Resonant Raman in ABINIT

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Y. Gillet, M. Giantomassi & XG, Phys. Rev. B88, 094305 (2013) – first-order
Y. Gillet, S. Kontur, M. Giantomassi, C. Draxl & X. Gonze, subm. to Sci. Reports
Y. Gillet, PhD thesis.
https://dial.uclouvain.be/downloader/downloader.php?pid=boreal:182880&datastream=PDF_01

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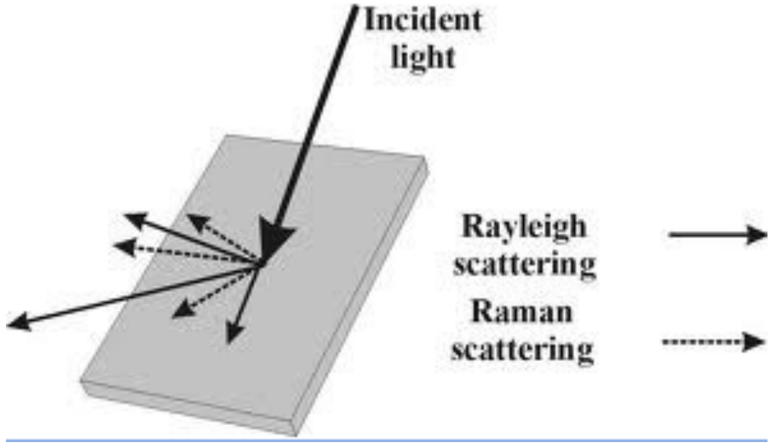
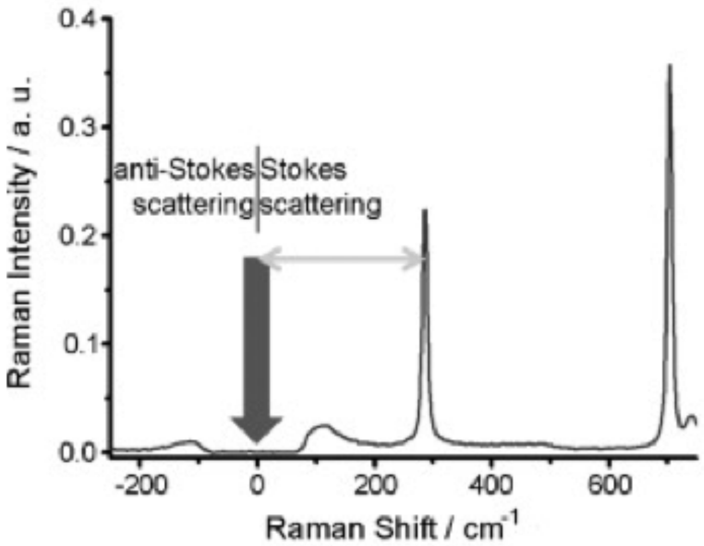
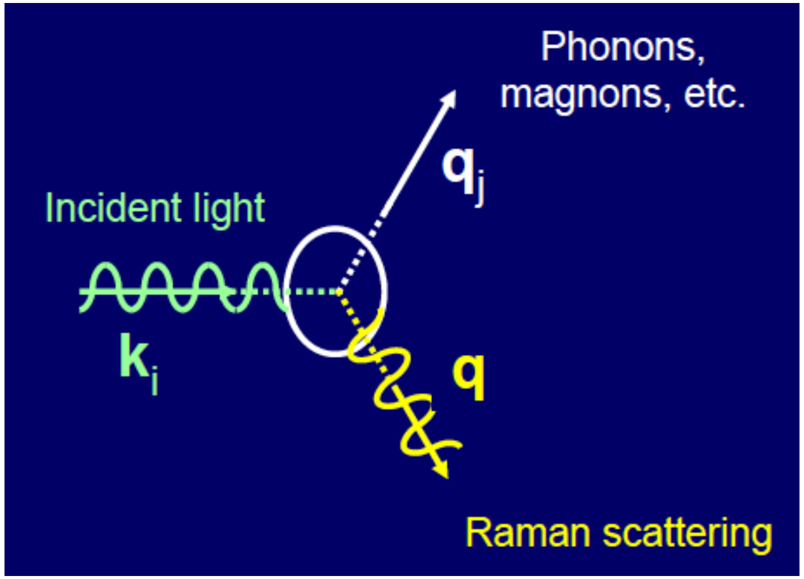
Raman spectroscopy

