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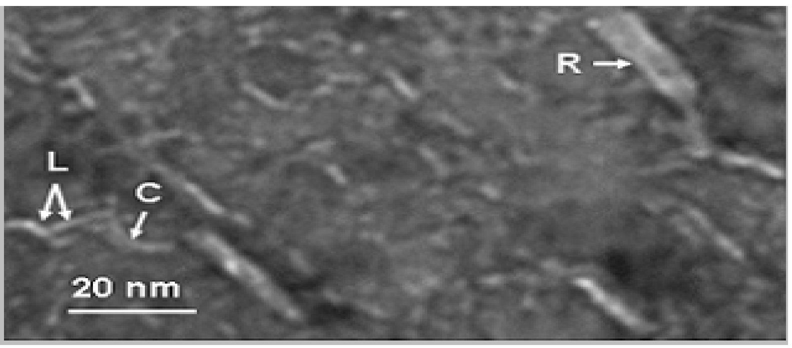

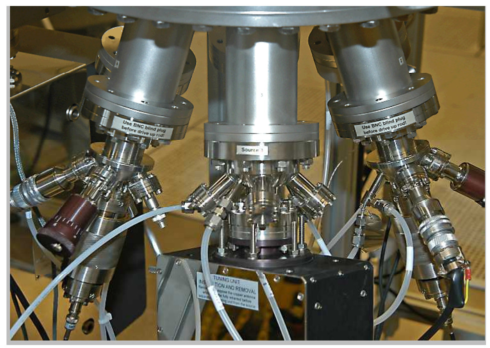

Tritium and He-3 in $ErT_{2-x}He_x$

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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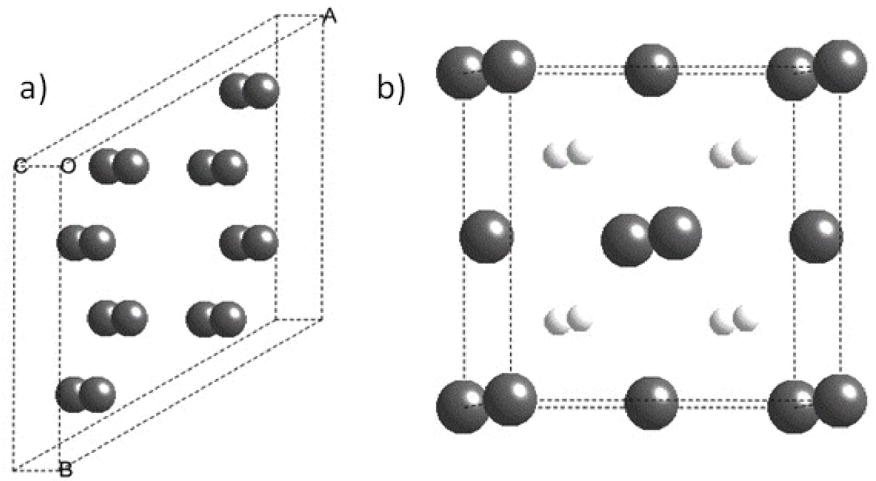
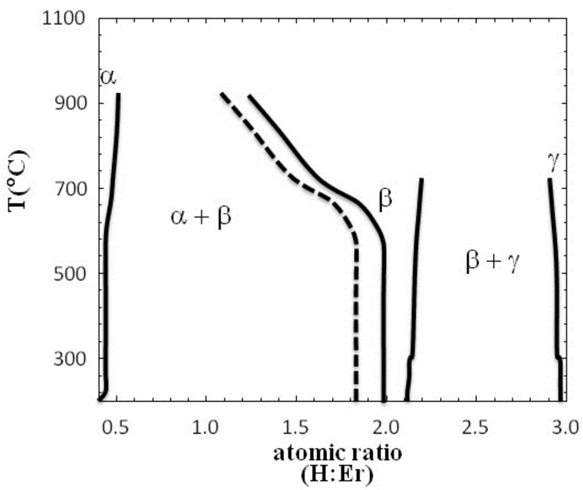
Properties of the Er:T system

- Erbium = HCP
- ErT_2 β-phase = FCC
- β-phase extends from 2.0 - ~2.2.
- Sub-stoichiometric β-phase due to stoichiometric deficiency.
  - We often observe 1-2% oxygen as large Er_2O_3 chunks and as nano-clusters.
  - Other impurities like other RE's





