SAND2007-4725C

Epitaxial Oxide Growth in ErH_2 Thin Films

Chad Parish*, Clark Snow, Jim Browning, and Luke Brewer
Sandia National Laboratories, Albuquerque, NM
*Contact: cmparis@sandia.gov

INTRODUCTION

- Neutron generators (NGs, Figure 1) are devices used to produce high-energy neutrons
- Neutron generators are important components of nuclear weapons
- Neutrons are generated by accelerating deuterium (D, isotope ^{2}H) into a rare-earth tritide (T, isotope ^{3}H) target
- Erbium ditritide (ErT_2) is a commonly used target material
- However, the microstructural properties of ErT_2 are not yet well understood
- In this work, the distribution and crystallography of erbium oxide (Er_2O_3) contamination in films of erbium dihydride (ErH_2) were examined to simulate the radioactive tritiated system ErT_2

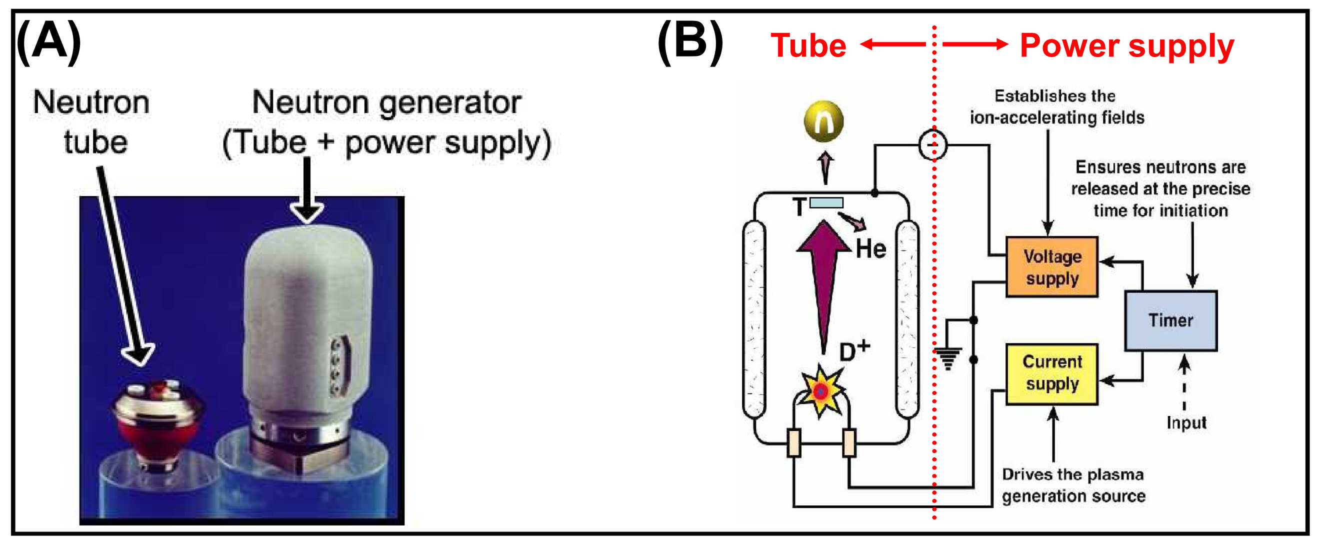

Figure 1: (A) Photograph of a neutron generator system. (B) Schematic illustration of a neutron generator.

