NAVAL NUCLEAR LABORATORY

Advancing TSL Generation Methods for ENDF/B-IX

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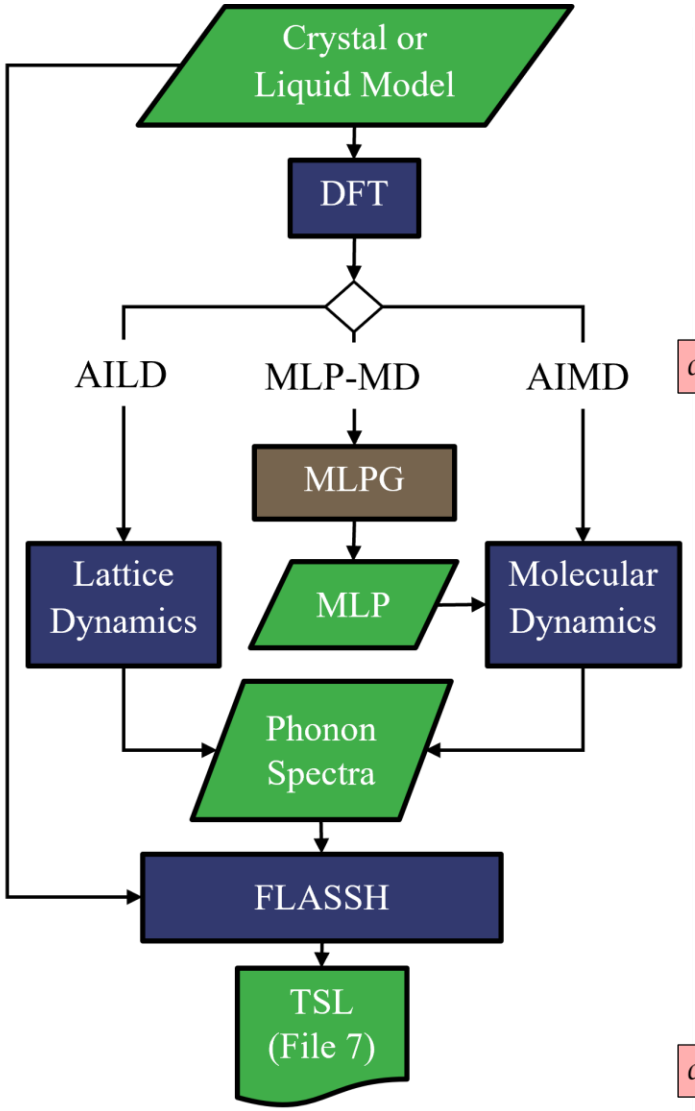
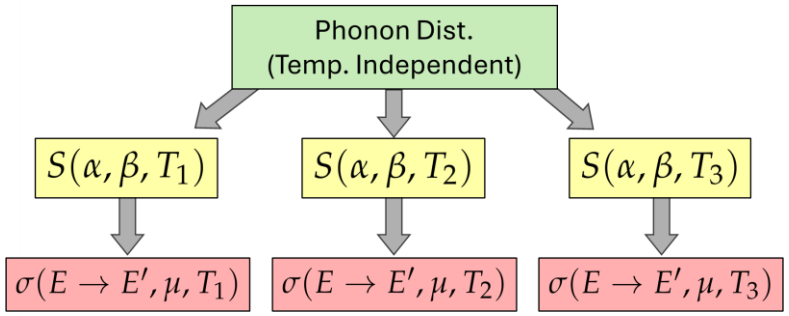
Nuclear Data Week 2026: CSEWG, Evaluations