Prediction A. Smekal (1923)
Discovery C.V. Raman & K.S. Krisnan (1928)

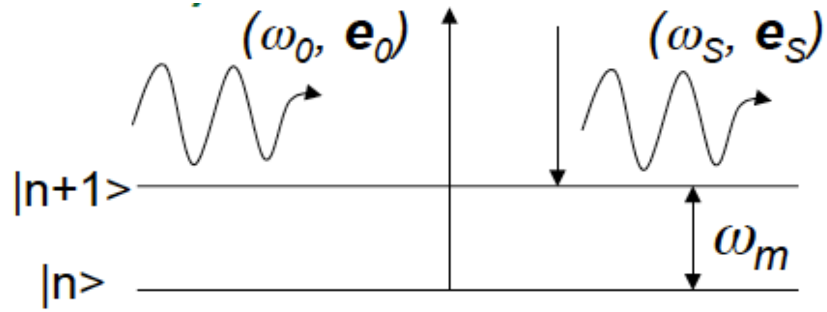






Stokes contribution from phonons

Incoming photon (ω_0, $\mathbf{e}_0$) scattered to an outgoing photon (ω_S, $\mathbf{e}_S$) by creating a phonon ω_m



$$\frac{dS}{dV} = \frac{(\omega_0 - \omega_m)^4}{c^4} \left| \mathrm{e}_S \cdot \alpha_m \cdot \mathrm{e}_0 \right|^2 \frac{\hbar}{2\omega_m} (n_m + 1)$$

Raman susceptibility :

$$\alpha_{jk}^{m} = \sqrt{\Omega_0} \sum_{\kappa\beta} \frac{\partial \chi_{jk}}{\partial R_{\kappa\beta}} \xi_m(\kappa\beta)$$

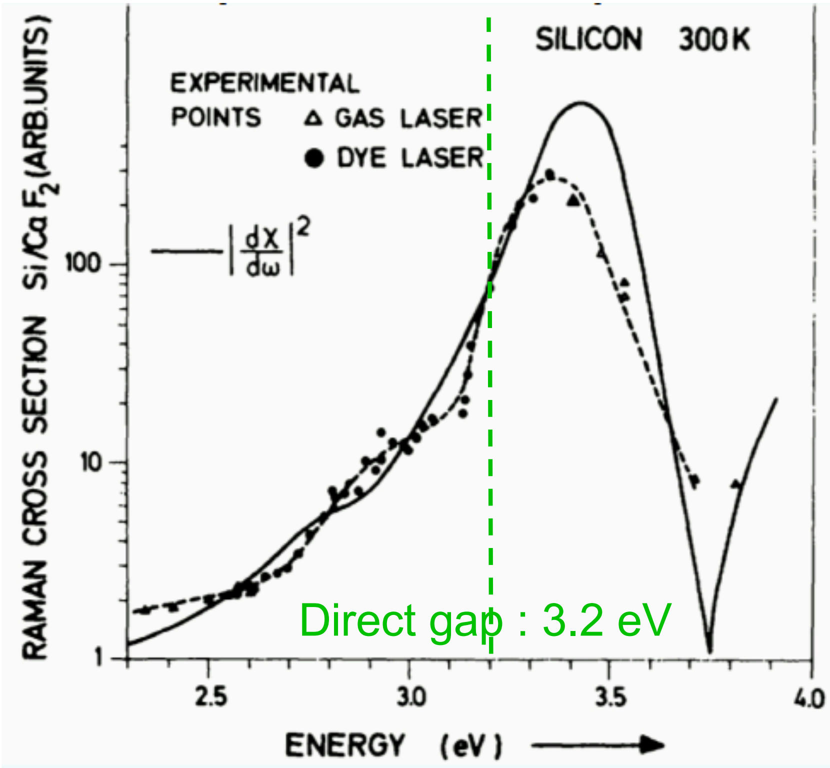
Boson factor :

