Photoluminescence

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Theory

Absolute luminescence intensity:

$$I(\hbar\omega) = \frac{n_D \omega^3}{3\epsilon_0 \pi c^3 \hbar} |\vec{\mu}_{eg}|^2 \sum_m |\langle \chi_{gm} | \chi_{e0} \rangle|^2 \, \delta(ZPL - E_{gm} - \hbar\omega)$$

Normalized luminescence intensity:

$$L(\hbar\omega) = C\omega^3 A(\hbar\omega), \qquad A(\hbar\omega) = \sum_m |\langle \chi_{gm} | \chi_{e0} \rangle|^2 \, \delta(ZPL - E_{gm} - \hbar\omega)$$

Fourier transform of generating function (with Lorentzian broadening):

$$A(ZPL - \hbar\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} G(t) e^{i\omega t - \gamma|t|} dt$$

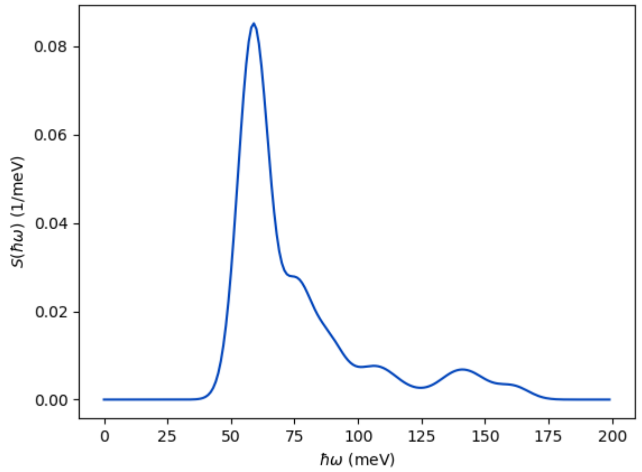
Generating function (where S(t) is the Fourier transform of S):

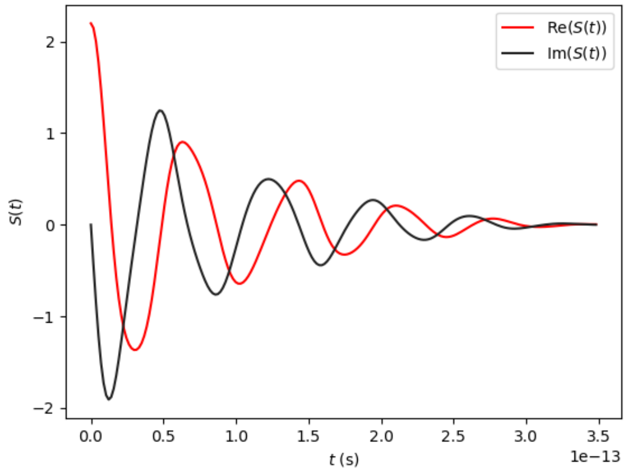
$$G(t) = e^{S(t) - S(0)}, \qquad S(t) = \int_0^{\infty} S(\hbar\omega) e^{-i\omega t} d(\hbar\omega)$$

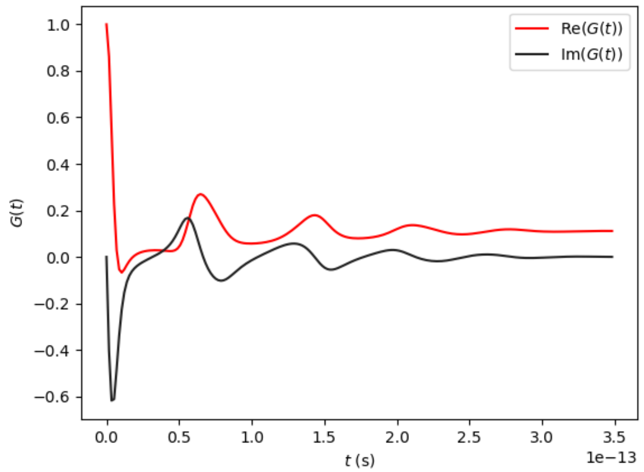
Huang-Rhys function:

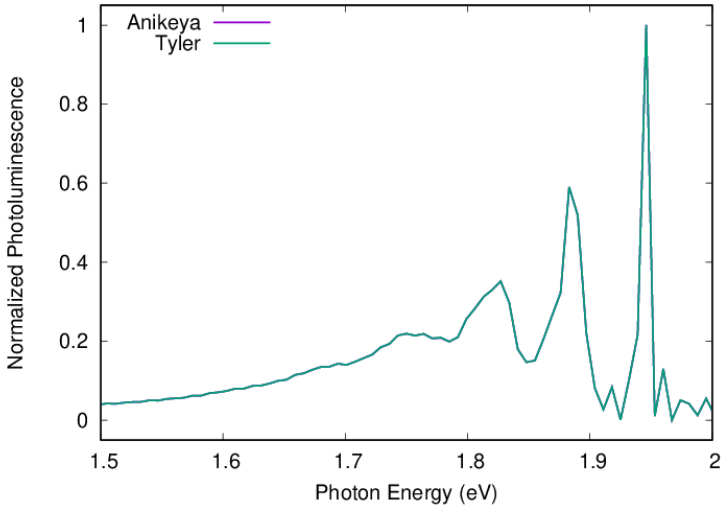
$$S(\hbar\omega) = \sum_k S_k \delta(\hbar\omega - \hbar\omega_k), \qquad S_k = \frac{\omega_k q_k^2}{2\hbar}, \qquad q_k = \sum_\alpha \sqrt{m_\alpha} (R_{e;\alpha} - R_{g;\alpha}) \Delta r_{k;\alpha}$$

