Two-temperature (TTM) molecular dynamics

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TTM implementation in LAMMPS
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Cascade damage example
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Electronic effects: two temperature model (TTM)

- Model radiation-excited carrier gas consisting of hot electrons and holes
- Provide a mechanism for energy exchange between radiation-excited carrier gas and atoms
- Conserves energy (electronic and atomic)
- Electron-ion energy transfer based on a Langevin thermostat
- Electronic temperature controlled by a heat diffusion equation

Duffy and Rutherford, *J. Phys. Condens. Matter* **2007**
Rutherford and Duffy, *J. Phys. Condens. Matter* **2007**

TTM theory

Langevin thermostat

$$m\mathbf{a} = F_i(t) - \gamma_i \mathbf{v}_i + \mathbf{F}'(t)$$

- $F_i(t)$: conservative force
- $\gamma_i \mathbf{v}_i$: friction term (energy loss)
- $\mathbf{F}'(t)$: random force (energy loss or gain) (usually a net gain)

Electron stopping effects

$$\gamma_i = \gamma_p + \gamma_s \qquad \text{For } v_i > v_o$$

Friction due to electron-atom interactions

Friction due to electron stopping

$$\gamma_i = \gamma_p \qquad \text{For } v_i \leq v_o$$

γ values are user-specified constants related to relaxation times (τ)

Energy transport in the electronic subsystem via the heat diffusion equation

$$C_e \frac{\partial \mathrm{T_e}}{\partial t} = \nabla(\kappa_e \nabla T_e) - g_p(T_e - T_a) + g_s T_a^{'}$$

- Electron temperature (T_e) varies locally.
- T_e is taken as an average in a spatial cell.
- Heat eqn is discretized and solved numerically.
- User supplies initial T_e values.
- LAMMPS keeps track of time evolution of $T_e$values.
- C_e and κ_e are user-specified constants.
- The T values and “coupling constants” (g_p and g_s) vary spatially and temporally.

Link between the electronic and atomic sub-systems

Energy loss of the atomic sub-system:

$$\Delta U_i = \mathbf{F}_i \cdot \mathbf{v}_i \Delta t = \gamma_i v_i^2 \Delta t \qquad \Delta U_l = \Delta t \sum_{i \in J} \gamma_i v_i^2 = \Delta t \sum_{i \in J} \gamma_\mathrm{p} v_i^2 + \Delta t \sum_{i' \in J} \gamma_\mathrm{s} v_i'^2$$

Energy gain of the electronic sub-system:

$$\Delta U_{\mathrm{eg}} = g_\mathrm{p} T_\mathrm{a} \Delta V \Delta t + g_\mathrm{s} T_\mathrm{a}' \Delta V \Delta t.$$

Equate the two:

$$\sum_{i \in J} \gamma_\mathrm{p} v_i^2 = g_\mathrm{p} T_\mathrm{a} \Delta V$$

$$\sum_{i' \in J} \gamma_\mathrm{s} v_i'^2 = g_\mathrm{s} T_\mathrm{a}' \Delta V$$

Define the atomic T values:

$$\tfrac{3}{2} k_\mathrm{B} T_\mathrm{a} = 1/N \sum_{i \in J} \tfrac{1}{2} m v_i^2$$

$$\tfrac{3}{2} k_\mathrm{B} T_\mathrm{a}' = 1/N' \sum_{i' \in J} \tfrac{1}{2} m v_i'^2$$

And solve for the coupling “constants”:

$$g_\mathrm{p} = \frac{3 N k_\mathrm{B} \gamma_\mathrm{p}}{\Delta V m}$$

$$g_\mathrm{s} = \frac{3 N' k_\mathrm{B} \gamma_\mathrm{s}}{\Delta V m}.$$

Implementation in LAMMPS

- **Generalized for use with multiple atom types.**
- **T_e varies in time and in all three spatial dimensions.**
- **User supplies physical constants and initial T_e values.**
- **Reasonable energy conservation achieved.**
- **Models thermal conductivity and finite heat capacity of the electron subsystem.**

