First principles site occupation and migration of helium in β-phase erbium hydride

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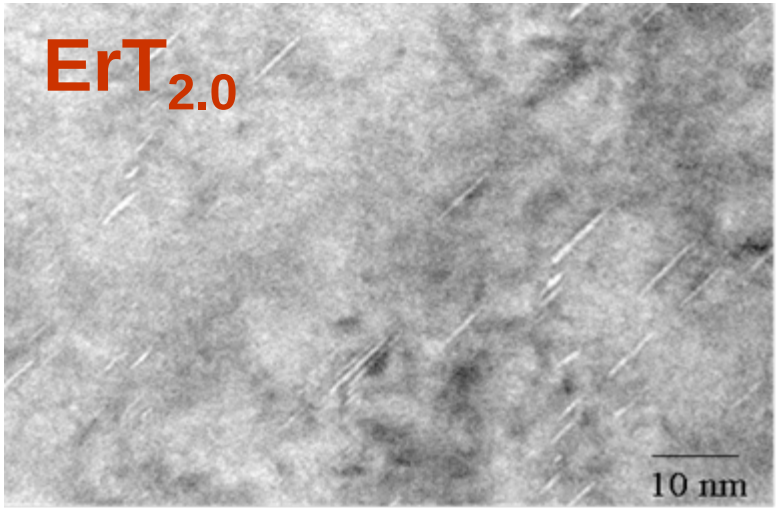
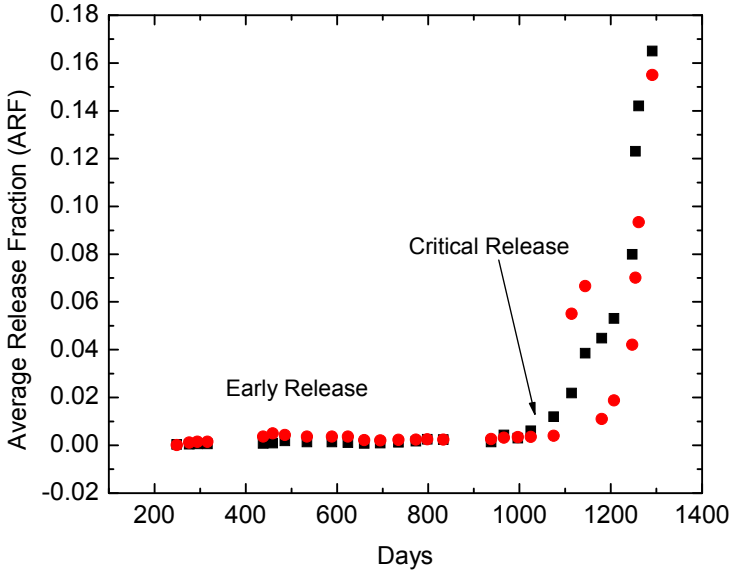
A helium release primer

- The majority of helium is stored in helium bubbles.



- Helium release breaks down into two phases, early and critical release.



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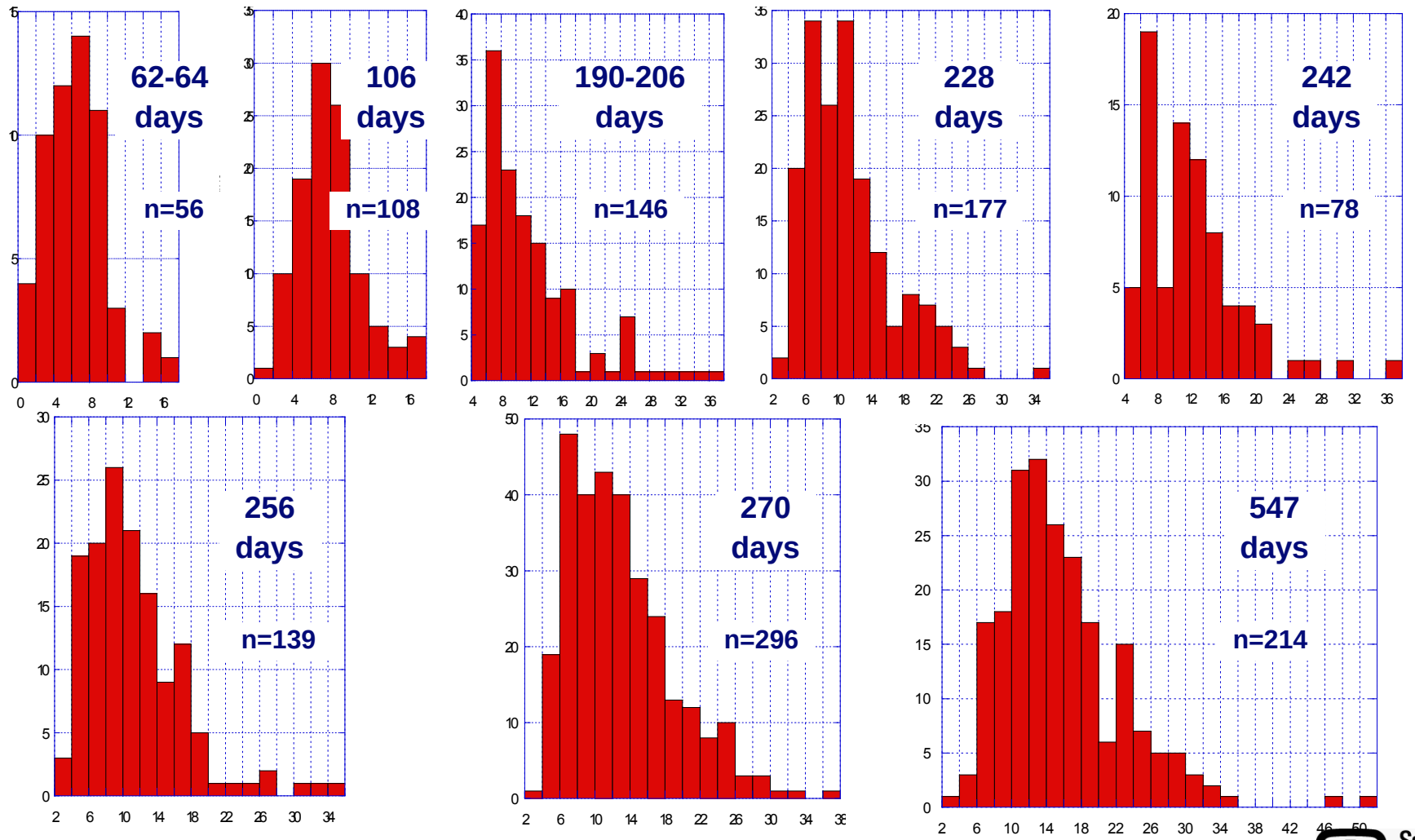
Helium Bubble Growth in $ErT_{2-x}He_x$



