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Predictions of novel features in x-ray scattering spectra for thermometry

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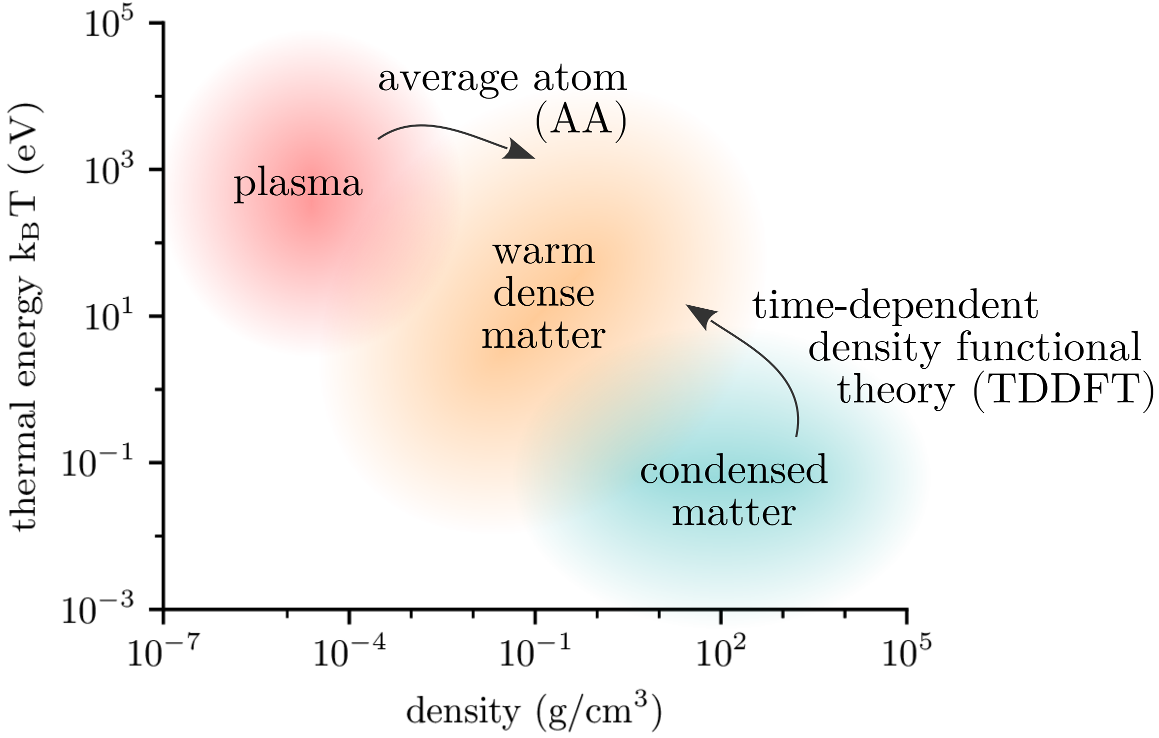
Bridging the Gap between Plasma and Condensed Matter

- Fusion fuel passes through poorly understood WDM regime on the way to ignition
- Competing physics challenges models
- Scarce experimental data, uncertain conditions limit model validation
- Validate modified AA against more accurate but expensive TDDFT
- Predict potential improvements for x-ray diagnostic techniques



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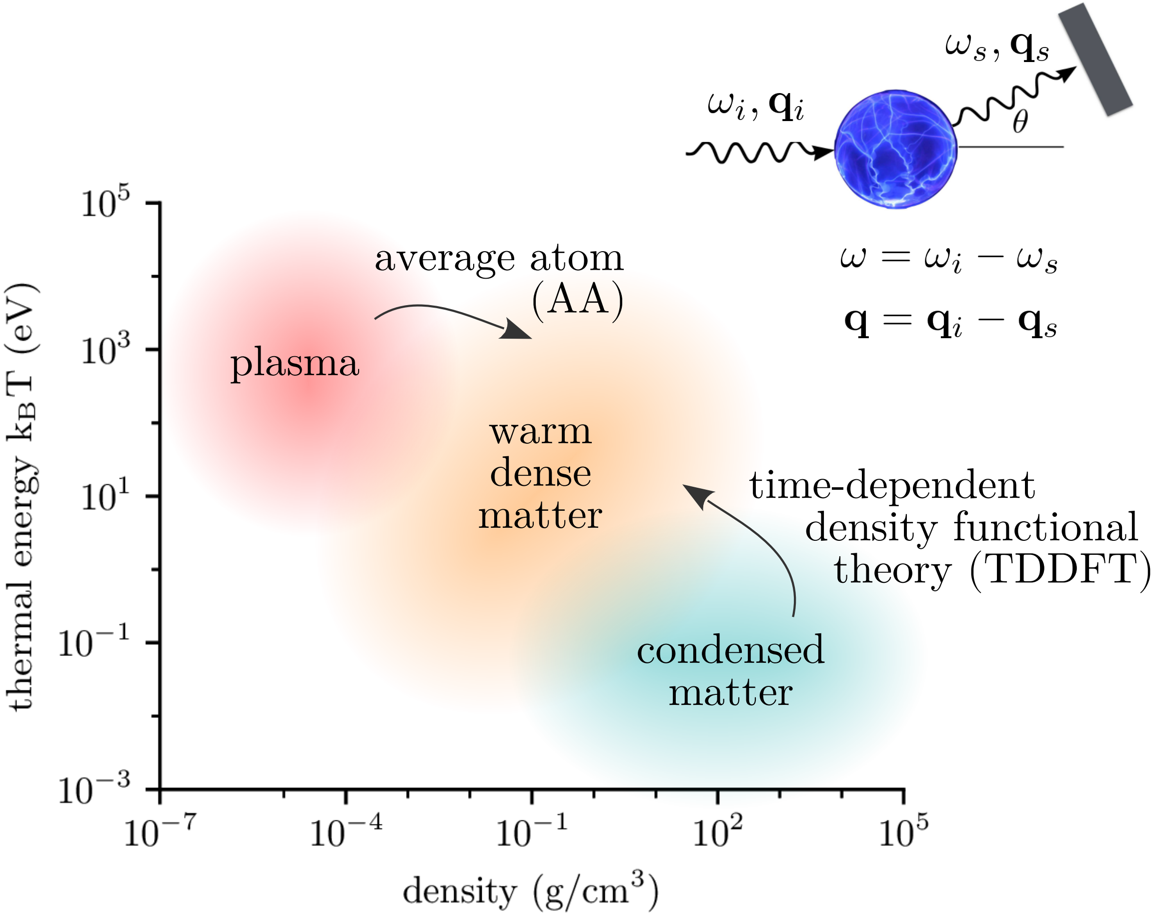


