Modeling and Simulation

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Stanford U.: Kiran Prasai, Riccardo Bassiri, Marty Fejer

References: Our Previous Modeling-Simulation of TLS and Loss

- Silica

Rashid Hamdan, Jonathan P. Trinastic, and H. P. Cheng, J. Chem. Phys.**141**,054501 (2014)

- Tantala and Ti-doped tantala

Jonathan P. Trinastic, Dustin J. Davis, Rashid Hamdan, and Hai-Ping Cheng. Phys. Rev. B95, 014109 (2017).

- Silica revisited

C. R. Billman, J. P. Trinastic, D. J. Davis, R. Hamdan, and H. P. Cheng, Physical Review B, 2017. **95**(1): p. 014109

- Potential energy functions

Jonathan P Trinastic, Rashid Hamdan, Yu-Ning Wu, L. Zhang and Hai-Ping Cheng, J. Chem Phys. **139**,154506 (2013)

Current Activities

I. Structure Model Generation

- Melting-quenching + Reversed MC
- Simulated growth

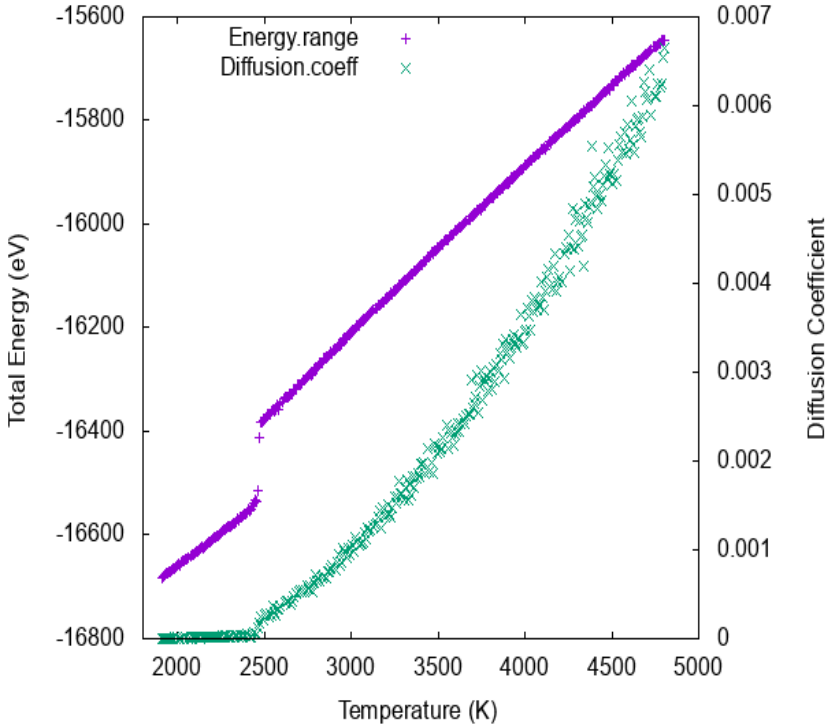
III. Potential Function Construction

- DFT calculation, parameter fitting

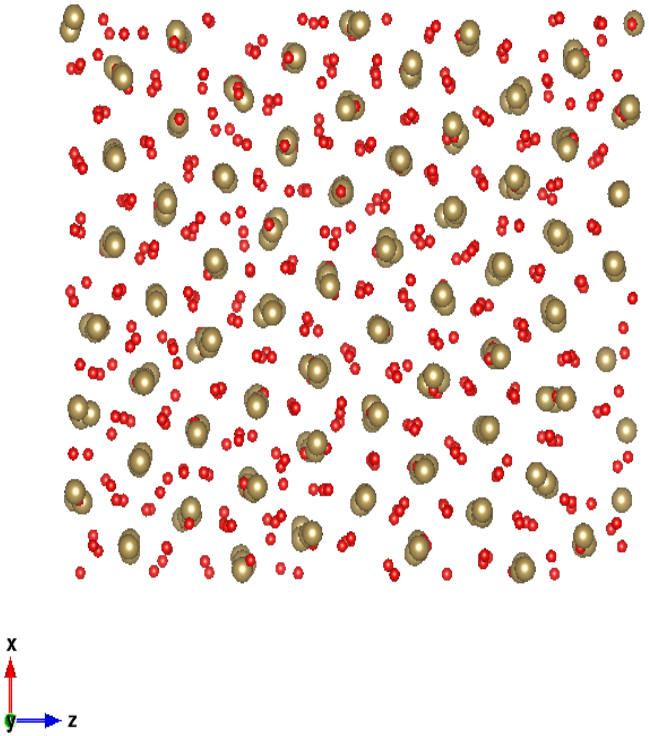
II. Loss Calculations

- Energy landscape, statistics, and loss prediction

Tantala (Ta_2O_5) Melting-Crystalli

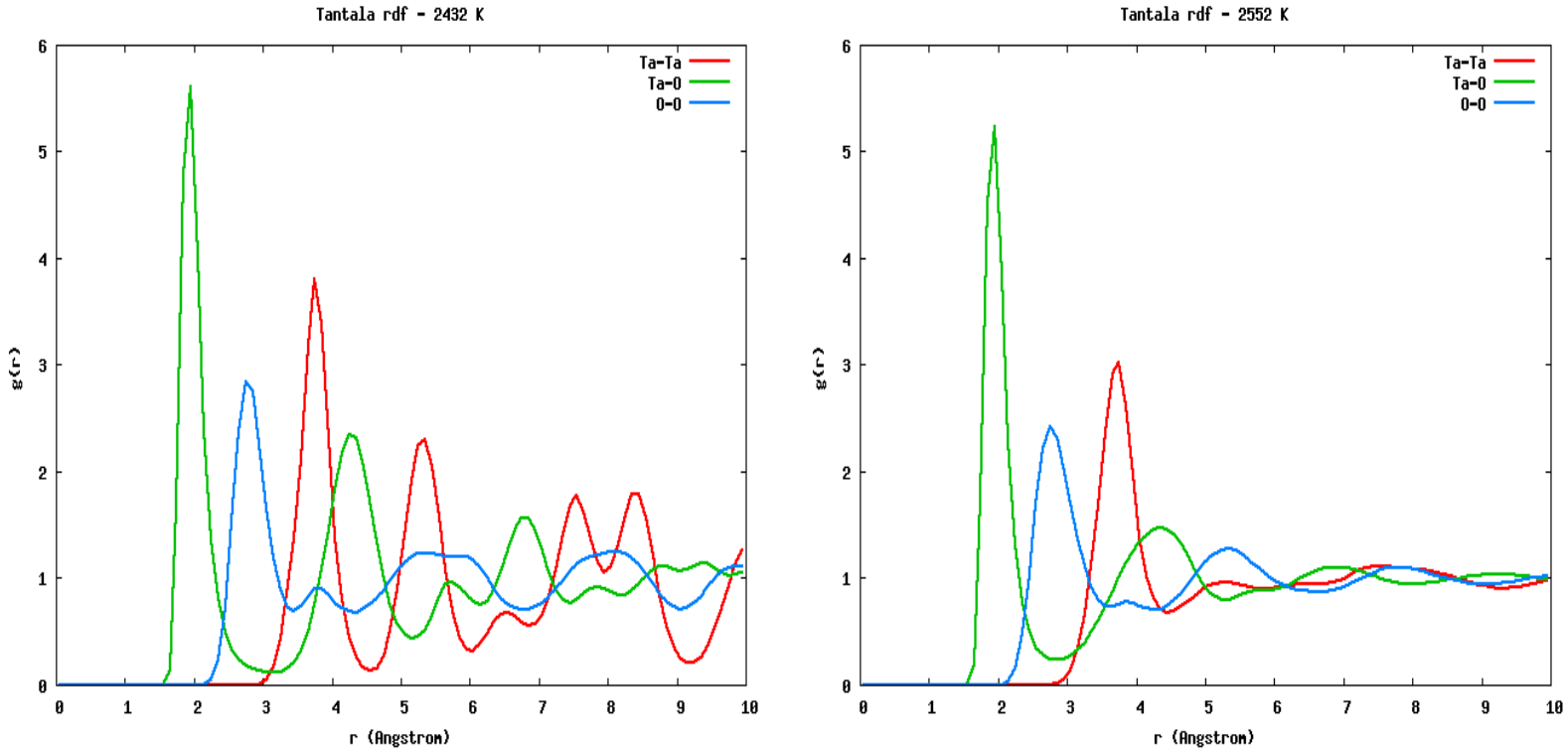


Quench of previously heated Tantala (Ta2O5) crystal starting from a temperature of 4800 K and cooled to 1912 K. The discontinuity in total energy and drop to zero in the diffusion coefficient indicates a phase transition at 2475 K.



