Hydrogen and Helium Isotopes in Materials

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A Neutron Reflectivity Study of ErT_2 Films

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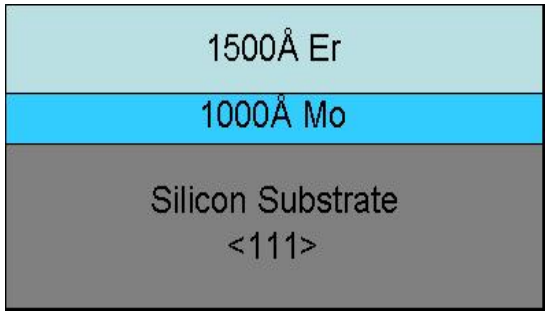
Motivation

- Can we distinguish near surface differences in the erbium tritide layer (denuded zone ??)

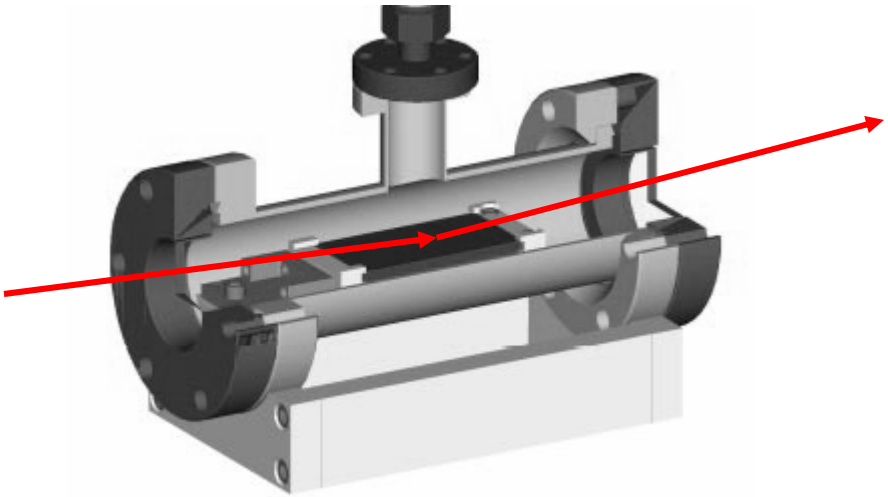
- How does the tritide layer change as a function of ^{3}He concentration?

Experiment

- Sample configuration:
  - Si <111> substrate
  - Deposition
    - 1000 Å Mo (to prevent formation of Er-Si compounds) deposited by e-beam PVD
    - 1500 Å of Er deposited by e-beam PVD
    - rate of 10 Å/s
    - Substrate temperature = 450°C
  - Hydriding
    - 450 °C at ~ 200 Torr
      - SS reactor
- Scattering Chamber:
  - Modified Nor-Cal 6” tee
  - Swagelok BW series valve
  - Sapphire windows
  - Sample sits on BN
  - Chamber evacuated ~ $1x10^{-7}$ Torr



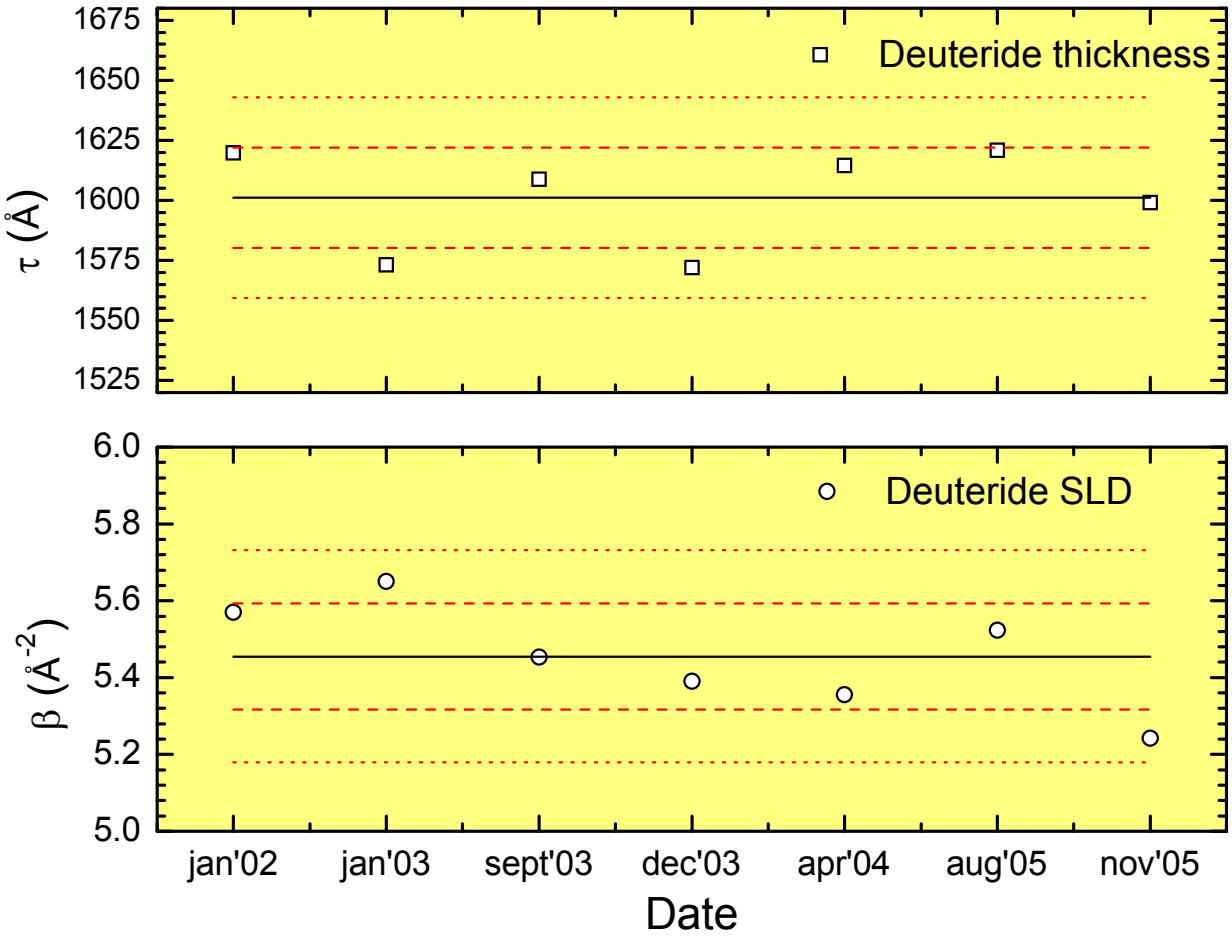
Si dimensions:
2.54 cm x 5.08 cm x 0.635 cm



