Hydrogen and Helium Isotopes in Materials Conference
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"In-situ Monitoring of Deuterium Site Occupancy in Erbium Deuteride Via Time-Of-Flight Neutron Diffraction Analysis"

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Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Overview

- Introduction
- Experimental
- Results
  - Overview of β phase (fluorite) formation
  - D_2 overpressure
- Summary

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Research objectives

- Research goal: to obtain a scientific and technical basis for understanding tritium decay in Er-tritide.
  - ^{3}He Bubble formation and growth
  - Influence of Microstructure on ^{3}He bubble retention
  - Structural analysis of ErT_2 phase
    - Lattice parameter values
    - Formation of fluorite phase
    - Tritium site occupancy in fluorite

Focus of this presentation

Use D as surrogate for T

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

We would like to understand the loading process of (D,T) as Er metal converts to the fluorite lattice.

- We want to probe the Er (α) and $Er(D,T)_2$ (β) phase while they form.
- We only used D_2 in this experiment.
  - What pressure / temperature conditions generate fluorite?
- D scatters well for neutron diffraction experiments.
- Loose powders are more random, providing straightforward analysis.

- We chose to analyze Er metal powder as it is loaded with D_2 and simultaneously collect neutron scattering data to observe in-situ changes in structure during the fluorite phase formation.
  - Gain insight into site occupancy without hazards of tritium

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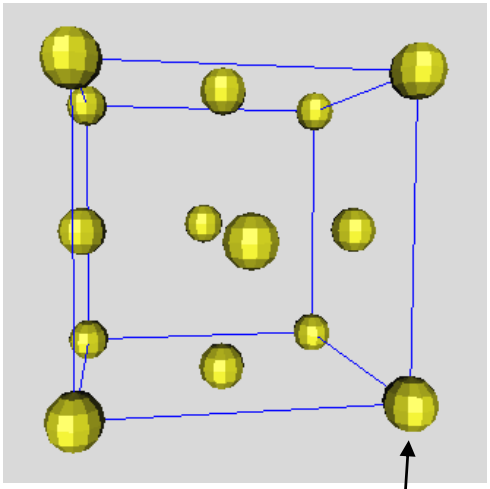
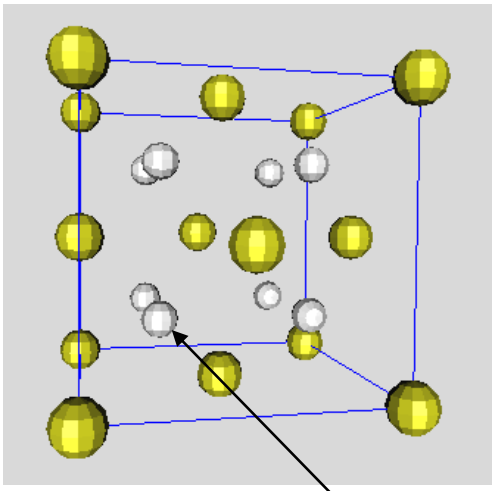
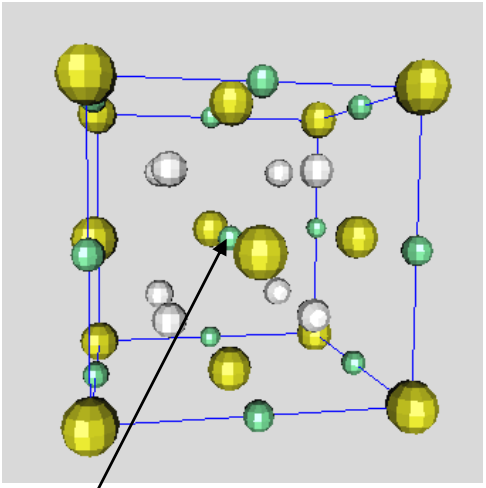
In particular, we wanted to see what happens to the tetrahedral (D_{tet}) and octrahedral (D_{oct}) site occupancy upon loading



FCC lattice of Er



D occupies tetrahedral (¼, ¼, ¼) sites



Octahedral (½, ½, ½) sites could also contain D*

* T. J. Udovic, J.J. Rush, and I. S. Anderson, *Phys. Rev. B,* **50,** pp.7144 (1994)

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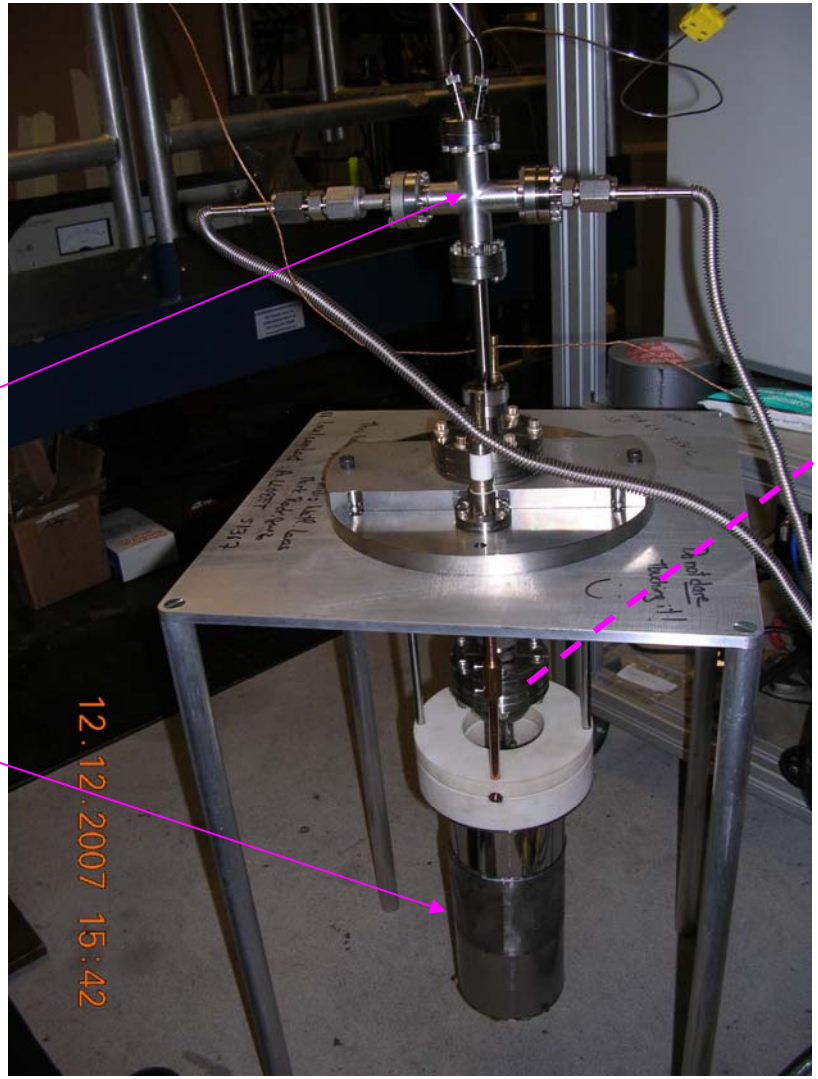
Experimental reactor was designed for safety and flexibility