Structure of Tantala (Ta2O5) below the phase transition at 2432 K.

Tantala (Ta_2O_5) Crystallization

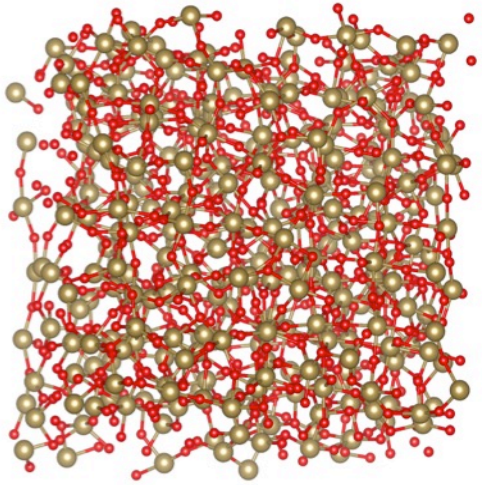


Left: Radial distribution functions for Tantala at 2432 K, after crystallization, and Right: in an amorphous configuration, at 2552 K, on the right.

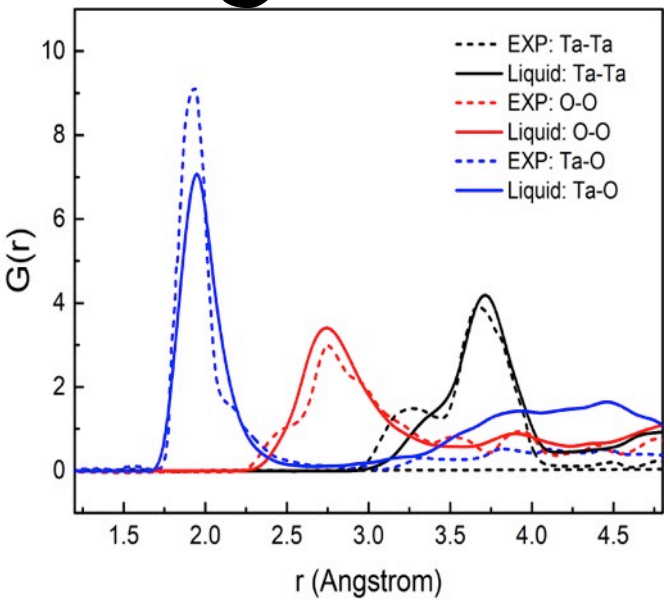
Tantala (Ta_2O_5) Crystallization

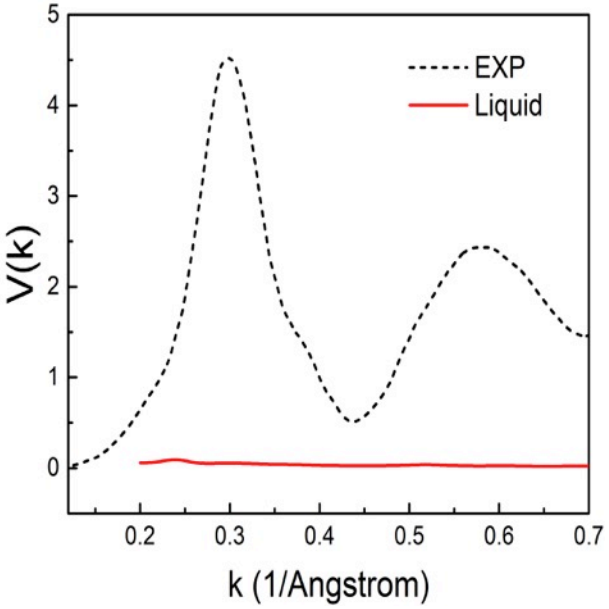
- Simulation was done using the LAMMPS classical molecular dynamics simulation package.
- Previously heated Ta_2O_5 was quenched in intervals of 8 K. Each data point consists of equilibration and cooling over 180,000 timesteps in an npt ensemble.
- Linear regression was used on the mean-squared distribution data collected to obtain the diffusion coefficient for each temperature interval.
- The phase transition can be seen at 2475 K from the discontinuity in energy drop in the diffusion coefficient.
- Crystallization of Tantala was observed below this point.