Reproduce Anikeya results









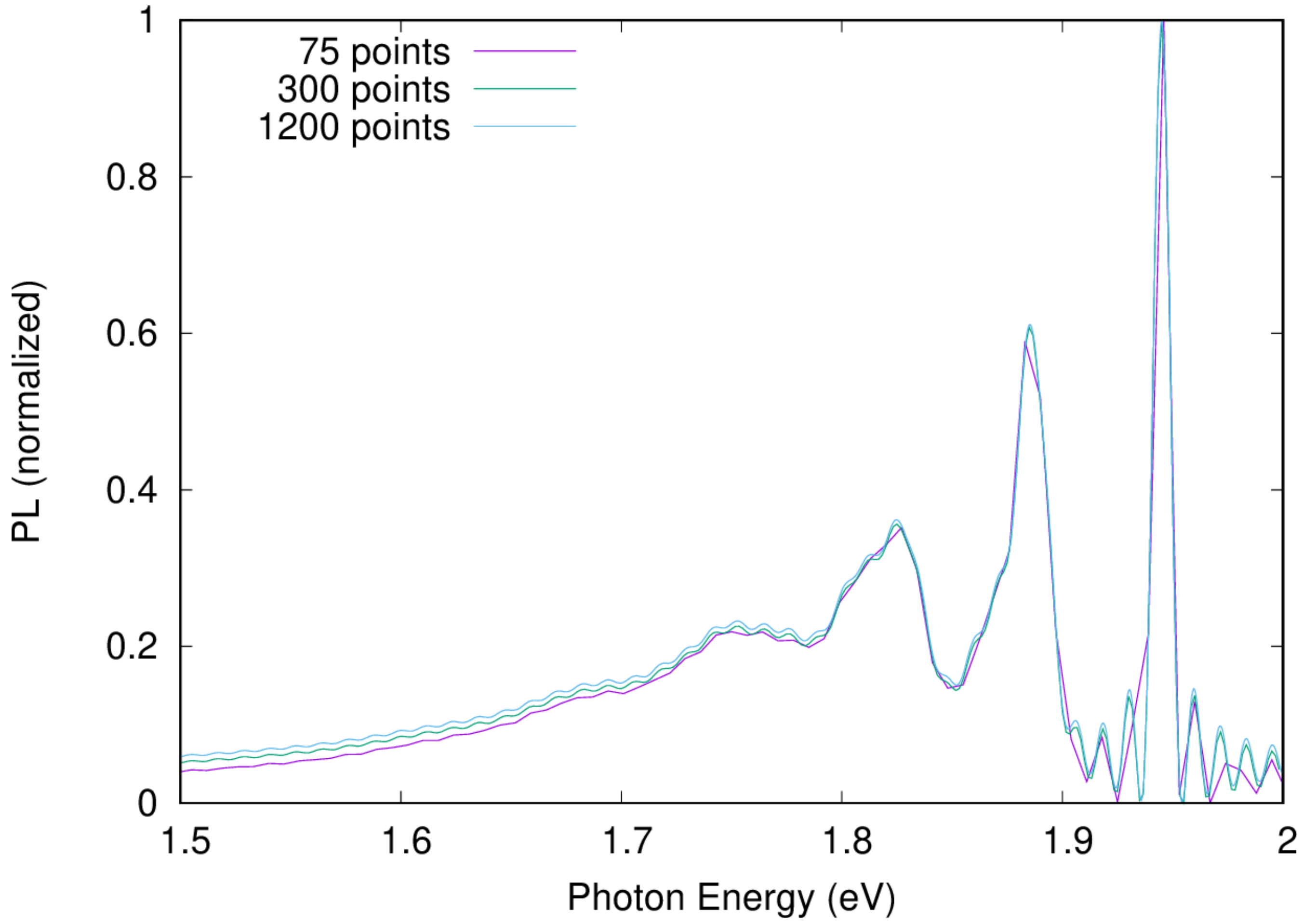
The Python Code

- Improved organization of code itself
  - More use of method makes it easier for future users to grab and modify chunks of the code
- Read directly from QE output
- Input file
  - Can specify: path_to_qe, zpl, skfile, smear, limit, gamma, tolerance, hw_min, hw_max, more…

