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Temperature-dependent x-ray diffraction of thermographic phosphors

Caroline Winters

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D. R. Lowry, J. C. Miers,
C. Winters, L. E. Hansen, E. R. Westphal
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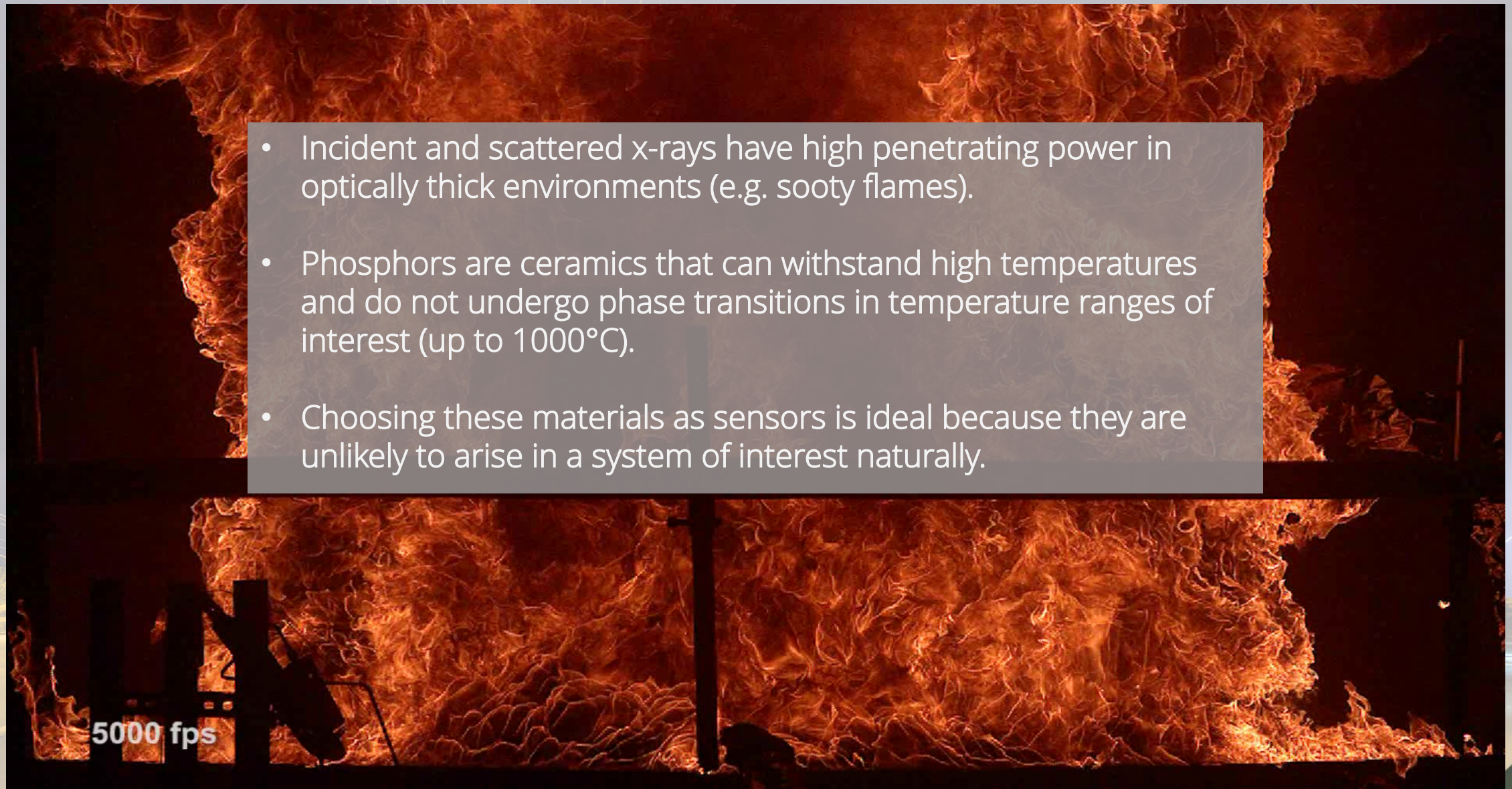
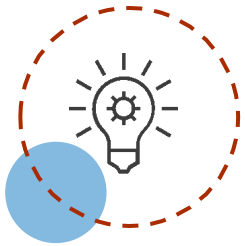
Extension of phosphor thermometry into complex environments can be hindered by optical obscurations, where UV/visible radiation is readily absorbed or occluded



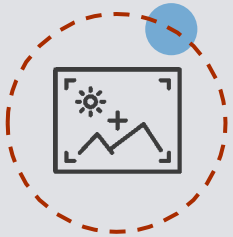
- Incident and scattered x-rays have high penetrating power in optically thick environments (e.g. sooty flames).
- Phosphors are ceramics that can withstand high temperatures and do not undergo phase transitions in temperature ranges of interest (up to 1000°C).
- Choosing these materials as sensors is ideal because they are unlikely to arise in a system of interest naturally.

The development of x-ray thermometry has relied on bright sources at user facilities or commercial instruments



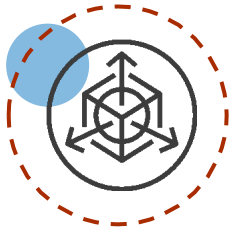
Flash X-ray diffraction

- Diffraction intensity
- Random phase shifts produce weaker diffraction lines
- [Morgan, D. V., et al., (2007)]



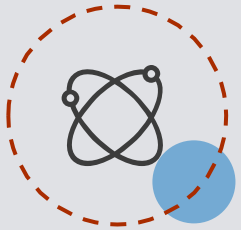
CT-based thermometry

- Temperature sensitivity due to volumetric changes in the material density
- Difficult to reproduce
- [Fani, F., et al., (2014)]



X-ray Scattering

- LINAC source
- [Sellberg, J. A., et al., (2014)]
- Synchrotron source
- Interference patterns from electronic elastic scattering
- [Rahman, N., et al., (2021)]
- [Halls, B. R., et al., (2021).]