IBA characterization of tritide films

Atom	Sample 1 (atoms/cm^2)	Sample 2 (atoms/cm^2)
H	1.65×10^{17}	2.31×10^{17}
D	4.54×10^{15}	4.89×10^{15}
T	7.59×10^{17}	7.14×10^{17}
Er	4.86×10^{17}	4.48×10^{17}
(HDT)/Er	1.91	2.08

Deuteride layer thickness and SLD as measured over the duration of the study



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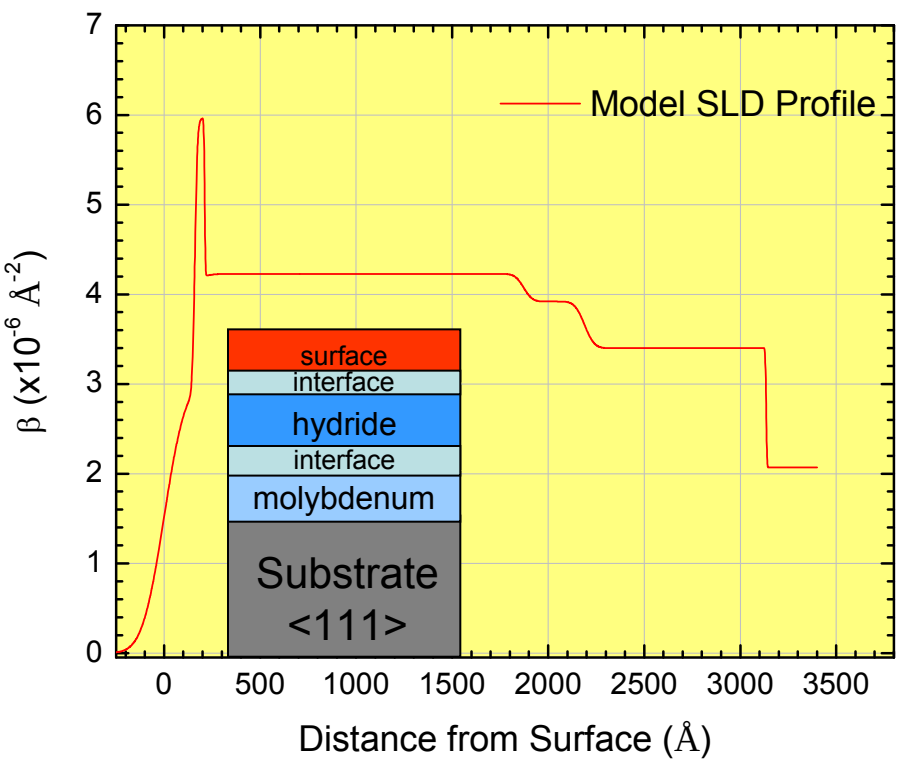
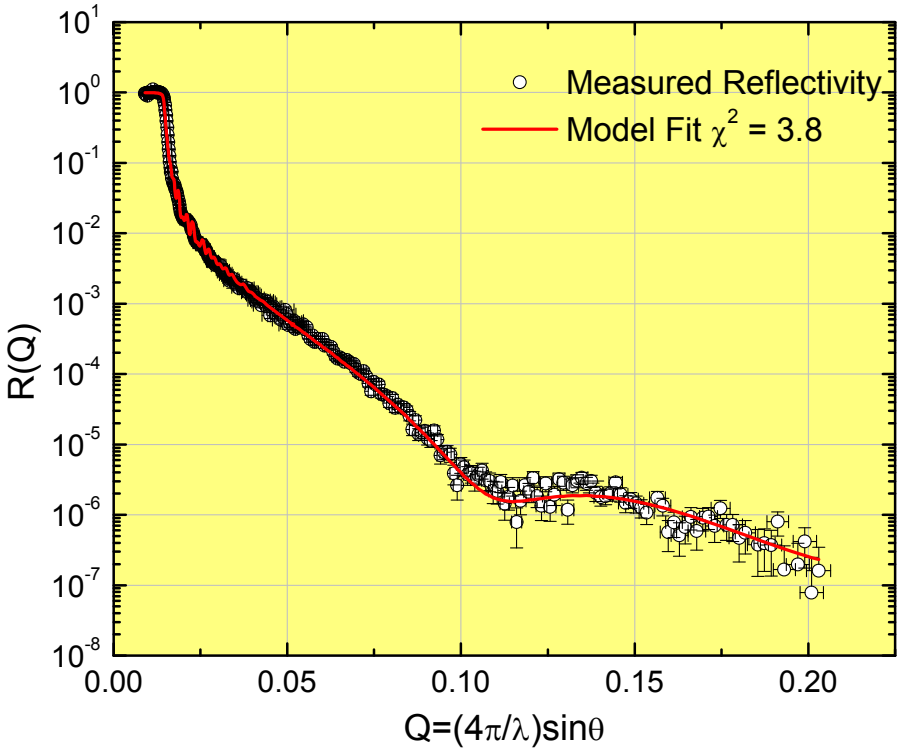
Example of experimental results and subsequent model fit to data