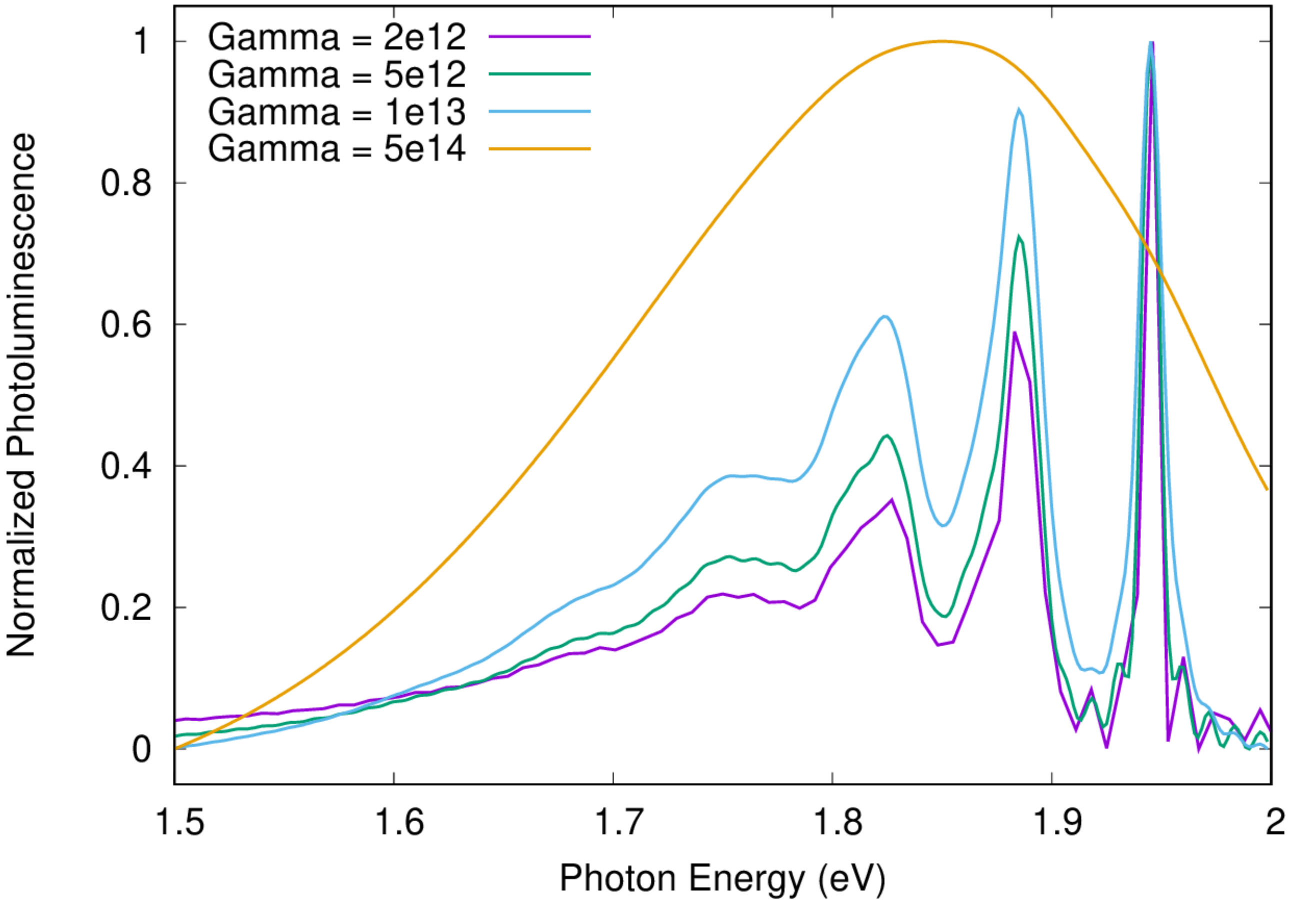
Spectrum tests

- Parameters to consider:
    1. Number of points
    2. Gamma – Lorentzian smearing for ZPL
    3. Smear – gaussian broadening which replaces direc delta
    4. Limit – finite limit used in integration