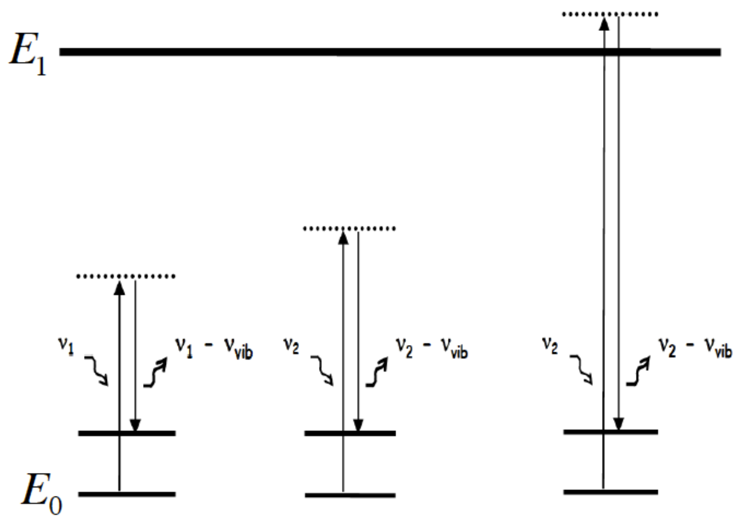
$$n_m = \frac{1}{e^{\hbar\omega_m / k_B T} - 1}$$

phonon eigenmode (only at Gamma)

Needed : phonon frequencies, eigenmode, Raman susceptibility

LASER frequency dependence





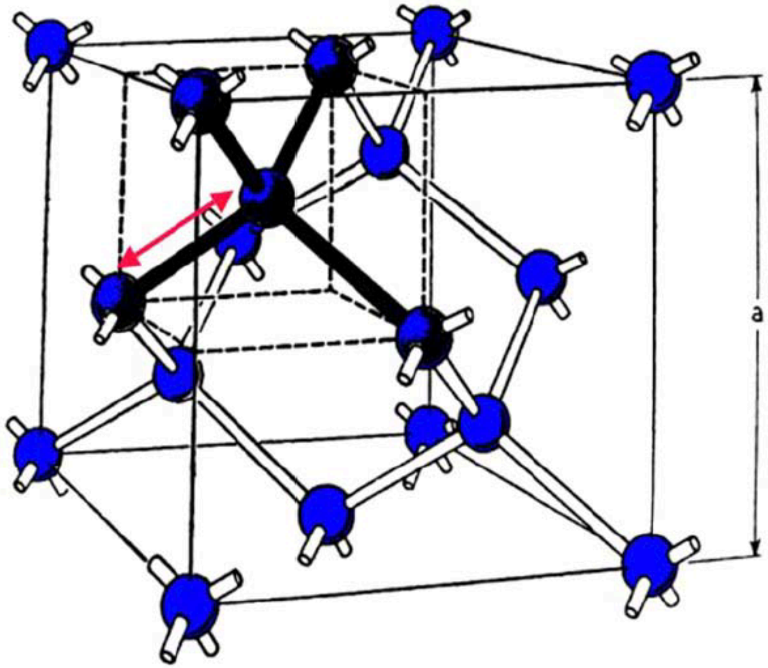
Between 2.3 eV and 3.3 eV, Si Raman cross section increases by about 200