Thermographic X-ray Fluorescence

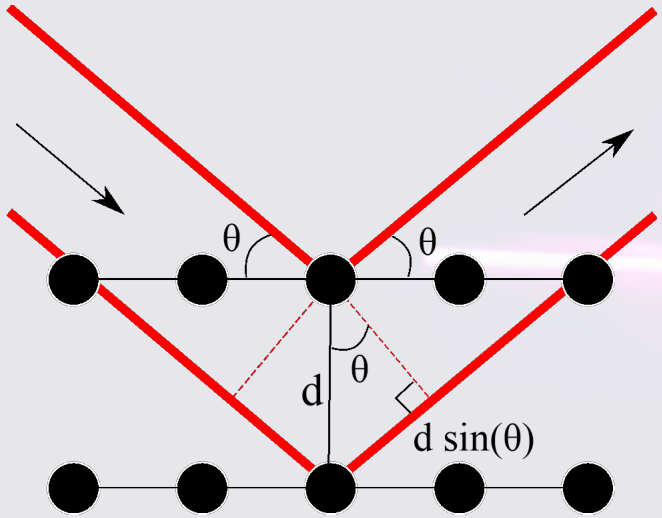
- Synchrotron source
- Trends observed, but wide spread in data uncertainty
- [Westphal, E. R., et al., (2019)]

Variable-Temperature X-ray Diffraction

- Commercial instrument
- Angle-dispersive x-ray diffraction
- [Halls, B., et al., (2022)]

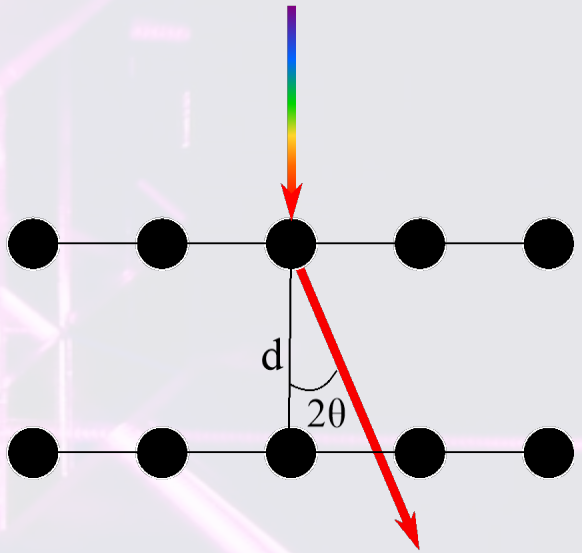
X-ray diffraction relies on Bragg's law of crystal diffraction

Angle-dispersive x-ray diffraction (ADXRD)



- Single x-ray wavelength (energy), collection at many angles
- Constructive interference occurs for $n\lambda = 2dsin(\theta)$

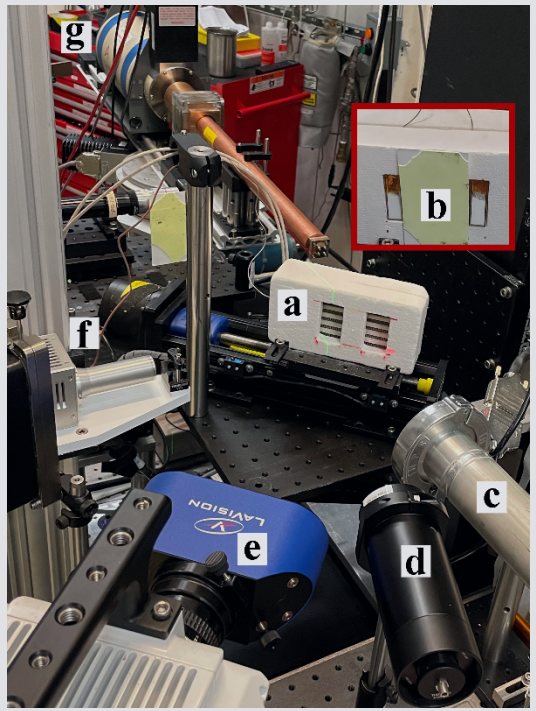
Energy-dispersive x-ray diffraction (EDXRD)



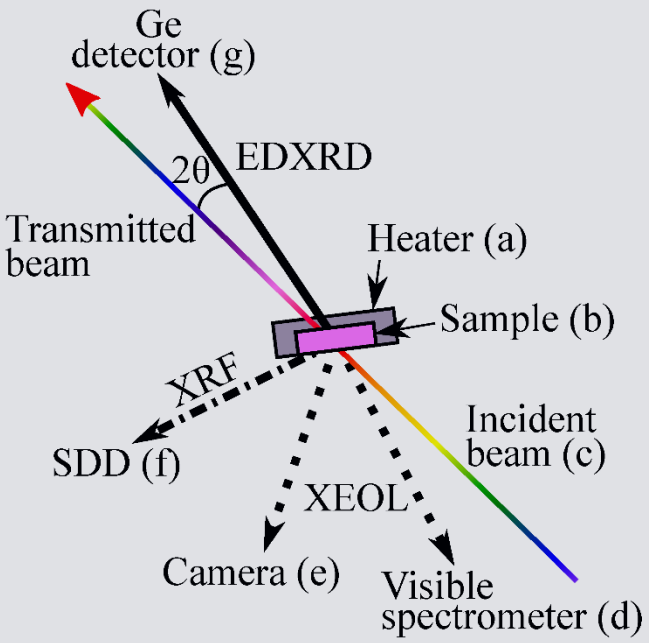
- Polychromatic x-ray beam, collection at a fixed angle (θ)
- Constructive interference occurs for $E = \frac{nhc}{2dsin(\theta)}$

Preliminary EDXRD studies were carried out at the Advanced Photon Source synchrotron at Argonne National Laboratory [1]