Sample 1

^{3}He:Er = 0.073

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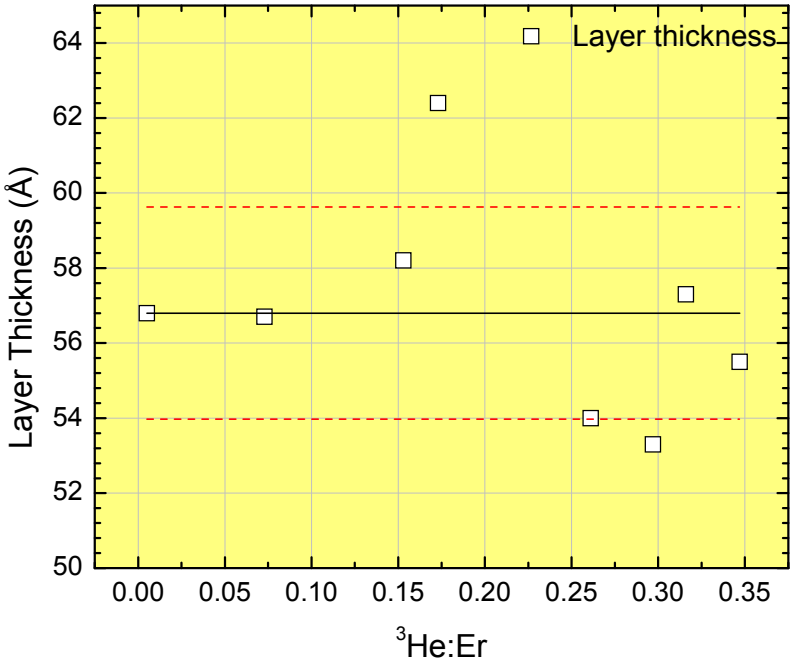
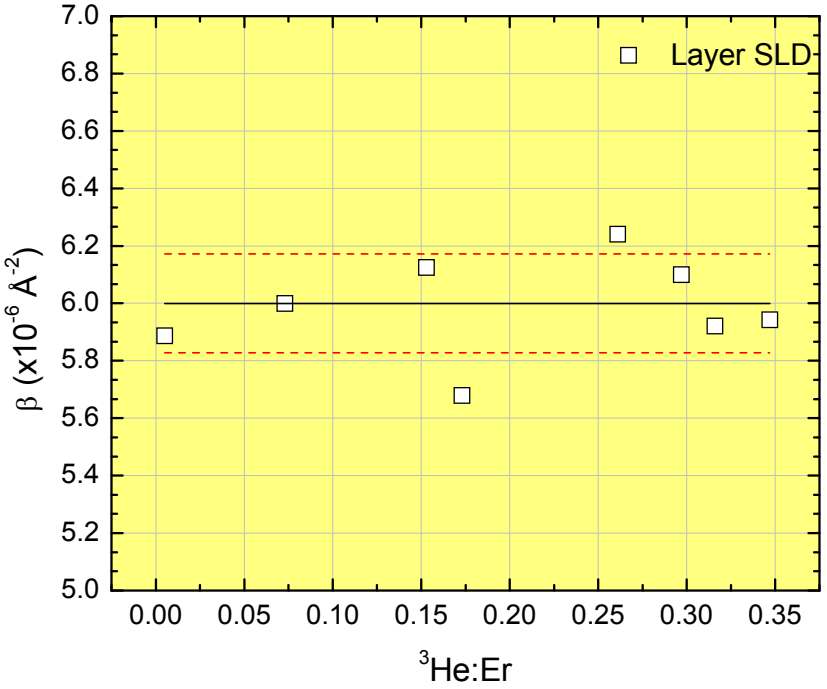
Sample 1: layer thickness and SLD of near surface region as a function of ^{3}He concentration