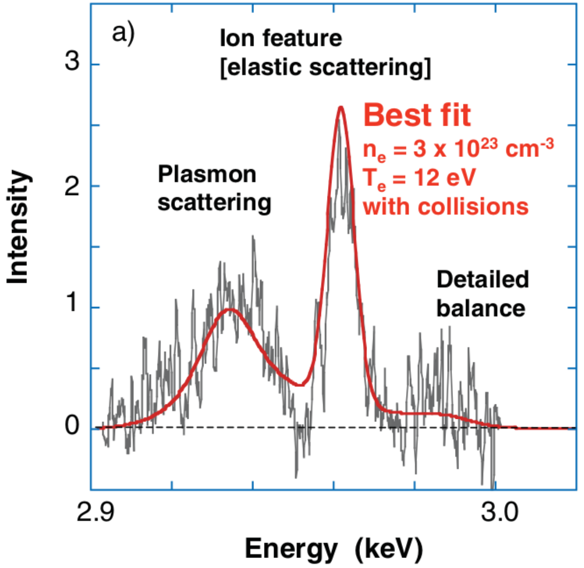
Diagnosing Temperatures with X-ray Scattering

Electron temperature:

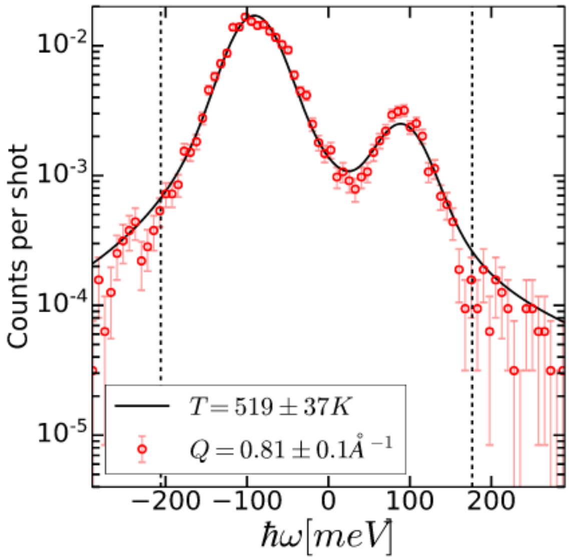
- detailed balance between ~10 eV red- and blue-shifted plasmon features
- sensitivity limited to T ~ 10 eV
- alternative: bound-bound features

Ion temperature:

- detailed balance between ~100 meV red- and blue- shifted phonon features
- detector limitations challenging
- complement: band structure effects



$$\frac{S(\mathbf{q},\omega)}{S(\mathbf{q},-\omega)} = e^{\hbar\omega/(k_B T)}$$



Glenzer et al., Phys. Rev. Lett. 98 (2007)

Descamps et al., Sci. Rep. 10 (2020)

Simulating X-ray Scattering with Time-Dependent DFT

- Initial condition: equilibrium state from Mermin-DFT
- Evolve response to probe in real time

$$i\frac{\partial}{\partial t}\phi_j(\mathbf{r},t) = \hat{H}[n(\mathbf{r},t)]\phi_j(\mathbf{r},t)$$

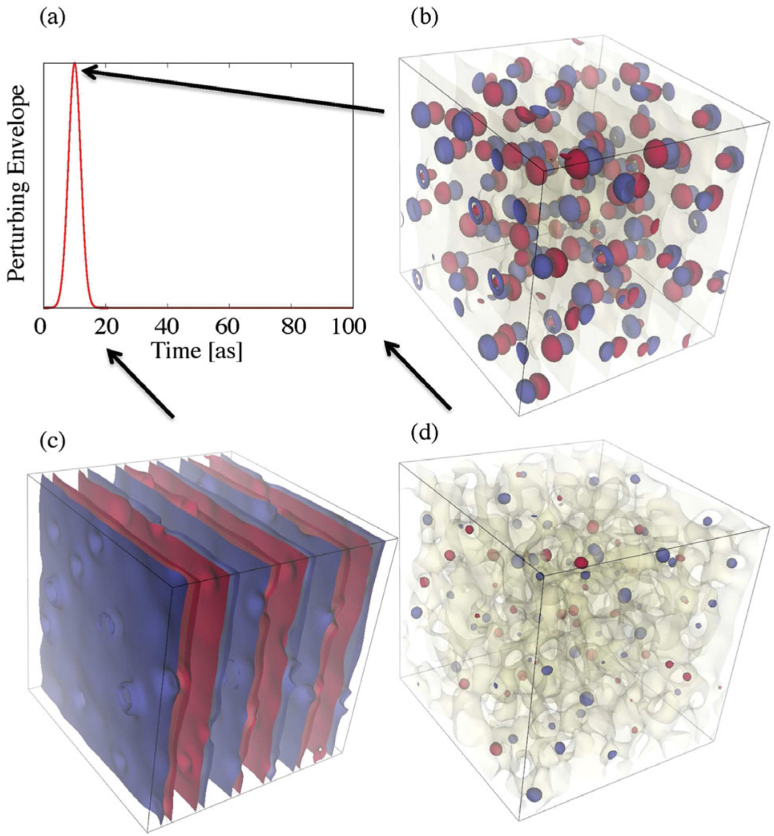
$$n(\mathbf{r},t) = \sum_j f_j(T)|\phi_j(\mathbf{r},t)|^2$$

$$V_{\text{probe}}(\mathbf{r},t) = V_0\, e^{i\mathbf{q}\cdot\mathbf{r}} f(t)$$

- Dynamic structure factor (DSF) related to Fourier transform of density response

$$\tilde{\chi}(\mathbf{q},-\mathbf{q},\omega) = \delta\tilde{n}(\mathbf{q},\omega)/(V_0\tilde{f}(\omega))$$

$$S(\mathbf{q},\omega) = -\frac{1}{\pi}\frac{\text{Im}\left[\tilde{\chi}(\mathbf{q},-\mathbf{q},\omega)\right]}{1-e^{-\omega/(k_B T)}}$$