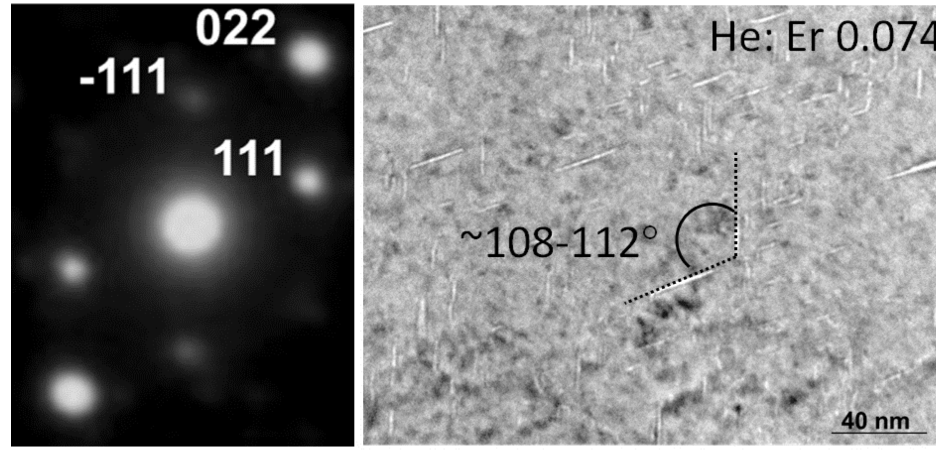
Helium Bubble Shape

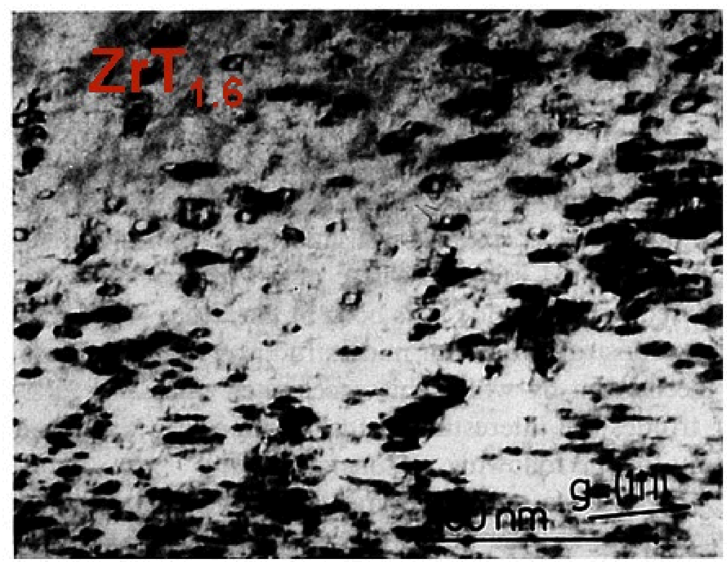
- Helium stored in platelets oriented along (111) planes.
- 4 (111) planes in FCC, only observe 2 at a time in TEM.
- Width ~ 1-2 nm.
- Platelets v. Spheres

$$\text{Ratio of } \frac{Surface\ Energy}{Strain\ Energy} = \frac{2\gamma}{\mu b}$$