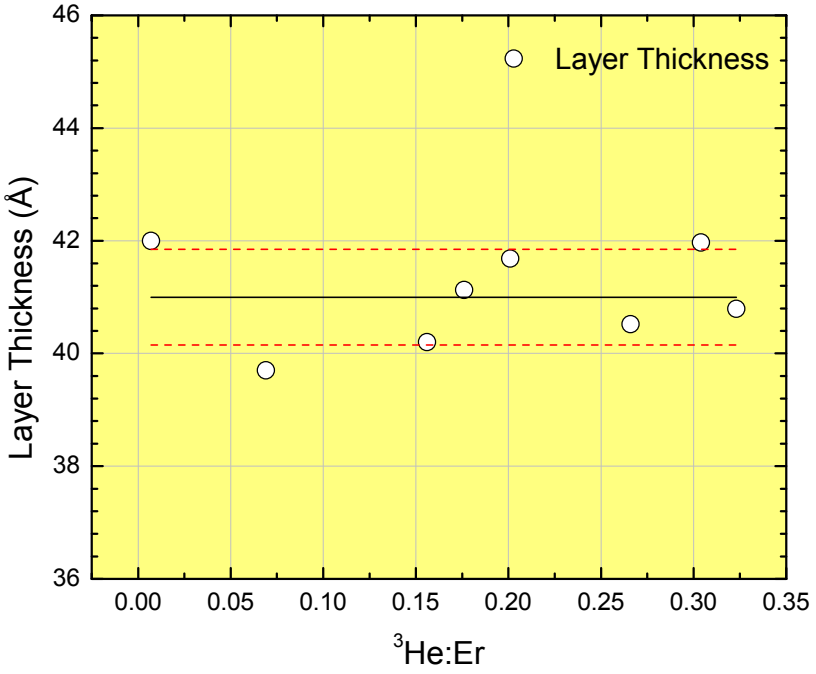


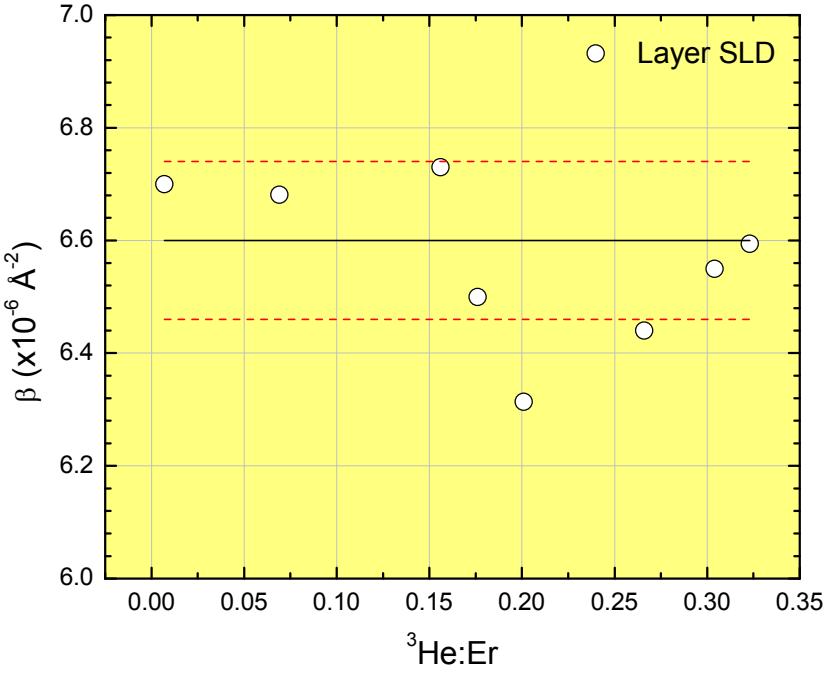


G:M=1.91

SLD consistent with high O and T concentration

Sample 2: layer thickness and SLD of near surface region as a function of ^{3}He concentration



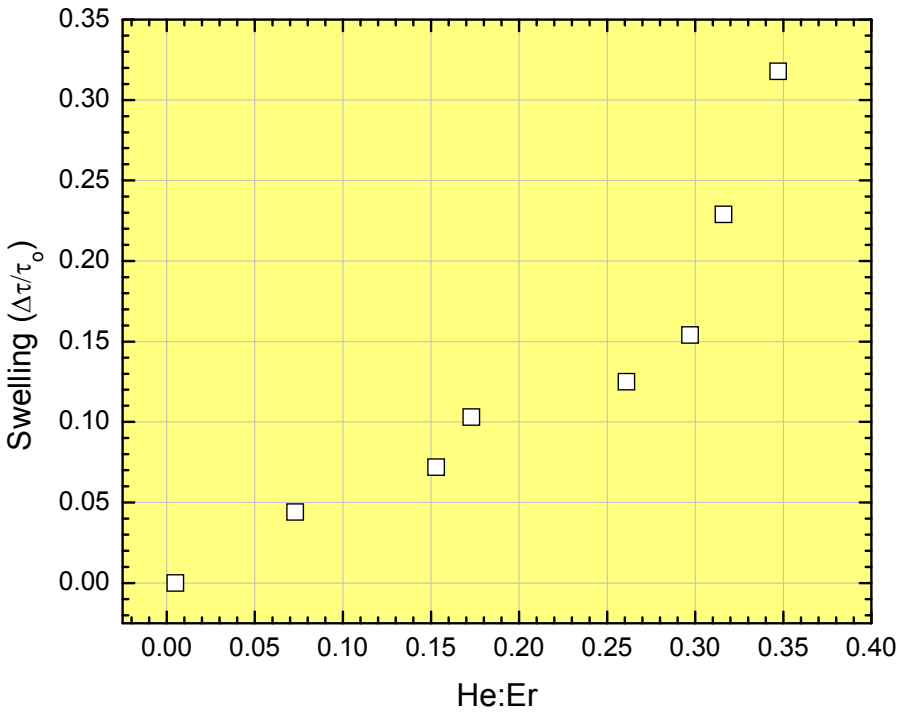


G:M=2.08

SLD consistent with high O and T concentration

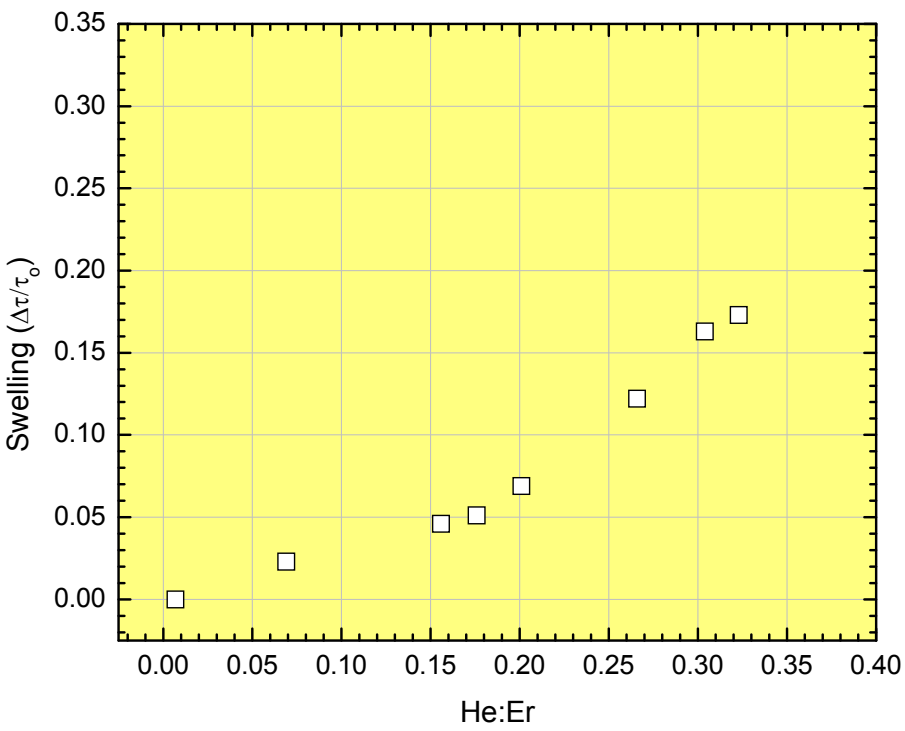
Tritide layer swelling as a function of ^{3}He concentration