Baczewski et al., Phys. Rev. Lett. 116 (2016)

Simulating X-ray Scattering with Average Atom

- Chihara decomposition: $S(q,\omega) = S_{ii}(q,\omega) + S_{FF}(q,\omega) + S_{BF}(q,\omega) + S_{BB}(q,\omega)$

- Modified S_{FF} captures non-Drude behavior
  - Mermin dielectric function
  - non-ideal, quantum DOS
  - T-matrix elastic + inelastic collisions

$$S_{FF}(q,\omega) = -\frac{1}{1-e^{-\omega/(k_B T)}}\frac{q^2}{4\pi n_e}\mathrm{Im}\left[-\frac{1}{\varepsilon(q,\omega)}\right]$$

$$\epsilon_{\mathrm{MA}}(k,\omega) = 1 + \frac{(\omega+i\nu)[\epsilon_{\mathrm{RPA}}(k,\omega+i\nu)-1]}{\omega + i\nu\frac{\epsilon_{\mathrm{RPA}}(k,\omega+i\nu)-1}{\epsilon_{\mathrm{RPA}}(k,0)-1}}$$

- Introduce bound-bound S_{BB} to achieve continuity under ionization of bound states

$$S_{BF}(q,\omega) = \sum_i g_i\, f(\varepsilon_i)(1-f(\varepsilon_i+\omega))\,\mathcal{M}_i^{BF}(q,\omega)$$

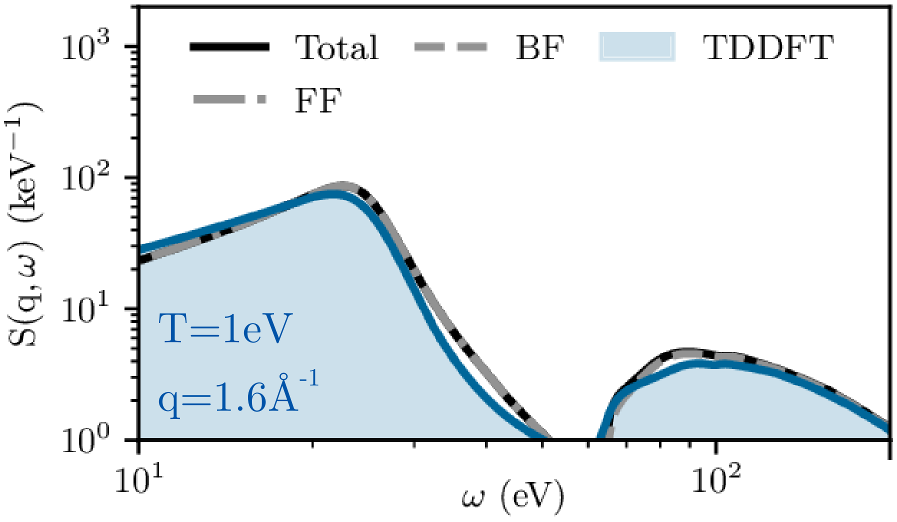
$$S_{BB}(q,\omega) = \sum_{i,f} g_i\, f(\varepsilon_i)(1-f(\varepsilon_f))\,\phi(\omega)\,\mathcal{M}_{if}^{BB}(q,\omega)$$

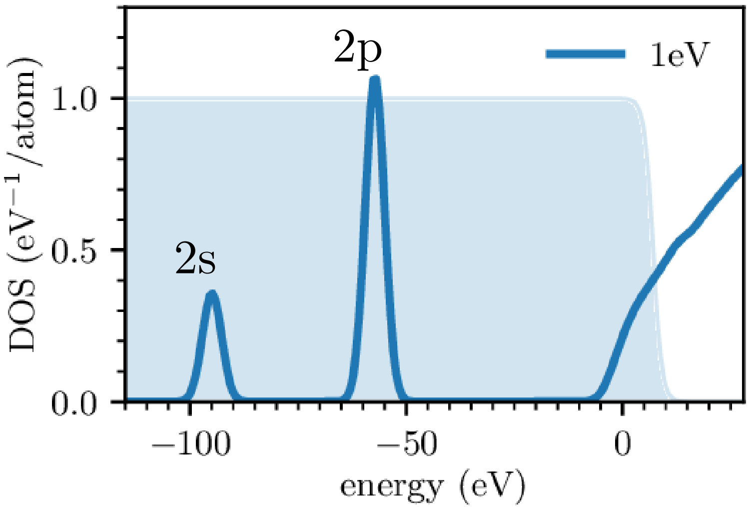
sum over initial and final bound states

line shape

Bound-Bound Transitions in Aluminum

arXiv:2109.09576





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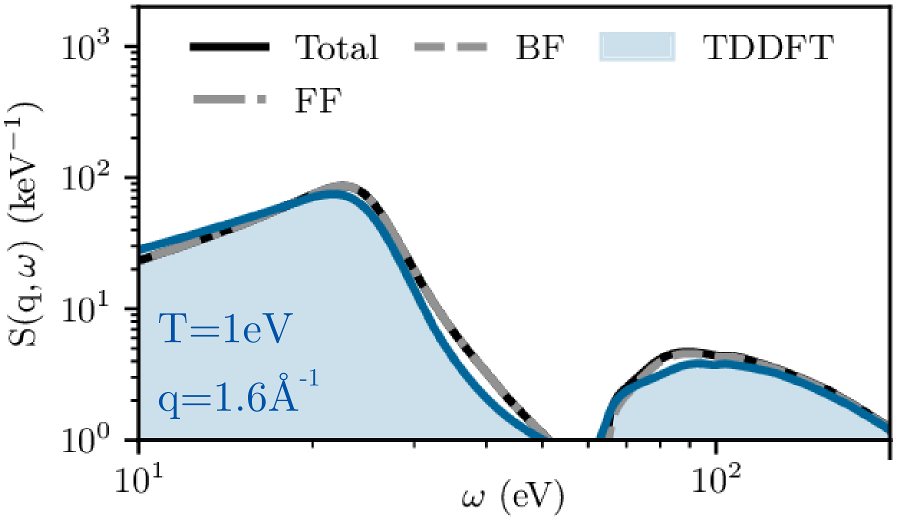
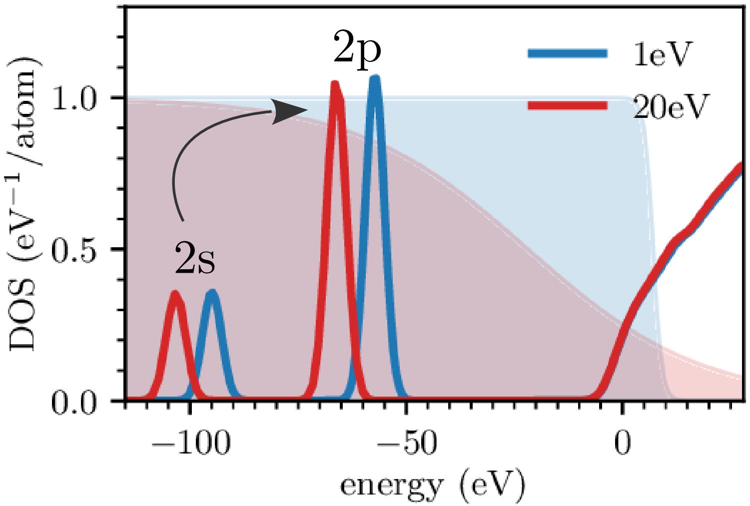
- Thermal depletion of 2p states for T $\gtrsim$ 20eV