Motivation

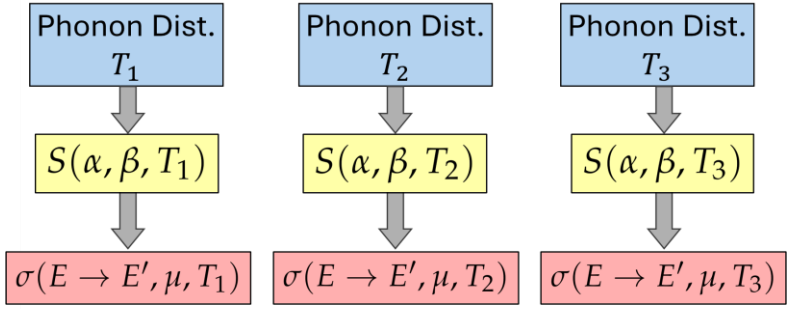
- ENDF/B-VIII.1 solid material TSLs evaluated with temperature-independent material models
- Aim to relax temperature-independent phonon assumption for ENDF/B-IX
  - Impact on criticality and temperature feedback (https://doi.org/10.1016/j.anucene.2024.111034)
- Relax several of the remaining approximations used in ENDF/B-VIII.1 quantum materials simulation
  - *Ab initio* lattice dynamics (AILD) omits key temperature-dependent effects
  - *Ab initio* molecular dynamics (AIMD) simulation sizes are too small
- Machine-learned potentials (MLPs) techniques address limitations of currently used force fields
  - MLP with molecular dynamics (MD) approach (https://doi.org/10.1016/j.anucene.2024.110978)
  - Trained on the same quantum force field as AILD and AIMD