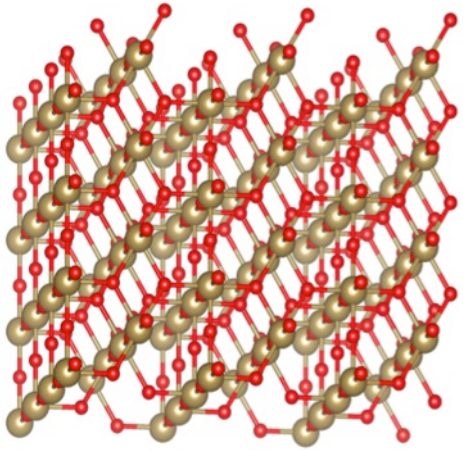
Structure, g(r) and Variance



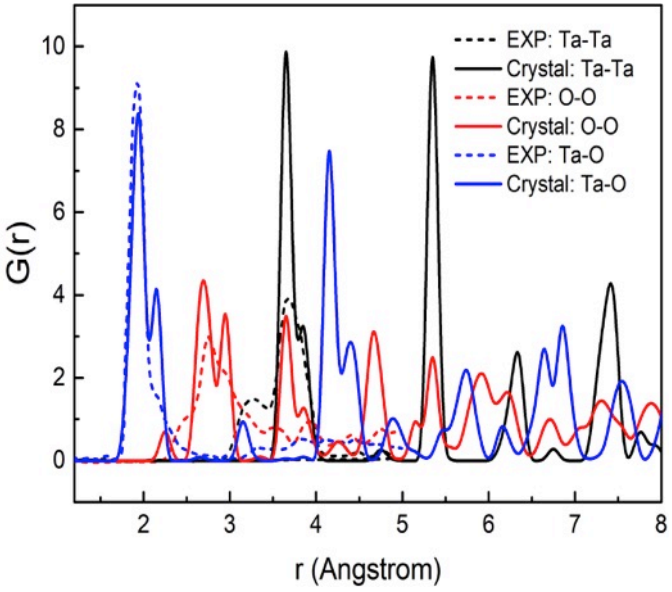
Amorphous Tantala

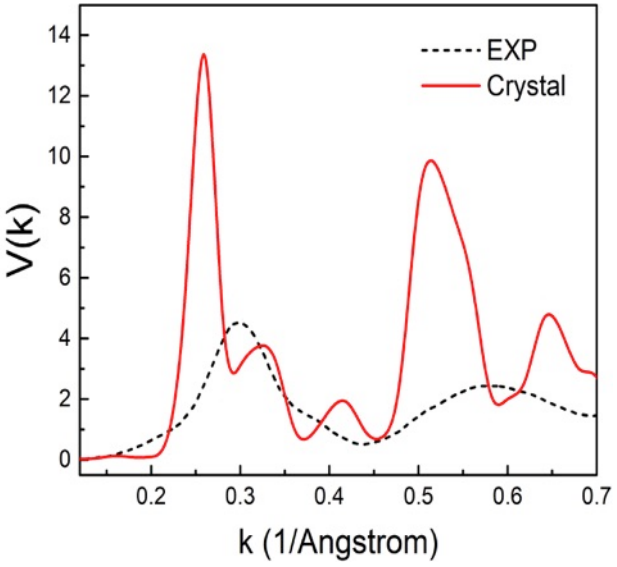




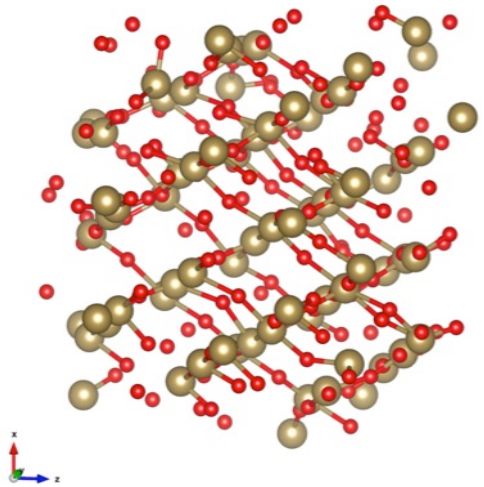


crystalline Tantala

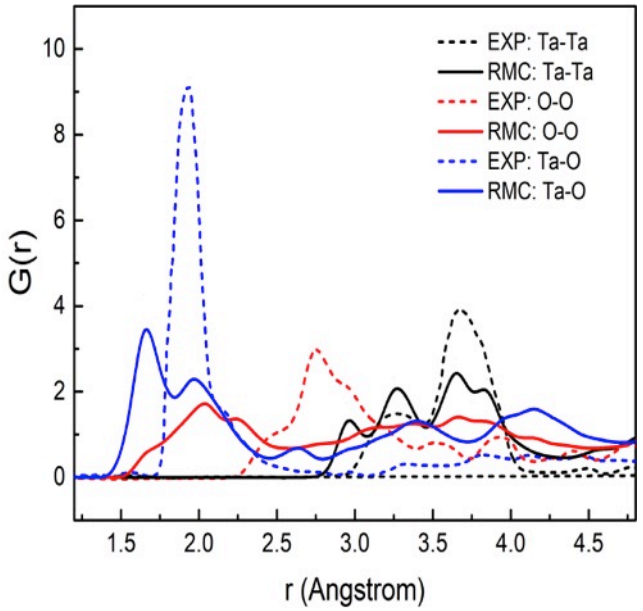


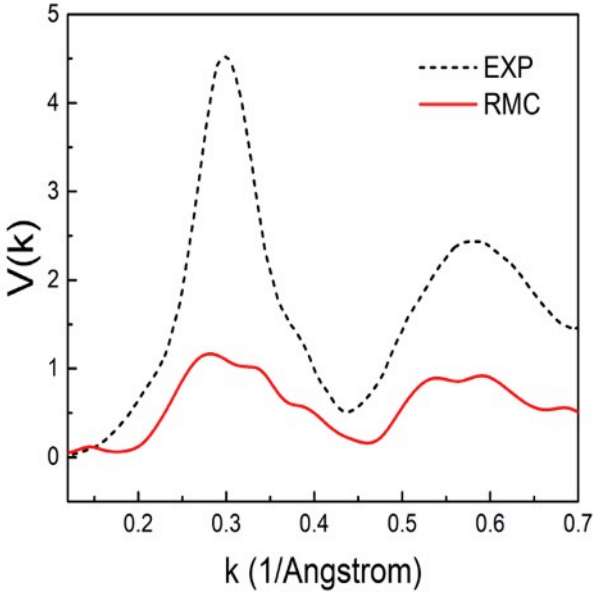


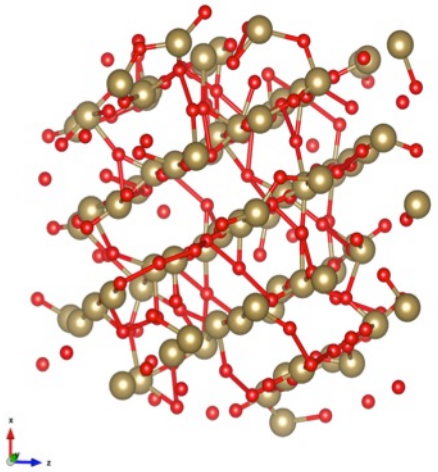
Matching Experiments using reversed Monte-Carlo

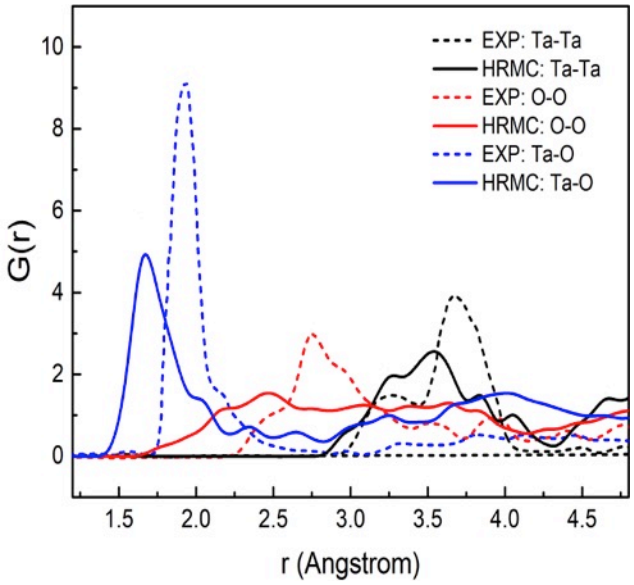


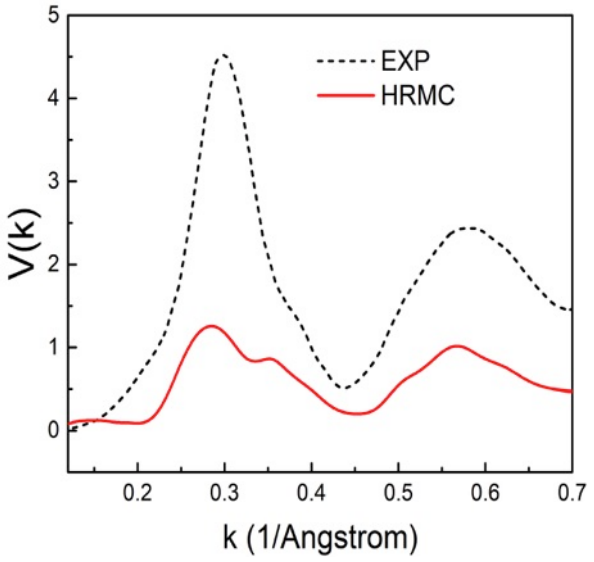
Above: RMC without energy constraint
Below: With energy



