



Heat transport across warm dense matter interfaces

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Yuan Ping	LLNL		
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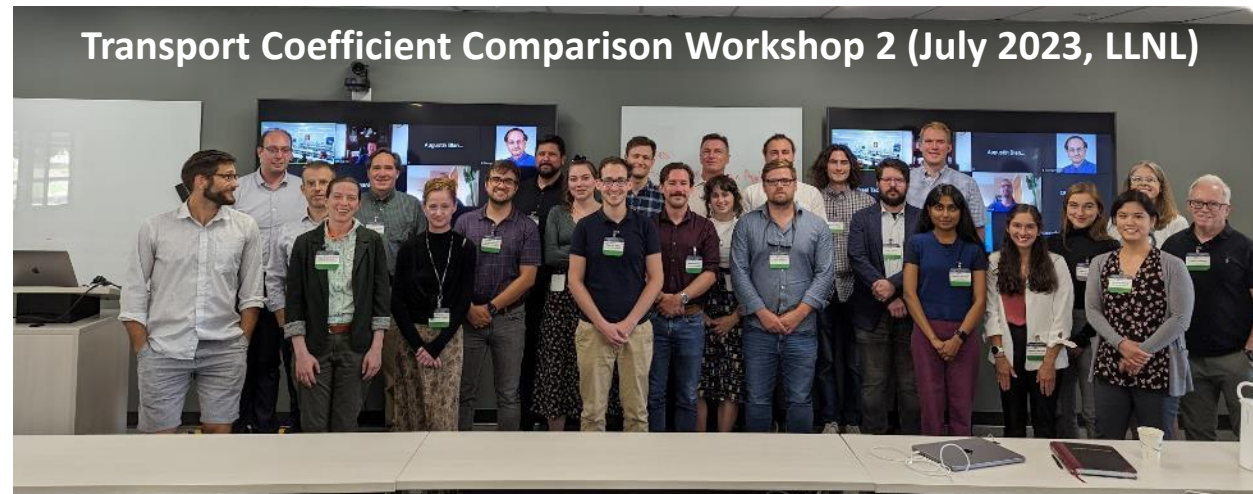
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University of Nevada, Reno

Physics of Non Ideal Plasmas
Tuesday | 17th September 2024



NSF Award
PHY-2045718

Key Question: Can we directly measure transport properties in warm dense matter?

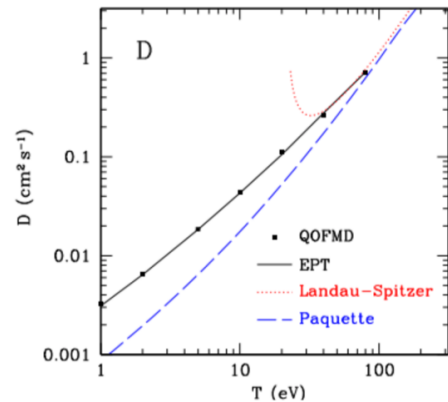
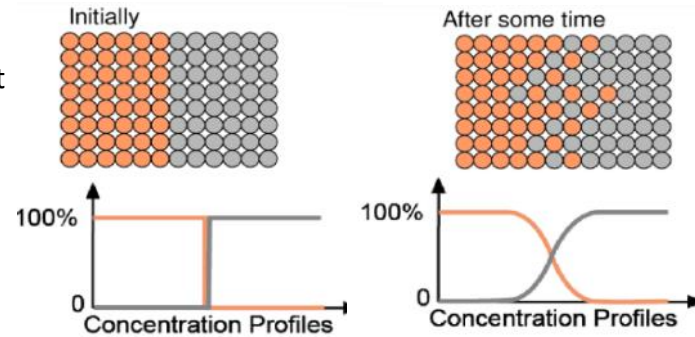


“transport coefficients including thermal and electrical conduction, electron–ion coupling, inter-ion diffusion, ion viscosity, and charged particle stopping powers.”

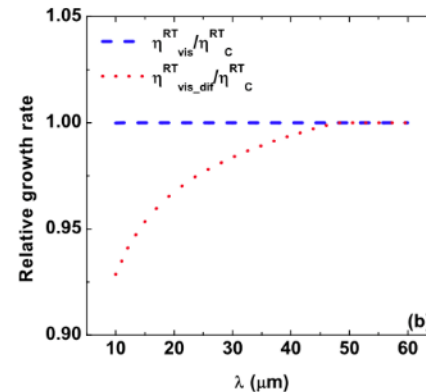
Motivation + Transport Properties

Particle Diffusion

- Mutual diffusion refers to movement of ions in response to concentration gradients.
- It is the basic microscopic quantity that governs the atomic level mixing of matter.
- The mutual diffusion coefficient is a fundamental dynamical plasma property.
- Diffusion is one of many processes hampering performance of ICF efforts.
- It is necessary to understand models of the interiors of Jovian planets.
- Large differences exist between different theoretical techniques.
- No experiments have attempted to measure diffusion properties in WDM.



Theoretical coefficients of self-diffusion in a deuterium plasma at 4.04 g/cc J. Daligault *et al.* PRL 116 (2016)

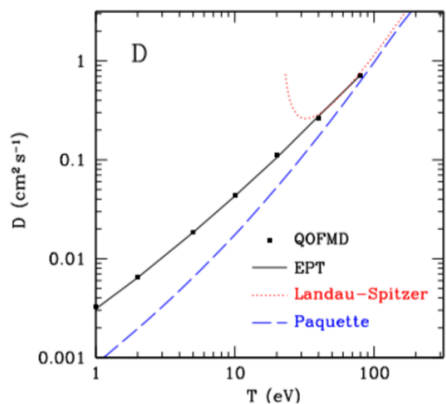
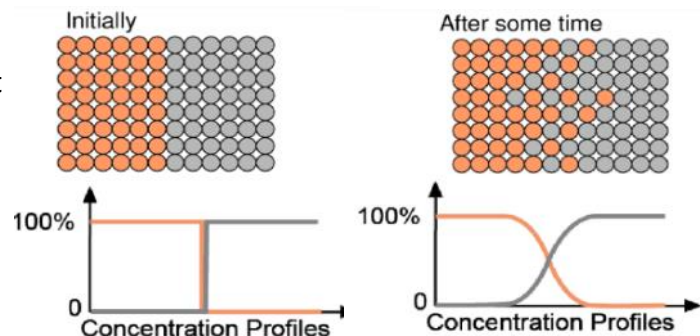


Growth rate of the RT instability with and without diffusion at a beryllium-deuterium interface. C. Wang *et al.* Physics of Plasmas 22, 102702 (2015)

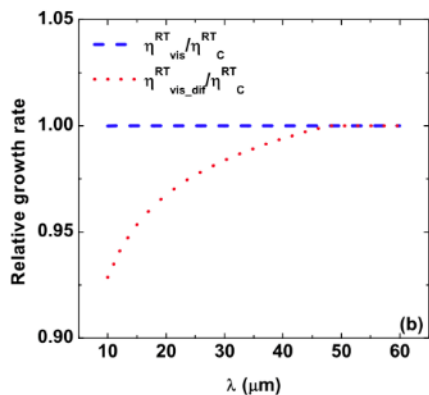
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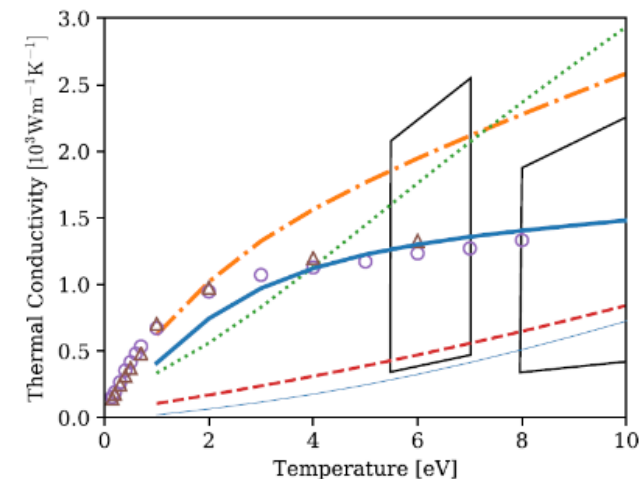
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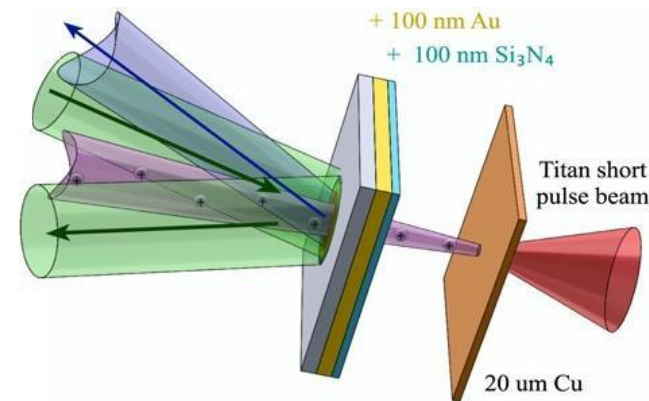
Growth rate of the RT instability with and without diffusion at a beryllium-deuterium interface. C. Wang *et al.* Physics of Plasmas 22, 102702 (2015)

Thermal Diffusion

- Thermal diffusion refers to transport of heat in response to temperature gradients.
- It is the basic microscopic quantity that governs the atomic level energy transport.
- In planetary interiors, thermal conductivity plays a vital role.
- The design of inertial confinement fusion (ICF) implosions depends on the thermal conductivity of both the ablator materials and the fuel mixture.
- Large differences exist between different theoretical techniques.
- Few experiments have attempted to measure thermal conductivity properties in WDM.



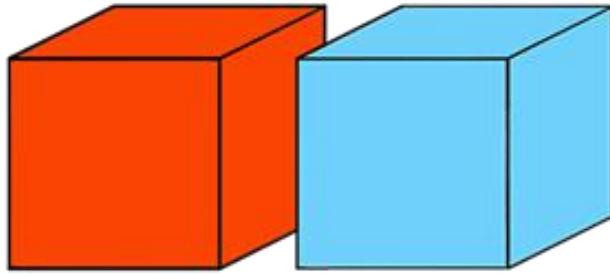
Thermal conductivity as a function of temperature for Al at solid density of $\rho = 2.7 \text{ g/cm}^3$, for a variety of models. The black boxes are experimental (McKelvey *et al.* Shaffer and Starrett, Phys Rev E 101, 053204 (2020))



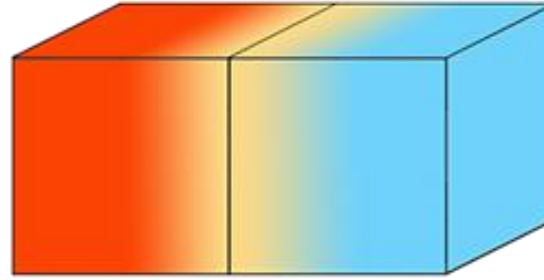
A. Mcelvey *et al.* Scientific Reports volume 7, Article number: 7015 (2017)

Thermal Conductivity affects the temperature profile of a system

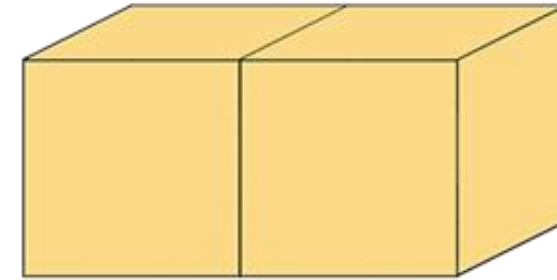
1. Not Touching - No Conduction



2. Conduction begins:
heat transfers to colder object

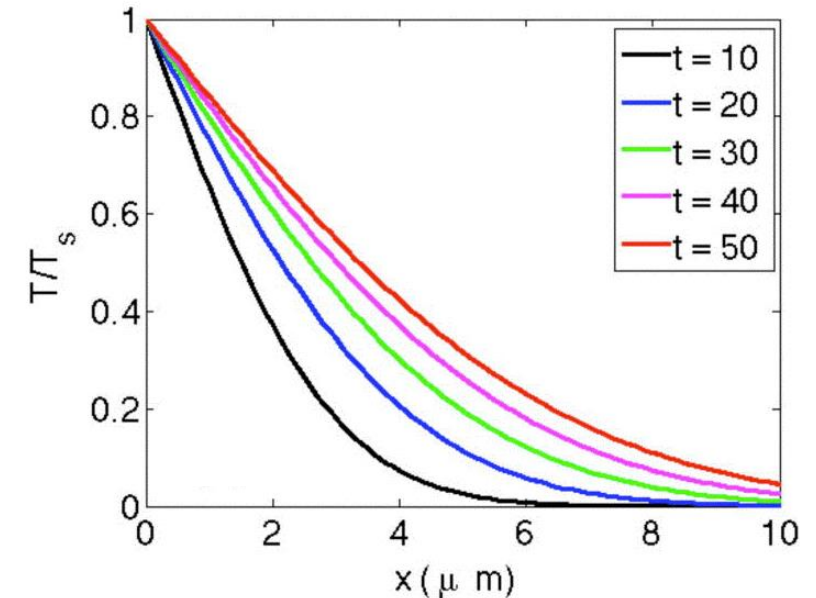


3. Conduction completed:
Two objects are in equilibrium



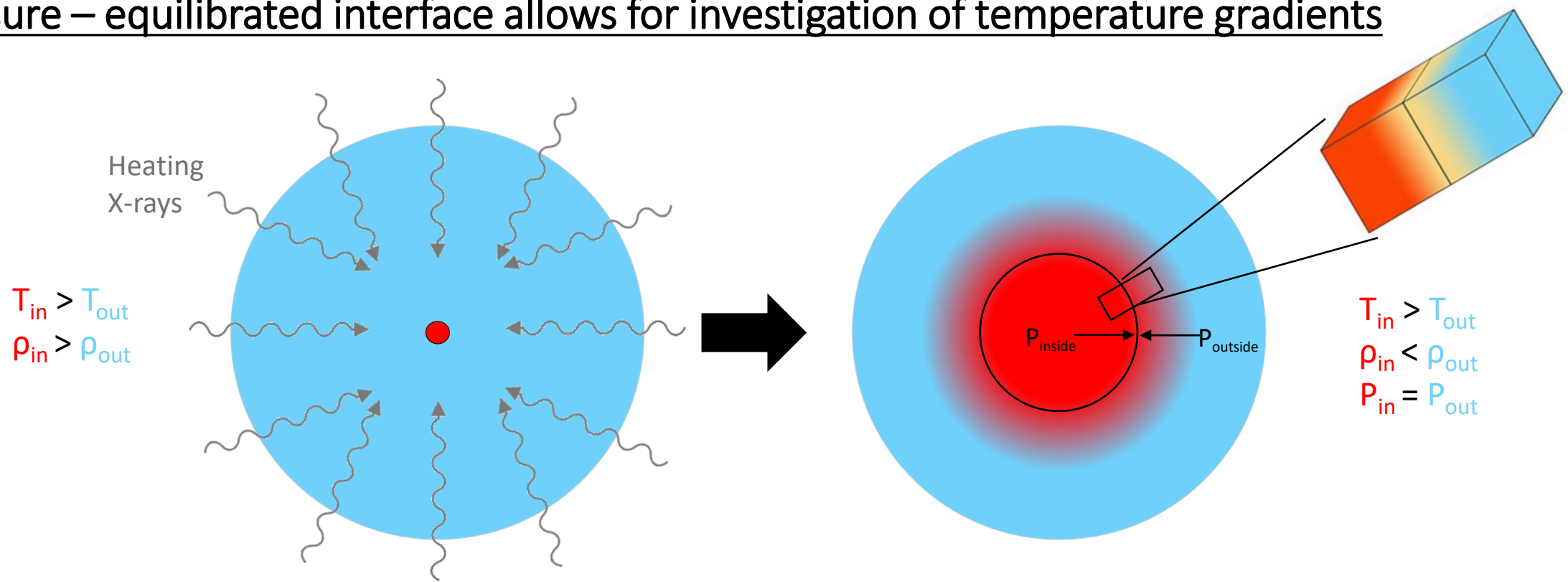
- Thermal conductivity refers to a material's ability to transport heat and is the basic microscopic quantity that governs the atomic level energy transport.
- How much heat is conducted, and how quickly it conducts, are dependent on the materials involved and the initial temperature gradient
- Characteristic scales: $\Delta r \approx \sqrt{\alpha \Delta t}$

- For WDM materials: $\Delta t \sim ns$, $\alpha = \frac{\kappa}{\rho C_P} \sim cm^2/s \rightarrow \Delta r \sim \mu m$



Temperature ratio as a function of distance, at various time delays.
Y. Ping *et al.* Phys. Plasmas 22, 092701 (2015)