AILD

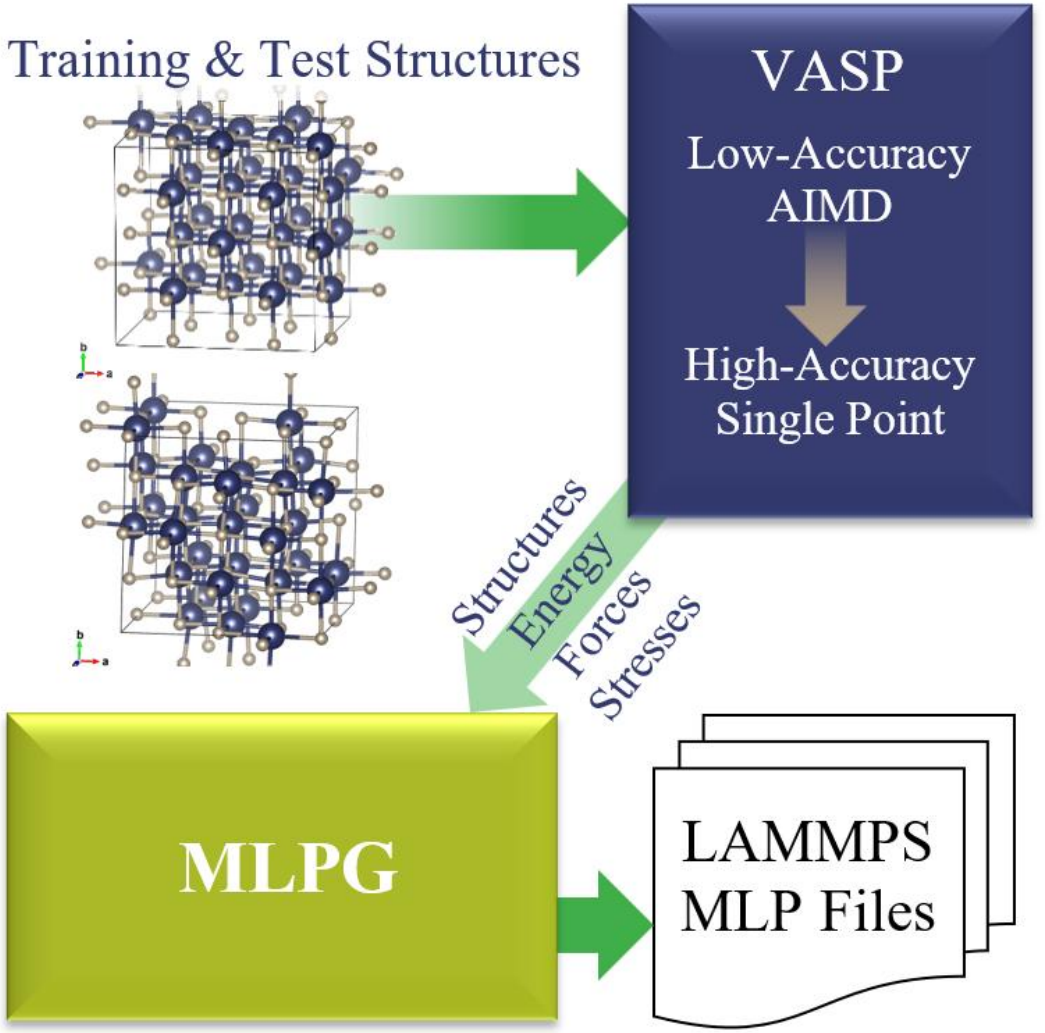


MLP-MD



MLP Generation

- MLP constructed from supervised machine-learning (many types)
  - Map atomic environment to energy
  - Accurate for wide range of properties
    - https://doi.org/10.1557/s43578-023-01239-8
- MLP generator (MLPG) developed in MedeA code
  - Sponsored by Naval Nuclear Laboratory (2020-2022)
  - Simplifies MLP generation
    - One environment for everything
    - MLP for TSL created in <1 week
- ZrH_x, YH_x, MgO, ZrC have been developed
- Simple AIMD training sets suitable for most materials; high-temperature systems may need more sophisticated training sets



Improved Numerical Accuracy in Phonon Generation Methods for TSLs

- TSL are generated from phonon spectra with the phonon expansion

