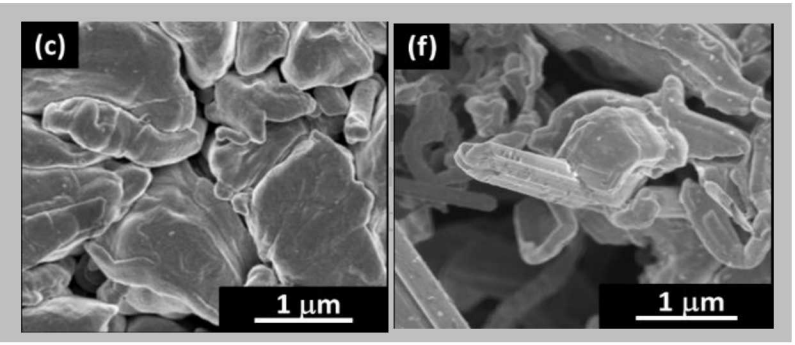
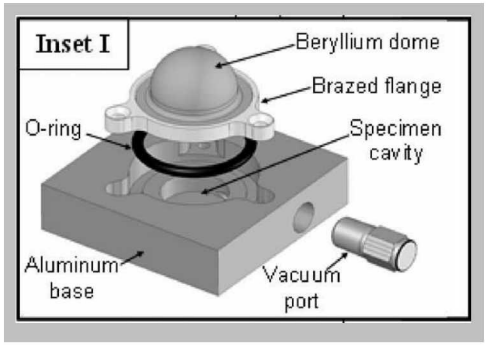
SAND2019-9103C

Exceptional service in the national interest

Sandia National Laboratories





USE OF A BERYLLIUM-DOME HOLDER FOR TEXTURE AND STRAIN CHARACTERIZATION OF LI METAL THIN FILMS VIA TILT-A-WHIRL METHODOLOGY

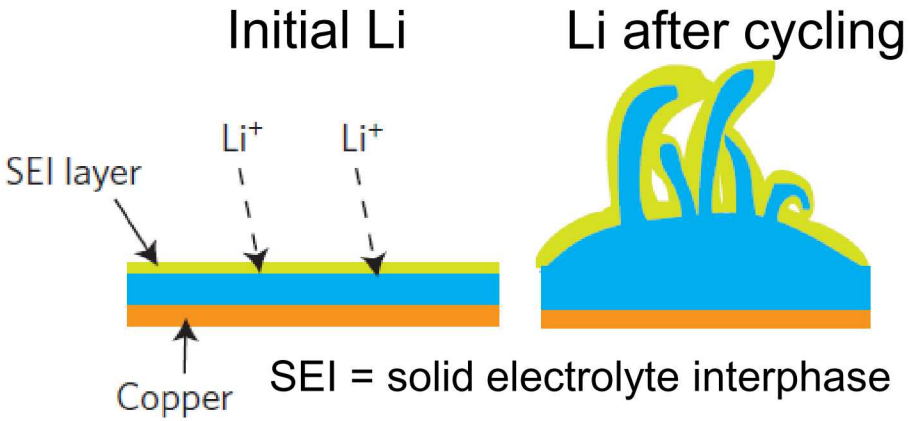
Mark A. Rodriguez, Katharine L. Harrison, Brian Perdue,
Subrahmanyam Goriparti, Brad Boyce, and Zach Casias
Sandia National Laboratories, Albuquerque, NM

U.S. DEPARTMENT OF ENERGY
NNSA National Nuclear Security Administration

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

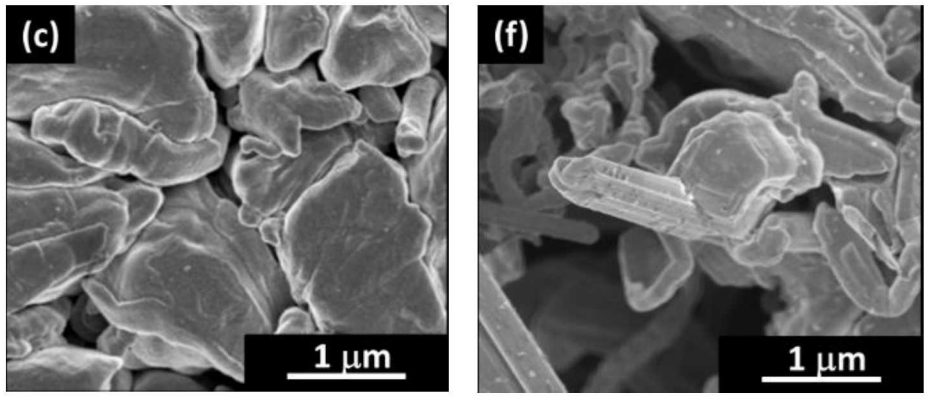
We have been tasked with testing Li films for residual macrostrain

Sandia National Laboratories



SEI = solid electrolyte interphase

Zheng et al., 2014, *Nature Nanotechnology*, *9*(8), p.618.



Harrison et al., *ACS Nano* 2017, 11 (11), 11194-11205.

High energy density batteries require Li anodes

↓

Li anodes are problematic because dendrites form when charging leading to short circuits and fires

↓

Compression at the lithium-electrolyte interface improves Li morphology and suppresses dendrites

↓

Initial work by Campbell *et al.* suggests that Li can be work hardened, which could impact how dendrites form and are prevented

Campbell *et al.*, Scientific Reports, 2018, 8:2514.

↓

If Li can be work hardened, we hypothesize that residual strain should be evident in XRD

Hey Mark, can you use your Tilt-A-Whirl for this?