J. B. Renucci, R. N. Tyte and M. Cardona. PRB 11, 3885 (1975)

Frequency-dependent Raman susceptibility

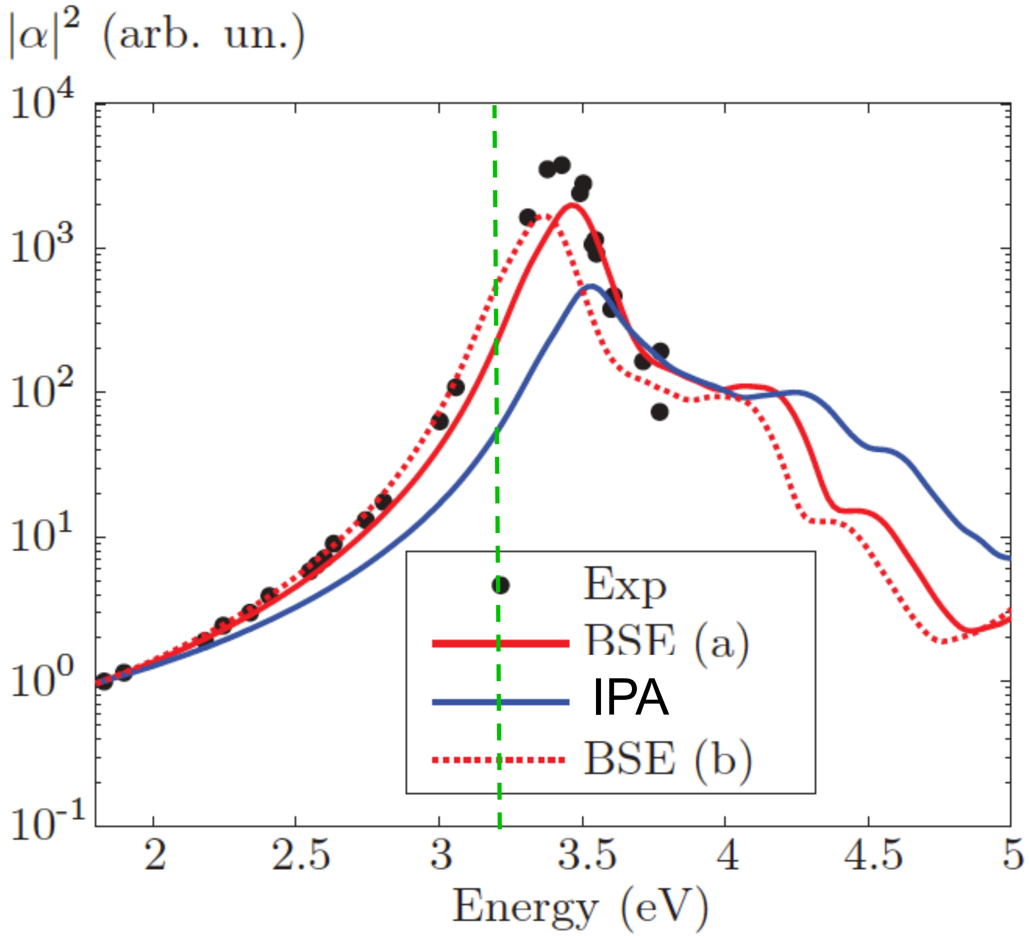
$$\alpha_{jk}^{m}(\omega=\omega_{LASER}) = \sqrt{\Omega_0}\sum_{\kappa\beta}\frac{\partial\chi_{jk}(\omega=\omega_{LASER})}{\partial R_{\kappa\beta}}\,\xi_m(\kappa\beta)$$

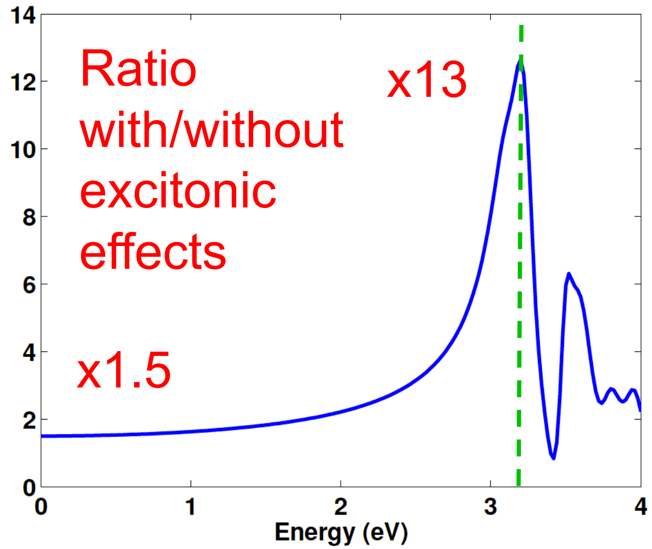
Frozen-phonon approach :
the phonon mode is known



$$\alpha_{jk}^{m}(\omega) = \sqrt{\Omega_0}\lim_{\lambda\to 0}\frac{\chi_{jk}(\{R+\lambda\xi_m\};\omega)-\chi_{jk}(\{R\};\omega)}{\lambda}$$

Raman intensity dependence on LASER frequency : 1st-order results





Large excitonic effect : one order of magnitude !