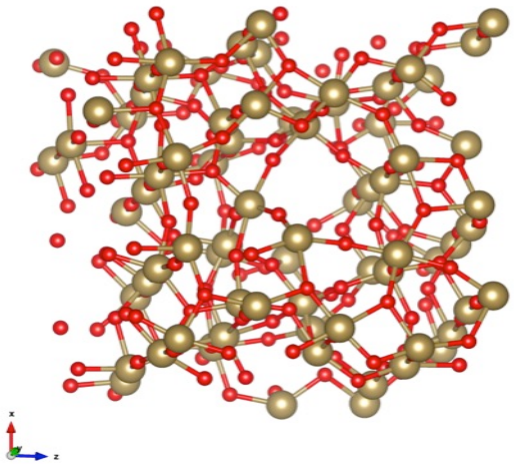


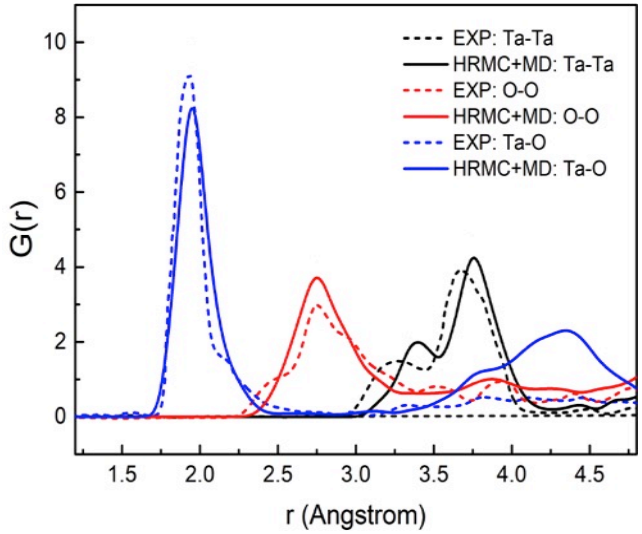


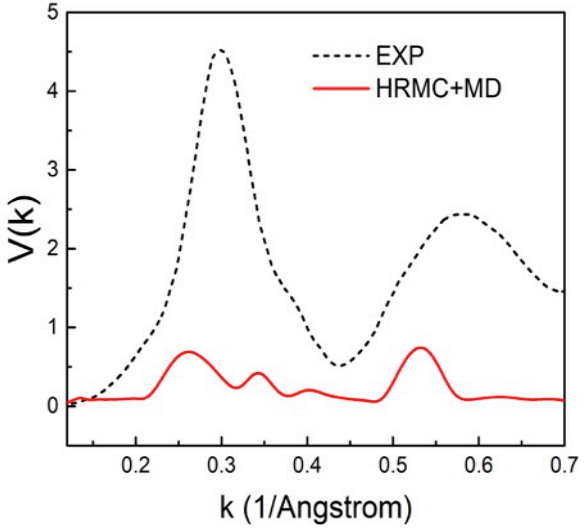




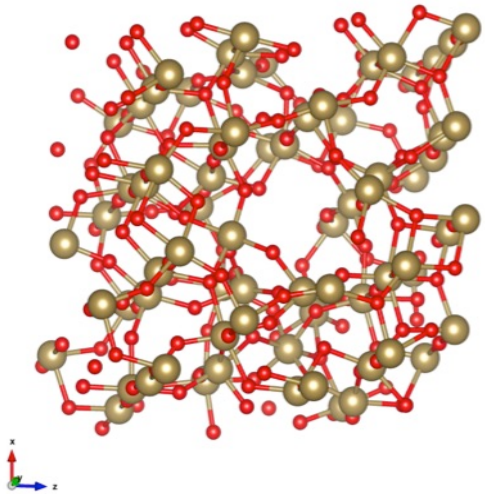
Matching Experiments using reversed Monte-Carlo

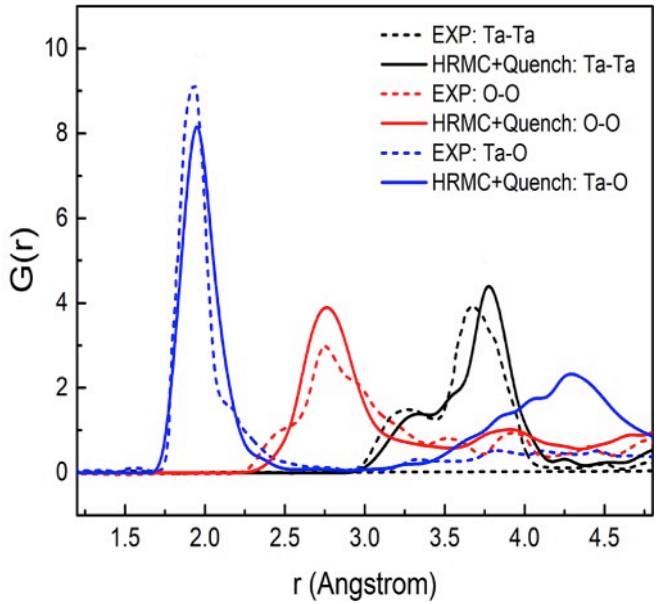


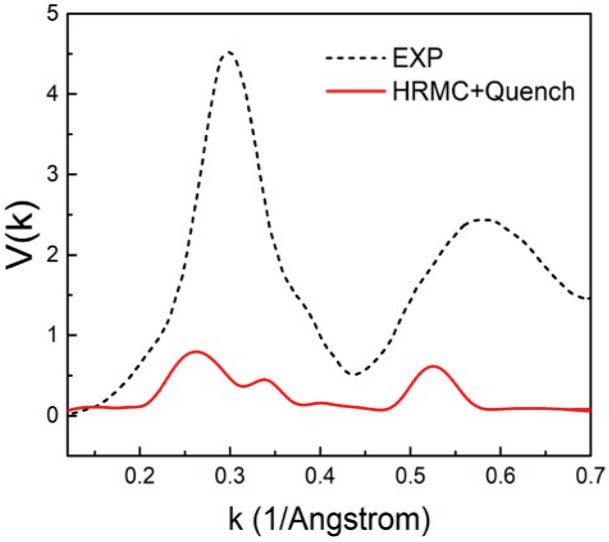




Above: MD after HRMC
Below: Quench after HRMC







Ta_2O_5 growth simulation with LAMPPS

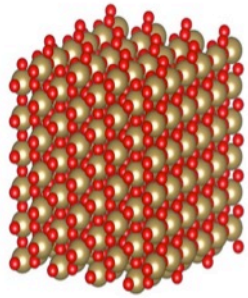
Simulation pair potential is based on previous references.
Unified interatomic potential and energy barrier distributions for amorphous oxides, JCP 139,154506(2013)
The pair potential of Ta-O Si-O have been provided by the reference. The pair potential is based on the charged atom.

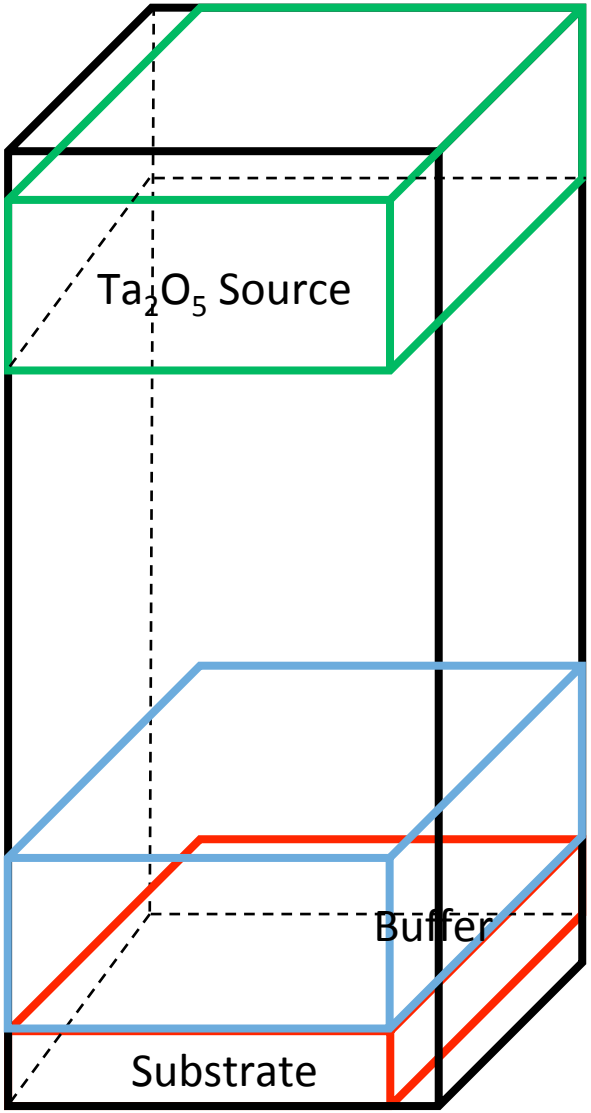
System setup:
Substrate: Silica(SiO_2), Silicon; experiment substrate is amorphous fused silica
Deposit material: Tantalite, tantalum pentoxide(Ta_2O_5), Ions in real experiment deposit
Deposit method: lammps (fix deposit, fix pour with g) PS: need add fix_deposit module in MISC lammps

Simulation region:
The box size would be: a a a+b+c (a is the cubic box for TLS, b is the substrate, c is the source of the deposit materials)
Boundary condition: p p f (Normal simulation), p p p (for long range Coulomb potential)

Calculated material properties: density, defect density, RDF, variance
Experimental electron beam radius: about 2nm = 20 A, which Q = 0.61/R = 0.0305 1/A
Deposit parameters: substrate density, temperature, incident atom speed
Deposit method: deposit ions, deposit atom, deposit molecular