> 0.1 Sphere
< 0.1 Platelet

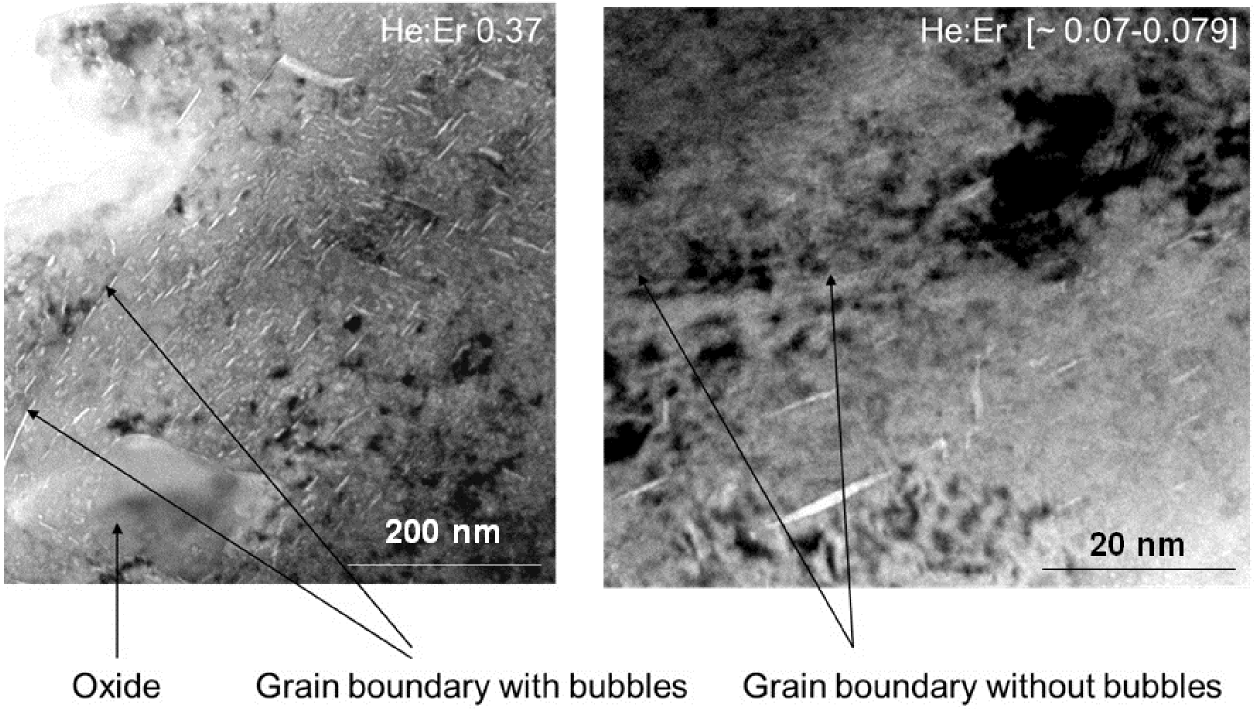
ErT_2 ~ 0.06
ZrT_2 ~ 0.26





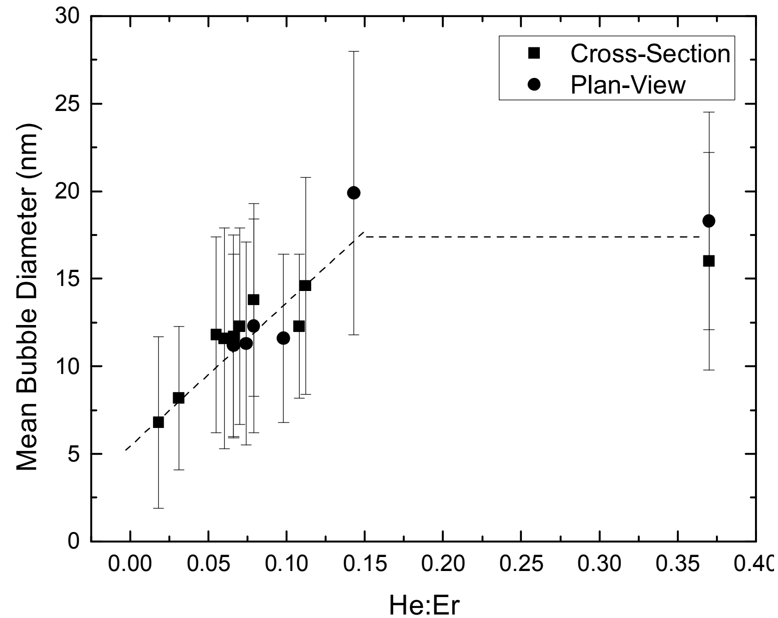
Helium Bubble Spatial Distribution

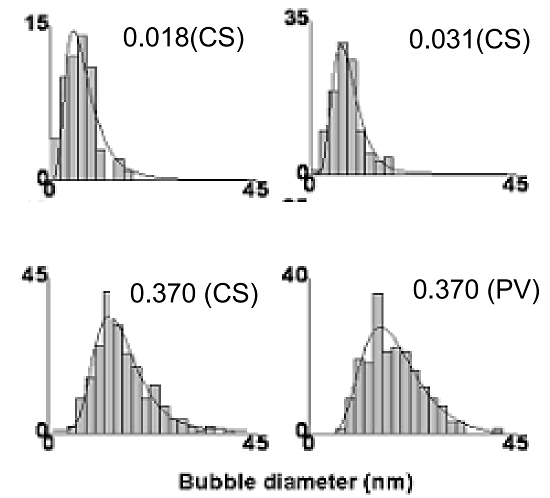
- Bubbles observed evenly distributed throughout film.
- Grain Boundary decoration only when GB aligns along (111) plane
- Bubbles observed around Er_2O_3 pieces.



Helium Bubble Growth and Interactions I

- Length increases with time up to He:Er ~ 0.15.
- Width doesn't change until He:Er ~0.15.
- Size distribution log-normal throughout life.
  - Tight distribution early
  - Larger distribution later