Bound-Bound Transitions in Aluminum

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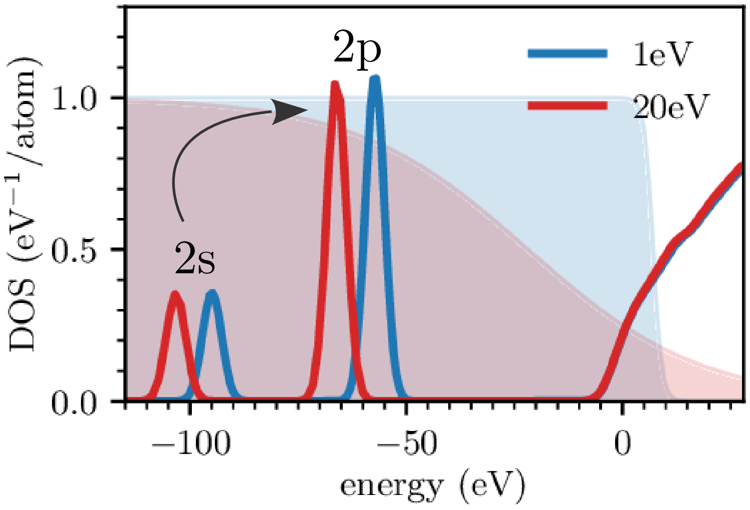
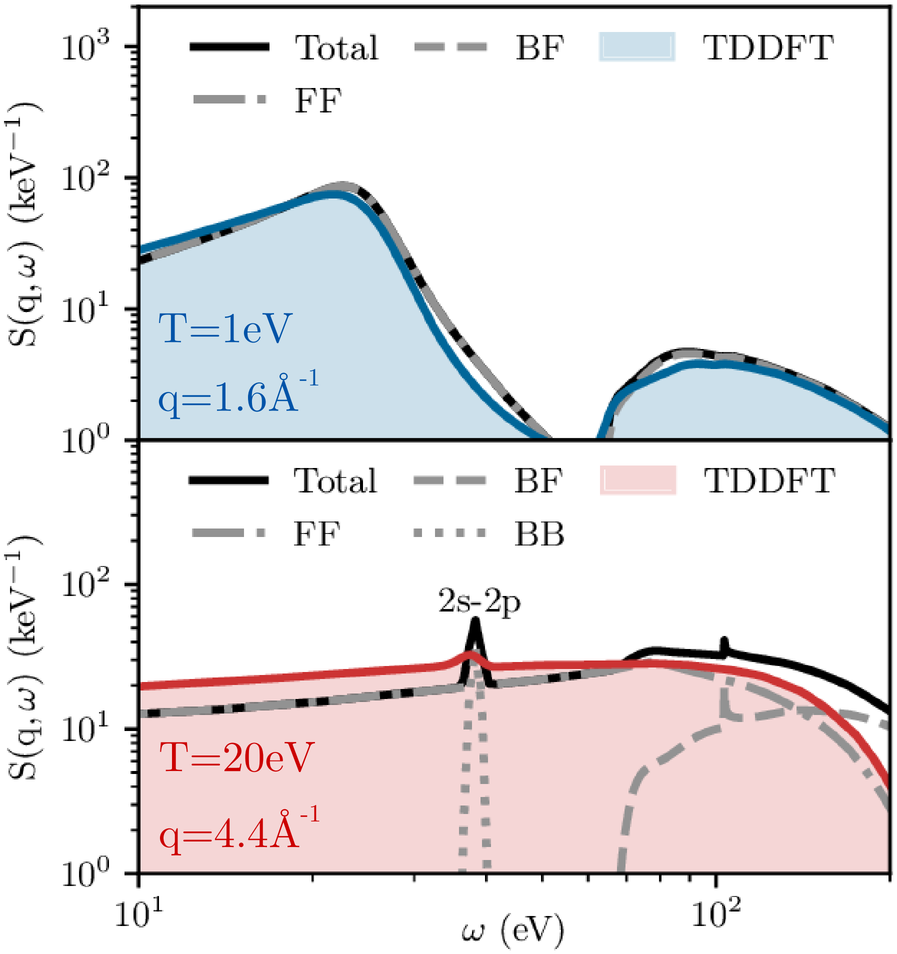
- Thermal depletion of 2p states for T $\gtrsim$ 20eV
- Prominent 2s $\rightarrow$ 2p feature at ~40eV in DSF
  - particularly towards higher angles
  - very sensitive to T via 2p occupation