G:M=1.91



Sample 1

G:M=2.08



Sample 2

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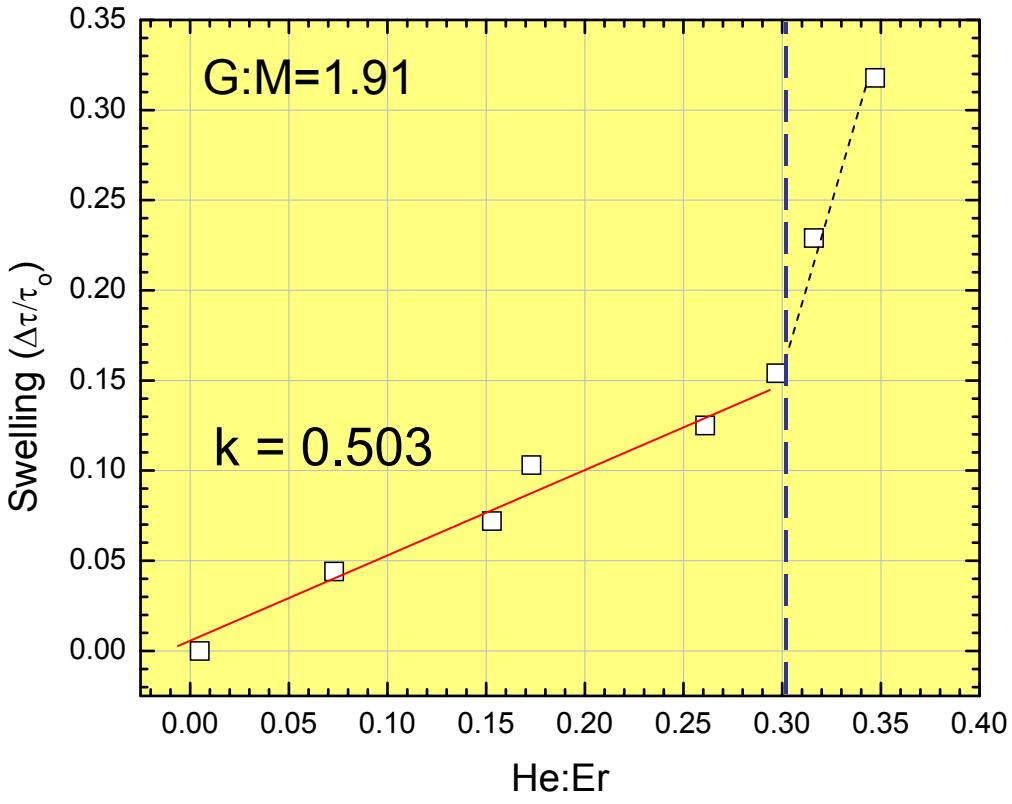
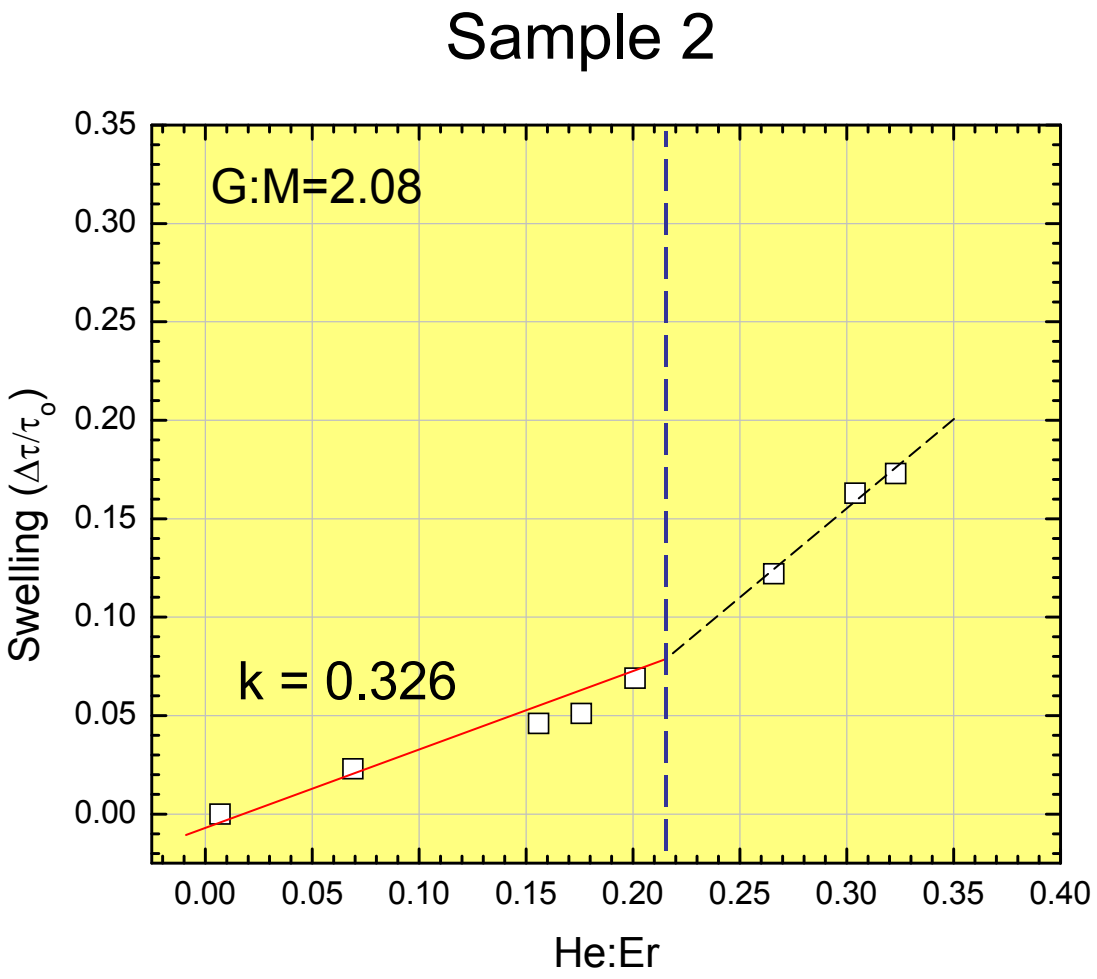
A closer look at the swelling data