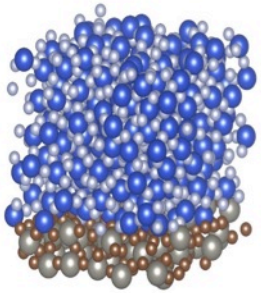
Model Structure



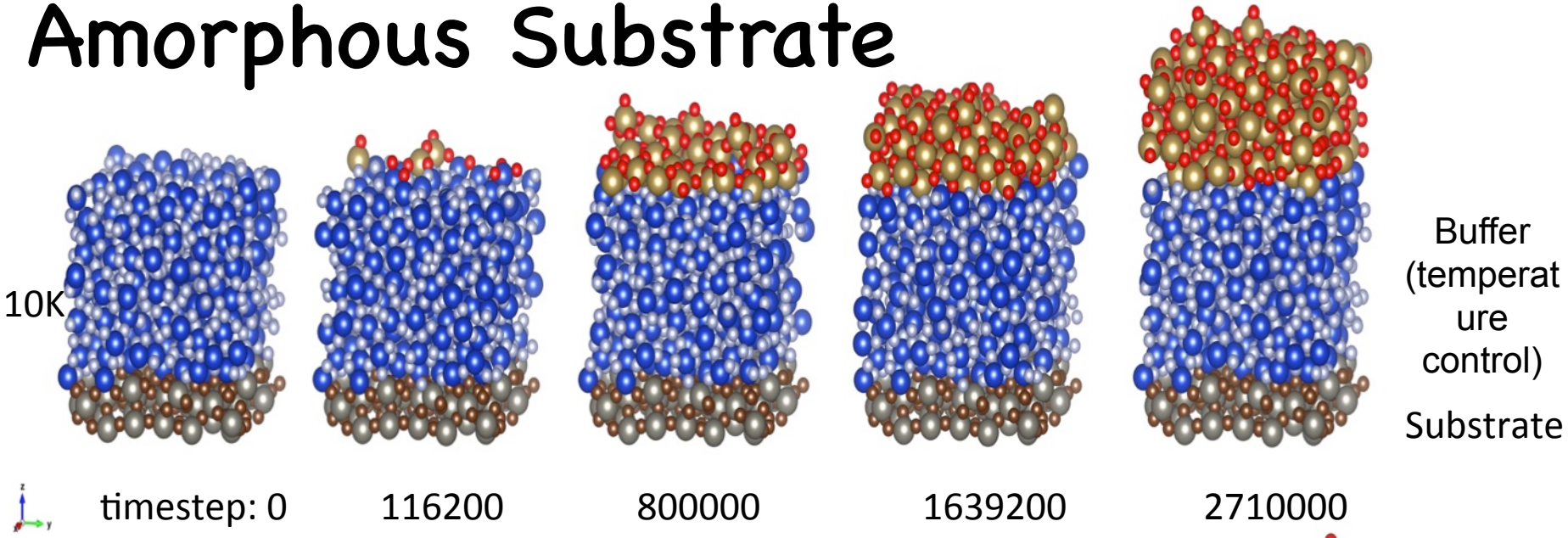


$2000K < V_o < 4000K$

V_o



Amorphous Substrate



MD deposit simulation references:

Structure: beta-Ta2O5

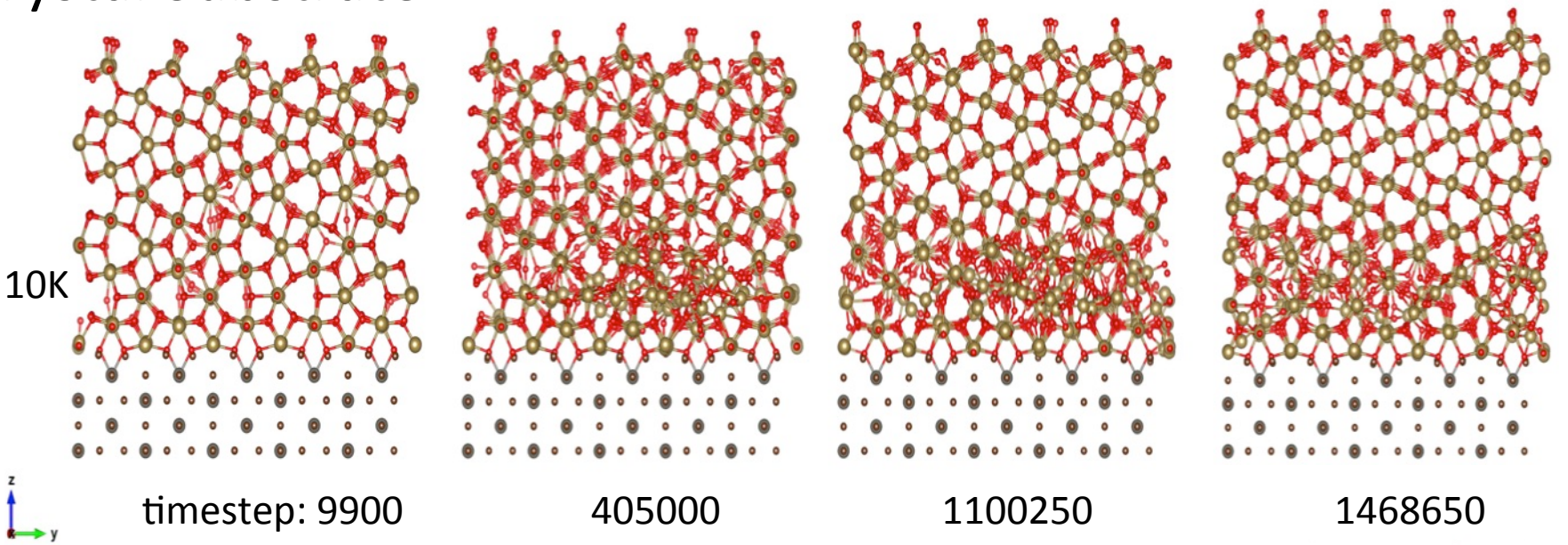
[1] Aleshina, L. A., and S. V. Loginova. "Rietveld analysis of X-ray diffraction pattern from β-Ta 2 O 5 oxide." *Crystallography Reports* 47.3 (2002): 415-419.

MD Potential:

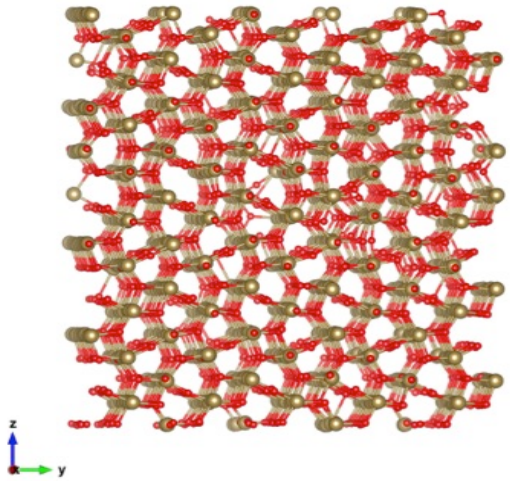
[1] Trinastic, J. P., et al. "Unified interatomic potential and energy barrier distributions for amorphous oxides." *The Journal of chemical physics* 139.15 (2013): 154506.