PROCEDURE

- Erbium metal films ~500 nm thick were deposited onto a 100 nm molybdenum interlayer on a (100) Si substrate
- The erbium films were then hydrided into ErH_2
- ErH_2 has the fluorite $Fm\bar{3}m$ crystal structure, Figure 2A
- Bixbyite-structured $Ia\bar{3}$ Er_2O_3, Figure 2B, forms unintentionally during crystal growth
- Samples were prepared by focused-ion-beam lift-out using an FEI dual-beam instrument, with 2-5 keV final polishing
- Samples were examined in a Phillips CM30 TEM/STEM operated at 300 keV

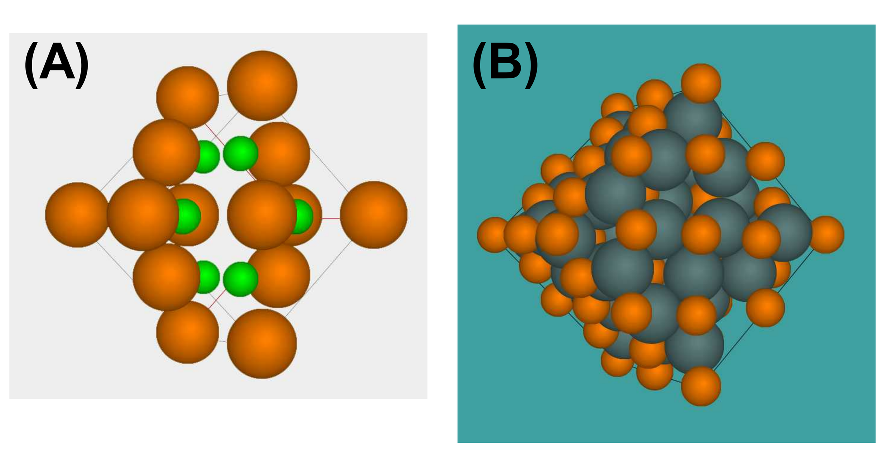

Figure 2: (A) Fluorite-structured ErH_2. (B) Bixbyite-structured Er_2O_3.

a_{ErH2}=0.512 nm a_{Er2O3}=1.054 nm

a_{Er2O3}/a_{ErH2}≈2.06

RESULTS

- Energy-filtered TEM (EFTEM) is expected to have superior signal/noise for low-atomic-number elements such as oxygen, compared to EDS
- Figure 3 shows that oxygen EFTEM maps indeed have significantly better signal/noise than EDS maps

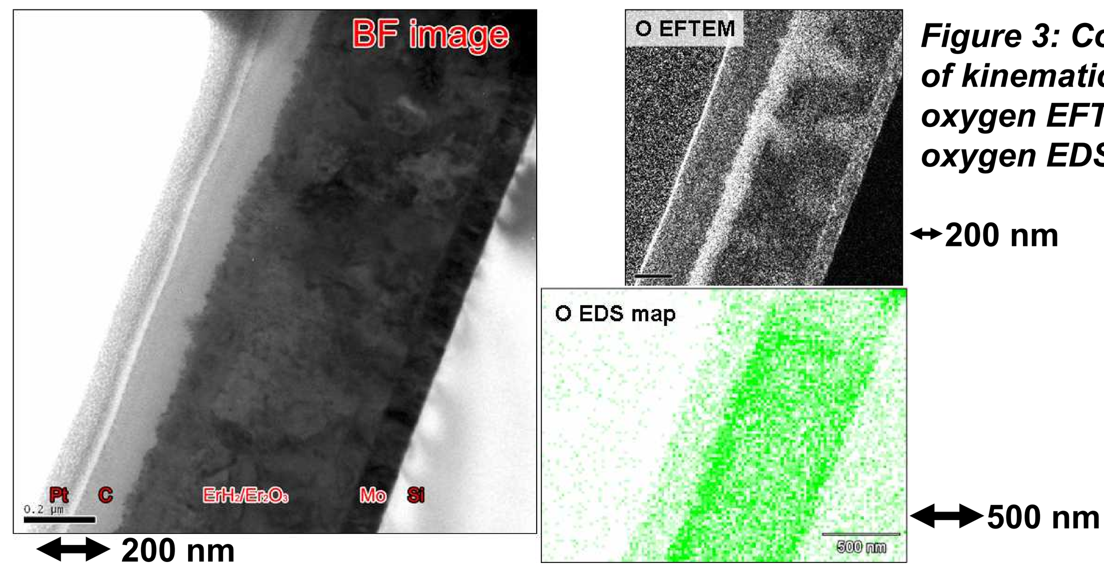

Figure 3: Comparison of kinematical BF-TEM, oxygen EFTEM, and oxygen EDS maps.