Data by Gillian Bond NMT, James Browning ORNL, and C. Snow

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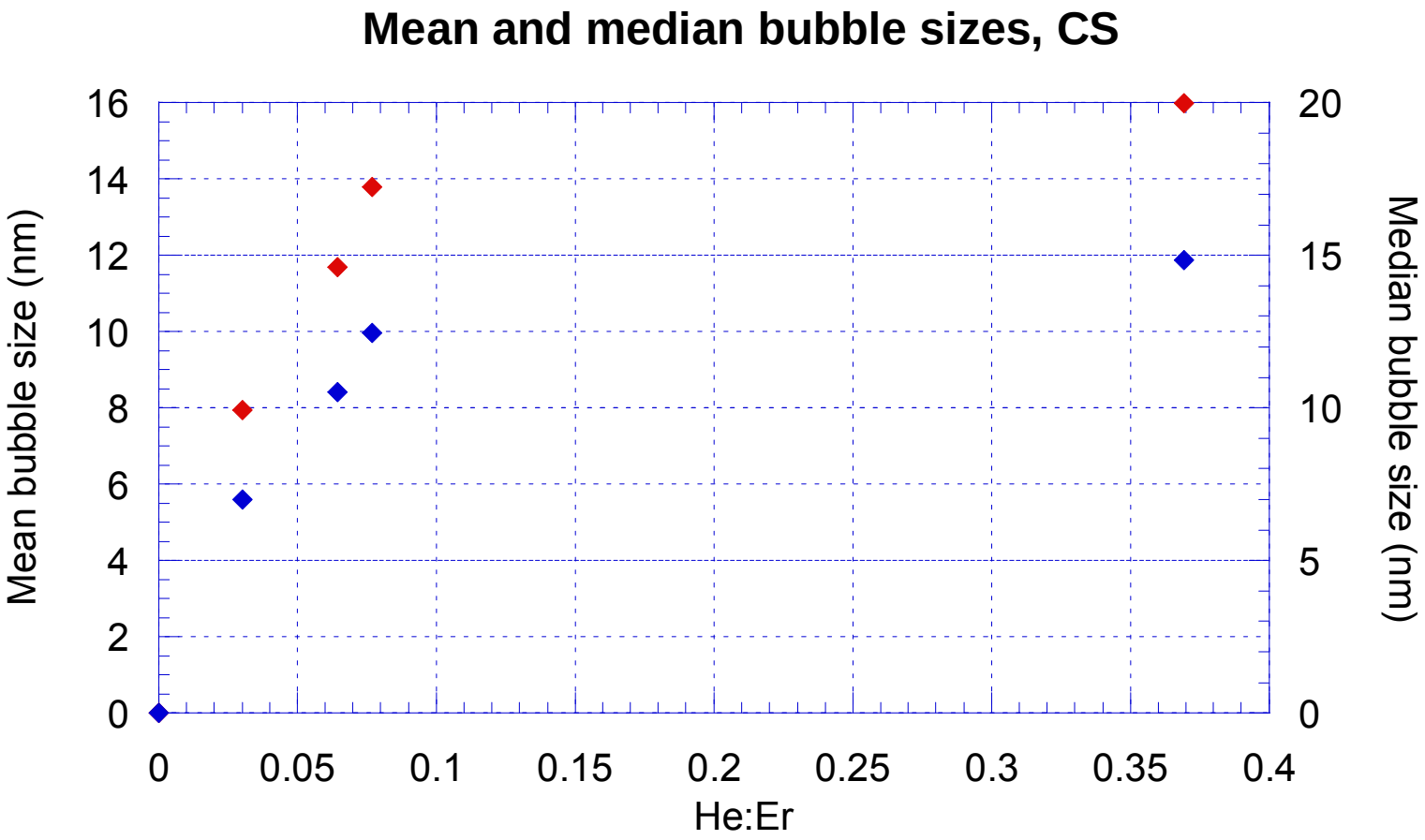
Bubble Sizes in $ErT_{2-x}He_x$



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Computational Methodology?