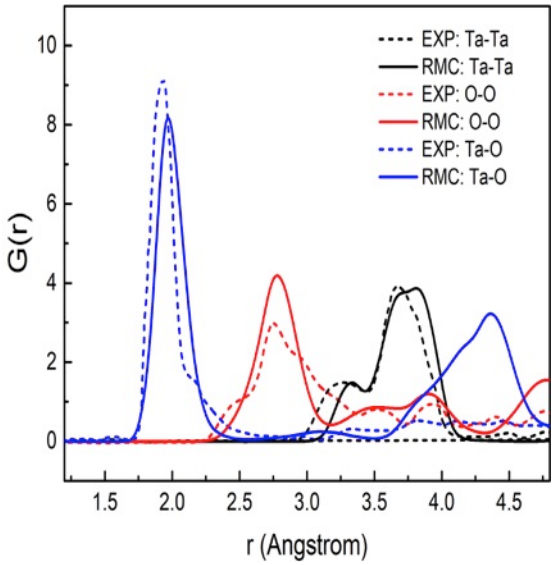
Crystal Substrate

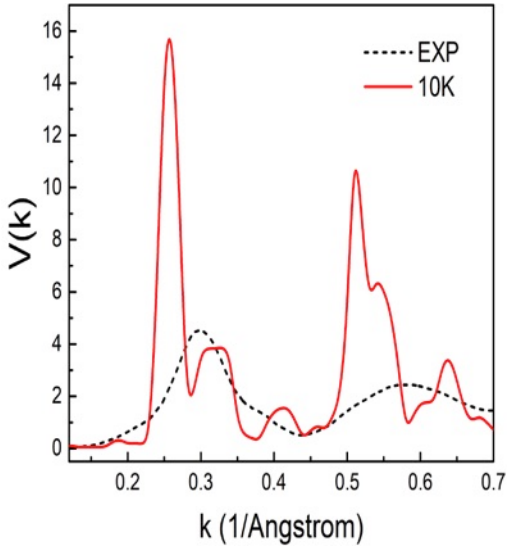
Deposit start timestep: 10000

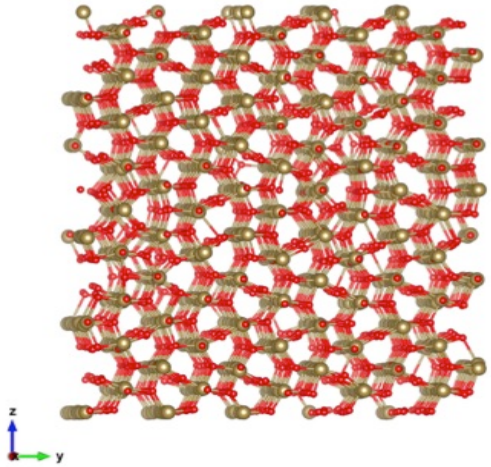


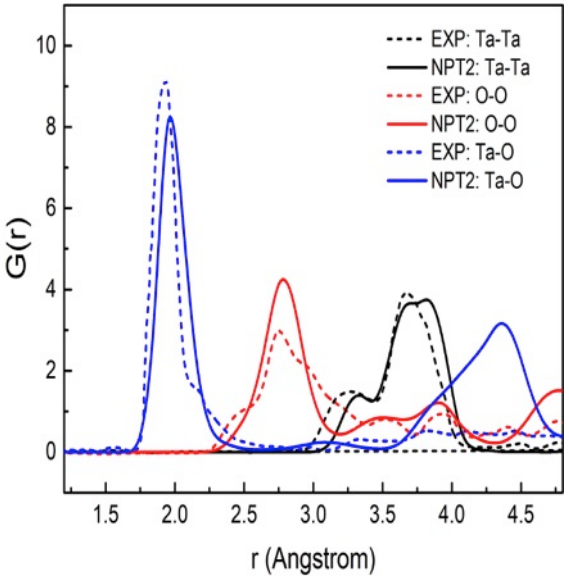
Crystal Substrate Charaterization

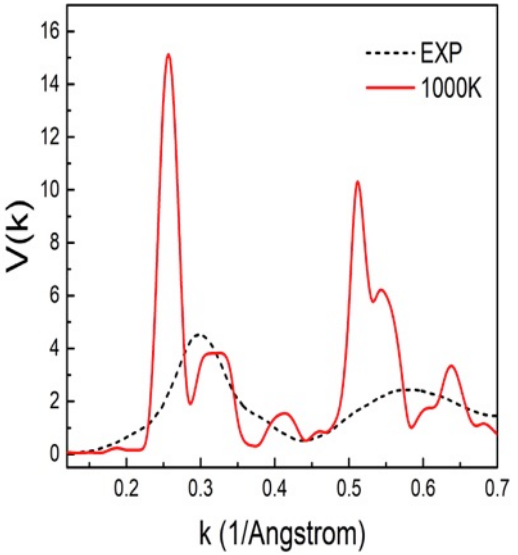












Interatomic Potential for Nb_2O_5

- A BKS-Morse potential was constructed for the B and H phase of Niobium Pentoxide (Nb_2O_5) using the General Utility Lattice Program (GULP).
- Parameters of the interatomic potential were adjusted to reproduce elastic properties of the two phases of Nb_2O_5 simultaneously, specifically the elastic tensor components and derived bulk, shear and young's moduli.
- The elastic properties from our fitted potential were compared to those obtained through DFT calculations using VASP, given the absence of experimental measurements for Tantala's desired properties.
- The DFT calculation results can be compared to results published by C. Valencia-Balvín et al.
- Table 1 lists the optimized parameters along with the analytical form of the Buckingham and Morse terms.
- Table 2 lists the percent error in the DFT calculations and those using our fitted potential.

Interatomic Potential for Nb_2O_5

$$U_{ij} = U_{ij}^{BKS} + U_{ij}^{Morse} - \left(\frac{q_i q_j}{r_{ij}} \right)$$

$$U_{ij}^{BKS}(r_{in}) = A\exp\left(-\frac{r_{ij}}{\rho}\right) - \frac{C_{ij}}{r_{ij}^6} + \ldots \qquad U_{ij}^{Morse}(r_{in}) = D\left(\left[1-\exp\left(-a\left(r_{ij}-r_o\right)\right)\right]^2 - 1\right)$$

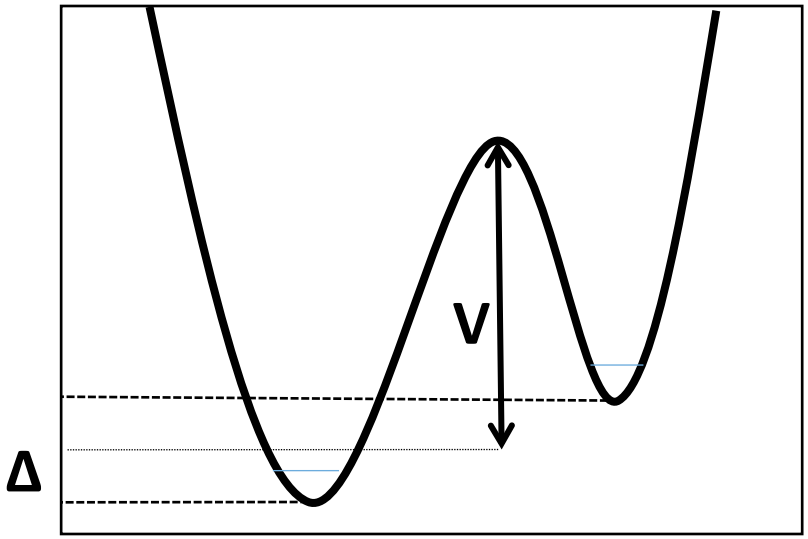
Interaction	A	Rho	C	D	a	r_0	q
Nb – O	100067.01	0.1319	6.05	0.293288	1.602698	2.617186	Nb: 3.0
O – O	740.84767	0.399508	175.0000	0.000	0.000	0.000	O: -1.2

Table 1: Optimized parameters for the M-BKS potential.

Material	Property (GPa)	DFT	Fit	Percent Error
Nb2O5 - B phase				
	Bulk	109.86	118.64014	7.992
	Shear	59.276	58.99578	0.473
	Young x	211.40	219.79628	3.972
	Young y	138.31	141.76593	2.499
	Young z	110.71	143.91642	29.994
Nb2O5 - H phase				
	Bulk	154.65	138.56986	10.398
	Shear	58.443	70.08824	19.926
	Young x	217.08	201.32302	7.259
	Young y	252.85	218.53058	13.573
	Young z	225.44	192.17224	14.757

Table 2: Bulk, shear and Young's moduli of Nb_2O_5 using M-BKS potential alongside fitting parameters (DFT values).

Two-Level Systems and Internal Friction: Old



- Amorphous solids modeled as an ensemble of two level systems (TLS) characterized by their energy asymmetry (Δ)and barrier height (V)
- Longitudinal or transverse (l/t) internal friction (Q^{-1}) based on excited mode in solid

$$Q_{l/t}^{-1} = \frac{\gamma_{l/t}^2}{k_B T E_{l/t}} \int_0^\infty \int_0^\infty \frac{\omega\tau}{1+(\omega\tau)^2} \operatorname{sech}^2\left(\frac{\Delta}{2k_B T}\right) g(V) f(\Delta)\, dV\, d\Delta$$