Reactor setup loads into HIPD chamber for neutron scattering

D_2 gas / vac multiplexer

Vanadium heating element and shielding



TC

Reactor Material: Fused silica

Er powder

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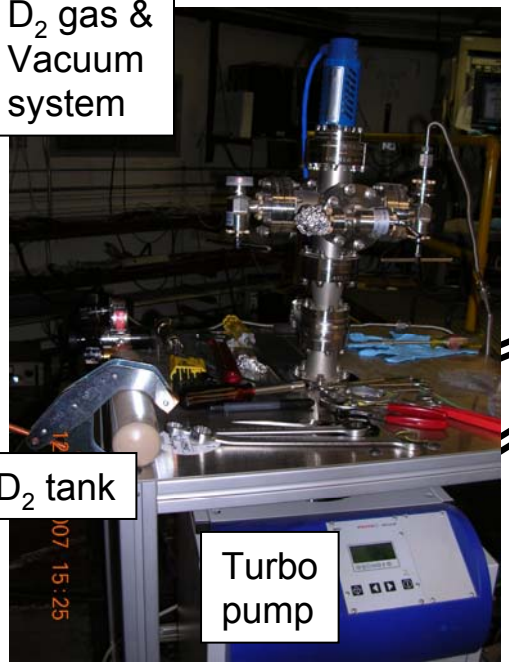
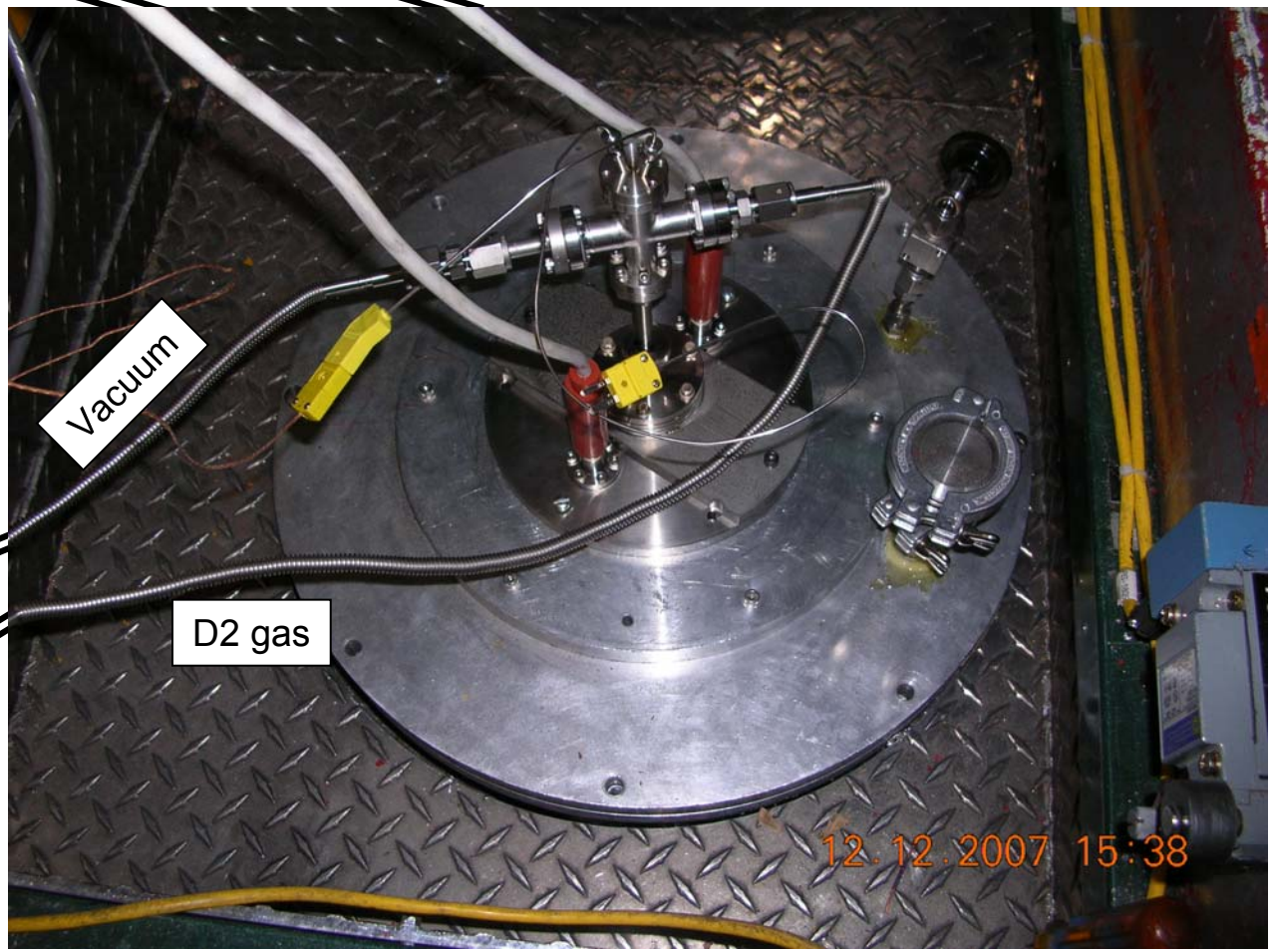
Illustration shows setup of reactor on HIPD beamline at LANSCE