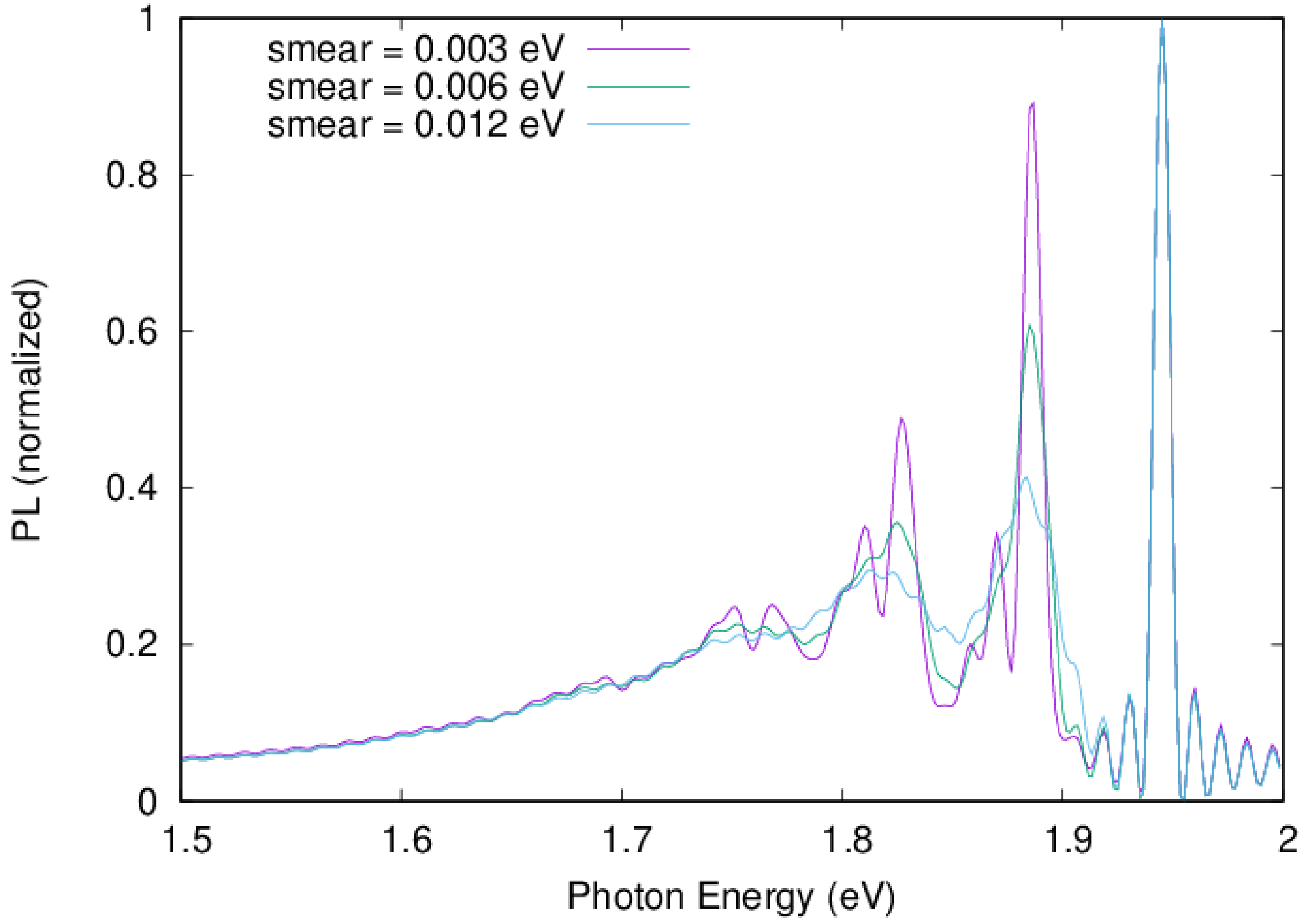
Spectrum tests



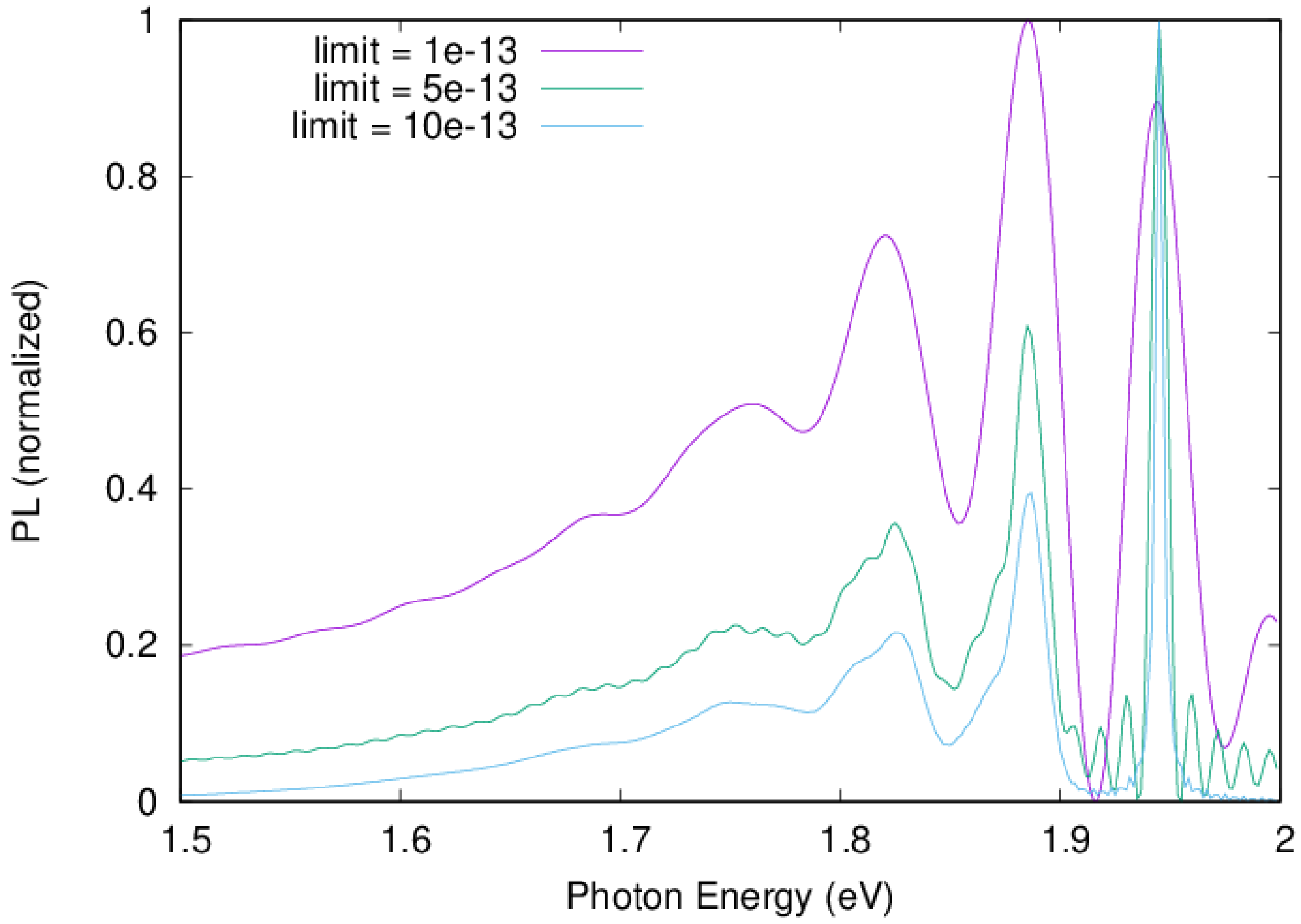
Spectrum tests



Spectrum tests



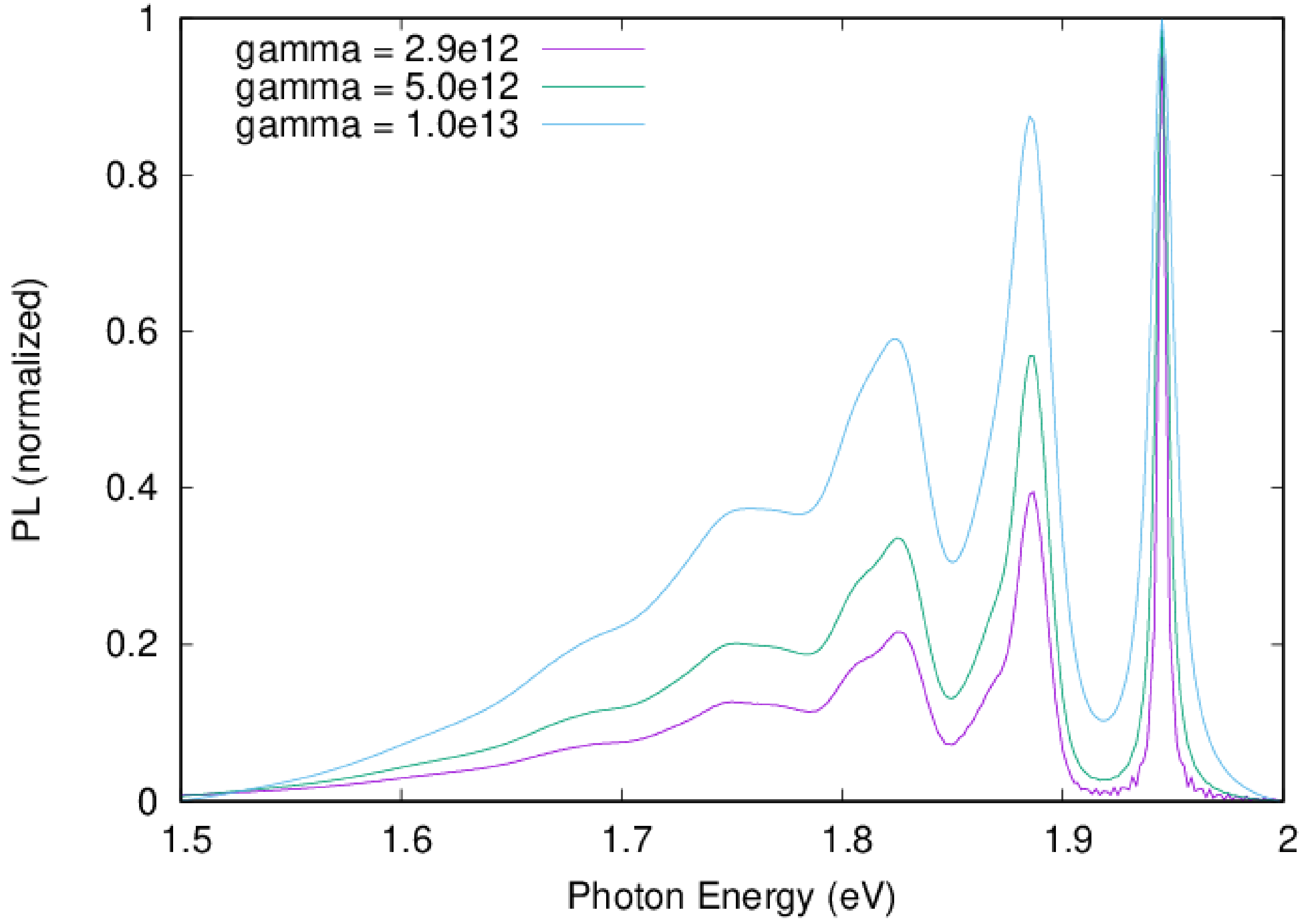
Spectrum tests



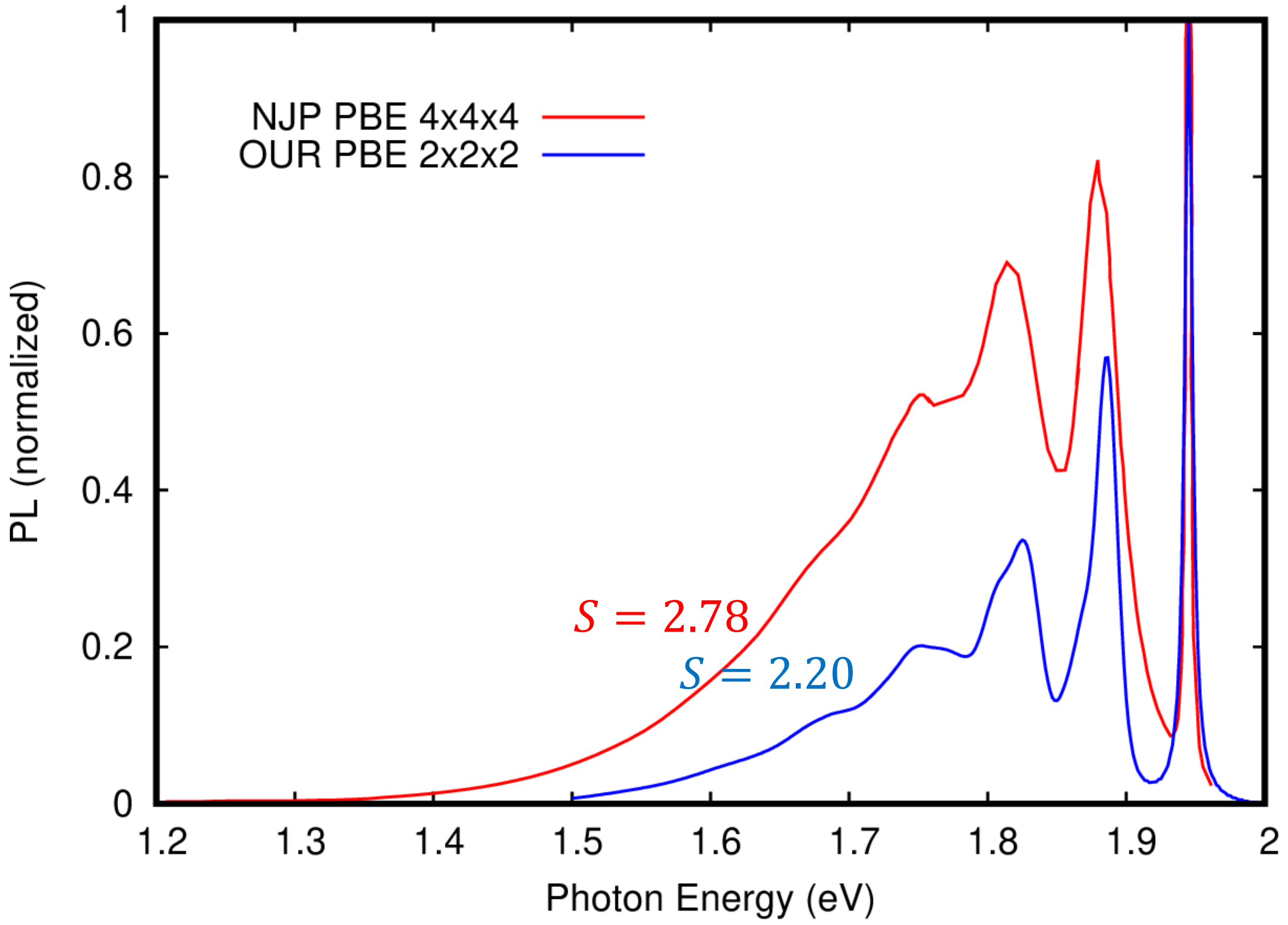
Spectrum tests

- Parameters to consider:
  1. Number of points
     → at least 600/eV
  2. Gamma – Lorentzian smearing for ZPL
     → around 1e12 to 1e13 improving other parameters make width fit more reliable
  3. Smear – gaussian broadening which replaces direc delta
     → around 6 meV seems good; does not effect ZPL