- We used two different DFT codes.
  - SeqQuest
    - Developed at Sandia by Peter Schultz
    - Gaussian basis sets.
    - Calculations done using double-zeta plus polarization basis sets.
  - VASP
    - Vienna Ab-initio Simulation Package
    - Plane wave ab-initio molecular dynamics code
    - Projector augmented wave functions.
    - Calculations done using the PBE functional.
- Results were consistent across both codes.

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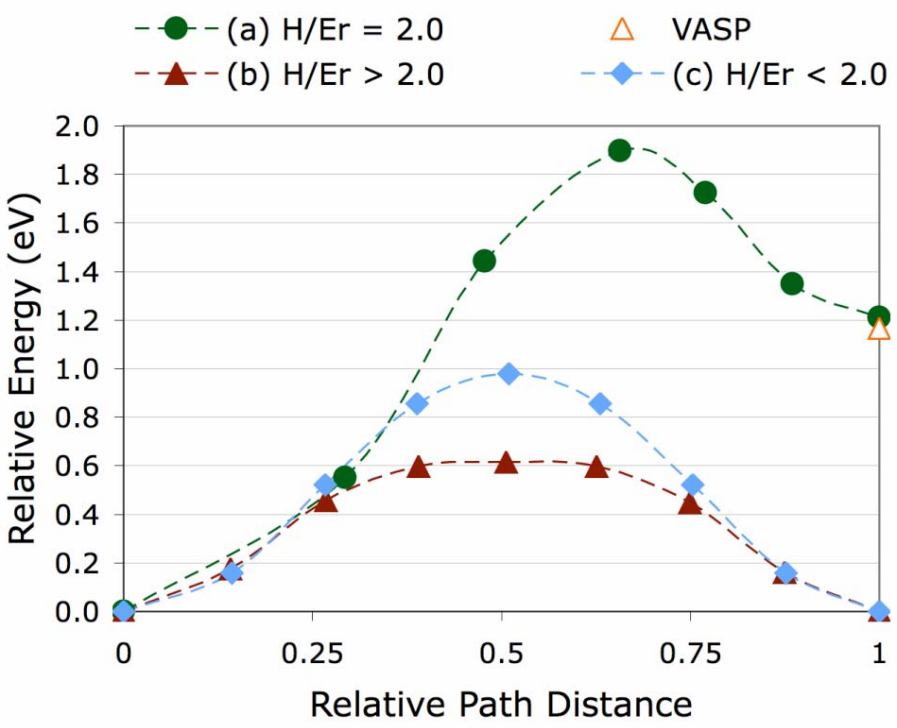
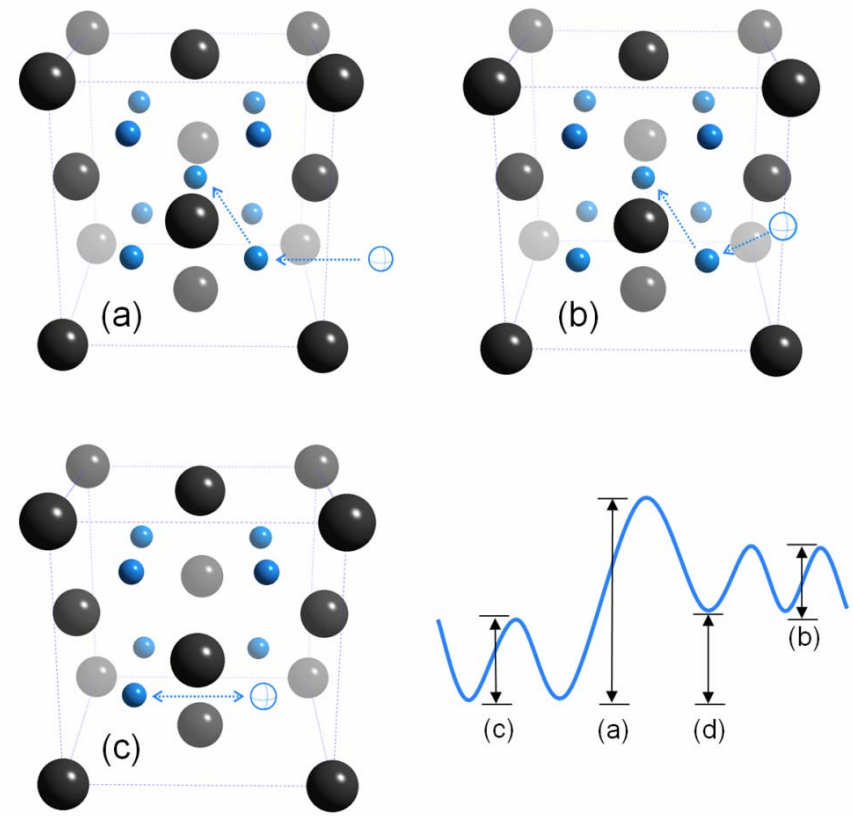
Hydrogen migration energies in ErH_{2+x}





