

The Effects of Pulse Shaping on Laser-Material Interaction Mechanisms

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Acknowledgements

Sandia Collaborators

Dr. David P. Adams

Dr. Robert V. Reeves

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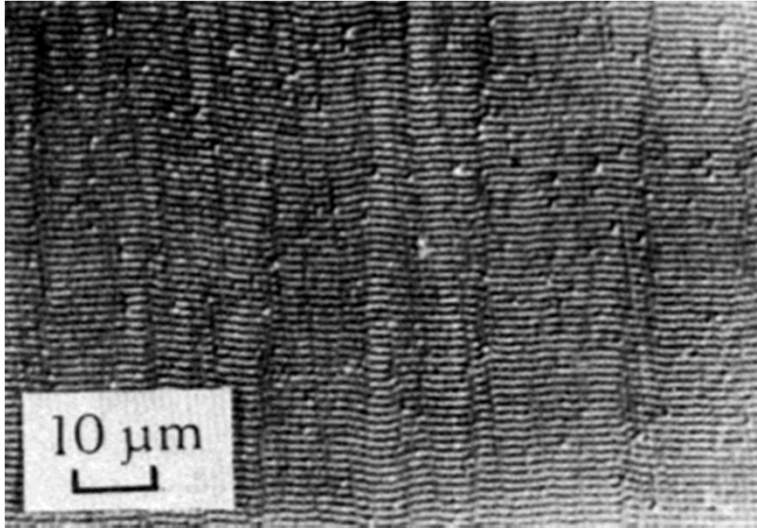
Dr. Ben Torralva

Michael J. Abere

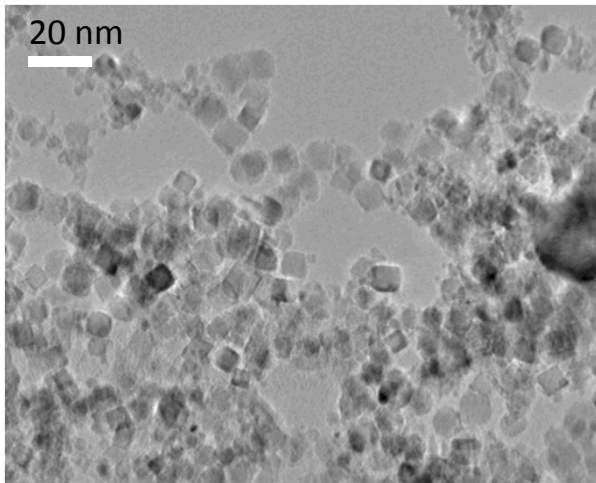
Keegan J. Schrider

Motivation

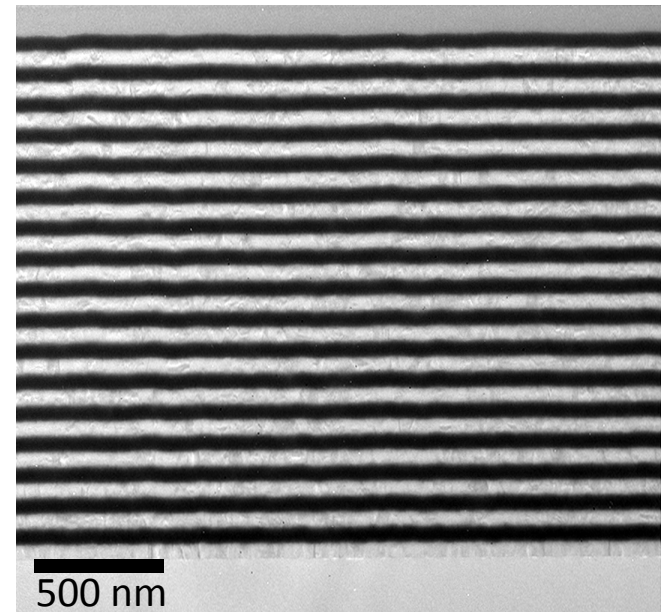
Machining



Nanoparticle Printing



Energetic Systems



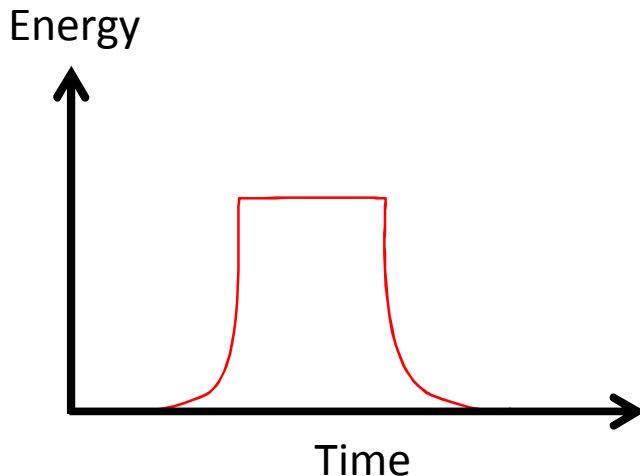
Laser Pulses Temporally Compressed

Pulse Shaping

- Laser light becomes more useful for changing materials when we compress light into *pulses*.
- Intensity of the laser pulse is directly related to pulse *energy* and *length*.
- Both Gaussian and “flat-top” spatial intensity distributions are utilized.

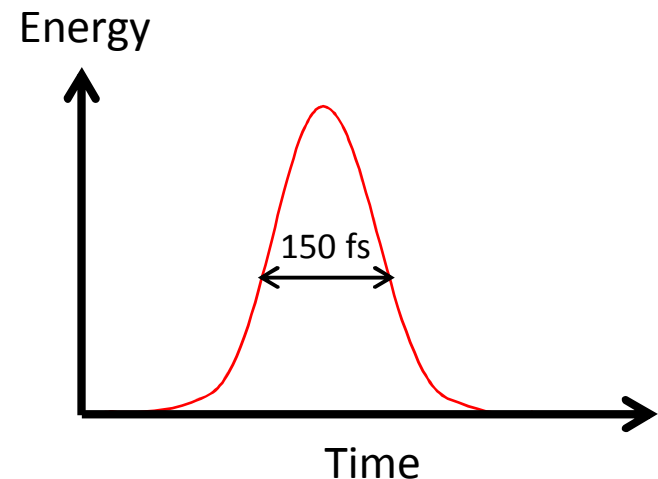
Continuous Wave (CW) + Quasi-CW

$\sim 10^3 \text{ W/cm}^2$



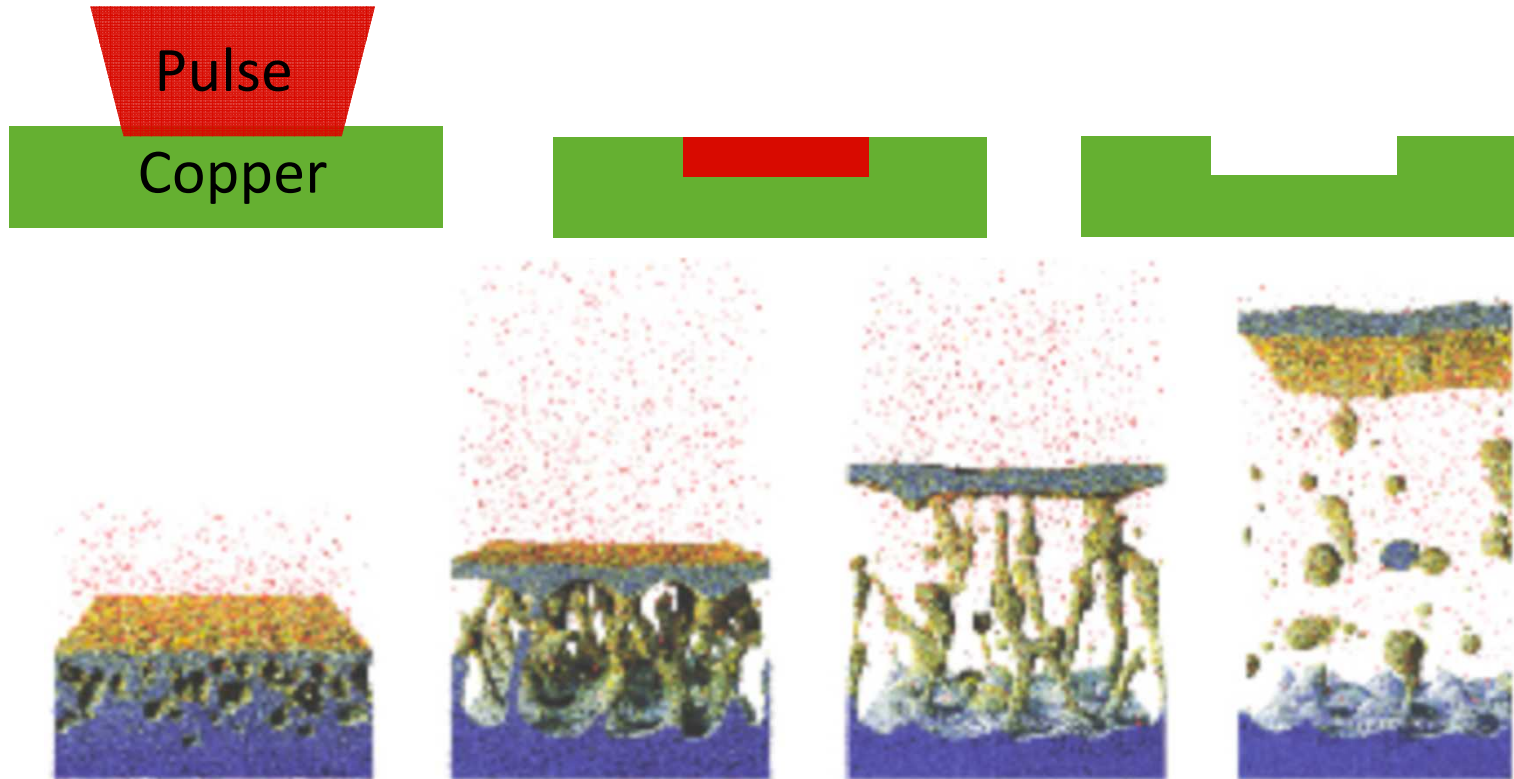
Ultrafast ($\sim 150 \text{ fs}$)

$\sim 10^{12} \text{ W/cm}^2$



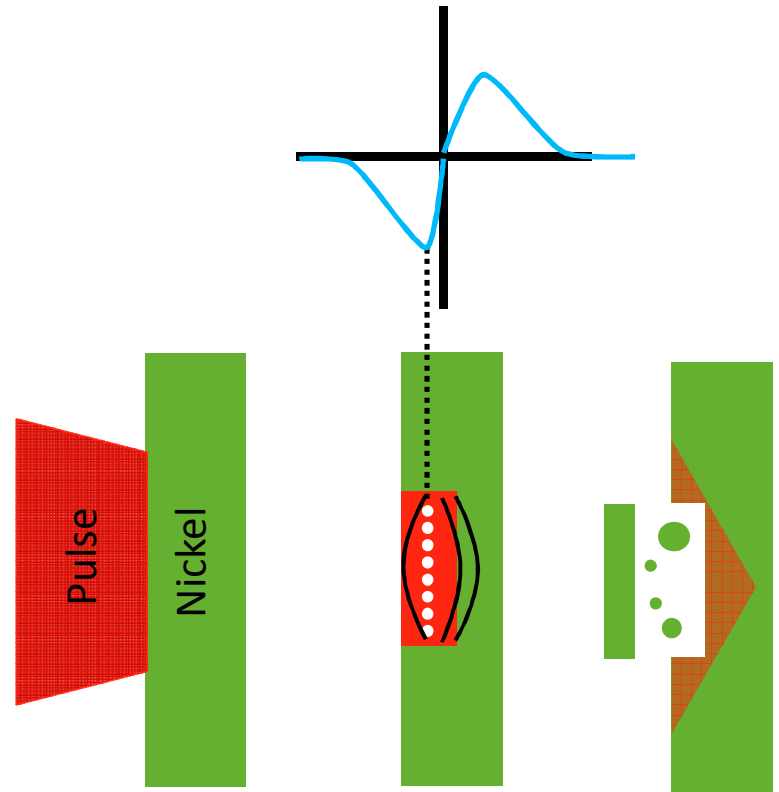
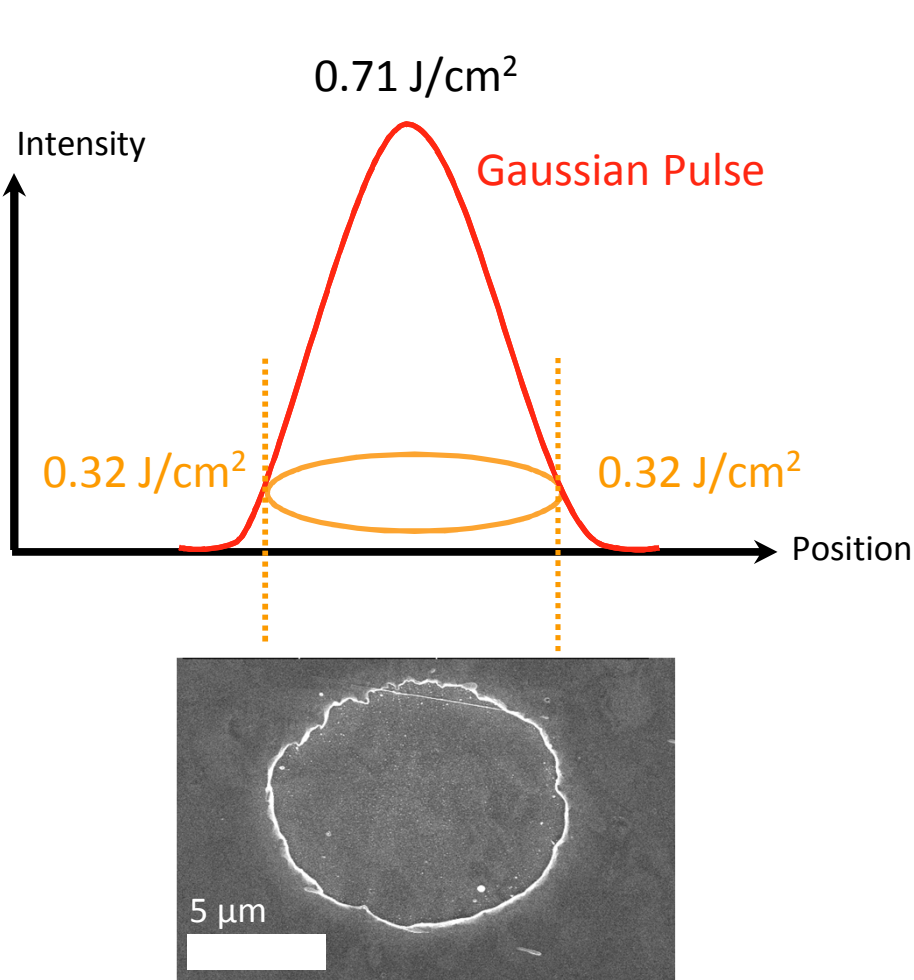
Ultrafast Laser Damage in Bulk Materials

- First few nanometers of material melts in ~ 3 ps.
- Rapid expansion leads to homogeneous void nucleation within melt.
- Material “breaks” where voids nucleate.

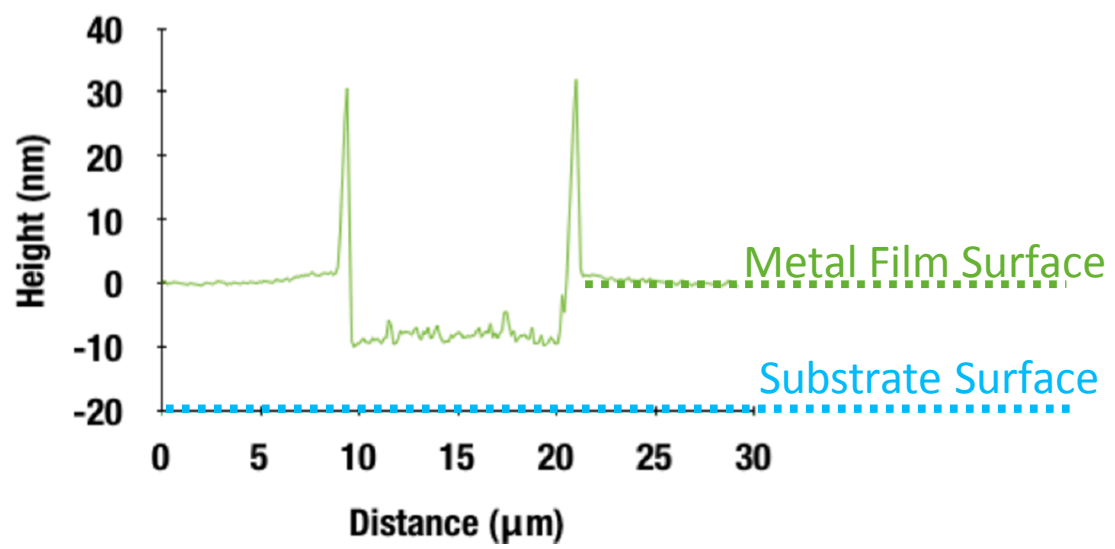
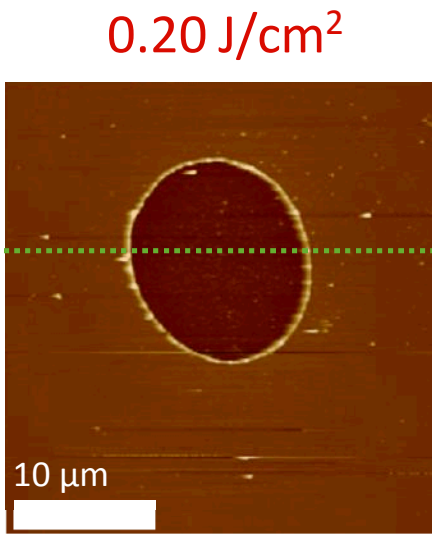
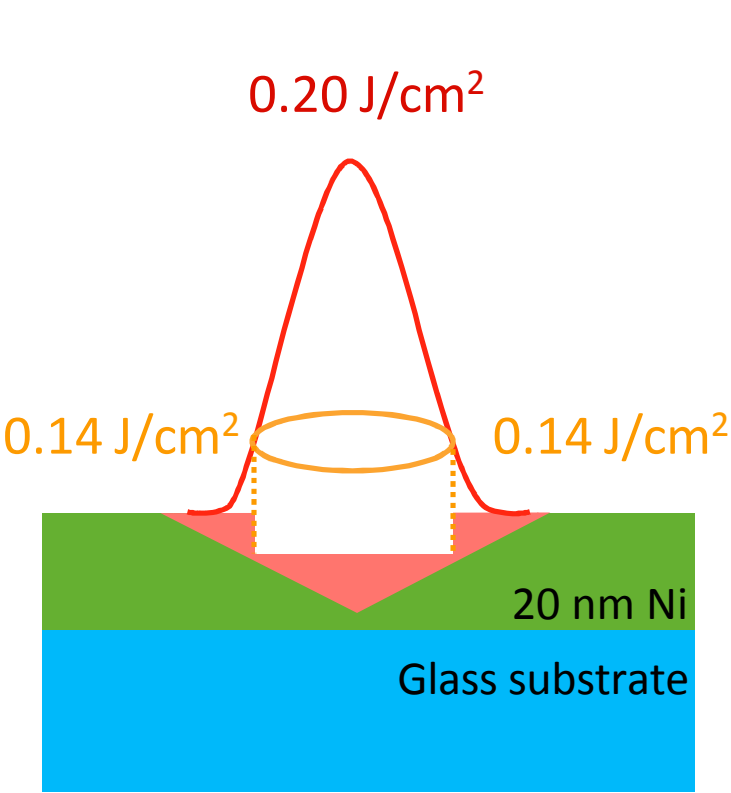


Gilmer (unpublished)

Deterministic Thresholds



Low Fluence Removal Within Film



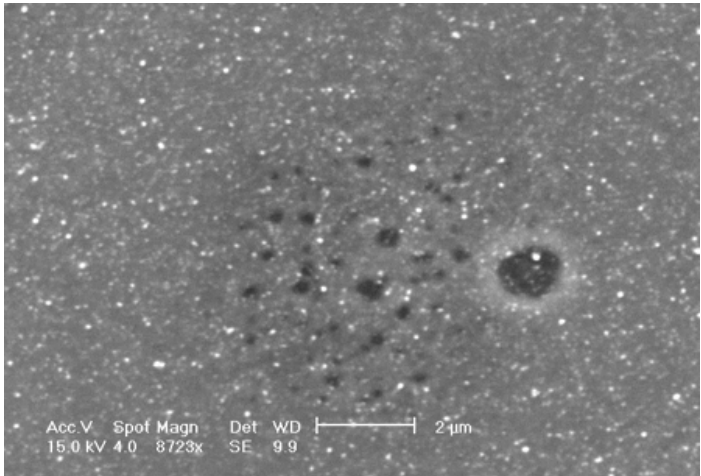
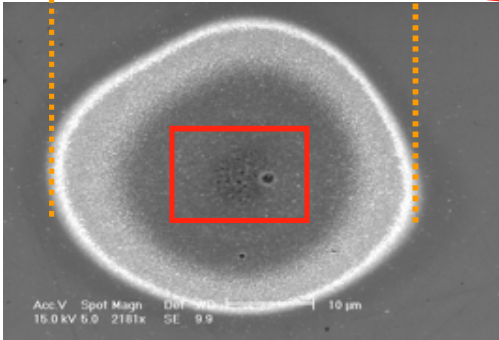
Interfaces Alter Material Removal Thresholds