Experimental Set-up



Experimental Schematic



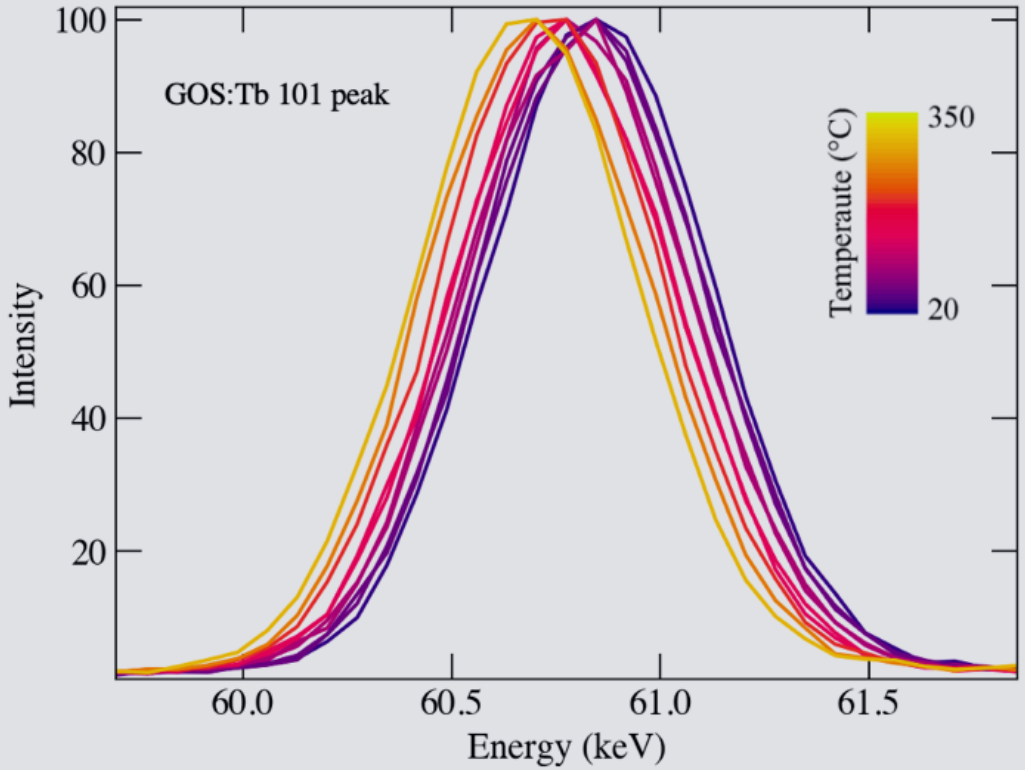
- Polychromatic x-ray beam energy range of 5 – 150 keV.
- Fixed diffraction angle of 2θ = 3.9°.
- Diffracted x-rays are collected with a high-resolution energy-dispersive semiconductor Germanium detector.
- Data is collected between room temperature and 300°C in 50°C increments.
- Terbium doped Gadolinium Oxysulfide (GOS:Tb) phosphor

EDXRD: energy-dispersive x-ray diffraction
XRF: x-ray fluorescence
XEOL: x-ray excited optical luminescence
SDD: silicon drift diode x-ray detector
Ge: germanium x-ray detector

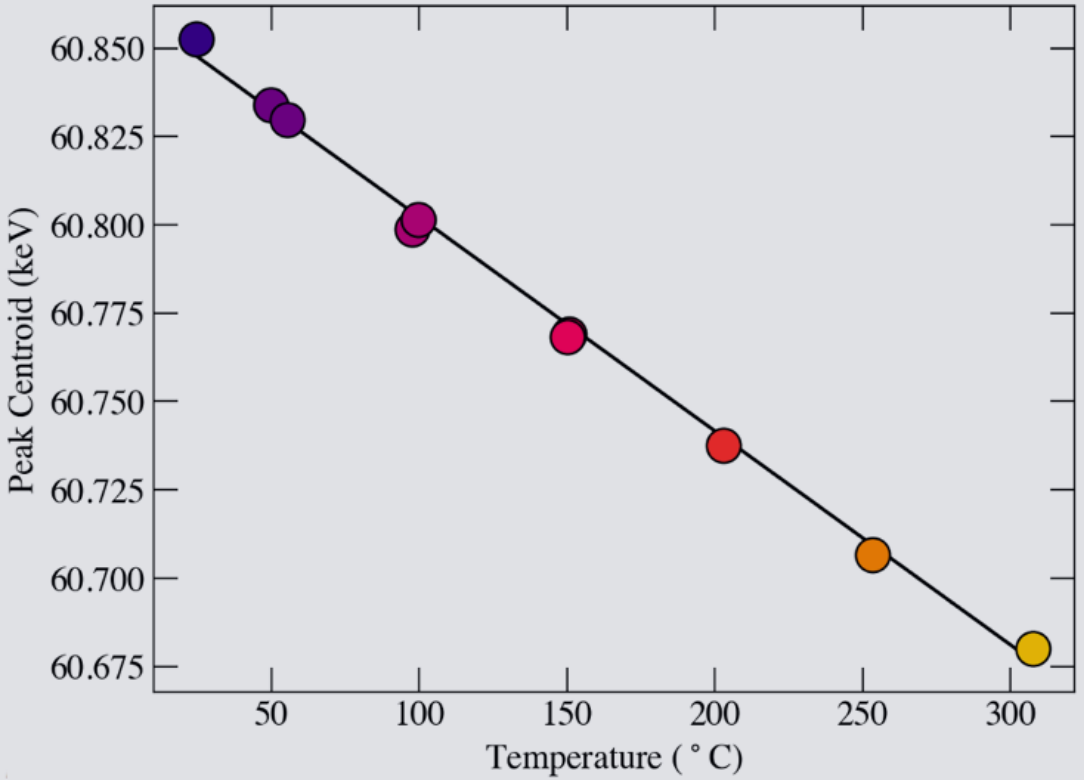
[1] L. E. Hansen et al, J. Appl. Phys. 132, 065105 (2022)