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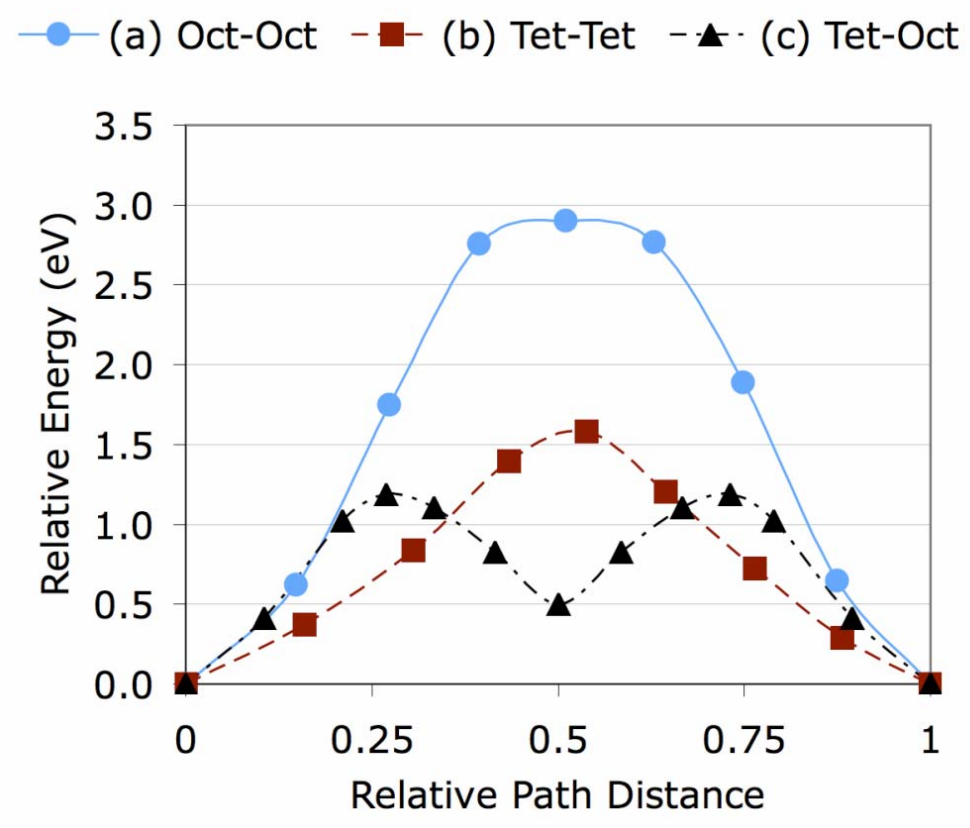
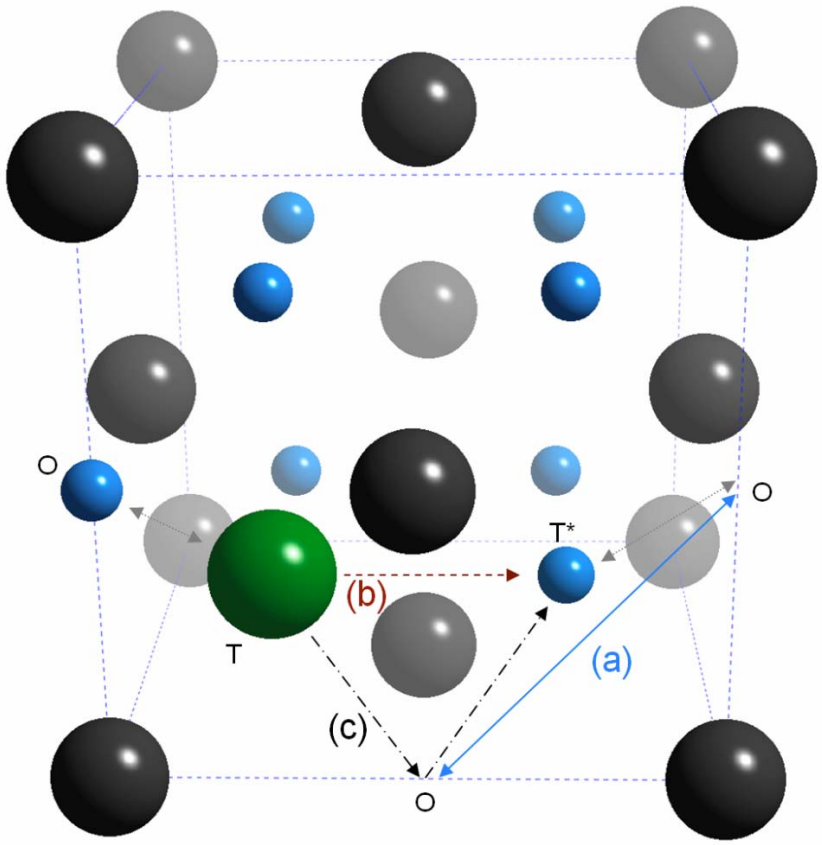
Oxygen migration in ErH_{2+x}