Sample 1



A closer look at the swelling data



Relationship between tritide swelling and ^{3}He atomic volume

$$\frac{\Delta\tau}{\tau_\circ} = \underbrace{\left(\frac{v_{He}}{v_{M_H}}\right)}_{k}\left(\frac{^3He}{Er}\right)$$

- Where
  - v_{He} ≡ helium atomic volume
  - v_{MH} ≡ erbium atomic volume in the hydride
- Given

$$v_{M_H} \approx 33.8 Å^3$$

- Then

$$Sample1: v_{He} = 17.2 Å^3$$

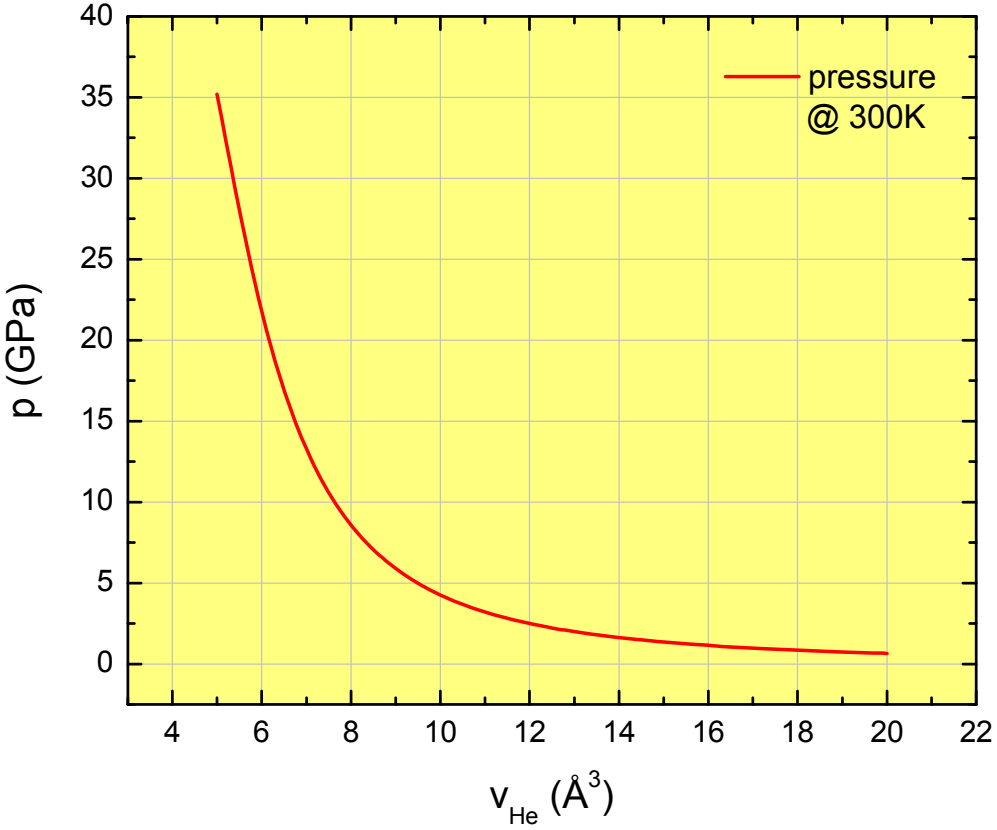
$$Sample2: v_{He} = 11.1 Å^3$$

(Shobert, et al., Phys. Rev B, 31(11), 7109, 1985. Shobert, et al., Phys. Rev B, 40(2), 1277, 1989. Cowgill, SAND 2004-1739, 2004.)

