Thermal Neutron Scattering Law Evaluations for Zirconium Carbide and Critical Mass Calculations

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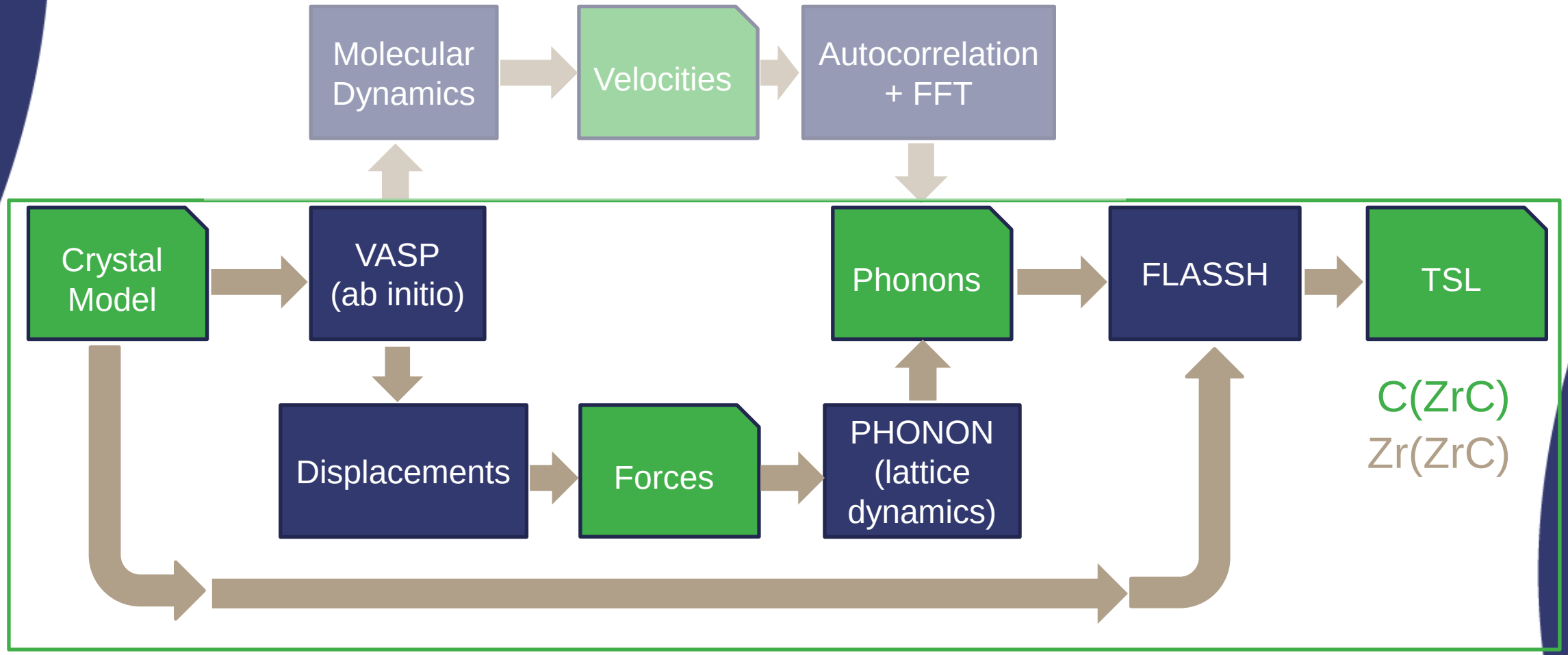
Overview