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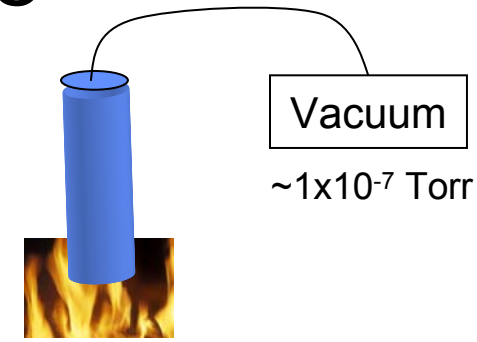
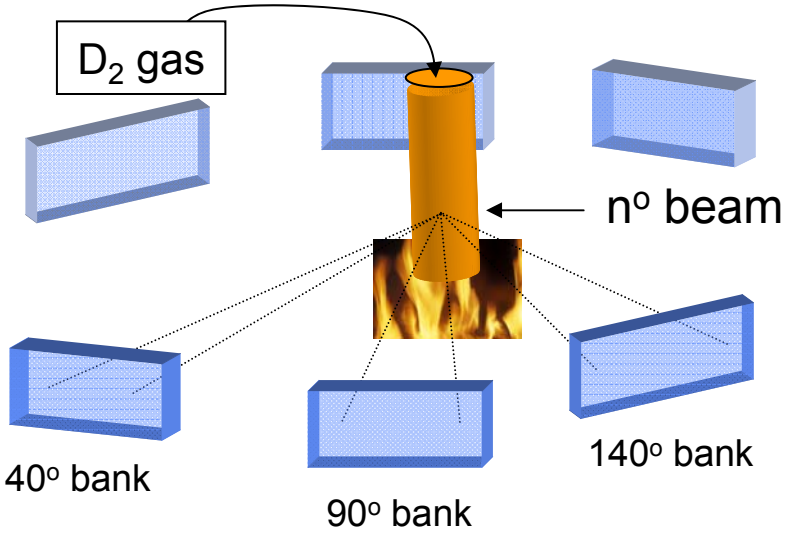
Experimental protocol for the reported results collected at 450°C

Heat Er powder to loading temp in vacuum



Hold at temperature and add D_2 gas:
Pressures: 0.1, 1.0, 10 Torr

Monitor structure as Er → ErD_2 fluorite



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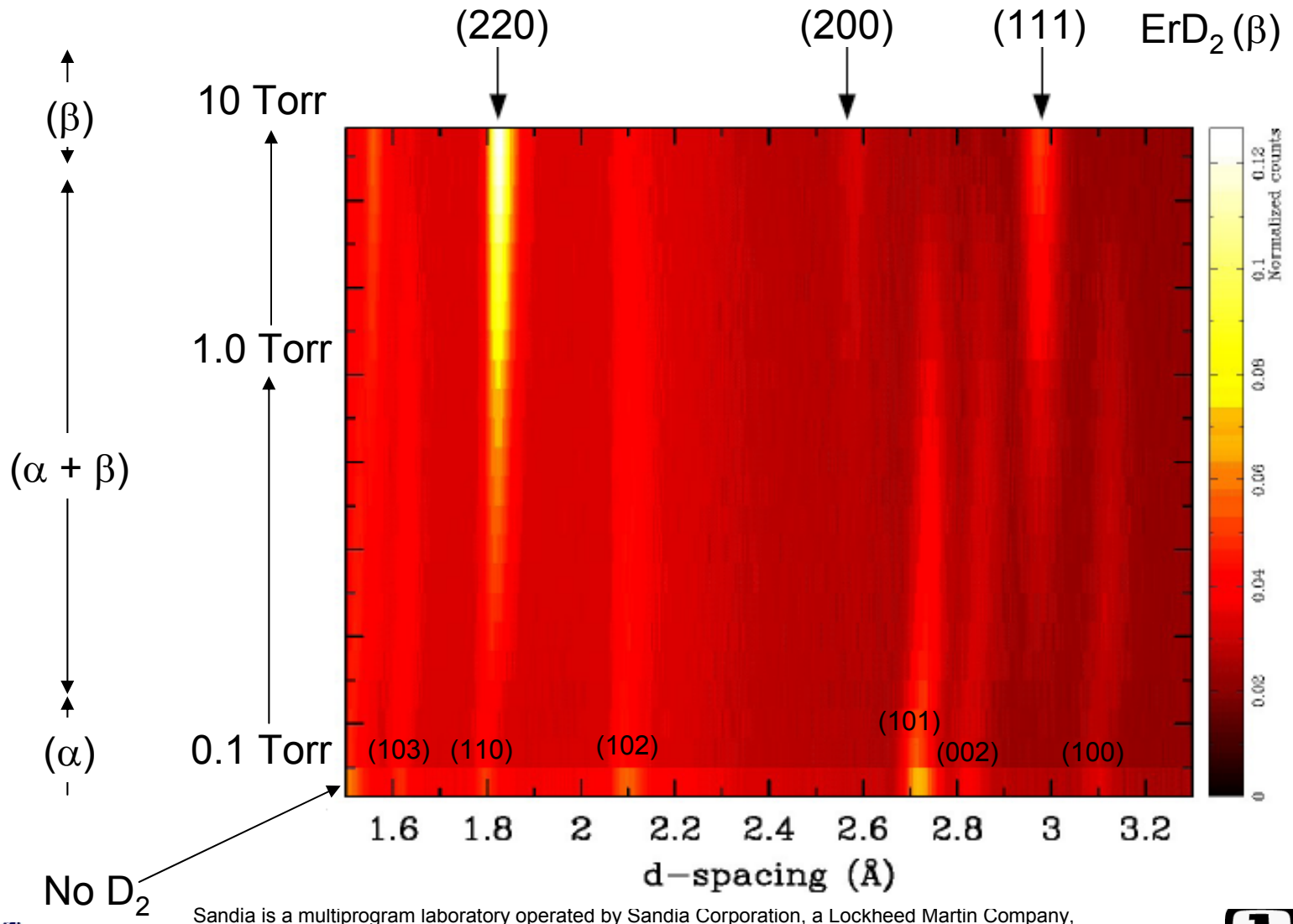
In-situ monitoring of Er loading at 450°C shows conversion to ErD_2 at ~1 Torr