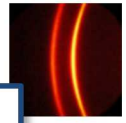
Data Processing and Analysis Flowchart for **TILT-A-WHIRL**

Sandia National Laboratories

batch.slm

Data collection via slam file

*.gfrm



3360 frames
20 hrs / 13 GB

integration.slm

Frame integration and file stitching

*.PLT

840 merged ASCII files
13 MB

Matlab.exe

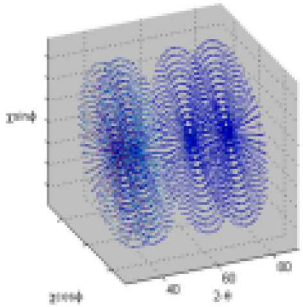
File clean-up by **Spaghetti/ Linguine**

*.rft

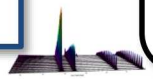
840 2θ scans with χ,ϕ tags "spaghetti data"

Matlab.exe

Pole Figure Explorer for visualization and plotting



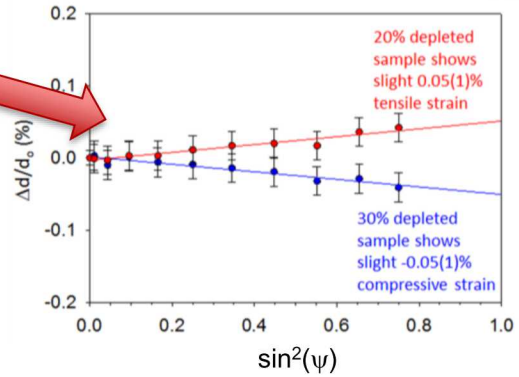
*.rft



14 raw data ϕ-merged 2θ scans

Jade.exe

Phase ID, LP refinement, macro-strain via **JADE**



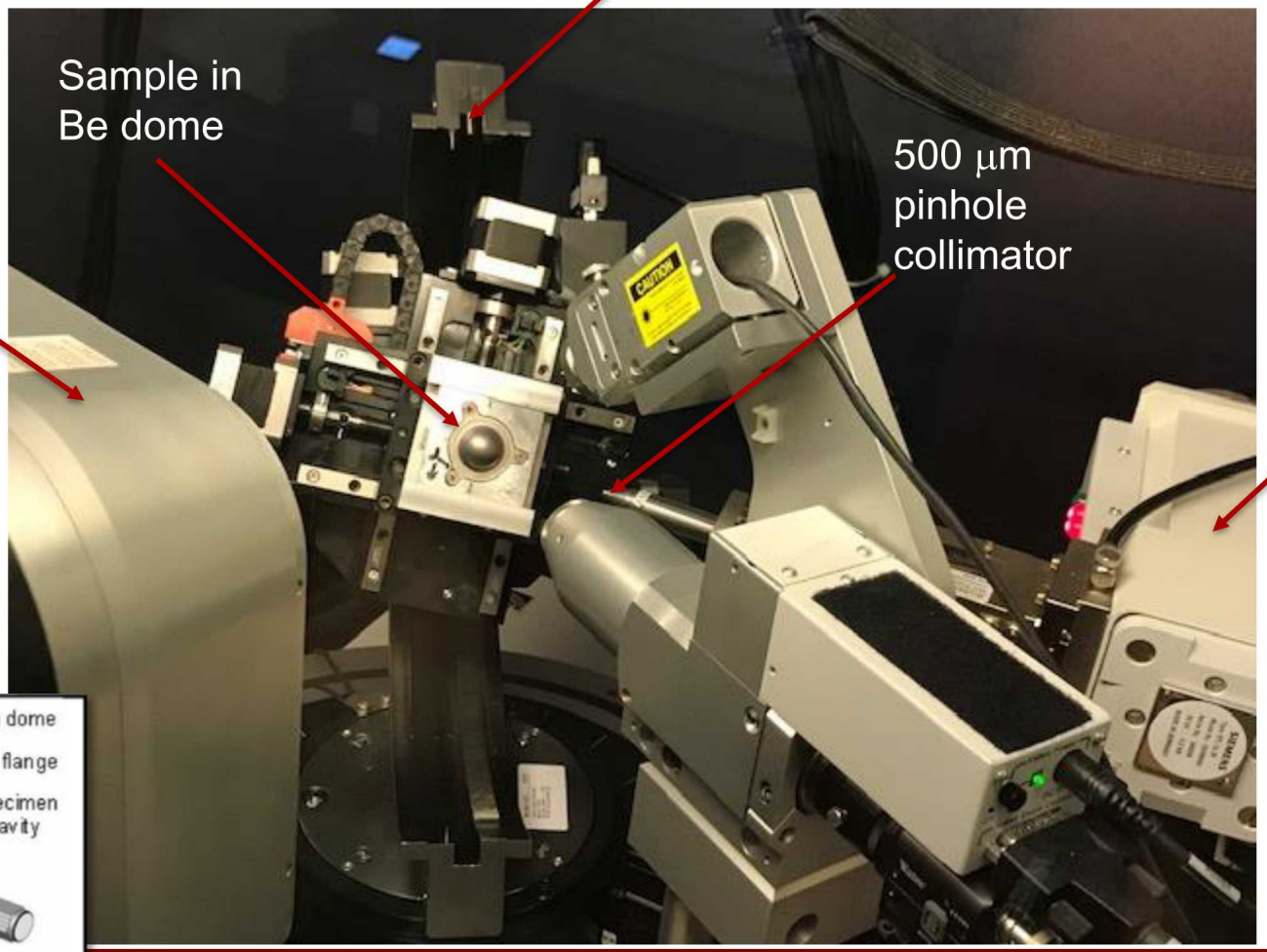
reference

TILT-A-WHIRL: a texture analysis package for 3D rendering of pole figures using MATLAB

Rodriguez, *et al.* (2013)
Powder Diffraction **28** (2), pp 81-89.

