Modelling the electronic excitation induced structural dynamics of tungsten

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Overview

• Background/Motivation

• High temperature DFT calculations
  • Soft phonon modes
  • Solid to solid phase transitions

• Electronic temperature dependent interatomic potentials

• Large scale MD simulations
  • Laser irradiation of W thin films
Motivation: *To include the effects of excited electrons in radiation damage simulations*

Radiation that interacts primarily with nuclei:
- Neutron irradiation
- Low energy ion irradiation

Radiation that interacts primarily with nuclei:
- Swift heavy ion irradiation
- Laser irradiation
2T-MD

- 2T model with thermal diffusion equation of lattice replaced by MD
- Lattice is thermostatted to the local electronic temperature

\[ m_i \frac{\partial v_i}{\partial t} = F_i - \gamma v_i + \tilde{F} \]

\[ C_e \frac{\partial T_e}{\partial t} = \nabla \kappa \nabla T_e - g_p (T_e - T_a) \]

- Thermal diffusion equation solved by FD technique
- Energy is exchanged between lattice and electrons at each MD step
- *Temporal and spatial* \( T_e \) *evolution is known throughout simulation*
2T-MD - Cascades

50 keV Fe cascades

- residual defect number very sensitive treatment of electronic loss
2T-MD – Laser irradiation

• Successful modelling/experimental project on Au nanofilms
  • Ultrafast electron diffraction (UED) of laser irradiated single crystal 10 nm gold films
  • 2T-MD modelling on same length and time scales
  • Exceptional agreement between calculated and measured time evolution of the Bragg peak intensities
  • **No adjustable parameters**

• Follow-on project on W
  • Calculated and measured e-p coupling constant $1.65 \times 10^{17}$ vs $1.4(3) \times 10^{17}$ W m$^{-3}$ K$^{-1}$
  • Noted soft modes at high electronic temperature

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W Phonon dispersion – high Te

**bcc**

\[ Te = 300K ; 20000K \]

**fcc**

\[ Te = 300K ; 20000K \]

**bcc 300 K – comparison with expt.**

**fcc**

\[ Te = 300K ; 20000K \]
bcc to fcc/hcp

Is there a barrier?

**Tetragonal Bain path**

**Hexagonal path**

**Trigonal Bain path**
Electron density - redistribution

Electron density difference between $T_e = 20,000$ K and $T_e = 300$ K
Solid to solid phase transitions - Summary

- $10000 K < T_e < 15000 K$
  - Strong directional softening of bcc but no instability
  - Free energy barrier for transformation
  - Both fcc and hcp are stable
  - Free energy of fcc and hcp are lower than bcc

- $T_e > 15000 K$
  - bcc structure becomes dynamically unstable
  - No free energy barrier for tetragonal Bain transformation from bcc to fcc or hexagonal transformation from bcc to hcp

- Redistribution of electron density
  - Increase in density along 2nd neighbour bonds
$T_e$ dependent interatomic potentials

• Aims
  • To derive a set of interatomic potentials that reflect the change in the interactions resulting from electronic excitations
  
  • To use the potentials, in conjunction with 2T-MD, to perform large scale simulations of:
    • Laser irradiation
    • Swift heavy ion irradiation
    • Neutron irradiation
$T_e$ dependent potentials - strategy

- Use DFT to calculate free energy of bcc and fcc W as a function of lattice parameter for a range of $T_e$

- EAM potential
  - Extended FS for ground state potential (Dai et al, JPCM 2006)
  - Normalize DFT free energies to give 0 at Dai potential cutoff
  - Subtract Dai repulsive $V(r_{ij})$ energy from DFT energies
  - Convert volume to density using Dai expression for density
  - Use cubic spline to fit embedding term to DFT data (100% bcc or 50% bcc / 50% fcc)

- Summary
  - Repulsive term and form of density unchanged
  - $F(\rho_i) \rightarrow F(\rho_i, T_e)$ - embedding term to fit calculated HTDFT data
$T_e$ dependent potentials - results

Fitted to bcc

Fitted to 50/50 bcc/fcc
2T-MD Simulations with $T_e$ dependent potentials

- Electronic heat conductivity considered as infinite
- $T_e$ dependent $C_e$ and e-p coupling evaluated from \textit{ab initio} calculations
- PES changes included by Te dependent potentials

\[ C_e(T_e) \frac{\partial T_e}{\partial t} = \nabla \cdot (\kappa_e \nabla T_e) - G(T_e) \cdot (T_e - T_i) + S(z, t) \]

\[ m \frac{\partial v_i}{\partial t} = F_i(t) - \gamma_i v_i + \tilde{F}_i(t) \]

\[ \tilde{F}_i(t) = \sqrt{\tau} \tilde{A}_i(t) \]
2T-MD simulation results: 40 mJ cm$^{-2}$ fluence

- 50x50x50 bcc unit cell simulation cell
- Periodic in x and y
- 800 fs laser homogeneous pulse
- Gaussian time distribution

- Blue – bcc
- Grey - disordered
2T-MD simulation results: 40 mJ cm$^{-2}$ fluence
2T-MD Simulations - 80 mJ cm$^{-2}$ fluence

- *Blue* – bcc
- *Green* – fcc
- *Red* - hcp
- *Grey* - disordered

Only showing fcc and hcp

0.4 ps  
0.6 ps
2T-MD Simulations - 80 mJ cm\(^{-2}\) fluence

Artificially low e-p coupling
- Blue – bcc
- Green - fcc
- Grey - disordered

1.2 ps
Summary

- High temperature DFT calculations find:
  - Soft modes in bcc W for Te > 10000 K
    - fcc and hcp both stable
  - Dynamic lattice instabilities for Te > 15000 K
    - No barrier for tetragonal Bain transformation

- Derived $T_e$ dependent potentials by fitting to HTDFT results
- Used to model laser irradiation of thin film W
- Observe bcc to fcc transformation for very low e-p coupling
  - *Non-thermal solid to solid phase transformation*
Discussion questions

- How should we treat electronic energy loss in cascades?

- How high would you expect $T_e$ to be in a plasma facing W?
  - Irradiation with n, e, $\alpha$, tritium
  - $10^4$ K for 300 keV cascades (Eva Zarkadoula et al to be published)

- Does HTDFT give a good description of interactions between excited atoms?
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