Silicon

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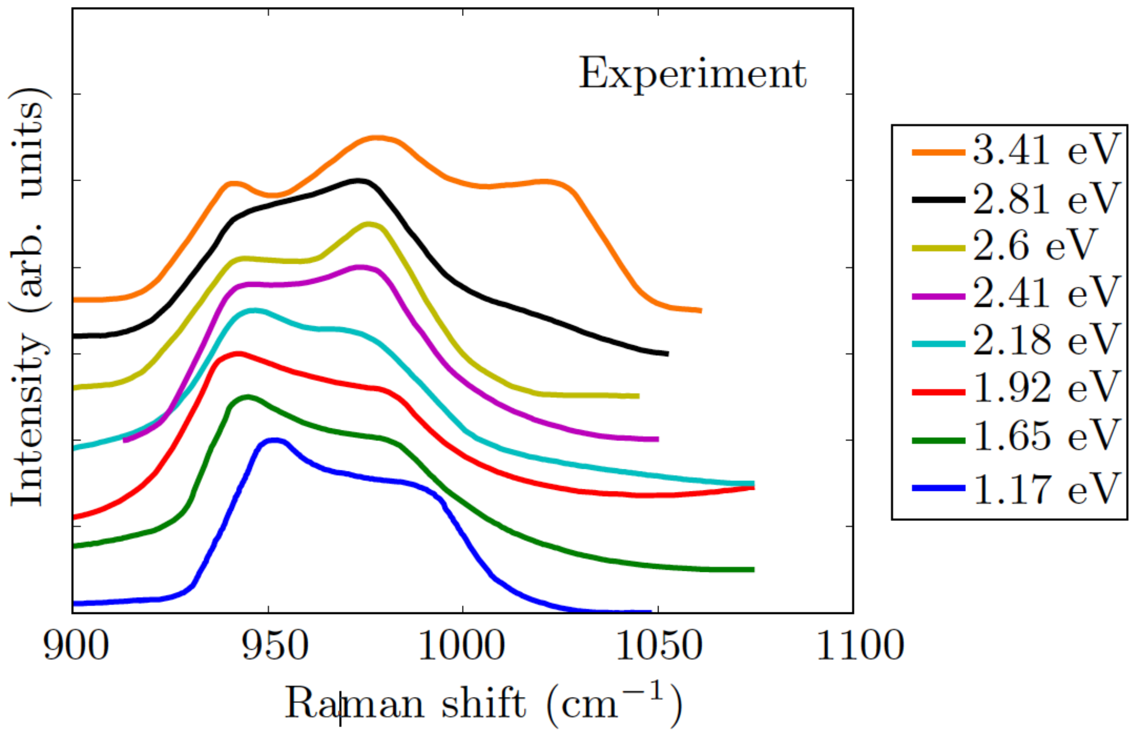
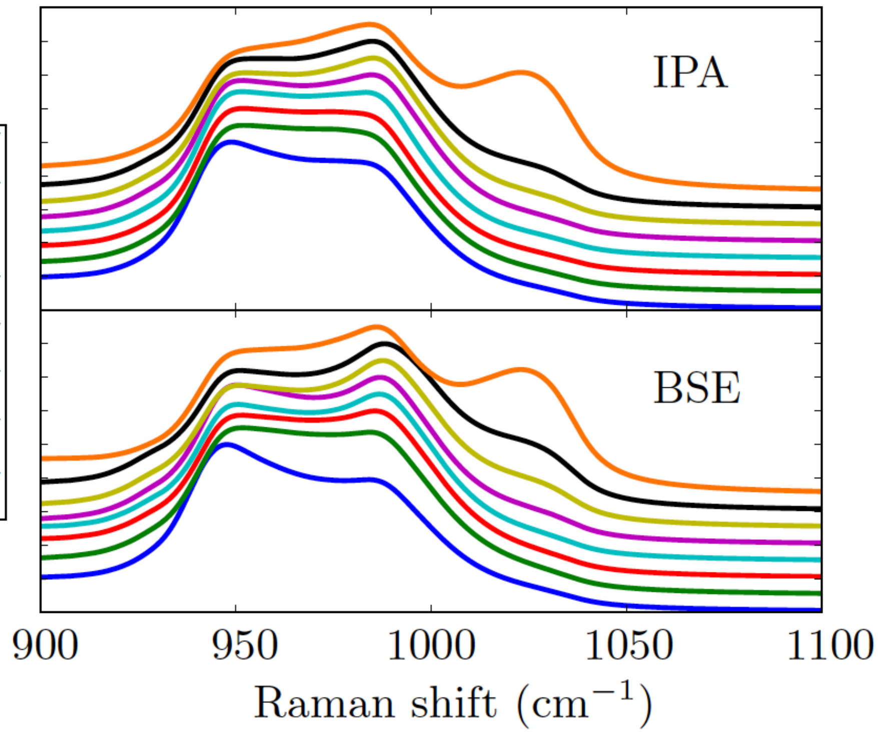
Phys. Rev. B 88, 094305 (2013)