$$S_s(\alpha,\beta,T) = e^{-\alpha\lambda}\sum_P \frac{\alpha^P}{P!}J^{\star P}\,;\quad J(\beta) = \frac{\rho(\beta)}{\beta(\exp(\beta)-1)};\quad \lambda = \int_{-\infty}^{\infty} J(\beta)\mathrm{d}\beta$$

- MD temperature-dependent phonons historically calculated with the velocity autocorrelation function (VACF)

$$\rho_{\mathrm{v}}(\omega) = \frac{\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{v}(t')\otimes\boldsymbol{v}(t+t')\rangle_{t'})]}{\frac{1}{2}\mathrm{Tr}(\langle \boldsymbol{v}(t')\otimes\boldsymbol{v}(t')\rangle_{t'})}$$

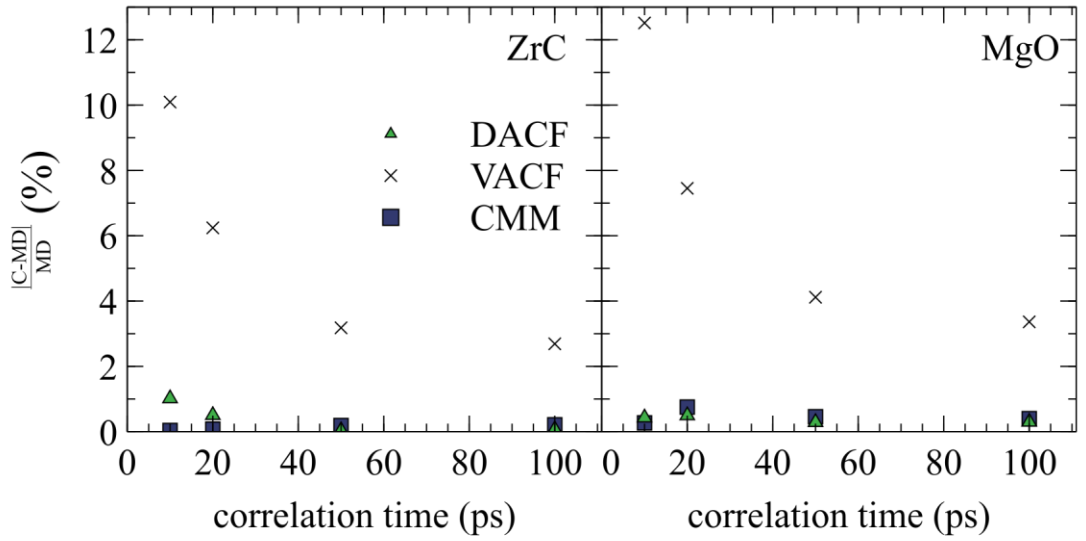
- Phonon spectral function, $J(\beta)$, is defined by the displacement autocorrelation function (DACF) not velocity. With classical MD