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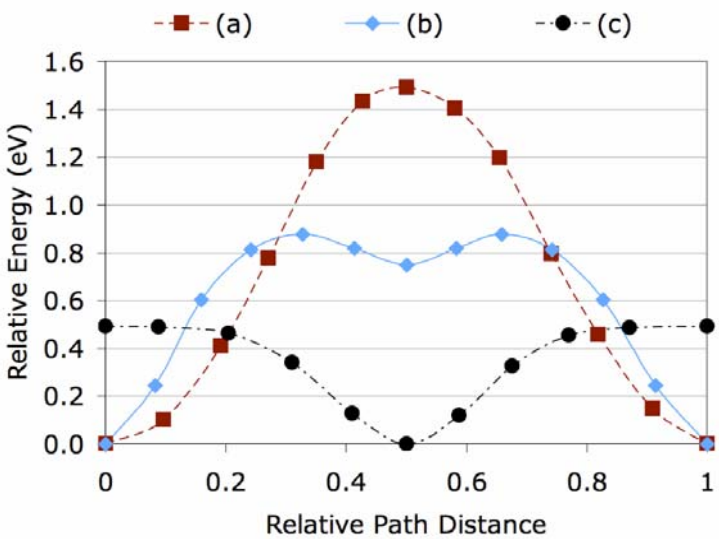
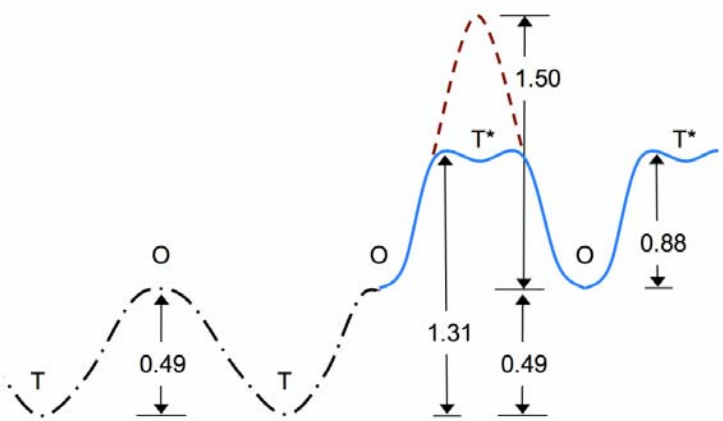
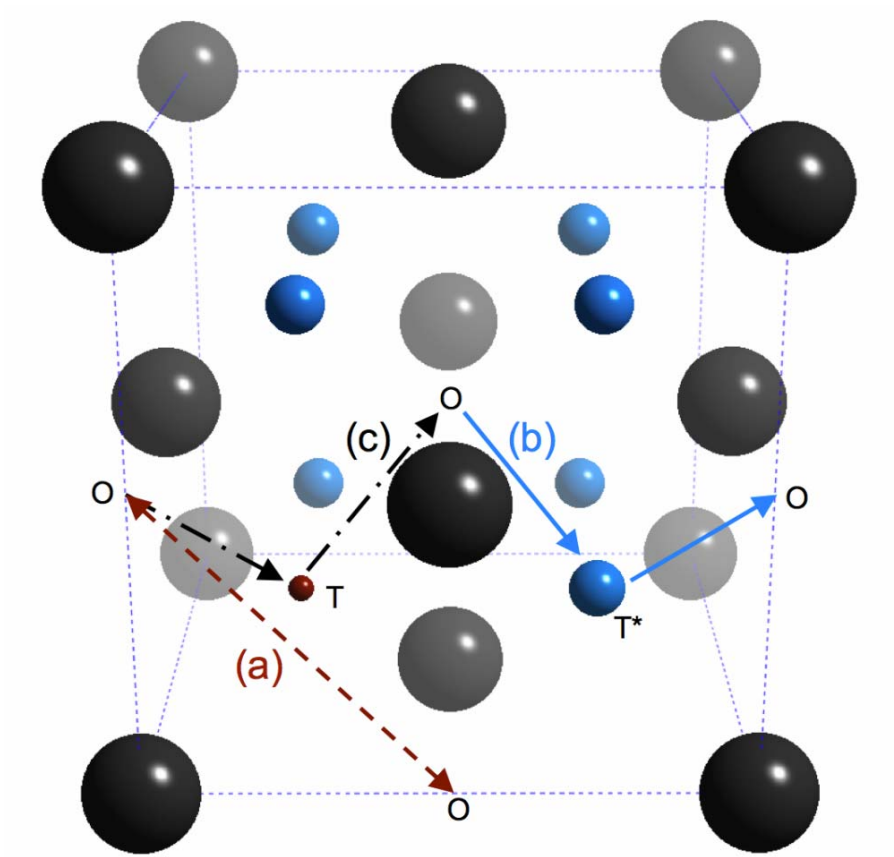
Helium migration in $ErT_{2-x}He_x$







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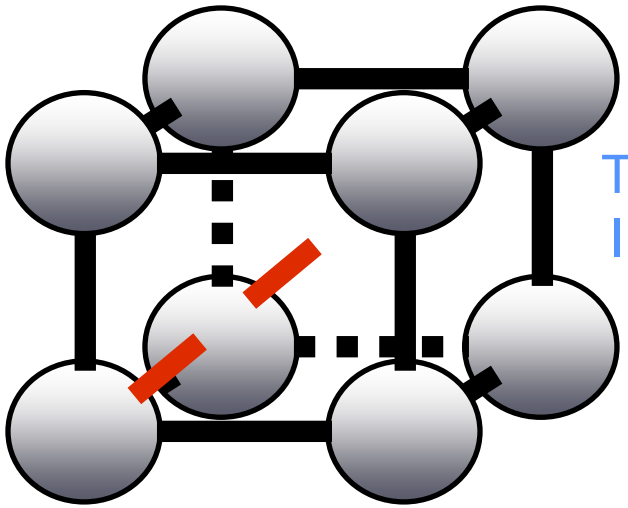
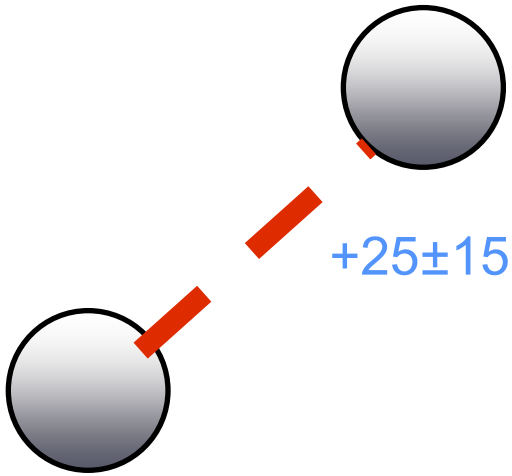
Hydrogen Vacancy Binding