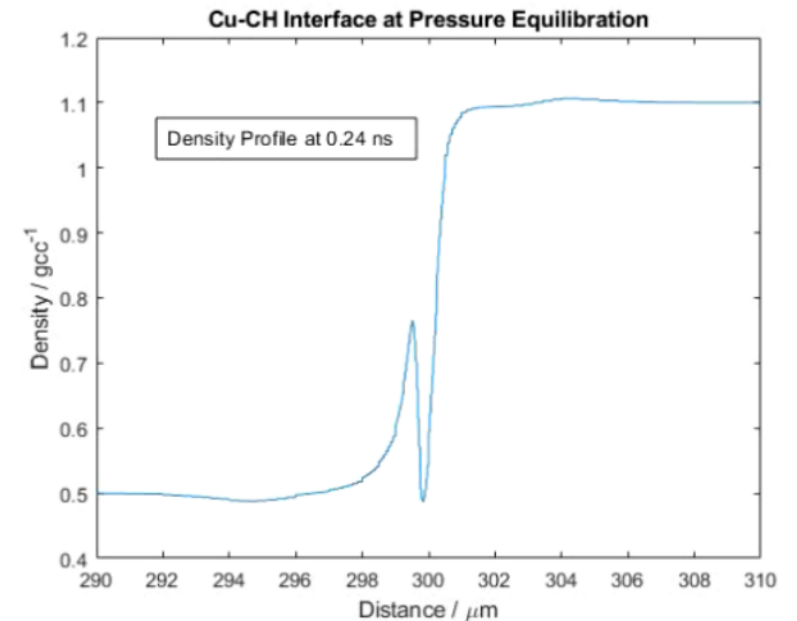
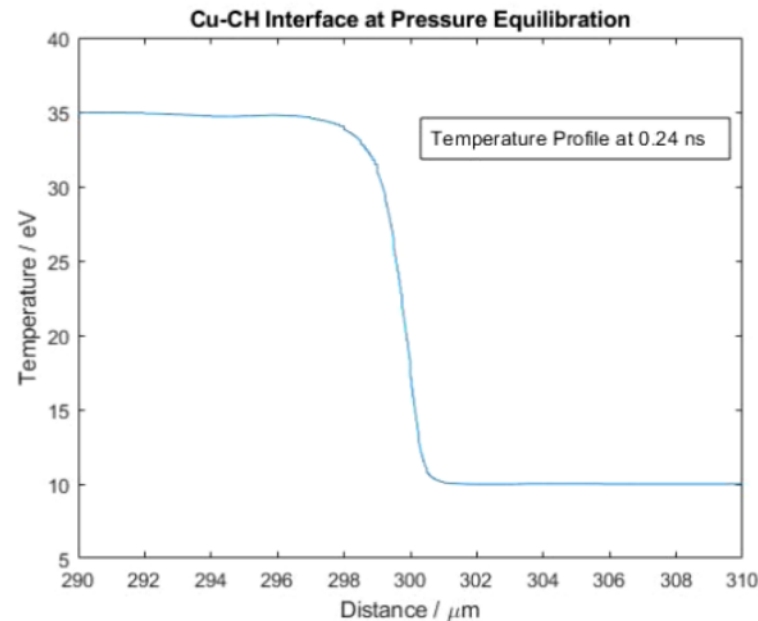
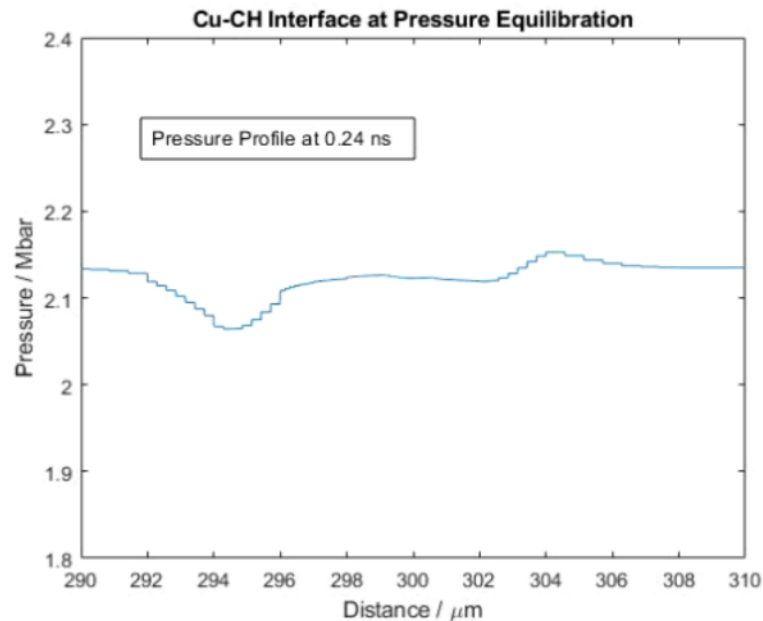
A pressure – equilibrated interface allows for investigation of temperature gradients



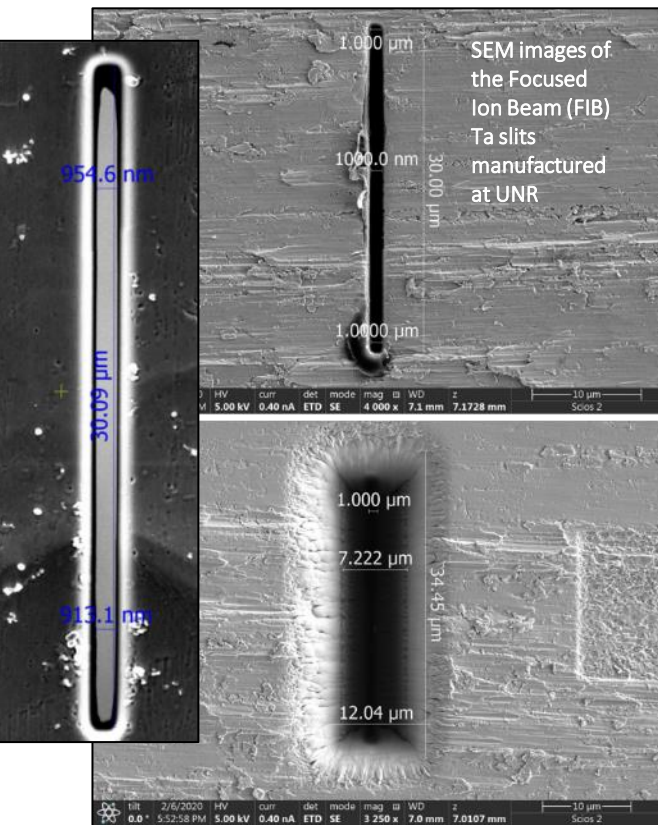
- Isochorically – heating a tamped system – such as a metal wire inside plastic – creates an expanding interface
 - As it expands, we have **hot metal** next to a **cooler plastic**, setting up the temperature gradient
- If the tamper can hold the expansion of the inner material, the pressure on either side of the interface is equilibrated $\rightarrow P_{inside} = P_{outside}$
 - Of course, we can't see heat, only its effects on density
 - For a constant pressure, a change in the temperature profile will inversely affect the density profile ($P \propto \rho T$)

Thermal conduction will alter density gradients – how much?

- At a pressure – equilibrated interface, the temperature profile is continuous, but the density develops a discontinuity at the interface
 - The metal next to the interface is hotter than the plastic, but cooler than metal further from the interface
 - The metal densifies towards the interface as it deposits heat into the plastic
 - The opposite happens for the plastic, which is hottest and least dense immediately next to the interface
- The scale lengths of the material on either side of the discontinuity are on order of a micron, and the shape will heavily depend on thermal conductivity

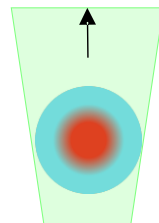


Using a 1 μm wide slit gives a micron-scale source for X-ray radiography



Top Down

Detector
(+100 cm)
 $M \sim 77$

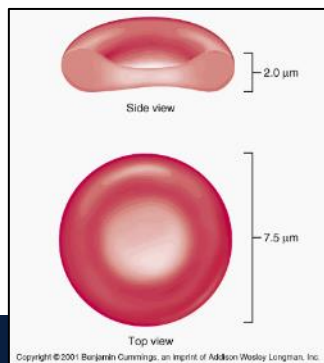


1 μm Slit in Ta Plate

V He- α 5.2 keV

V Backlighter Foil

10x450J, 1 ns
BL Beams



- By placing a micron-wide slit in front of the source, we have the spatial resolution to see micron-scale length changes
- A micron-scale source will also have the spatial coherence to be sensitive to interference (refraction, diffraction) effects
- At a sharp boundary between two WDM materials, we will have absorptive, refractive, *and* diffractive effects
 - The combination of these features will be unique to the density profile across the boundary and the imaging geometry
 - From the evolving density profile of a system, we can determine the thermal conductivity for the involved materials
- We call this “Fresnel Diffractive Radiography” (FDR)

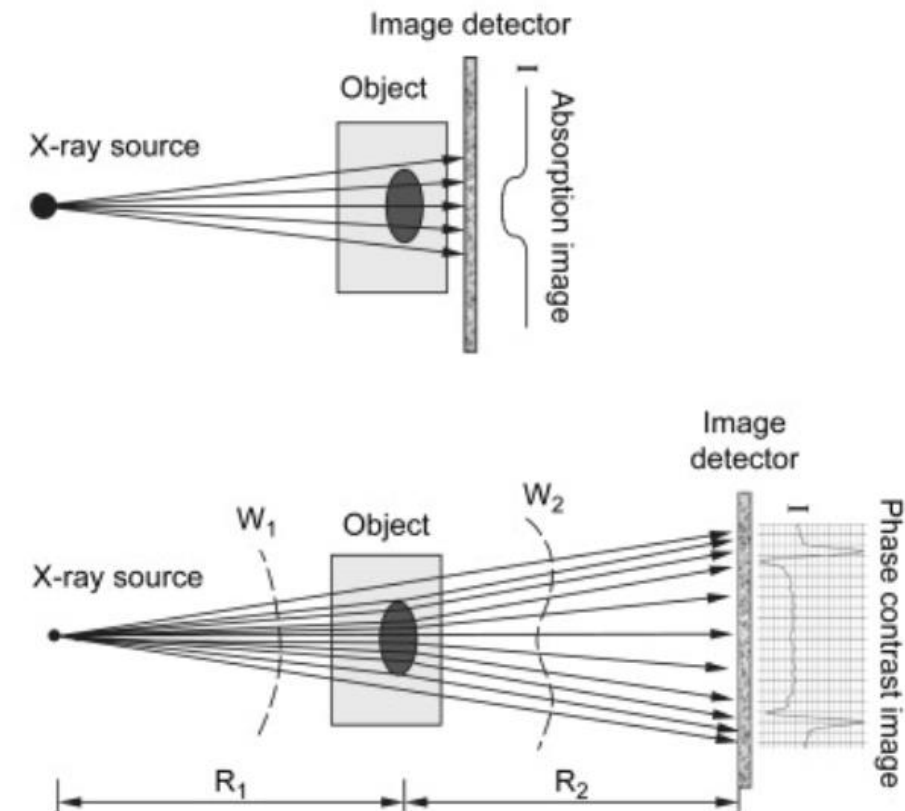
The micron-sized source introduces interference effects

- Imaging geometry or source characteristics means that most X-ray imaging is based upon absorption contrast
- For small source sizes and large propagation distances, we move into the phase-contrast imaging (PCI) / **refraction**-enhanced radiography (RER) regime, with improved contrast at weakly absorbing interfaces
- Additionally, **diffraction** fringes occur from material boundaries, but are frequently overshadowed due to larger refraction fringe scales*
 - Diffraction becomes important at Fresnel numbers $F \sim 1$

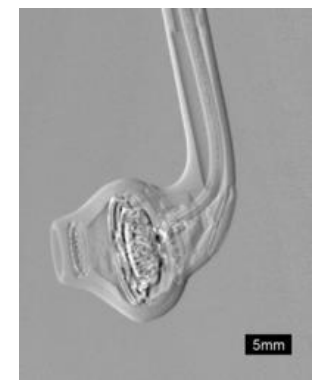
$$F = a^2 / f\lambda$$

where f is related to geometry and a is the scale length
“Fresnel Diffraction”

- The major difference between this work and previous RER experiments is **the sensitivity to diffractive effects**

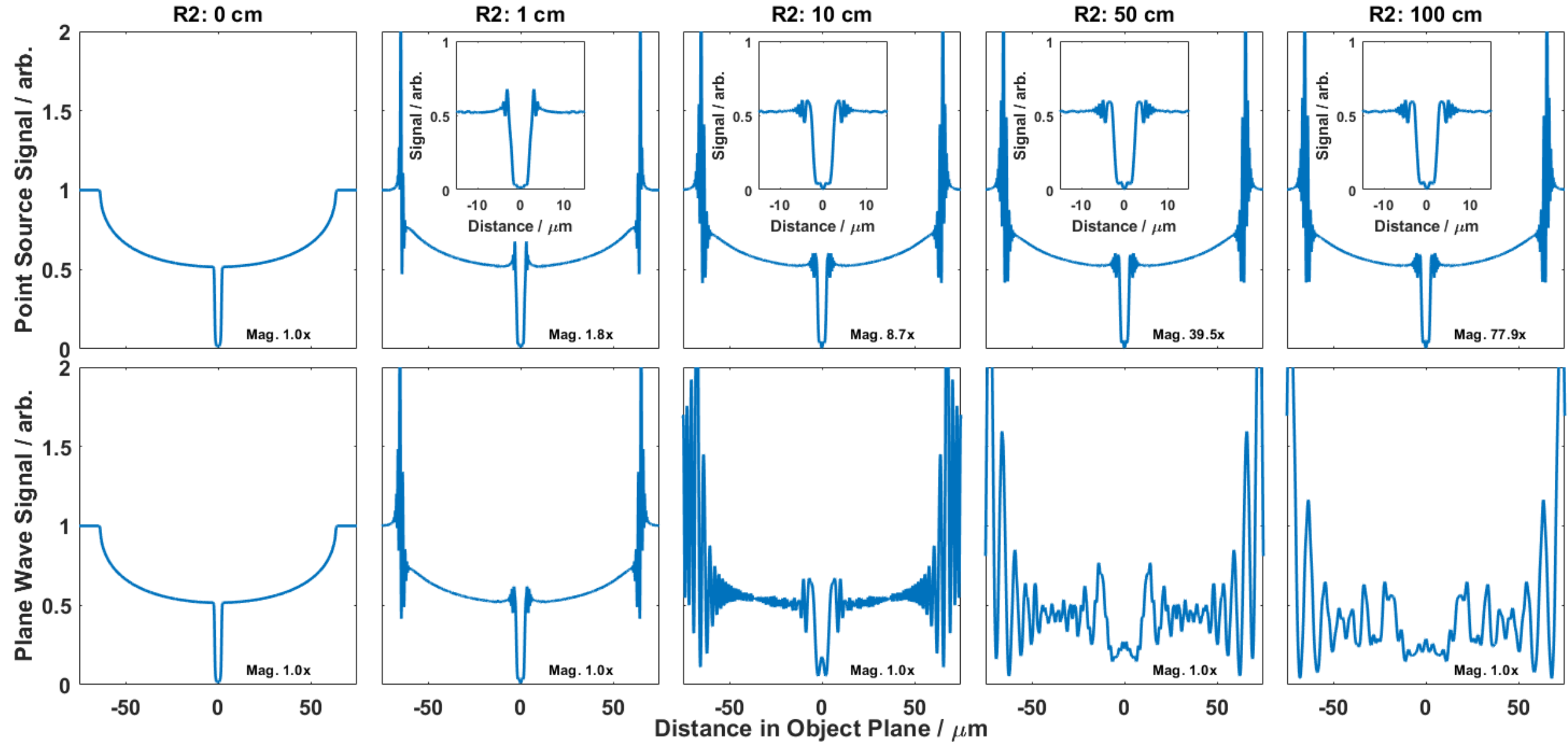


Absorption



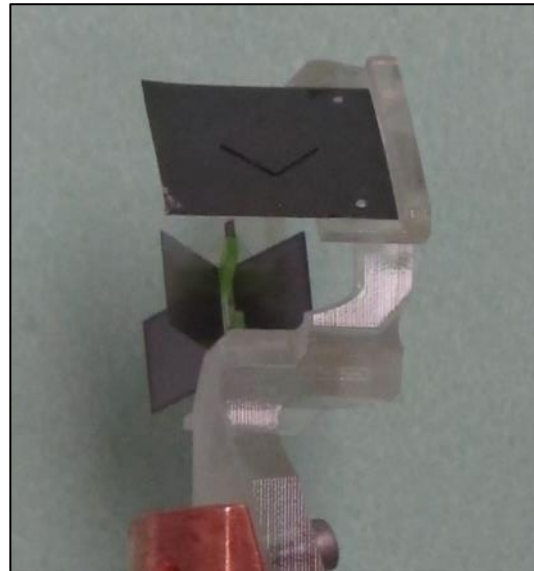
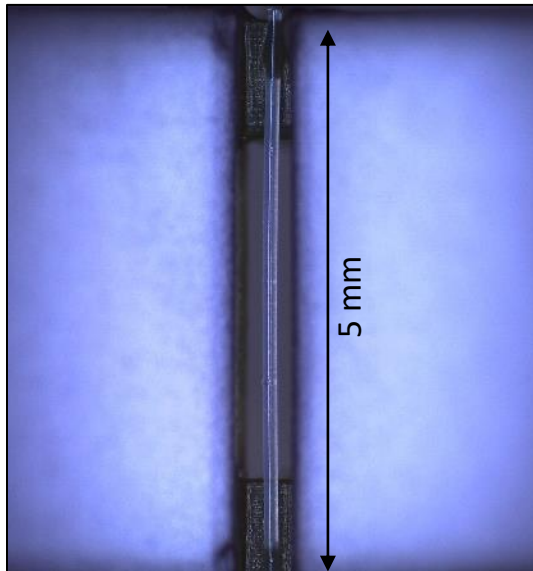
PCI