Clear thermographic behavior was identified in the EDXRD spectra of GOS:Tb with 1% energy resolution [1]

GOS:Tb 101 EDXRD peak



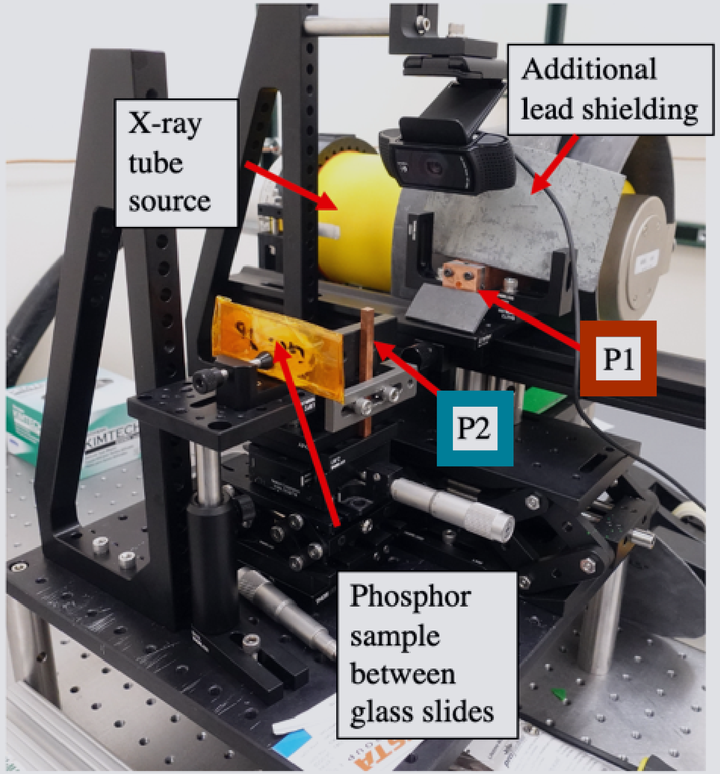
Peak centroid versus temperature

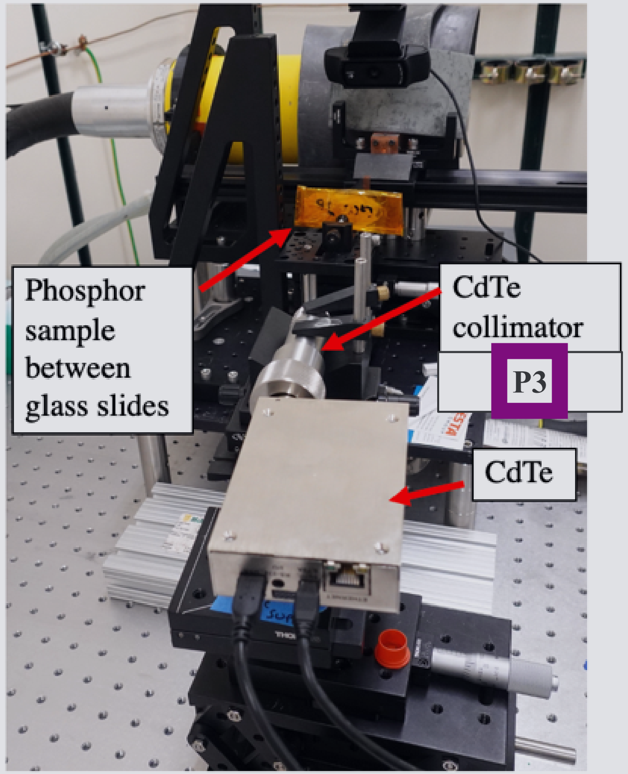


As temperature increases, the crystal expands, and the peaks shift to lower energy

[1] L. E. Hansen et al, J. Appl. Phys. 132, 065105 (2022)

We are developing fieldable EDXRD in a laboratory setting



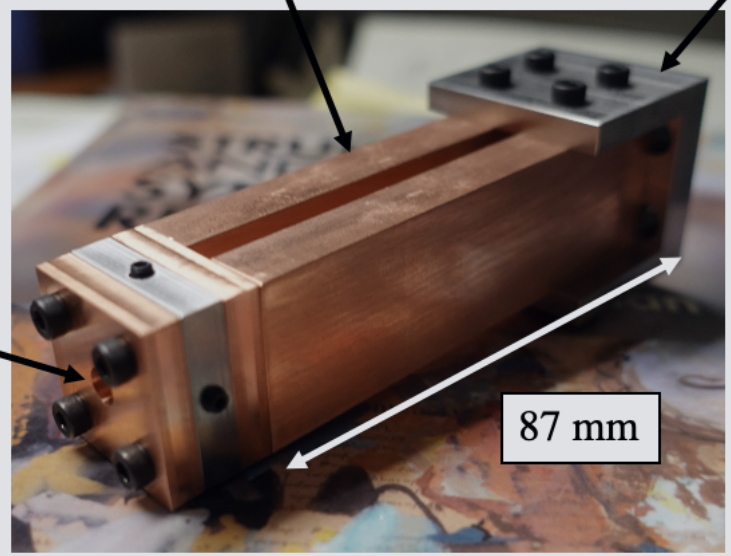


- Polychromatic 225 keV, 15 mA Phillips MCN x-ray tube.
- Fixed diffraction angle of 2θ = 6°.
- Diffracted x-rays are collected with a portable CdTe detector (Amptek, XR-100).
- P1: Source Pinhole
- P2: Sample Pinhole
- P3: Detector Pinhole

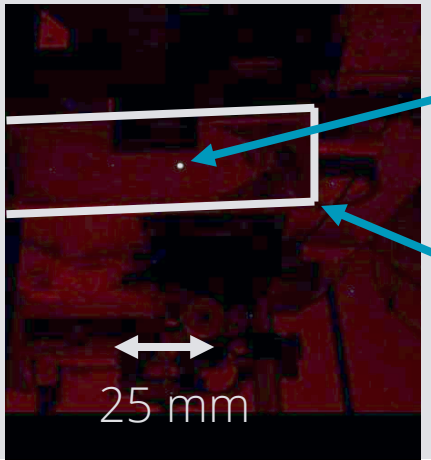
Challenges:

- We lack the flux achievable at a synchrotron.
- CdTe detectors have lower resolution than the Ge detector used at the synchrotron.
    - It can be placed as close to the sample as desired.
- Tube sources are highly dispersive, so the x-rays must be heavily collimated.