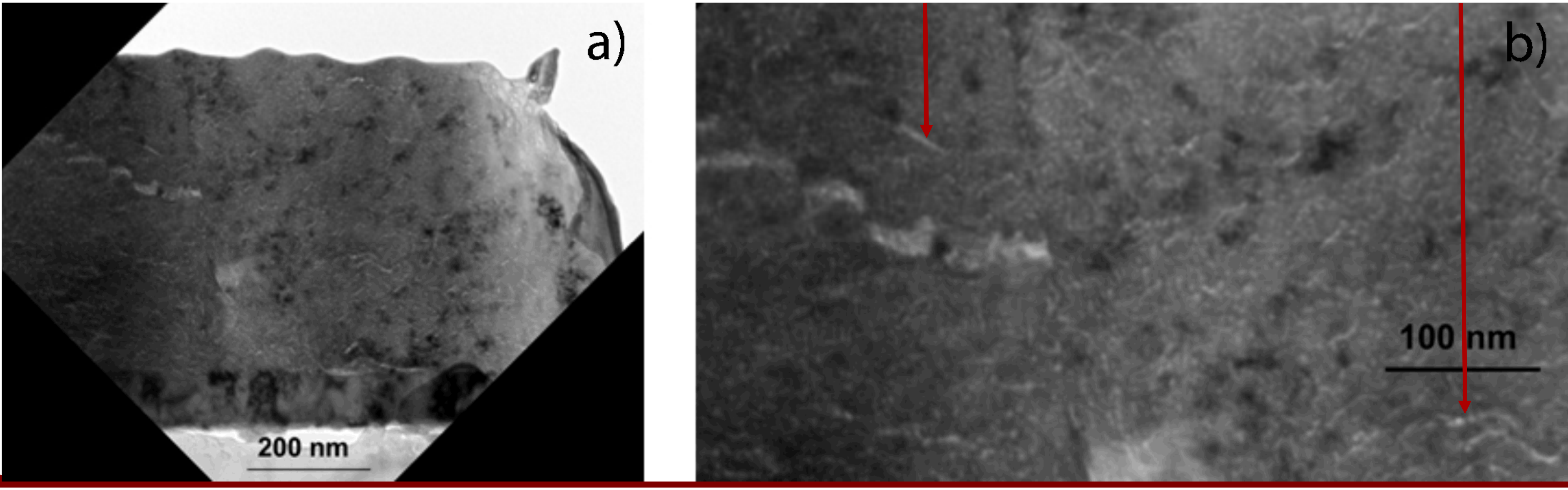




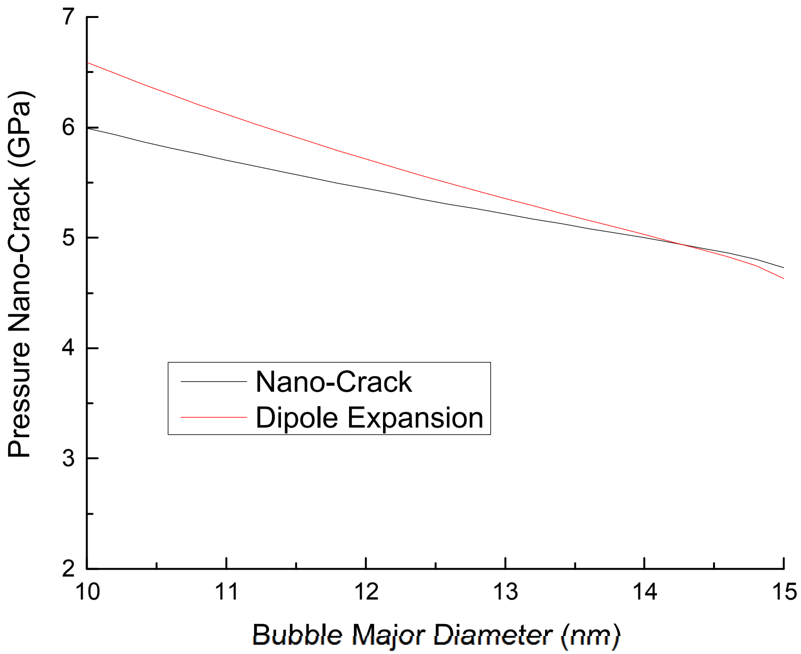
Bond et al., J. Appl. Phys. 107, 083514 (2010)

Helium Bubble Growth and Interactions II

- Bubbles begin to link later in life.
- Length stops growing, width begins to increase.
- Becomes very difficult to even define what is a bubble.



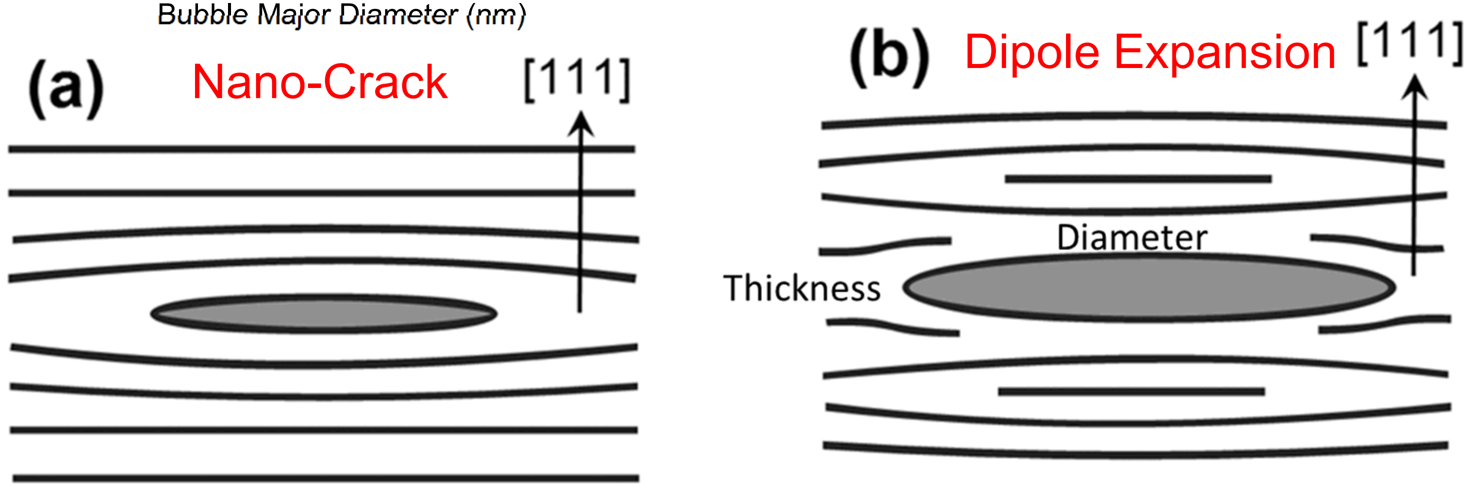
Helium Bubble Transition Point



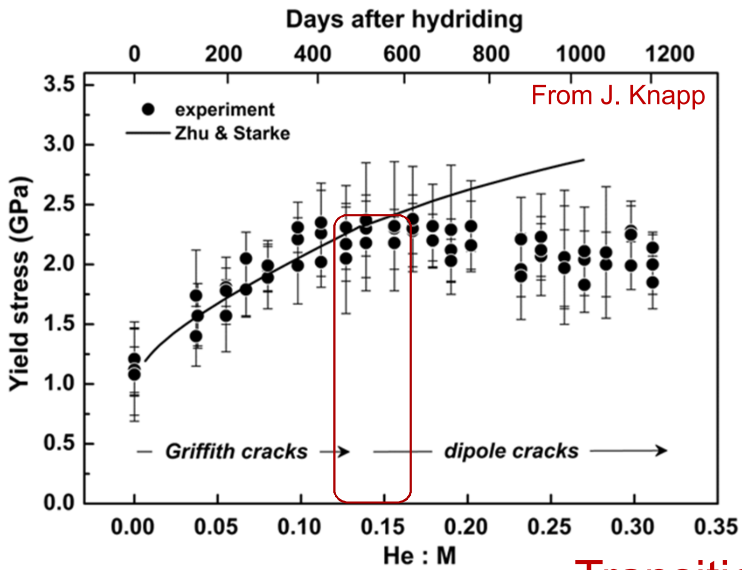
$$P_{Nano\_Crack} = \frac{\pi G m}{2(1-\nu)} \frac{s}{d}$$