70 nm Ni film

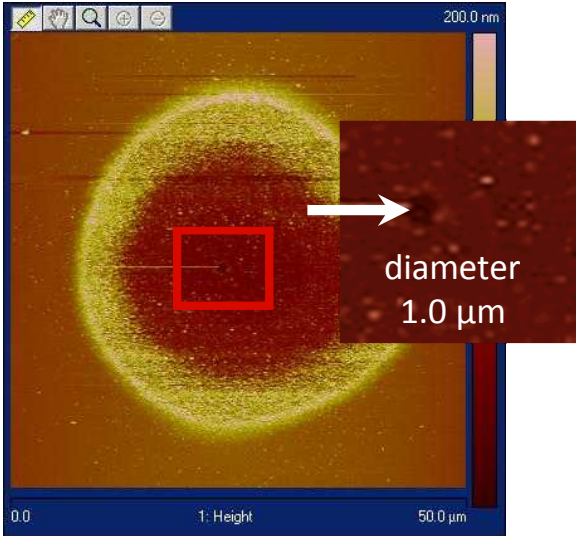
0.95 J/cm²

0.26 J/cm²

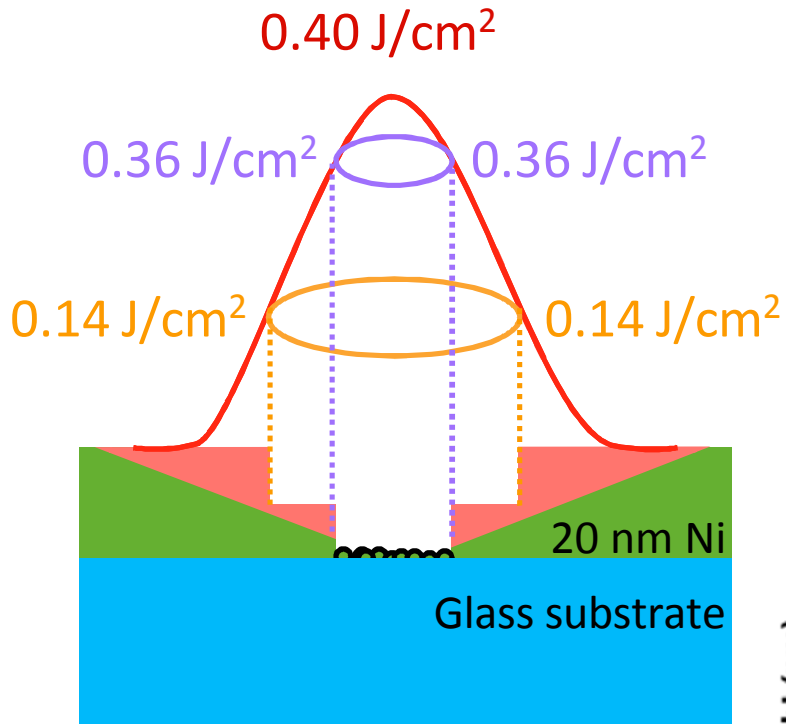
0.26 J/cm²



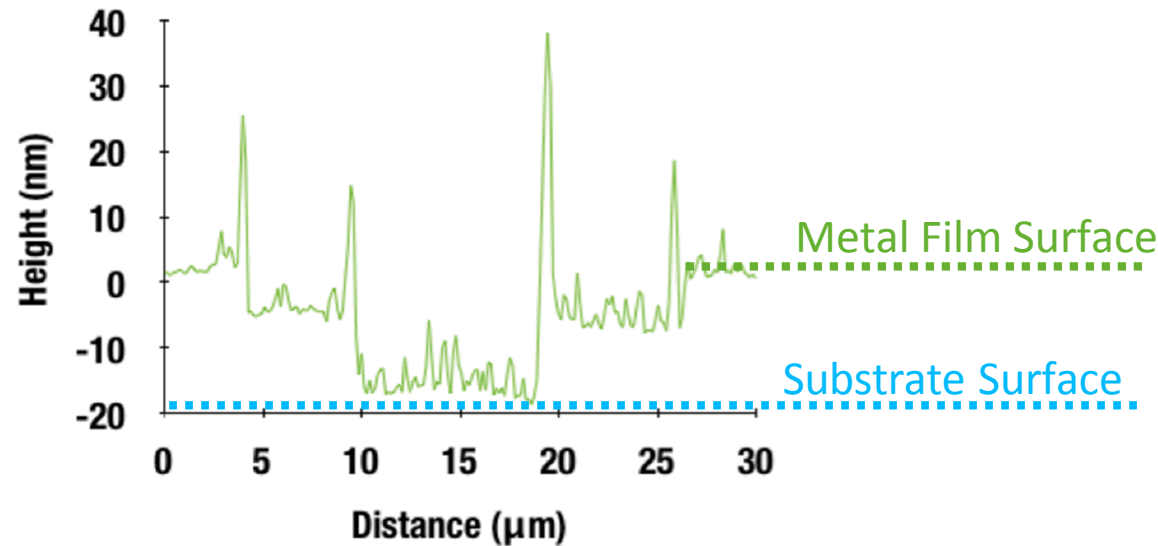
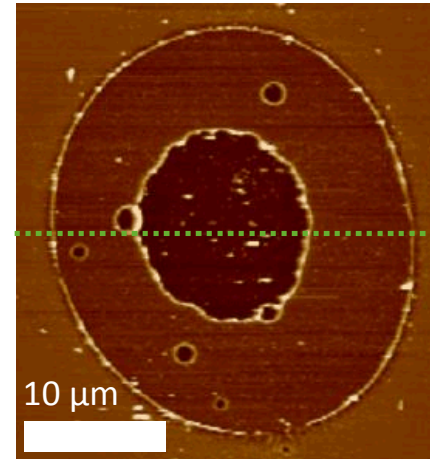
0.95 J/cm²



High Fluence Removal Within Film + at Interface

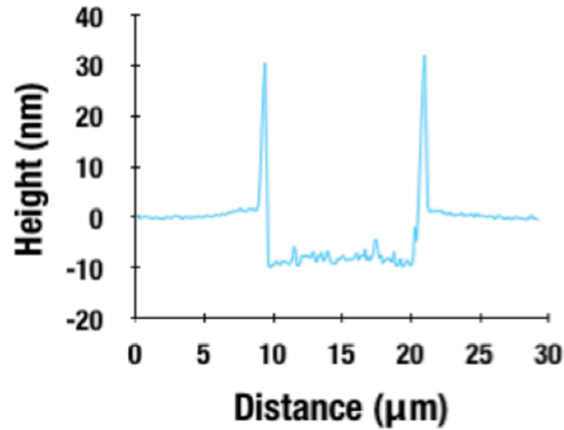
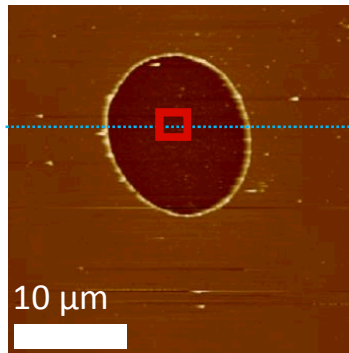


0.40 J/cm^2

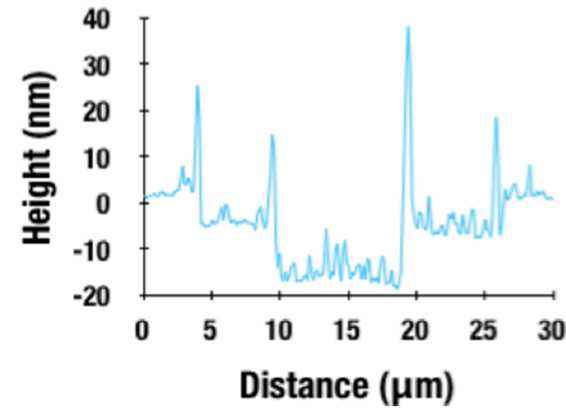
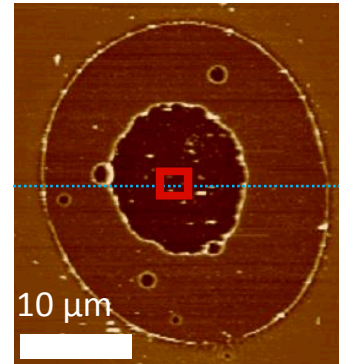


Void Nucleation Controls Surface Roughness

0.20 J/cm²

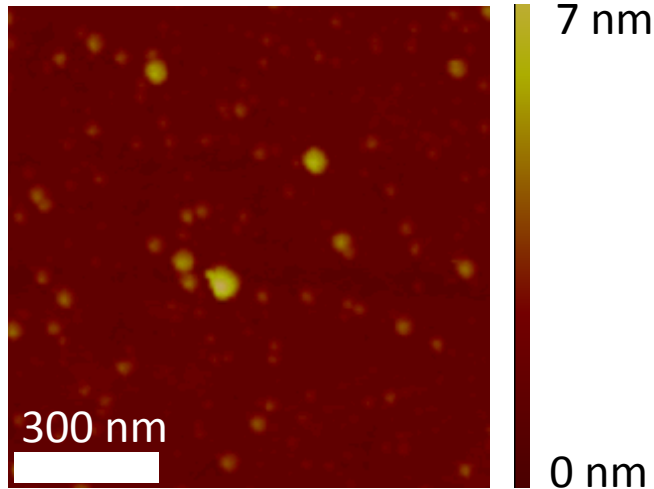


0.40 J/cm²



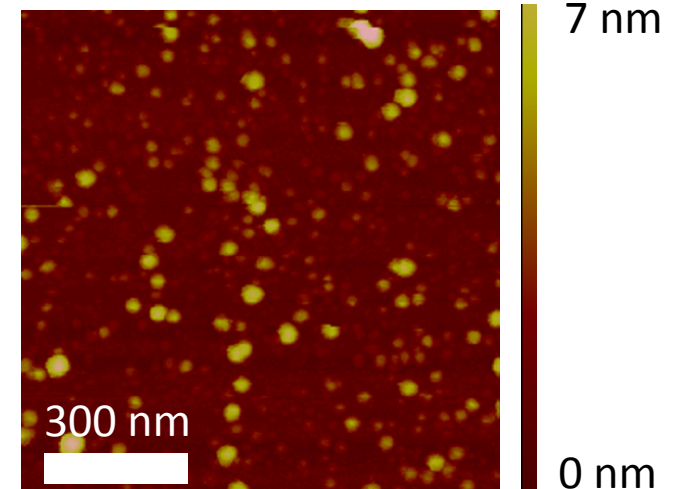
Intra-film Separation

RMS Roughness = 1.27 nm

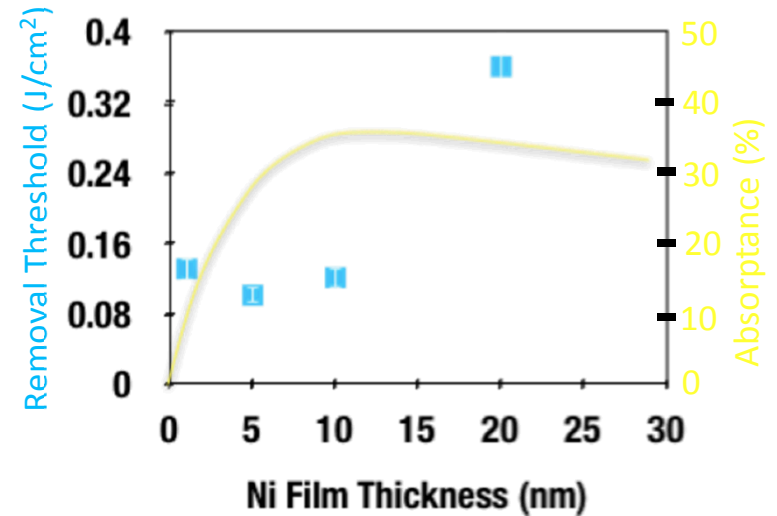
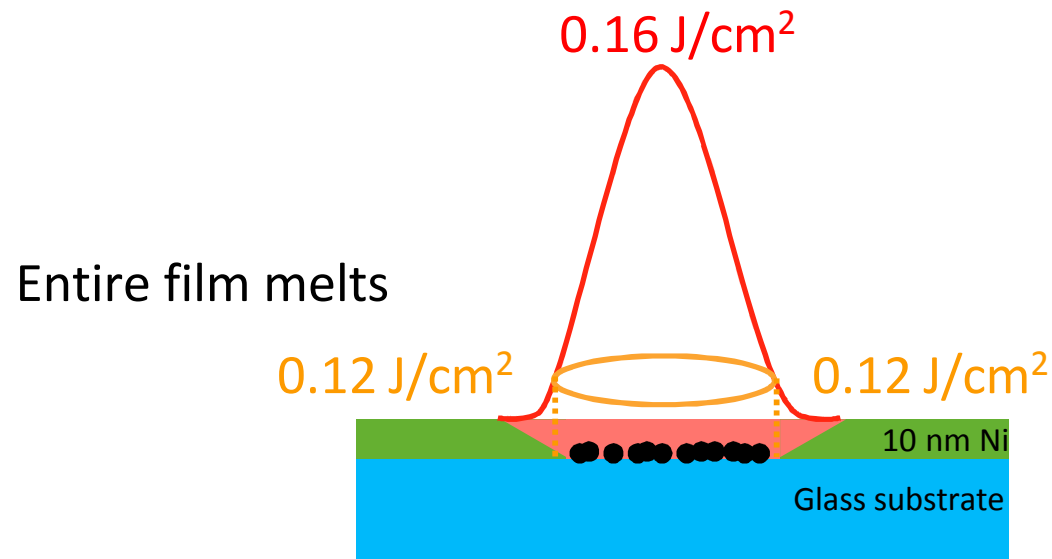


Interface Separation

RMS Roughness = 3.06 nm

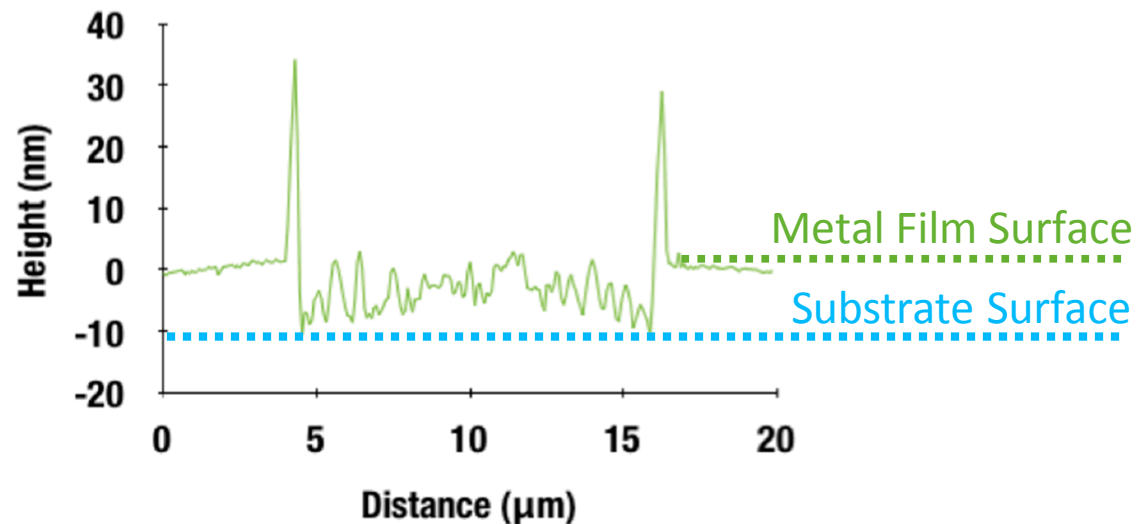
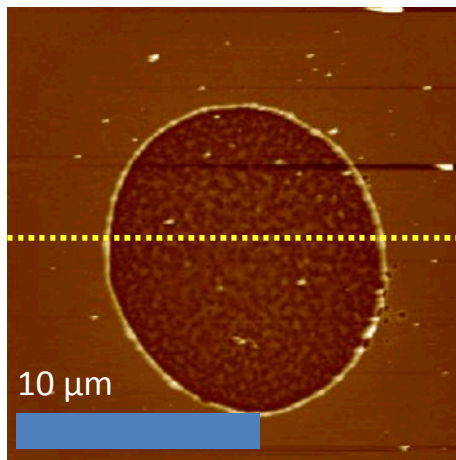


Heterogeneous Nucleation Dominates in Thinnest Films

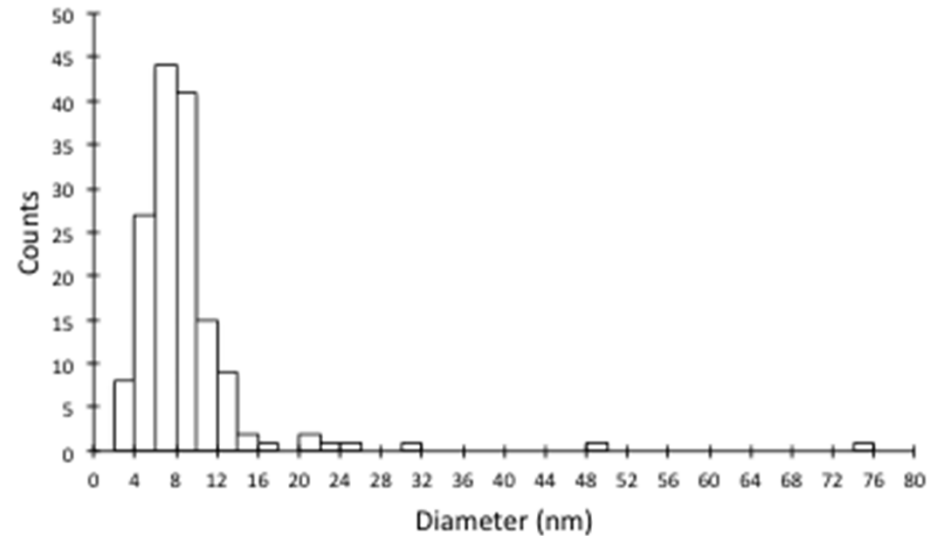
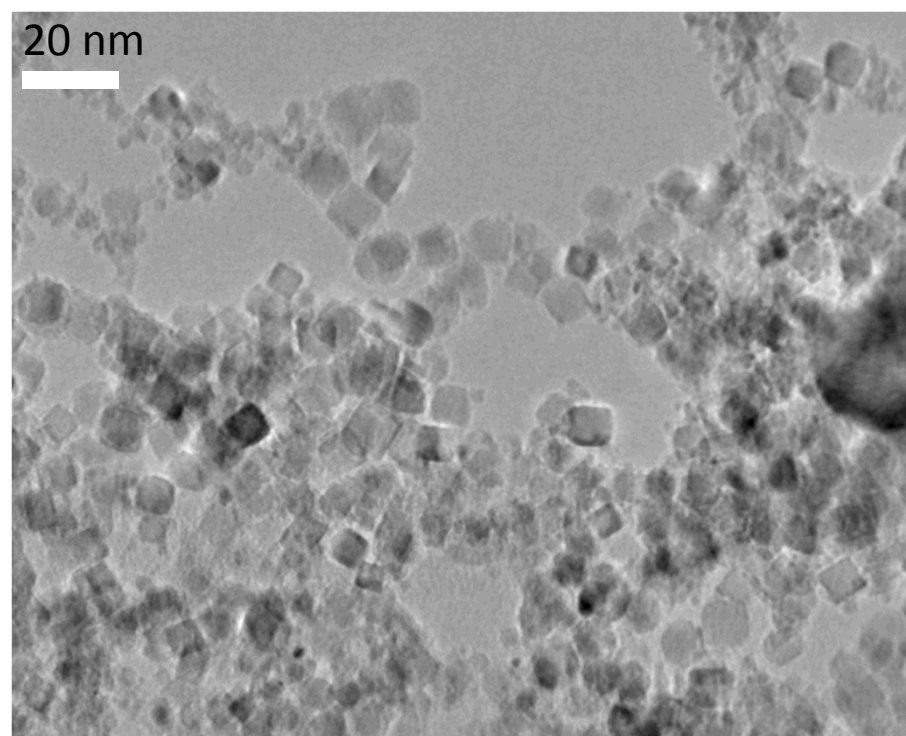
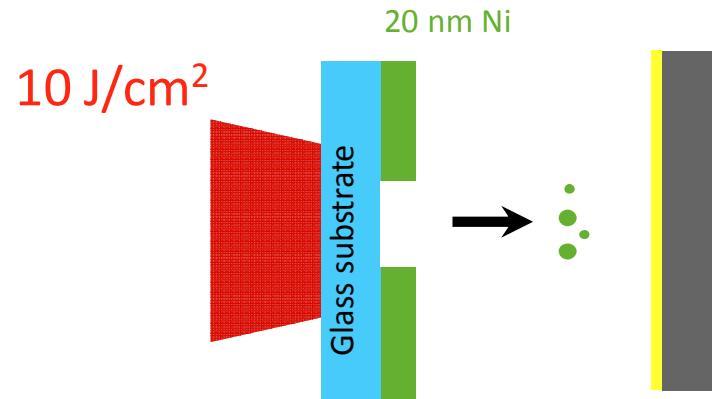


0.16 J/cm²

Heterogeneous Nucleation



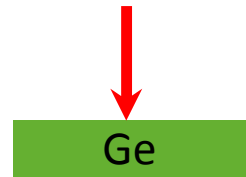
Ni Nanoparticle Printing



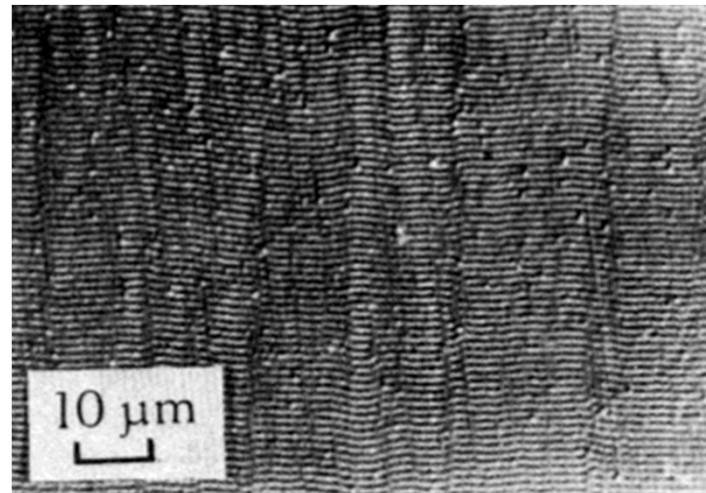
Surfaces Spatially Shape Laser Pulses

Typical Laser Induced Periodic Surface Structures (LIPSS)

- Randomly rough surfaces couple to laser light.
- Interference pattern is formed when scattered light interferes with incident laser light.



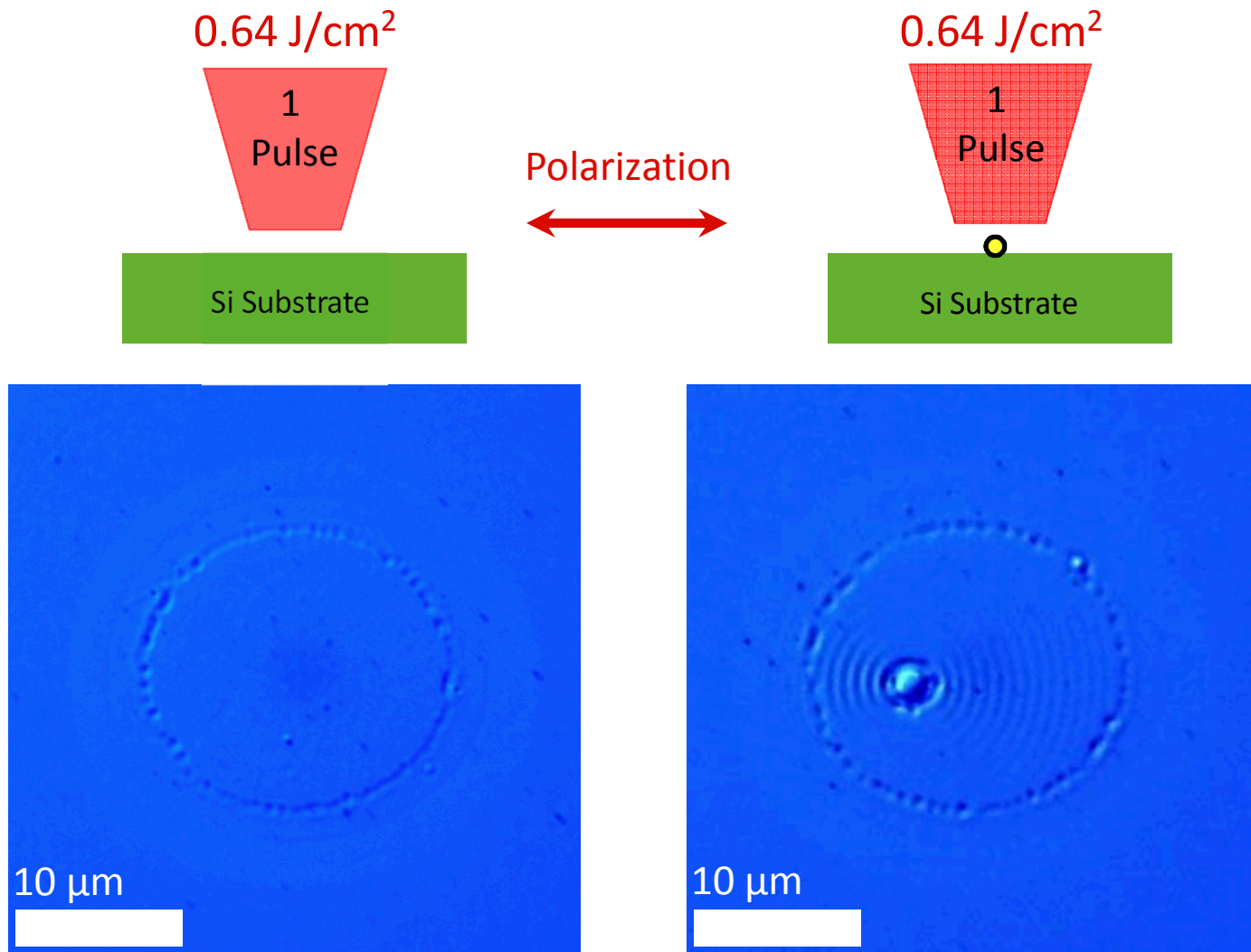
20 ns pulses
Incident $\lambda = 1.06 \mu\text{m}$
 $\Lambda \sim 1.06 \mu\text{m}$
30 pulses



↑↓ Polarization
Direction

LIPSS After 1 Pulse

- LIPSS formation observed near surface defects.