  4. Limit – finite limit used in integration
     → at least 1e-12

Spectrum tests (limit = 1e-12)



Comparing with Alkauskas 2014



NV center in diamond calculations - Stockpile

Code	Functional	Cell Size	ZPL (eV)	HR
QE	PBE	2x2x2	1.641	2.20
QE	PBE	3x3x3	1.720	*phonon IP*
QE	PBE	4x4x4	1.704	*phonon IP*
QE	HSE06	2x2x2	2.025	--
QE	HSE06	3x3x3	1.994	--
QE	HSE06	4x4x4	1.965	--
VASP	PBE	2x2x2	1.618	*phonon IP*
VASP	HSE06	2x2x2	1.993	--
VASP	PBE	4x4x4	1.706 [1]	--
VASP	HSE06	4x4x4	1.955 [1]	--
VASP	PBE	4x4x4	1.757 [2]	2.78*
VASP	HSE06	4x4x4	2.035 [2]	3.67* (3.63, 3.02)
Experiment	--	--	1.945 [3]	3.5** [4]

[1] A. Gali, E. Janzén, P. Deák, G. Kresse, E. Kaxiras, *Phys. Rev. Lett.* **103**, 186404 (2009).

[2] A. Alkauskas, B. B. Buckley, D. D. Awschalom, C. G. Van de Walle, *New J. Phys.* **16**, 073026 (2014). *phonon always at PBE & supercell implant method (3.63@4x4x4 3.02@2x2x2)

[3] G. Davies and M. F. Hamer, *Proc. R. Soc. A* **348**, 285 (1976).

[4] A. Gali, *arXiv* 1906.0047 (2019). **approximated from experimental debye walle factor (DW) where S = -ln DW

Conclusions & Outlook

- Conclusions
  - Code improved for better usability
  - Tests parameters show some parameters needed better converging (# of points, limit) others can be tuned as discussed by Alkauskas (gamma, smear)
  - NV center in diamond is useful testing—having many calculations is useful for PL, ZFS, and more
- Outlook
  - We decided last week to switch to DLTS so this work will be done in the background as focus is now on implementing full-phonon
  - Phonon dispersion for NV diamond supercell are great calculations to run on the weekend or whenever the cluster is empty