$$\rho_{\mathrm{u}}(\omega) = \frac{\omega^2\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{u}(t')\otimes\boldsymbol{u}(t+t')\rangle_{t'})]}{\frac{1}{2}\int_{-\infty}^{\infty} d\omega\,\omega^2\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{u}(t')\otimes\boldsymbol{u}(t+t')\rangle_{t'})]}$$

- Phonons must be physically and numerically accurate. A proposed correlation mixing method (CMM) using a switching function $f(\omega)$

$$\rho(\omega) = f(\omega)\rho_{\mathrm{u}}(\omega) + \big(1-f(\omega)\big)\rho_{\mathrm{v}}(\omega)$$

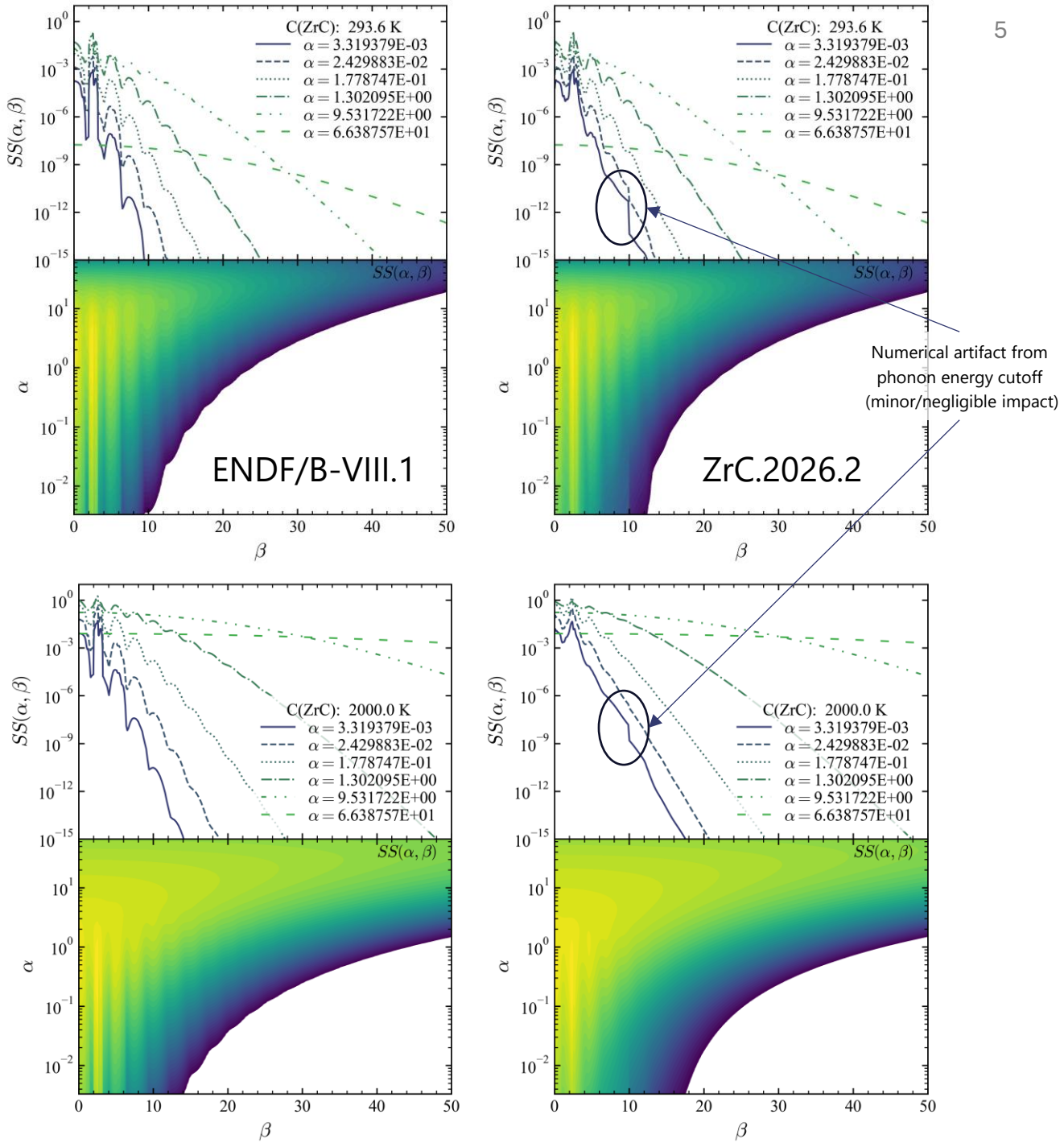
Theoretical Definition	Computation
$\int_{-\infty}^{\infty}\frac{\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{v}(t')\otimes\boldsymbol{v}(t+t')\rangle_{t'})]}{\omega^2}\mathrm{d}\omega = \int_{-\infty}^{\infty}\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{u}(t')\otimes\boldsymbol{u}(t+t')\rangle_{t'})]\mathrm{d}\omega \triangleq 3W = \frac{3\lambda}{Ak_{\mathrm{B}}T}$	Sensitive to $\rho(\omega)$ at low energies
$\int_{-\infty}^{\infty}\omega^2\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{u}(t')\otimes\boldsymbol{u}(t+t')\rangle_{t'})]\mathrm{d}\omega = \int_{-\infty}^{\infty}\mathcal{F}[\mathrm{Tr}(\langle \boldsymbol{v}(t')\otimes\boldsymbol{v}(t+t')\rangle_{t'})]\mathrm{d}\omega \triangleq \frac{3k_BT}{M}$	Insensitive to $\rho(\omega)$ at low energies