Phillips and Crozier, *J. Chem. Phys.* **2009**, *131*, 074701

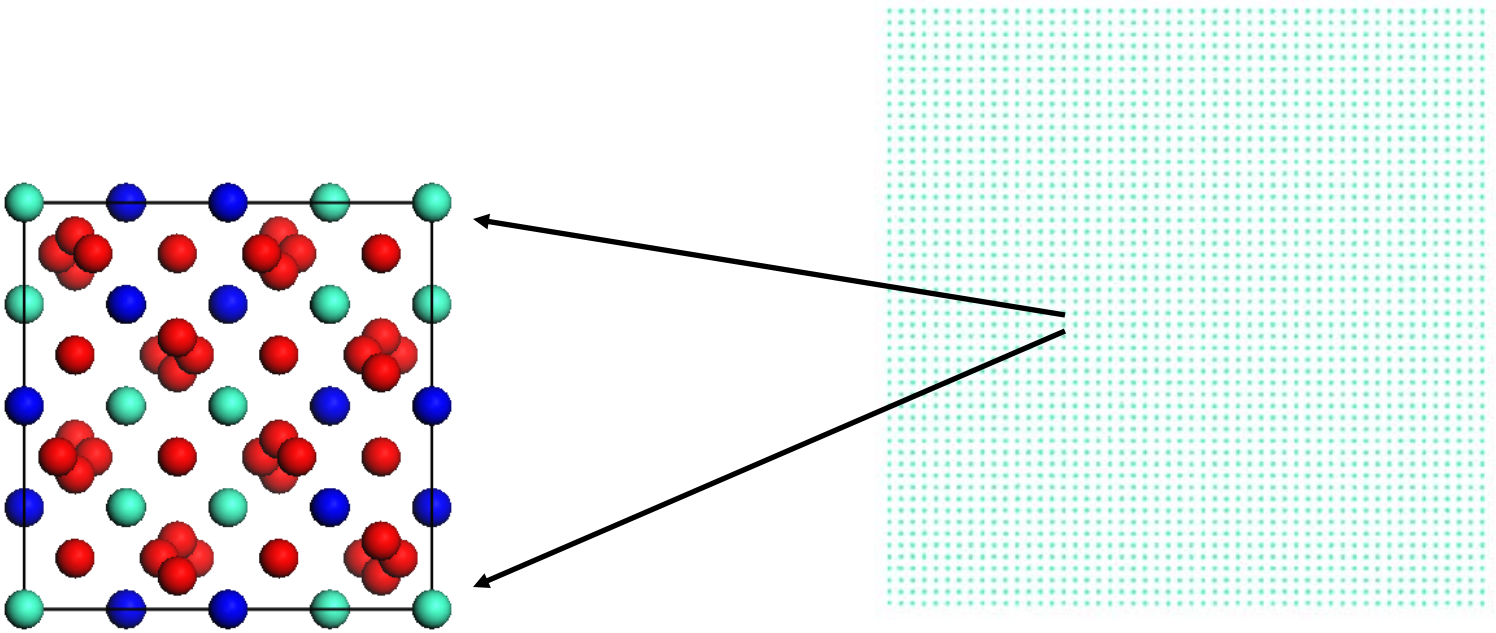
TTM input parameters (fix ttm)

- Electron specific heat*
- Electron density*
- Electron thermal conductivity
- Electron-ion interaction coefficient, γ_p*
- Electron stopping friction coefficient, γ_s (SRIM tables, www.srim.org)
- Electron stopping critical velocity, v_0
- Number of electronic grid points
- Initial electron temperature

*** DFT calculations can be used to estimate these values**

Example: cascade damage simulation

- Gadolinium pyrochlore waste form ($Gd_2Zr_2O_7$)
- Large initial velocity (10 keV) imparted to the primary knock-on atom (PKA) to simulate a radiation recoil event



