

# In-Situ Optical Diagnostics of Neutron Generator Target Films

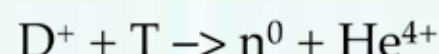


## Sandia National Laboratories

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### RESEARCH PROBLEM

- Neutron tubes are small electrostatic accelerators that produce neutrons. Deuterium ions are accelerated into a target containing tritium to produce neutrons via the reaction:

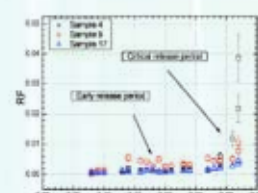
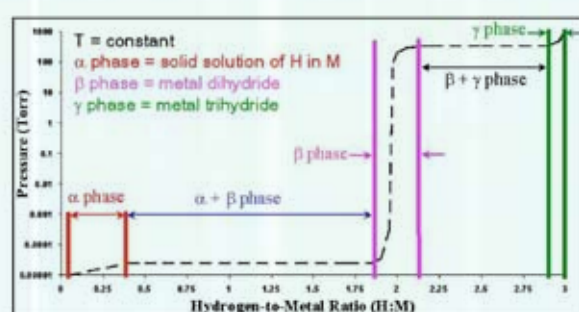


- Target and source store the hydrogen isotopes deuterium (D) and tritium (T) in metal occluder films.
- Tritium is radioactive and decays into  $^3He$ , which if released from the target film will poison the vacuum environment of the neutron tube.



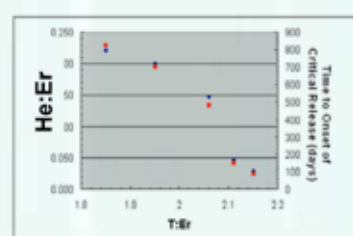
### WHY DO THE OCCLUDERS NEED SPECIAL ATTENTION?

- Hydrogen occluders of the rare-earth and transition metals have complex phase diagrams. The diagram below shows an Er-H isotherm for an erbium powder.
- Three distinct crystallographic phases are possible.



- A link between helium release and target film stoichiometry has been reported.

- Helium produced by the radioactive decay of tritium needs to be retained in the target. RFi = Instantaneous Release Fraction



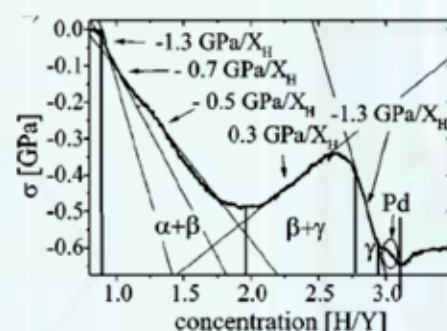
(From Beavis et al., J. Vac. Sci. Technol., 14 (1977), 509)

### HOW DO YOU MEASURE THE IMPORTANT PARAMETERS OF METAL HYDRIDE FILMS?

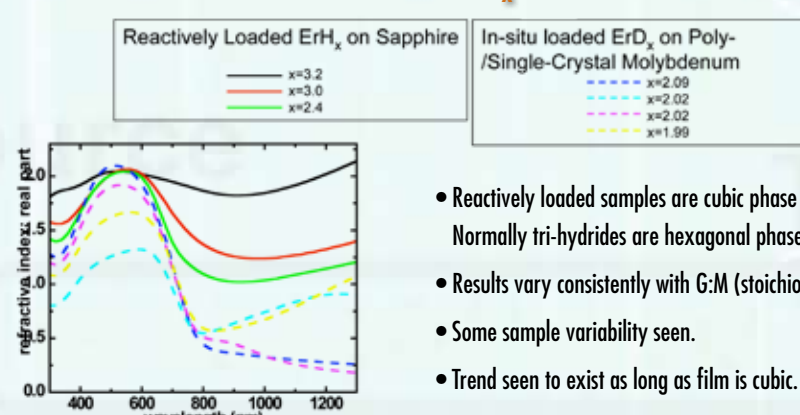
Use optics to in-situ monitor the hydriding process to determine when the film is

1. in the property crystallographic phase
2. has the right stoichiometry.

- Optical properties of metal hydrides change with film stoichiometry.
- Stress state of metal hydride films change with film stoichiometry.
- Notice the color change of the target films below for different Gas:Metal ratios.
- Example below is for YHx.