- EDS analysis does show an increased oxygen signal in areas that EFTEM shows to be oxygen rich, further confirming the identity as oxides (Figure 4)

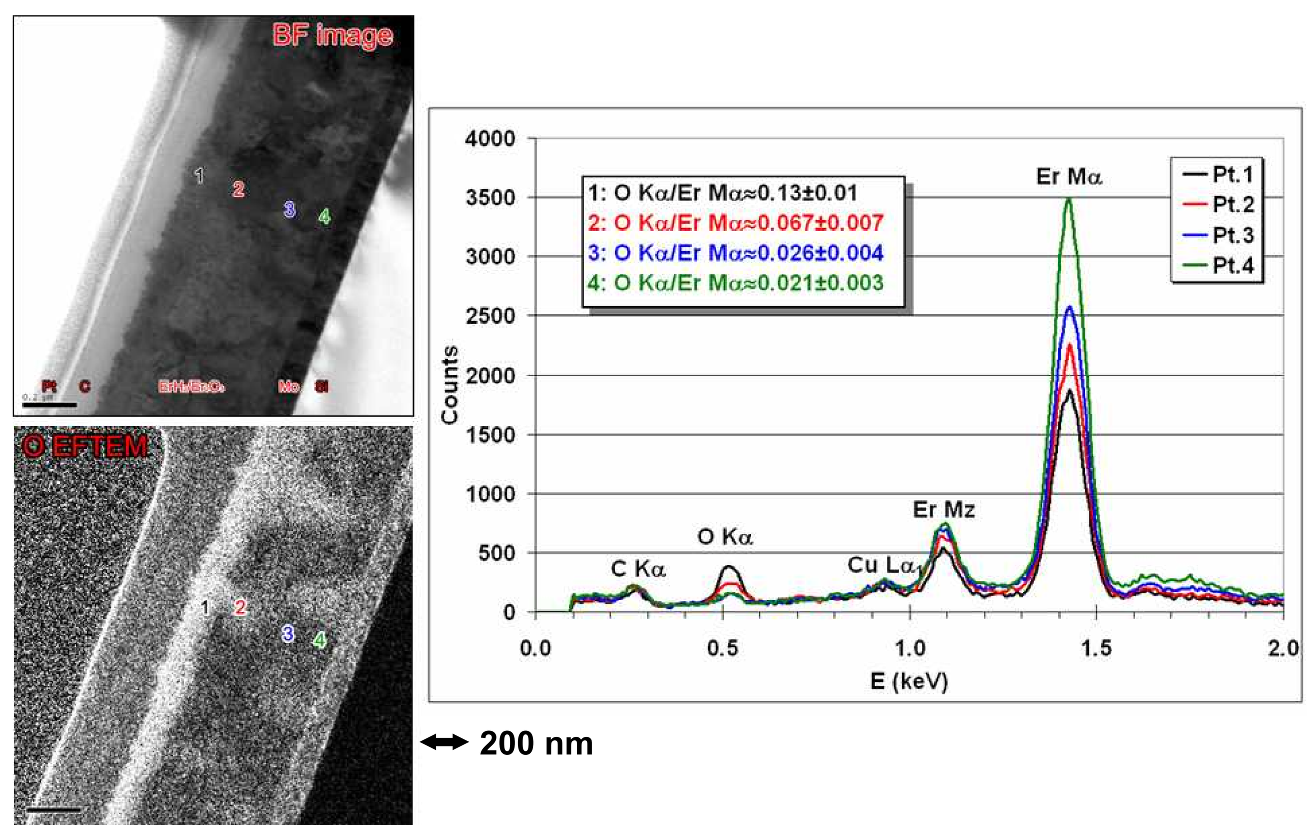

Figure 4: Comparison of BF image, EDS point-spectra, and EFTEM oxygen map.