10.8 Å

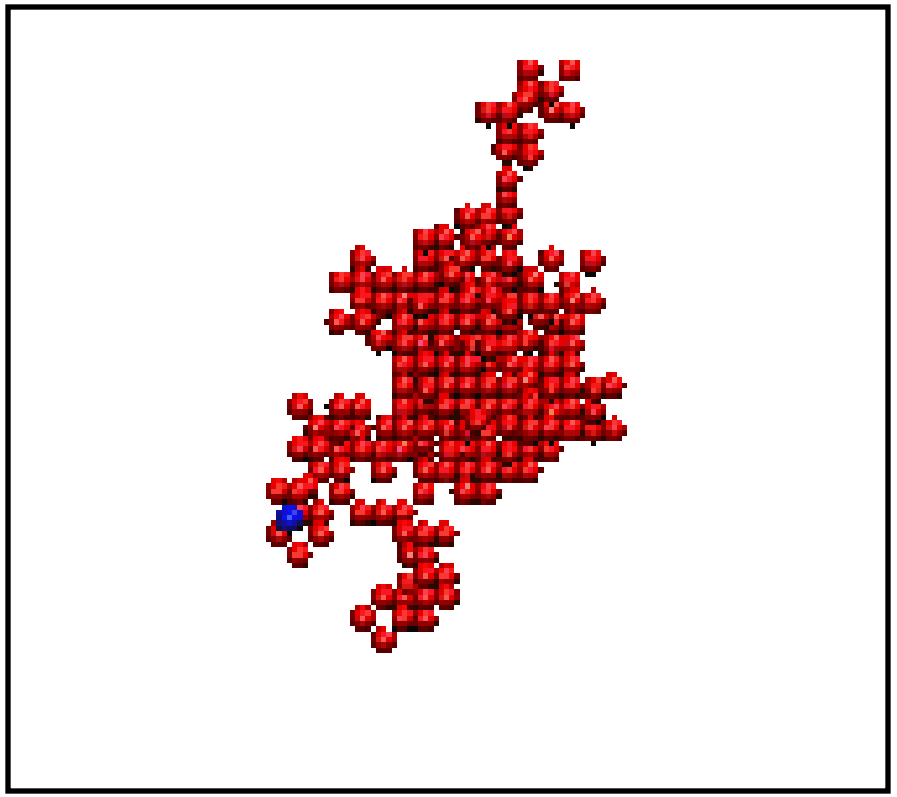
1 unit cell, 88 atoms

162 Å

15 x 15 x 15 supercell, 297k atoms
(only Gd atoms shown)