Point source X-ray radiography maintains a consistent shape with increasing propagation distance



The experiments rely on precise alignment between the slits and the wires

- The initial experimental goal for the Omega shots was to achieve micron-scale spatial resolution with FDR
- The alignment of the wire and the slit is critical to maximize the resolution capabilities of the platform
 - Each degree of relative tilt of with respect to parallel introduces $\sim 0.5 \mu\text{m}$ of source broadening
- We achieve $\sim 2 \mu\text{m}$ source size, due in part to the tapering of the slit



Diffraction enhanced imaging utilizing a laser produced x-ray source

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Submitted: 14 March 2022 • Accepted: 27 July 2022 • Published Online: 07 September 2022

M. Oliver, C. H. Allen, L. Divol, et al.

Developing a platform for Fresnel diffractive radiography with $1 \mu\text{m}$ spatial resolution at the National Ignition Facility

Cite as: Rev. Sci. Instrum. 94, 013104 (2023); [doi: 10.1063/5.0101890](https://doi.org/10.1063/5.0101890)
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Toward an integrated platform for characterizing laser-driven, isochorically heated plasmas with $1 \mu\text{m}$ spatial resolution

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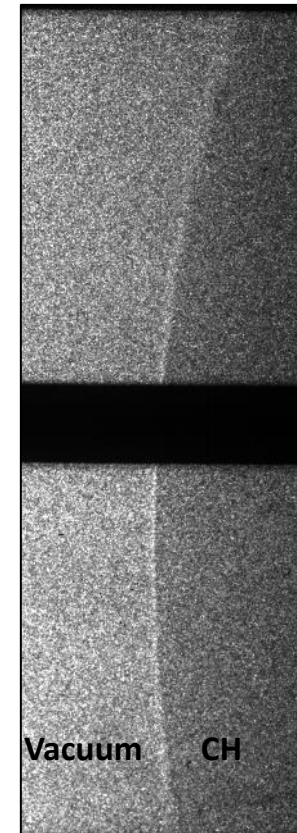
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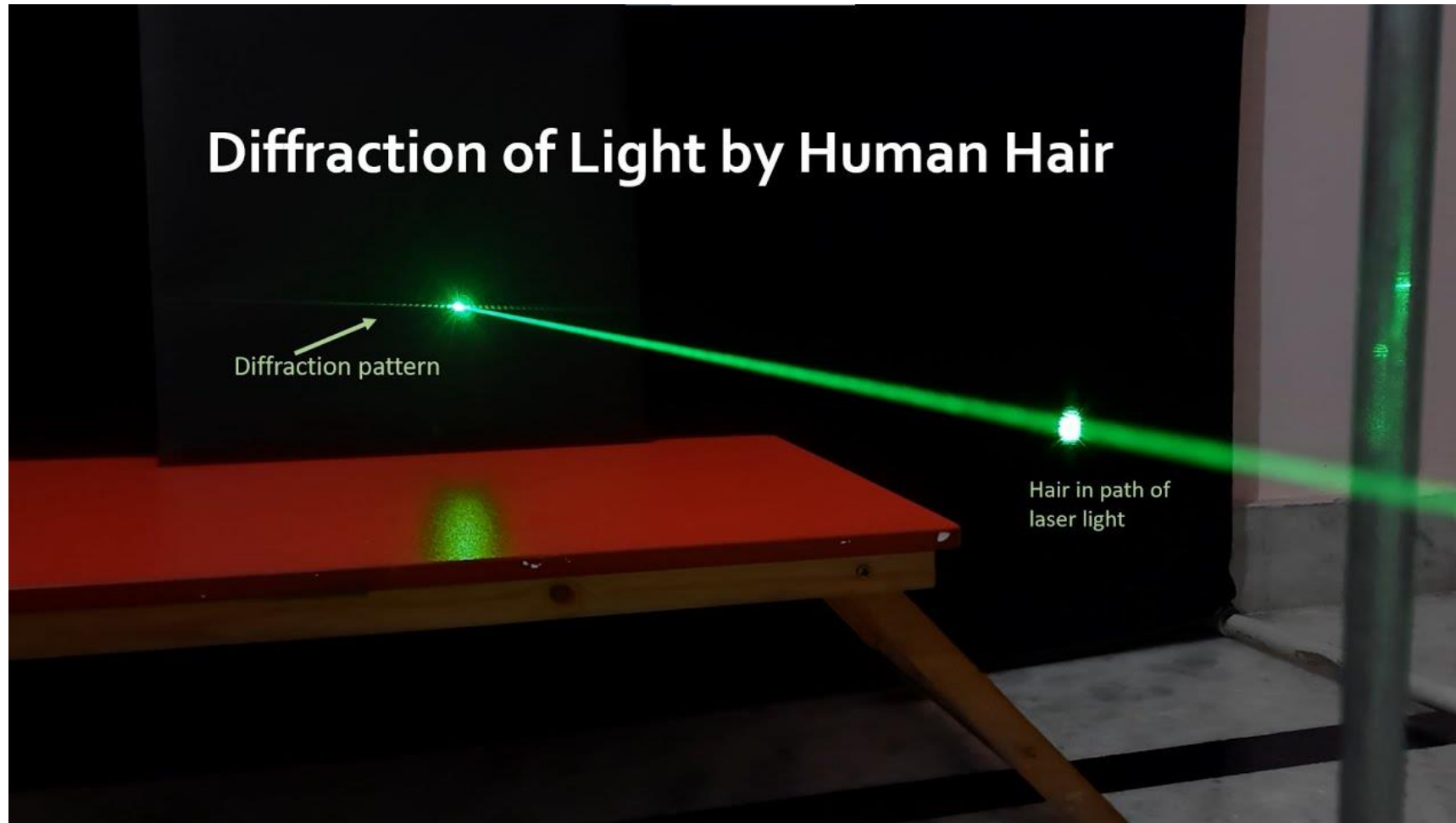
Received 18 October 2021; revised 4 February 2022; accepted 15 February 2022; posted 15 February 2022; published 3 March 2022



Preliminary data from Schölmerich/Döppner March 2023

Imaging a sphere with a slit demonstrates that the alignment between the slit and the interface is important

Diffraction patterns are very useful for measuring small objects

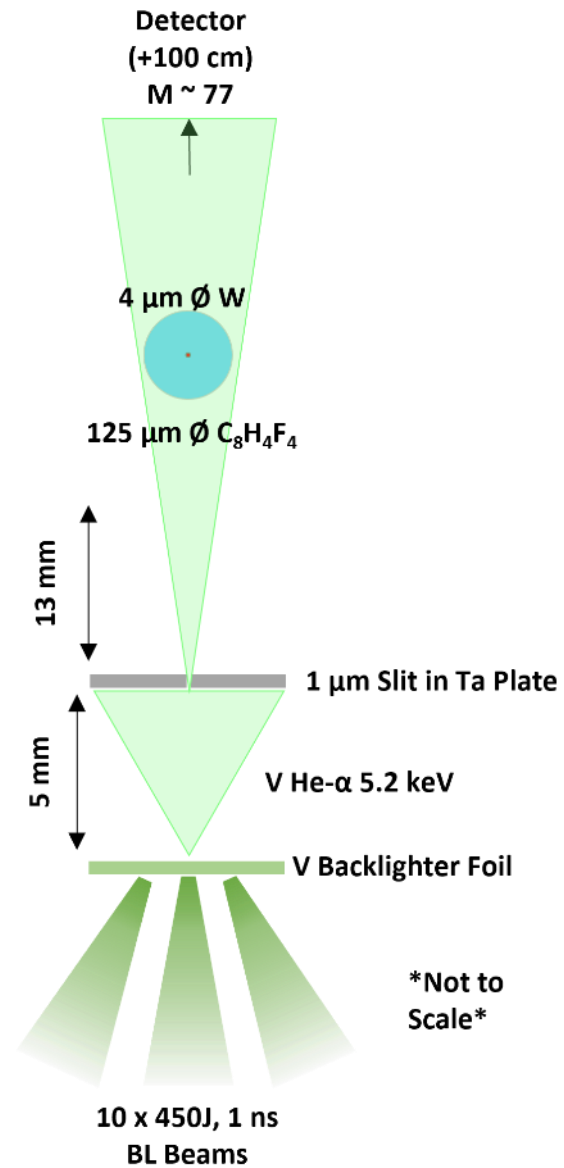


Human Hair: $\sim 100 \mu\text{m}$ diameter

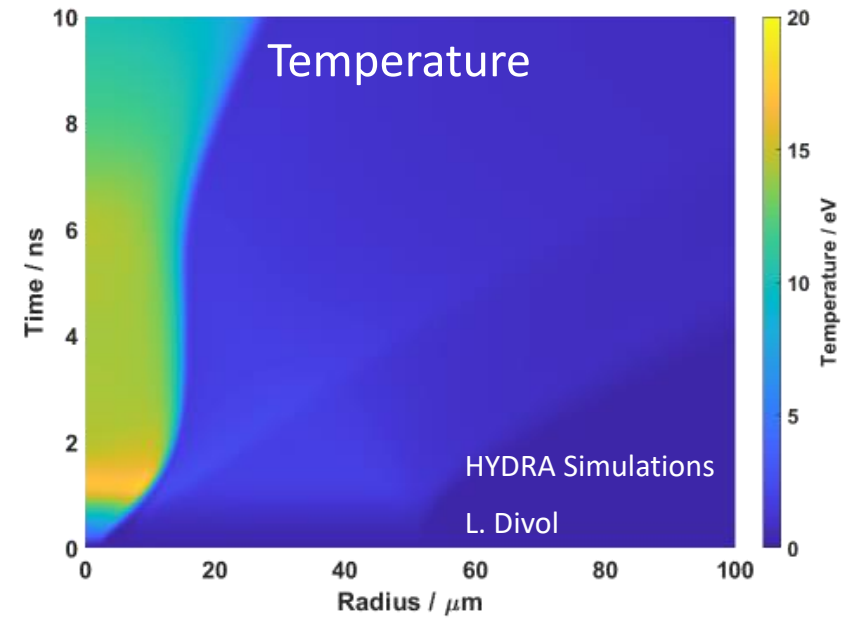
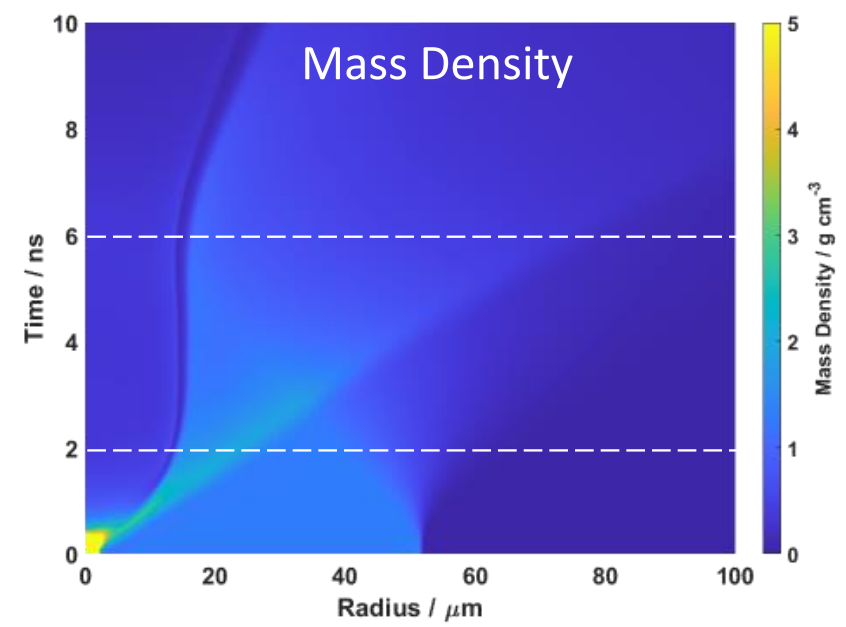
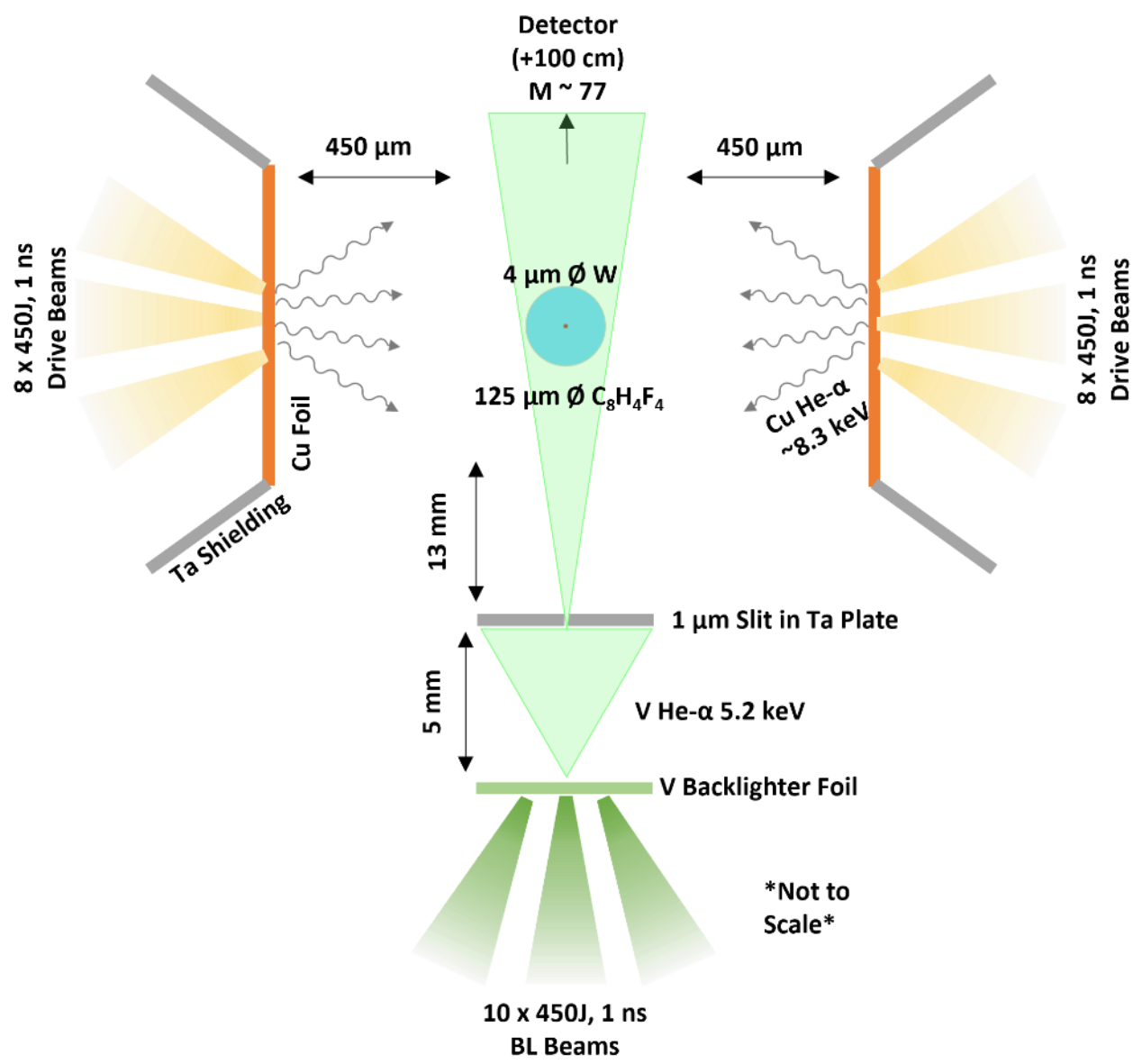
Optical wavelength: 532 nm