Raman intensity dependence on LASER frequency : 2nd-order results

J.B.Renucci et al, PRB **11**, 3885 (1975)



Théorie



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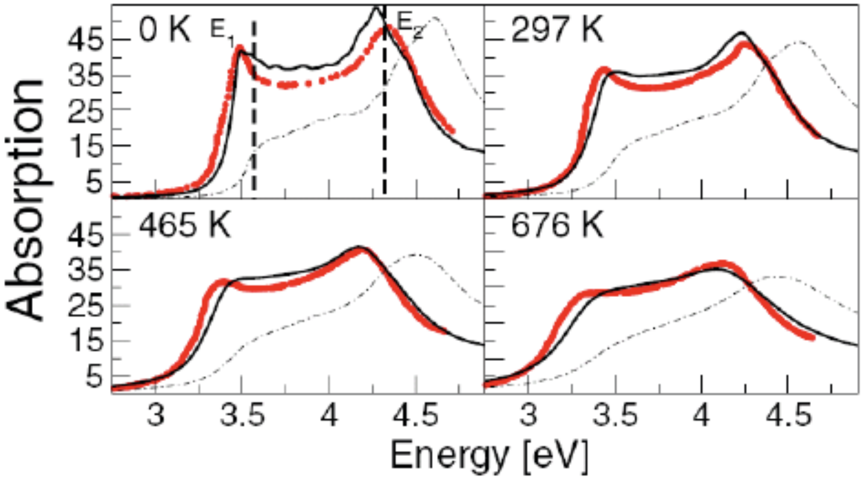
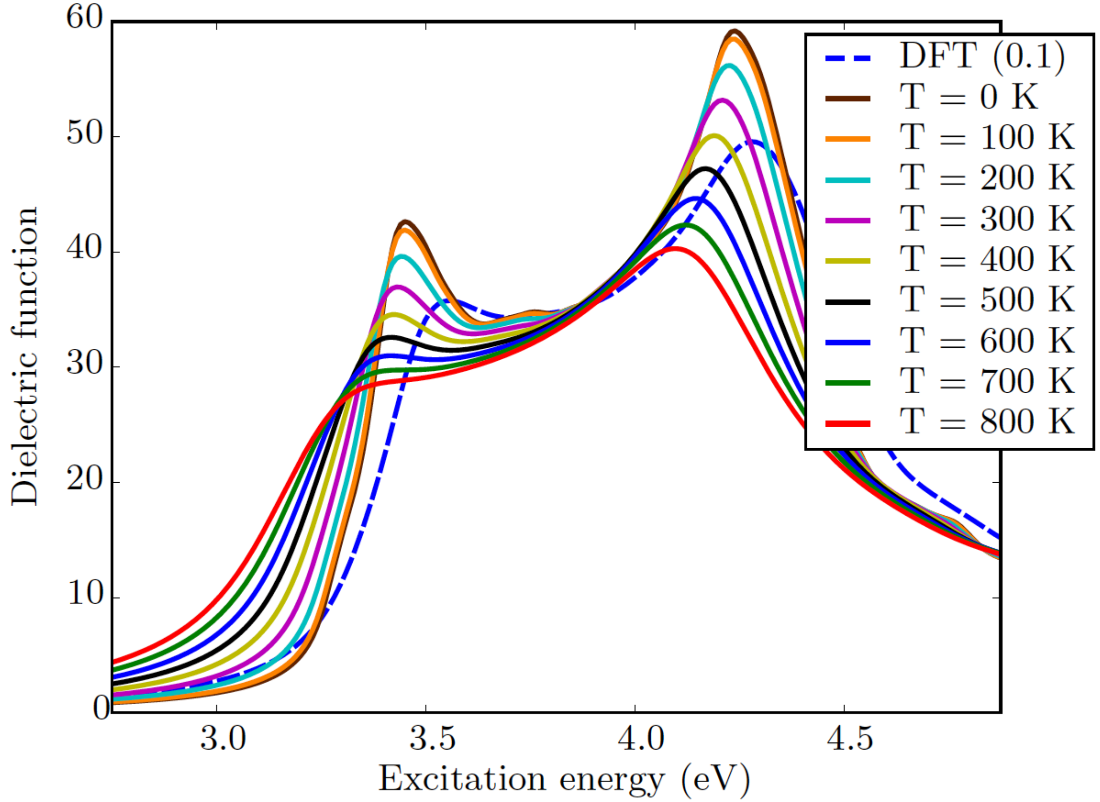
Y. Gillet, et al, Sci. Reports, subm.