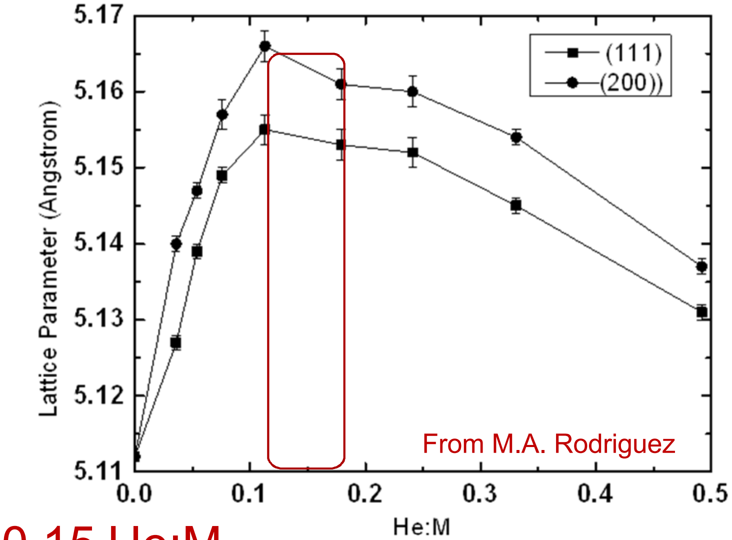
$$P_{dipole} = \frac{2\gamma}{s} \frac{(d+b+s)}{(d+b)} + \frac{G_m d_{111}}{(d+b)}$$

- γ = Surface Energy
- ν = Poisson's ratio
- d = platelet diameter
- s = platelet thickness
- b = Burger's vector
- d_{111} = 111 plane spacing
- G_m = effective Shear modulus