Ismail et al, *J. Phys. Condens. Matter*., submitted

Non-TTM results: Gd defects

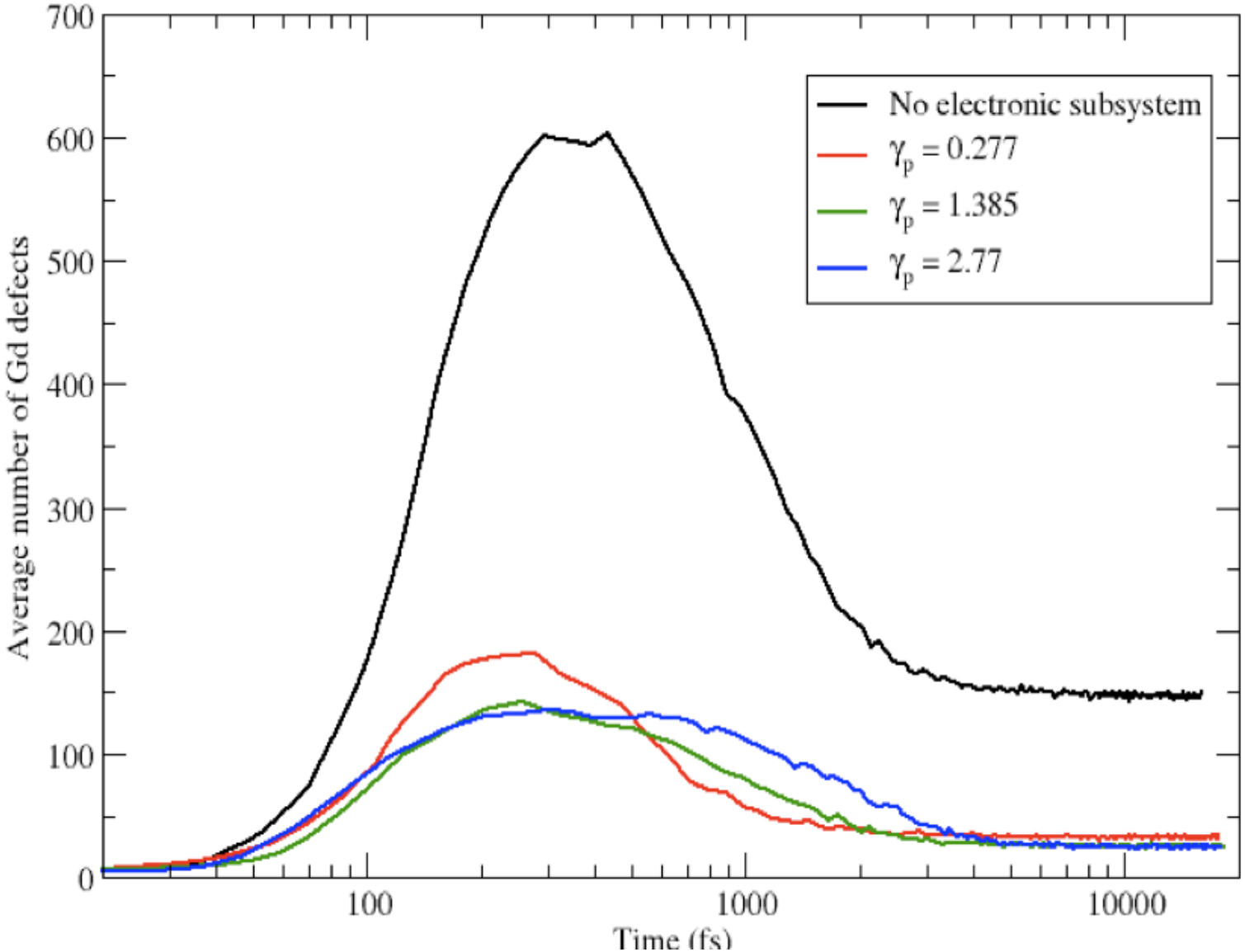


Blue = PKA (U)