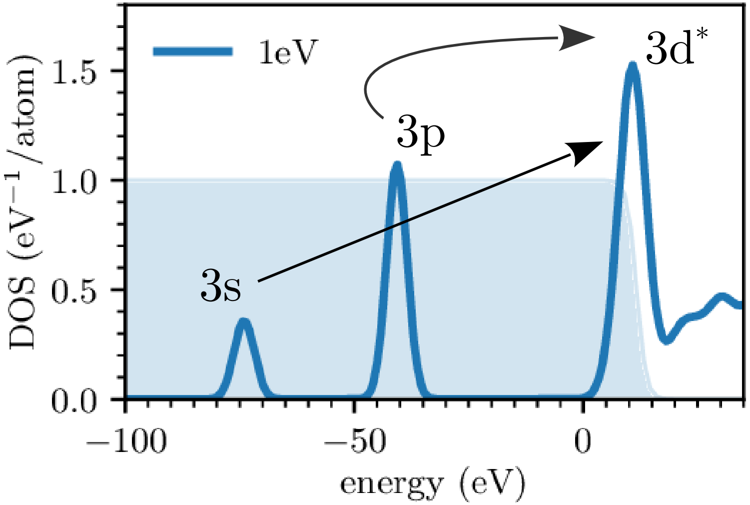


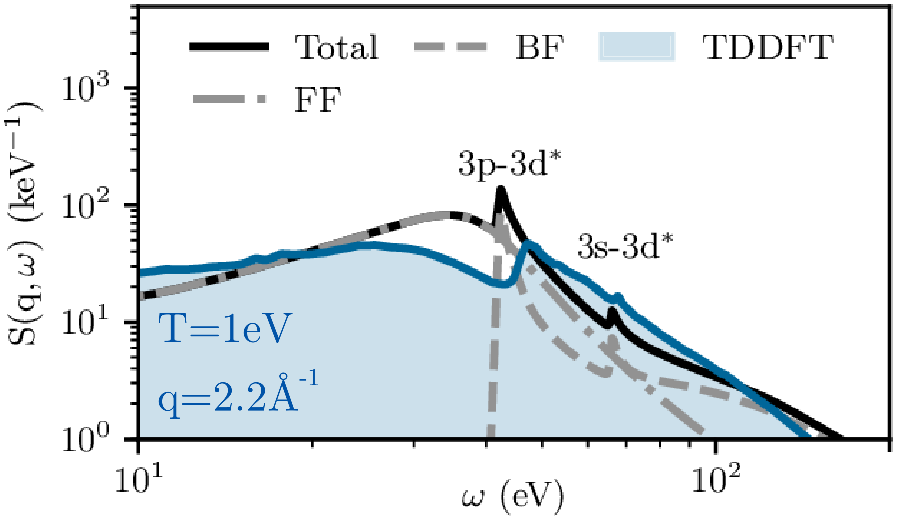


Bound-Bound Transitions in Iron

arXiv:2109.09576

- Richer set of transitions:
  - $3p \rightarrow 3d$ at ~55eV
  - $3s \rightarrow 3d$ at ~85eV





Bound-Bound Transitions in Iron

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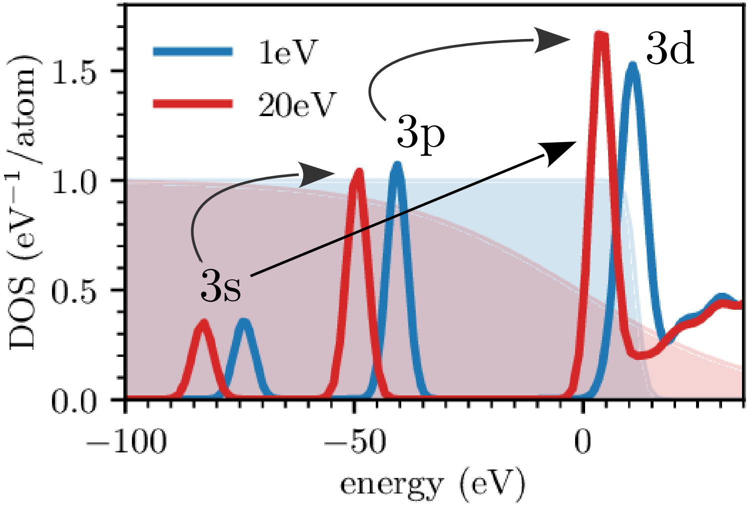
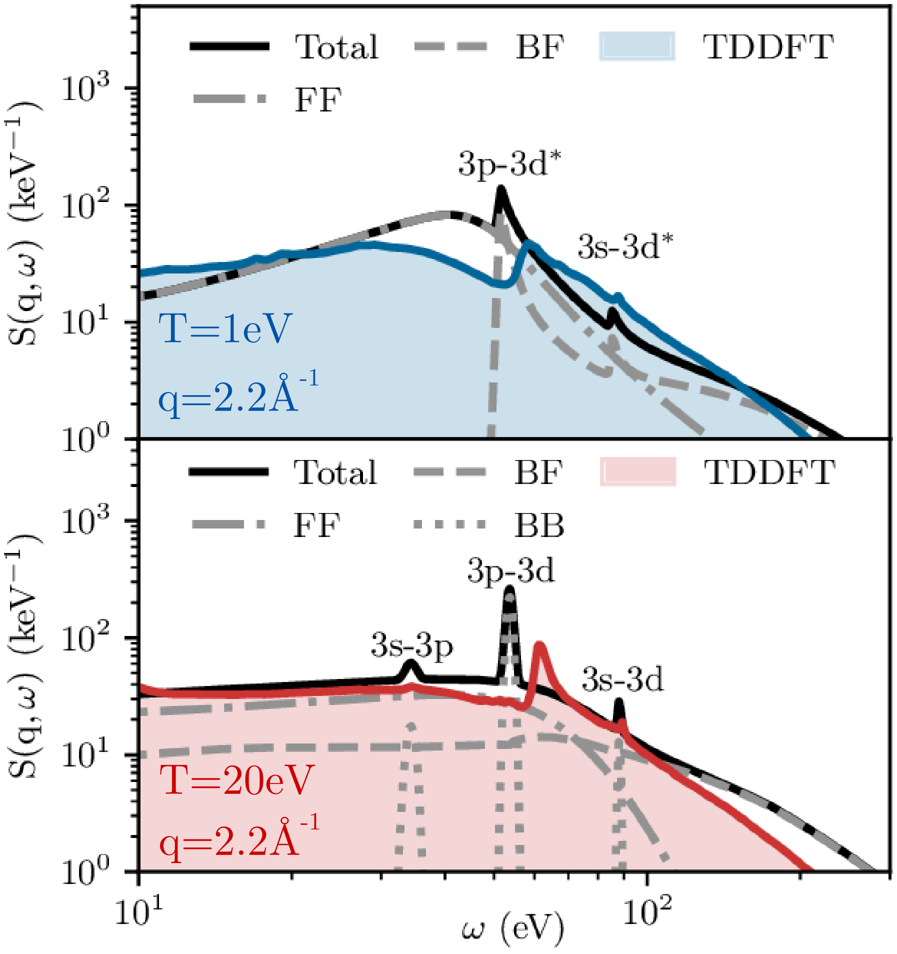
- Richer set of transitions:
  - 3p → 3d at ~55eV
  - 3s → 3d at ~85eV
  - 3s → 3p at ~35eV
- Apparent discrepancy in 3p-3d energy!
  - ~5eV higher in TDDFT than AA