Dependence on temperature of frequency-dielectric response



A. Marini, *Physical Review Letters* **101**, 106405 (2008)



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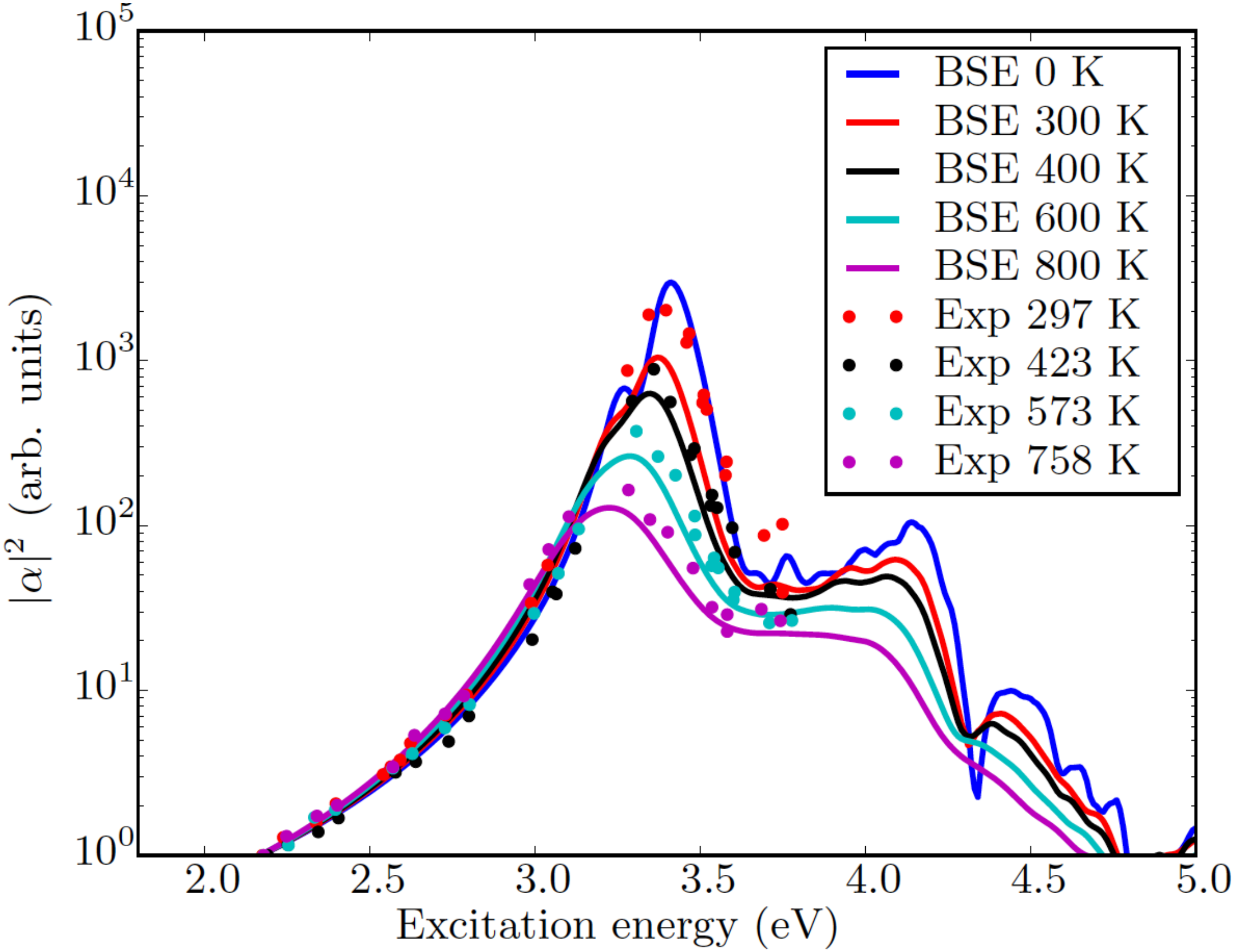
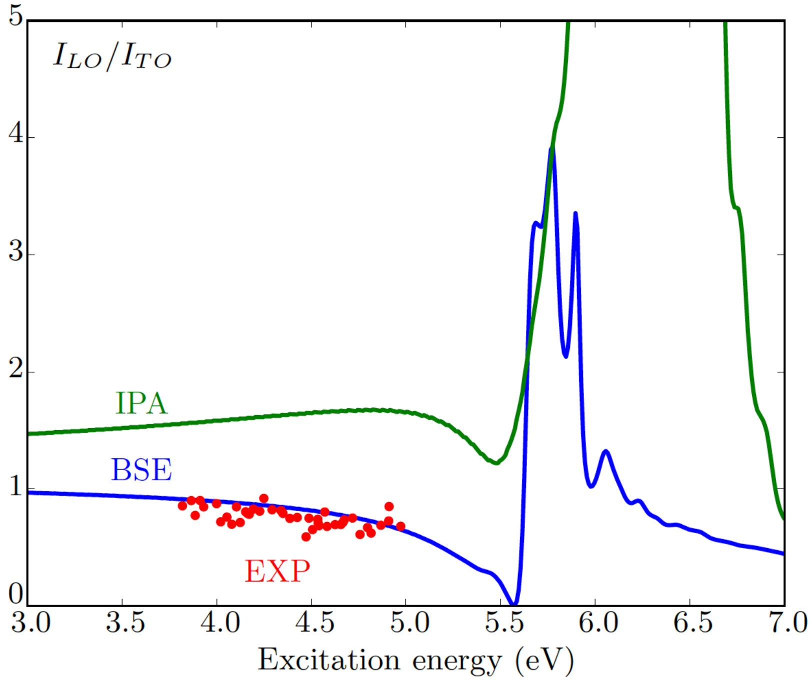
Temperature-dep. Resonant Raman