LIPSS Formation After 2 Pulses

0.64 J/cm²

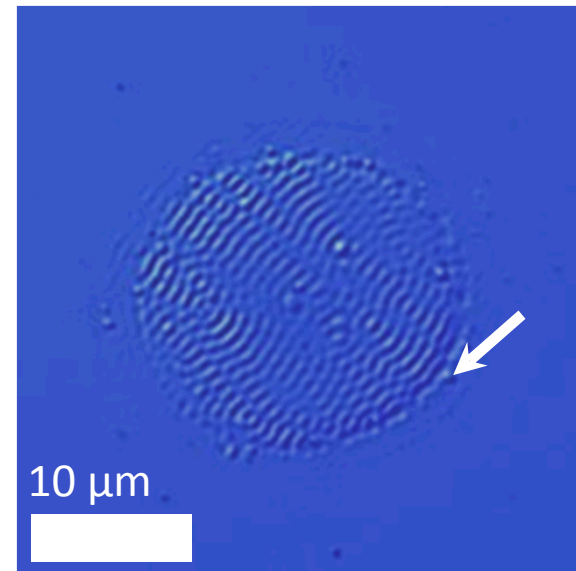
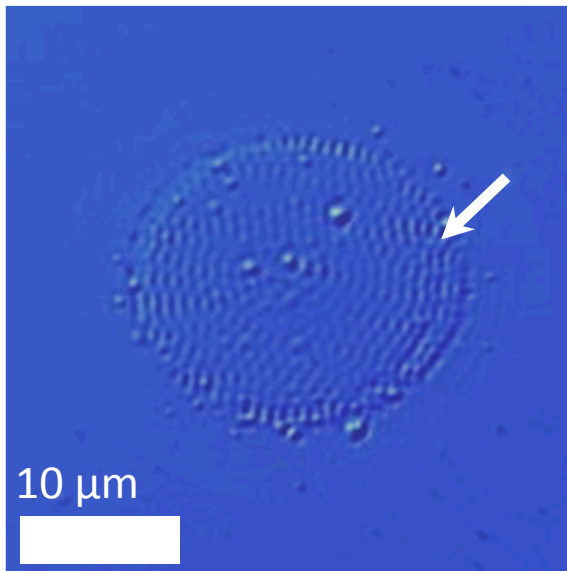
2
Pulses

Si Substrate

Average Period = 757 nm
Standard Deviation = 100 nm



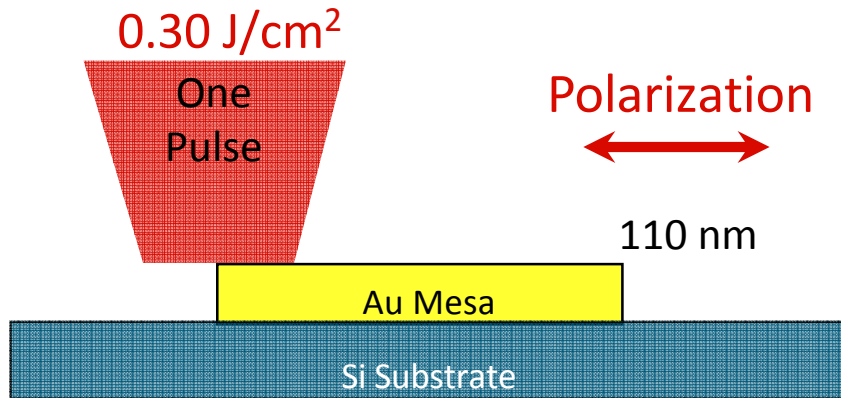
Polarization



LIPSS Formation Near Au Microstructures

Side View

Fundamental Wavelength = 775 nm

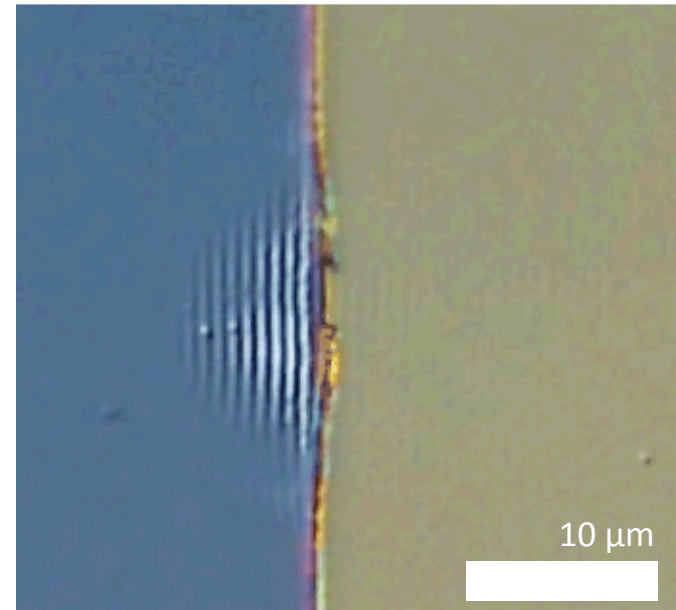
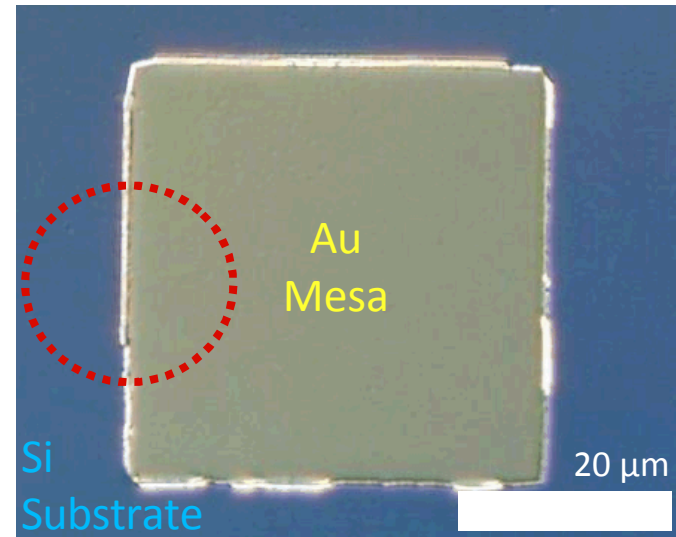


Au melt threshold $\sim 0.55 \text{ J/cm}^2$

Si melt threshold $\sim 0.20 \text{ J/cm}^2$

- Below crater threshold
- Above melt threshold

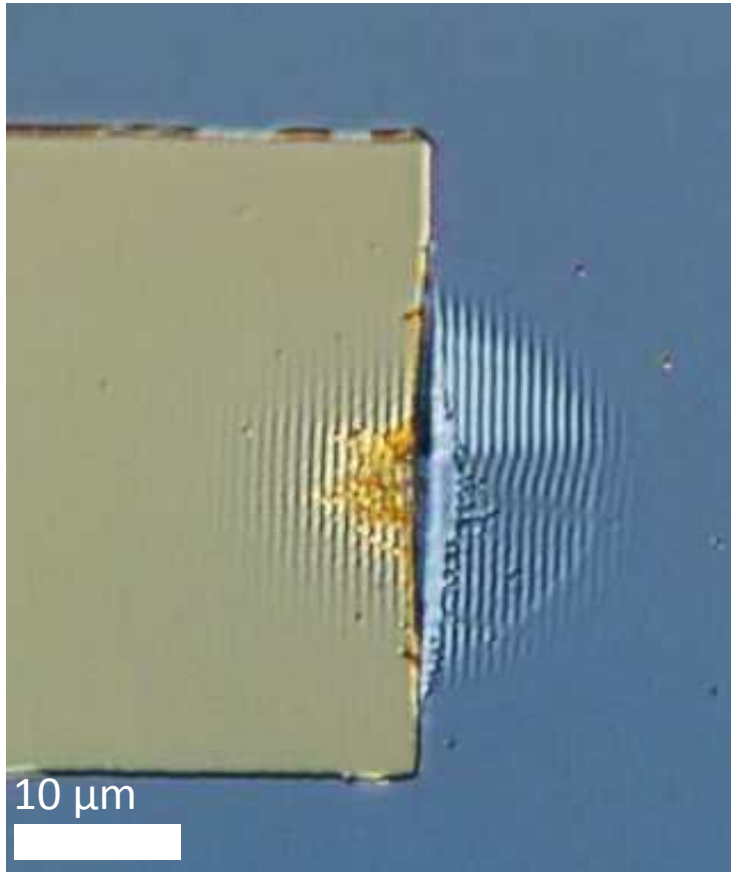
Top View



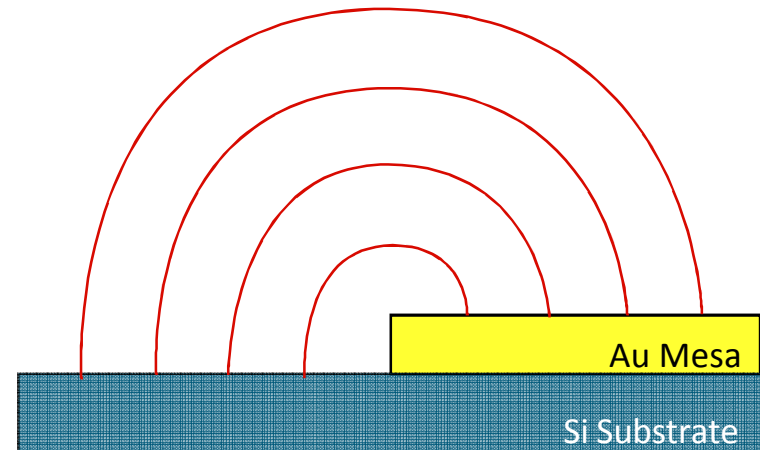
LIPSS Formation Near and On Top of Au Microstructures

0.60 J/cm²

Polarization



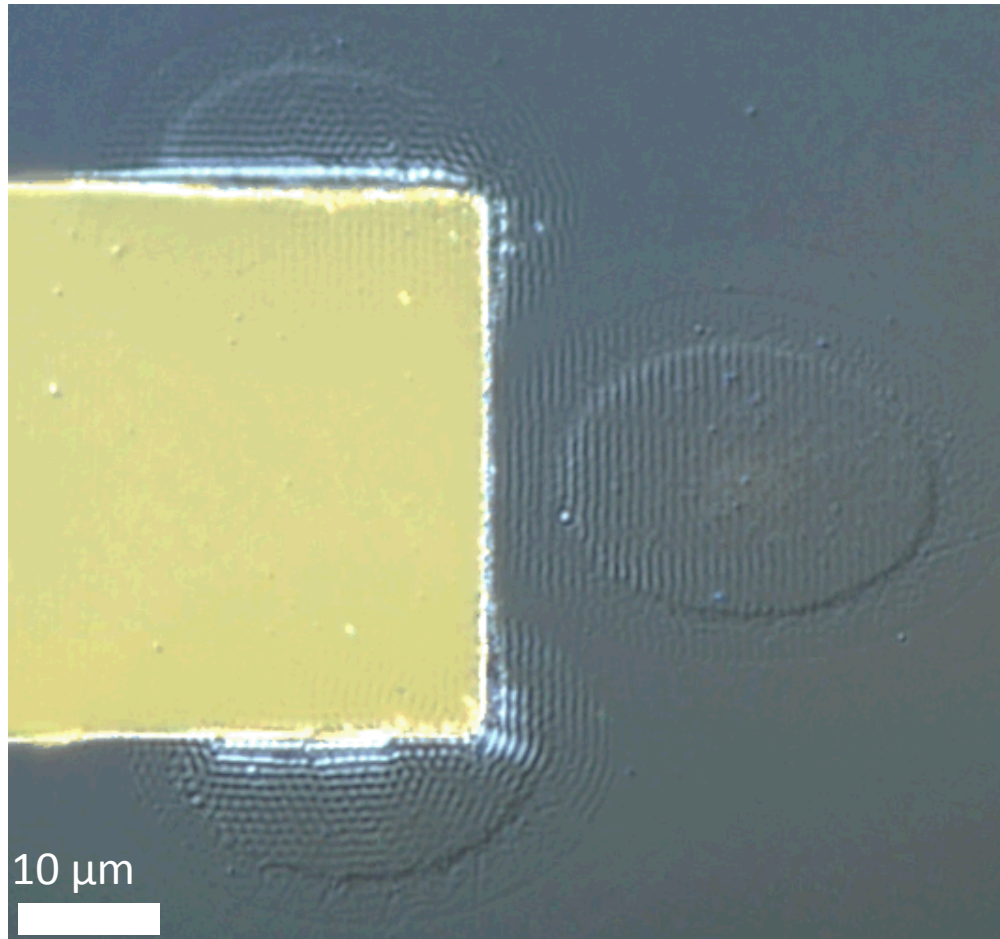
Diffracted Light
Side View



Mesa Edges Act As Sources for 1 pulse LIPSS Formation

0.75 J/cm²

Polarization

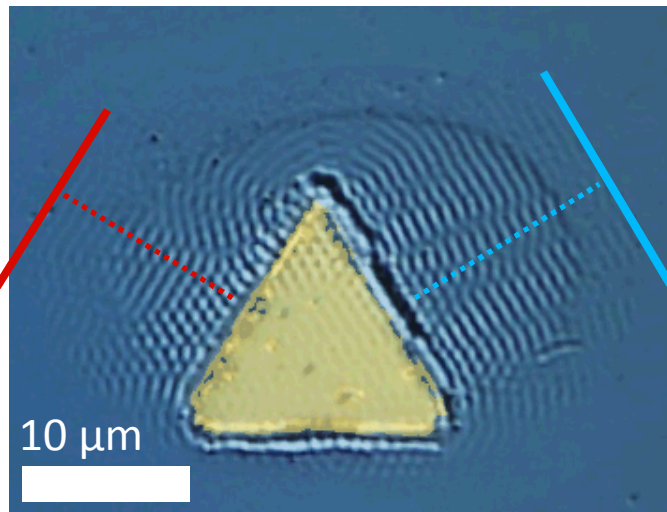


10 μm

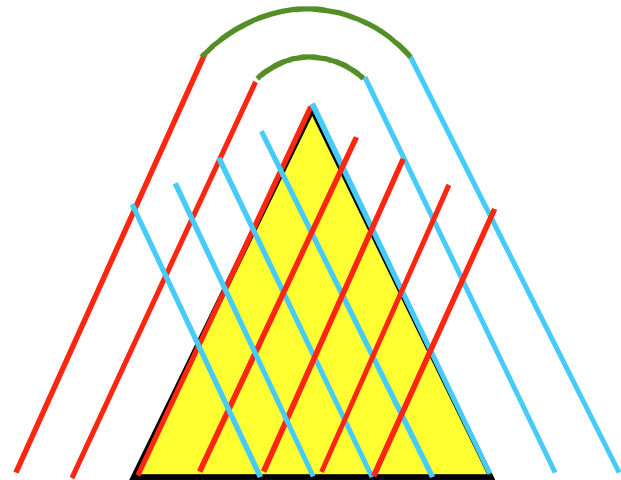
LIPSS Formation from Multiple Sources

0.50 J/cm²

Polarization



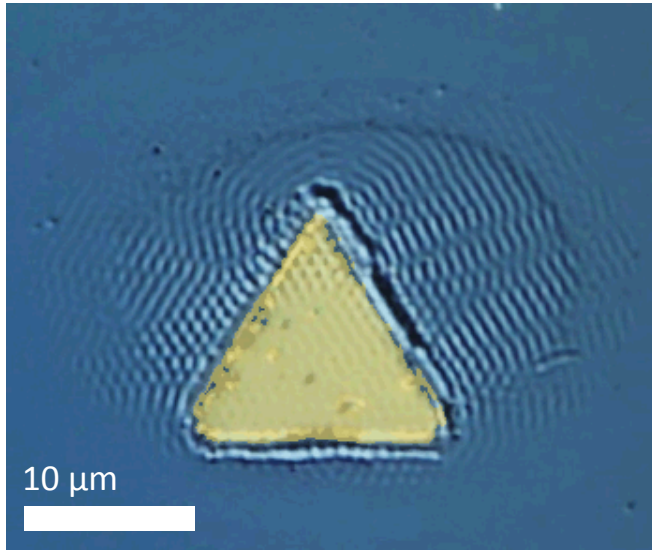
Diffracted Light
Top View



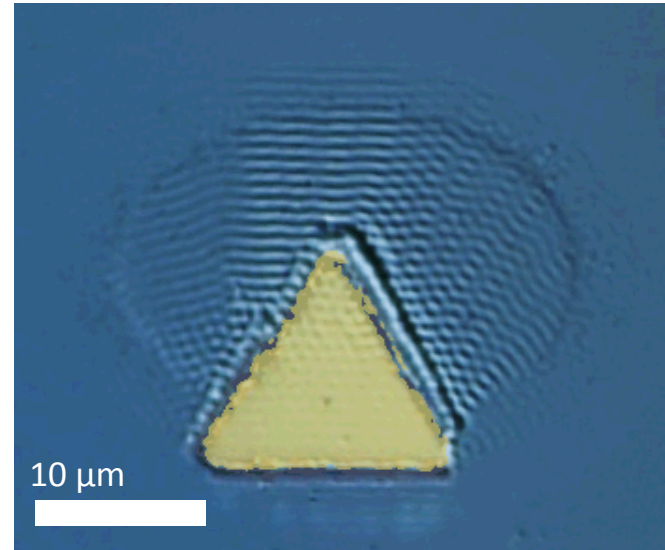
Dependence on Polarization

0.50 J/cm²

Polarization



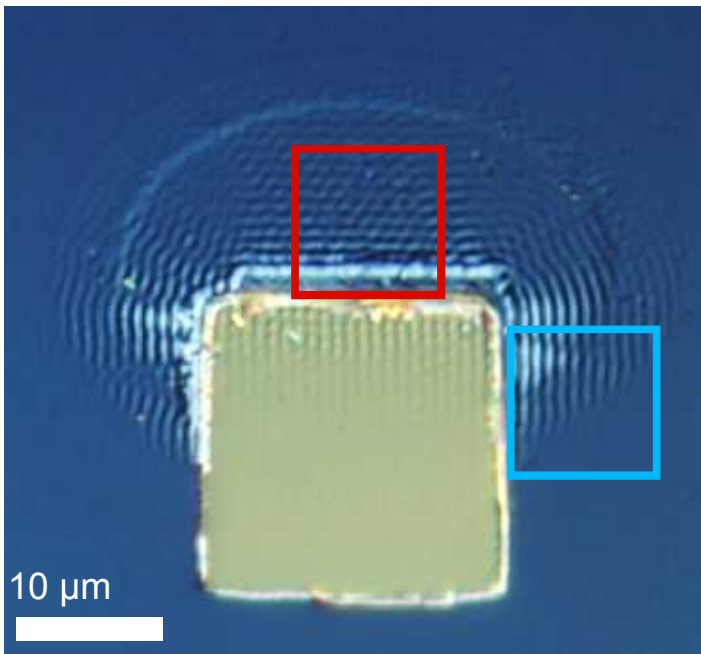
Polarization



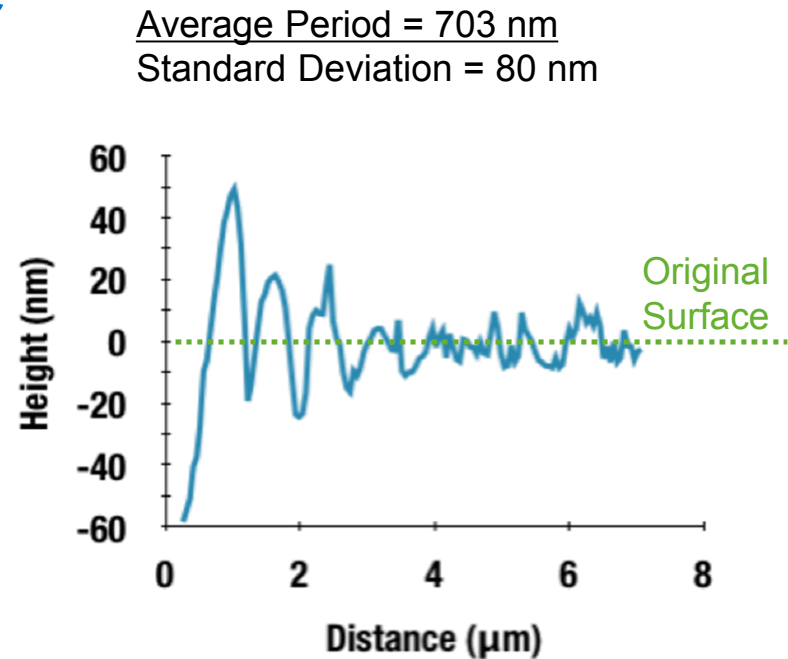
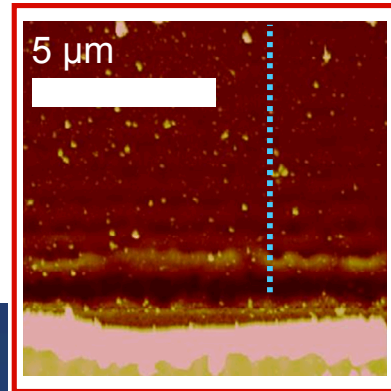
Two Mechanisms Playing a Role