We perform the same experiment, but at 1000x smaller scale

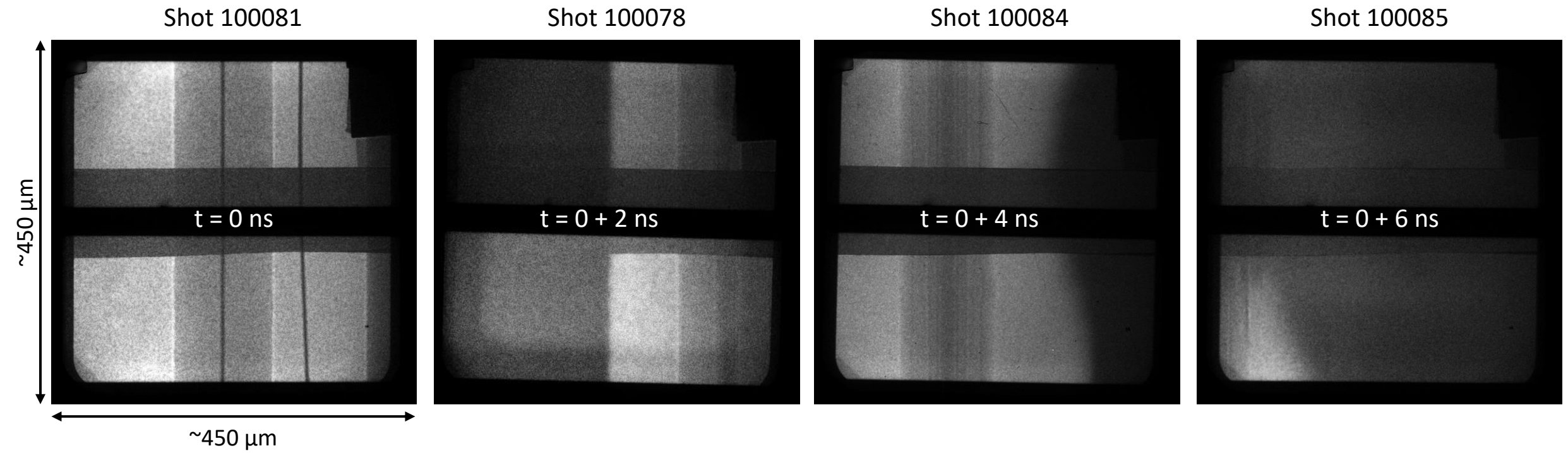
We use FDR to image an isochorically heated buried wire



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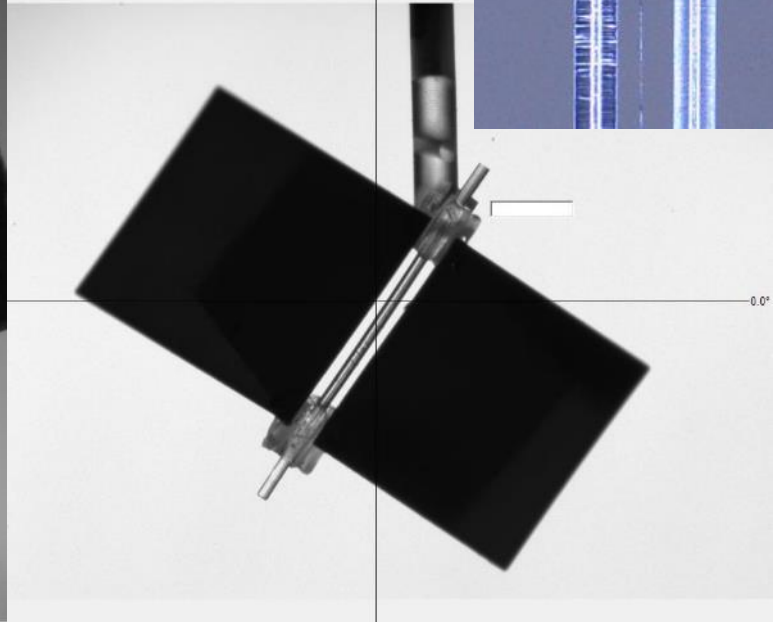
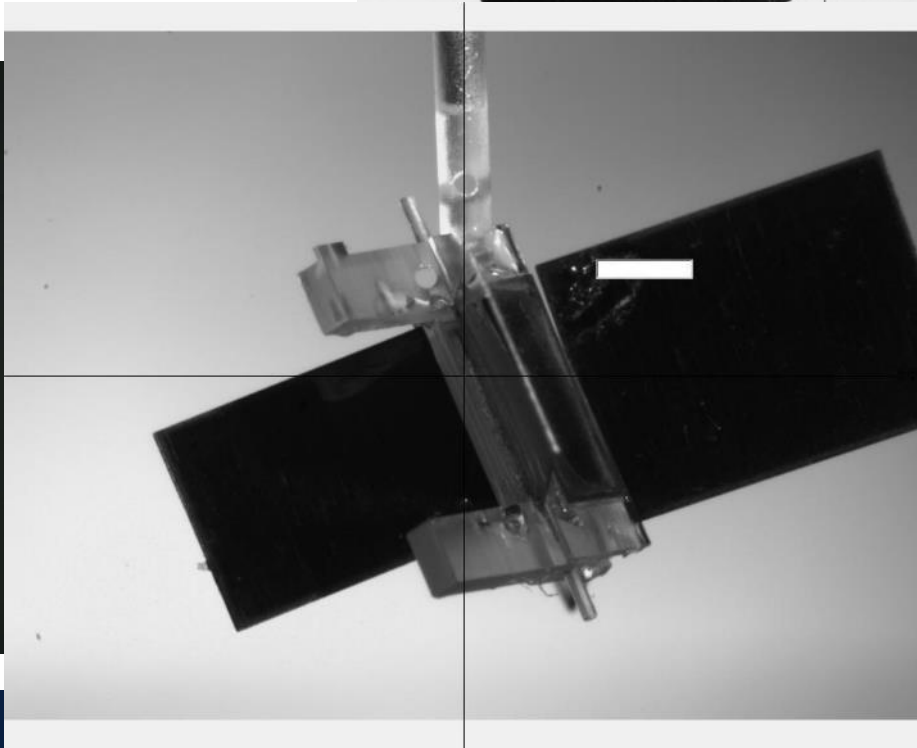
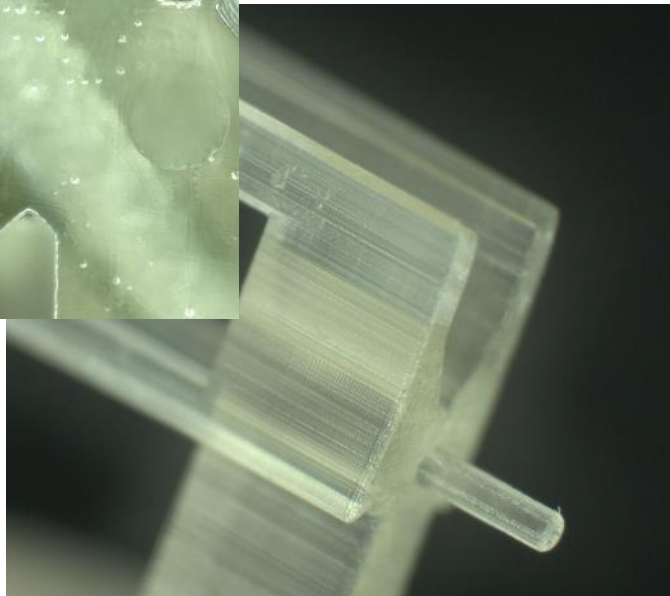
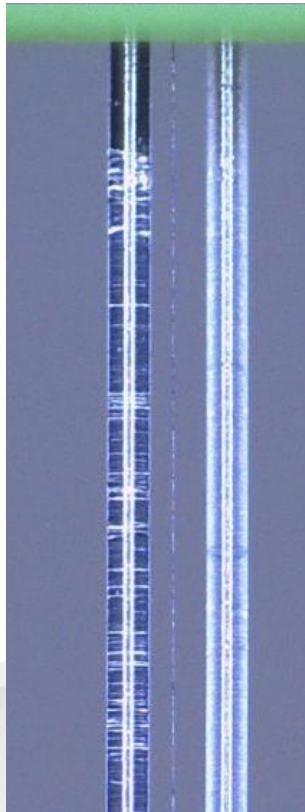
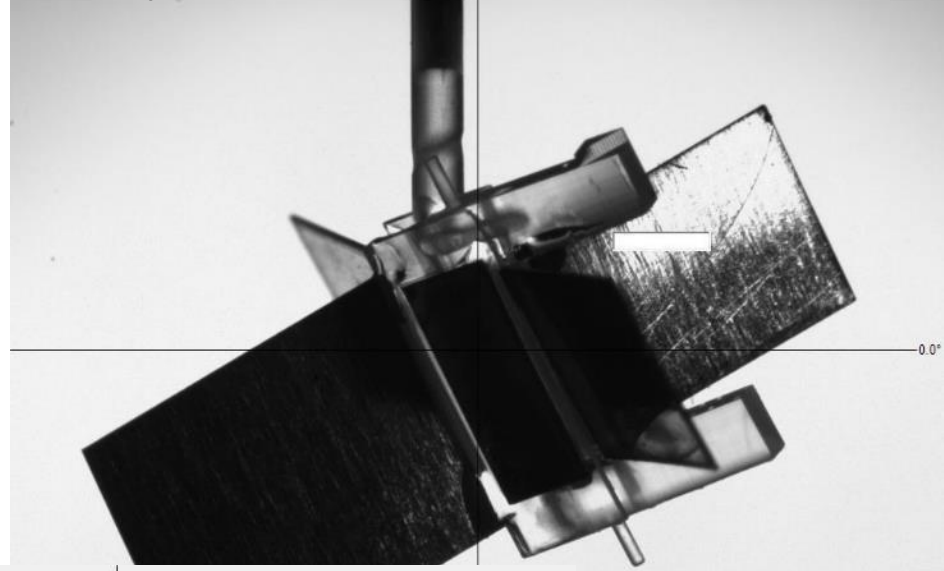
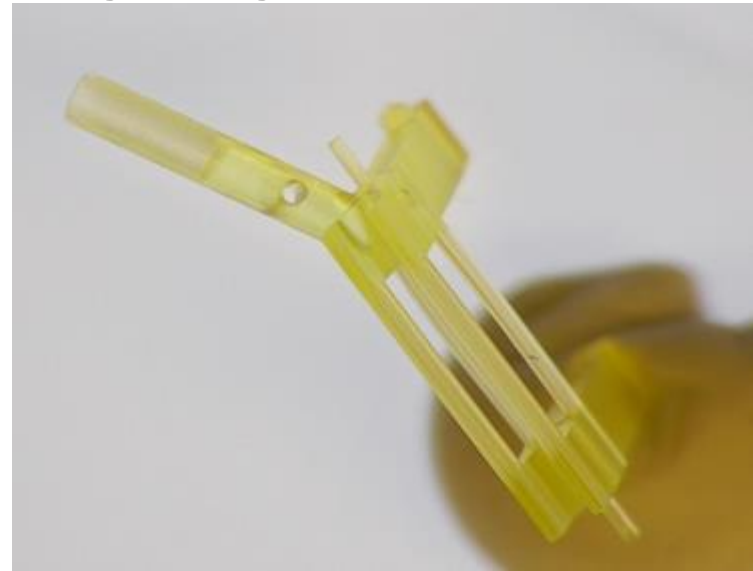


The FDR platform has achieved amazing results



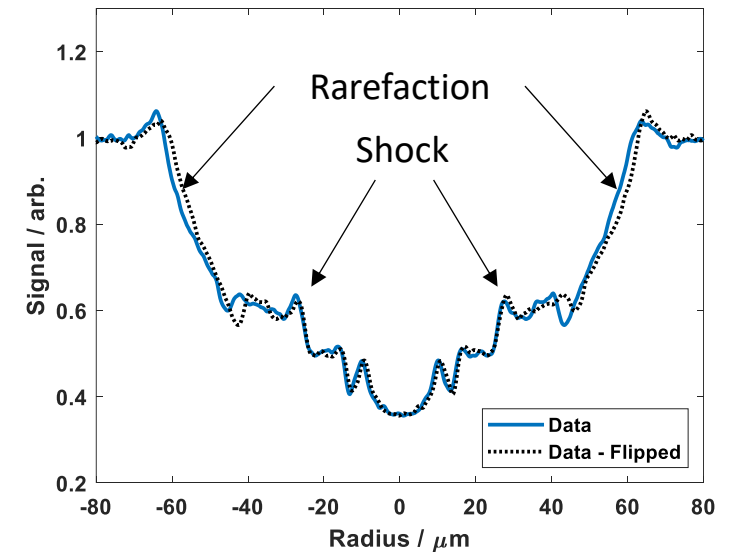
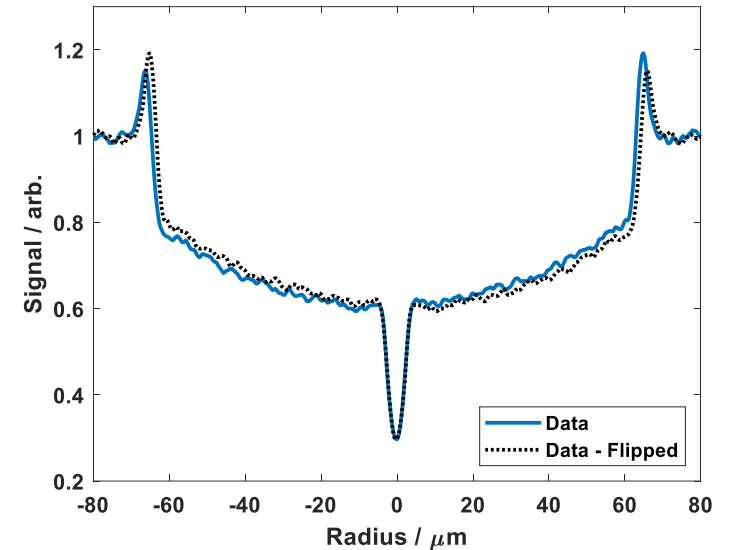
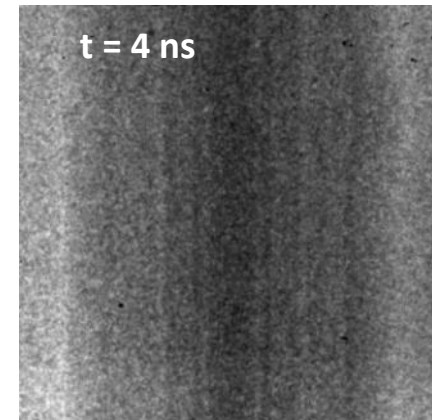
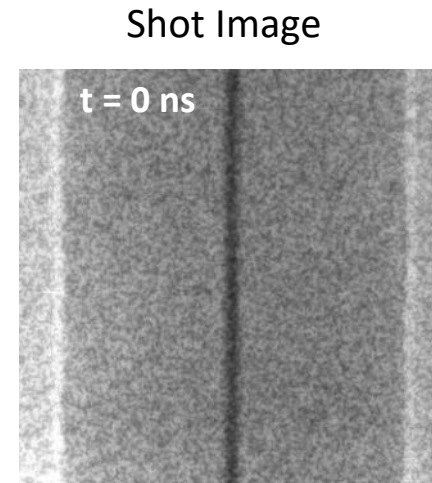
- The primary XRF diagnostic recorded excellent data, allowing for a time sequence over multiple shots/targets
- Target positioning on the detector sometimes left data on the edge (as in 100085) – more recent target designs have improved fiducials allowing for much more repeatable placement