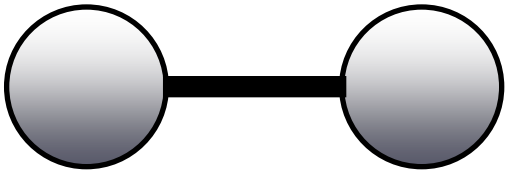
(Peter Schultz)



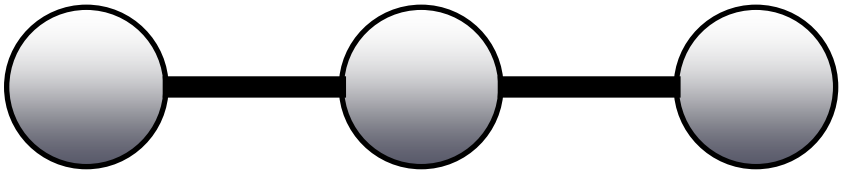
FCC
Tetrahedral
Interstitials



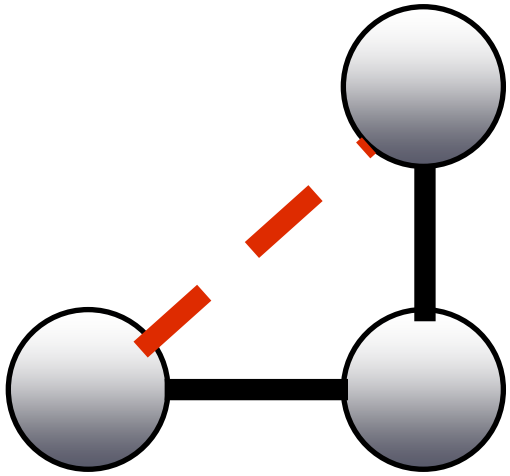
+25±15 meV



-70±40 meV



-175±50 meV



-100±70 meV

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Kinetic Monte Carlo Modeling

(Corbett Battaile)

- Kinetic Monte Carlo uses the inputs from the DFT calculations to calculate a probability of atomic motion.
- The atoms are all given the same statistical opportunity to move but only some will be successful.
- Can model millions of atoms moving simultaneously.
- Can follow the atoms for very long time scales, years in many cases.
- May be able to develop a model of helium release and tritium loss using KMC.

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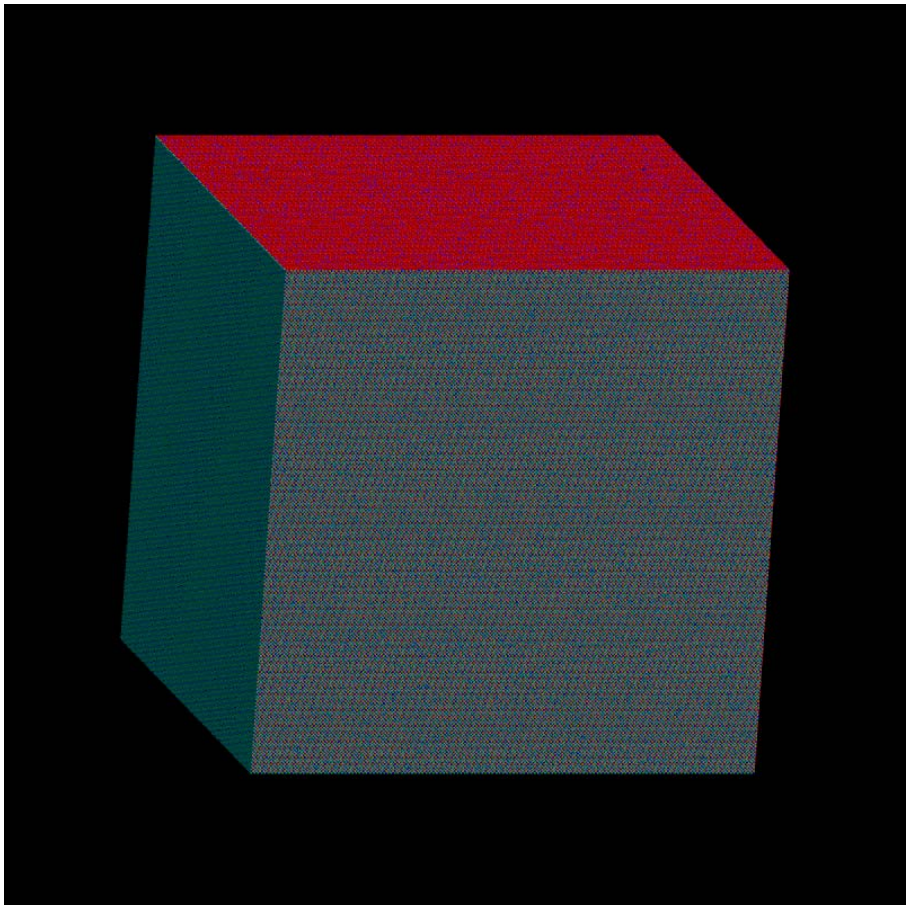
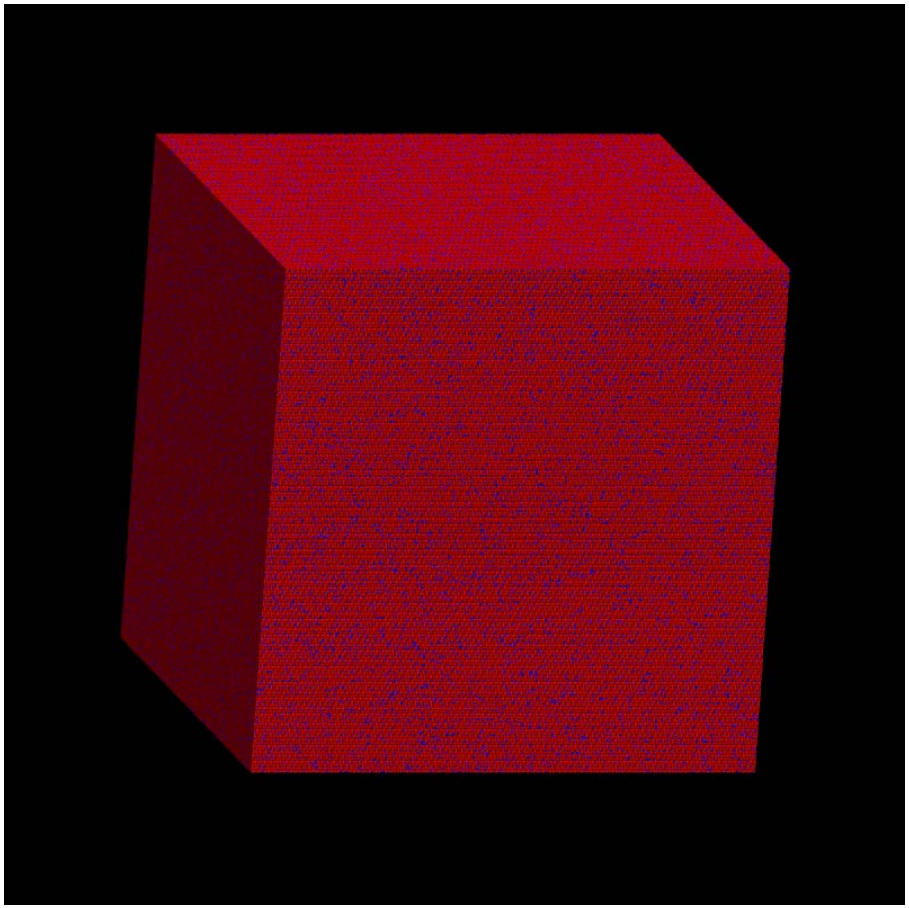
“Large” Massively Parallel Simulations