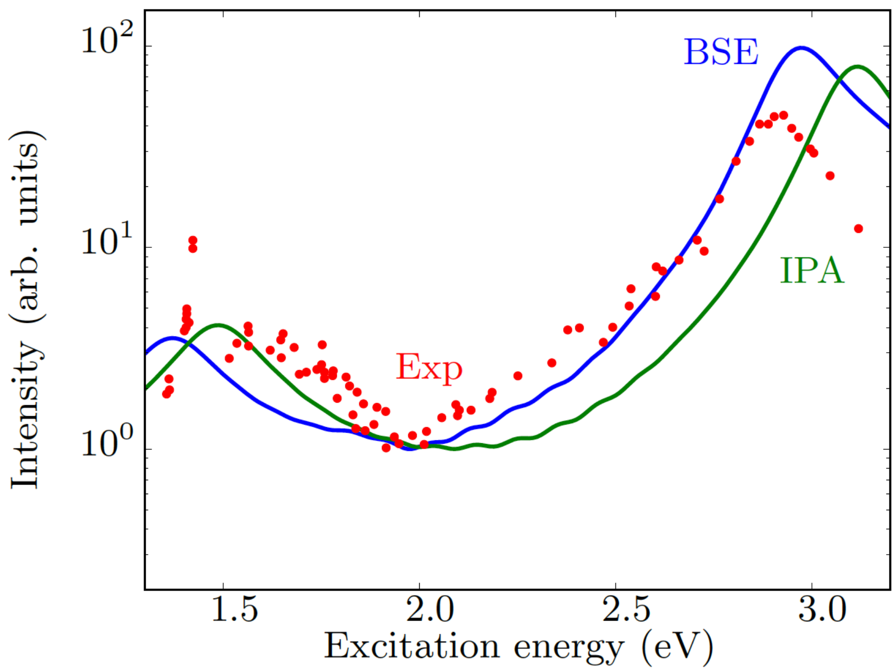
Silicon

Polar materials

Needs LEO (Linear electro-optic) contribution.



Silicon carbide



Gallium arsenide

Implementations within ABINIT

Many finite-difference calculations : AbiPy

In main ABINIT :

- Bethe-Salpeter, many improvements incl. Haydock
- Also T-dep (non-hermiticity)
- Linear Electro-Optic

Thanks for your attention !