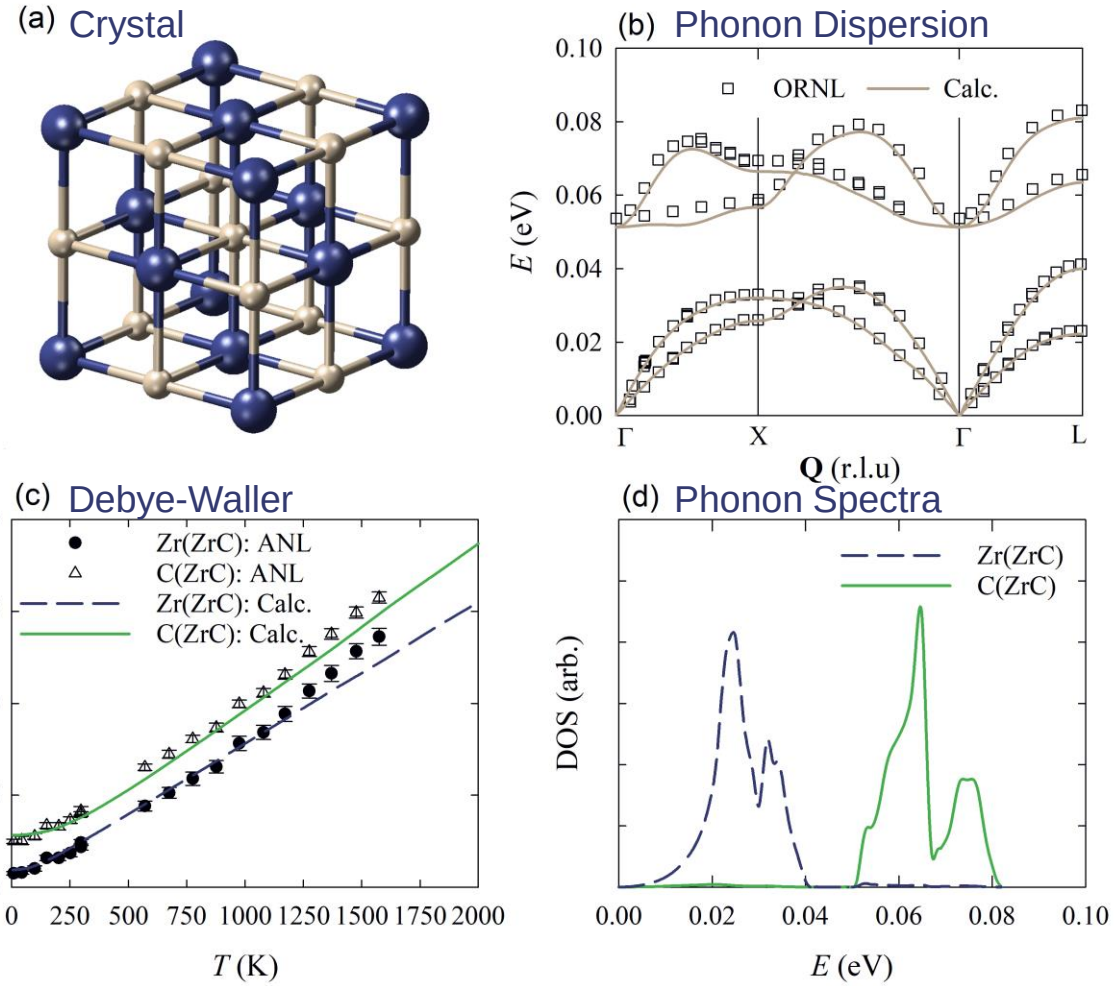
- Zirconium Carbide (ZrC) is a refractory material currently investigated for high temperature reactor applications
  - Fission product barrier in advanced TRISO
  - Corrosion barrier in space nuclear thermal propulsion (SNTP)
- ZrC studied as base of (U,Zr)C solid solution and UC_2 composite fuels for NERVA-type cores during Rover program
  - Testing of material performed at Nuclear Furnace 1 (NF-1) located at Los Alamos
- Thermal Scattering Law (TSL) evaluations generated with FLASSH as new contribution to ENDF/B-VIII.1
  - Standard incoherent approximation
  - New mixed elastic scattering format
  - Disordered alloy theory introduced to coherent elastic scattering to capture natural isotopic abundances

TSL Evaluation Methods



Zirconium Carbide Phonon Model Validation

- Crystal structure lattice parameter in agreement with experiment.
  - 0.471 nm vs. 0.470 nm
- Phonon dispersions relations in good agreement with experiment
  - Differential validation of phonon spectra
- Debye-Waller coefficient in reasonable agreement with experiment from 0 K – 1600 K
  - Integral validation of phonon spectrum
  - *W* is a measure of mean square displacement used directly in TSL
- C(ZrC) spectra localized around higher energy modes separated from lower energy modes
  - Similar behavior in metal-hydrides introduce regular structure $S(\alpha, \beta)$ (quantum oscillations) that strongly influence thermalization