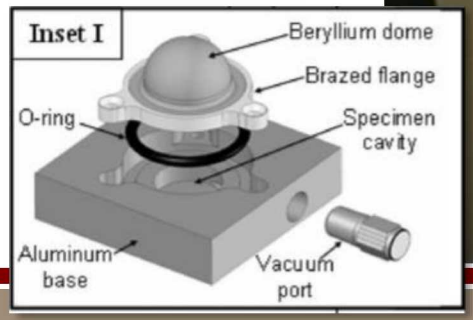
Setup for data collection employs Be dome holder on texture cradle

Sandia National Laboratories

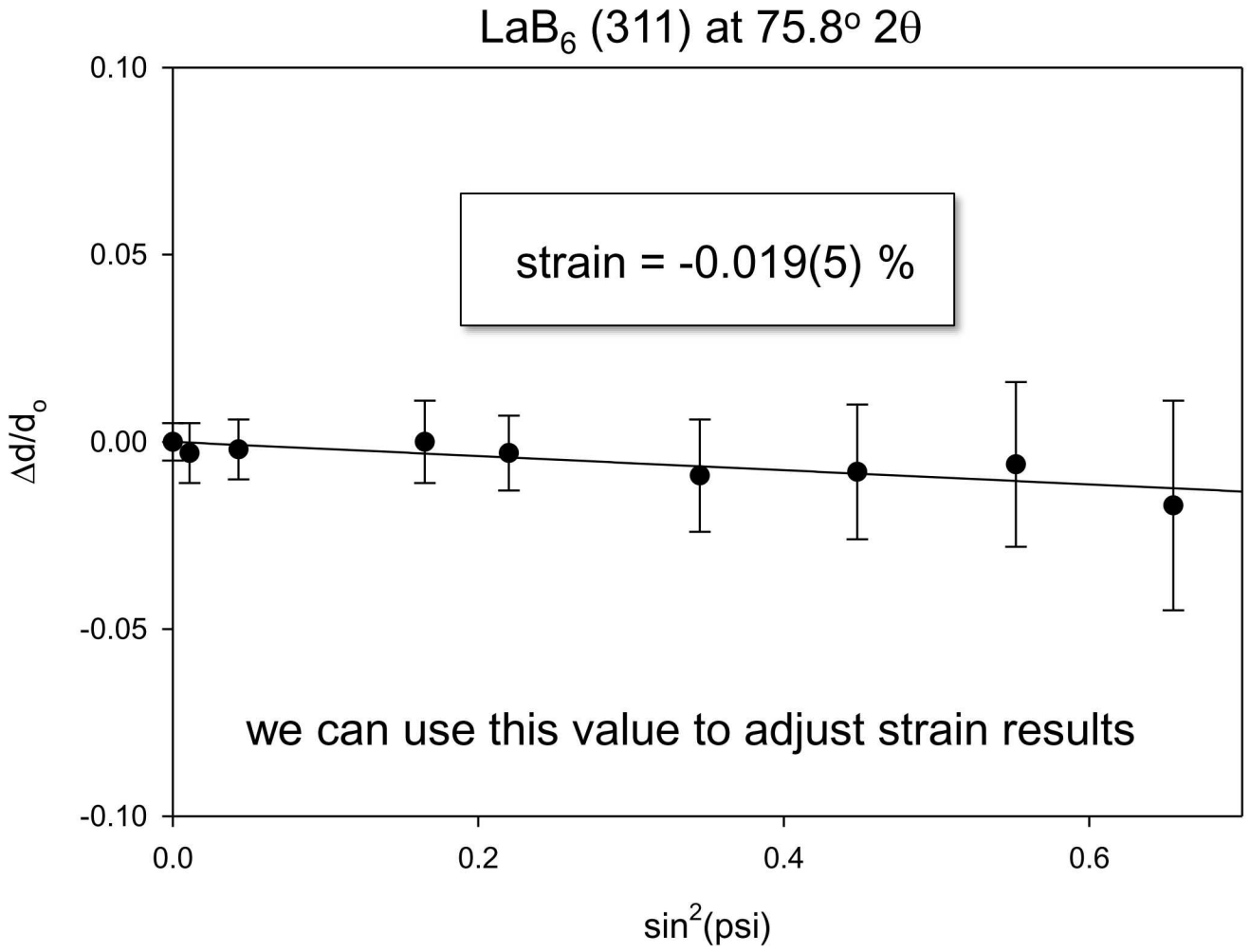


A beryllium dome specimen holder for XRD analysis of air sensitive materials

Rodriguez, *et al.* (2008) *Powder Diffraction* **23** (2), pp. 121-124.



