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
# IBA characterization of tritide films

<table>
<thead>
<tr>
<th>Atom</th>
<th>Sample 1 (atoms/cm²)</th>
<th>Sample 2 (atoms/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>$1.65 \times 10^{17}$</td>
<td>$2.31 \times 10^{17}$</td>
</tr>
<tr>
<td>D</td>
<td>$4.54 \times 10^{16}$</td>
<td>$4.89 \times 10^{16}$</td>
</tr>
<tr>
<td>T</td>
<td>$7.59 \times 10^{17}$</td>
<td>$7.14 \times 10^{17}$</td>
</tr>
<tr>
<td>Er</td>
<td>$4.86 \times 10^{17}$</td>
<td>$4.48 \times 10^{17}$</td>
</tr>
<tr>
<td>(HDT)/Er</td>
<td>1.91</td>
<td>2.08</td>
</tr>
</tbody>
</table>
Deuteride layer thickness and SLD as measured over the duration of the study
Example of experimental results and subsequent model fit to data

Sample 1
$^3$He:Er = 0.073
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 $^3\text{He}$ 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

Swelling ($\Delta \tau/\tau_0$)

0.00 0.05 0.10 0.15 0.20 0.25 0.30 0.35 0.40

He:Er

Sample 1

G:M=2.08

Swelling ($\Delta \tau/\tau_0$)

0.00 0.05 0.10 0.15 0.20 0.25 0.30 0.35 0.40

He:Er

Sample 2
A closer look at the swelling data

Sample 1

Swelling ($\Delta \tau/\tau_o$)

G:M=1.91

k = 0.503
A closer look at the swelling data

Sample 2

Swelling ($\Delta \tau/\tau_0$)

G:M=2.08

$k = 0.326$
Relationship between tritide swelling and $^3$He atomic volume

$$\frac{\Delta \tau}{\tau_0} = \left( \frac{V_{He}}{V_{MH}} \right) \left( \frac{3\text{He}}{Er} \right)$$

Where
- $v_{He} \equiv$ helium atomic volume
- $v_{MH} \equiv$ erbium atomic volume in the hydride

Given
$$v_{MH} \approx 33.8\text{Å}^3$$

Then
Sample 1: $v_{He} = 17.2\text{Å}^3$
Sample 2: $v_{He} = 11.1\text{Å}^3$

Bubble pressure determined by use of Mill’s EOS

\[ p^{-1} = 0.1196 - 0.04801v_{He} + 0.005955v_{He}^2 \]


\[ \Rightarrow p_{\text{sample}_1} = 0.95 \text{GPa} \]

\[ \Rightarrow p_{\text{sample}_2} = 3.12 \text{GPa} \]

Summary of tritide layer swelling data

<table>
<thead>
<tr>
<th></th>
<th>Sample 1</th>
<th>sample 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDT:Er</td>
<td>1.91</td>
<td>2.08</td>
</tr>
<tr>
<td>T:Er</td>
<td>1.56</td>
<td>1.59</td>
</tr>
<tr>
<td>Slope</td>
<td>0.503</td>
<td>0.326</td>
</tr>
<tr>
<td>$^3\text{He}:\text{Er}$</td>
<td>0.297</td>
<td>0.201</td>
</tr>
<tr>
<td>$\Delta\tau/\tau_0$</td>
<td>0.15</td>
<td>0.11</td>
</tr>
<tr>
<td>$v_\text{He} (\text{Å}^3)$</td>
<td>17.2</td>
<td>11.1</td>
</tr>
<tr>
<td>P (GPa)</td>
<td>0.95</td>
<td>3.12</td>
</tr>
</tbody>
</table>
Measured verses predicted SLD

\[ \beta_{\text{layer}} = x \beta_{\text{ErT}_2} + (1 - x) \beta_{\text{bubbles}} \]

Sample 1

Sample 2
Measured verses predicted SLD
(corrected for oxygen)

![Graph showing measured versus predicted SLD](image)
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 \(^3\text{He}:\text{Er}\) 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:

$$ErT_2 \rightarrow ErT_{2-x}^xHe$$

$$(T \rightarrow ^3He + \beta^- + \bar{\nu})$$

• 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.