Thermal Neutron Scattering Formalism

- TSL contains distinct (interference) and self (non-interference) components
  - Momentum transfer (α) and energy transfer (β) represented with unitless parameters

$$S(\alpha,\beta) = S_d(\alpha,\beta) + S_s(\alpha,\beta) \qquad \alpha = \frac{E + E' - \mu\sqrt{EE'}}{Ak_BT} \qquad \alpha = \frac{E' - E}{k_BT}$$

- Phonon expansion used in incoherent approximation for inelastic scattering ($p > 0$)

$$S_s(\alpha,\beta) = \sum_p S_s^p(\alpha,\beta)$$

- C(ZrC) and Zr(ZrC) both include: coherent elastic, incoherent elastic, and inelastic
  - ^{91}Zr and ^{13}C have small incoherent nuclear potential

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi k_BT}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S^0(\alpha,\beta) + \sigma_{inc}S_s^0(\alpha,\beta) + (\sigma_{coh} + \sigma_{inc})\sum_{p>0} S_s^p(\alpha,\beta)\right]$$

  - Disordered alloy theory invoked to treat coherent elastic

$$\sigma_{coh,el}(E) = \frac{1}{E}\sum_{E_I<E}\frac{1}{G_I}|F(\boldsymbol{G}_I)|^2$$

Past Method: $$F(\boldsymbol{G}_I) = \sum_\mu \sqrt{\langle\sigma_{coh,\mu}\rangle}\exp(i\boldsymbol{G}_I\cdot\boldsymbol{R}_\mu)\exp(-\langle W_\mu\rangle G_I^2)$$

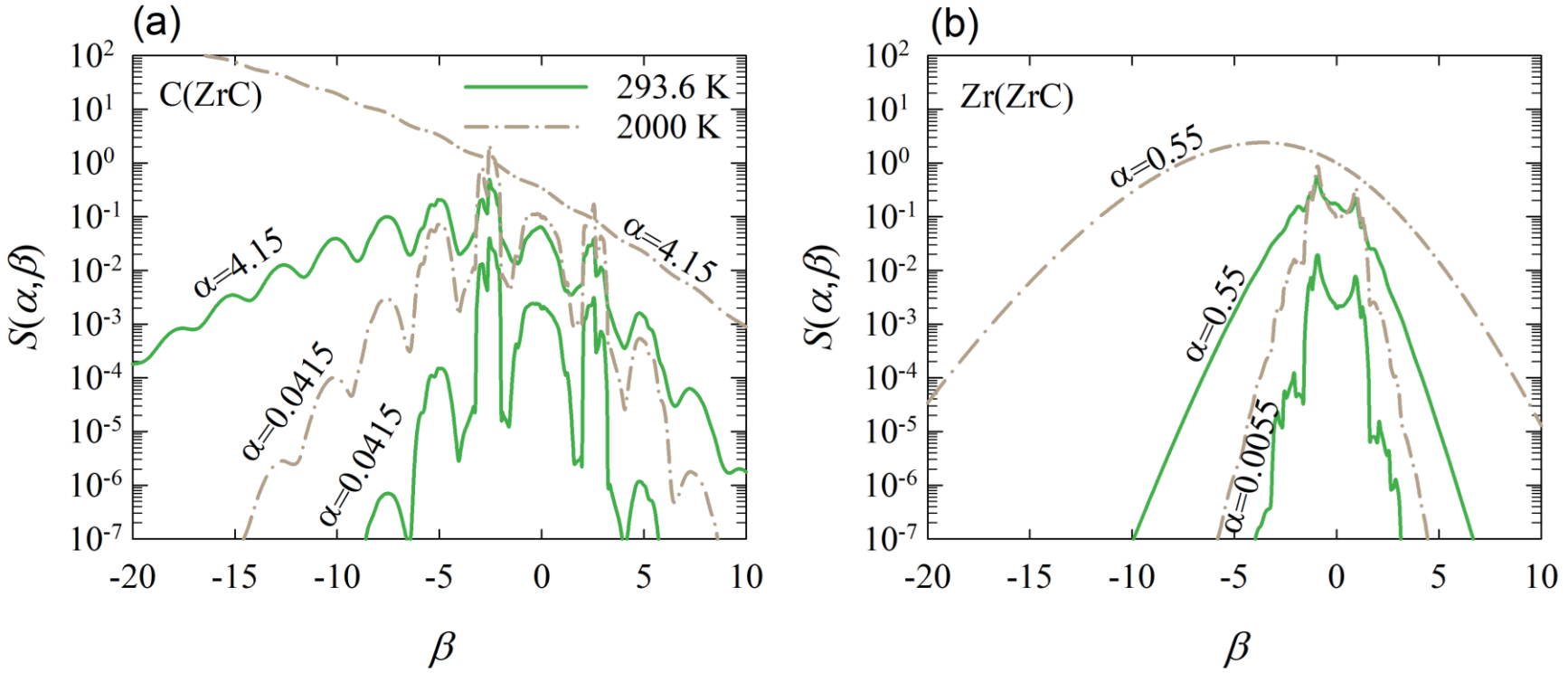
Disordered Alloy: $$F(\boldsymbol{G}_I) = \sum_{\mu,j} f_j\,\bar{b}_{\mu,j}\exp(i\boldsymbol{G}_I\cdot\boldsymbol{R}_\mu)\exp(-W_{\mu,j}G_I^2)$$