An adaptable collimator was machined that mounts to the head of multiple x-ray tube sources



This collimator will produce a 0.5—2 mm x-ray beam spot with exchangeable pinholes.

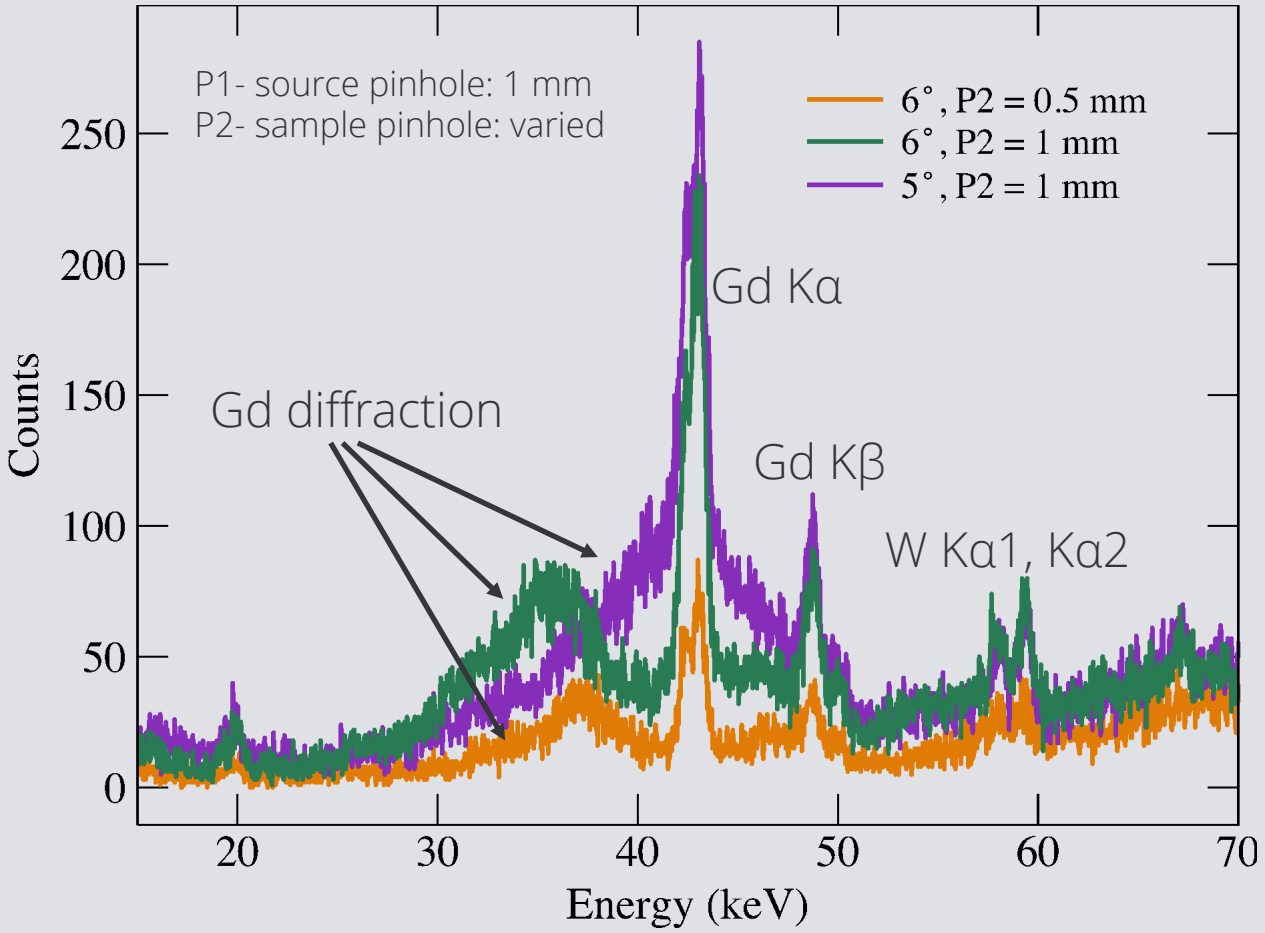


Glowing spot on a phosphor sample produced by the collimated x-rays

Outline of glass slides

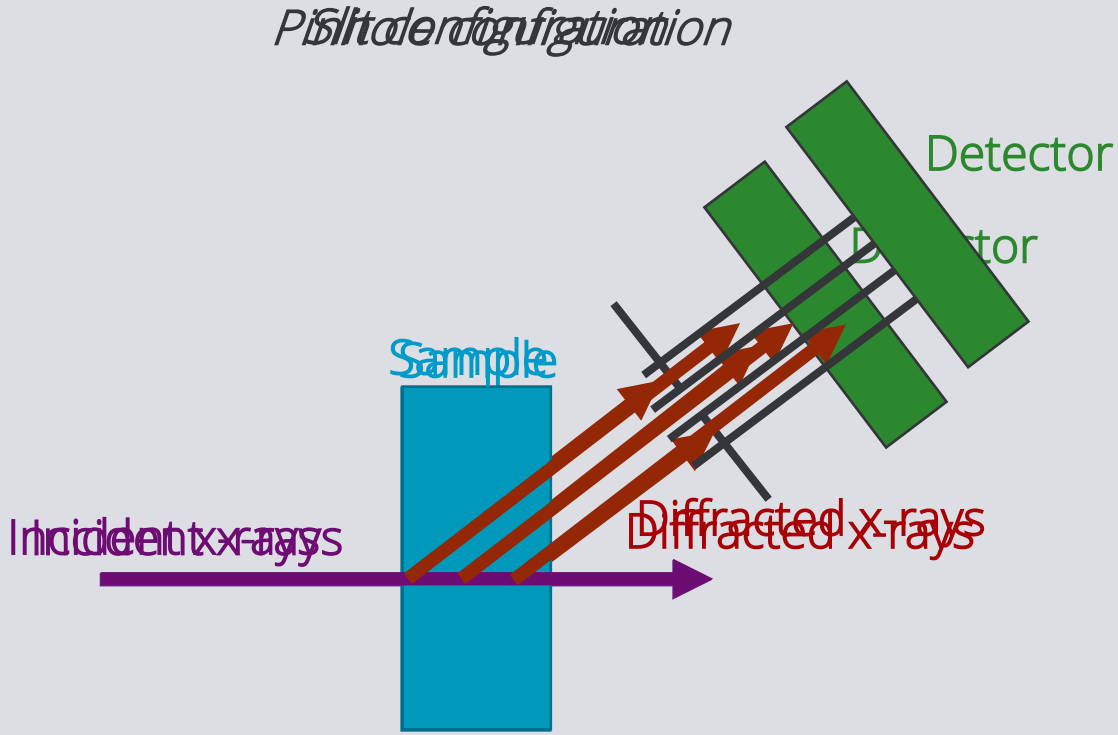
The first diffraction measurements showed poor resolution

Compare diffraction angles and source pinhole sizes



- 3 minute integration time