- EFTEM shows a 100-150 nm thick oxide layer on the surface of the film, as well as large oxides within the film (Figure 5)

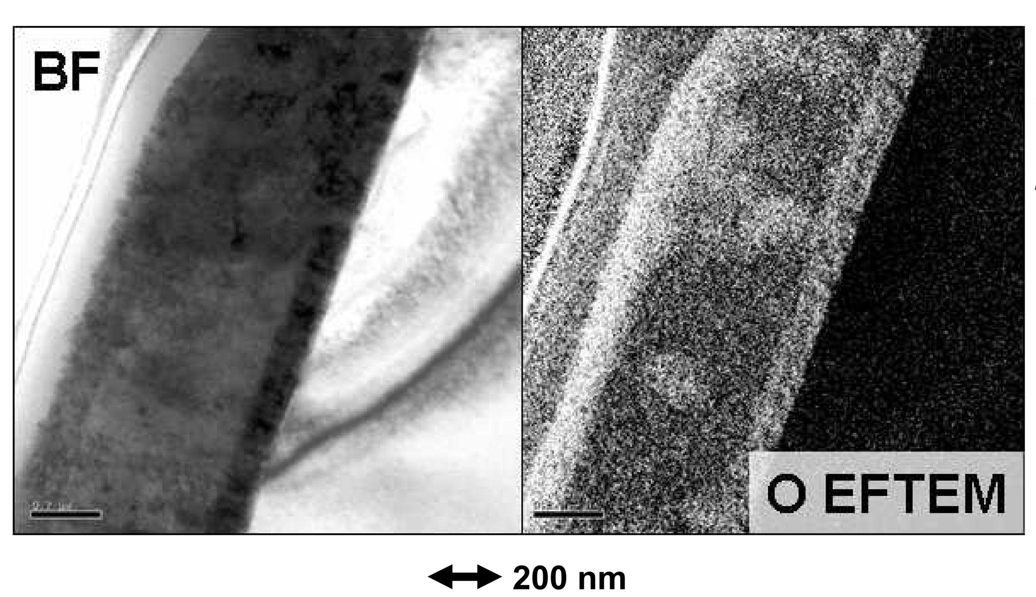

Figure 5: EFTEM shows a surface oxide layer and penetration of oxides into the hydride film.

- Tilting to a zone-axis orientation results in strong dynamical contrast in both top (oxide) and bottom (hydride) layers (Figure 6)

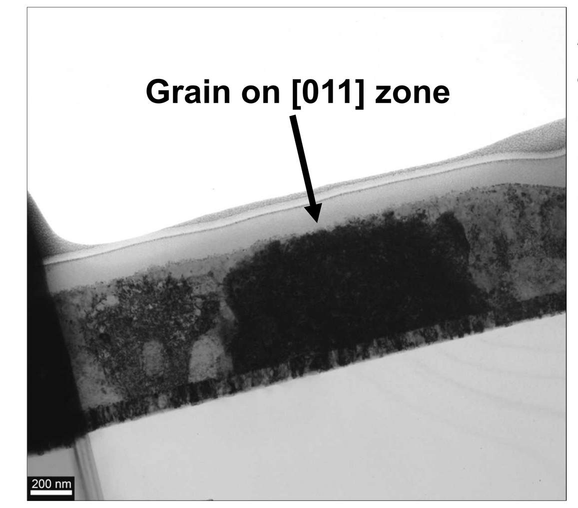

Figure 6: Oxide and hydride crystals showing dynamical diffraction under the same tilt conditions.