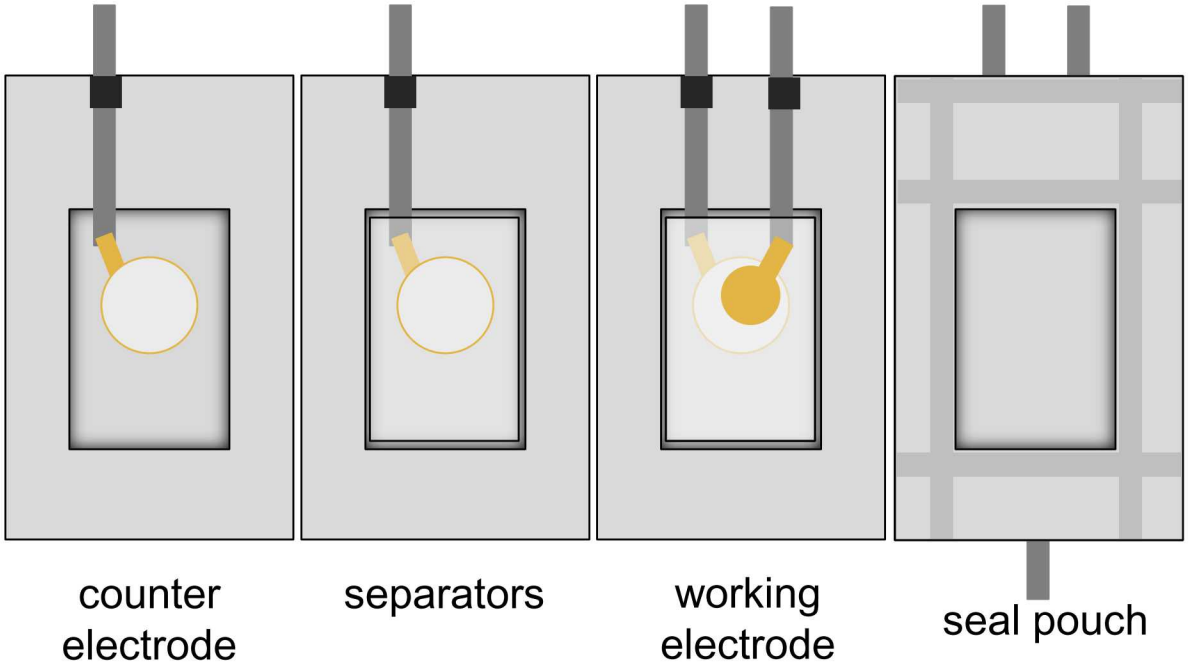
LaB_6 powder shows near zero strain



Pouch Cell Configuration

Electrolyte: 4 M lithium bis(fluorosulfonyl)imide:1,2-dimethoxyethane (4M LiFSI DME)

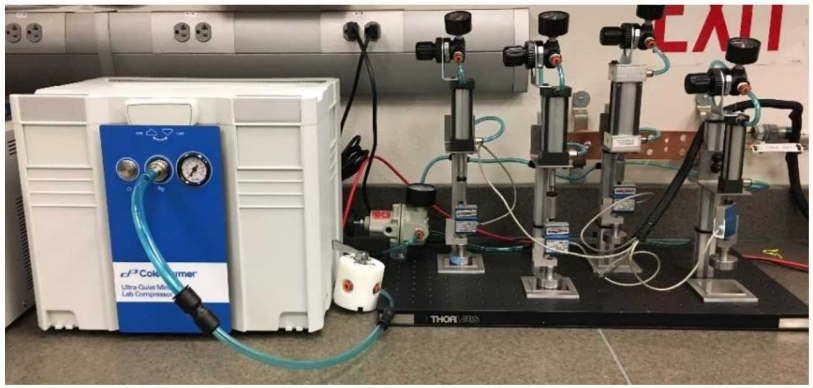
- Fabricate pouch cells and test
- Disassemble and extract working electrodes with electrodeposited lithium on it
- Wash with electrolyte solvent to remove salt
- Place in Be dome holder

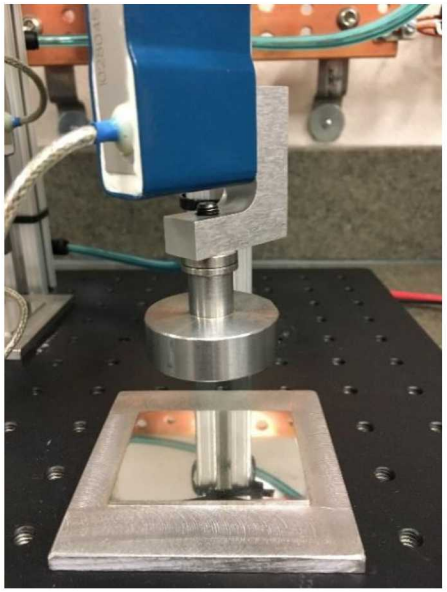


House-Built Four-Station Tester to test pouch cell batteries under compression

Sandia National Laboratories

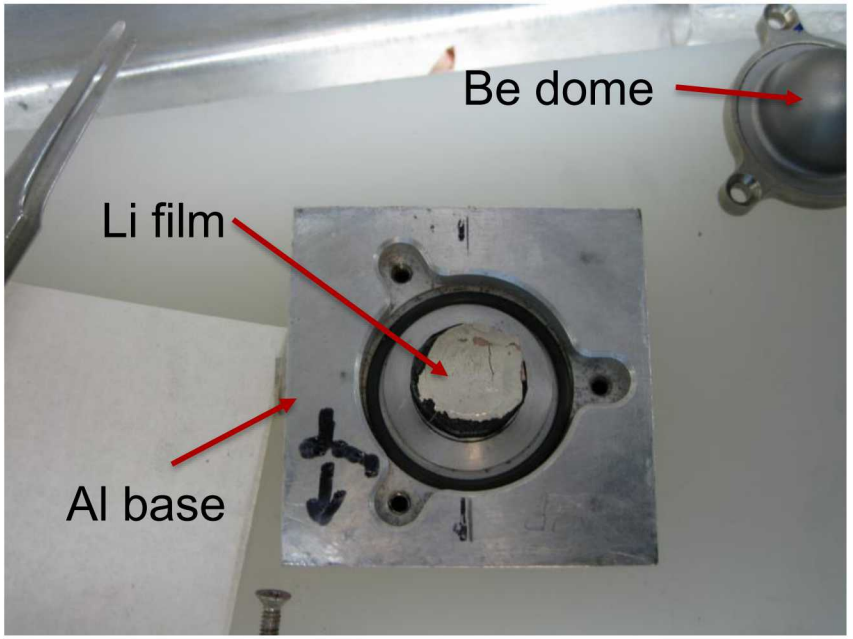
- Pneumatic Compression Cylinders
  - 2 Cylinders 100 lbf @ 100 psi
  - 1 Cylinder 44 lbf @ 100 psi
  - 1 Cylinder 123 lbf @ 100 psi
  - Individually controlled with independent regulators
- Ultra-Quiet (52dB) Mini Lab Compressor
  - Independent air supply MAWP 160 psi
- Interface 100 lbf Load Cells
  - BSC4 multi-channel bridge amplifier and signal conditioner
  - BlueDAQ user friendly interface for data acquisition
- Custom mirror-bright compression platens designed to fit/hold pouch cells





What do these Li films look like?

- Li Film on Cu substrate after cell dis-assembly.
- Sample is mounted in holder and sealed with Be dome
- All sample preparation is done within a glovebox (inert gas conditions).
- Surface of Al base serves as reference plane for sample height.