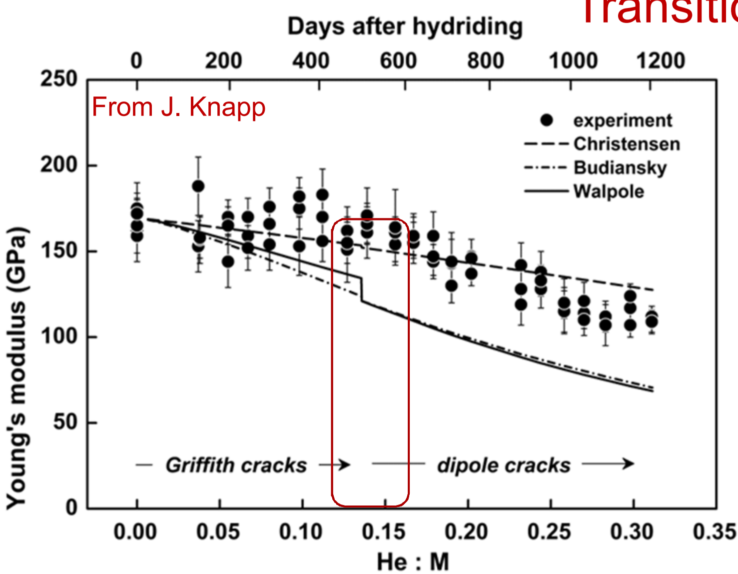


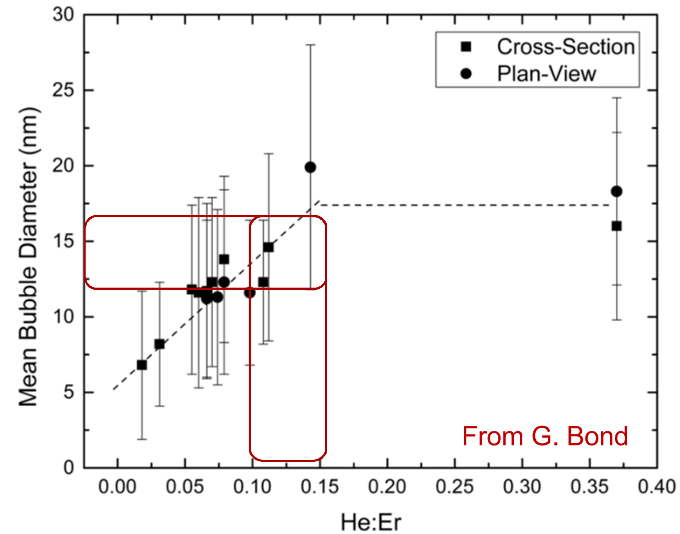
Evidence for Bubble Growth Model





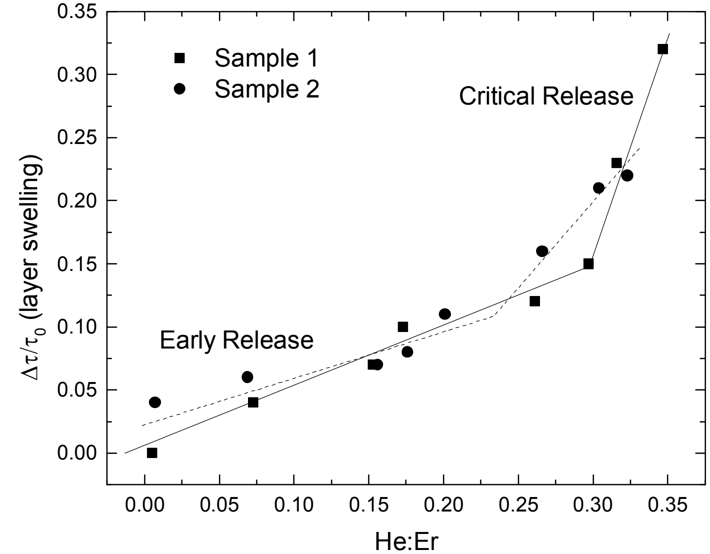
Transition ~ 0.12-0.15 He:M

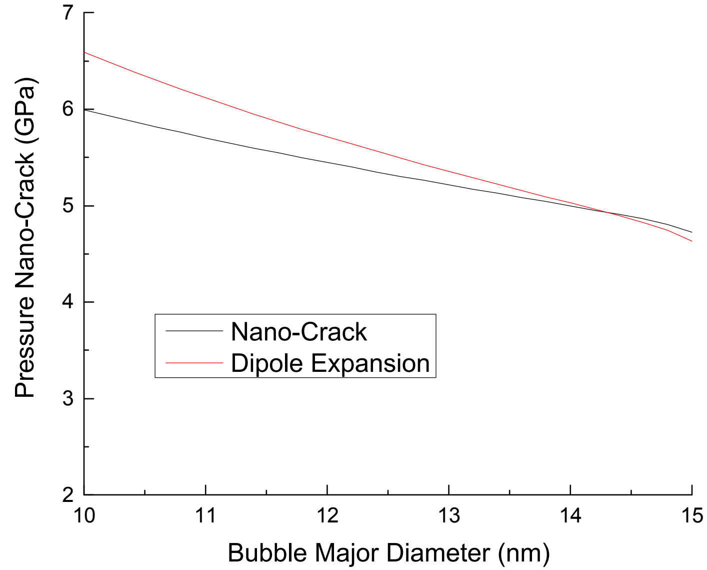




Helium Bubble Pressure

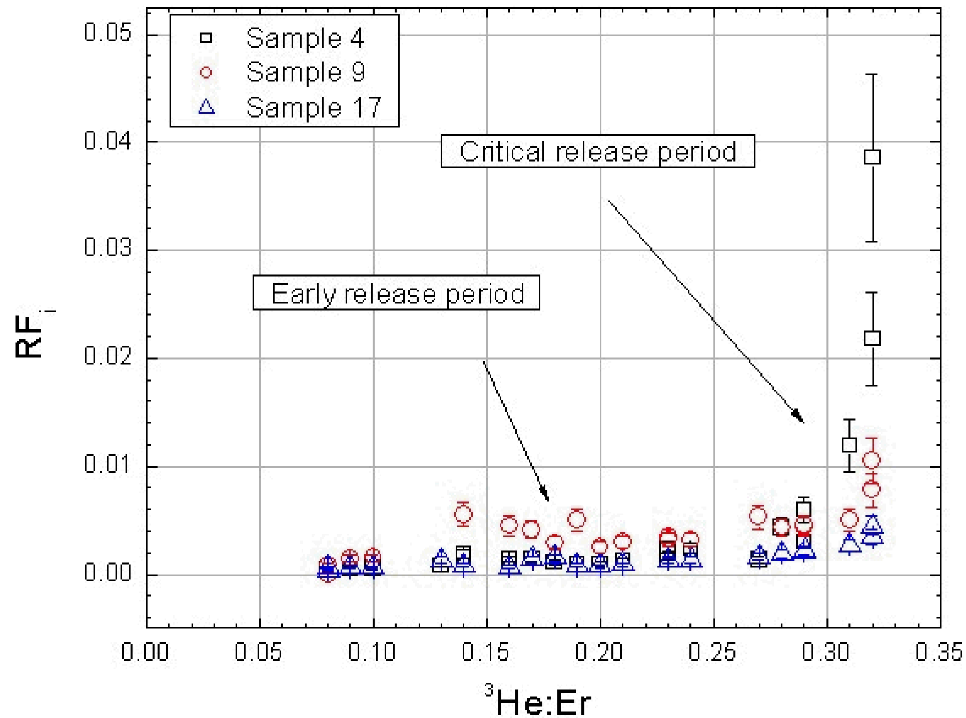
- Pressure in bubble
- $$\frac{\Delta V}{V} = \frac{c_{T0} t \lambda \Delta v T H_e}{\Omega}$$
  - $\sim cT_0\, \lambda\, t\left[\left(\frac{v_{He}}{\Omega}\right) * \left(\frac{\Delta v I}{\Omega}\right) - \left(\frac{\Delta v T}{\Omega}\right)\right]$
  - Ω = atomic volume (volume of the tritide per metal atom)
  - v_{He} =volume required by 3-He in the high pressure bubbles
  - $\Omega = \Omega_0[1 + cT(\Delta v/\Omega_0)_T]$
  - Using EOS for 3-He can extract bubble pressure
- Using Neutron Reflectivity to measure swelling P ~1-3 GPa
- Models predict 5 GPa