450°C

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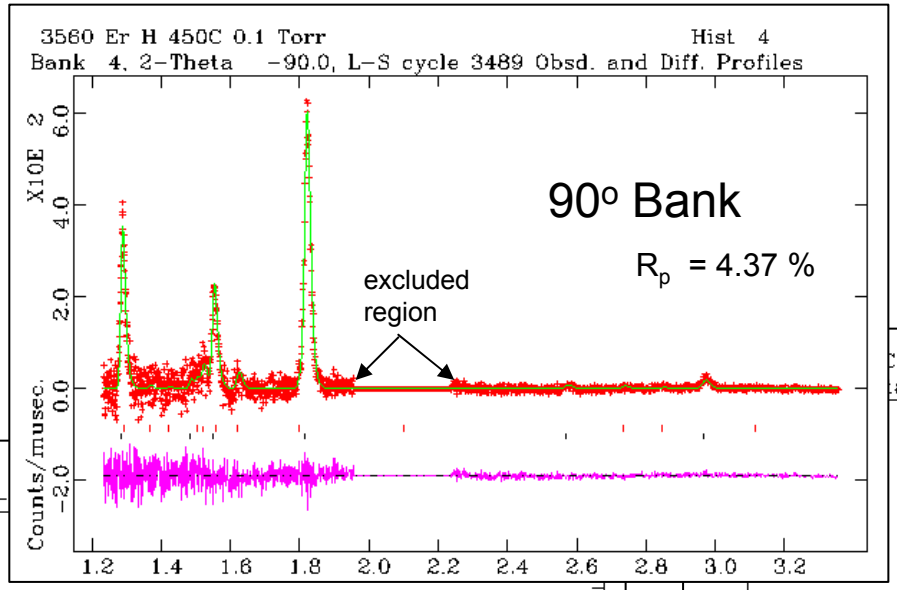
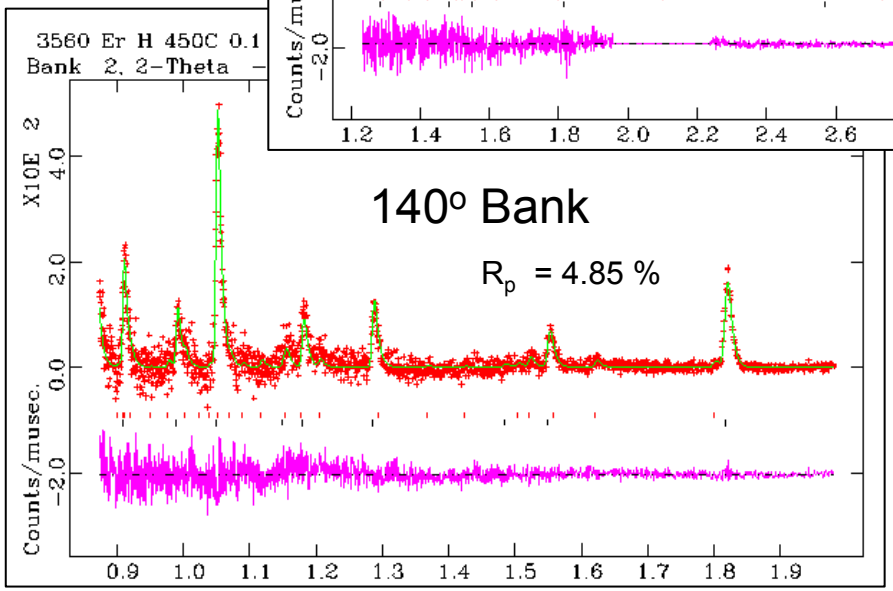
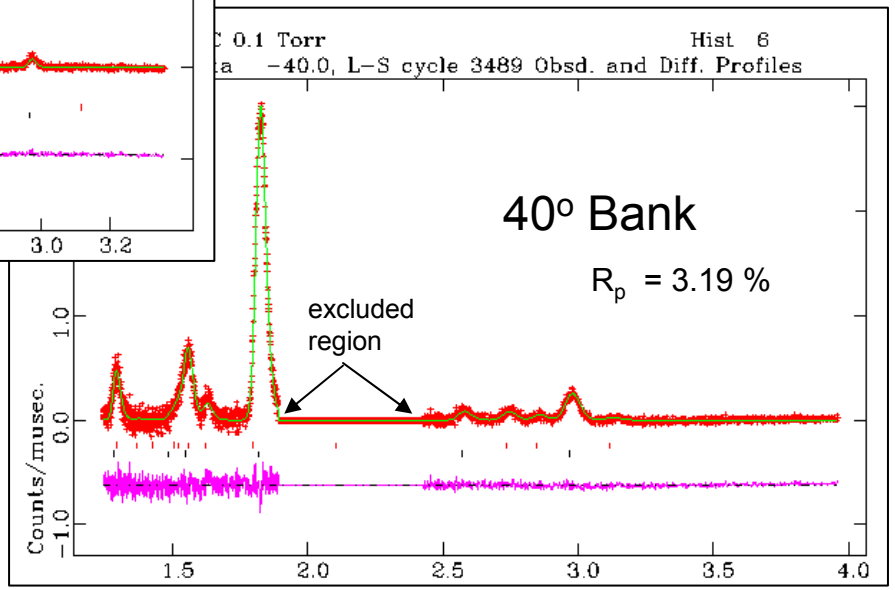
Rietveld structure refinements of observed histograms showed good modeling of α and β phases with low residuals