Bubble pressure determined by use of Mill's EOS

$$p^{-1} = 0.1196 - 0.04801 v_{He} + 0.005955 v_{He}^2$$

(Cowgill, SAND 2004-1739, 2004.)



$$\Rightarrow p_{sample1} = 0.95 GPa$$

$$\Rightarrow p_{sample2} = 3.12 GPa$$

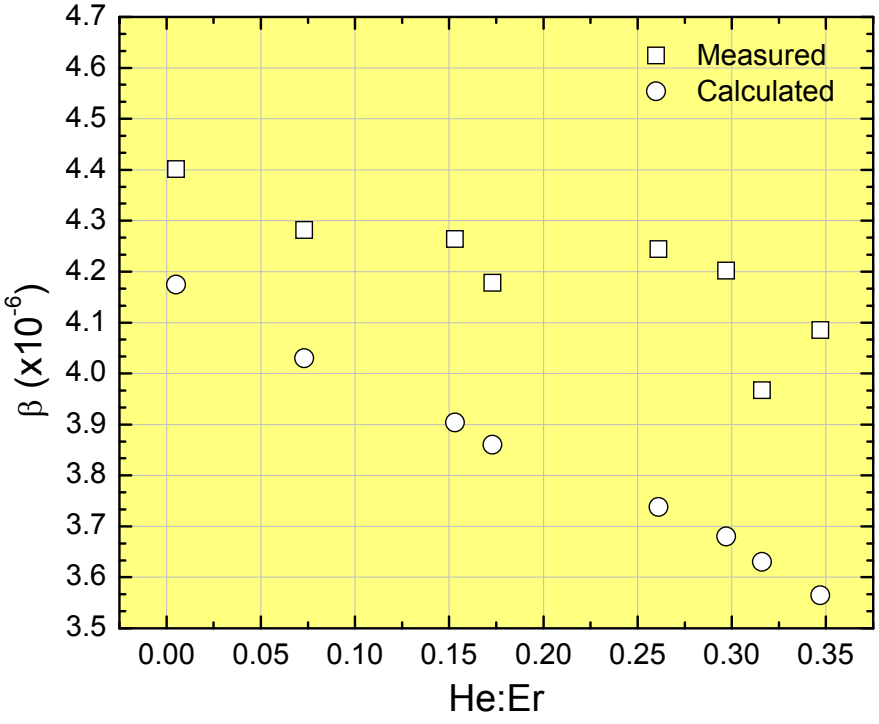
Mills, et al., Phys Rev B, 21(11), p.5137, 1980.

Summary of tritide layer swelling data

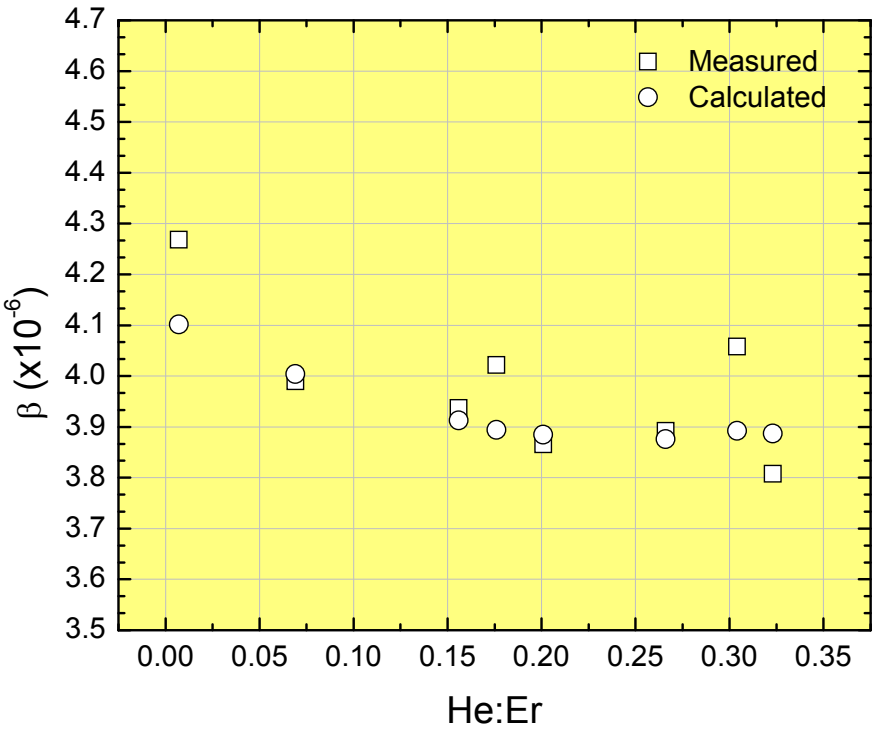
	Sample 1	sample 2
HDT:Er	1.91	2.08
T:Er	1.56	1.59
Slope	0.503	0.326
^{3}He:Er	0.297	0.201
$\Delta\tau/\tau_o$	0.15	0.11
v_{He} (Å^3)	17.2	11.1
P (GPa)	0.95	3.12