ZrC TSL Evaluation Parameters

- C and Zr potential scattering cross sections extracted from ENDF/B-VIII.0 nuclide evaluations at incident neutron energy of 0.0253 eV
  - Incoherent cross section from V. F. Sears, *Neutron News*, **29**, (1992) 26-37
  - Isotopic average cross section for incoherent elastic and inelastic scattering
  - Isotopic average scattering length for coherent elastic scattering
- C(ZrC) and Zr(ZrC) evaluated at 13 temperature:
  - 77 K, 293.6 K, 400 K, 500 K, 600 K, 700 K, 800 K, 1000 K, 1200 K, 1400 K, 1600 K, 1800 K, 2000 K
- Phonon expansion order of 300 used for convergence of $S(\alpha,\beta)$
  - Eliminate the need for short collision-time-approximation in FLASSH
- (α,β) grid valid to 5 eV incident neutron energy
  - Default automatic grid in FLASSH
  - α-grid of 200 points $\left[\frac{0.415322}{A},\frac{790.5139}{A}\right]$
  - β-grid of 283 points $[0,197.63]$

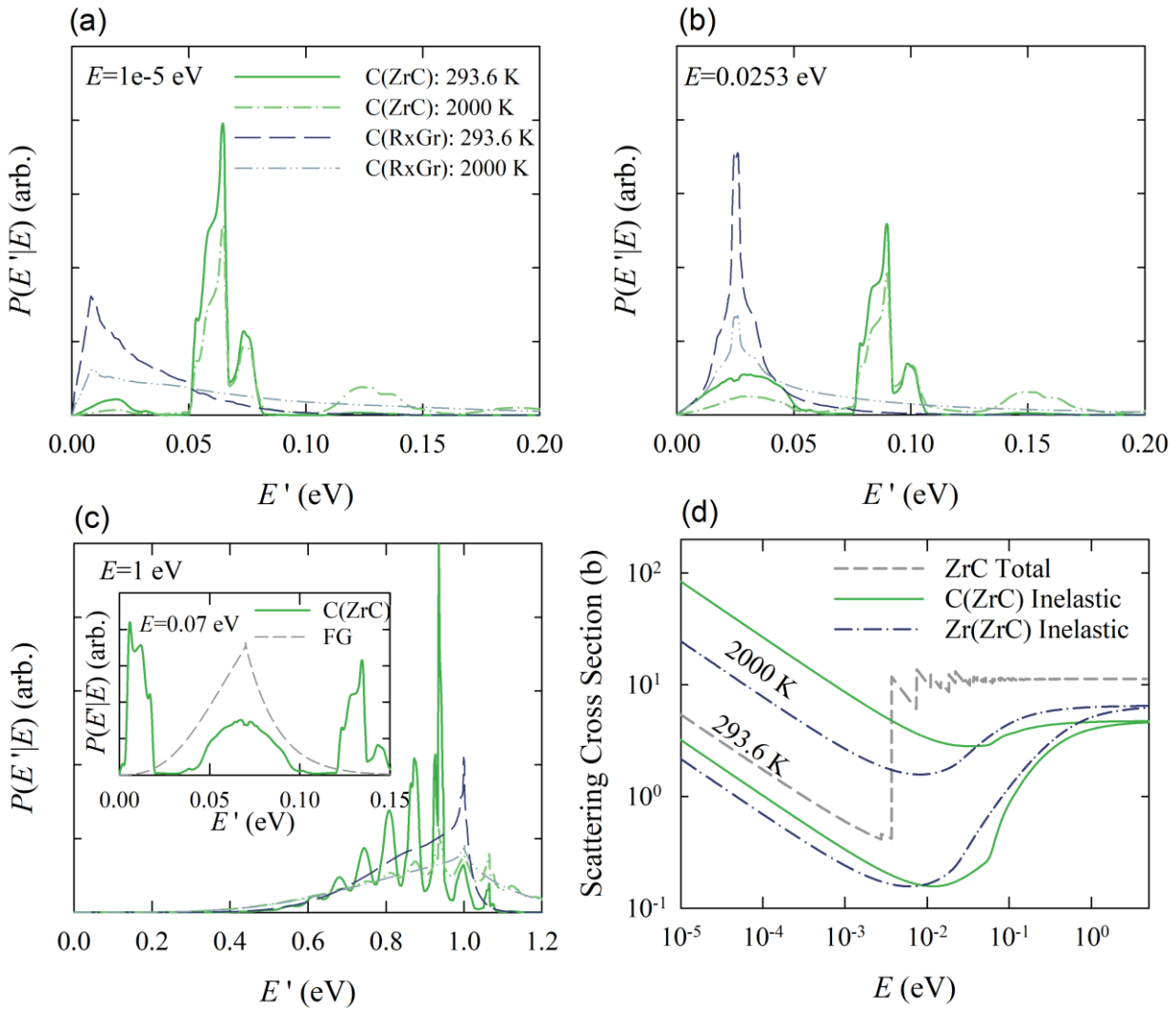
ZrC TSL Evaluation

- Zr(ZrC) TSLs characteristic of heavy elements in crystalline compounds
  - Structure up to 1-phonon energy transfer and relatively featureless beyond
  - Approximating free-gas (FG) for high momentum transfer
- C(ZrC) has strong quantum oscillator effect to multiple phonon orders
  - Oscillations in TSL correspond to phonon order
  - Energy transfer involves quantized exchange of higher energy phonons with substantial downscattering and upscattering
    - Differs from H(ZrH_x) where upscattering is unlikely at room temperature
  - Shallow oscillations persist as FG behavior present for large momentum and energy