Error of computed W for each method from direct output of classical MD simulation as a function of correlation time.

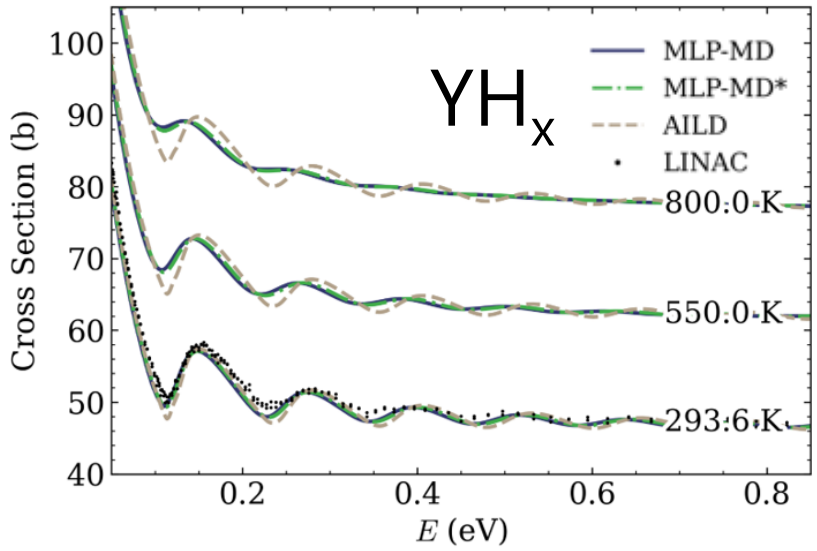
MLP-MD TSL

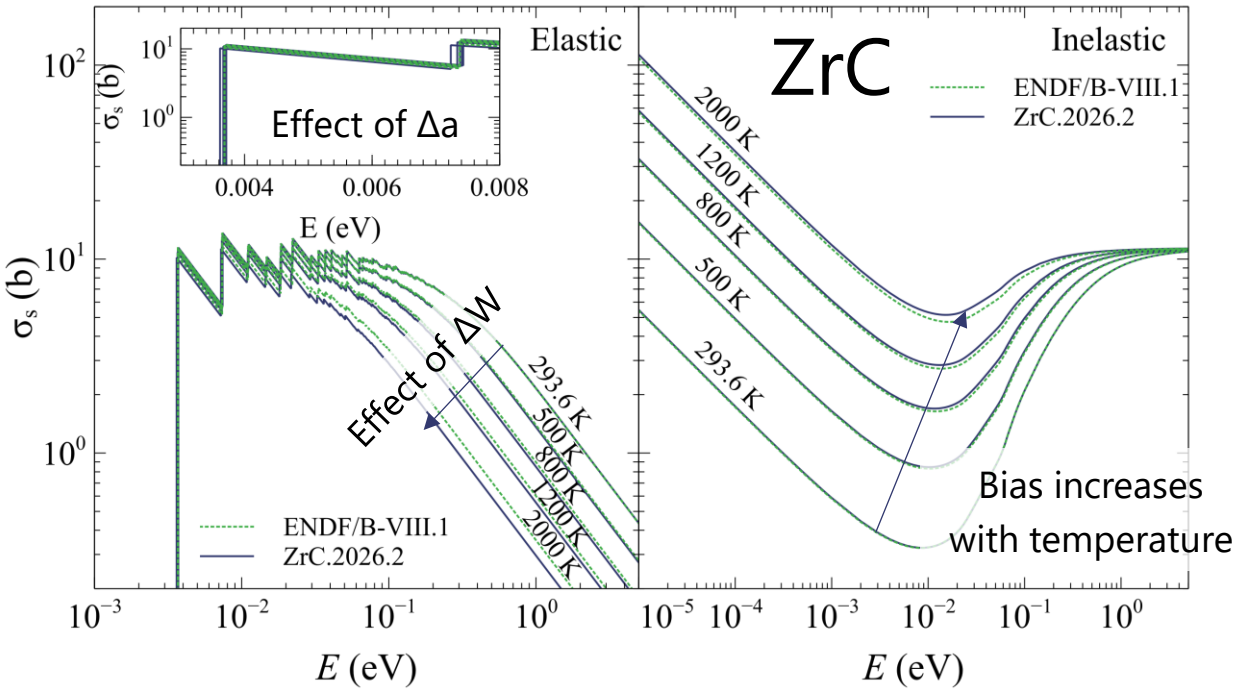
- Reevaluated ZrC with FLASSH using $\rho(\omega)$ for experimental thermal dilation
  - 293.6 – 2000 K
  - Max energy of 0.25 eV
  - ENDF/B-VIII.1 nuclear/mass parameters
  - Selected $\rho(\omega)$ energy cutoff has negligible impact on TSL
- Quantized peaks (oscillations) in C(ZrC) occur at integer multiples of optical phonon peak
- Temperature-dependent phonons impact C(ZrC) and Zr(ZrC)
  - Decrease spacing of quantized peaks
  - Diminished features/oscillations with increasing temperature



MLP-MD Cross Section

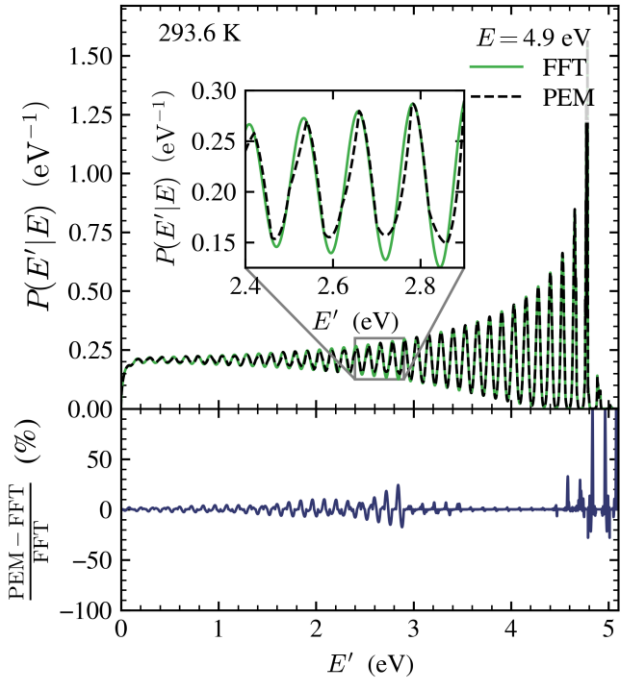
- YH_x reevaluation uses temperature-dependent $\rho(\omega)$
  - https://doi.org/10.1016/j.anucene.2024.110978
- ZrC reevaluation uses temperature-dependent $\rho(\omega)$ and experimental lattice parameters
- Effect of temperature-dependent phonons
  - Larger effect on coherent elastic scattering through W than lattice parameter
  - Minimal effect on inelastic cross section at low temperatures but appreciable at high temperature