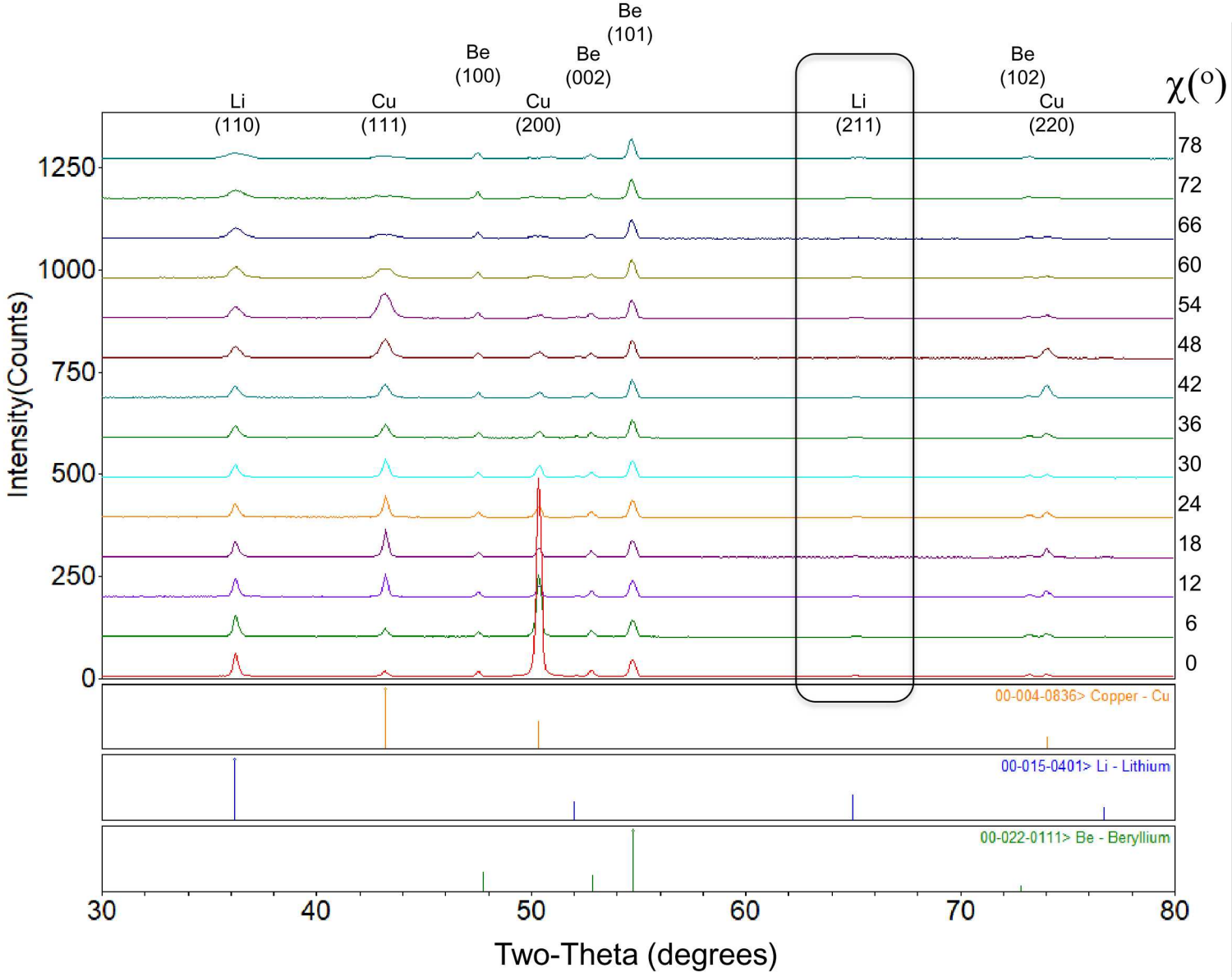
1000 kPa pressure Li film

Challenges to overcome to obtain data

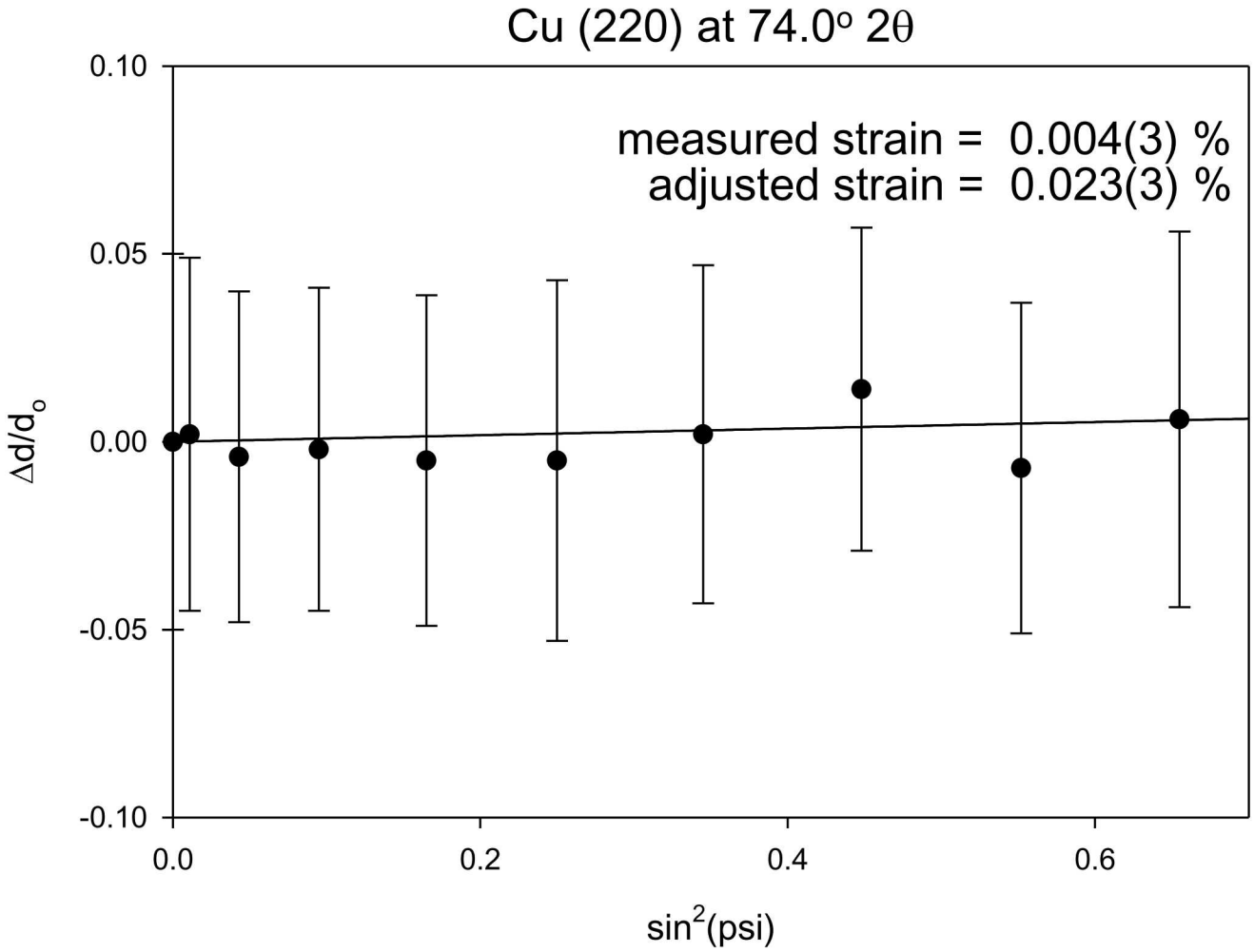
Sandia National Laboratories

- What if Li peaks are **too weak** to see?
  - analyze Li films with more thickness (e.g. 100 – 200 μm)
- What if Li film sample is **not flat**?
  - optimize sample to be as flat as possible and use only data for strain where χ tilt < 55° to avoid x-ray beam elongation on sample.
- Where are the Be and Cu peaks going to show up relative to the Li peaks and what if there is **peak overlap**?
  - run LaB_6 and Cu substrate material alone with and without the Be-dome enclosure to determine artifact peak locations