- SAD of the hydride shows a [011] type zone-axis diffraction pattern (ZADP) with unexpected satellite spots (Figure 7); satellite spots were observed in <u>all</u> hydride diffraction patterns
- SAD of the oxide shows streaking of the major oxide spots, which may indicate a slight mosaic structure (Figure 8)
- SAD of both layers indicates a strongly epitaxial relation (Figure 9)

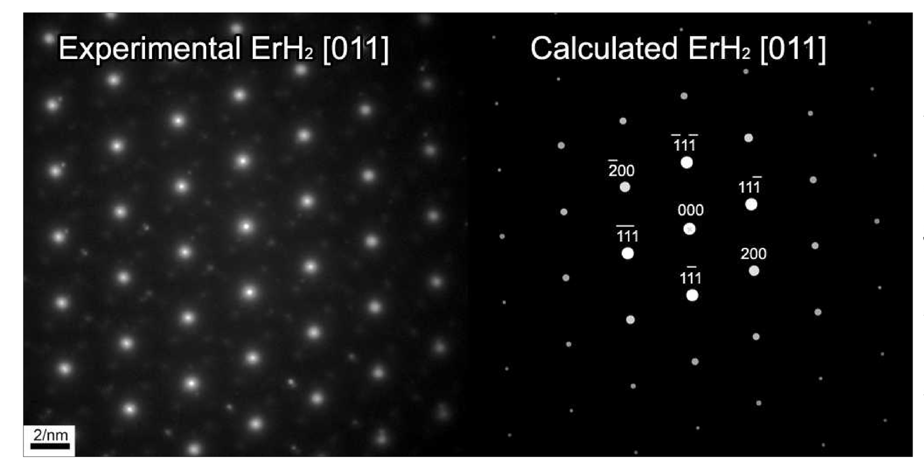

Figure 7: [011] type SAD pattern of the hydride grain in Figure 6. Satellite spots are not expected from the fluorite-based ErH_2 structure

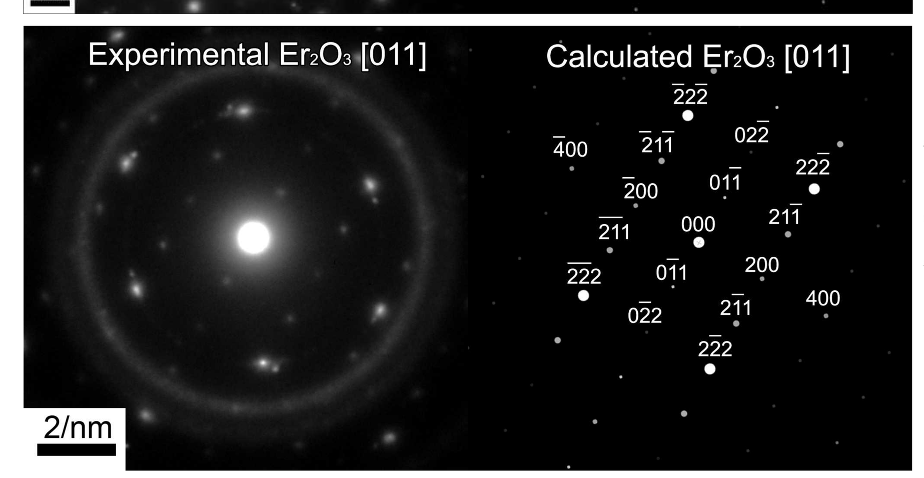

Figure 8: [011] type SAD pattern of the oxide layer in Figure 6.