Measured verses predicted SLD

$$\beta_{layer} = x\beta_{ErT_2} + (1-x)\beta_{bubbles}$$



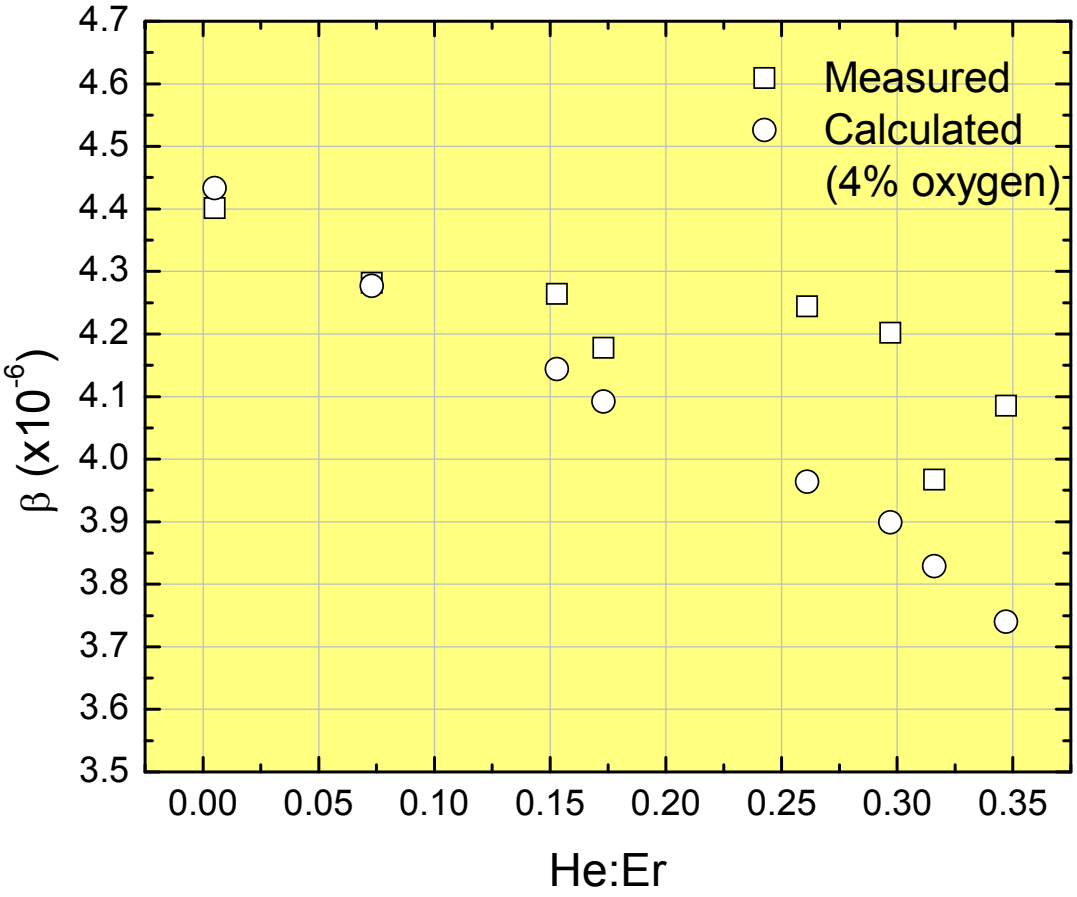
Sample 1



Sample 2

Measured verses predicted SLD

(corrected for oxygen)



Summary

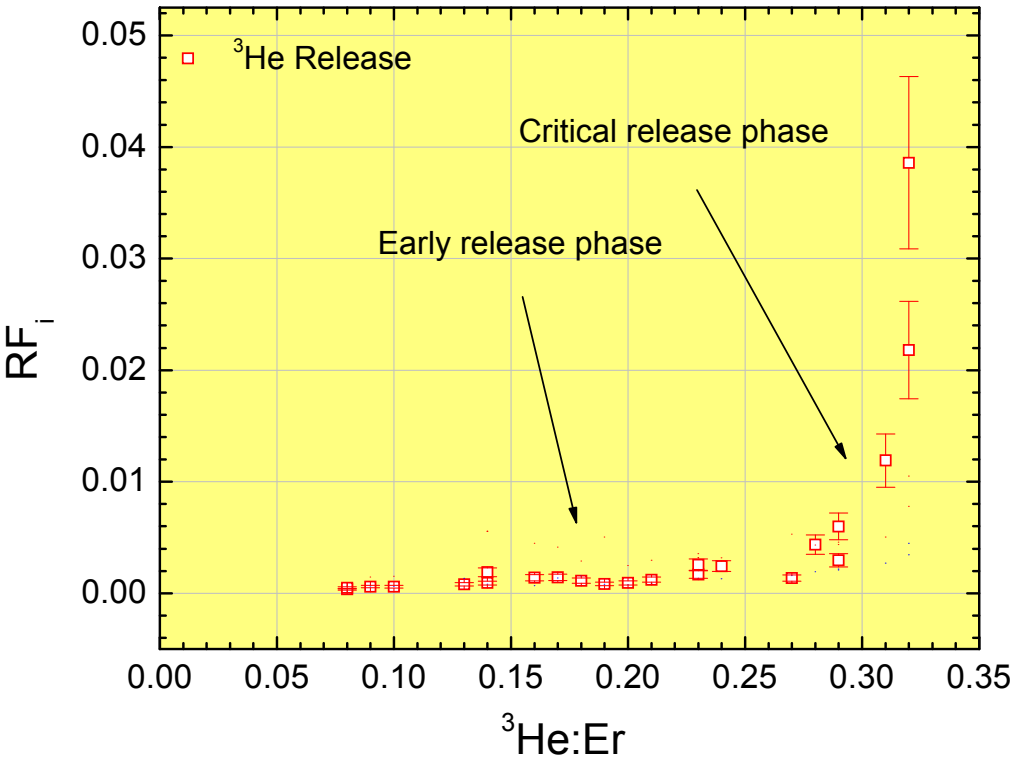
- The data indicate the existence of a near surface region with a length scale of ~ 50 Å and of high scattering length density (consistent with high oxygen/tritium concentration).
- Tritide layer expansion yields information on helium atomic volume within a bubble.
- The helium atomic volume obtained in the experiment is used with Mill's EOS to estimate pressure within a bubble during the constant expansion stage.
- The sample with higher hydrogen isotope-to-erbium ratio shows a higher bubble pressure and indicates transition to higher helium atomic volume within a bubble occurring at a lower ^{3}He:Er ratio.
- Film structure – many interfaces make it difficult to analyze.
- Repeat experiment using sapphire or quartz substrate.

Introduction

- ^{3}He out-gassing in many metal/metal hydride systems is characterized by two distinct regions
- Given the bulk of the ^{3}He produced remains in the lattice

$$ErT_2 \rightarrow ErT_{2-x}\,{}^3He_x$$
$$(T \rightarrow {}^3He + \beta^- + \bar{\nu})$$

- The intent of this work has been to determine how the ErT_2 film structure (interfacial and hydride layer) evolves with increasing ^{3}He concentration in the lattice



Structure of erbium hydride

- The beta phase of erbium hydride assumes the fluorite structure
- The hydrogen-to-erbium atomic ratio in the fluorite structure extends from about 1.85 to 2.15