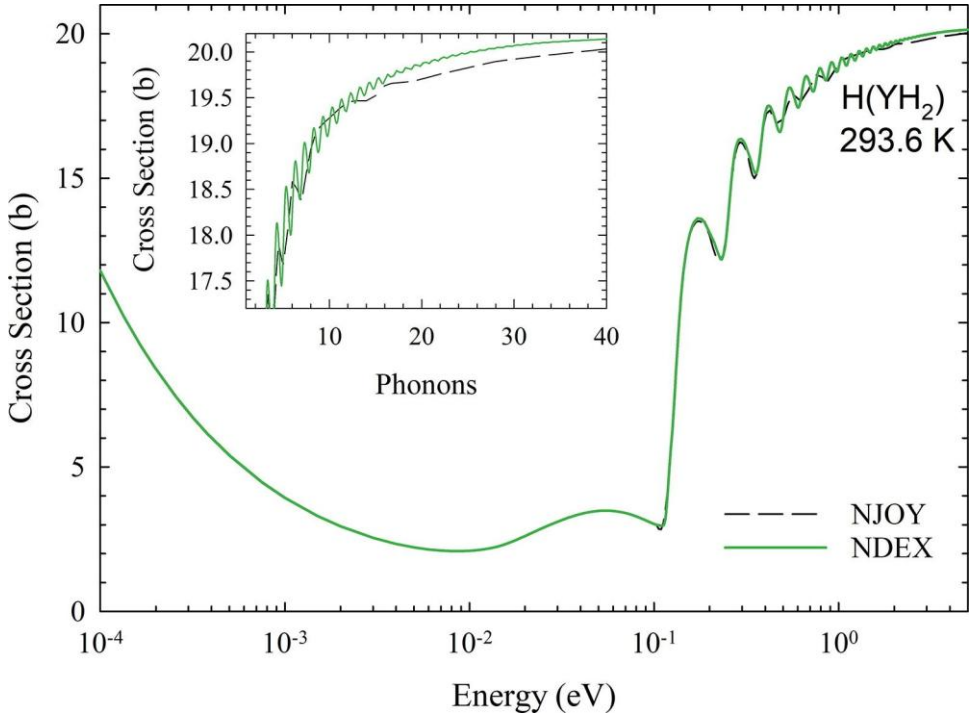


Other TSL Generation/Processing Methods

- High-resolution TSLs using FFT
  - Computed infinite phonon expansion; eliminates phonon order
  - Important for structured TSLs and Doppler broadening applications beyond 5 eV
  - https://doi.org/10.1016/j.anucene.2024.110553
- Adaptive cross section energy grid
  - NDEX method to improve resolution of integrated cross sections
  - Significantly improves treatment of oscillatory cross section of metal hydrides
  - Adaptive grid ensures converged cross section in contrast to fixed grid
  - https://doi.org/10.1016/j.anucene.2020.107773



FFT method compared to traditional phonon expansion method (PEM) for the ENDF/B-VIII.0 YH_2 TSL phonon spectrum



Adaptive NDEX energy grid compared to fixed energy grid in NJOY2012 when processing the ENDF/B-VIII.0 YH_2 TSL evaluation