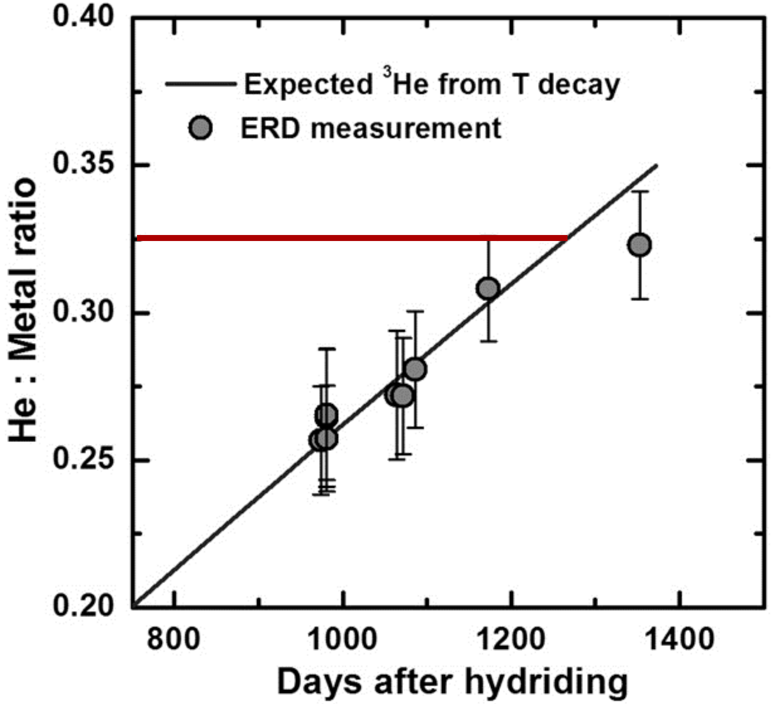




Helium Release

- Early life helium storage ~100%.
- Critical release occurs at He:M ~0.33



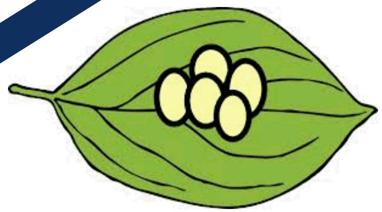


Conclusions and Further Questions

At ~0.33 He:M
“critical release”
then it dies

Nano-cracks

At 0.14 He:M growth mode transitions from nano-crack to dipole-expansion

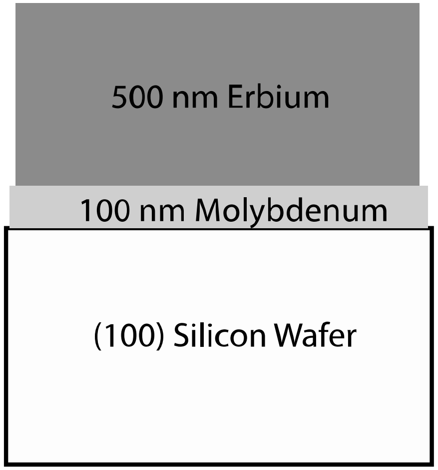


Don’t know how the eggs get there.

Question: Is the self-trapping mechanism appropriate for stoichiometric metal tritides?

Overview of this Study

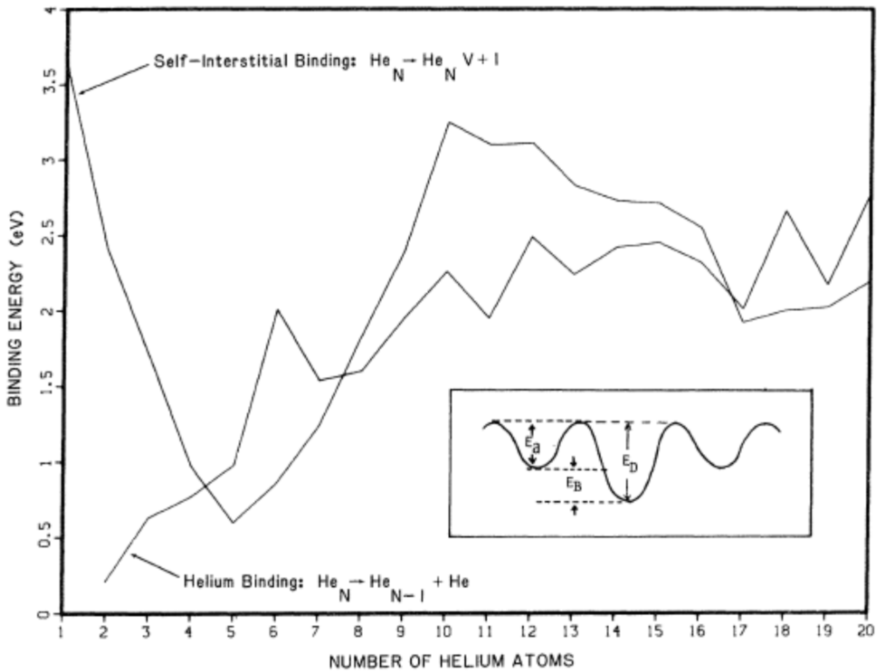
- 500 nm thick Erbium film deposited via e-beam PVD on Silicon wafer with Molybdenum interaction barrier.
- Expect 10-15% swelling upon conversion to ErT_2.
- Average stoichiometric deficiency of δ ~ 0.1.
- TEM to image bubbles
- XRD for lattice changes
- Nano-Indentation for mechanical property changes
- IBA/ERD for helium retention