Run 3560
450°C
0.1 Torr
χ^2 = 1.128



Excluded region removed
scatter from glass Rx chamber





d-spacing (Å)

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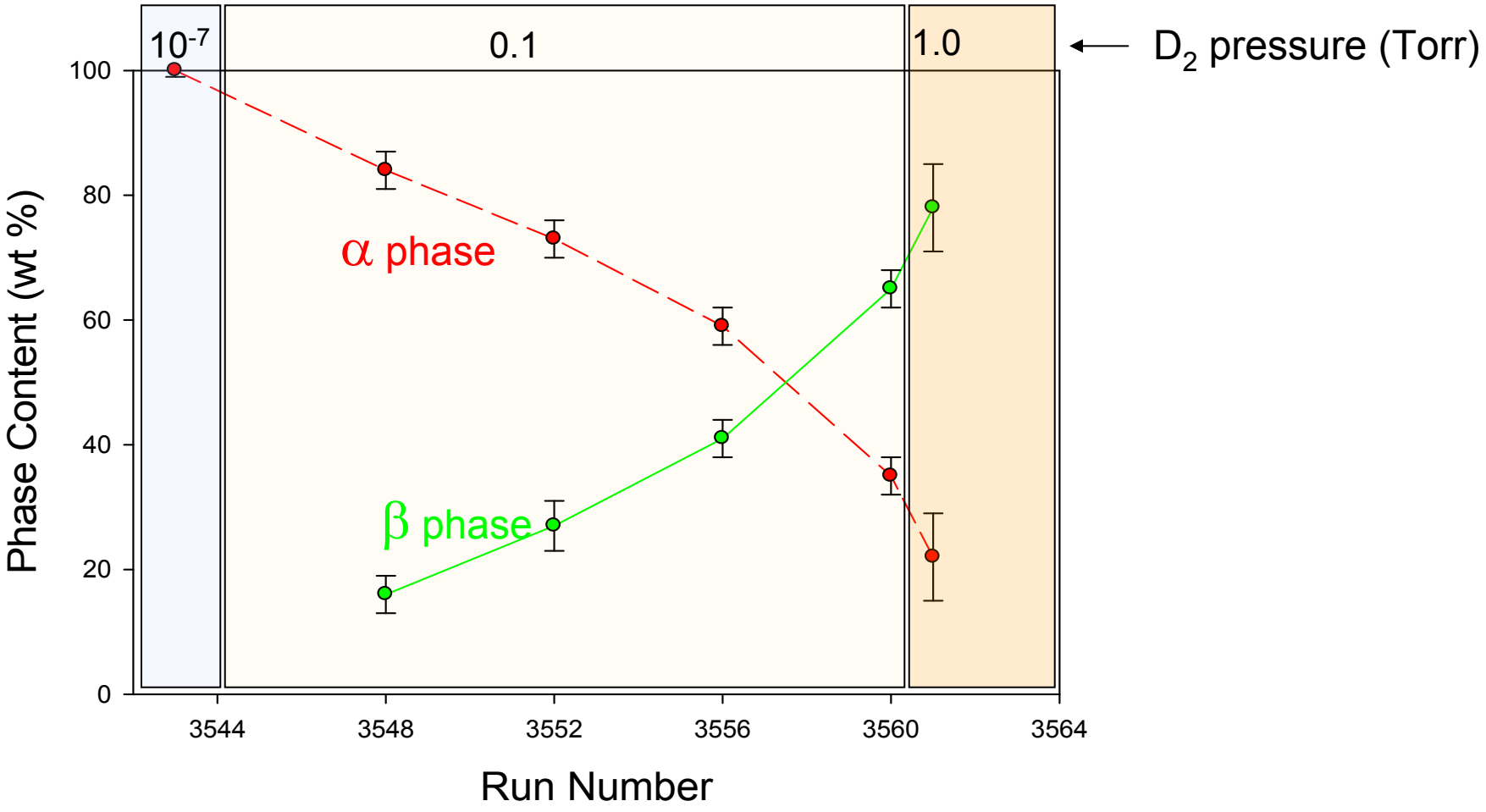
Structure refinements yielded results concerning content of α vs. β phase during D_2 loading



