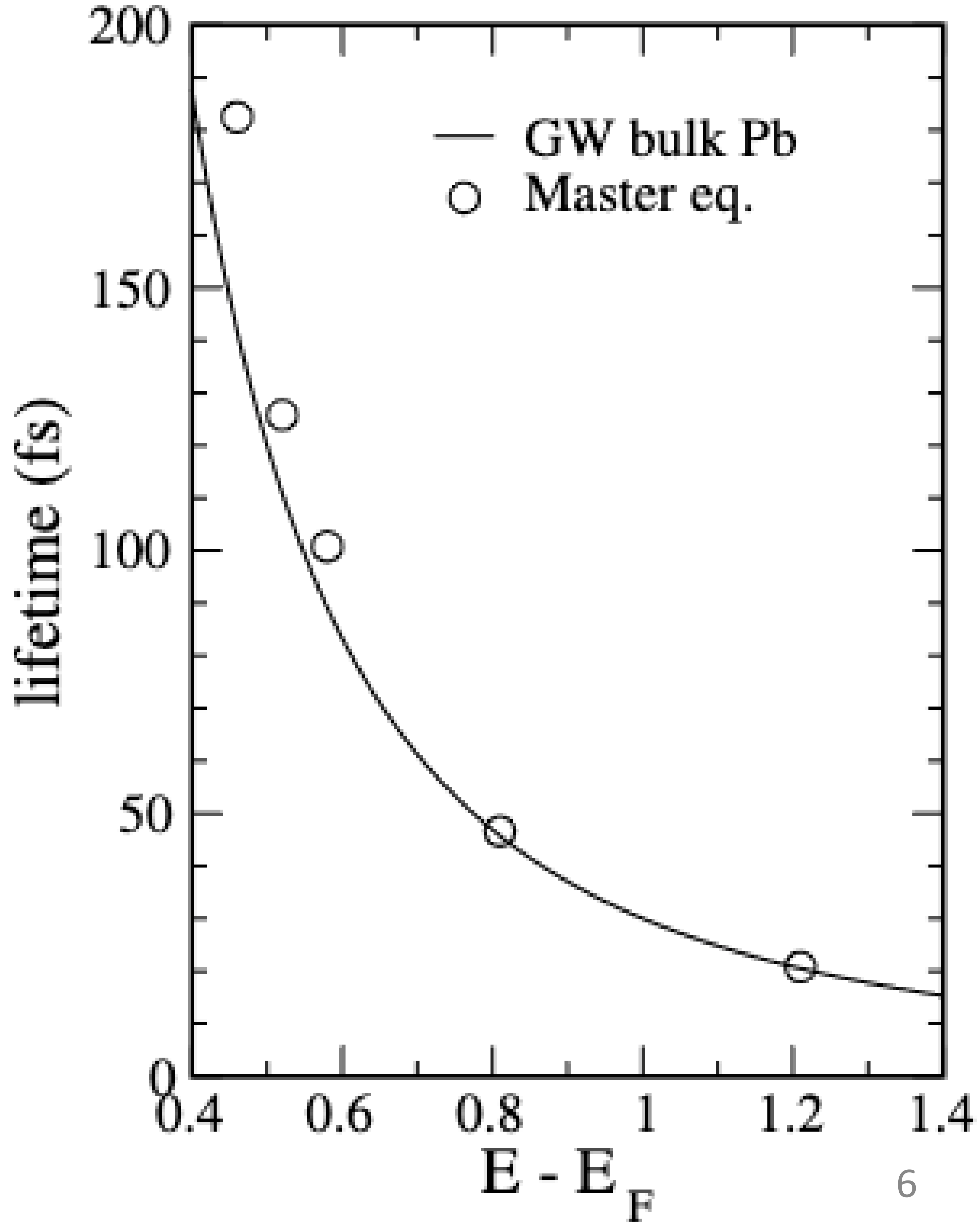
2. The result for -2ImΣ in the conduction band can be fitted to the Landau theory of the Fermi liquid

$$-2\text{Im}\Sigma(\epsilon_{n\boldsymbol{k}}) = \alpha(\epsilon_{n\boldsymbol{k}} - E_F)^2$$

3. Through the fit, α can be determined: in this example it was done for Pb

$$\Gamma_{n\mathbf{k}}^{\text{out, (ee)}} = \frac{(\varepsilon_{n\mathbf{k}} - E_F)^2}{30\text{fs}(\text{eV})^2}$$