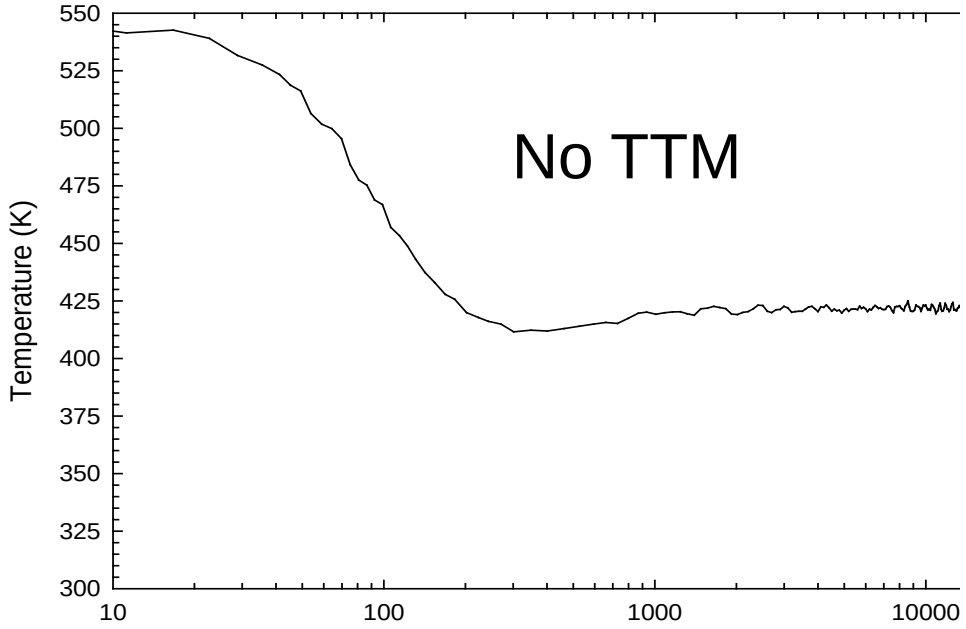
Red = defect Gd

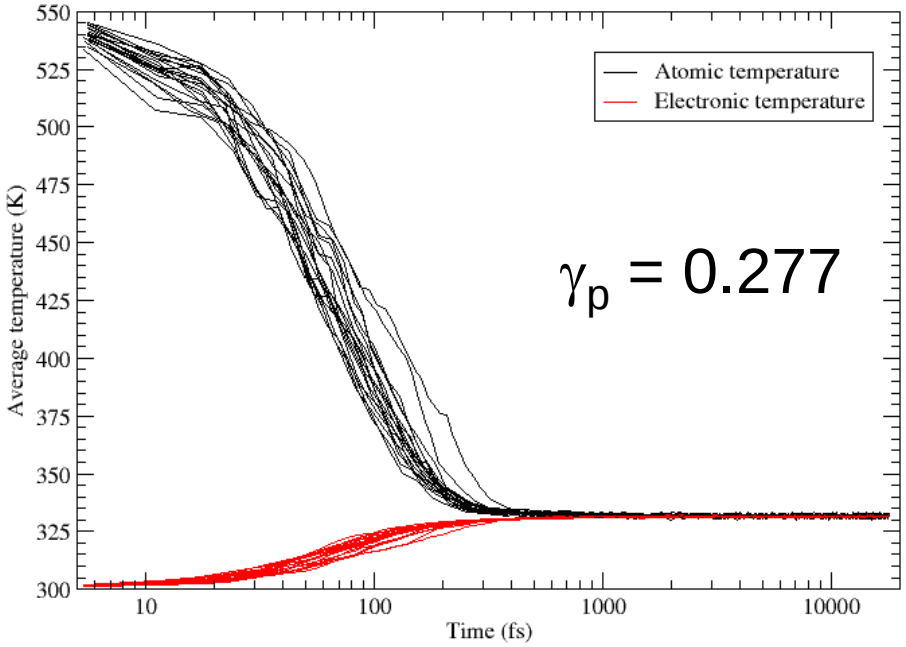
Results of defect analysis

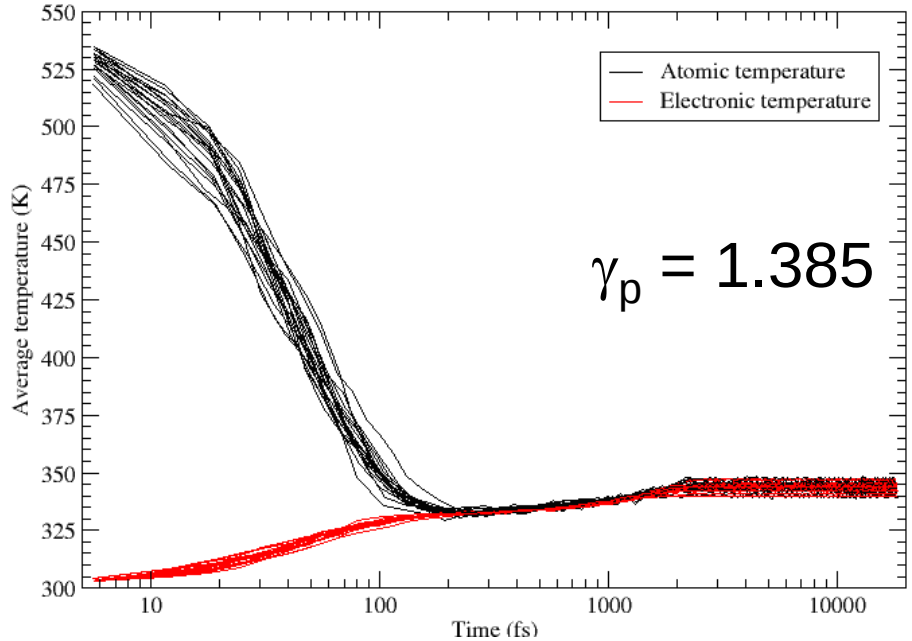
Averages from 20 random PKA directions