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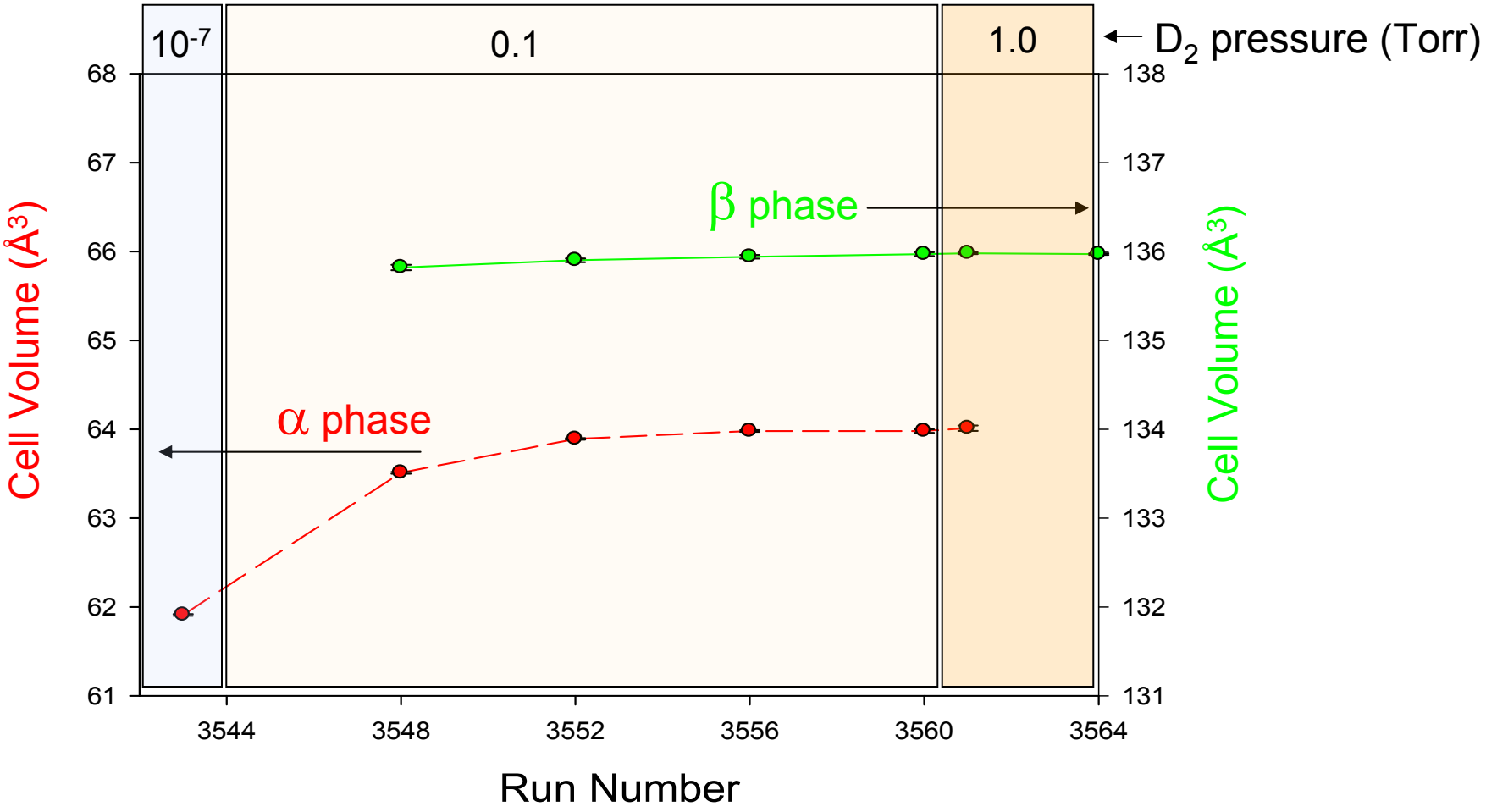
We observed a dramatic volume expansion for the α phase upon D_2 exposure. The β phase shows little change in cell volume.