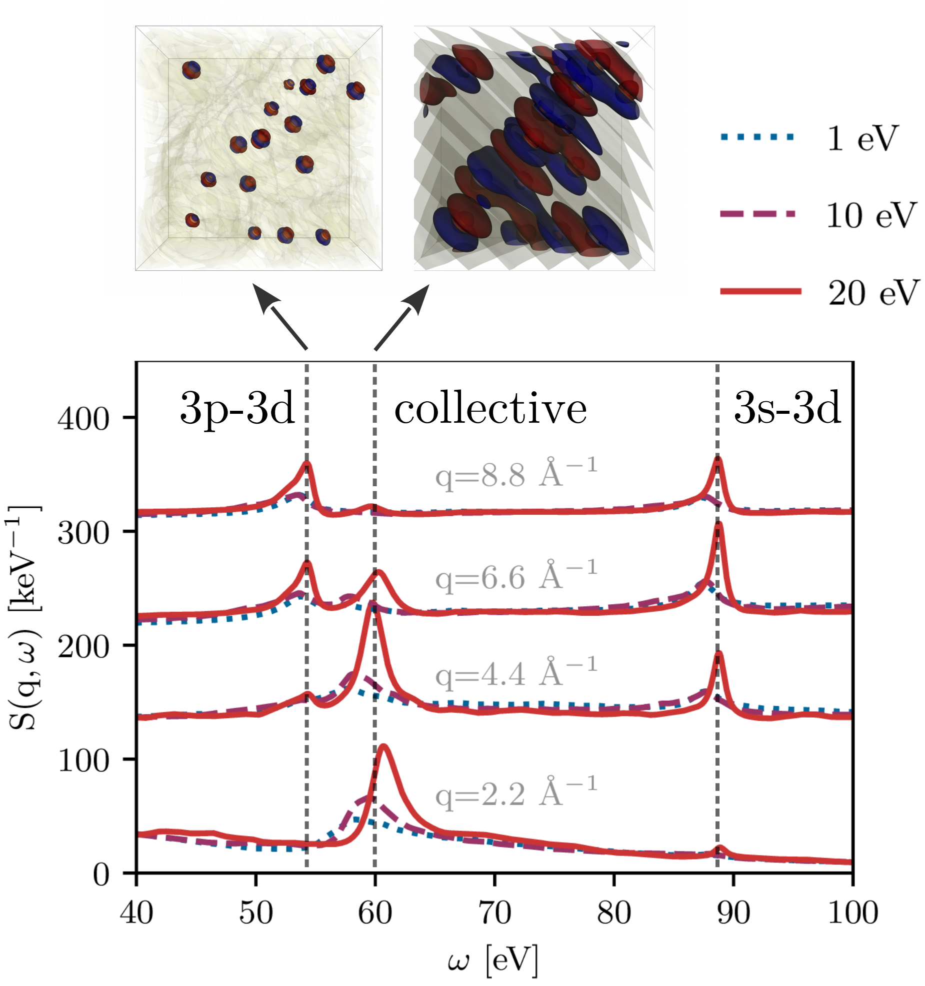




New Collective Feature in Iron from TDDFT

arXiv:2109.09576

- For high q:
  - 3p-3d feature appears at expected energy
  - density response localized
- For low q:
  - density response delocalized
  - higher-energy feature arises from new collective mechanism!



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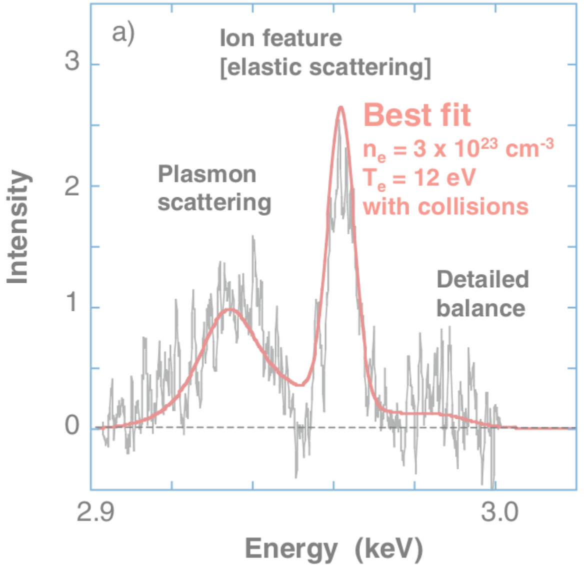