- Near-field diffraction
- Surface plasmon excitation

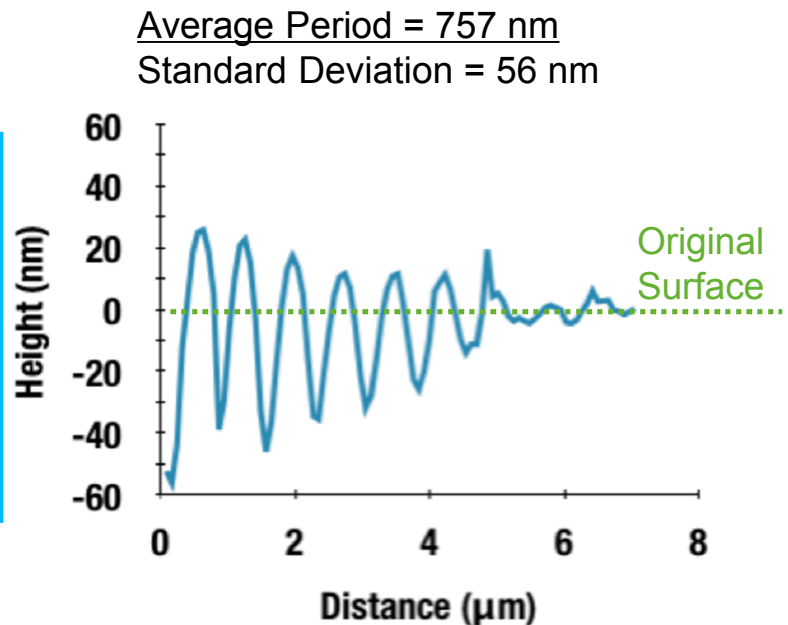
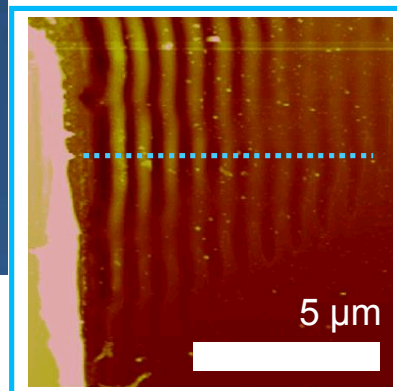
0.50 J/cm^2
Polarization



0.50 J/cm^2



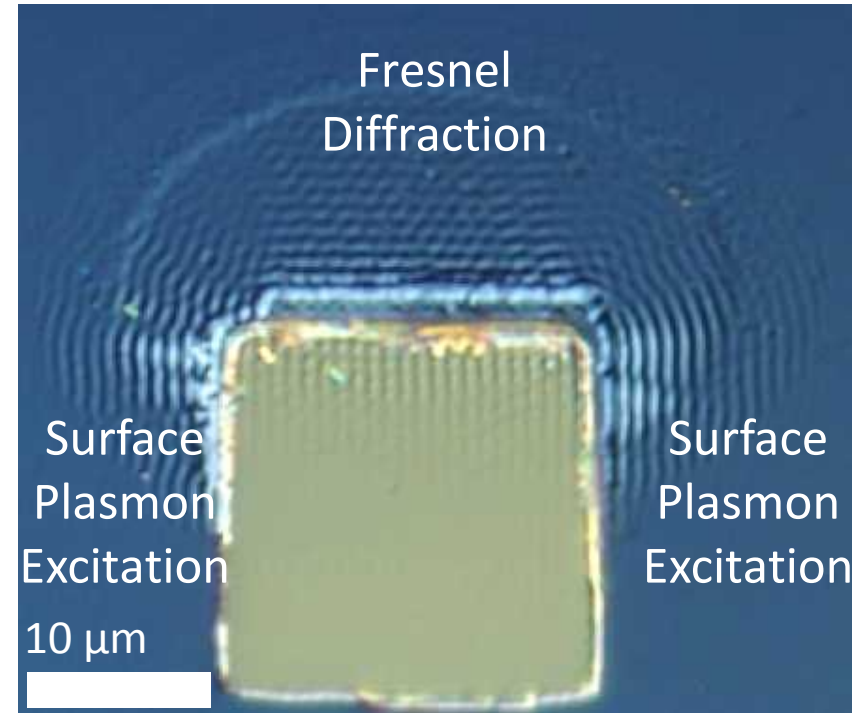
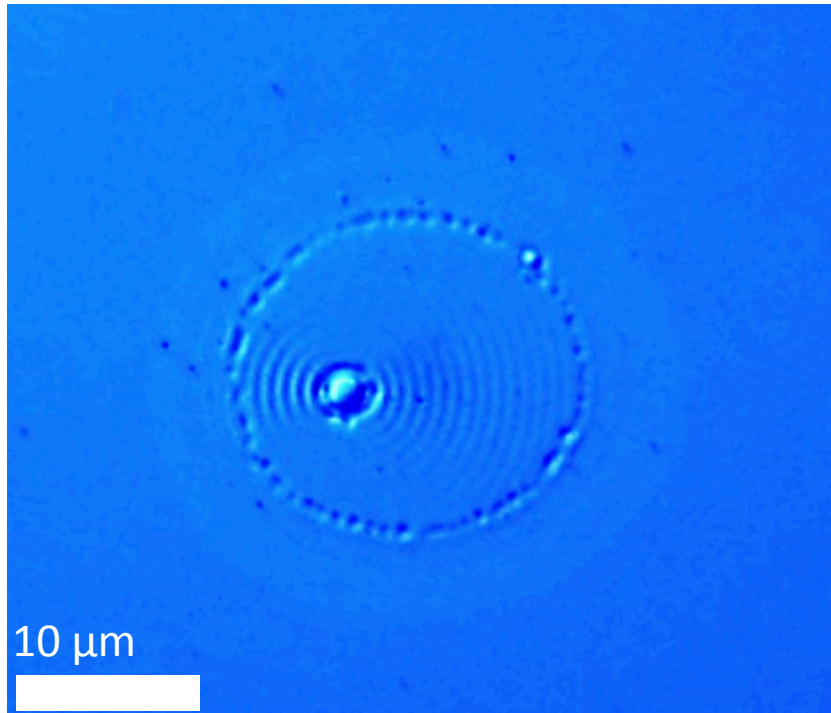
0.34 J/cm^2



LIPSS Formation: Sources + Material Removal

- The active LIPSS formation mechanism depends on the angle of the polarization vector with respect to surface features.
- LIPSS formation is due to material removal.

Polarization



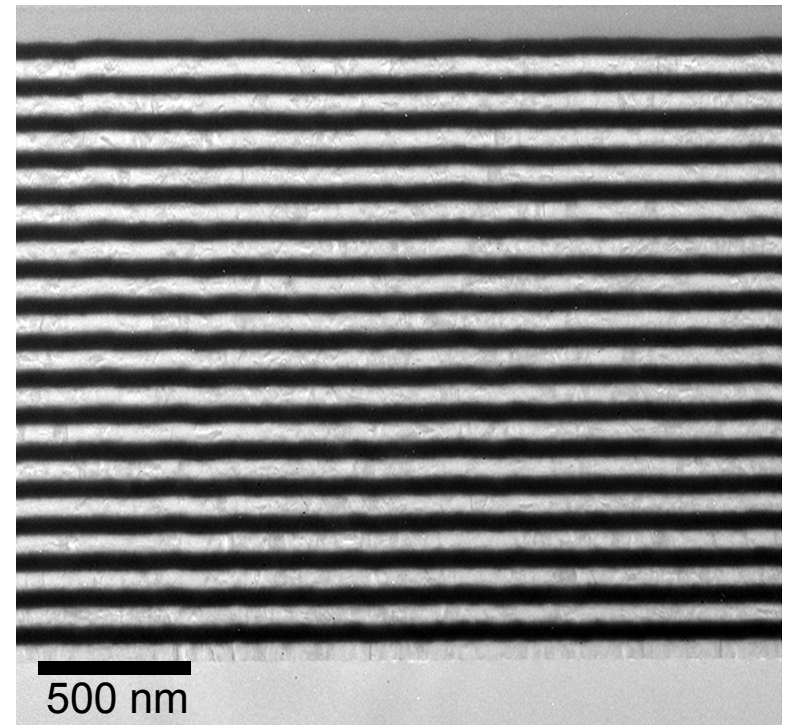
Temporal and Spatial Pulse Shaping

Reactive Multilayers



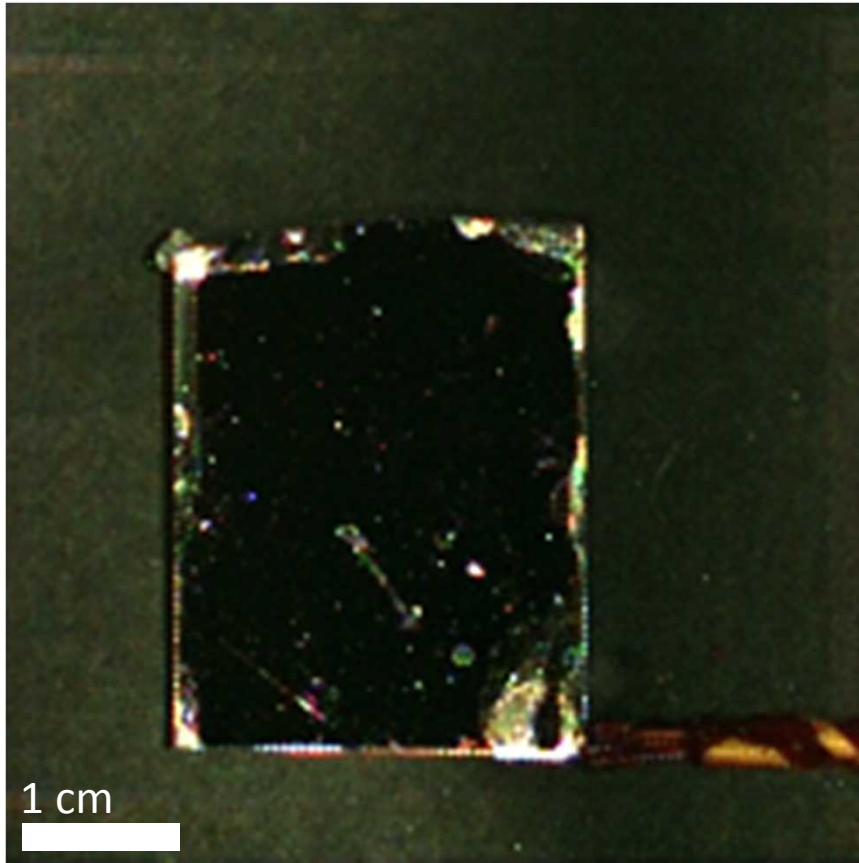
- Exothermic heat generation upon ignition.
- Self-propagating reaction.
- DC Magnetron sputtered layers
- 10 - 15 Å thickness variation
- 1 to 1 Al/Pt ratio
- Adiabatic reaction temperature = 2798 °C
- Melting not required for ignition

Al/Pt multilayer
TEM Cross-section

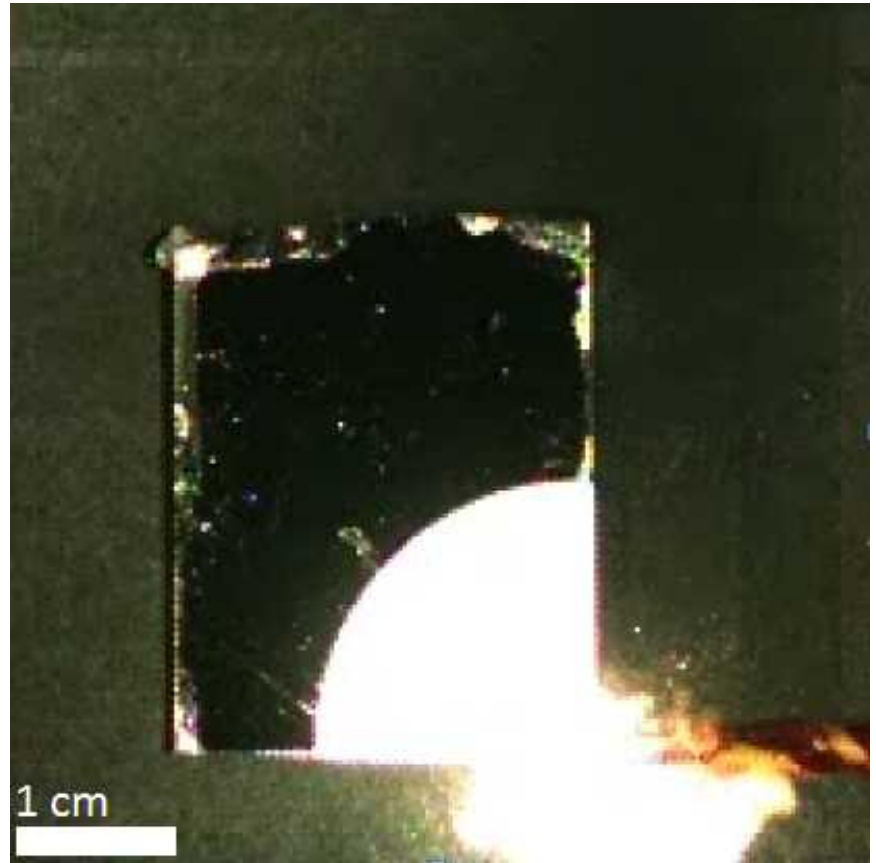


Ignition and Reaction Propagation

Ignition by capacitive discharge

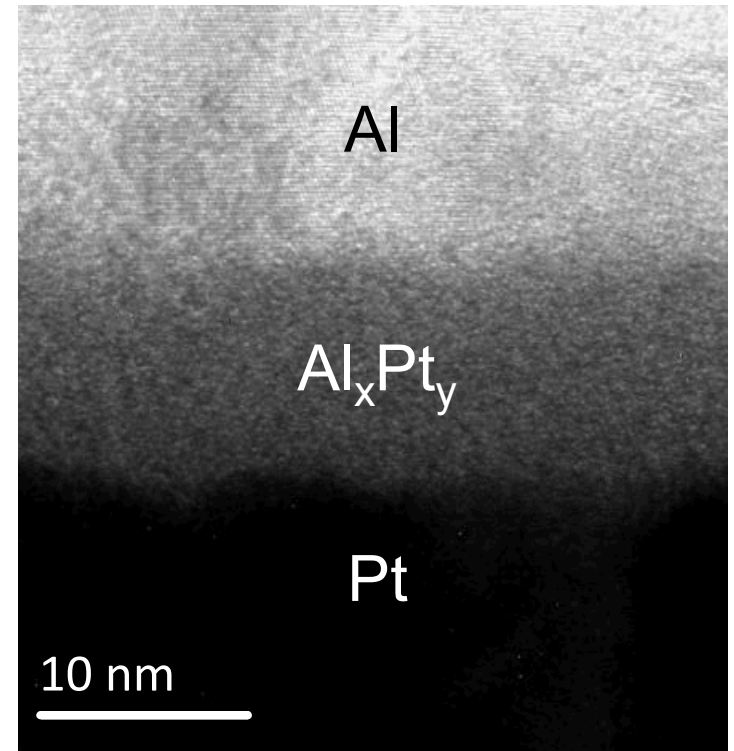
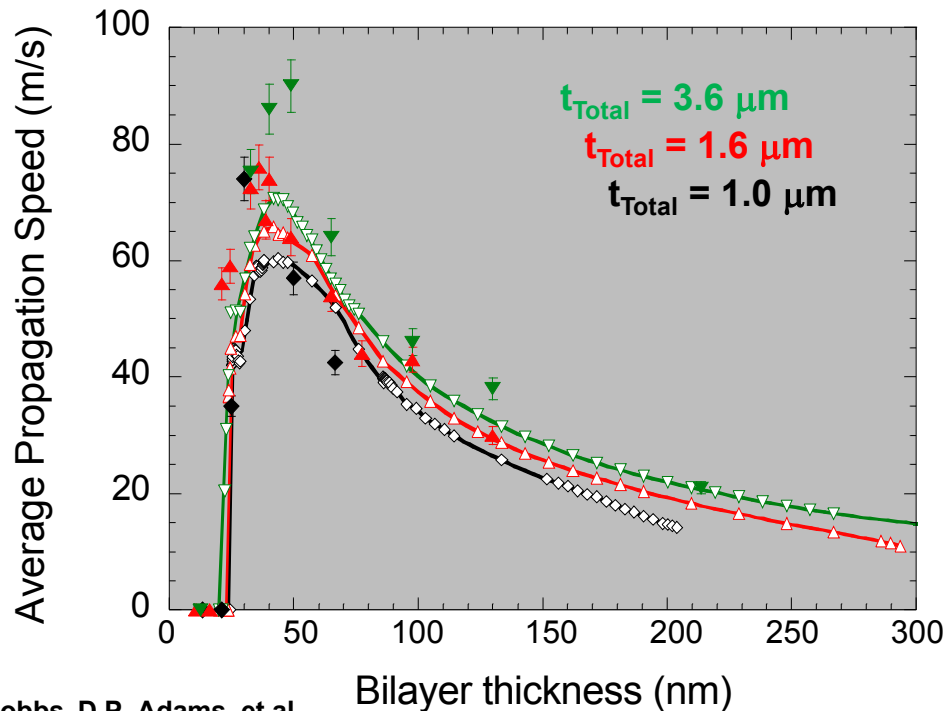
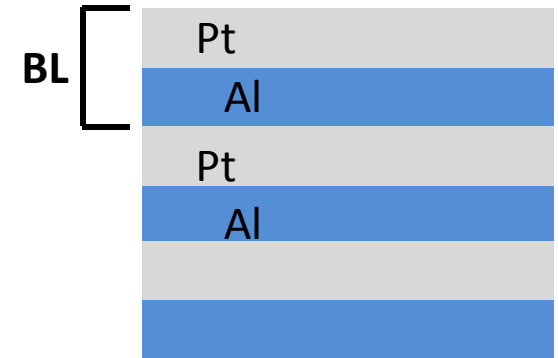


~ 600 microseconds after ignition



Bilayer Dependence

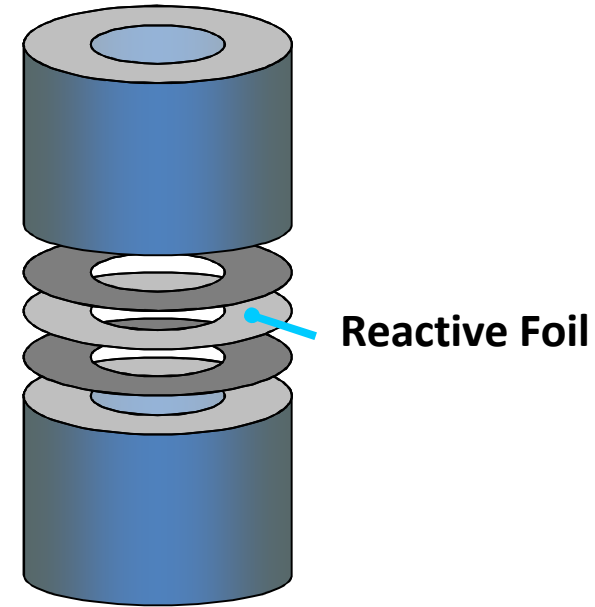
- Propagation speed increases with decreasing bilayer thickness.
- Shorter diffusion distances lead to shorter reaction times.
- Pre-mixing affects propagation speed of thinnest bilayers.



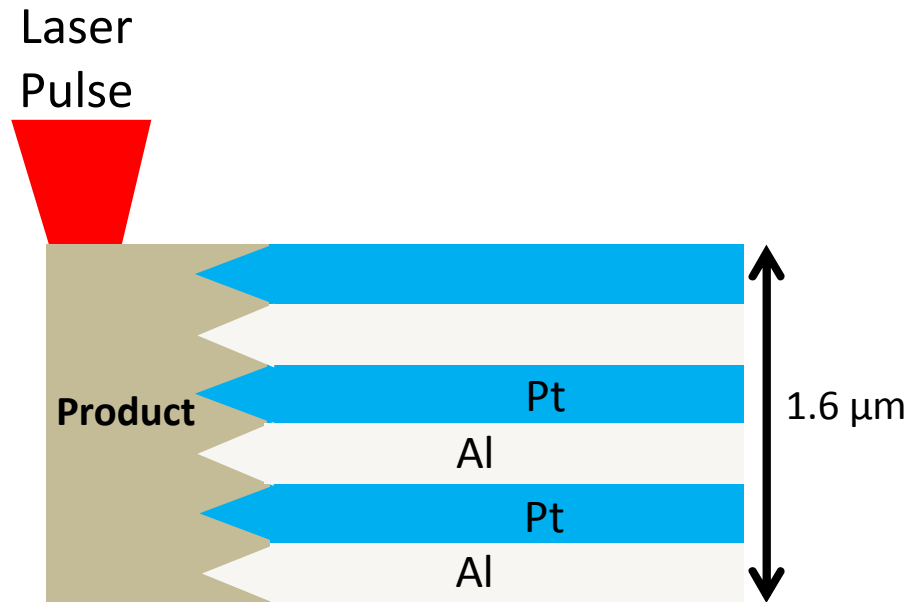
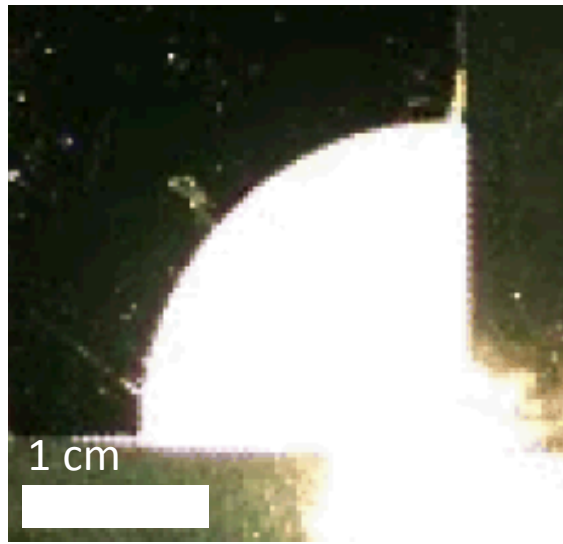
Laser Ignition

- Laser irradiation leads to more control over energy delivered to foil.
- Laser irradiation allows for remote ignition.
- Study effects of rate of heat input on ignition.
- Vary pulse length from femtosecond to millisecond to study effects of heating rate on ignition.

Joining

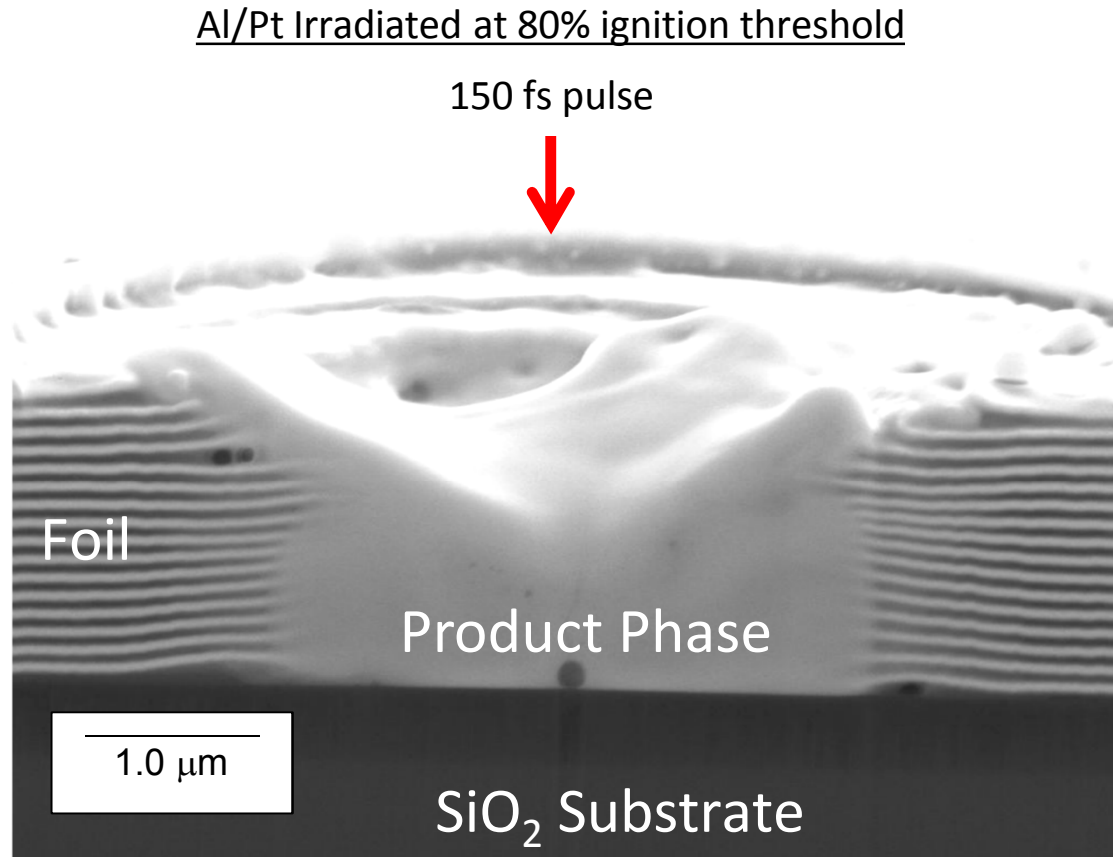


~ 600 microseconds after ignition



Laser Irradiation

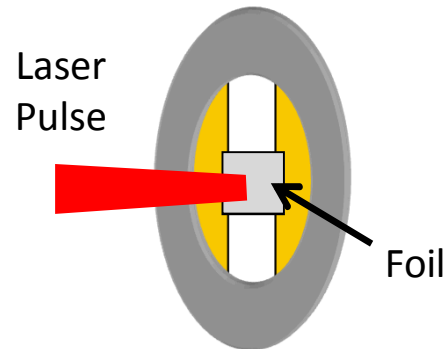
- Laser irradiation can modify foils without ignition.
- Pulse length dependent.