C(ZrC) Secondary Neutron Distributions

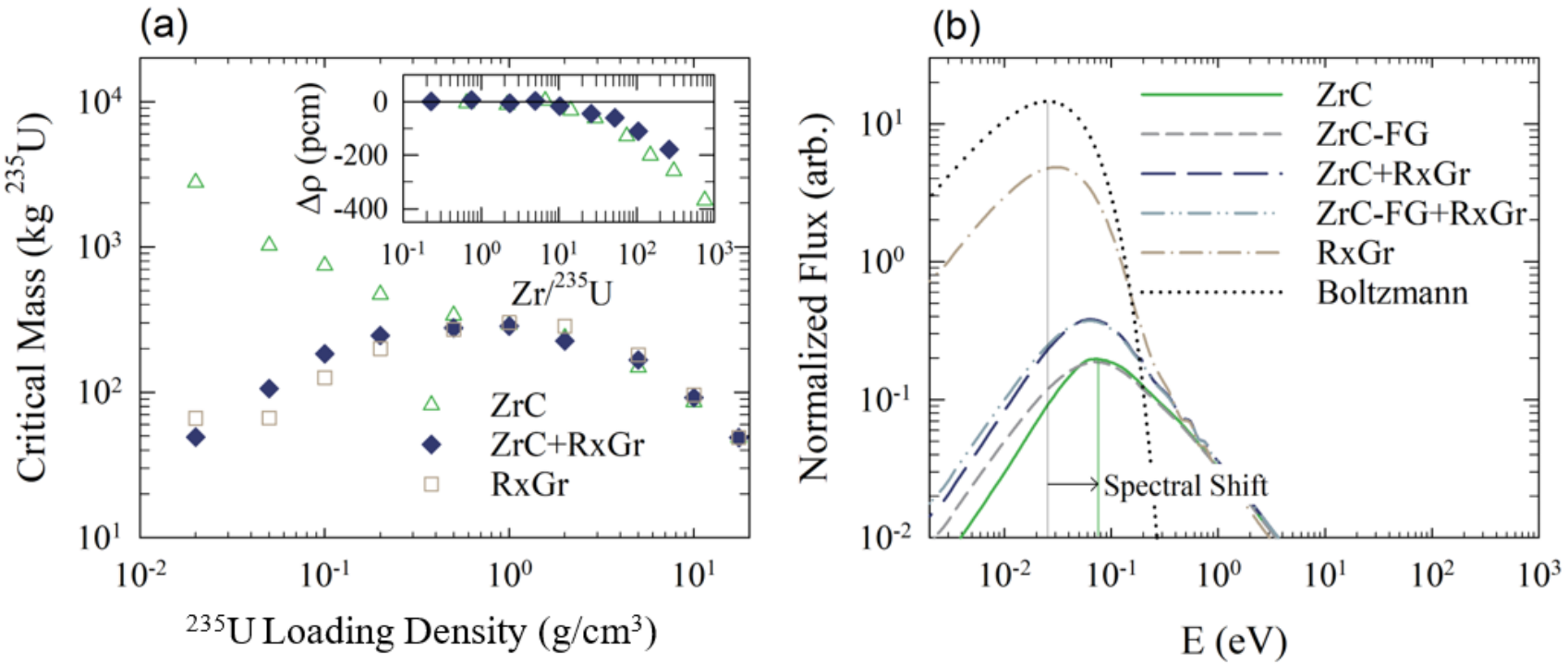
- C(ZrC) is compared to reactor-grade graphite (RxGr)
  - RxGr represents conventional TSL for chemical binding of C
- Epithermal range: ZrC down-scattering is quantized and greater than RxGr
  - Behaviors more similar at higher temperature but some quantization remains
- Neutrons in the phonon energy range have unusually probable high energy losses
  - Near complete energy loss
    - Atypical of non-hydrogenous solids
  - Improbable for FG or typical carbon compounds
- Thermal range: Quantized energy loss is forbidden in thermal energy range and below
  - Up-scattering is dominant
    - Spectral hardening is expected
  - Increase probability of quantized upscattering with temperature
    - Consistent with metal hydrides



*(a)-(c) secondary spectra at labeled energy; (d) integrated cross section at labeled temperatures

Critical Mass Calculations

- MC21 critical mass calculations performed for bare sphere configurations with homogeneous mixtures:
  - ZrC + HEU
  - ZrC + RxGr + HEU (approximation of NF-1)
  - RxGr + HEU
- Neutron multiplication sensitive to ZrC for ^{235}U loading below 0.2 g/cm^3 (Zr/^{235}U > 10)
  - Criticality in thermal drive systems (Zr/^{235}U > 100) substantially differs between TSL and FG
- ZrC introduces spectral shift to thermal spectrum (e.g., ^{235}U loading = 0.02 g/cm^3)
  - Zr absorption has a base impact
  - Quantum oscillator effect hardens spectrum compared to FG treatment
  - For ZrC as secondary moderator, primary moderator (e.g., RxGr) may soften spectrum



Conclusions

- New TSL evaluations for ZrC have been generated for contribution to ENDF/B-VIII.1
  - Support criticality safety analyses for high temperature reactor systems
- New evaluations extend physics of elastic scattering used in ENDF evaluations
  - Use of mixed elastic scattering
  - Introduction of disordered alloy theory to improve coherent elastic treatment
- C(ZrC) exhibits quantum oscillator behavior that is atypical of non-hydrogenous compounds
  - May be important for evaluation of systems with significant ZrC content
    - TRISO coatings
    - SNTP applications
  - Initial critical mass calculations demonstrate a C(ZrC) impact compared to FG
  - Further effects testing is in progress