Target alignment has been improved greatly with 3d printed target frames



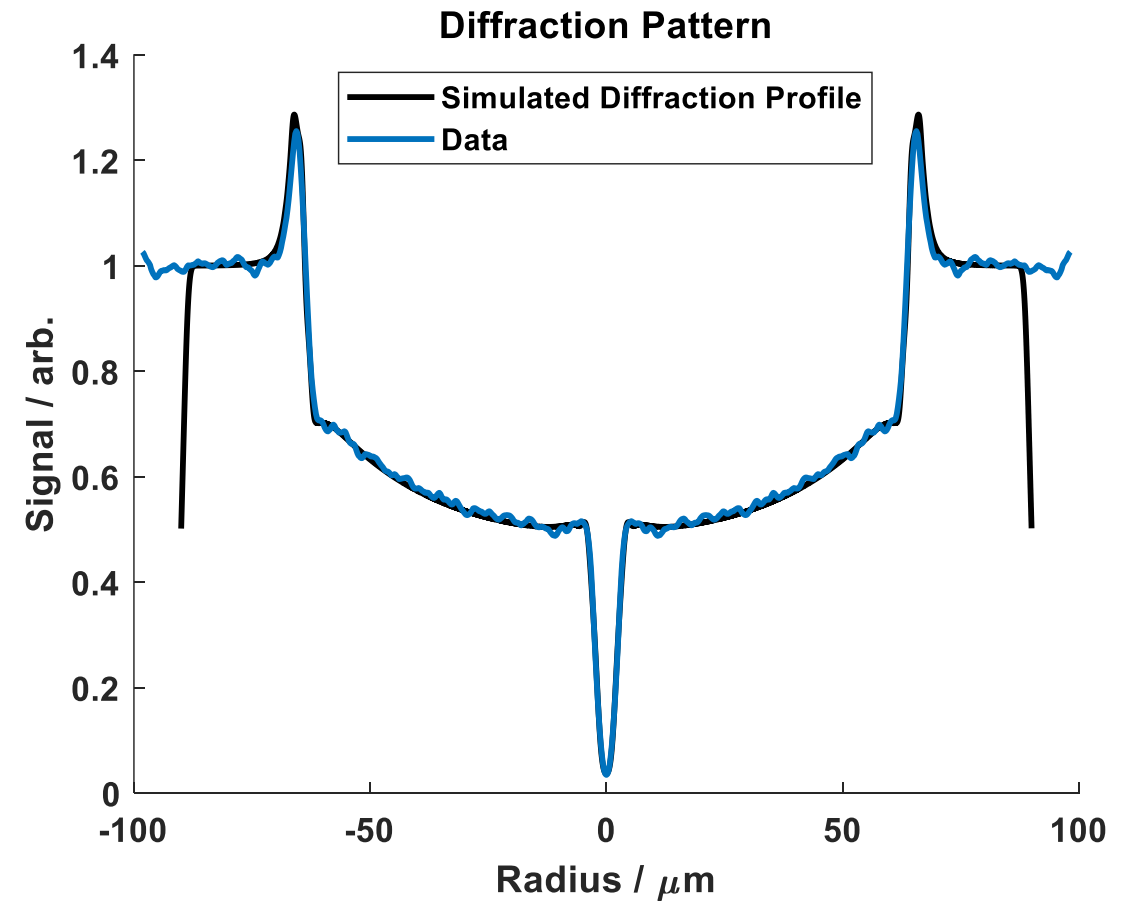
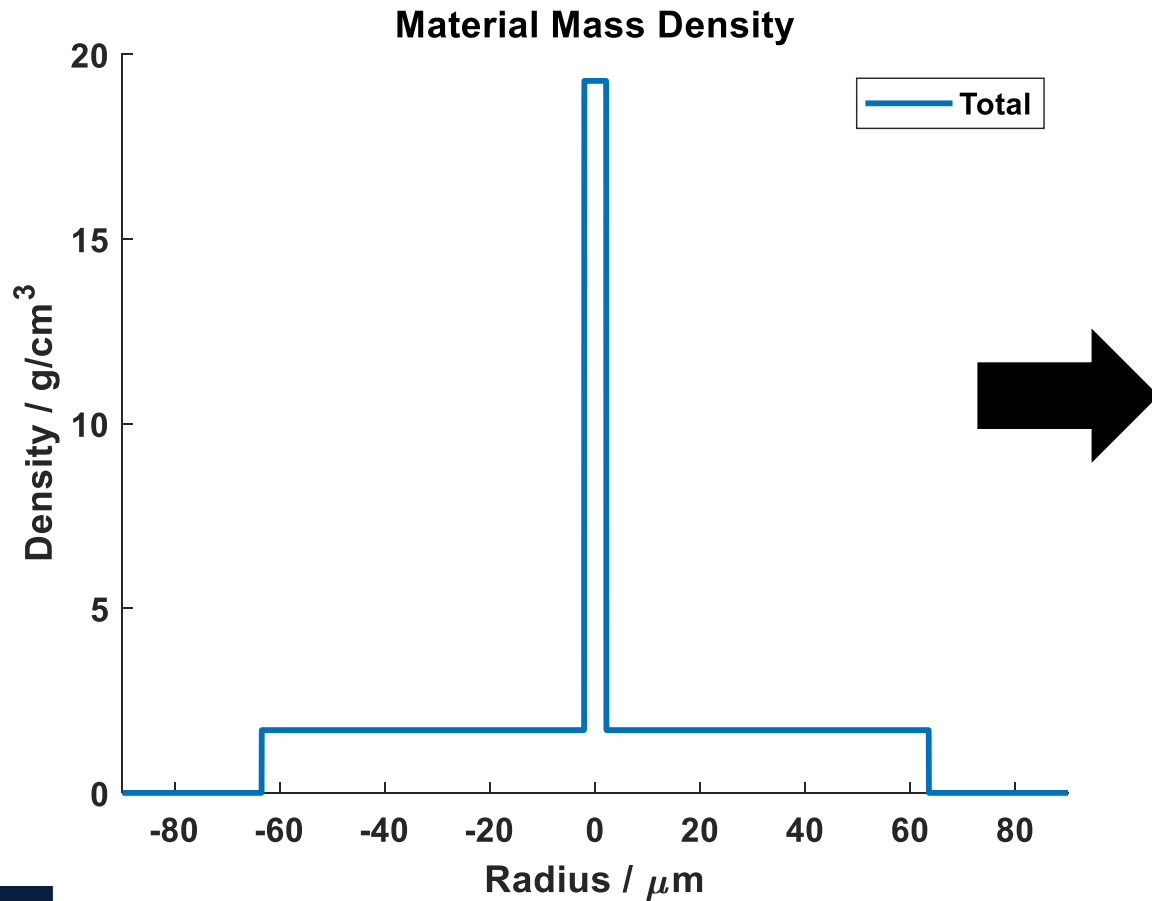
The evolving system and its features can tell us about the properties of the materials

- The notable features in the expanded system include:
 - W wire expansion ($\sim 4 \mu\text{m} \rightarrow \sim 20 \mu\text{m}$)
 - Outgoing shock waves
 - Rarefaction features from expanded outside edge
- The data is *extremely* symmetric, especially around the interface region we are interested in
- By taking data at different times, we can track the evolution of the interface changing over time
 - Goal is to determine the shape of the interface to determine thermal conductivity



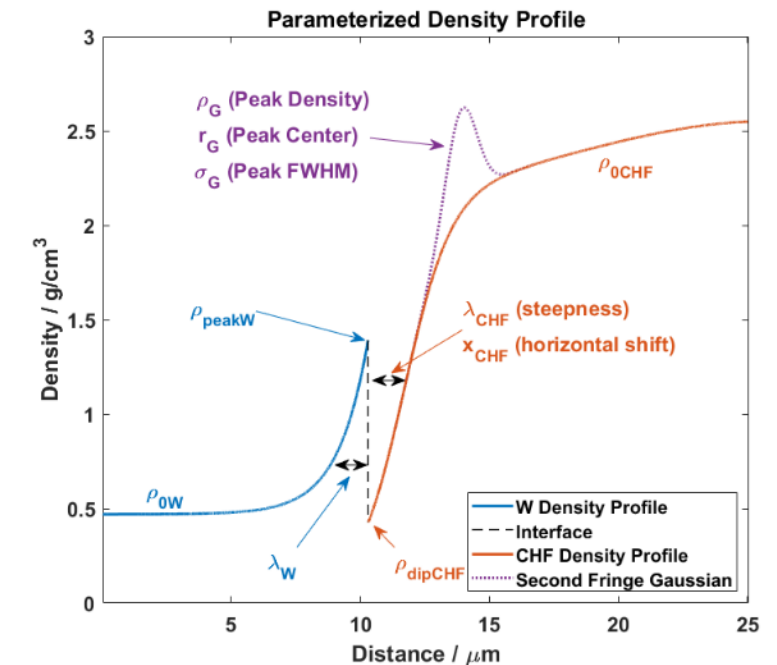
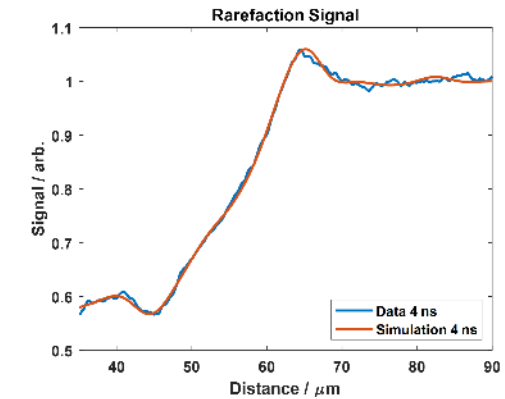
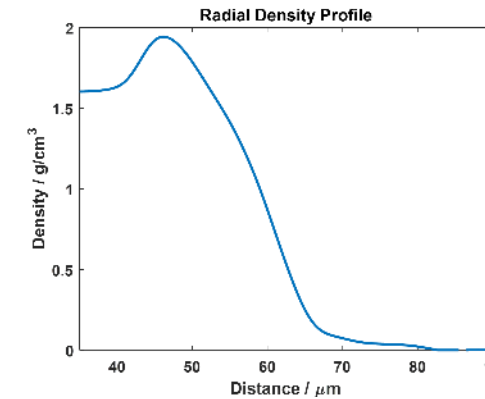
The key piece of analysis is forward modeling of the diffraction-refraction patterns

- We have developed a code that solves the Fresnel-Kirchoff Equation* for a parameterized density profile, calculating both refractive and diffractive effects
- The cold data gives us excellent agreement with a known step-like density profile and a **<2 μm source size**

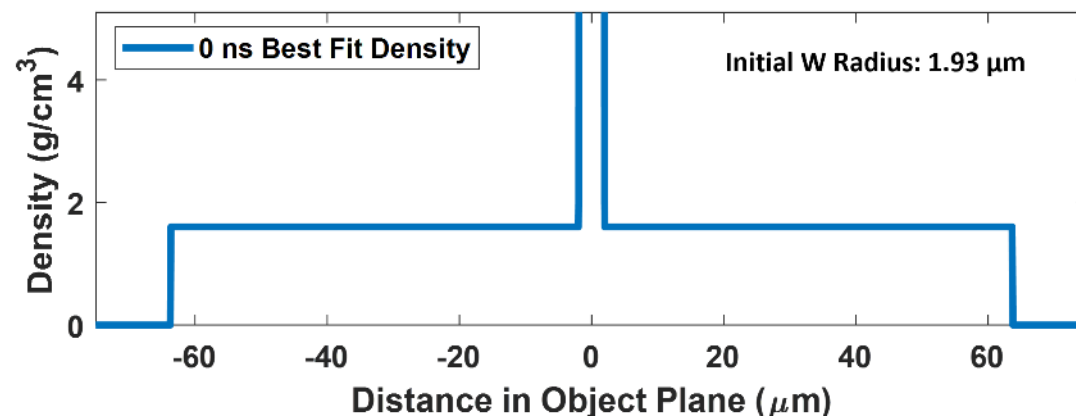
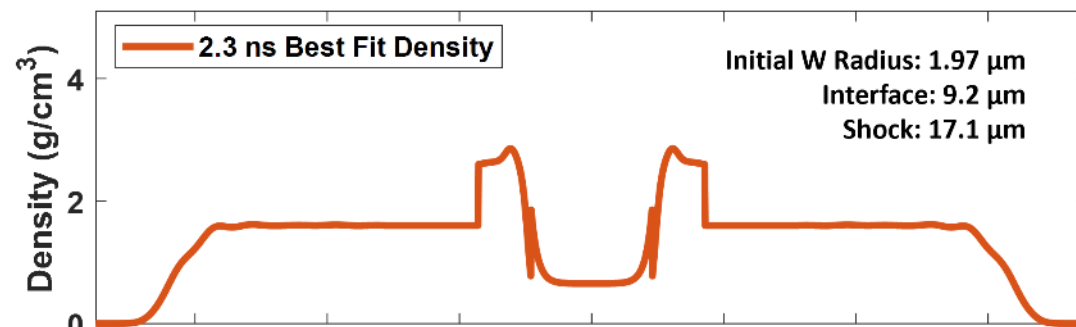
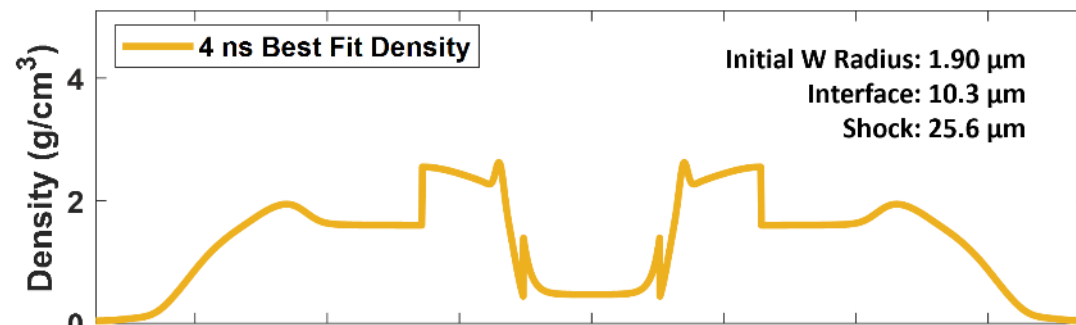


For the hot data, we utilized parameterized density profiles at the interface

- We created parameterized density profiles and simulated the diffraction patterns to match the data
- Requires material and density as functions of radius
- Going from outside in to be self-consistent:
 - Use the plastic data to determine the radial density profile for the target up to the shock wave
 - Parameterize the W-CHF interface to include a discontinuity, expanded W radius, and CHF density after the shock
- Initial material parameters need to be the consistent for all shots – the coated wires were made at the same time
 - Similar W radius, similar CHF radius

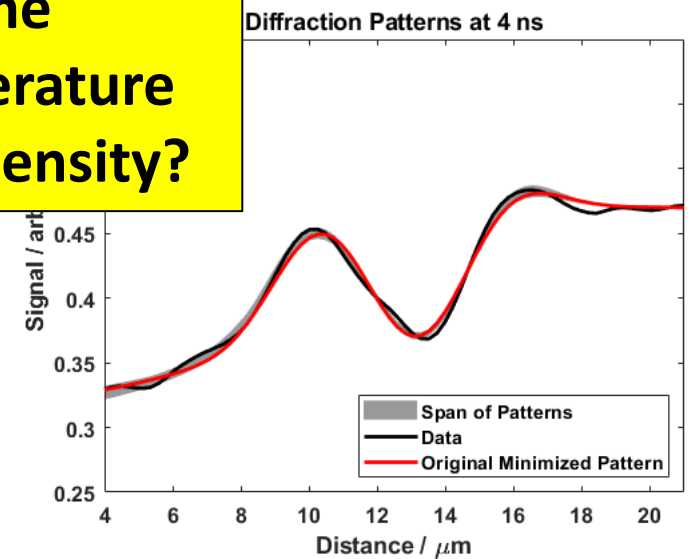
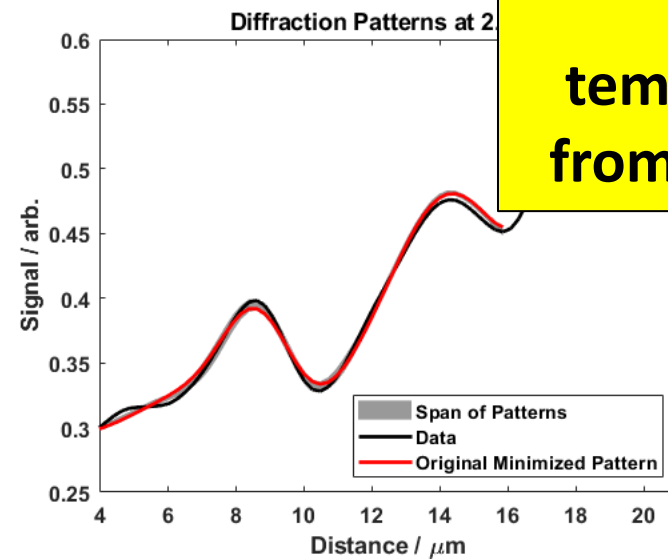
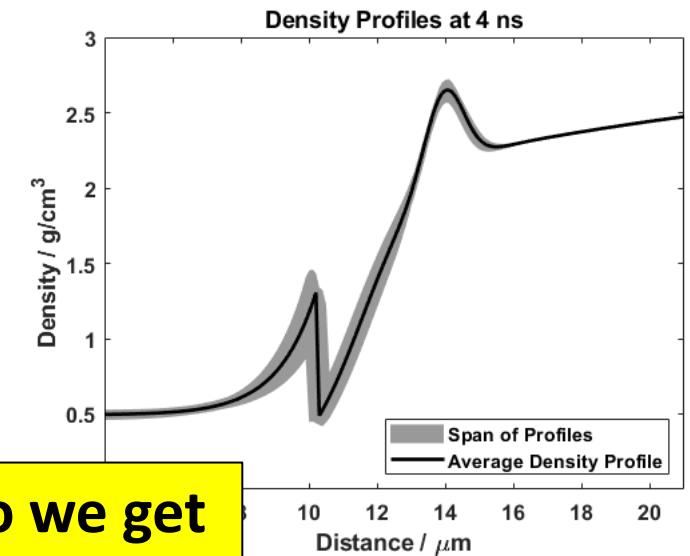
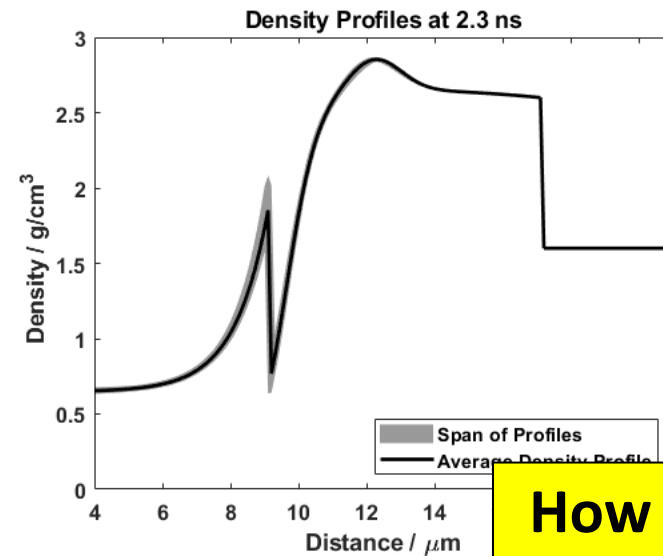


These parameterized density profiles have been fantastically successful



Use of Bayesian Inference to Determine Uniqueness

- To help determine the accuracy of our density profiles, we use Bayesian inference / MCMC in the style of Kasim *et al.** to sample phase space for the parameterized interface and provide error estimates



How do we get the temperature from density?