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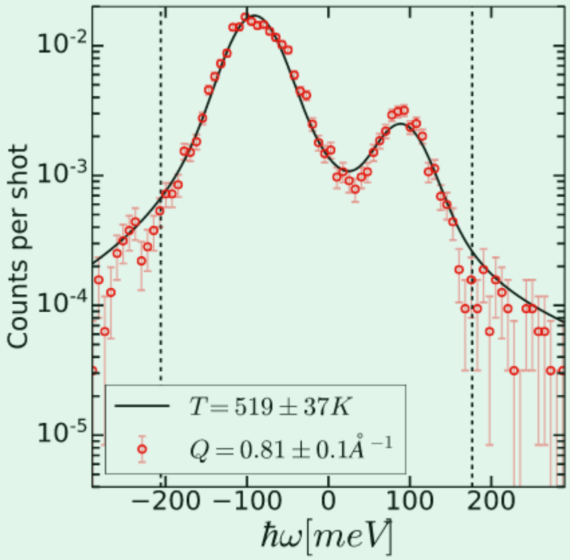
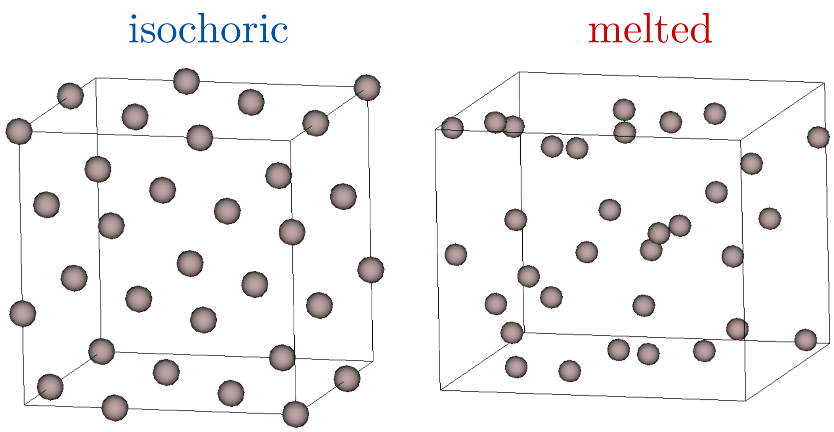
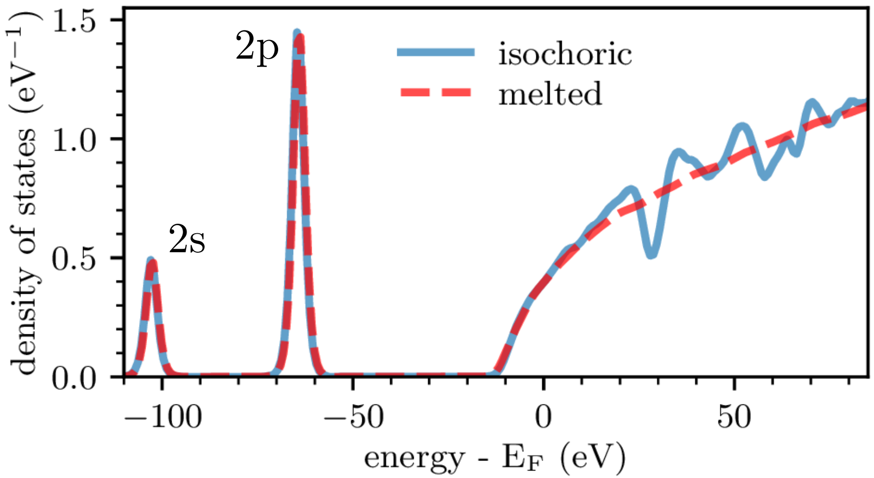


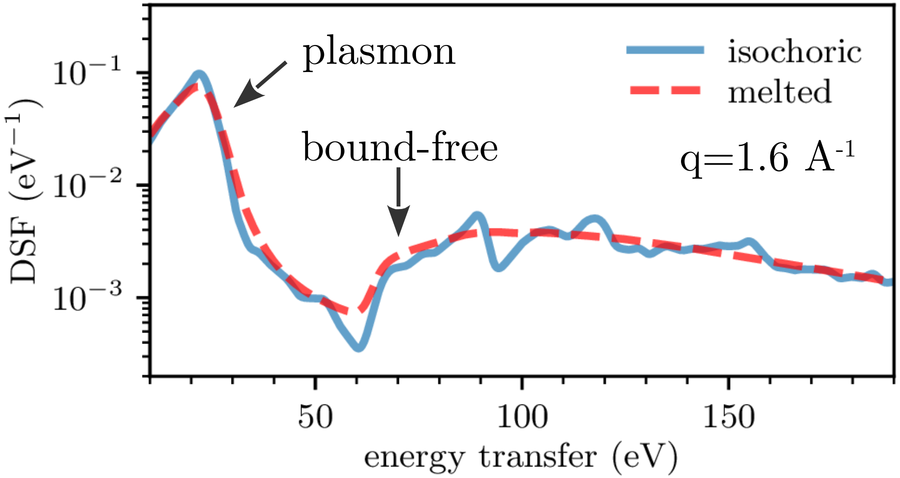
Descamps et al., Sci. Rep. 10 (2020)

Band Structure Effects out of Equilibrium

- Melted Al (T_i=T_e=1eV)
    - ~ideal free-electron gas
- Isochorically heated Al (T_i=0, T_e=1eV)
    - gaps among high-energy bands
- DSF reflects details of band structure
- May help simultaneously infer T_e and $T_i < T_{melt}$!