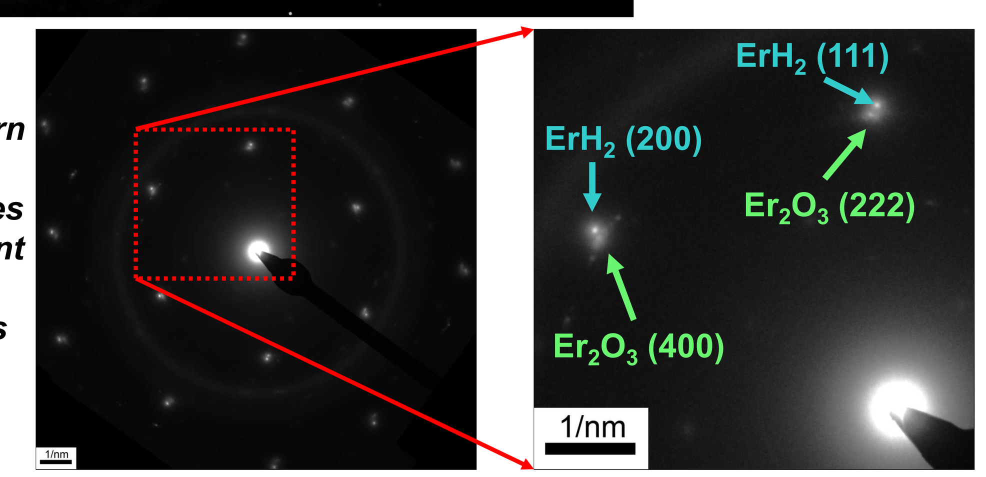

Figure 9: [011] type SAD pattern of both the grains in Figures 7 & 8. Alignment between ErH_2 and Er_2O_3 spots indicates epitaxy.

- To determine the orientation relation, an oxide layer and associated hydride grain (Figure 10) were tilted to a series of zone axis orientations (Figure 11)

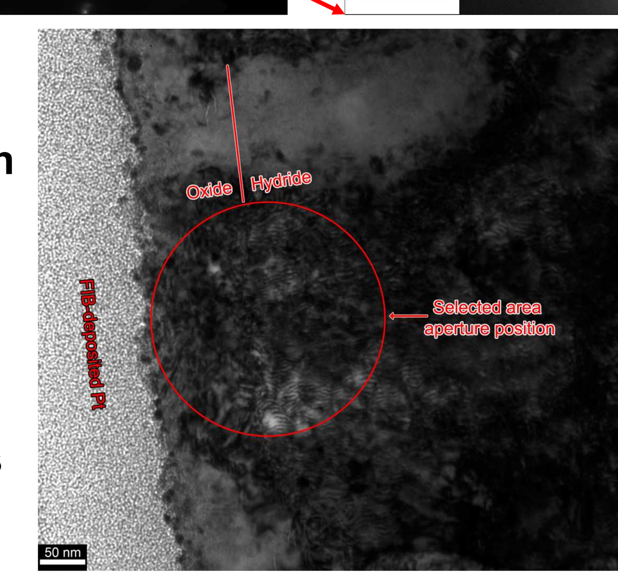

Figure 10: Hydride and oxide grains for diffraction shown in Figure 11

- The registry between the $(hkl)_{hydride}$ and $2(hkl)_{oxide}$ reflections in Figure 11 are indicative of a cube-on-cube epitaxial relationship