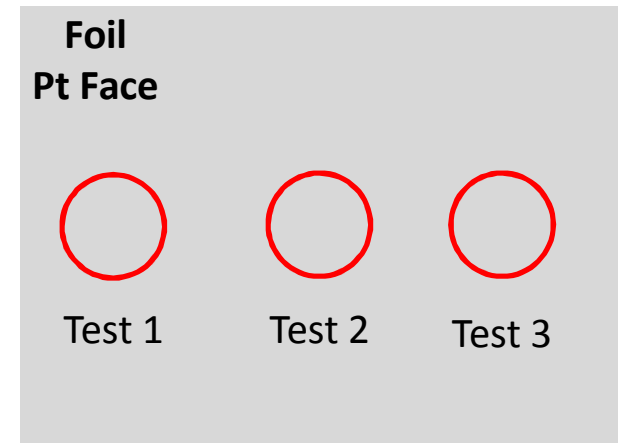
Picard, Yoosuf N. Ph.D. thesis University of Michigan, 2006.

Determining Laser Ignition Threshold

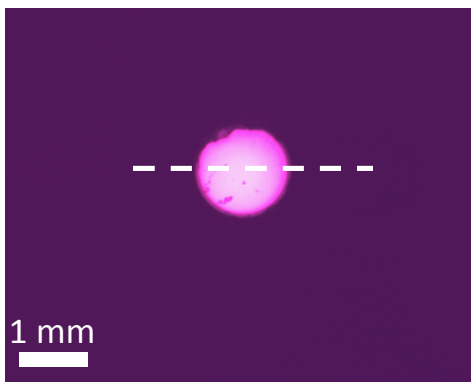
- Foil not on substrate
- Single Pulse Irradiation
- “Flat-top” Beam Profile
- Irradiate Pt side



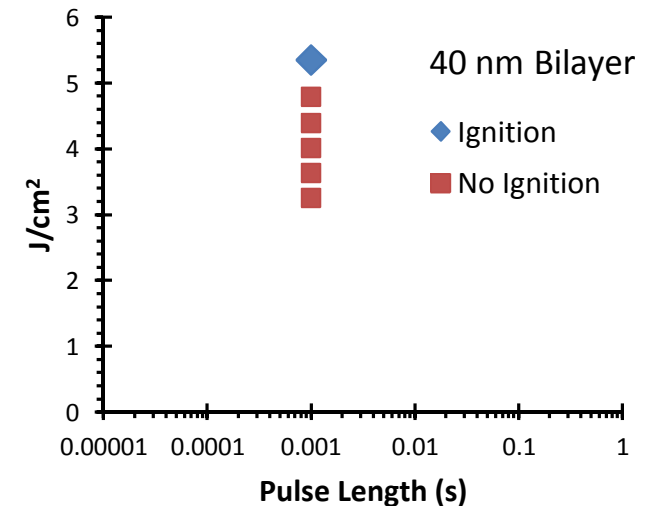
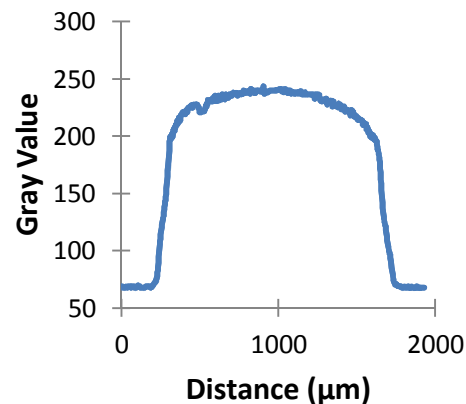
- Laser energy is increased until foil ignites.
- Non-irradiated region of sample is used for each test.



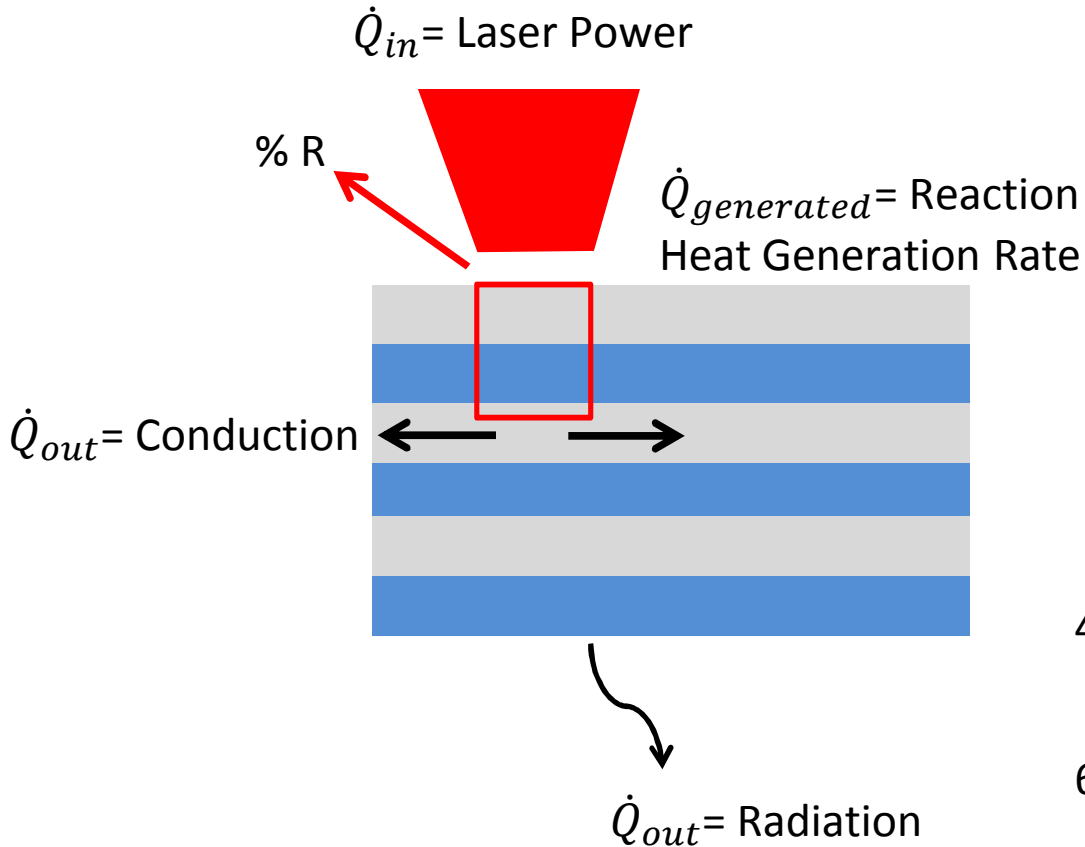
Focused Beam



“Flat-top”



Heat Flow and Interaction Volume

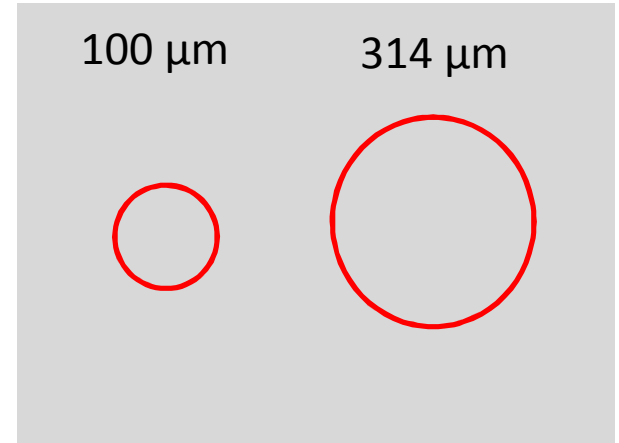


Change interaction volume

Laser Spot Size

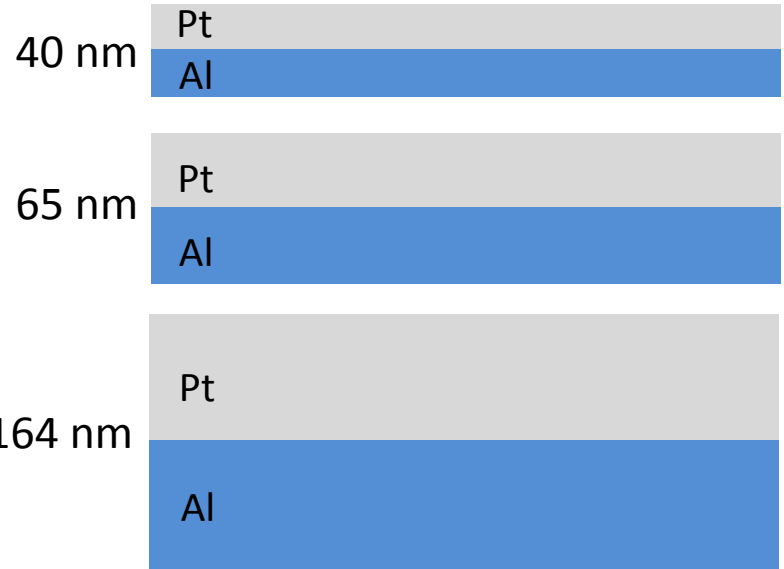
100 μm

314 μm



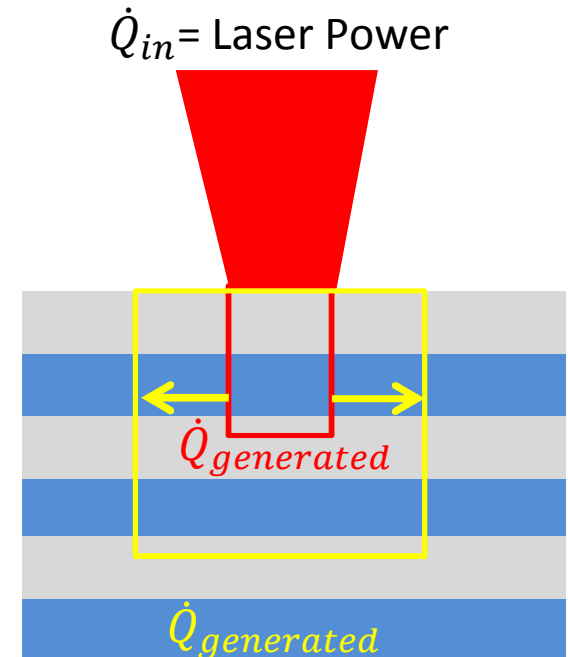
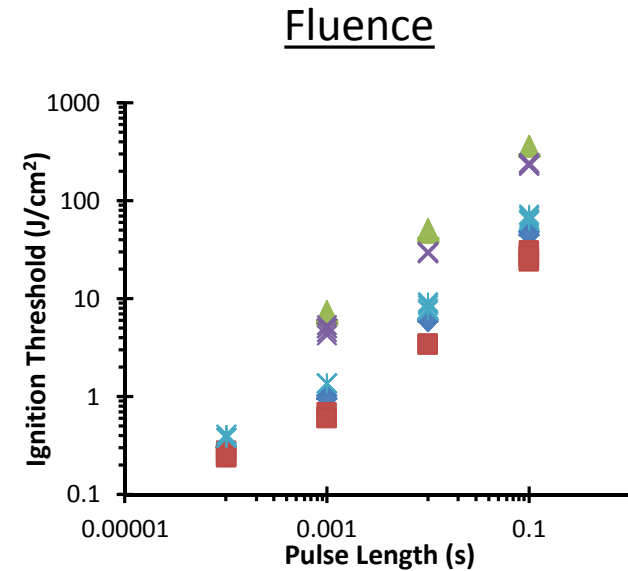
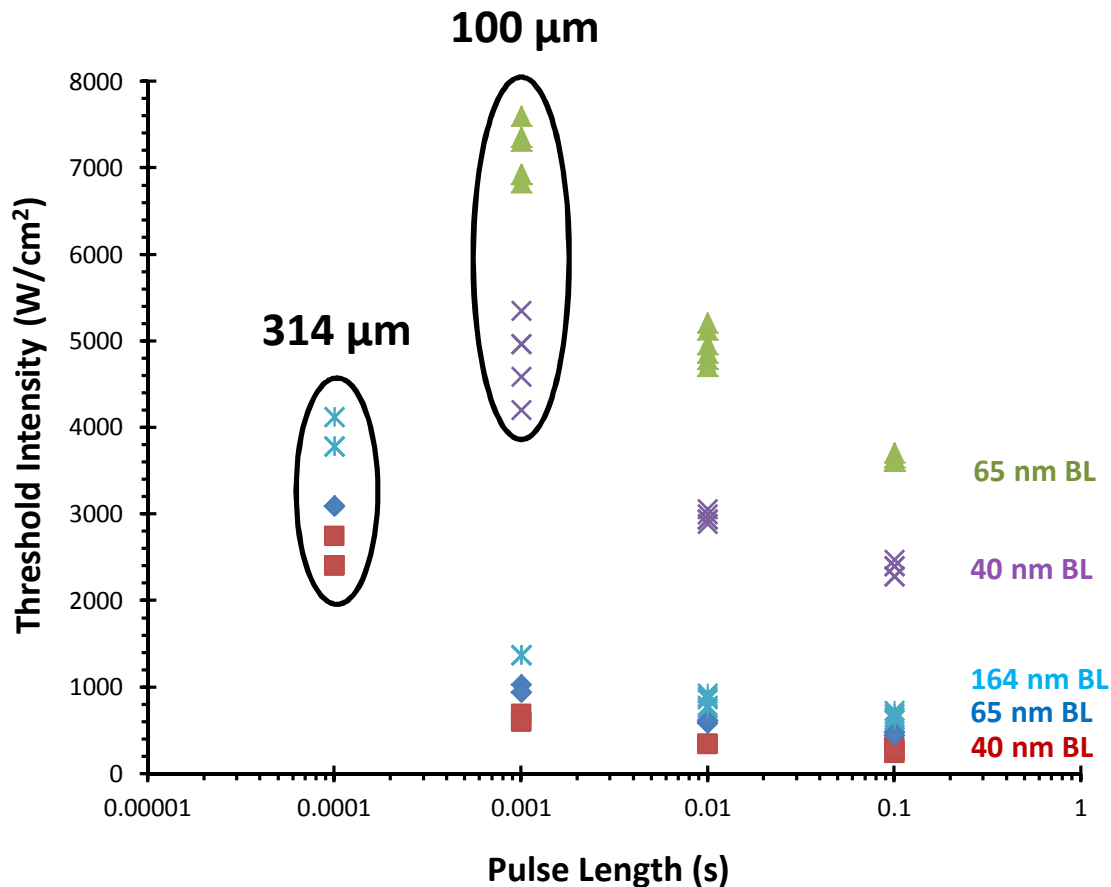
Total thickness = 1.6 μm

Bilayer Thickness



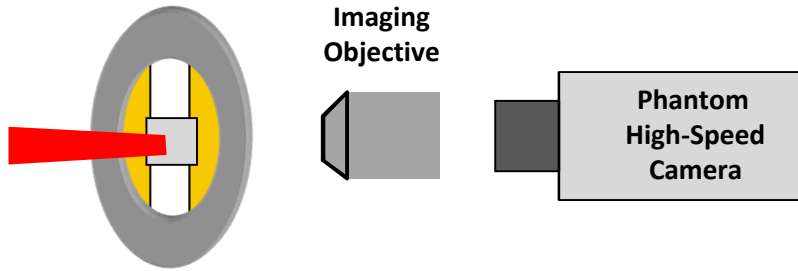
Intensity Threshold

- Intensity (W/cm^2) calculated using energy density and pulse length.
- Ignition threshold depends on intensity.
- Longer pulse lengths lower the intensity threshold.
- Longer pulse length may increase interaction volume via conduction.



Imaging Ignition

High-speed imaging of Al/Pt foil from the Al side

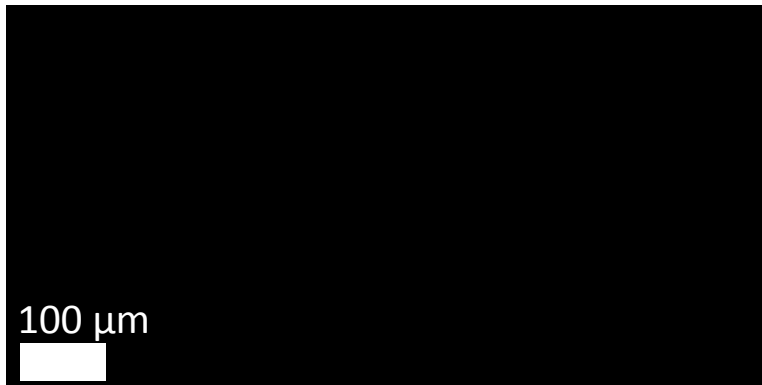


10 ms Incident Pulse

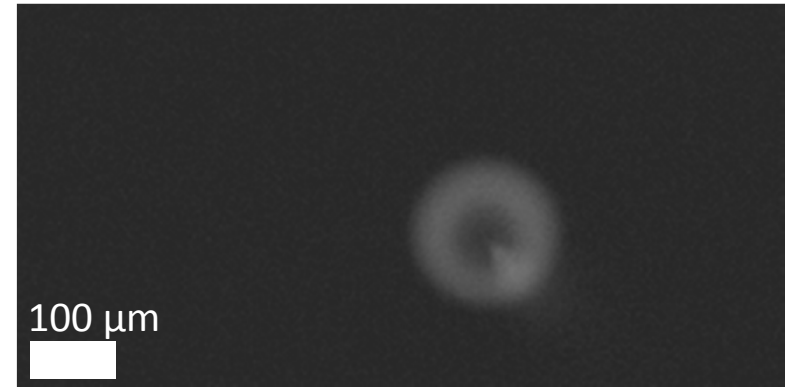
65 nm Bilayer

619 W/cm²

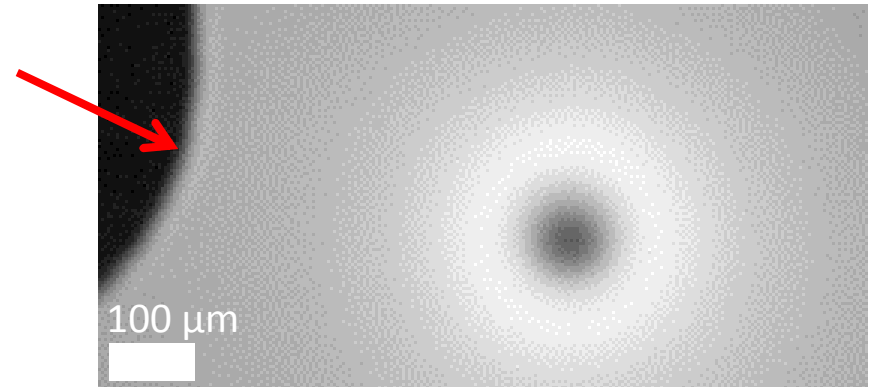
t = 0 ms Laser Turns On



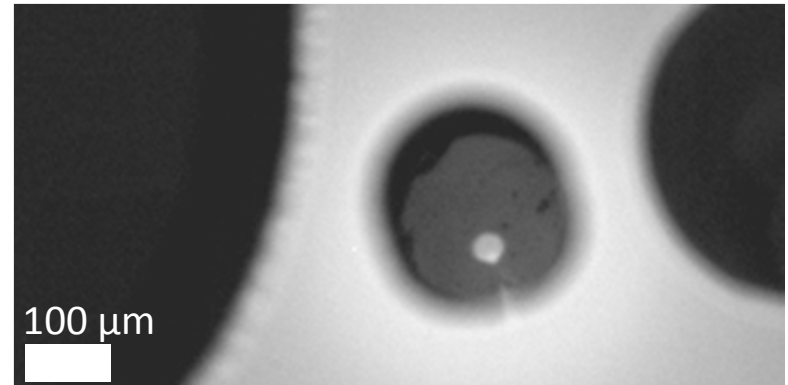
t = 2.88 ms Foil Ignites



t = 2.89 ms Entire Foil Melts

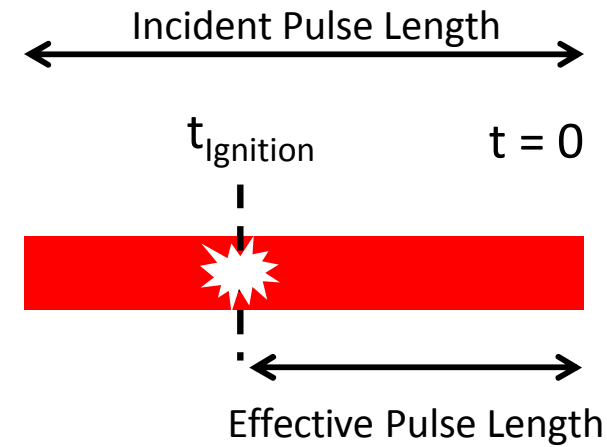


t = 2.91 ms Laser Penetrates Foil

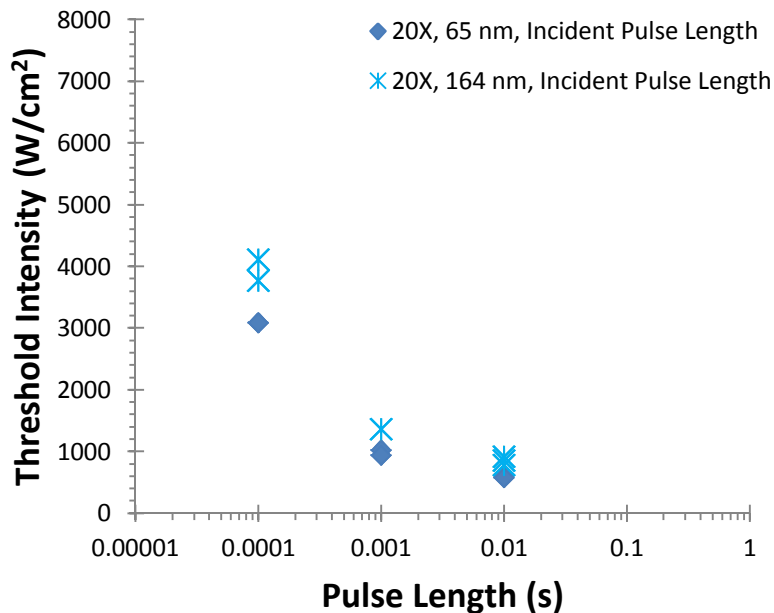


Laser-Foil Interaction Time

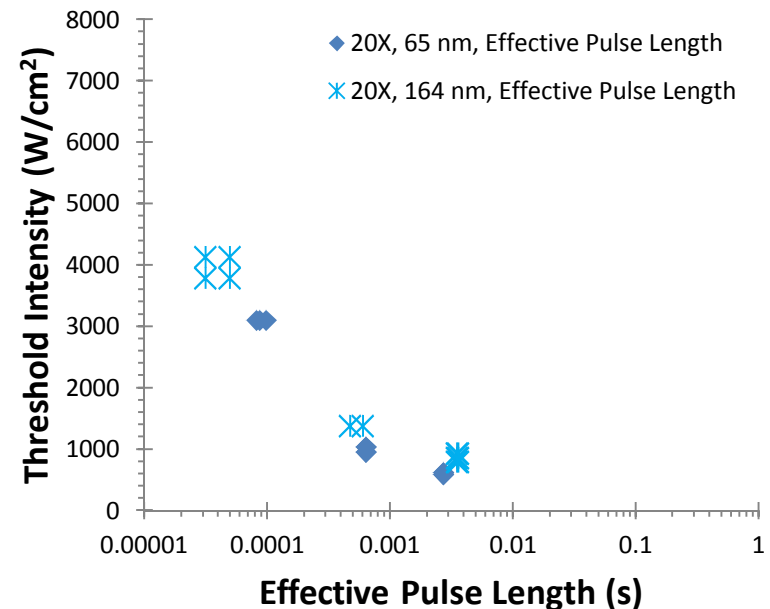
- Effective pulse length calculated by observing when ignition begins.
- Effective pulse length is usually shorter than incident pulse length.
- *Shorter pulses require energy to be delivered at a higher rate (power).*