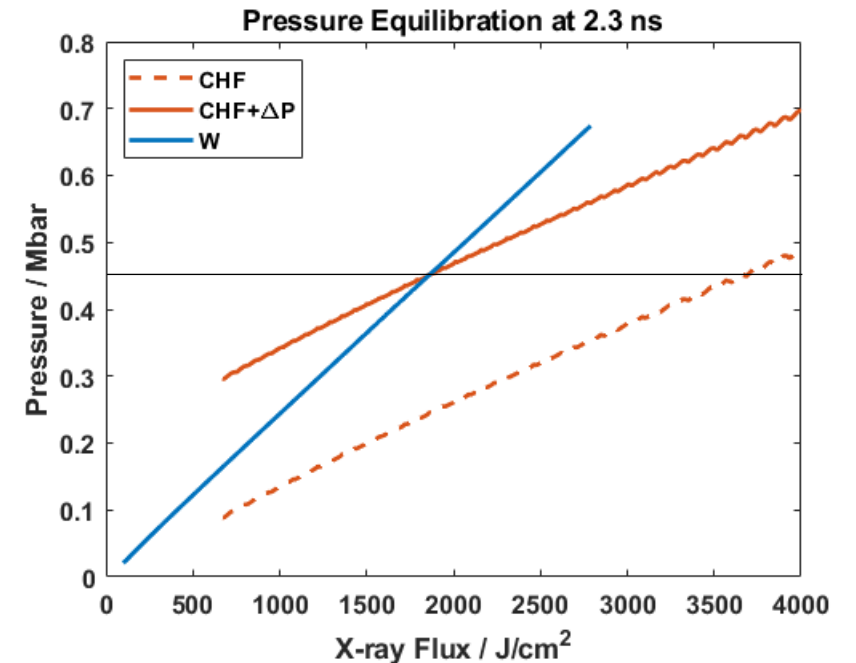
Capturing the shock in our data allows us to find a temperature

- **The plan:** If we have a density and a pressure, we can calculate a temperature
- Using FEOS tables for both the W and CHF, we can determine the matching pressure across the interface
 - Both the W and CHF should be subjected to the same X-ray flux – cross-over point is the equilibrium pressure

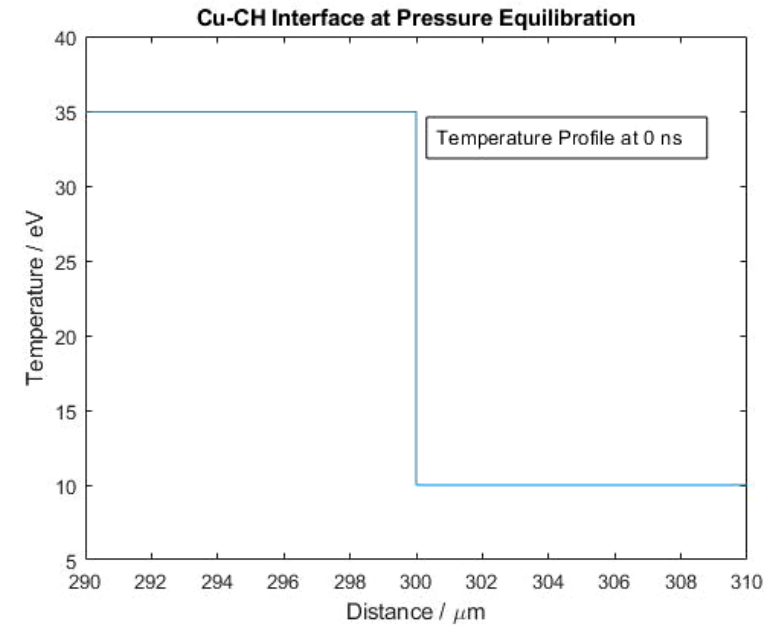
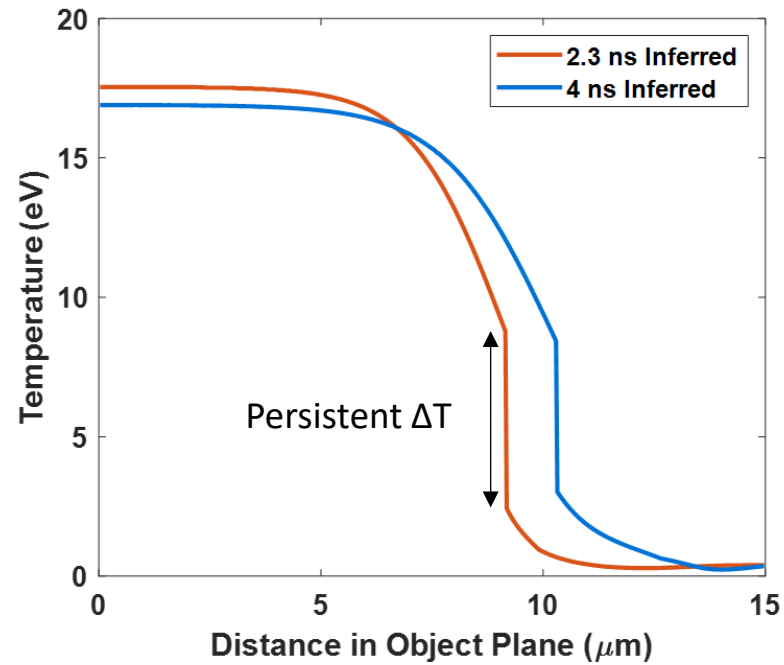
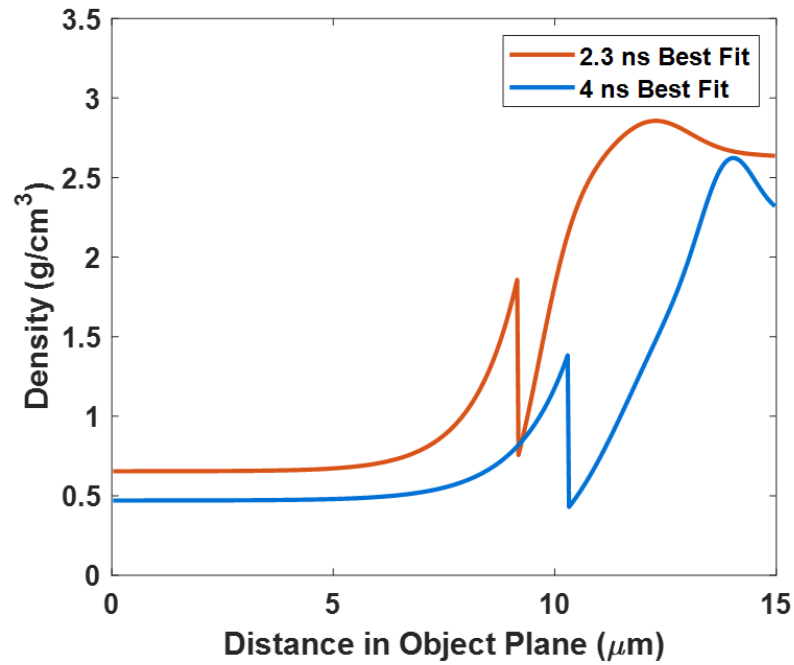
- The outward travelling shock in the CHF can be used to determine the pressure after the shock and near the interface from a form of Hugoniot Relations:

$$\Delta P = u_s^2 \left(\rho_1 - \rho_1^2 / \rho_2 \right)$$

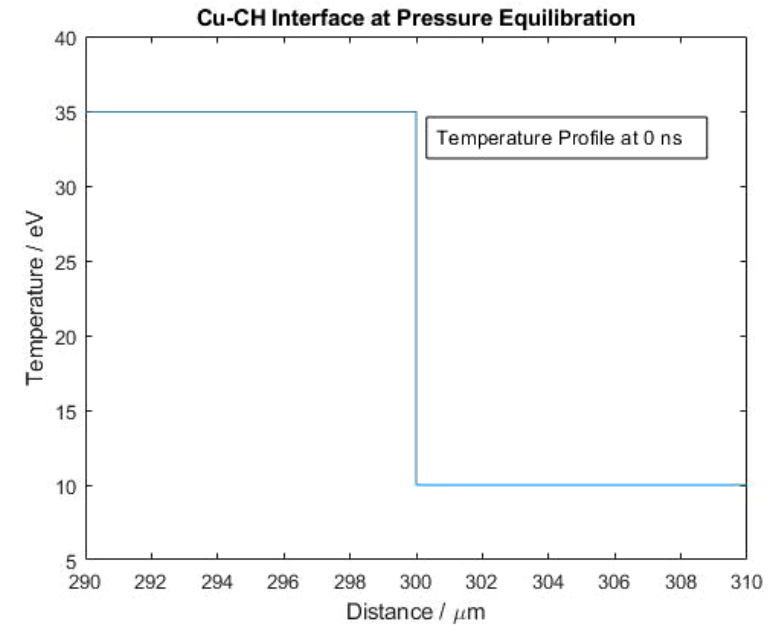
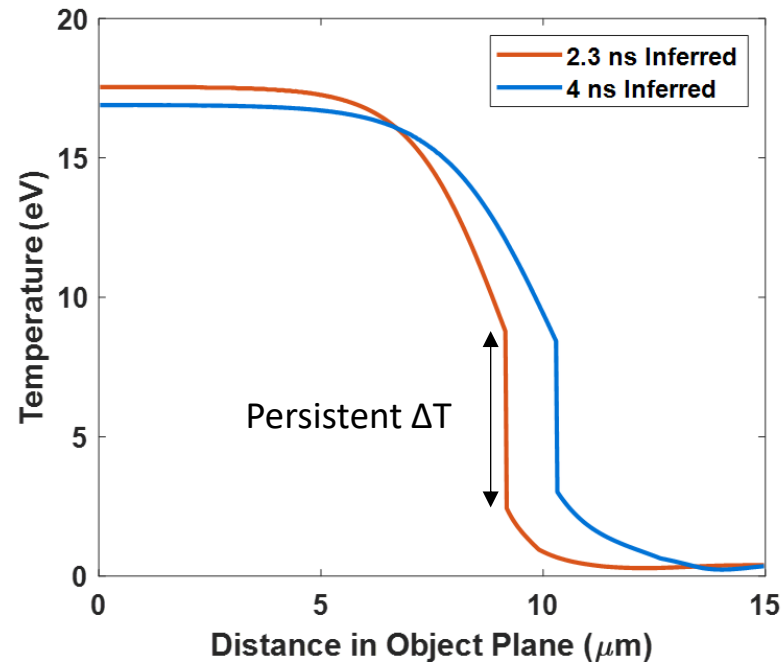
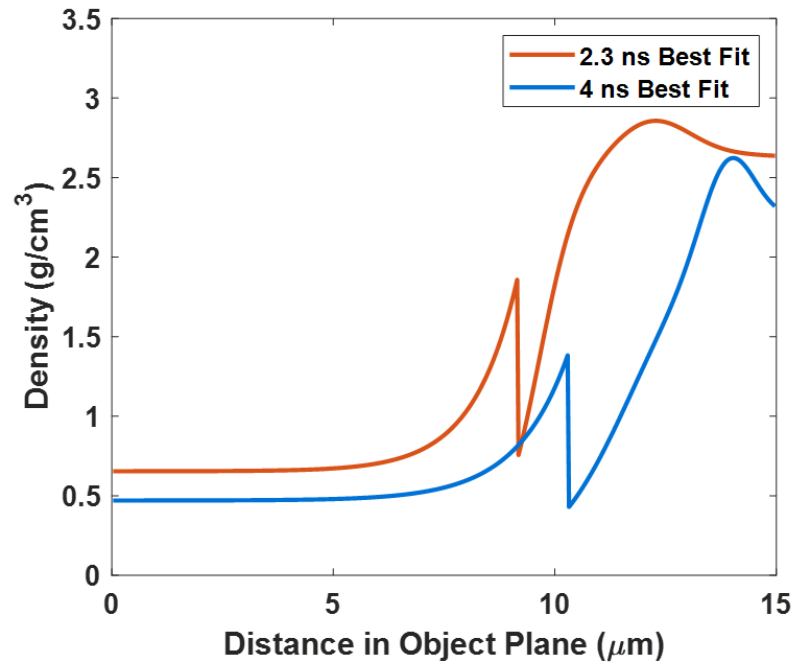
- The three data points (0, 2.3, 4 ns) are used to estimate the shock velocity at the 2.3 ns
 - This ends up being $r \propto t^{0.8}$ from previous wire explosion work on pulsed power machines



The temperature profile features a discontinuity at the interface



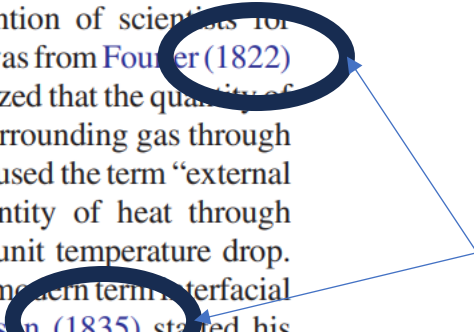
The temperature profile features a discontinuity at the interface



Interfacial thermal resistance: Past, present, and futureJie Chen[✉] and Xiangfan Xu[✉]*Center for Phononics and Thermal Energy Science, China–EU Joint Lab for Nanophonics, MOE Key Laboratory of Advanced Micro-structured Materials, School of Physics Science and Engineering, Tongji University, Shanghai 200092, China*Jun Zhou[✉]*Phonon Engineering Research Center of Jiangsu Province, Center for Quantum Transport and Thermal Energy Science, Institute of Physics Frontiers and Interdisciplinary Sciences, School of Physics and Technology, Nanjing Normal University, Nanjing 210023, China*Baowen Li[✉]*Department of Material Science and Engineering, Department of Physics, Shenzhen Institute for Quantum Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China and Paul M. Rady Department of Mechanical Engineering and Department of Physics, University of Colorado, Boulder, Colorado 80305-0427, USA*

This problem has attracted the attention of scientists for centuries. The first recorded discussion was from Fourier (1822) in the early 19th century. Fourier recognized that the quantity of heat that the solid bodies lose to their surrounding gas through the surface obeys the same principle. He used the term “external conductivity” to characterize the quantity of heat through surface per unit time per unit area per unit temperature drop. This definition is exactly the same as the modern term interfacial thermal conductance (ITC). Later Poisson (1835) started his study with the following continuity of the heat flux at the interface:

$$J = \kappa_1 |\nabla T|_1 = \kappa_2 |\nabla T|_2 = h_I \Delta T. \quad (1)$$

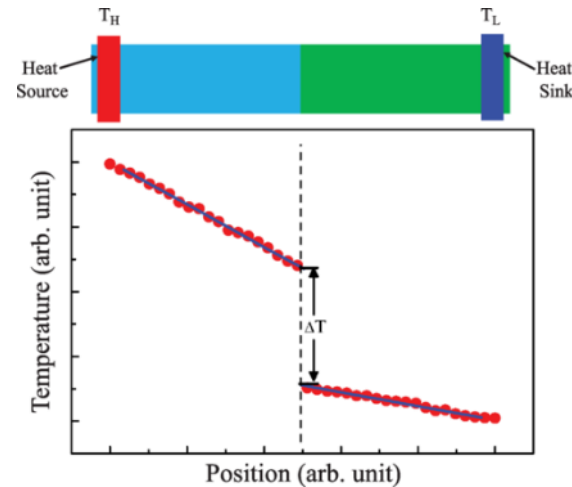
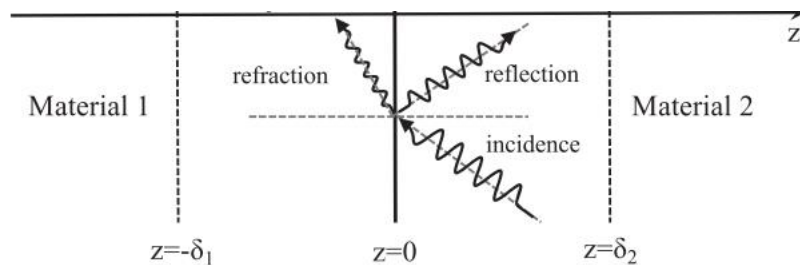


This is not a new phenomena!