  - Verify expected Li peak positions do not overlap observed Cu and Be peaks
- How do I get sample positioned correctly if I **can't use the laser** for z-position (height) adjustment?
  - Pre-align holder with mock-up sample and keep track of location in x,y,z
  - perform final height adjust via Li peak location on actual film sample

Li (211) is not overlapped and would work for macrostrain



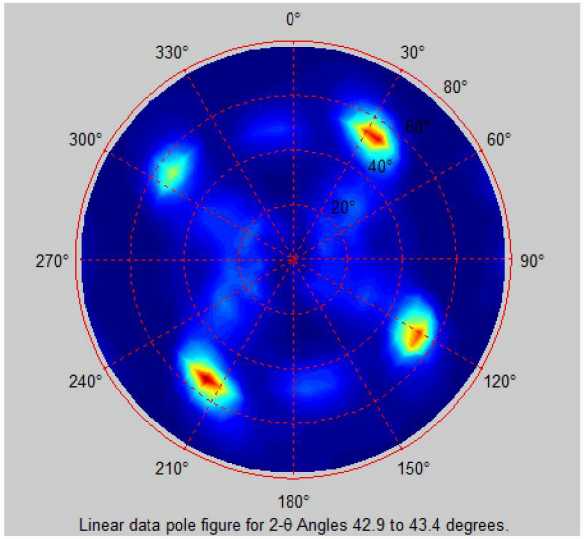
Cu under Li film is nearly unstrained



Cu substrate shows bi-axial texture

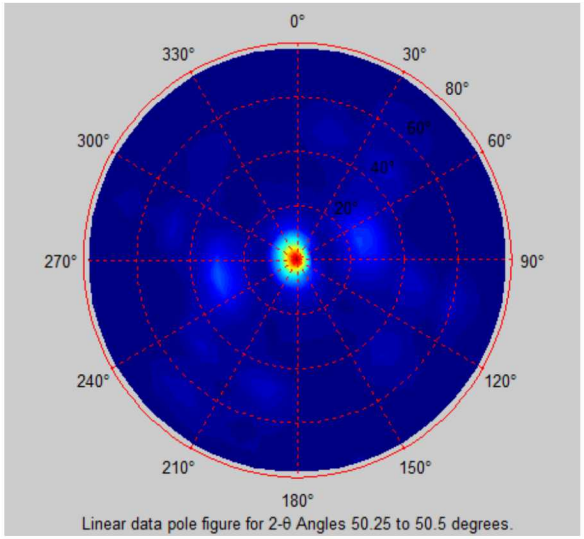
(111)

2D Pole Figure



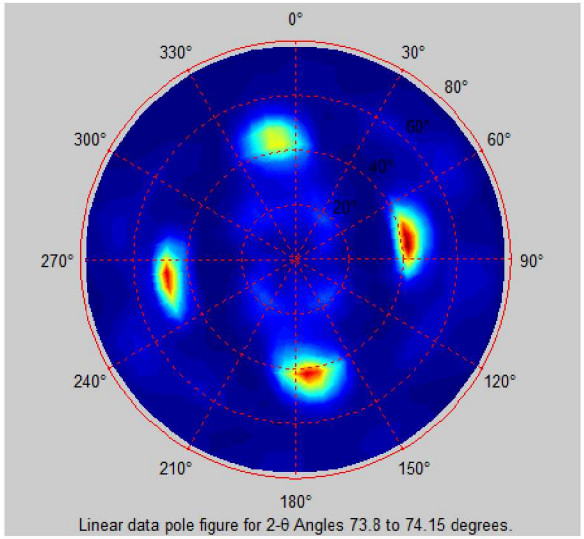
(200)

2D Pole Figure



(220)