Incident Pulse Length



Effective Pulse Length



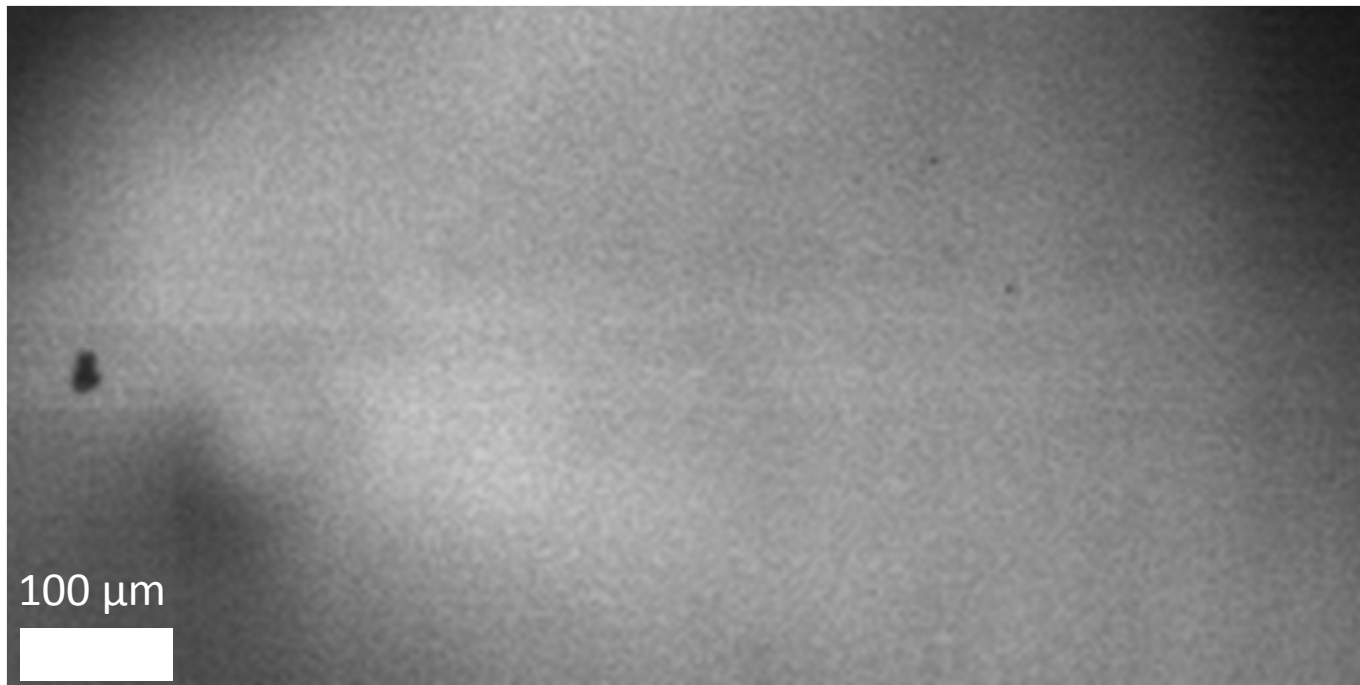
Sub-threshold Irradiation

95% Ignition Threshold

2.94 ms Incident Pulse

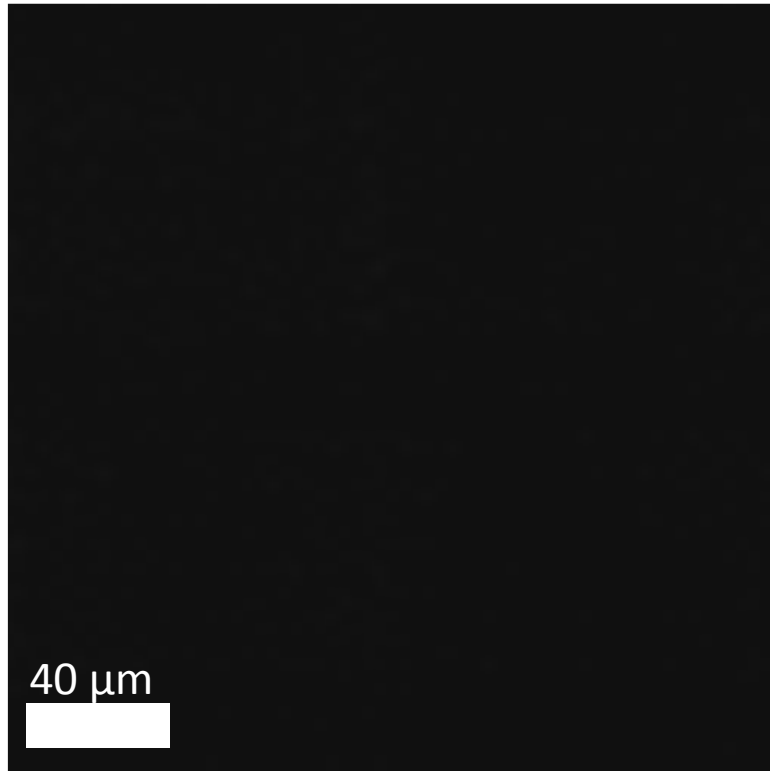
65 nm Bilayer

10 μs /frame

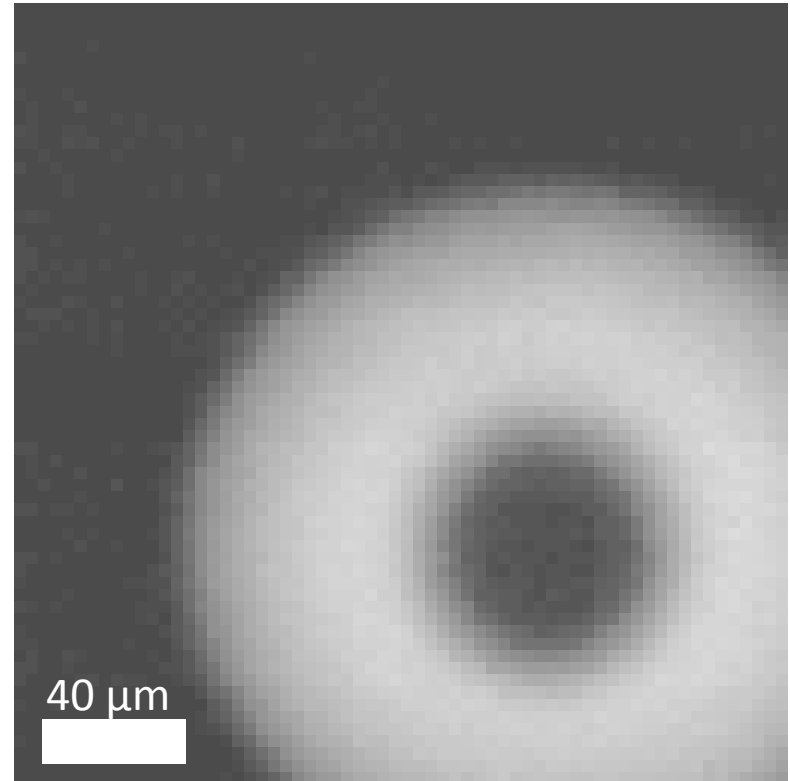


Self-Propagating Reaction after Ignition

10 ms Incident Pulse
164 nm Bilayer
894 W/cm² 4 μs/frame

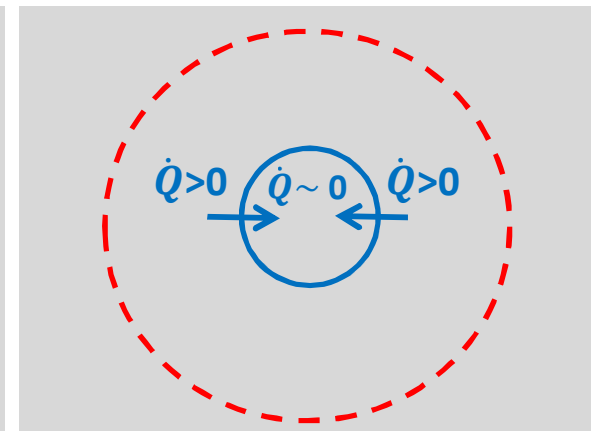
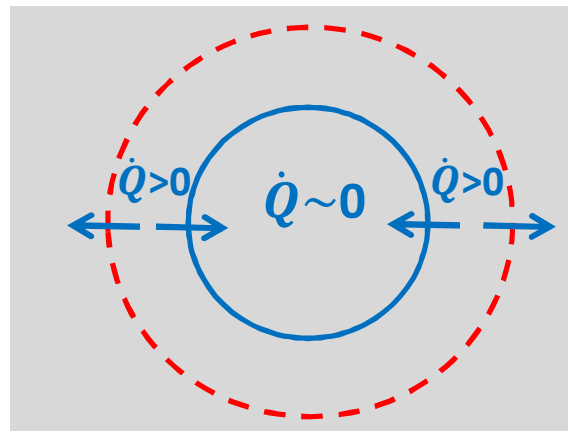
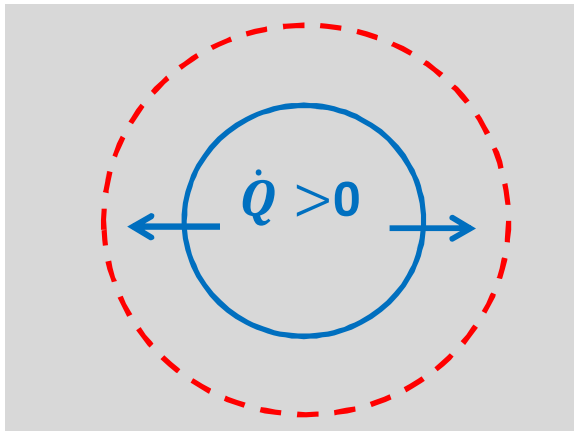
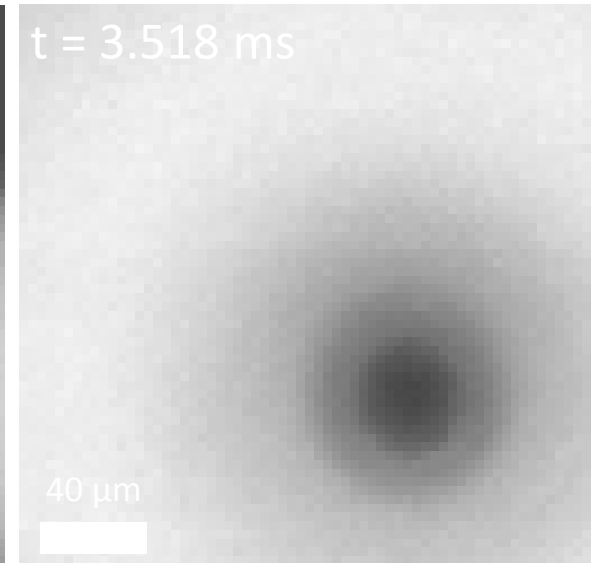
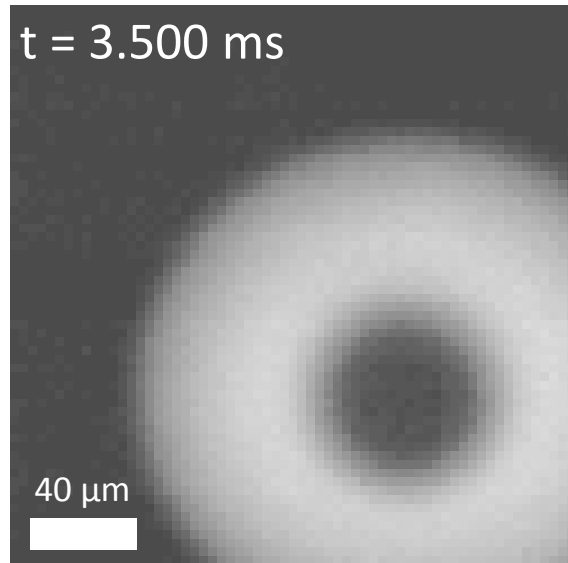
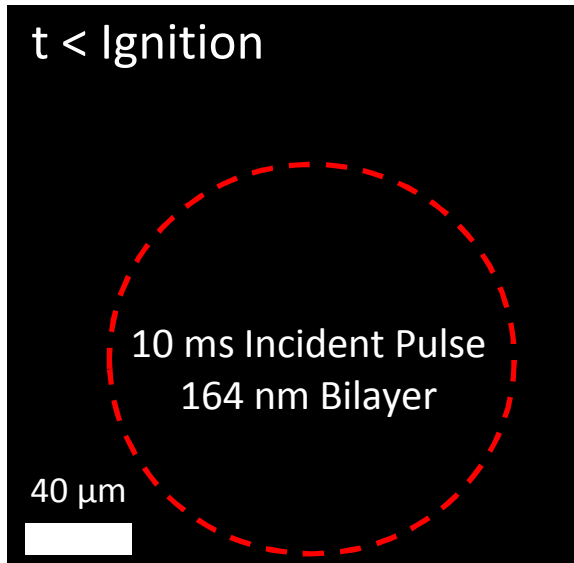


Ignition ~ 3.500 ms after laser turn on



Solid-State Reaction vs. Self-Propagating Reaction

- Dark center suggests ignition begins as a slow, non-propagating reaction.
- Bright annulus suggests these regions are hotter than surrounding regions.



Shorter Pulses – Mechanisms Change

100 μ s Pulse

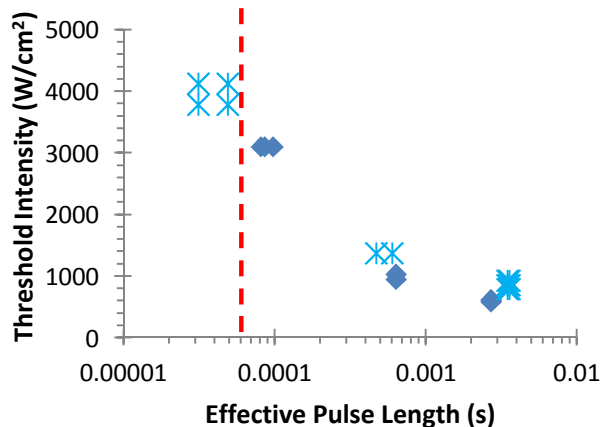
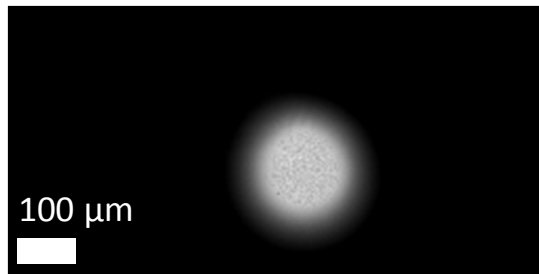
- Ignition occurs – no annulus observed.
- Central, irradiated area ignites.
- Mechanisms are *intensity* dependent

100 μ s Incident Pulse

164 nm Bilayer

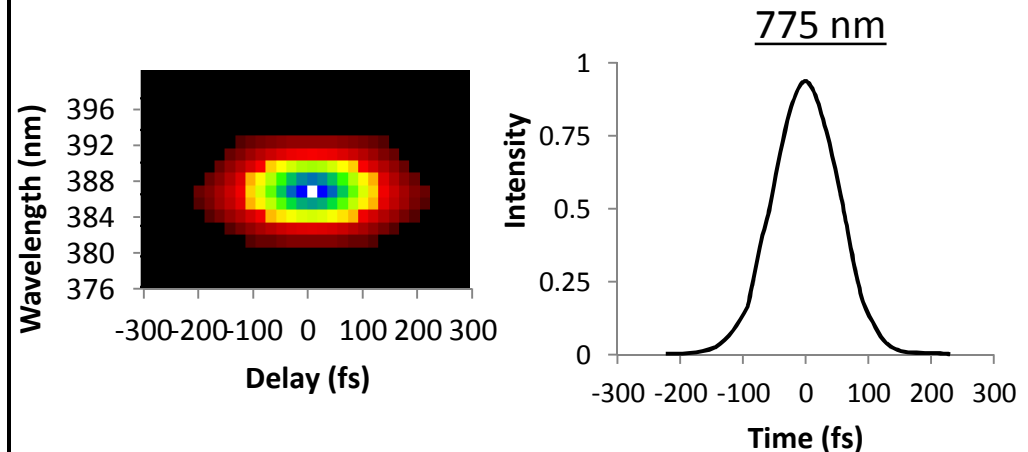
3.8 kW/cm²

t = 49 μ s

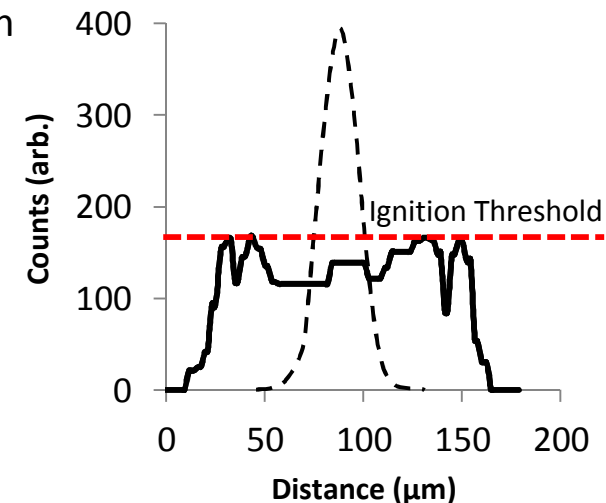
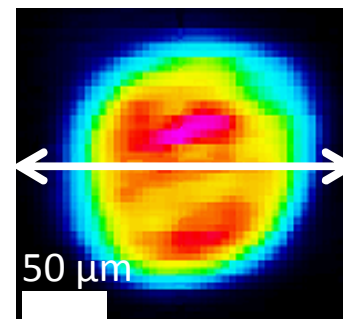


150 fs Pulse

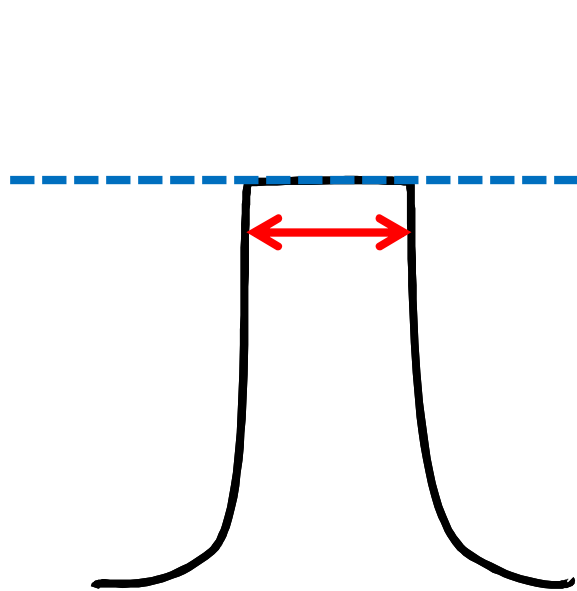
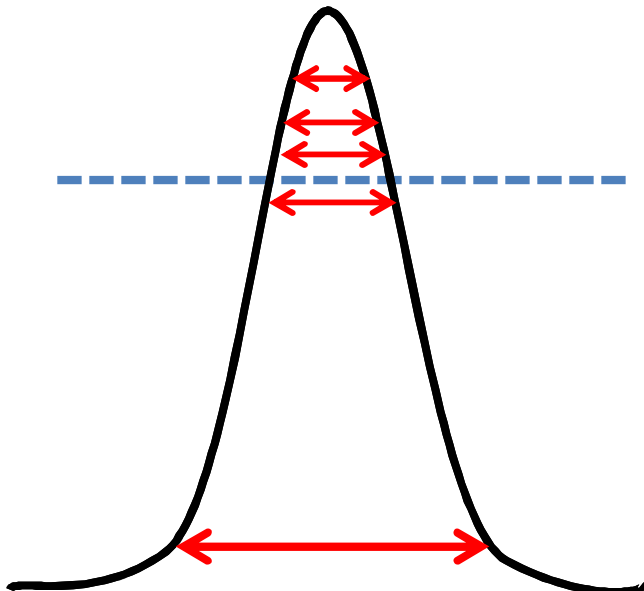
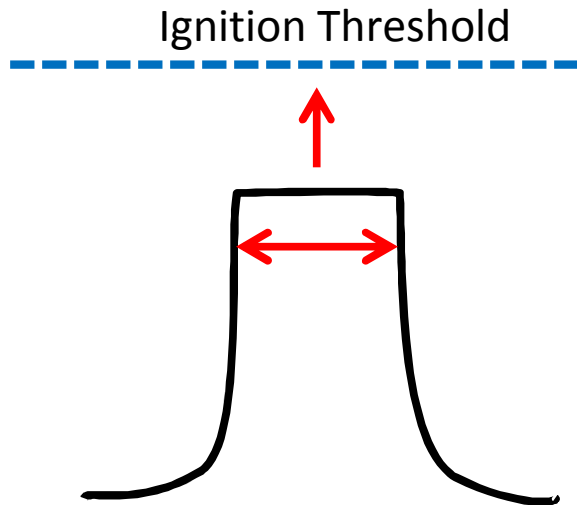
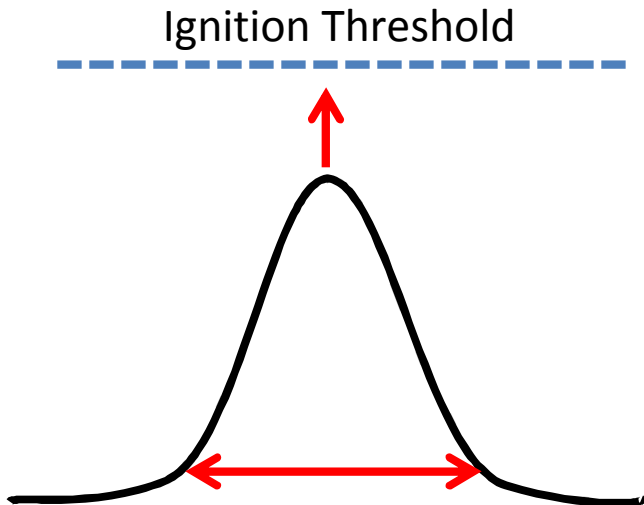
- Measured *temporal* distribution is Gaussian.
- “Flat-top” *spatial* distribution.