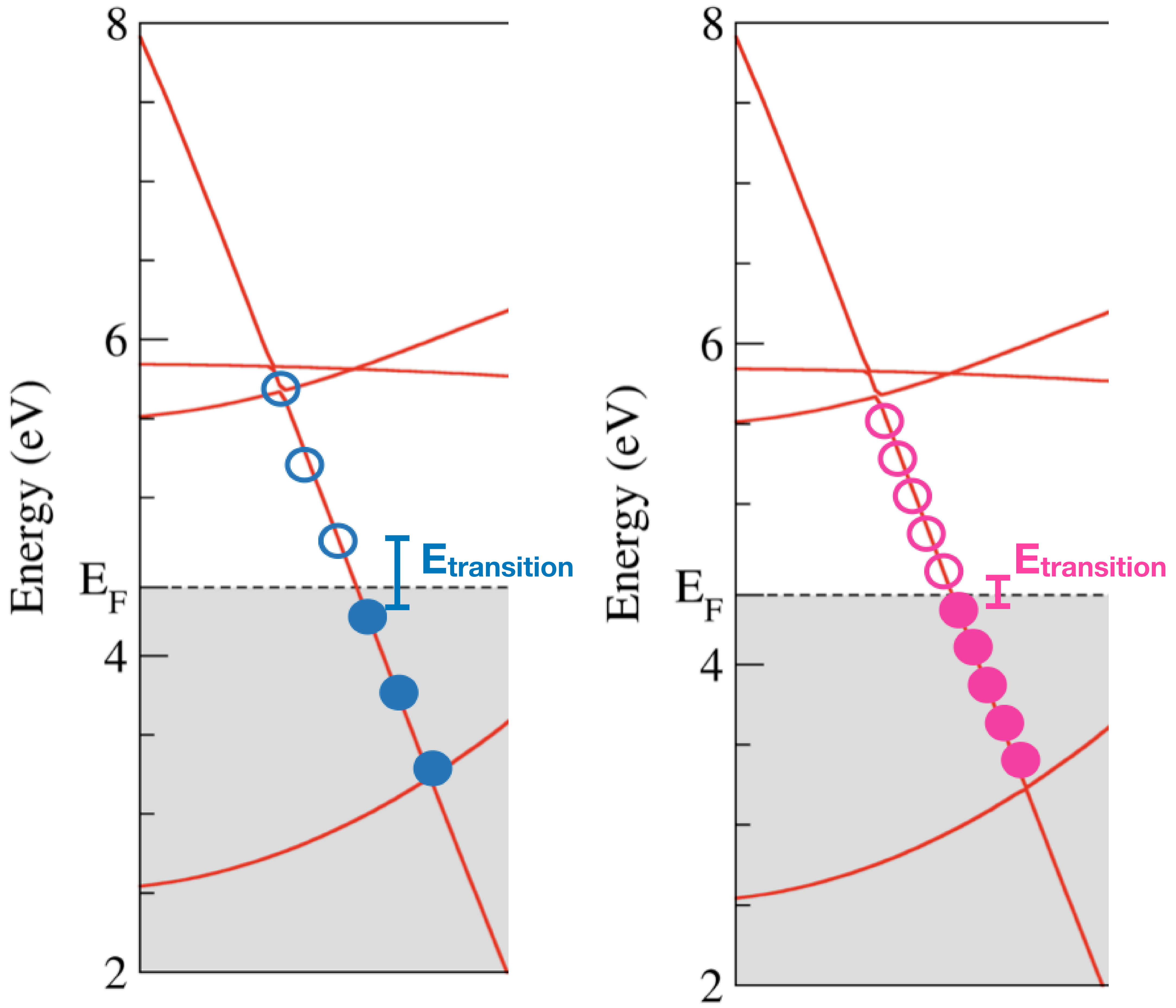
(GW bulk Pb)



# Electron-Electron Relaxation Time

## Possible issues with the *GW* Approach

- *GW* calculations are not well studied for metals - typically done for semiconductors for band gap correction
- Screening may be extremely sensitive to kpoint density because the fermi energy is within the bands
  - Low density:
  - High density:
  - Could make kpoint convergence difficult
- Self-energy from the *GW* calculation may be unreliable
  - The screening term (*W*) may not properly incorporate the influences from intraband transitions that are present in metals