Deformation potential

Elastic modulus

Illuminating frequency

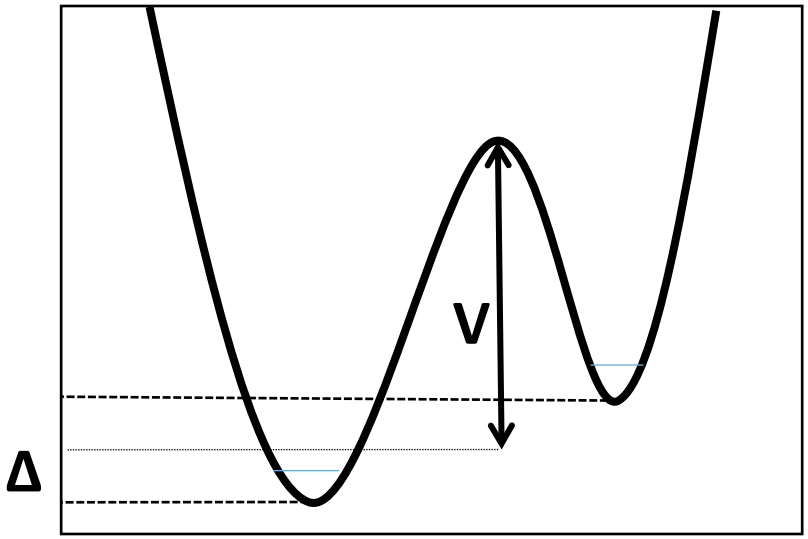
Relaxation time (function of V)

Barrier height (V) distribution

Asymmetry (Δ) distribution

Gilroy, K. S. and Phillips, W. A., Philos Mag. B, **38,** 735 (1981)

Two-Level Systems and Internal Friction: New



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Deformation potential

Elastic modulus

Illuminating frequency

Relaxation time (function of V)

Barrier height (V) distribution

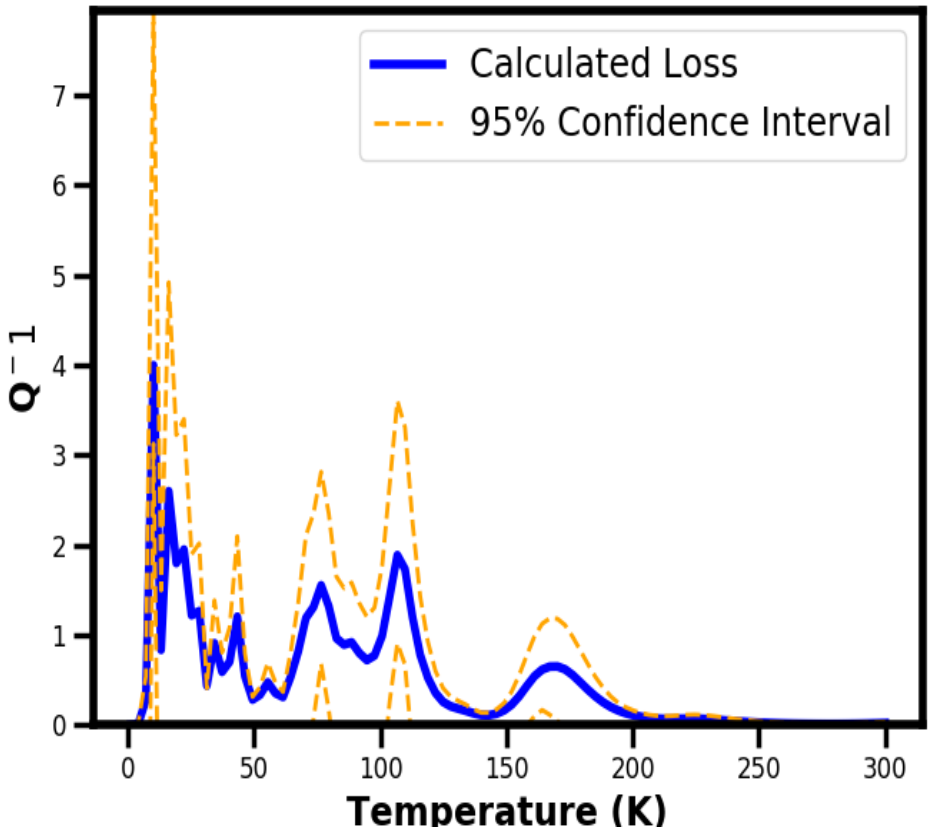
Asymmetry (Δ) distribution

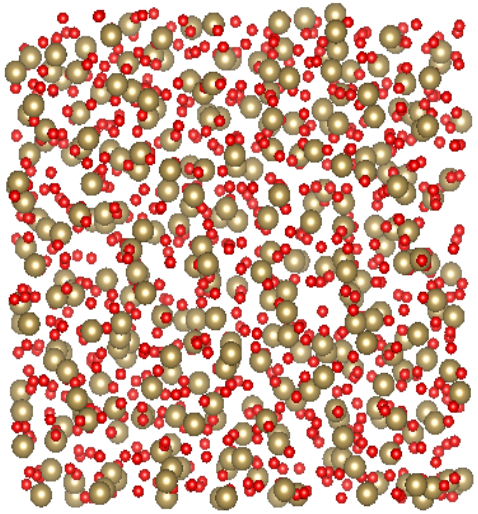
Gilroy, K. S. and Phillips, W. A., Philos Mag. B, **38,** 735 (1981)

Ta_2O_5 and Zr-dope Ta_2O_5

- Not converged yet!
- See the comparison with old formulation

Mechanical Loss – Pure Ta_2O_5

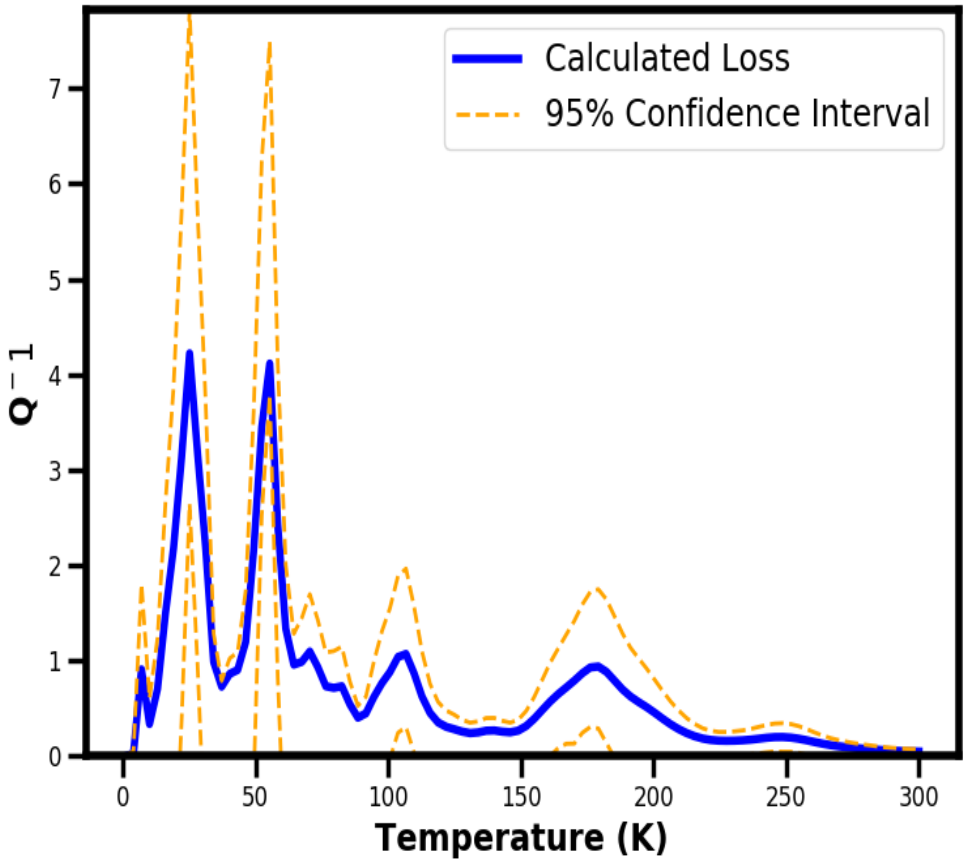


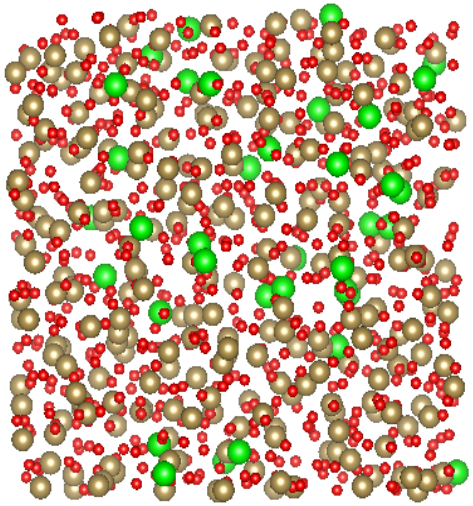


Longitudinal

Calculated internal friction (Q^{-1}) of pure Tantala (Ta_2O_5) as a function of temperature (T). Magnitude on the order of $x10^{-3}$ with observation frequency ω = 1 kHz.