Gray = Er, Red = H, Blue = H_{Vac}

(144^3 unit cells = 47M+ lattice sites, using SPPARKS)

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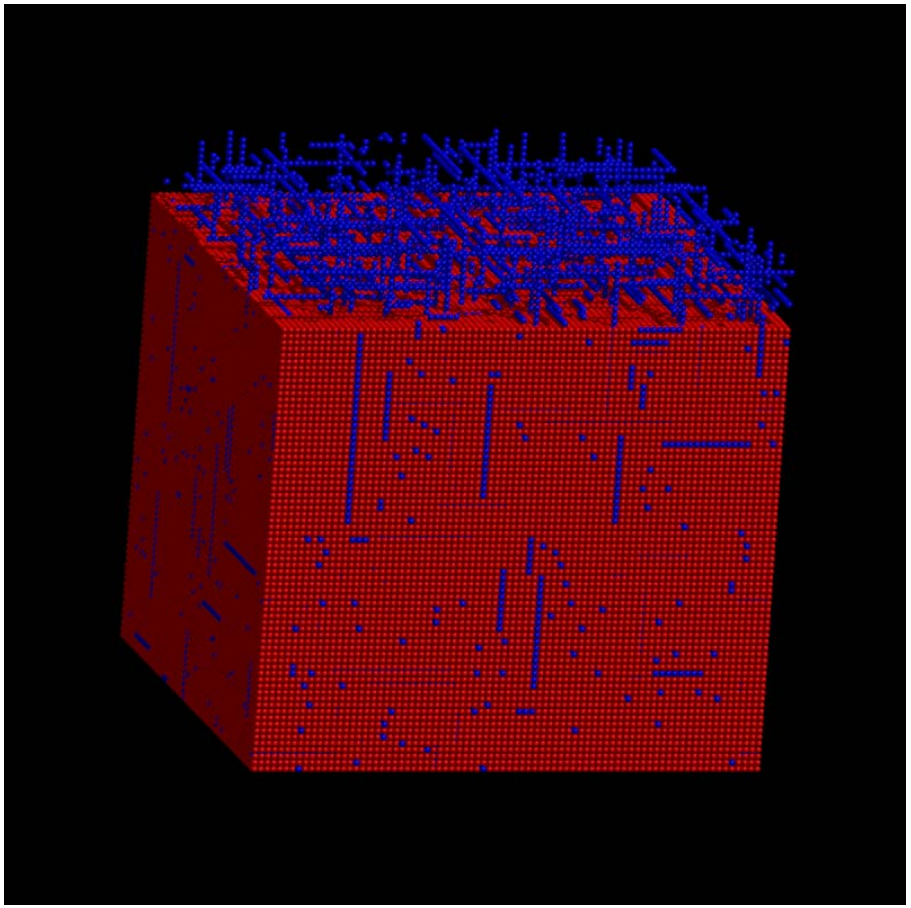
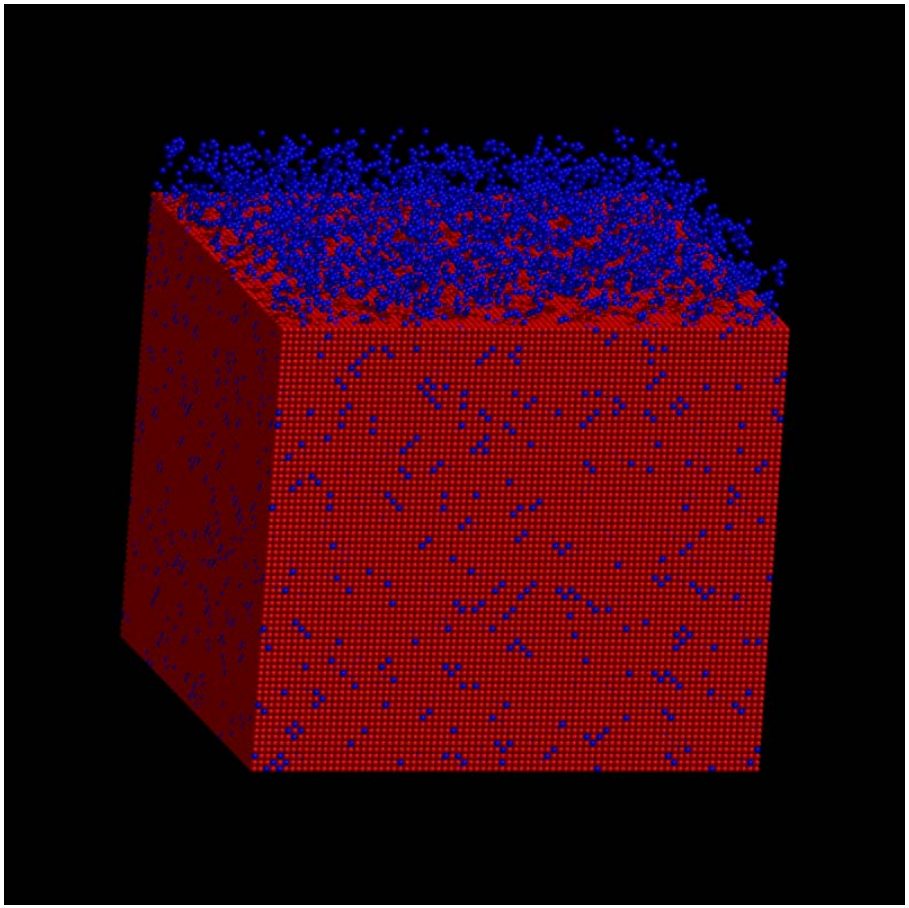
Hydrogen Bubble Morphologies