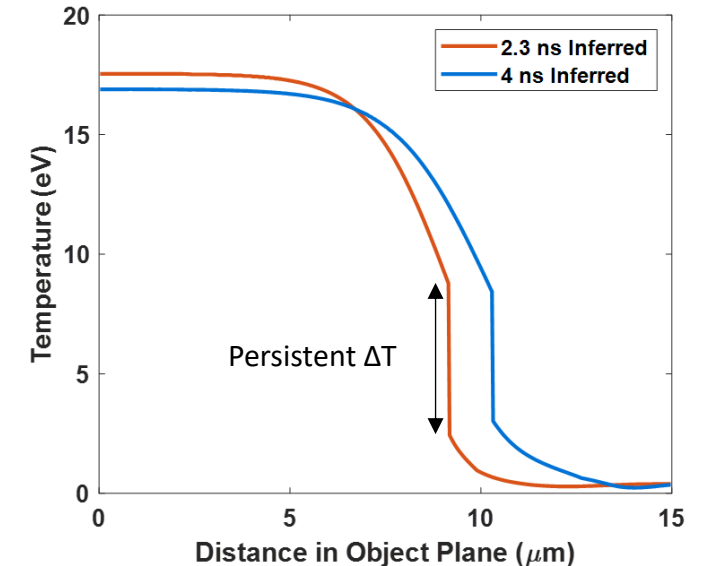
The temperature profile features a discontinuity at the interface

- We infer the radial temperature profile of our system from the FEOS* tables, and find a discontinuous temperature jump at the interface
- This is a temperature discontinuity resulting from *interfacial thermal resistance* (ITR)**
 - Due to differences in heat carriers between materials
 - Experimentally measured for solid-solid¹, solid-liquid², and solid-gas³ interfaces; MD simulations for liquid-liquid⁴
 - To our knowledge, it has not been explored for WDM or dense plasmas

1. E. T. Schwartz & R. O. Pohl. *App. Phys. Lett.* **51**, 2200 (1987)
2. G. L. Pollack. *Rev. Mod. Phys.* **41**, 48-81 (1969)
3. M. S. de Smolan. *Phil. Mag.* **46**, 279 (1898)
4. H. A. Patel *et al.* *Nano Let.* **5**, 2225 (2005)



Schematic Diagram of interfacial thermal resistance.
J. Chen *et al.* *Review of Modern Physics* **94**, 025002 (2022)



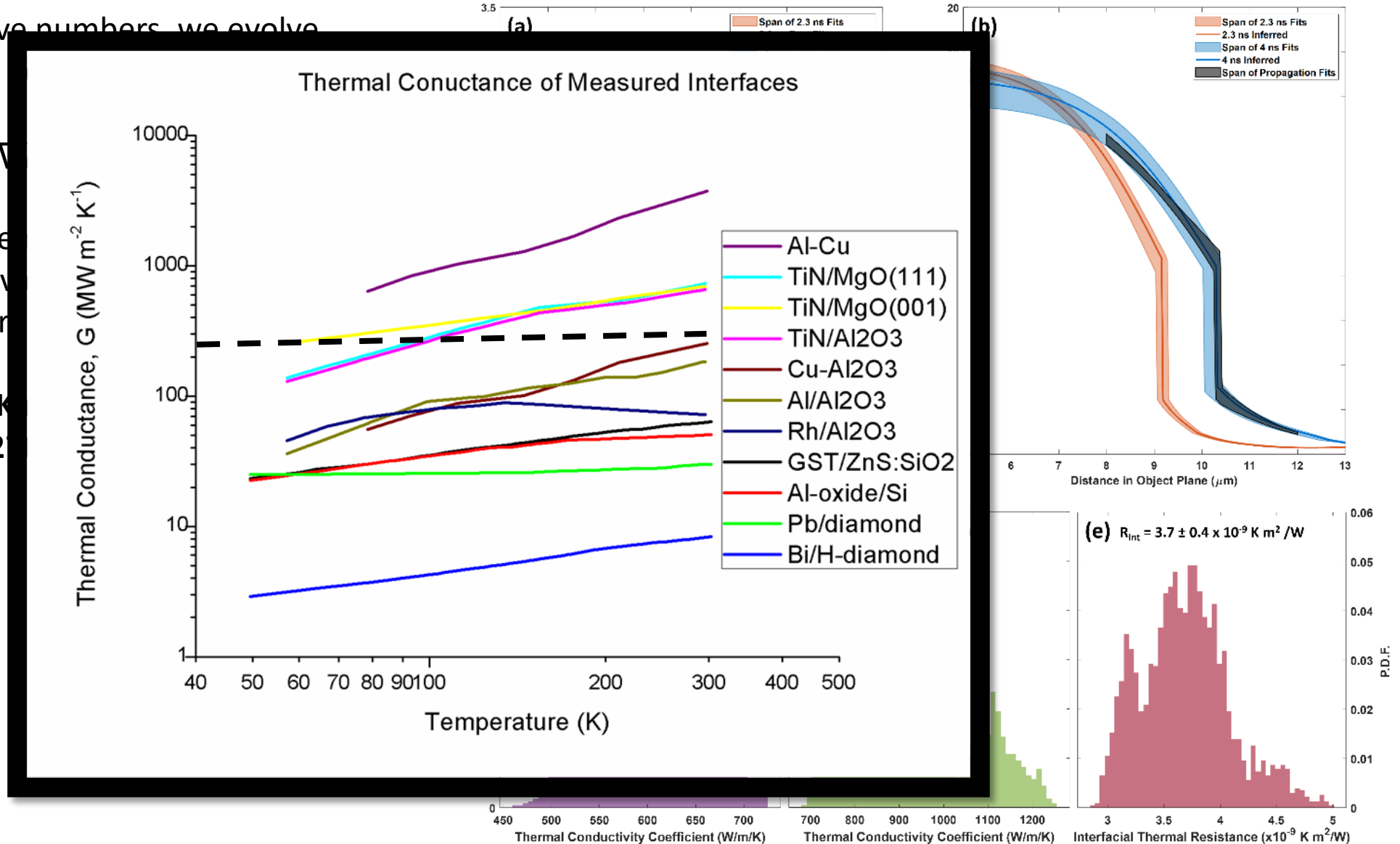
The data at 2.3 ns can be evolved to match the data at 4 ns

- To extract quantitative numbers, we evolve one time step to the

$$q(r, t) = \kappa \nabla T$$

- We get a quantitative number that is comparable to previous insulator systems at room temperature

- $R_{\text{Int}} \approx 3.7 \times 10^{-9} \text{ K m}^2/\text{W}$
- $G_{\text{Int}} = 1/R_{\text{Int}} \approx 2.7 \times 10^8 \text{ W m}^2/\text{K}$



The data at 2.3 ns can be evolved to match the data at 4 ns

- To extract quantitative numbers, we evolve one time step to the next using Fourier's Law:

$$q(r, t) = \kappa \nabla T = \frac{\Delta T}{R} \left[\frac{J}{m^2 s} \right]$$

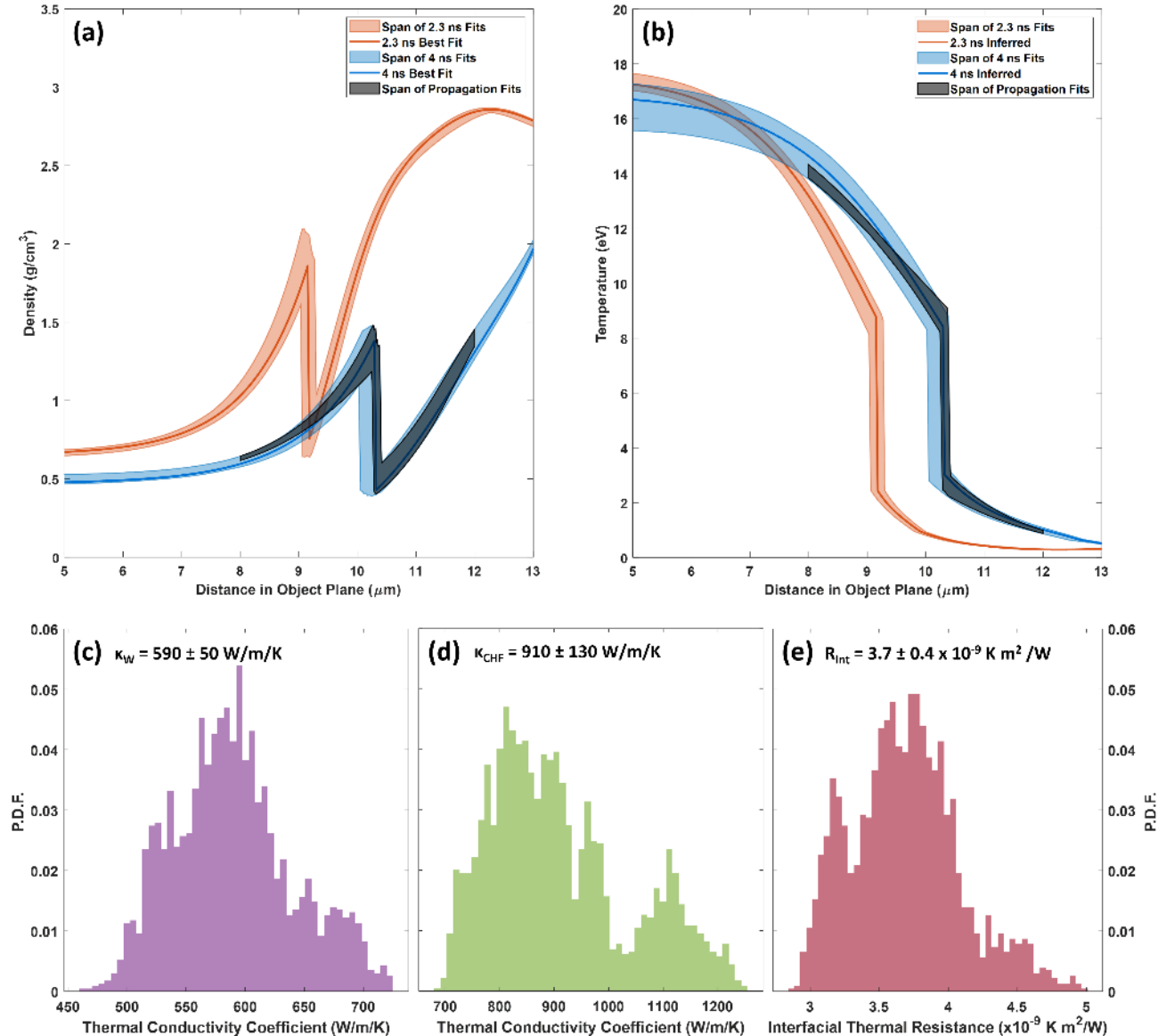
- We get a quantitative number for the ITR that is comparable to previously measured metal-insulator systems at room temperatures*:

- $R_{\text{Int}} \approx 3.7 \times 10^{-9} \text{ K m}^2/\text{W}$
- $G_{\text{Int}} = 1/R_{\text{Int}} \approx 270 \text{ MW/m}^2/\text{K}$

We expect similar effects at many interfaces in the HED regime, however the theory is still a work in process. For e^-e^- scattering:

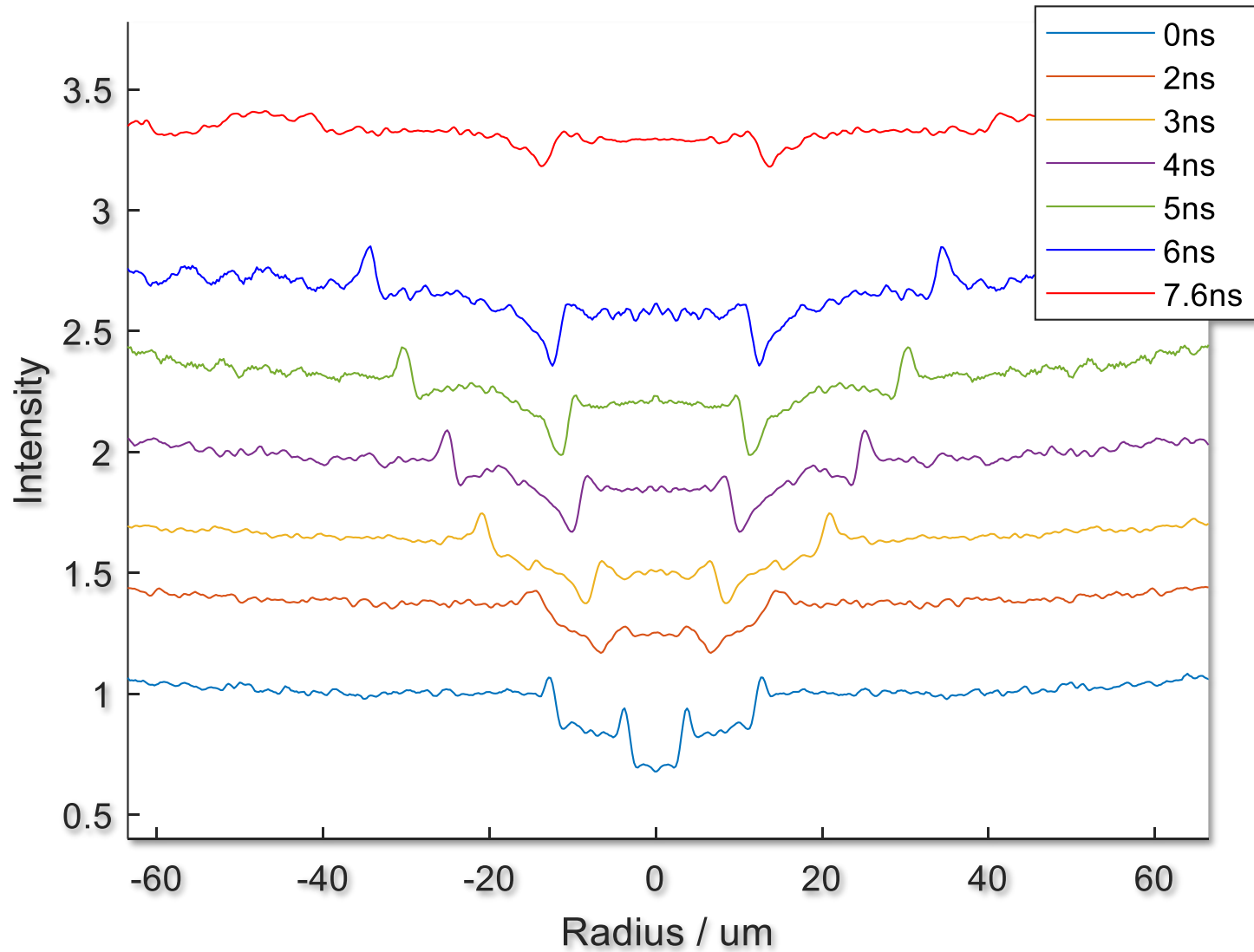
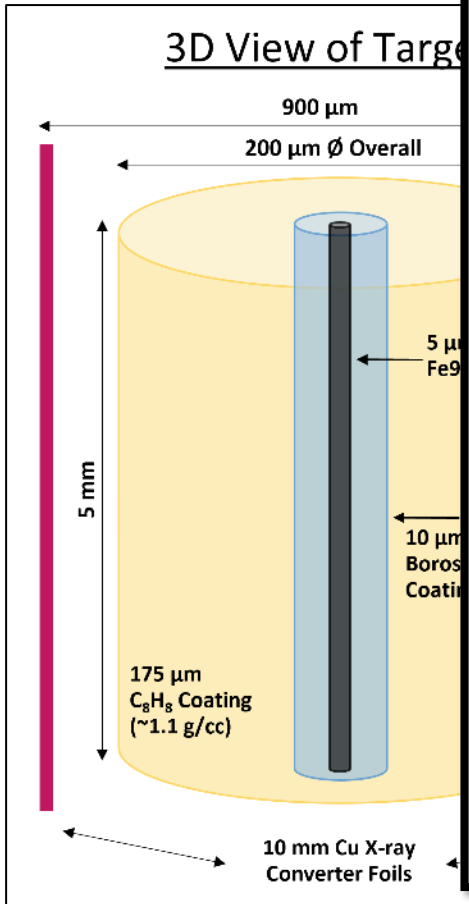
$$h_{I,e} = \frac{1}{4} \int_0^\infty (\varepsilon - \varepsilon_{F,1}) D_1(\varepsilon) \frac{\partial f_1(\varepsilon)}{\partial T} v_{e,1} \zeta_{e,1 \rightarrow 2}(\varepsilon) d\varepsilon,$$

$$\zeta_{e,1 \rightarrow 2}(\varepsilon) = \frac{D_2(\varepsilon) [1 - f_2(\varepsilon)] v_2(\varepsilon)}{D_1(\varepsilon) f_1(\varepsilon) v_1(\varepsilon) + D_2(\varepsilon) [1 - f_2(\varepsilon)] v_2(\varepsilon)}.$$