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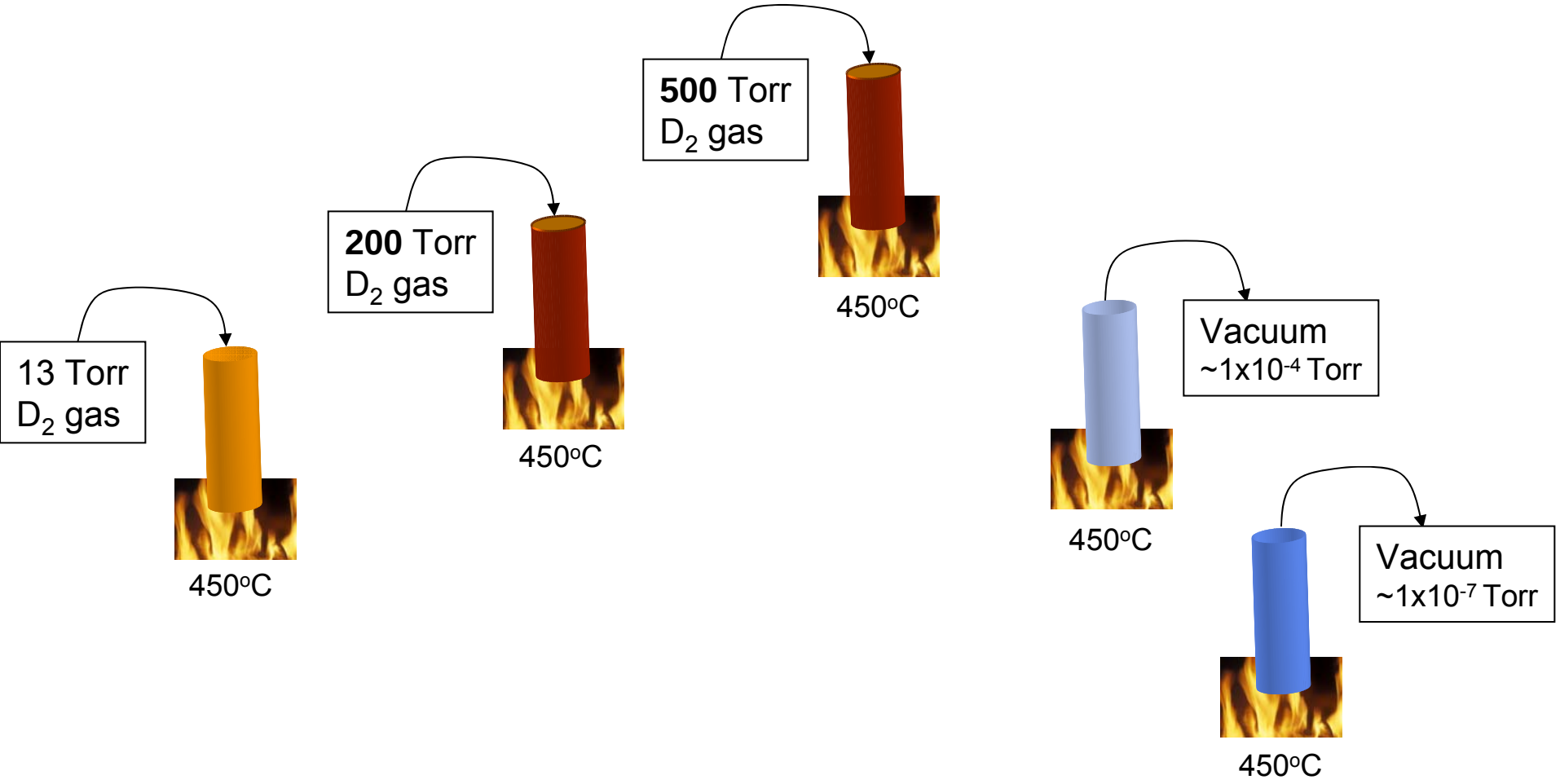
We monitored the structural changes in the β phase at 450°C with varying D_2 overpressures



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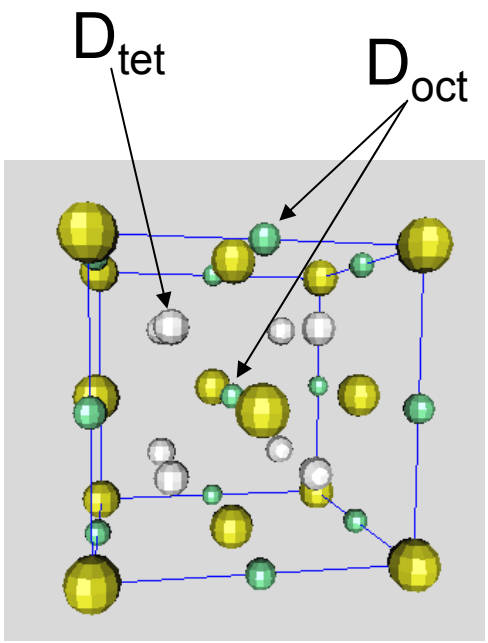
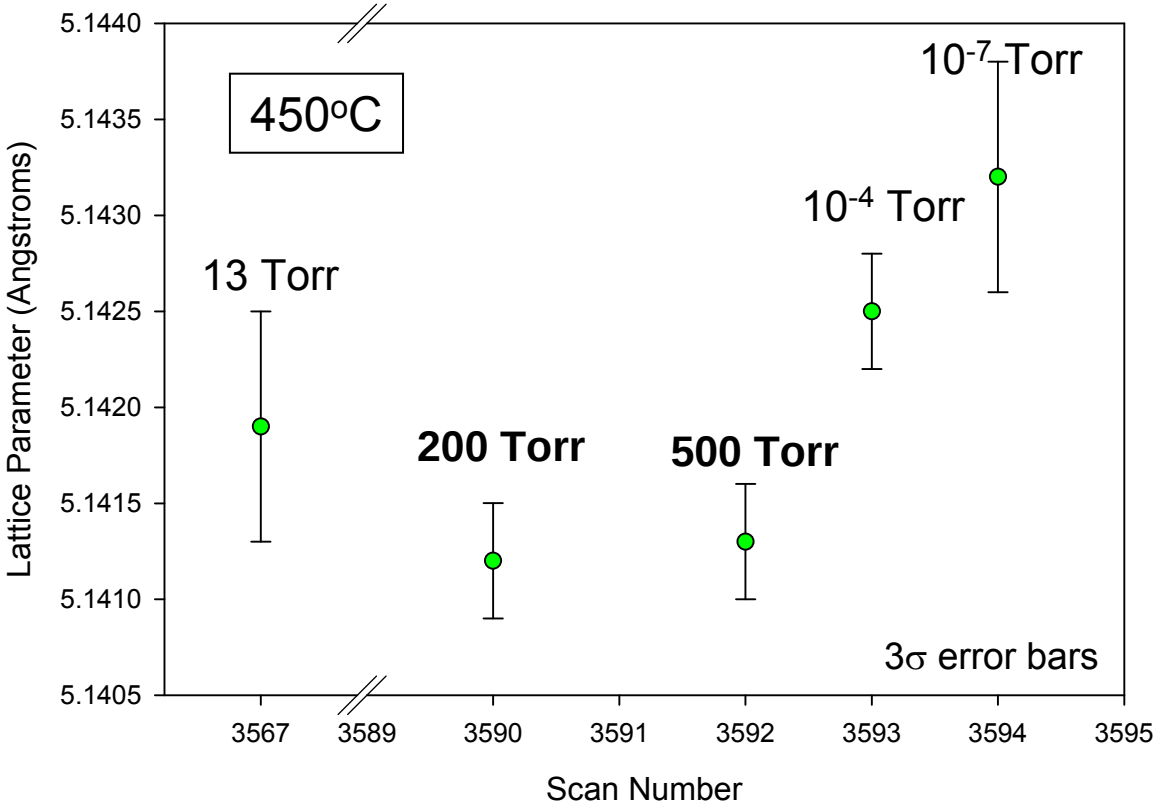
We observed small changes in the β phase lattice parameter with D_2 overpressure