<110> Repels, <100> Attracts (per DFT)



<110> Attracts, <100> Repels

(40^3 unit cells = 1M+ lattice sites, using serial code)

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Calculations of Elastic Constants of Cubic Di-Hydrides

(Peter Schultz and Clark Snow)

Cubic $(X)H_2$	C11 (GPa)	C12 (GPa)	C44 (GPa)	K0-Cij (GPa)	K0 Experimental (GPa)
ErH_2 k-limit k=12^3	140(4) 145.4	63(2) 61.1	78(1) 78.5	89.1(.1) 89.2	97 +/- 4
ScH_2 k-limit k=12^3	166(5) 168.8	63(2) 61.7	80(1) 76.9	97.4(.2) 97.5	
YH_2 k-limit k=12^3	132(3) 132.6	58(2) 56.2	73(1) 73.6	82.8(.1) 82.8	90 +/- 7
LaH_2 k-limit k=12^3	95(2) 95.7	48(1) 47.5	53(1) 54.1	63.5(.1) 63.6	
TiH_2 k-limit k=12^3	119(1) 120.5	155(1) 154.3	-20(4) -21(2)	143.1(.1) 143.0	
ZrH_2 k-limit k=12^3	88(2) 89.0	157(1) 156.3	-30(4) -30(3)	133.9(.1) 133/9	115 +/- 7
HfH_2 k-limit[b] k=12^3	90(4) 81.5	178(3) 182.1	-40(10) -49	148.5(.1) 148.6	

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Calculations of Elastic Constants of Tetragonal Di-Hydrides

Tetragonal XH_2	C11 (GPa)	C12 (GPa)	C13 (GPa)	C31 (GPa)	C33 (GPa)	C44 (GPa)	C66 (GPa)	K0-C	K0-E
TiH_2 k-limit k=12^3	178(2) 178.1	149(2) 146.2	119(2) 120.9	120(2) 122.1	159(2) 155.5	59(2) 47.5	17(1) 18.7	140.2(.1) 140.0	n/c 139.8
ZrH_2 k-limit k=12^3	177(2) 181.0	143(2) 138.7	106(2) 106.9	107(1) 106.9	151(3) 149.4	144(4) 149.4	40(2) 35.4	130.7(.2) 130.5	n/c 131.1
HfH_2 k-limit k=12^3	190(2) 195.6	159(2) 154.8	118(2) 117.4	118(2) 117.4	175(4) 175.3	76(6) 66(3)	49(2) 44.0	146.3(.2) 146.3	n/c 147.0

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Conclusions

- Sandia is actively involved in helium release research.
- We have mapped the growth and size of helium bubbles in $ErT_{2-x}He_x$
  - can be used for Model Verification and Validation
- First-principles calculations have provided insight into migration and site occupancy of:
  - Hydrogen
  - Oxygen
  - Helium
  - Elastic Constants of many di-hydrides
- Building a larger scale model using the Kinetic Monte Carlo method:
  - Millions (billions) of atoms
  - Long time scales up to years

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