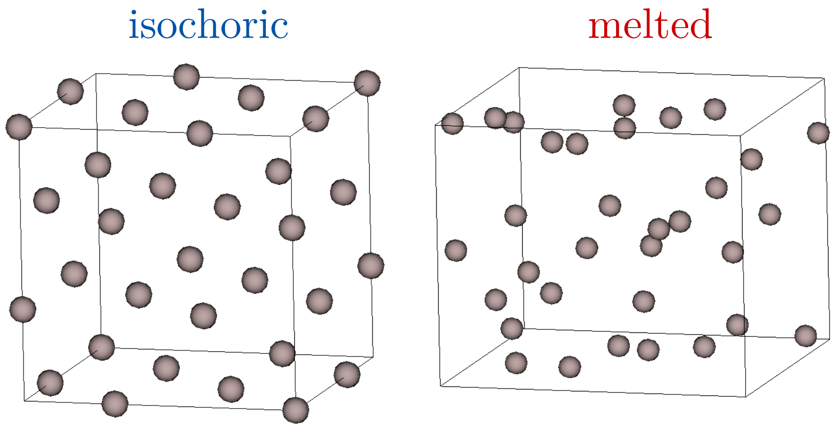


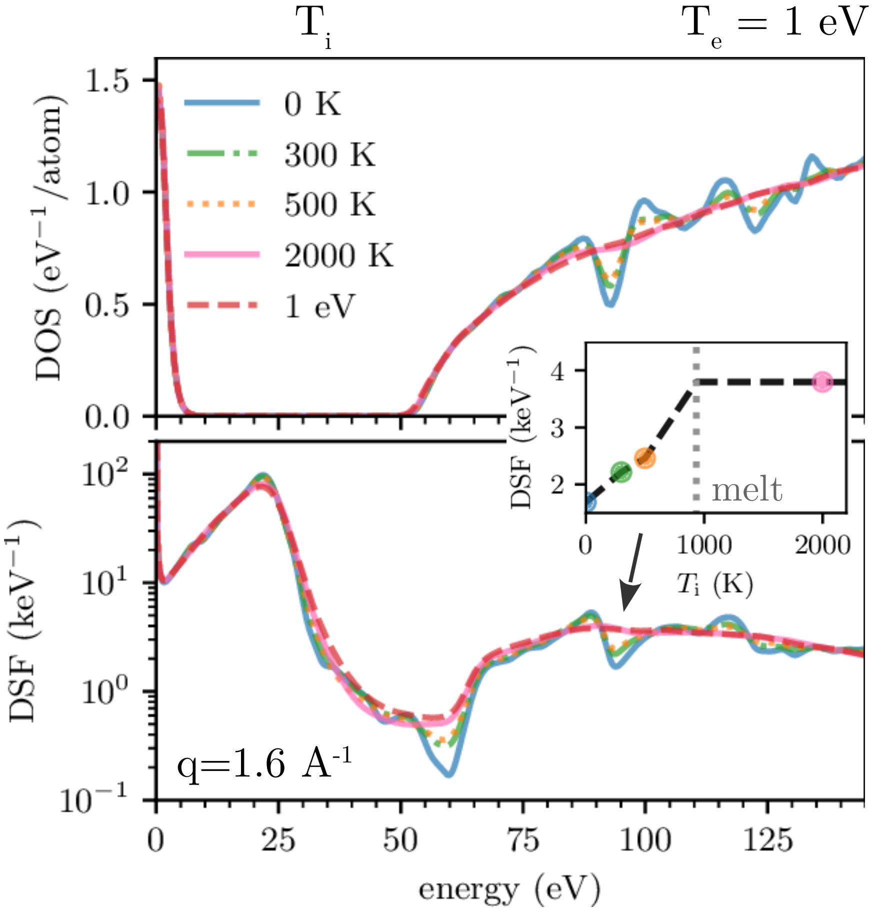




Band Structure Effects

- Lower amplitude features with increasing T_i
- T-sensitivity appears limited
  - may improve at higher scattering angles





Band Structure Effects

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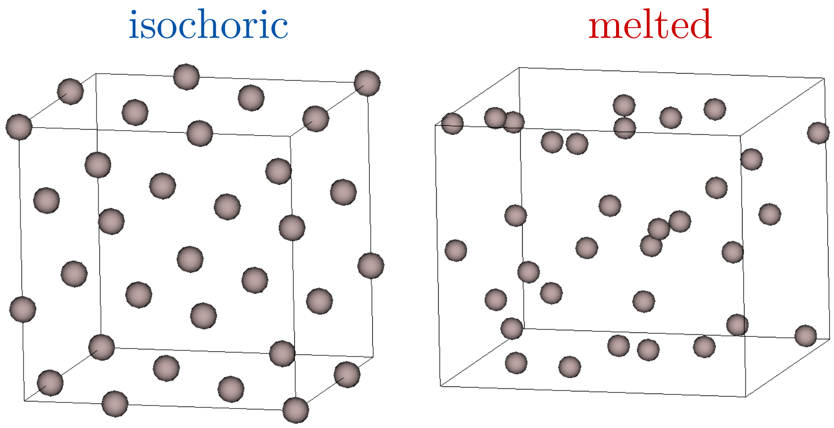
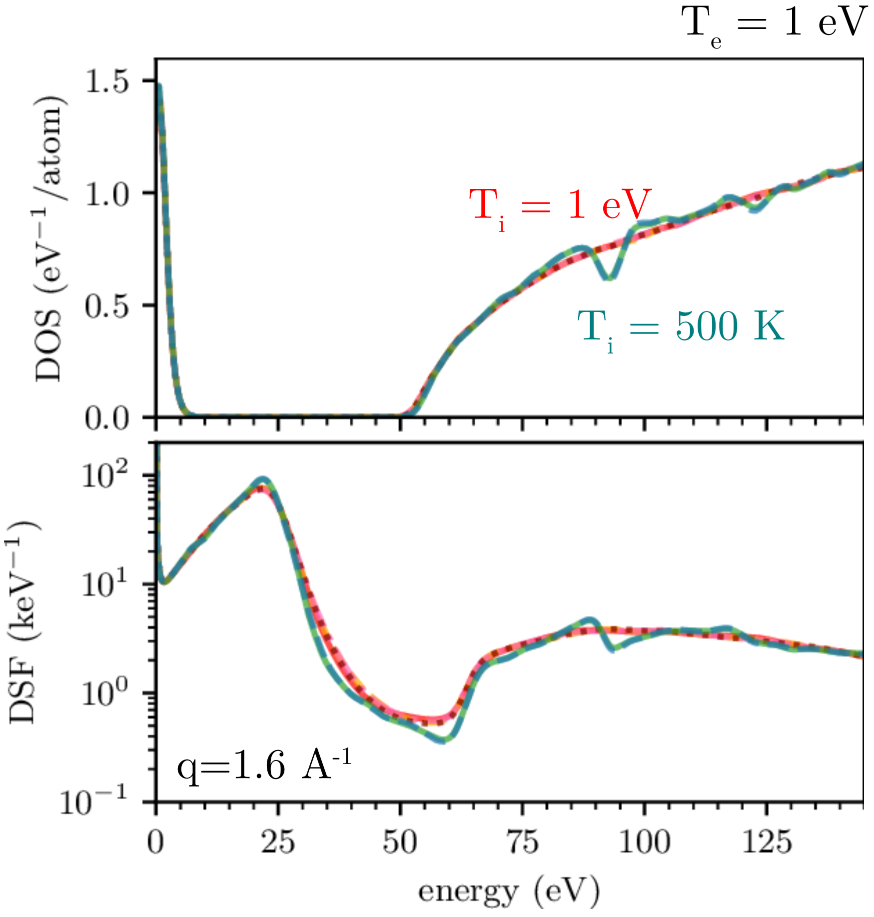
  - may improve at higher scattering angles
- Results insensitive to atomic configuration at given T





Conclusions

- Predict prominent bound-bound features in DSFs at high T
  - promising alternative to plasmon-based thermometry for $k_B T_e > \hbar\omega_p$
  - novel collective behavior in iron's 3p – 3d transition
- Predict subtle band structure features in DSFs of isochorically heated materials
  - promising alternative to phonon-based thermometry for $T_i < T_{melt}$
  - possibilities for simultaneously constraining T_e and T_i
  - promising for studies of ultrafast melting