$E_{transition}$ > $E_{transition}$

**How much does screening depend on T?**

# Electron-Electron Relaxation Time

## Screening with and without Drude term

- Look at real part of the dielectric function to find the dielectric constant which can be used to describe screening (without *GW*)

| Dielectric Constant | | |
|---|---|---|
| | **Sign** | **Magnitude** |
| No Drude term | + | Small |
| With Drude term | - | Large |

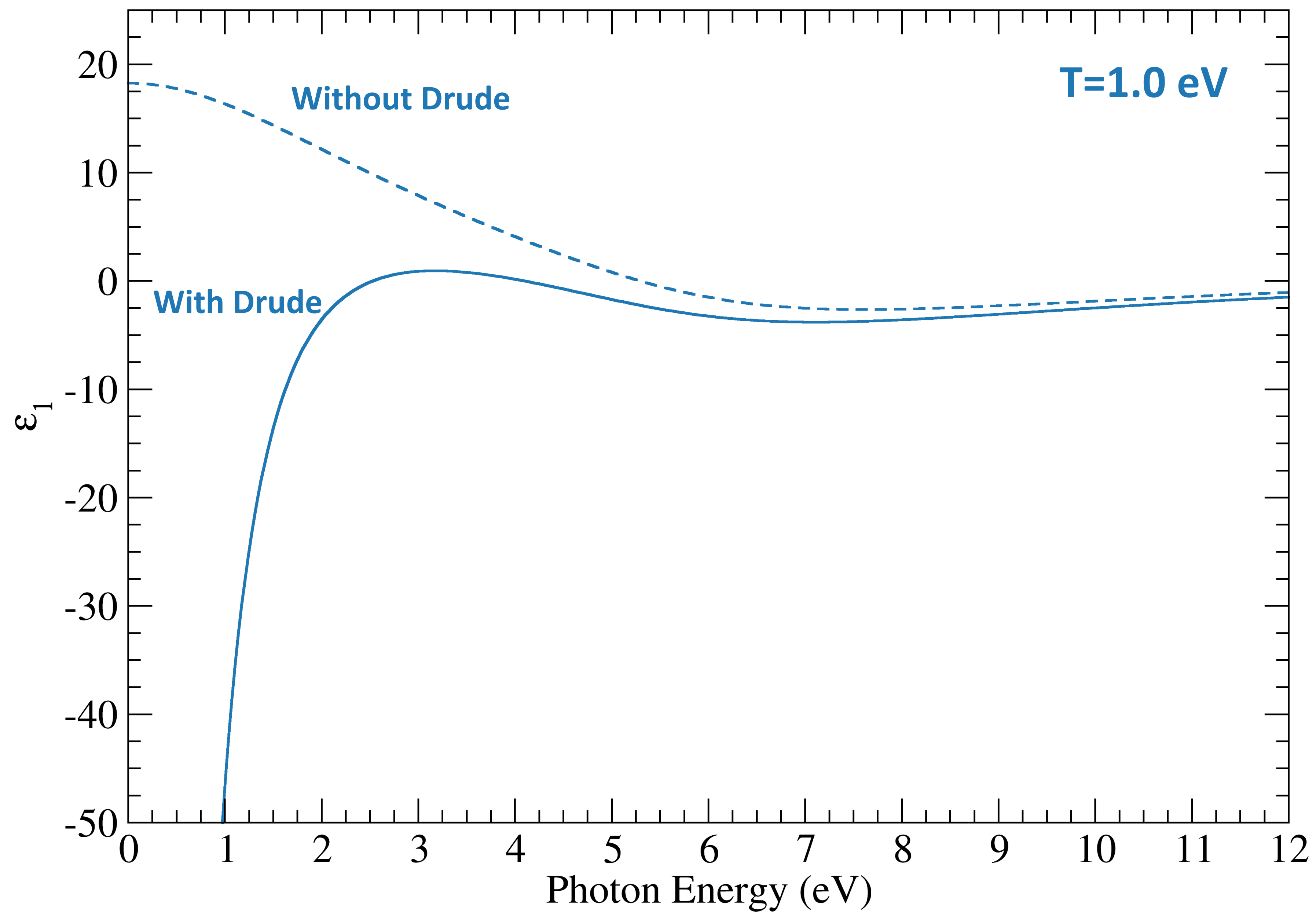
  - Intraband transitions have a large influence on screening