- Detector pinhole (P3) = 1 mm
  - Collimated signal into CdTe
  - Resolution is 13%.
- In order to achieve 3% resolution: *1000 µm → 25 µm CdTe pinholes.*
- At 3% resolution, the diffraction x-ray signal of GOS:Tb is too weak.

Ongoing work to improve signal will use more of the detector area

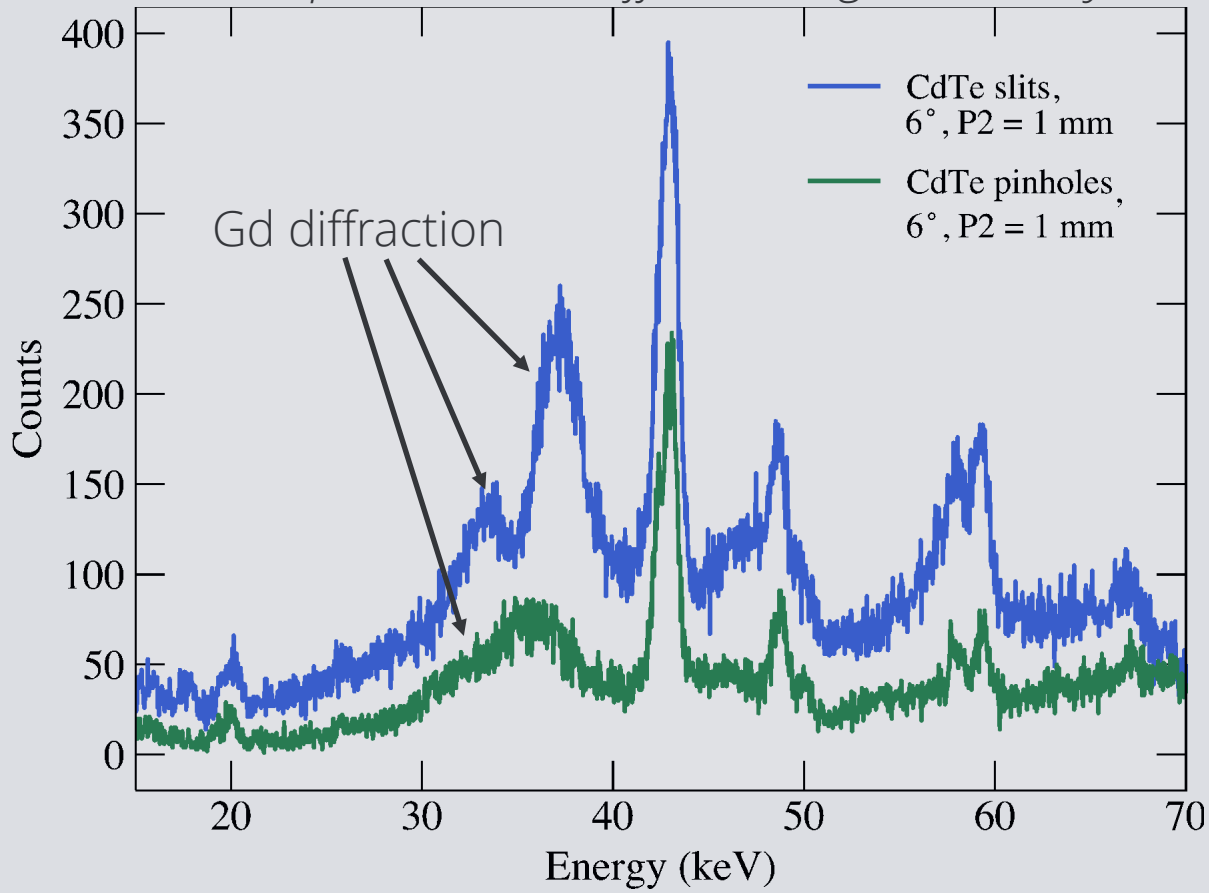


Moving to soller slit collimation on the CdTe rather than pinholes will allow us to collect more photons and utilize a larger area of our detector [2].