12% ZrO_2 doped Ta_2O_5

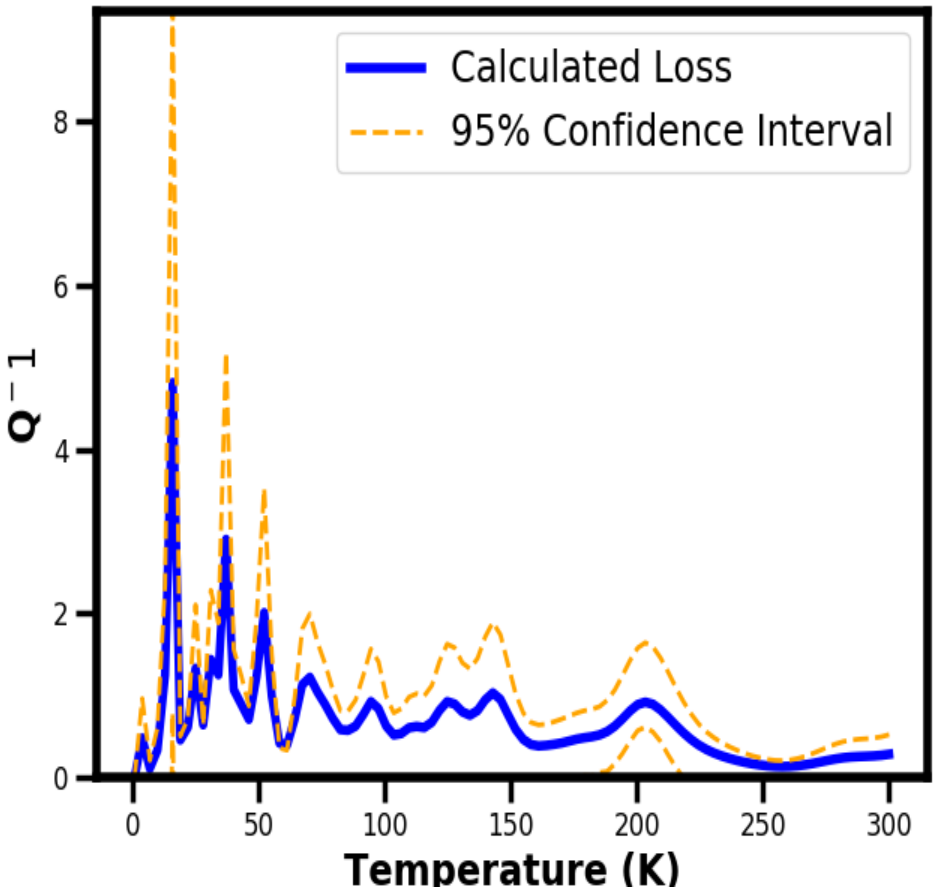


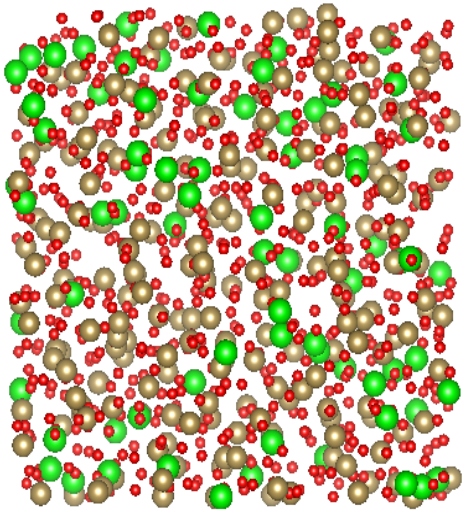


Longitudinal

Calculated internal friction (Q^{-1}) of 12% Zirconia ZrO_2 –doped Tantala (Ta_2O_5) as a function of temperature (T). Magnitude on the order of $x10^{-3}$ with observation frequency ω = 1 kHz.

25% ZrO_2 doped Ta_2O_5

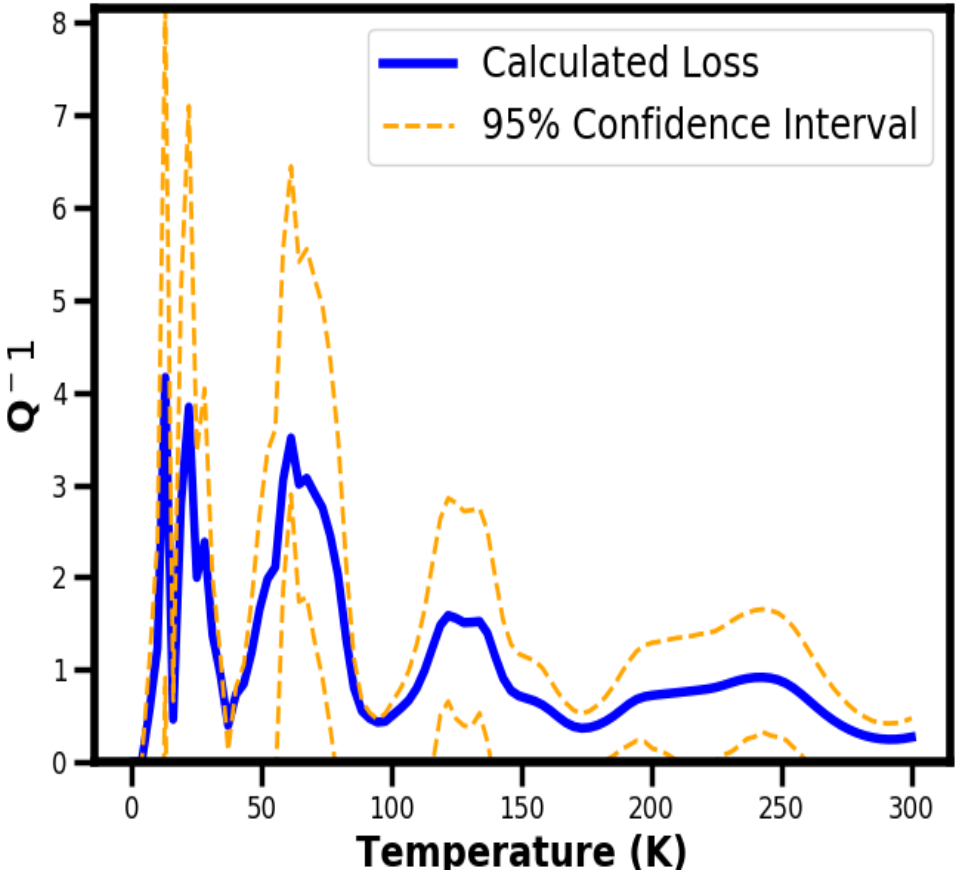


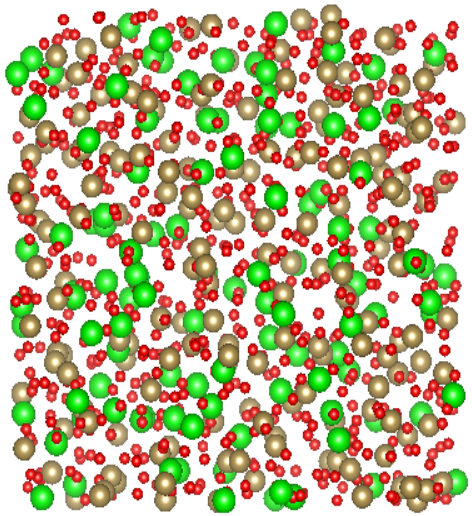


Longitudinal

Calculated internal friction (Q^{-1}) of 25% Zirconia ZrO_2 doped Tantala (Ta_2O_5) as a function of temperature (T). Magnitude on the order of $x10^{-3}$ with observation frequency ω = 1 kHz.

38% ZrO_2 doped Ta_2O_5





Longitudinal

Calculated internal friction (Q^{-1}) of 38% Zirconia ZrO_2 doped Tantala (Ta_2O_5) as a function of temperature (T). Magnitude on the order of $x10^{-3}$ with observation frequency ω = 1 kHz.

Longitudinal vs. Transverse Q^{-1}