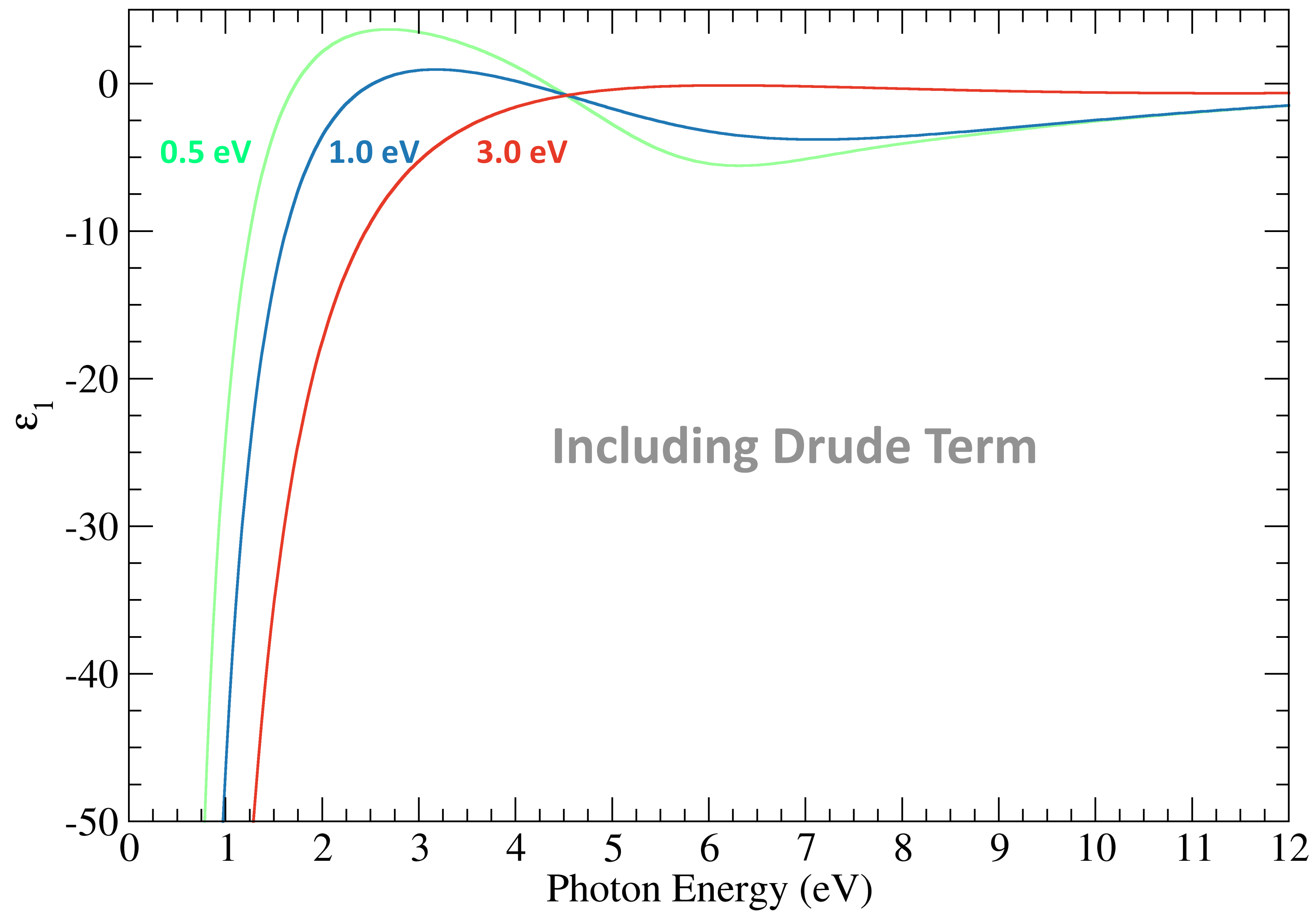
## Screening Temperature Dependence

- As temperature increases, the dielectric constant becomes larger in magnitude

## What does this mean for the *GW* calculation?

- Screening from intraband transitions have a significant influence on the system
- This must be accounted for properly in the *GW* calculation in order to produce reliable electron-electron relaxation times



# Conclusion

## Summary

- PBE and LDA calculations produce similar results for the electronic density of states and electronic band structure
  - Justifies using either the PBE or LDA functional
- Intraband transitions have a strong influence at all temperatures on absorption up to 2.5 eV
- We can take a *GW* approach to obtain electron-electron relaxation times
- Must completely converge kpoint grid
  - Must account for the influence intraband transitions on screening for all temperatures

## Future Work

- Study the influence of electron-phonon interactions in beryllium
- Benchmark and constrain the average atom method against density functional theory

# References

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