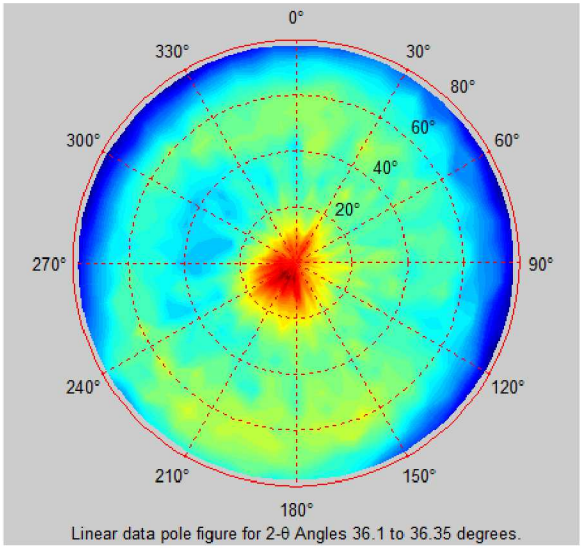
2D Pole Figure



Li substrate shows (110) out-of-plane texture and in-plane fiber texture

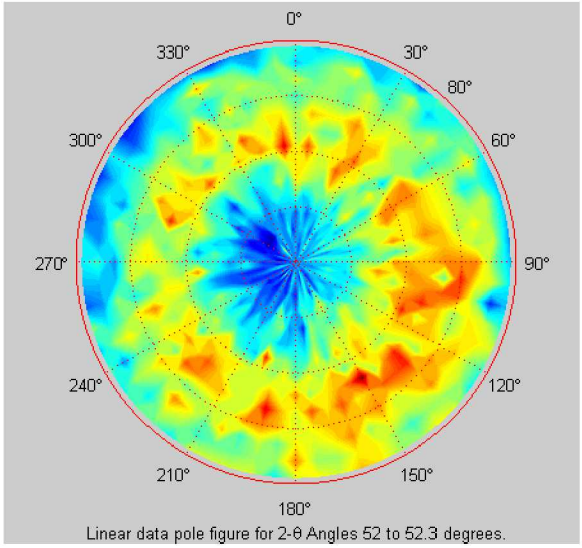
(110)

2D Pole Figure



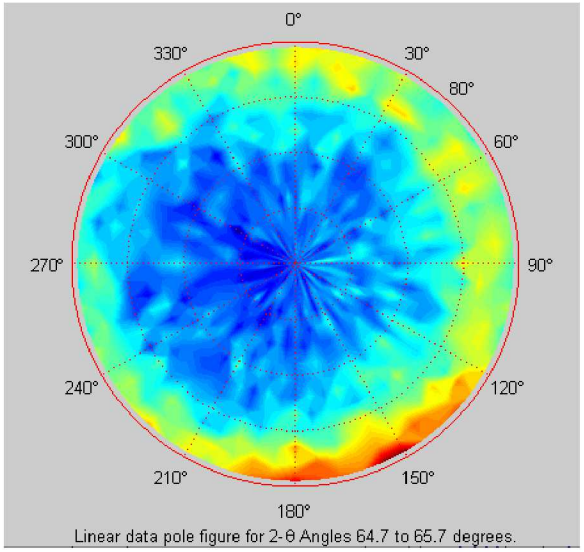
(200)

2D Pole Figure



(211)