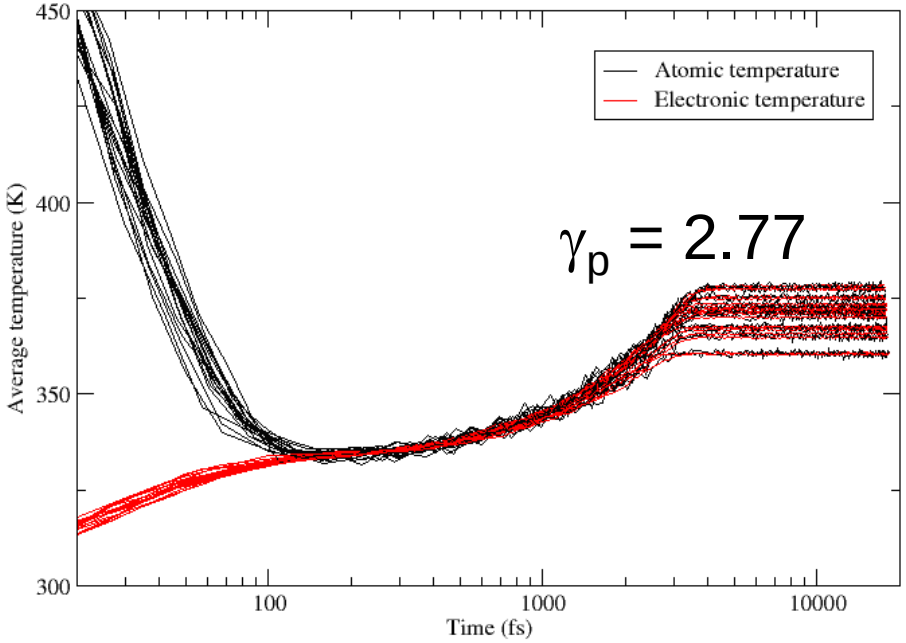


Atomic and electronic temperature

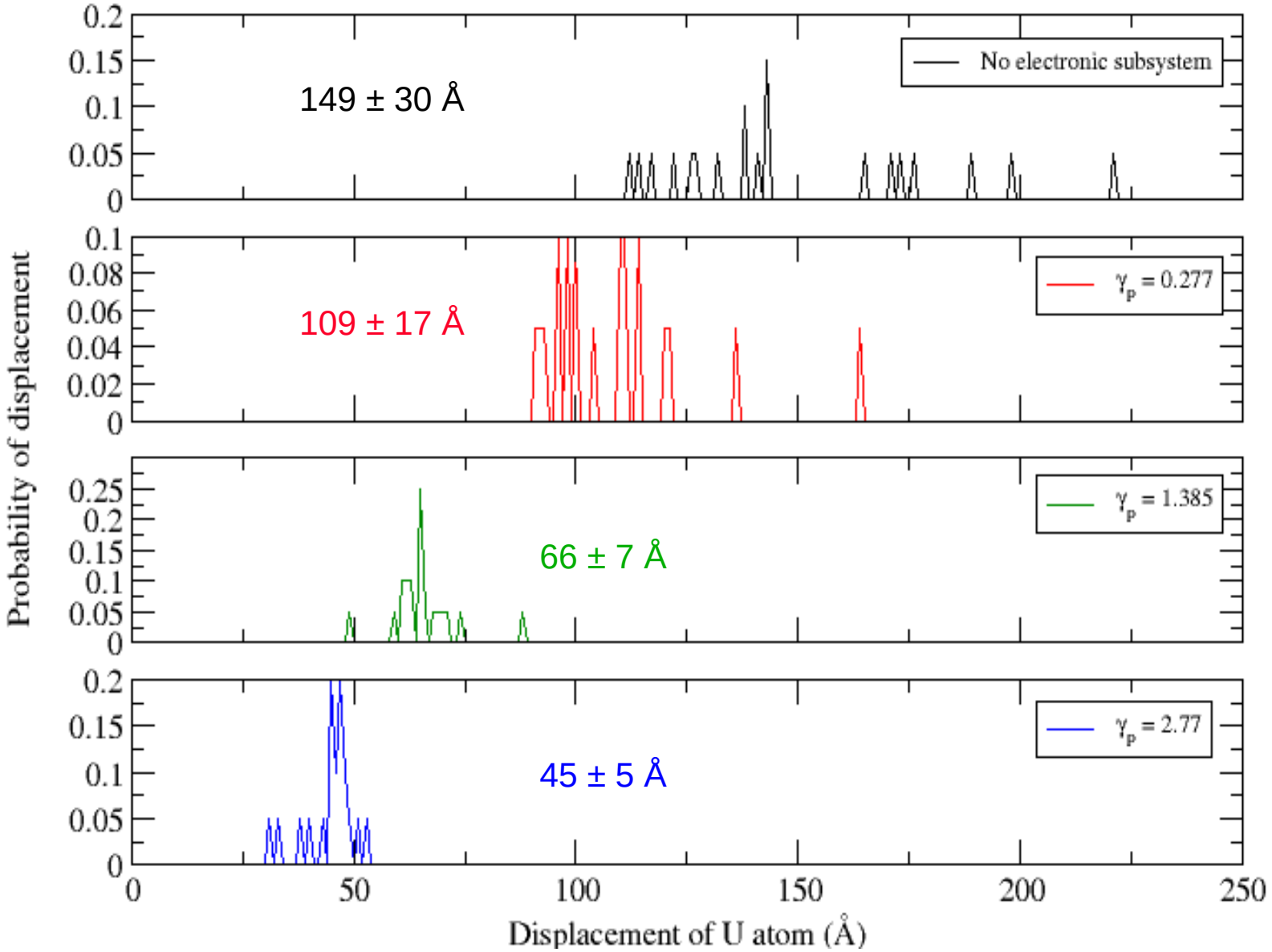








PKA (U atom) displacement results



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