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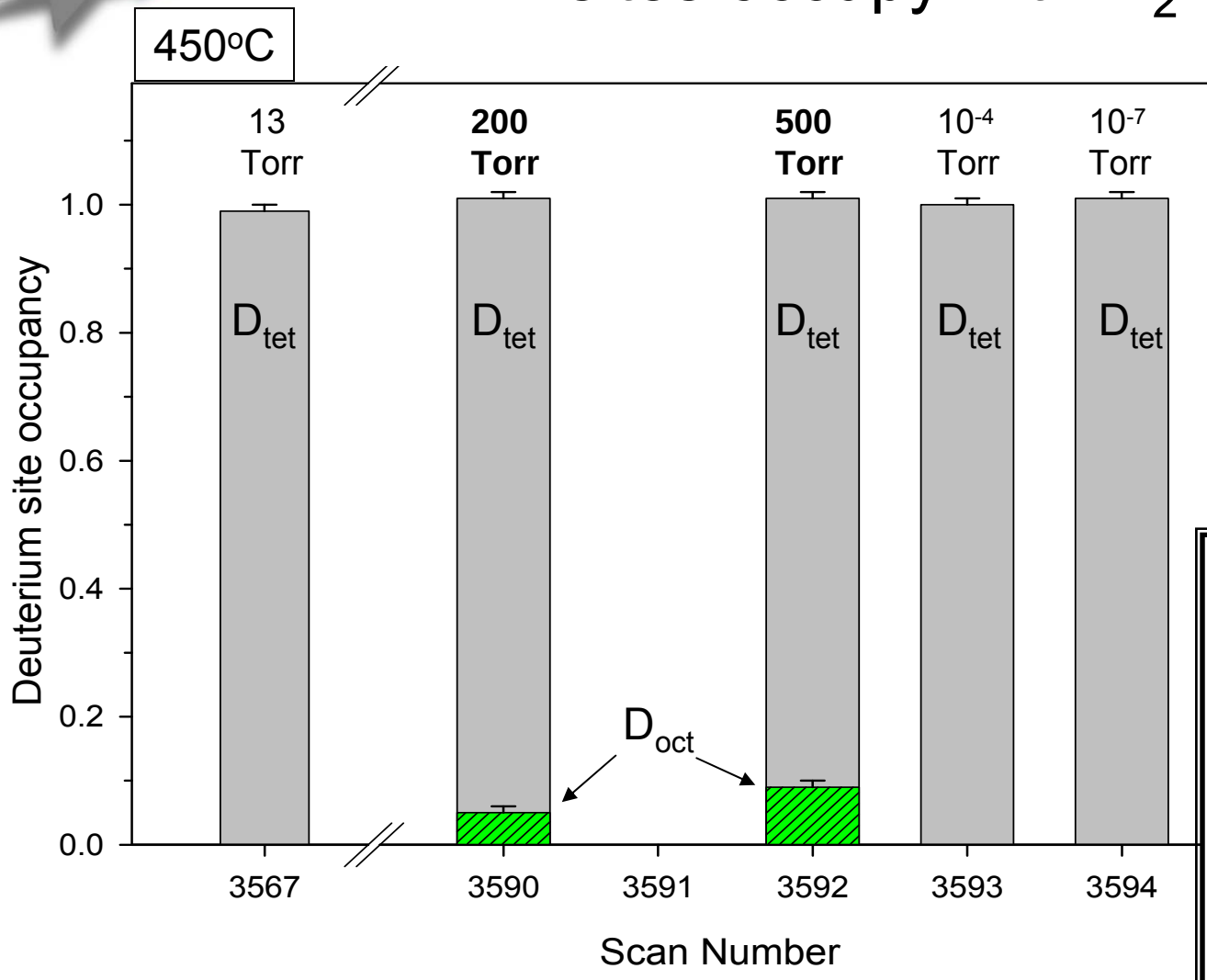
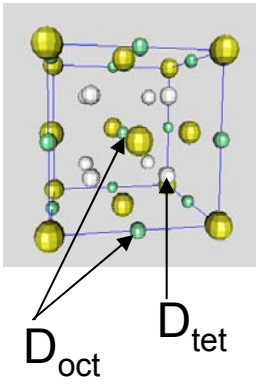
We detected as much as 10% octahedral sites occupy with D_2 overpressure





We observed the removal of D_{oct} occupancy at 450°C with vacuum pumping without loss of D_{tet}

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Summary

- In-situ neutron diffraction yields structural information regarding deuterium loading for Er metal.
- The α phase shows dramatic cell expansion upon exposure to D_2.
- The β phase shows little structural change in ($\alpha + \beta$) two-phase region.
  - suggests formation of the β phase near ErD_2 stoichiometry.
- D_2 overpressures of 200 to 500 Torr indicate 5 to 10 % occupation of D_{oct} sites, respectively.
- Pumping to vacuum (<10^{-4} Torr) at 450^{o}C shows removal of D_{oct} sites, with no reduction of D_{tet} sites, thus forming fully D_{tet} ($ErD_{2.0}$) β phase.

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  - **Michael Eatough (Sandia) and Alan Hurd (LANSCE) for serving as advocates for this analysis.**

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