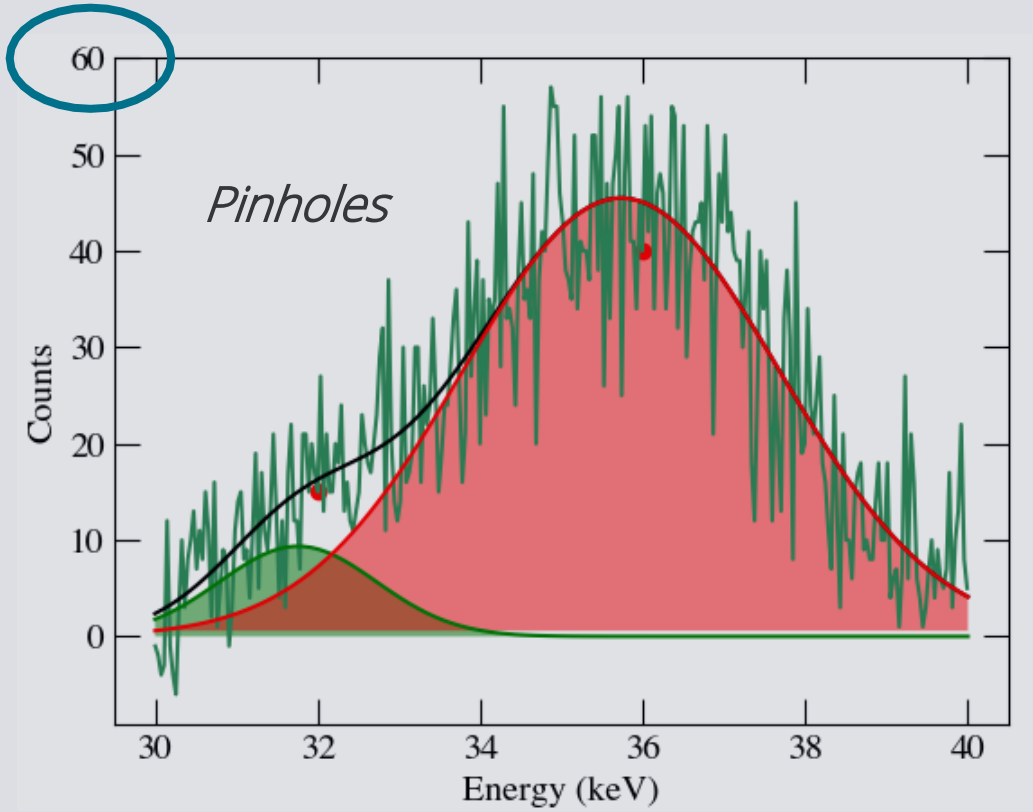
[2] Y. Chen et al, AIP Advances 8, 105113 (2018).

Slit configuration improved the diffraction resolution and signal levels

Compare signal through slits and pinholes --
4x improvement in diffraction signal intensity

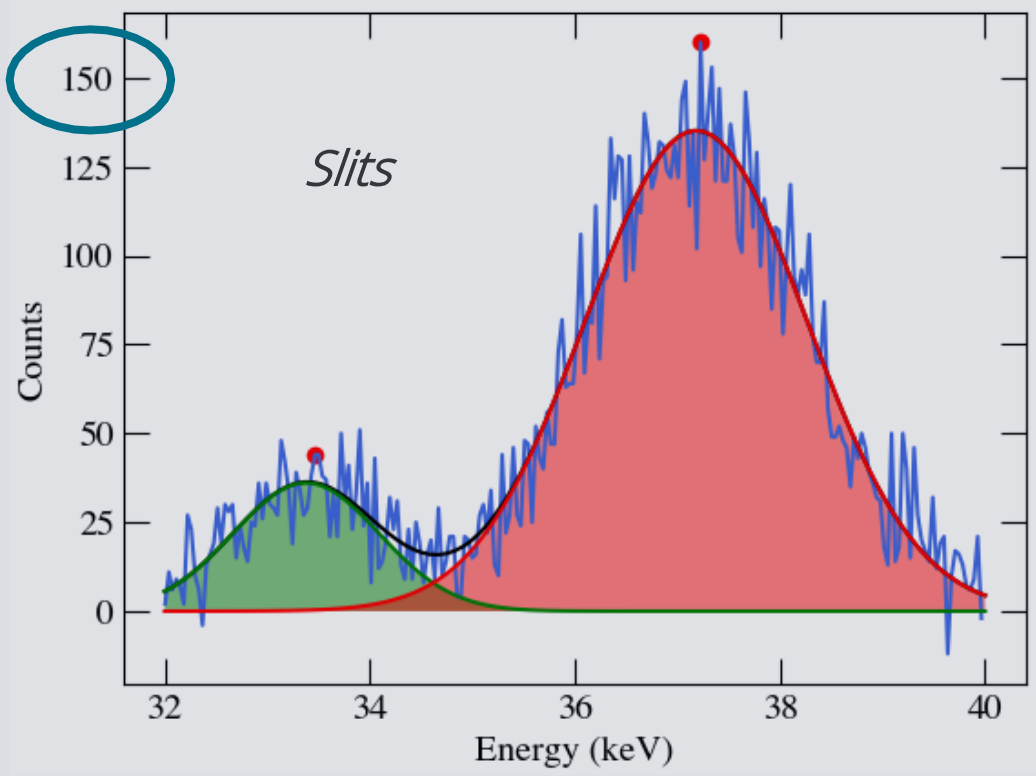


Gaussian fits quantified energy resolution improvement



Most intense peak:

$$\frac{\Delta E}{E} = 13\%$$



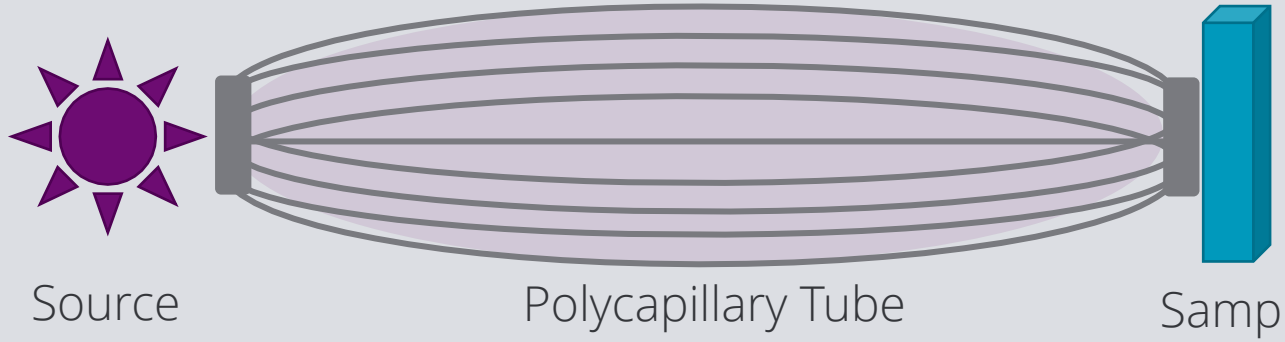
Most intense peak:

$$\frac{\Delta E}{E} = 7\%$$

Future work to improve resolution and signal

- The soller slits tested were off-the-shelf Cu slits, which are made for Cu Ka energy (8 keV). Our high-energy x-rays are not well absorbed by Cu. We are having custom W slits machined.

- Polycapillary x-ray focusing optics are being explored to increase flux.



- A lower-Z phosphor (ZnO:Ga) will allow us to use a thicker sample and achieve more scattering with less attenuation.

Temperature-dependent x-ray diffraction of thermographic phosphors

This work was supported by the Laboratory Directed Research and Development program at Sandia National Laboratories.

Thank you!

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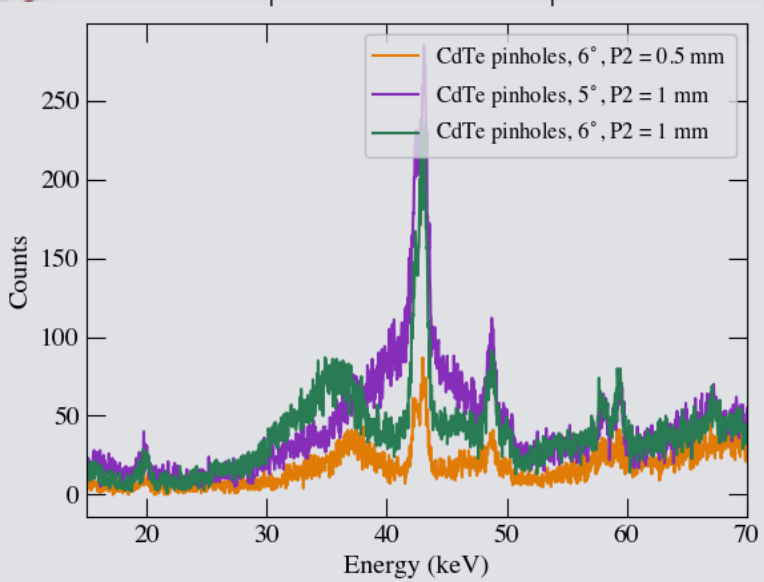
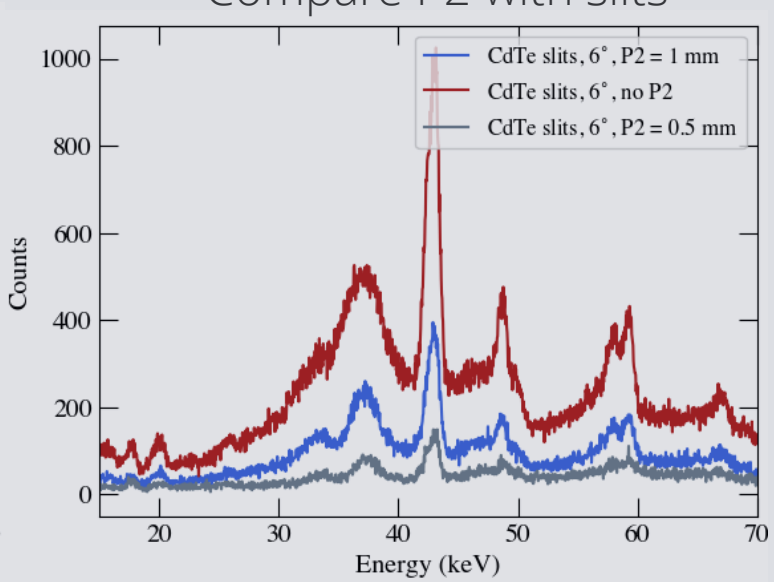
Backup slide

Compare P2 with pinholes

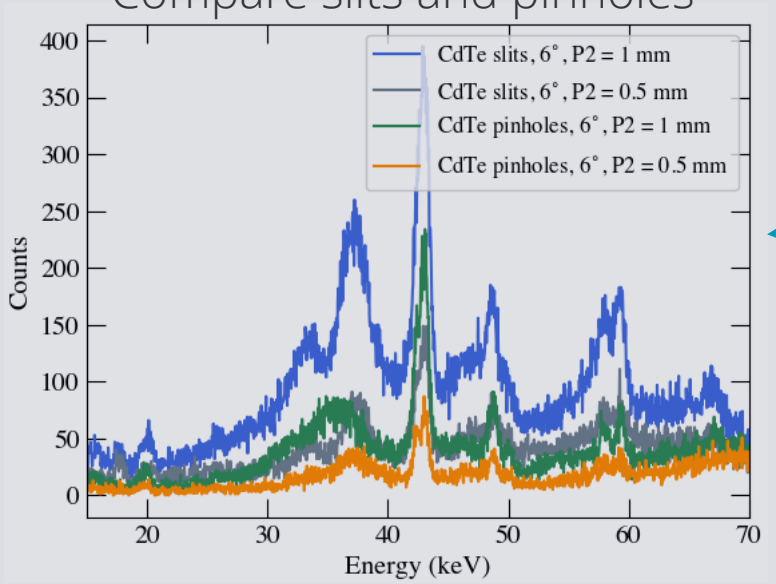


Compare P2 with slits



All EDXRD shown has 3 minute integration time (not showing the one longer scan)

Compare slits and pinholes



This looked busy to me show I just showed the P2=1mm data

These are convolved Gaussian fits