~ 30% Intensity Variation

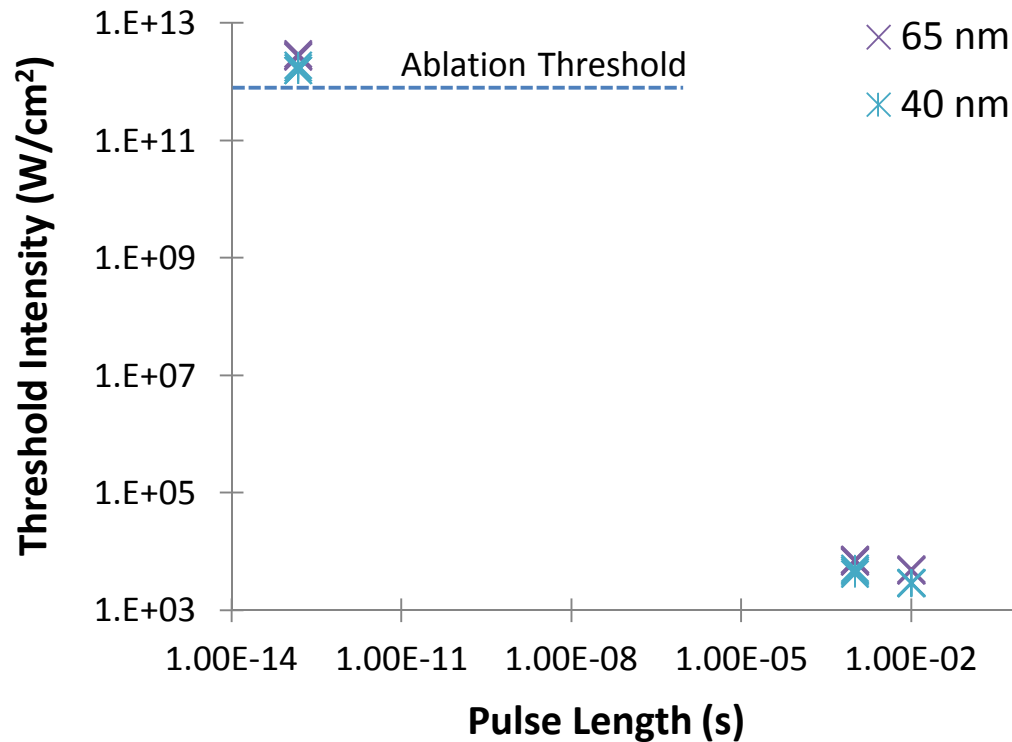


Fixed Interaction Volume



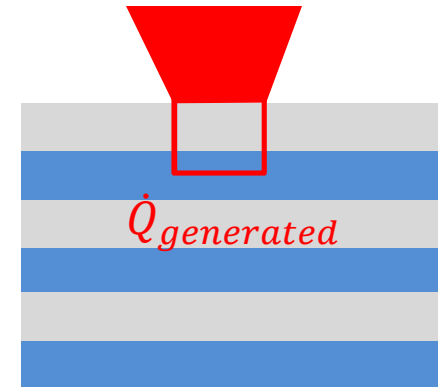
Ignition Threshold

- “Flat-top” beam is used for all pulse lengths – defines the interaction area
- 150 fs: mechanism changes from solid-state ignition to laser-induced melting and ablation.



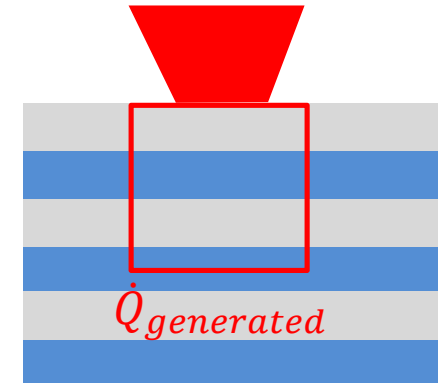
Shorter Pulses

$$\dot{Q}_{in} = \text{Laser Power}$$



Longer Pulses

$$\dot{Q}_{in} = \text{Laser Power}$$



Conclusions

- Material removal occurs at interfaces
- Void nucleation linked to surface roughness
- LIPSS formation is affected by surface features
- Diffraction and surface plasmons are responsible for LIPSS formation
- LIPSS begin forming on same time scales as material removal
- Laser pulse lengths ranging from femtoseconds to milliseconds can ignite foils.
- Laser ignition threshold depends on pulse duration, laser spot size, and foil bilayer thickness.
- Ignition mechanism depends on the laser pulse length.

Extra Slides

Pump-Probe Microscopy: Time Scales

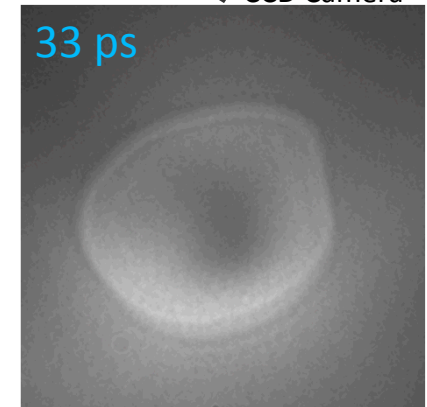
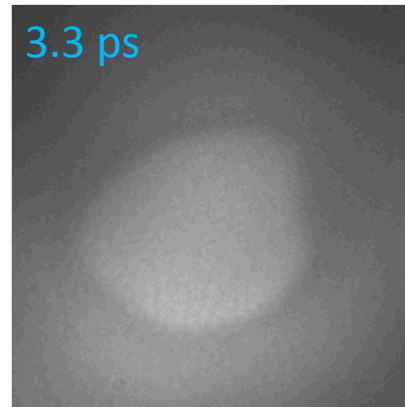
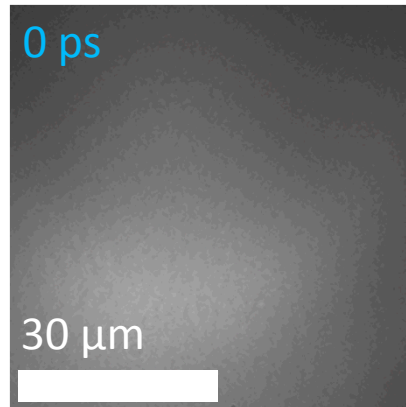
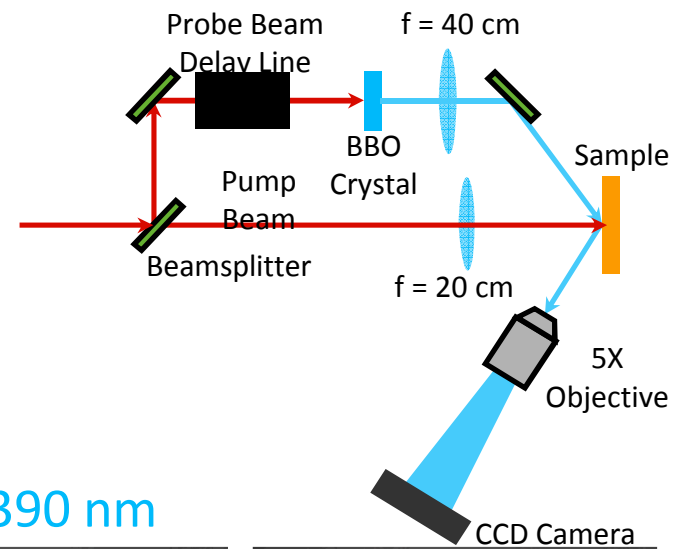
Pump Beam
 0.60 J/cm^2

Probe beam wavelength = 390 nm

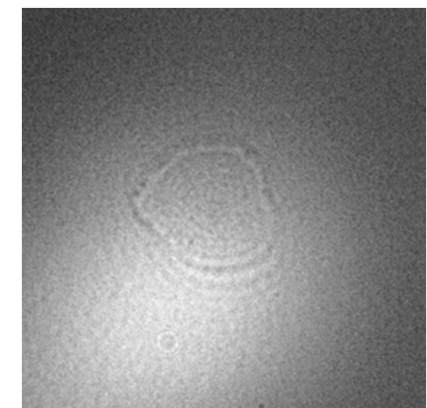
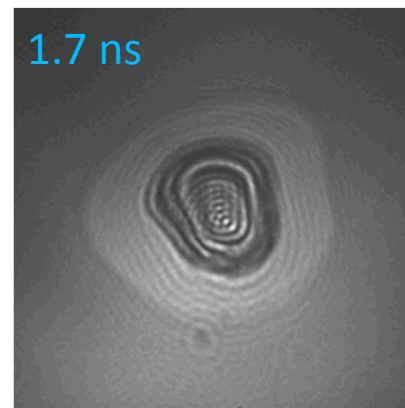
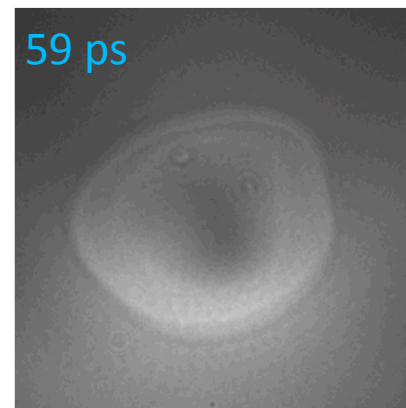
Silicon Substrate

Crater threshold
 0.32 J/cm^2

Melt threshold
 0.20 J/cm^2

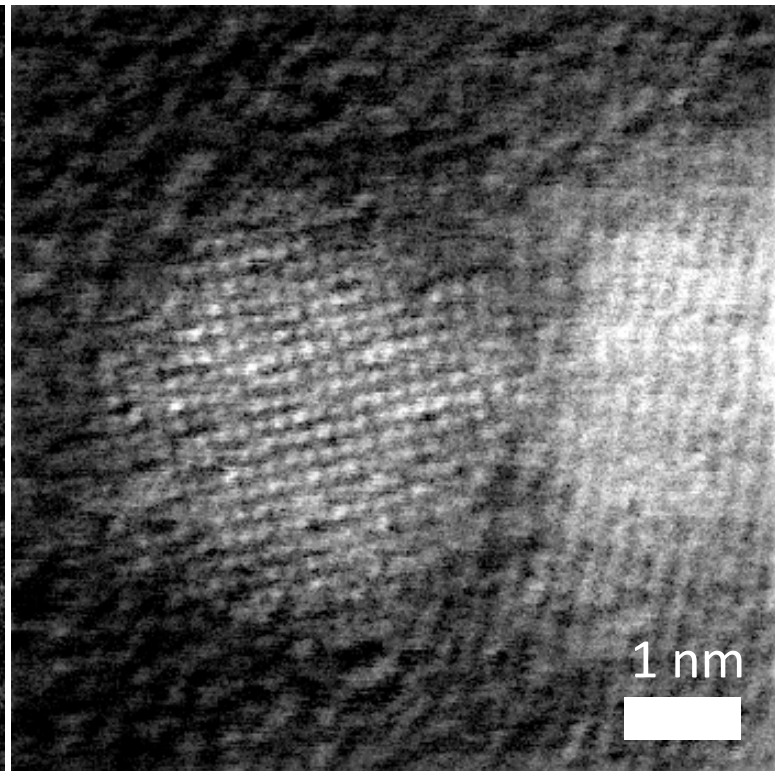
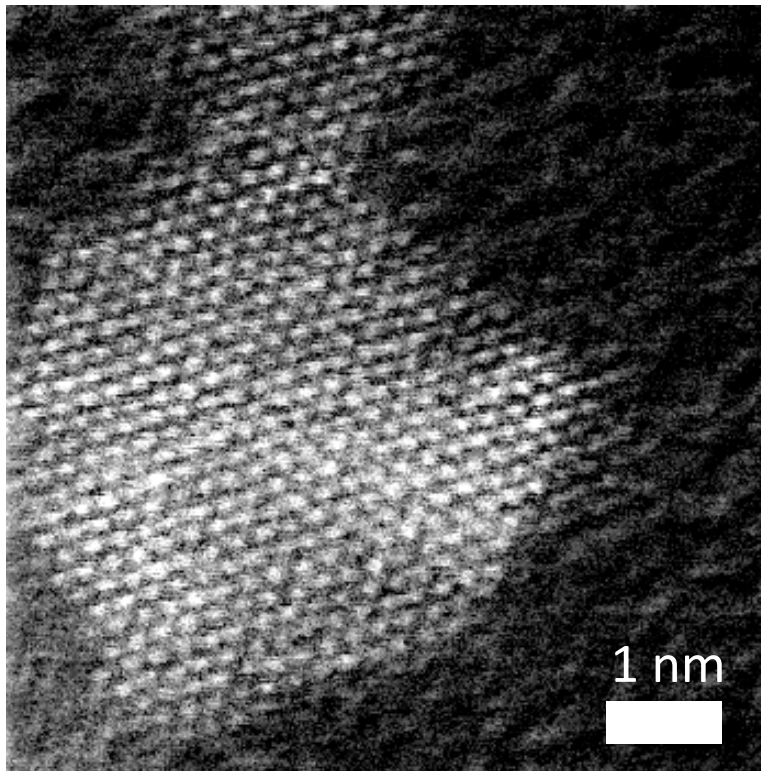


Resulting Crater

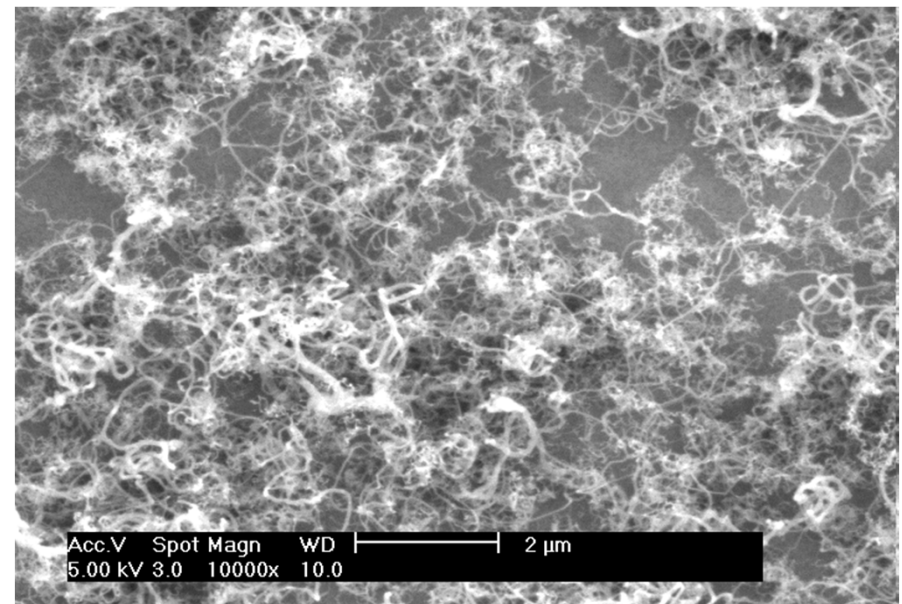
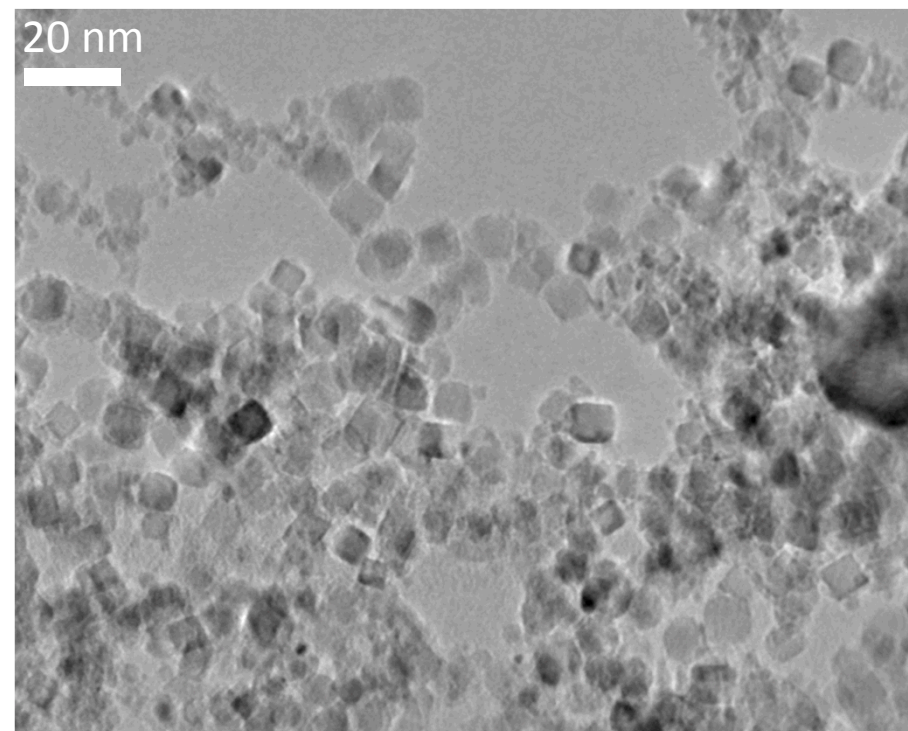
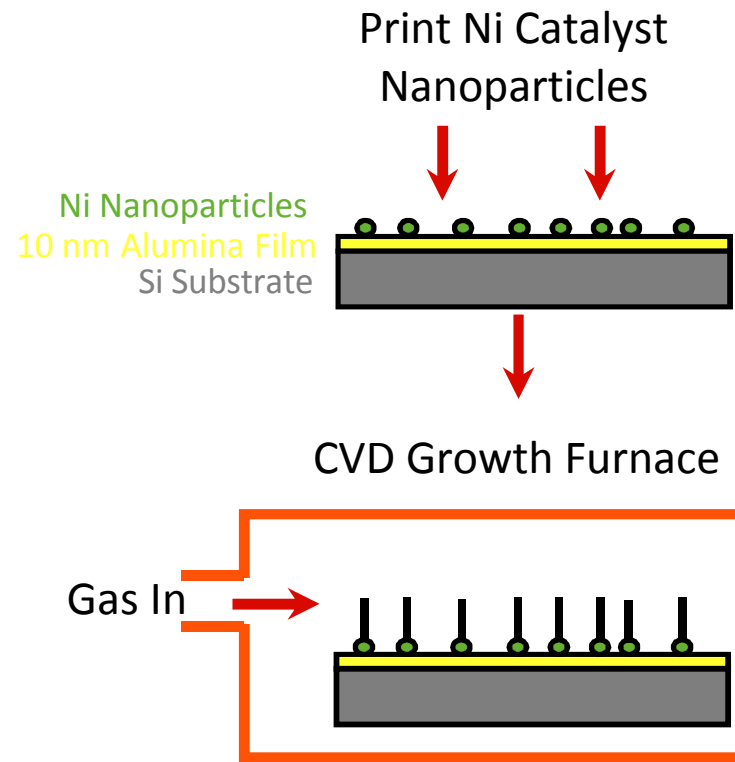
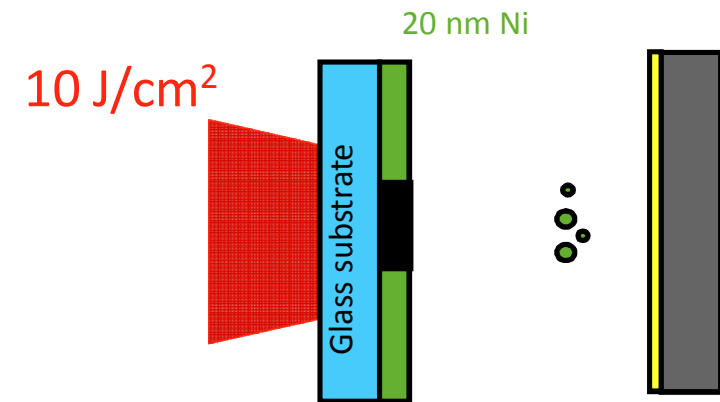