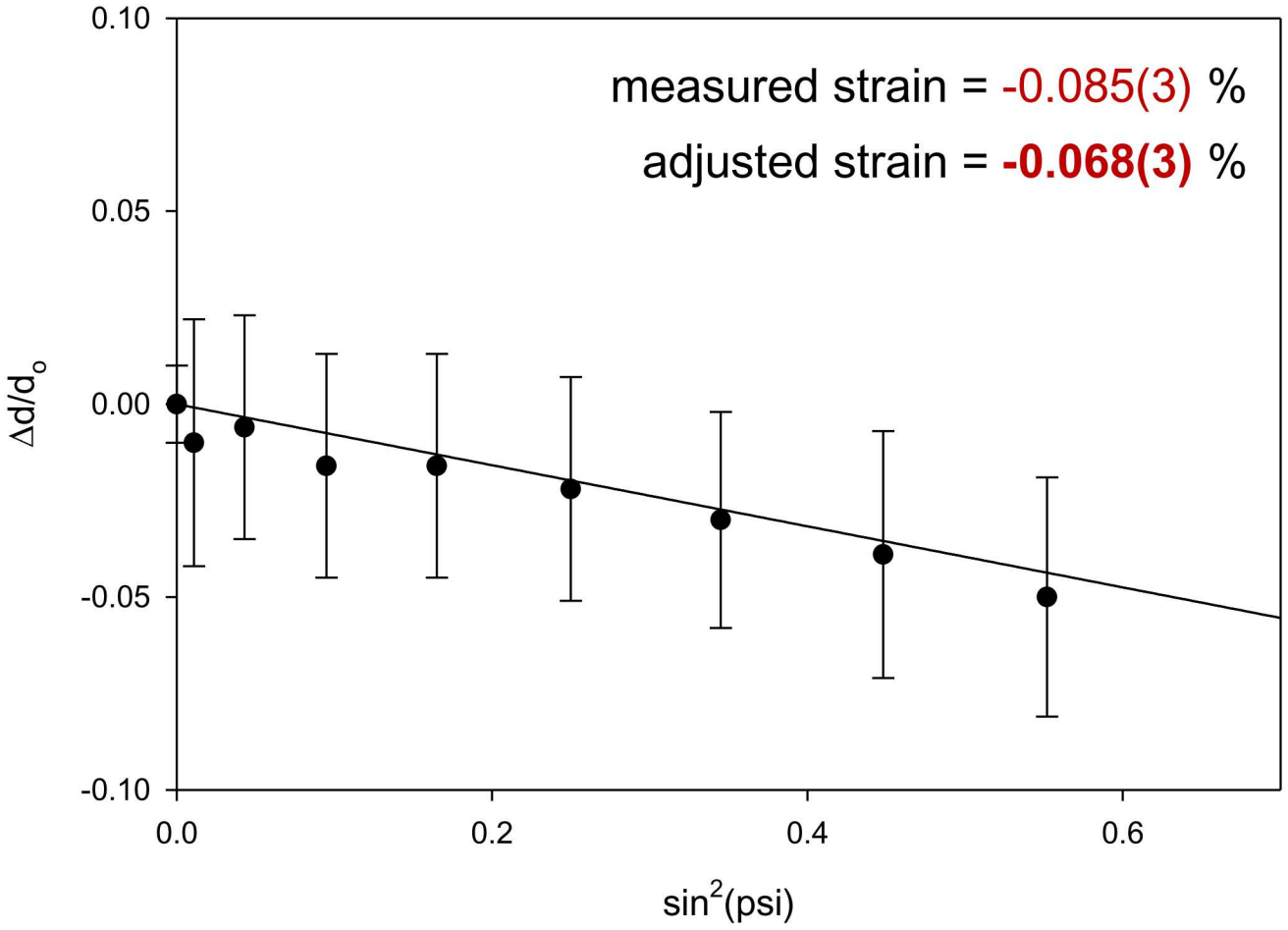
2D Pole Figure



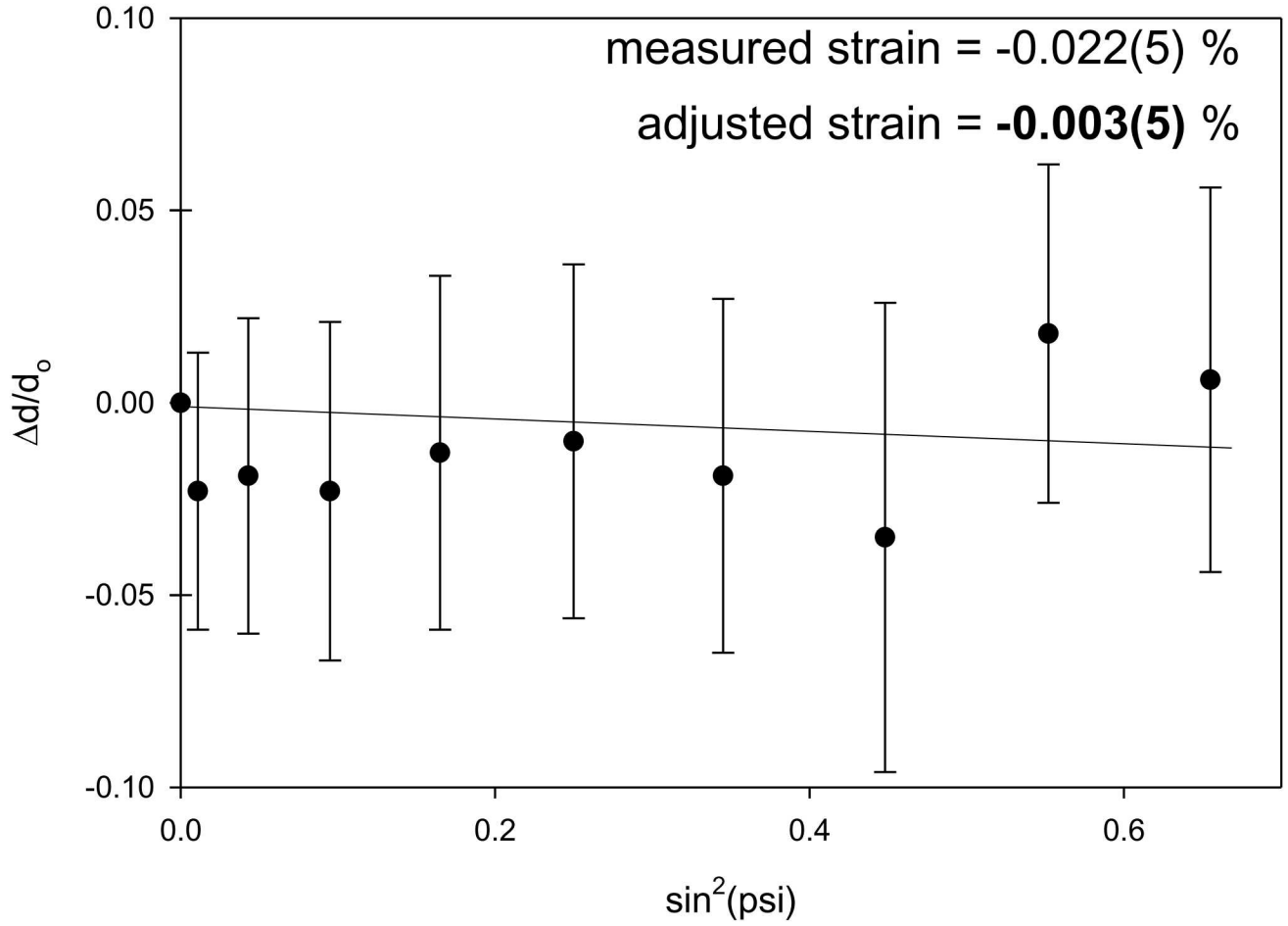
Li film from 1000 kPa cell shows a detectable compressive strain of approximately 0.07%

Sandia National Laboratories

Yeah!



Li film from cell with 10 kPa shows essentially no strain



Summary

Sandia National Laboratories

- We have demonstrated a means of determining residual strain in Li films via our Tilt-A-Whirl XRD methodology.
- The Be dome allows for isolation of the Li film during XRD measurement to prevent reaction of sample with atmosphere.
- Li Films exposed to high external pressure during cycling demonstrate a detectable in-plane compressive strain after removal from the pouch cell.
- Li films cycled at low pressure showed low or no detectable strain upon analysis after removal from the pouch cell.

Acknowledgments

- A special thanks to Kerry-Ann Stirrup for help with Tilt-A-Whirl data collection and analysis, Paul Cuillier for help pouch cell preparation,
- Supported in part by the Laboratory Directed Research and Development (LDRD) program at Sandia National Laboratories.
- Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.