The future of the FDR platform at Omega

- There are additional goals:
 - Goal is to compare nickel and iron
 - Glass-coated



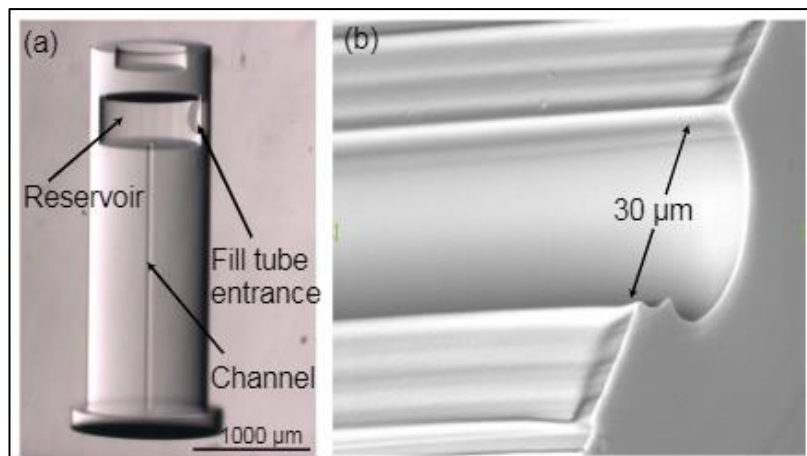
— metals like

The future of the FDR platform – at NIF

- We were awarded time as a part of the NIF Discovery Science allocation
 - Goal is to use cryogenic, liquid deuterium-plastic targets to continue investigating transport properties in materials relevant to ICF
- First shot used a CH-coated W wire, cryo shot is currently in assembly



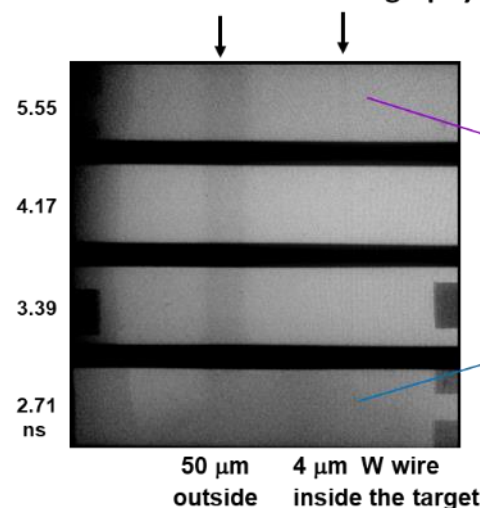
2PP Cryo Target



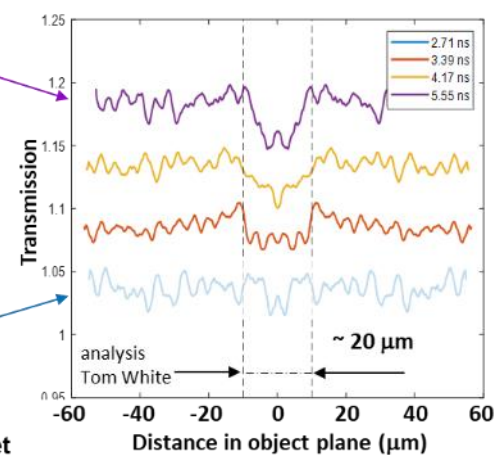
CH-Coated W Wire



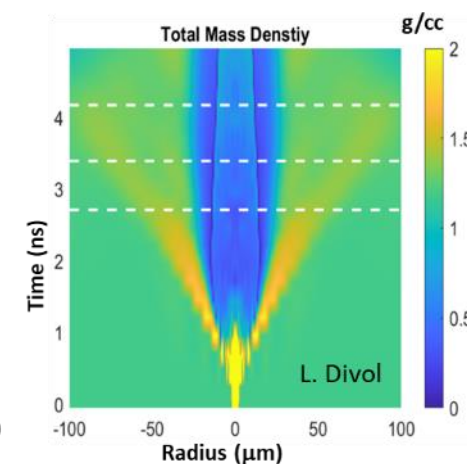
GXD 90-78: RER radiography



Processed lineouts

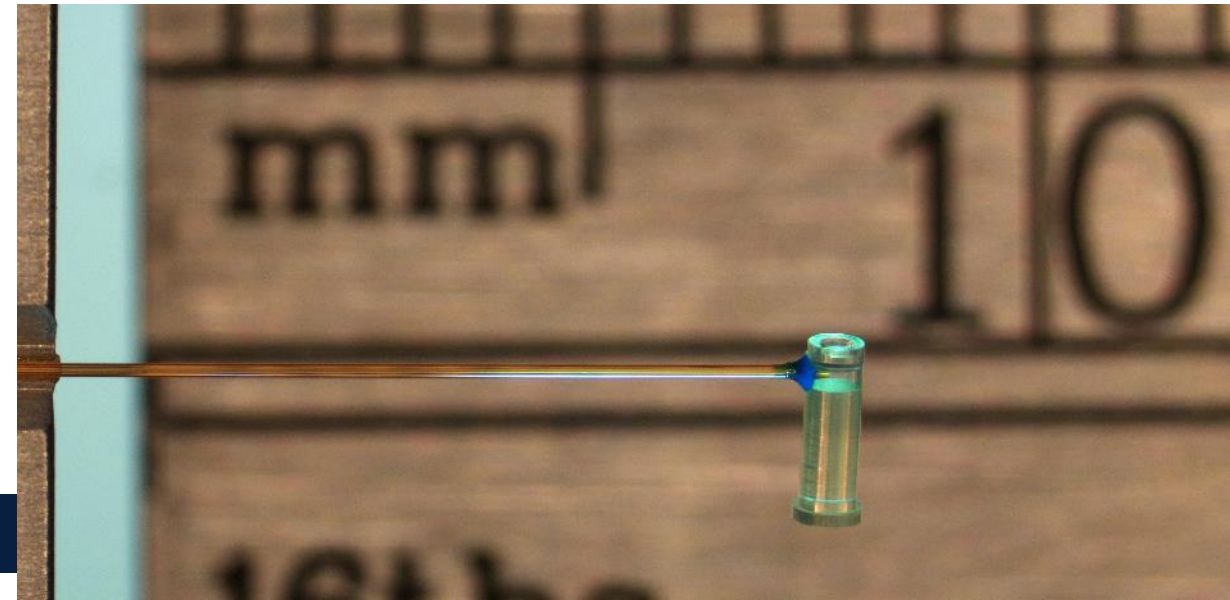
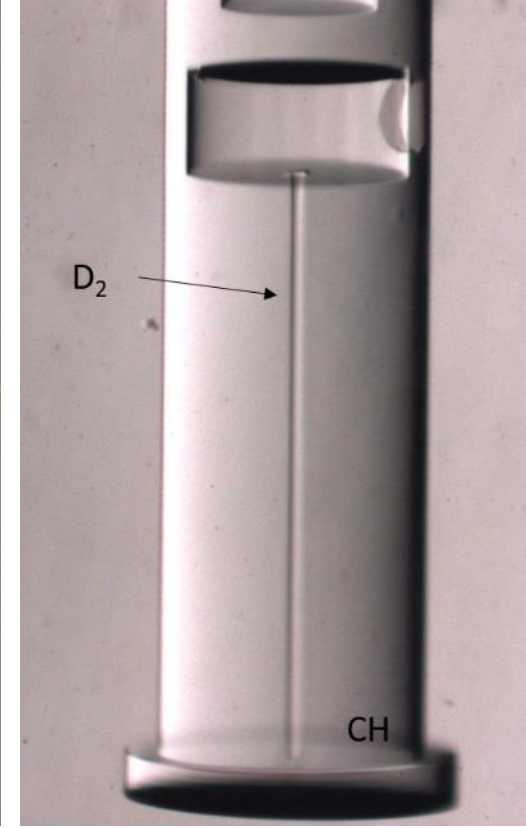
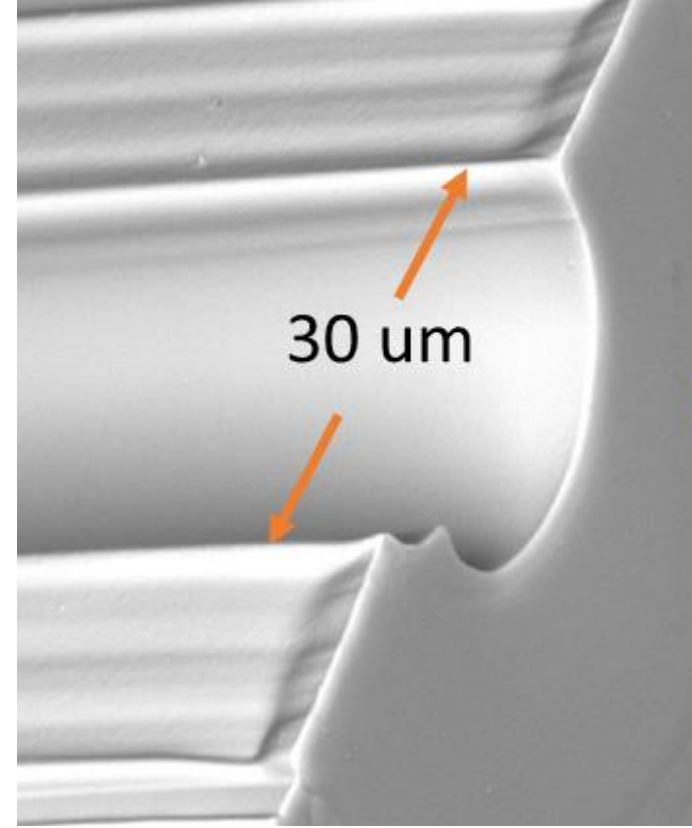


Hydrodynamic simulations



2PP (Two-Photon Polymerization) print

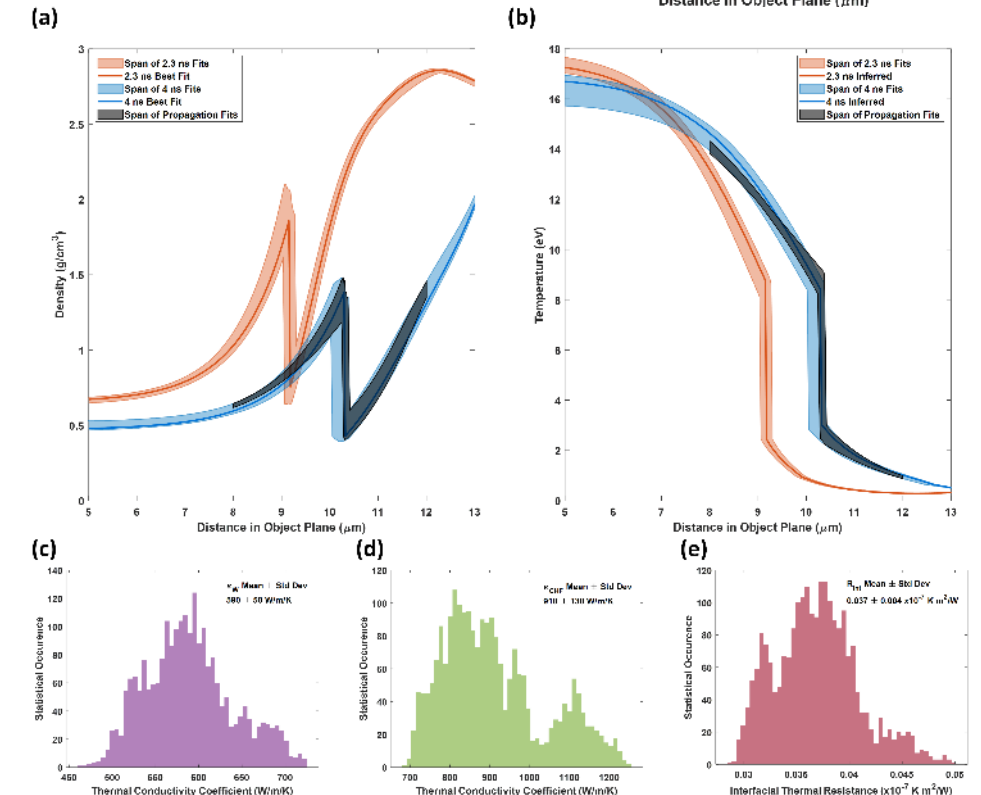
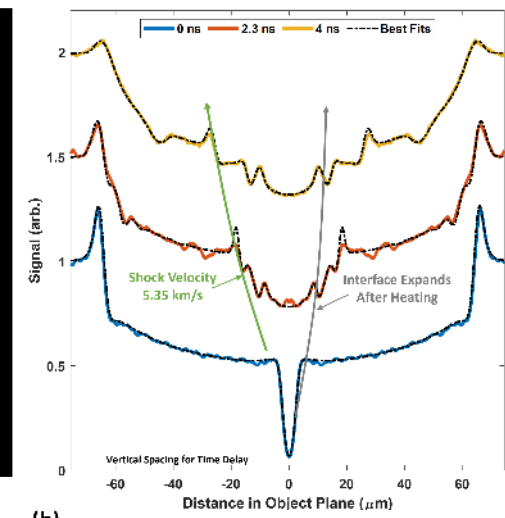
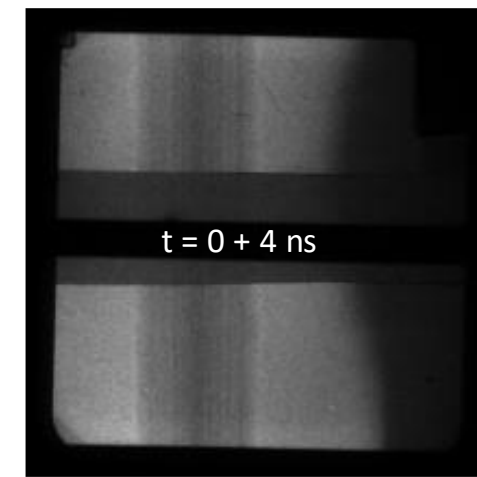
The two-photon polymerization (2PP) technique is based on the interaction of femtosecond laser radiation which induces a highly localised chemical reaction leading to polymerization of the photosensitive material with current resolution down to 100 nm.



Summary

- Using our Fresnel Diffractive Radiography (FDR) setup, we have taken data of an evolving WDM system at multiple times at the OMEGA Laser Facility with $\sim 2 \mu\text{m}$ spatial resolution
- We can accurately simulate the data by using a synthetic density profile, and confirmed the relative uniqueness of the profile with Bayesian analysis
- Using FEOS tables, we propagated our data from 2.3 ns to match the data at 4 ns by flowing heat through the system, and extracted values of thermal conductivity for warm dense tungsten and plastic
- We measure interfacial thermal resistance in warm dense matter for the first time and find it to play an important role in the evolution of our interface.
- ITR may be more important than previously thought

Thank you for listening!
Questions?
tgwhite@unr.edu





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Strongly Coupled Coulomb Systems (SCCS) is a major series of international conferences for scientists drawn from the Plasma Physics, Astrophysics, and Condensed Matter Physics communities.

SCCS 2025 will provide a forum for the presentation and discussion of research achievements and ideas relating to a variety of plasma, liquid, and condensed matter systems that are dominated by strong Coulomb interactions between their constituents. Each meeting has seen an evolution of topics and emphases that have followed new discoveries and techniques. The field has continued to see new experimental tools and access to strongly coupled conditions, most recently in the areas of warm matter, dusty plasmas, condensed matter, and ultra-cold plasmas.