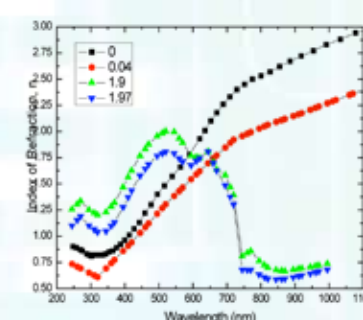


### ELLIPSOMETRY ON SMOOTH REACTIVELY LOADED $ErD_x$ FILMS



- Reactively loaded samples are cubic phase tri-hydrides. Normally tri-hydrides are hexagonal phase.
- Results vary consistently with G:M (stoichiometry).
- Some sample variability seen.
- Trend seen to exist as long as film is cubic.

### ELLIPSOMETRY OF SMOOTH $ErD_x$ FILMS ON Moly/SILICON SUBSTRATES

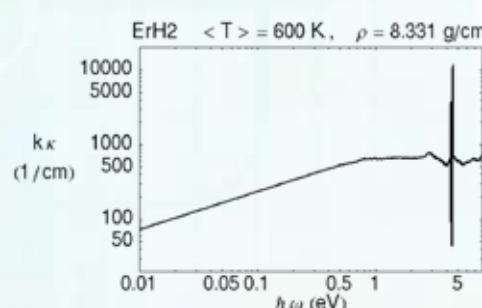
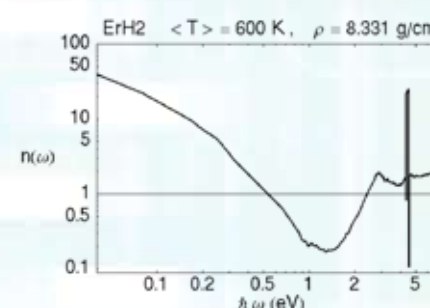


| Pressure (Torr) | Measured G.M | Expected G.M |
|-----------------|--------------|--------------|
| 0               | 0            | 0            |
| 7               | 0.04         | 1.85         |
| 116             | 1.9          | 2.05         |
| 336             | 1.97         | 2.15         |

- Same trends seen as from other sample sets.
- Expected G:M predicted from older thermodynamics on powders does not match measured values.
- We have not been able to consistently produce predicted lower stoichiometry films.
  - Are the thermodynamics wrong for films?
  - Films more pure than older films?
  - Differences between powders and films?
  - Are the kinetics driving the problem?

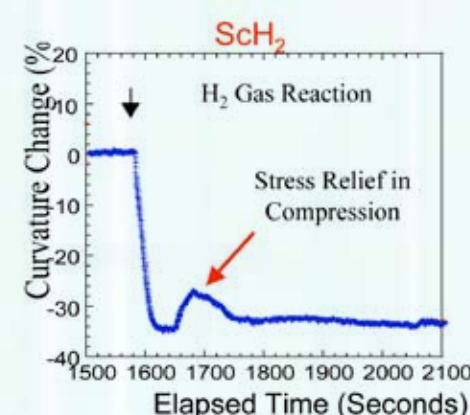
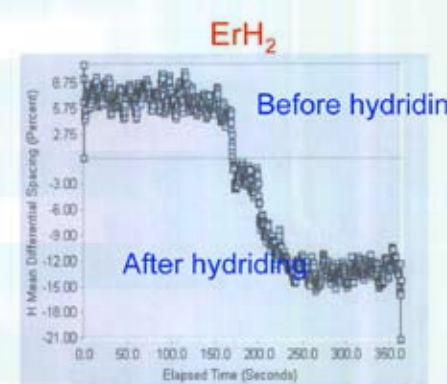
### THE OPTICAL PROPERTIES OF FILMS CAN ALSO BE MODELED

- Technique developed at Sandia by T. Mattsson and M. Desjarlais
- Equilibrium positions of atoms in large  $3 \times 3 \times 3$  cell computed with molecular dynamics.
- Electronic transitions of large cell computed with very high accuracy using VASP, an ab-initio electronic structure code.
- Optical properties calculated from the electronic transitions using the Kubo-Greenwood formalism.
- Results compare favorably to experimental results in spite of the differences in temperature.



### STRESS STATE OF FILM IS MEASURED USING SANDIA DEVELOPED MOSS

- MOSS=Multi-beam Optical Stress Sensor
- Measures curvature of substrate
- Thin film after deposition is in tensile stress
- Upon heating/hydriding film is in compressive stress
- After hydriding and cooling film develops in-plane tensile stress



### SIGNIFICANCE AND IMPACT OF RESULTS

- Our results call into question the validity of the thermodynamics/kinetics used to produce neutron tube targets.
- We are using the optical techniques developed in this LDRD to answer this critical question.
  - We are performing in-situ spectroscopic ellipsometry and reflectivity to monitor the uptake and release of hydrogen at differing temperatures and pressures.
  - We are performing MOSS measurements as film is hydriding to monitor the uptake and release of hydrogen at differing temperatures and pressures.