Faceted Nanoparticles

Atomic Resolution



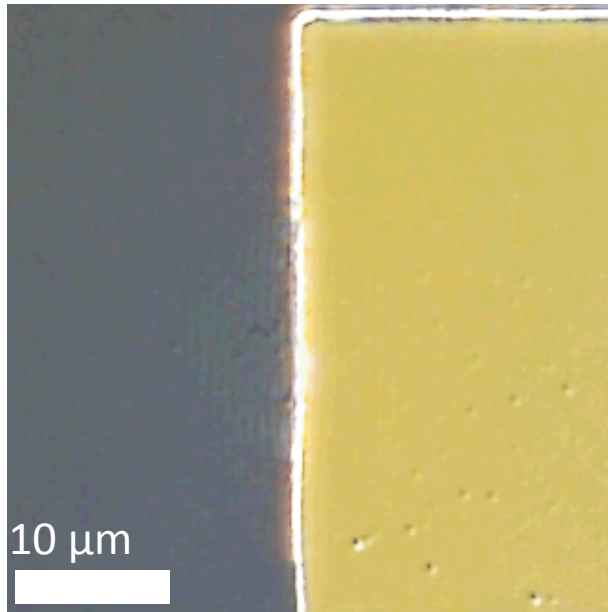
Ni Nanoparticle Printing



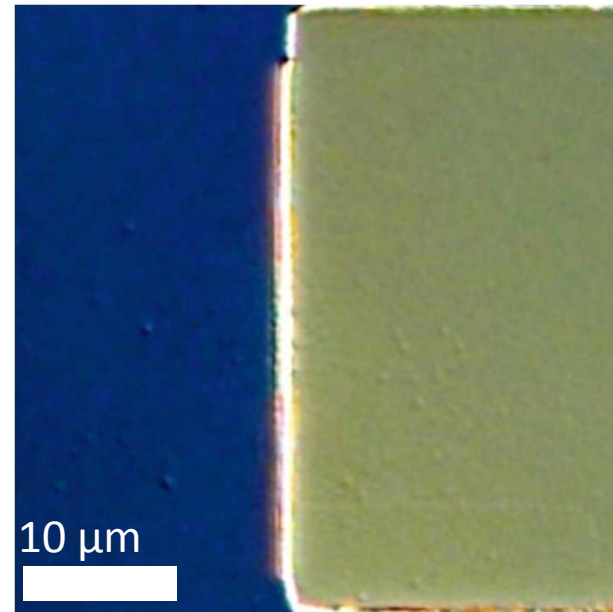
Amorphous Si Formation

0.20 J/cm²

Polarization



Polarization

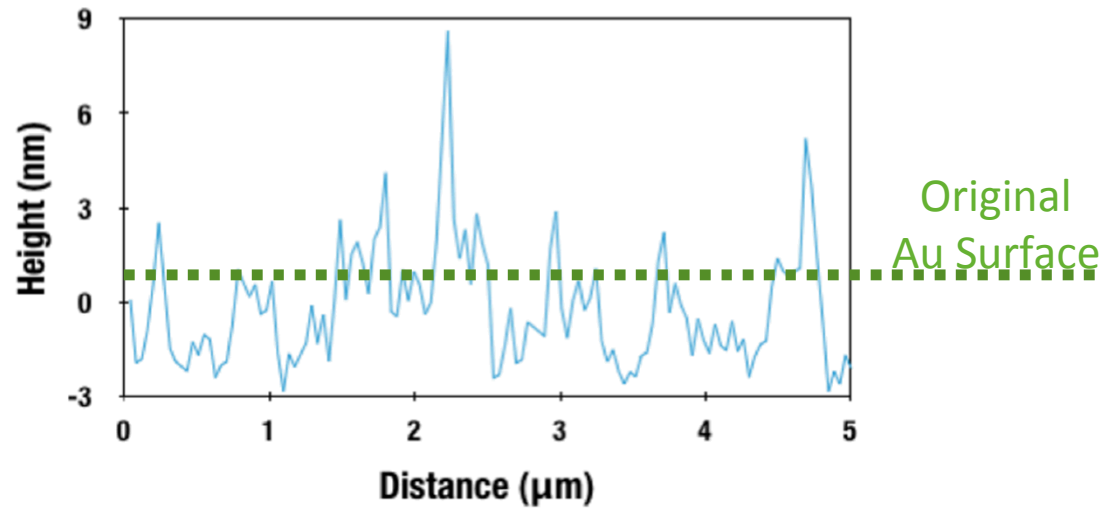
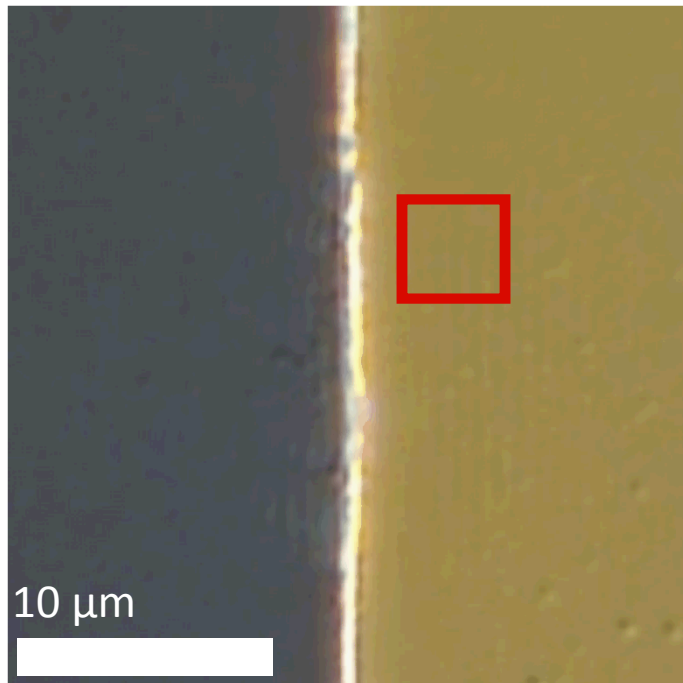
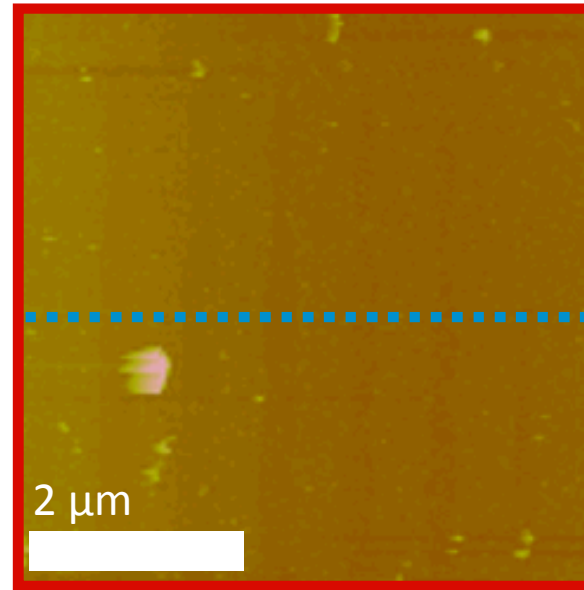


SPP Excitation

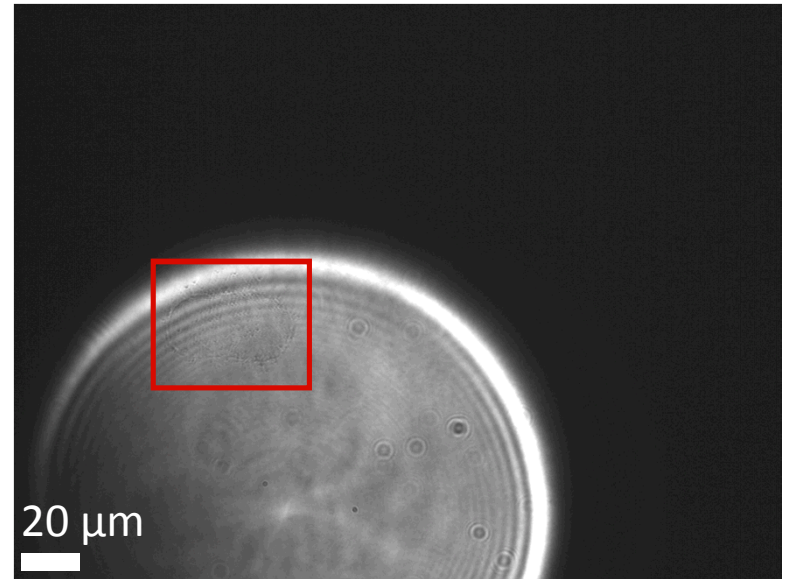
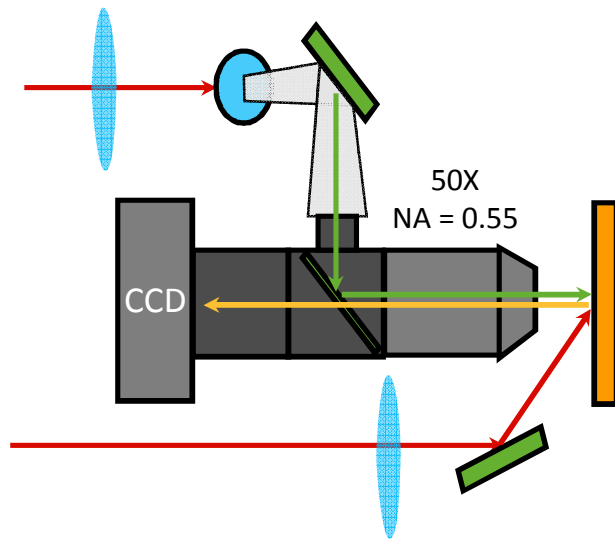
Au melt threshold $\sim 0.55 \text{ J/cm}^2$

0.20 J/cm^2

Polarization

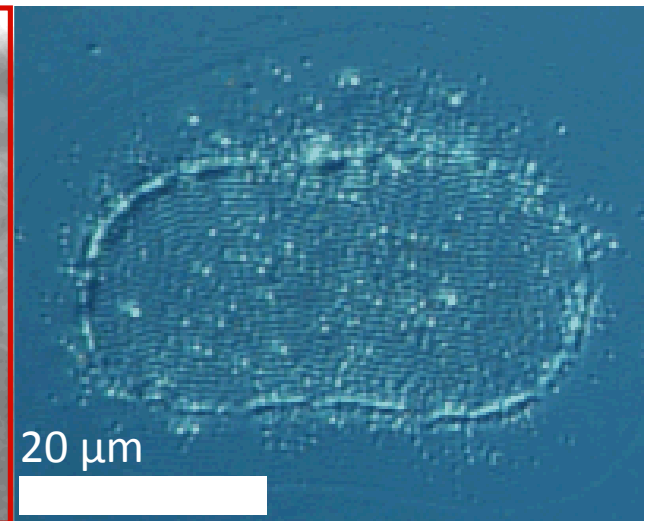
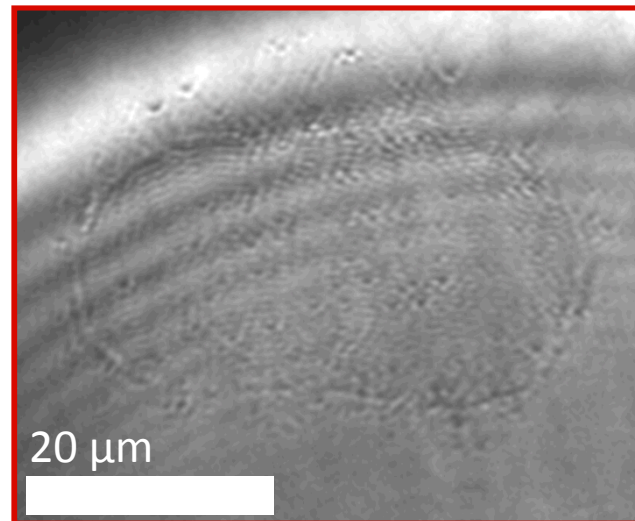


White Probe Light



Polarization

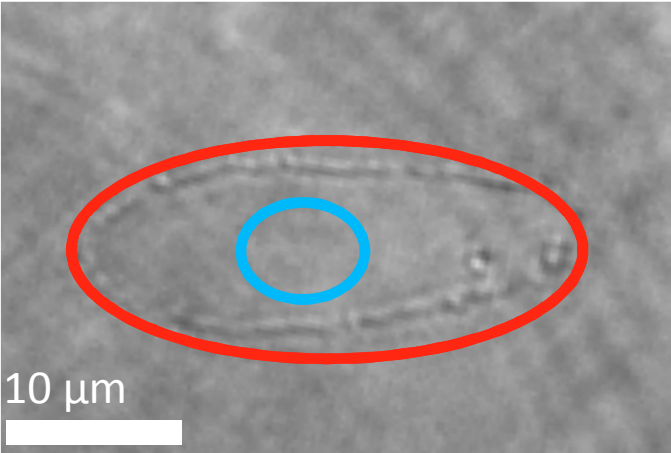
A red double-headed vertical arrow, indicating a relationship or comparison between the micrographs above and below it.



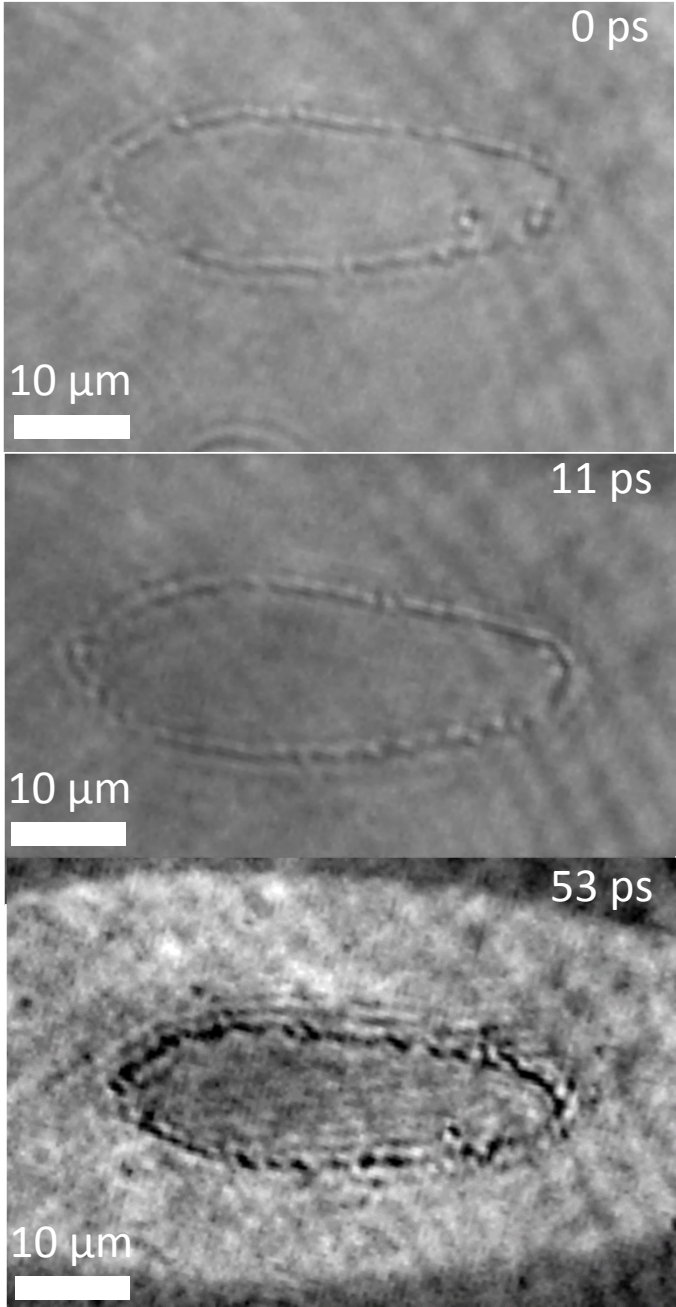
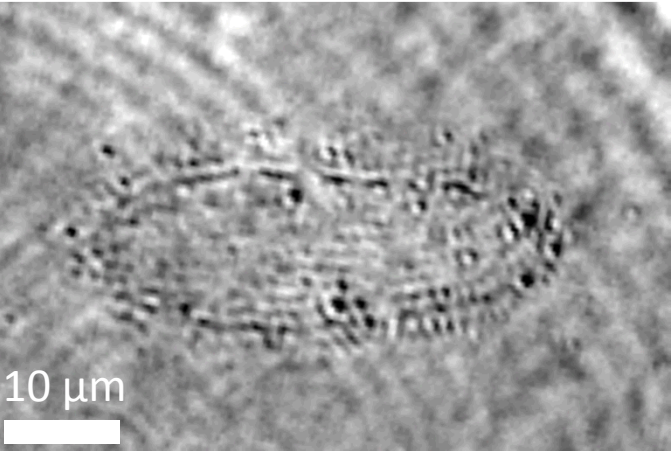
LIPSS Formation Dynamics

1st shot 2nd shot
0.45 J/cm² 0.32 J/cm²

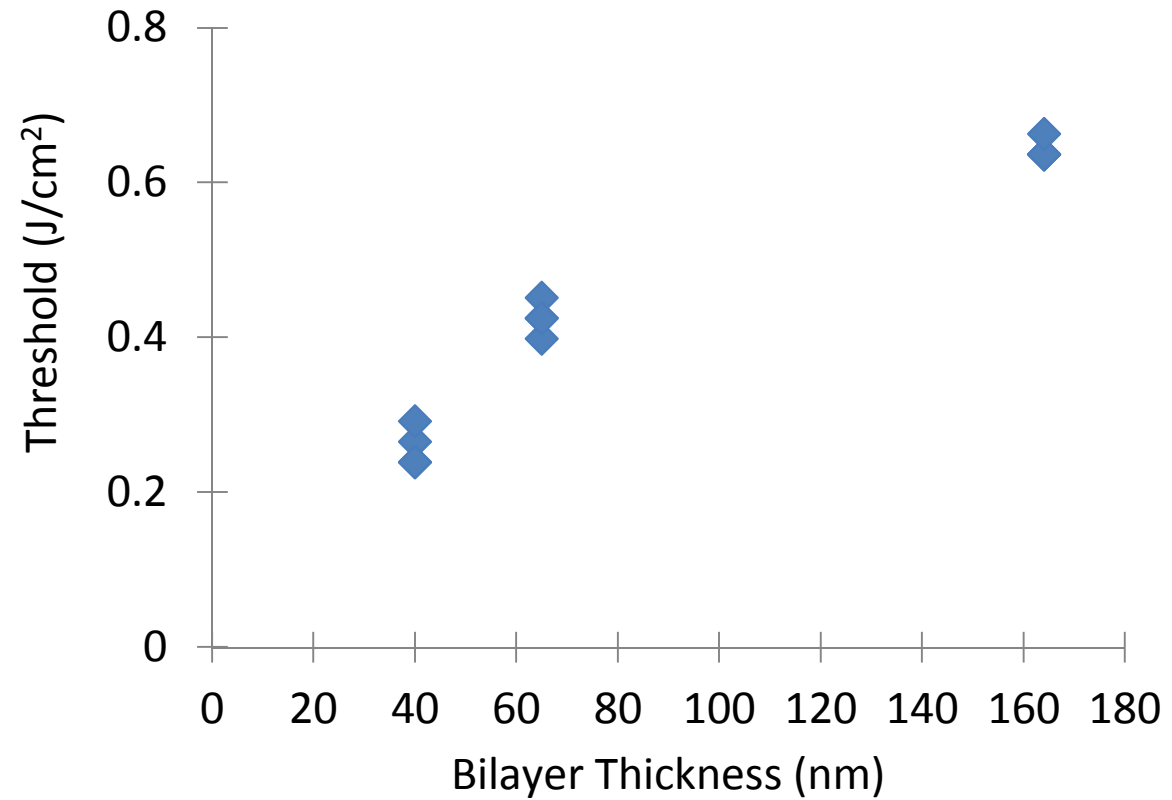
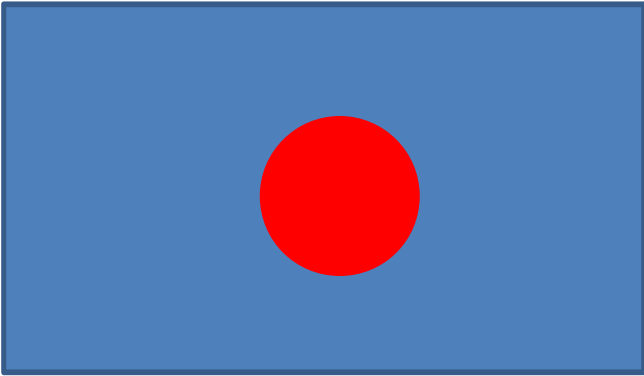
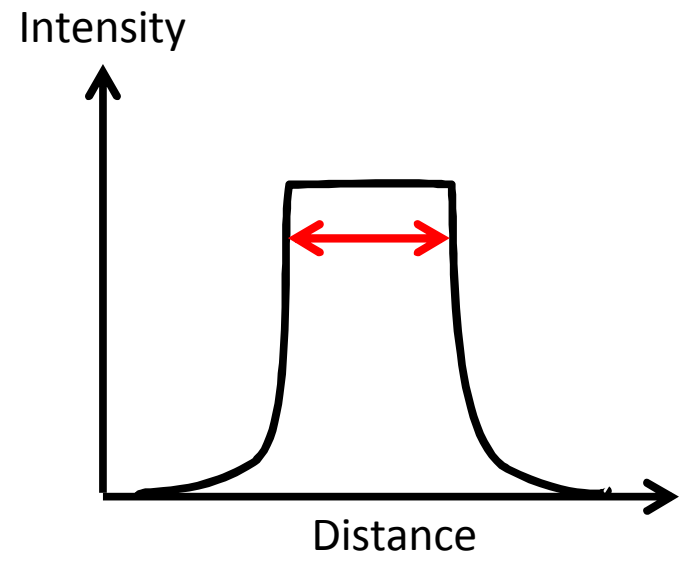
Polarization



Resulting LIPSS



150 fs Ignition Threshold



Effective Pulse Length Depends on Intensity

1 ms Incident Pulse
65 nm Bilayer

% Above Threshold	Effective Pulse Length (ms)
0	0.892
49	0.440
100	0.300

Diffusion Zone Size

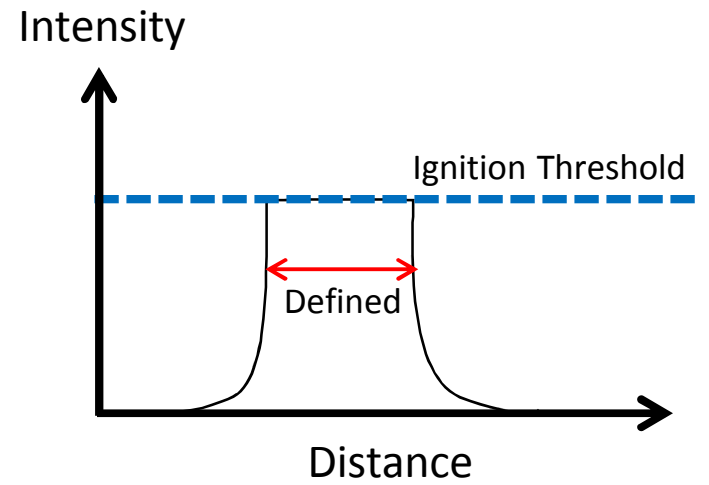
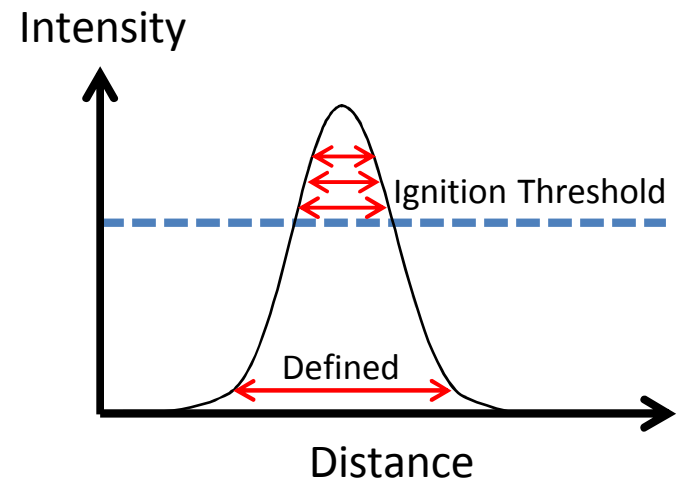
Depends on Pulse Length

65 nm Bilayer

Incident Pulse Length (ms)	Diffusion Zone Diameter (μm)
0.1, 9 mW	18
0.1, 9 mW	18
1, 9 mW	42
1, 4.5 mW	44
1, 5.5 mW	28
1, 5.5 mW	22
10, 36 mW	73
10, 36 mW	36
10, 36 mW	44

164 nm Bilayer

Incident Pulse Length (ms)	Diffusion Zone Diameter (μm)
0.1, 12 mW	0
0.1, 12 mW	0
1, 8 mW	18
1, 8 mW	22
10, 52 mW	49
10, 52 mW	52



Ignition and Melting

10 ms Incident Pulse

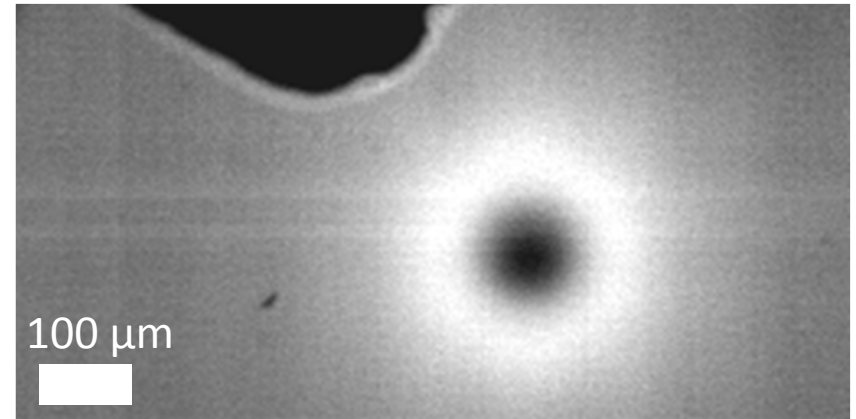
65 nm Bilayer

36 mW

~ 2.60 ms after laser turns on



~ 2.61 ms after laser turns on



Reflectance Measurements

10 ms Incident Pulse
65 nm Bilayer