Load Run	T:Er
1	1.844
1	1.927
2	1.842
2	1.987
3	1.851
3	1.909
Average	1.893
Std. Dev.	0.058

Helium Bubble Nucleation

Self-Trapping in Nickel



Wilson et al. PRB 24, p.5616 (1981)

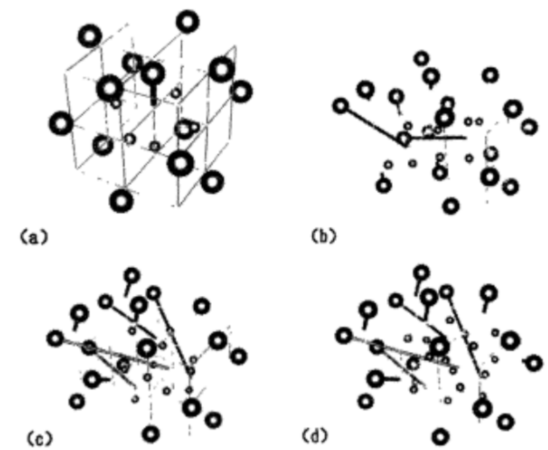
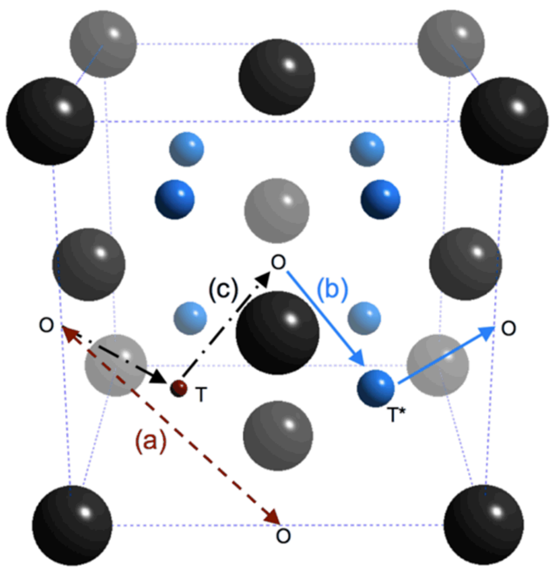
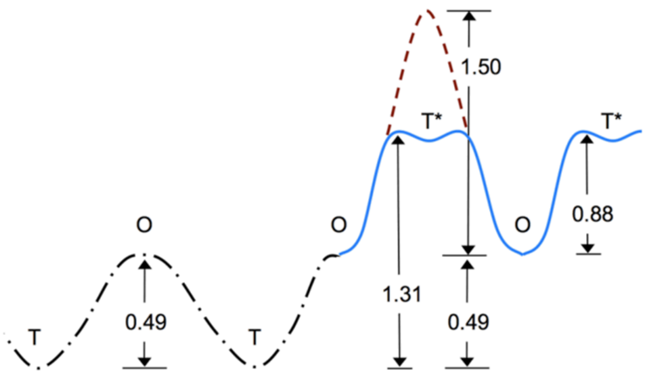
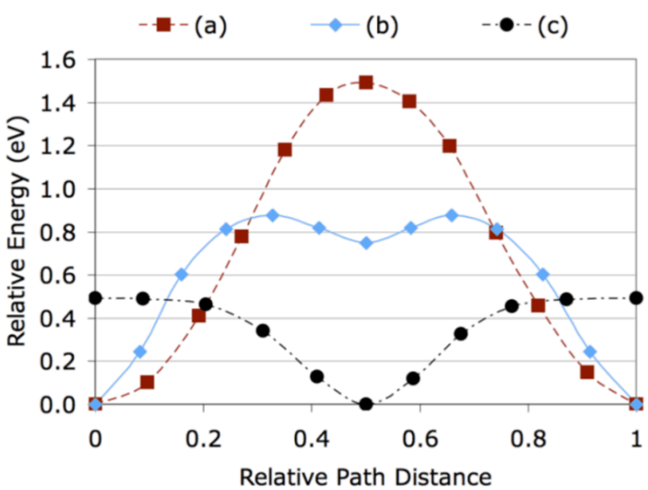


FIG. 2. Computer-drawn sketches of the minimum-energy configurations of (a) $He_5 \rightarrow He_5 V^* I^*$; (b) $He_8 \rightarrow He_8 V_2^* I_2^*$; (c) $He_{11} \rightarrow He_{11} V_7^* I_7^*$; and (d) $He_{16} \rightarrow He_{16} V_{10}^* I_{10}^*$ (see the Appendix) without introducing initial vacancies into the calculation. Near-Frenkel pairs are denoted $V^* I^*$.

Helium Diffusion in ErT_2







1. Helium migrating along path (a) represents direct Octahedral to Octahedral motion, this path requires ~1.5eV to overcome the energy barrier and move.

2. Helium migrating along path (b) represents an Octahedral-Tetrahedral-Octahedral path but with a hydrogen atom residing in the Tetrahedral space.

    - Helium motion along this path requires ~0.9eV to move to the tetrahedral site which itself is pseudo-stable as shown by the small decrease of energy in the tetrahedral site.

3. Helium migrating along path (c) represents an Octahedral-Tetrahedral-Octahedral path with an unoccupied tetrahedral site.