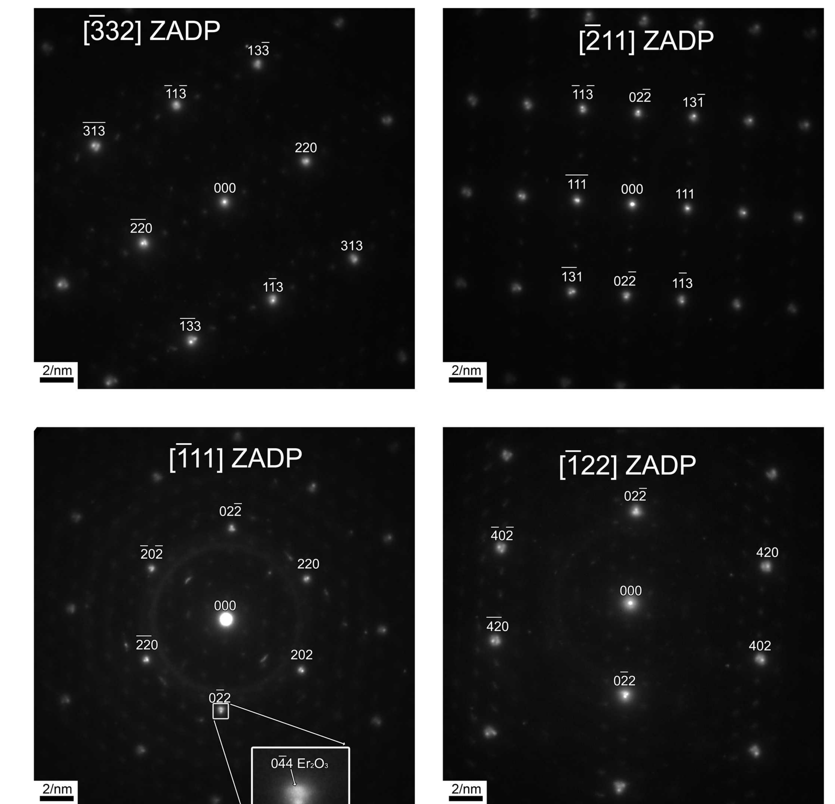

Figure 11: ZADPs of the grains shown in Figure 10. Labels are the ErH_2 reflections. All major spots show an associated Er_2O_3 reflection, as illustrated in the inset to [$\bar{1}$ 11].

- As seen in Figure 7, ErH_2 SADPs have satellite spots not predicted from the hydride crystal structure
- It is speculated that these satellite spots arise from double-diffraction off Er_2O_3 nanocrystals in a hydride matrix
- In Figure 12, a grain was tilted ~4° of <011> into a near {111} two-beam condition
- Figure 12 shows the BF image, and DF image taken with the reflection marked in the diffraction pattern
- The DF image shows small (~10 nm) particles, possibly Er_2O_3
- High-resolution TEM is currently underway to determine if the small particles are oxides

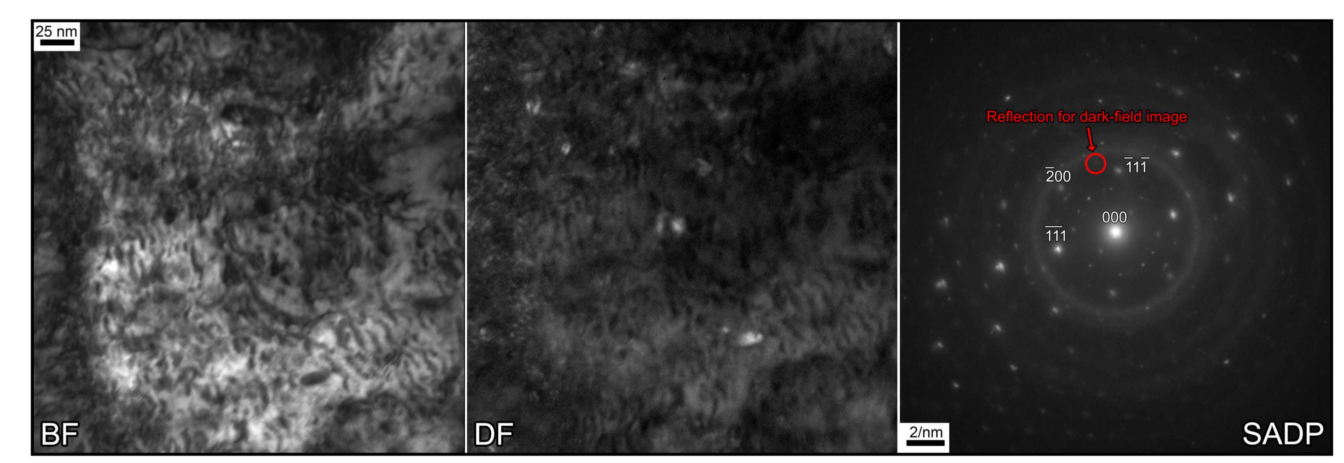

Figure 12: A BF image of a hydride grain, and associated DF image showing small inclusions arising from a diffraction spot not associated with the hydride crystal structure. It is speculated the spots may be double diffraction, and the inclusions are oxide.

CONCLUSIONS

- Erbium hydride films are confirmed by EFTEM, EDS, and SAD to suffer from growth of unwanted oxide particles
- An oxide surface film tends to grow in a cube-on-cube orientation
- It is possible that the hydride matrix is contaminated with small oxide inclusions; further investigations are underway

National Nuclear Security Administration

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

Sandia National Laboratories