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Pulsed laser irradiation of reactive metal multilayers

D.P. Adams
Sandia National Laboratories
Albuquerque, NM

Sandia collaborators:
J.P. McDonald†, E. Jones, Jr.

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† Currently at Dow Corning, Midland, MI

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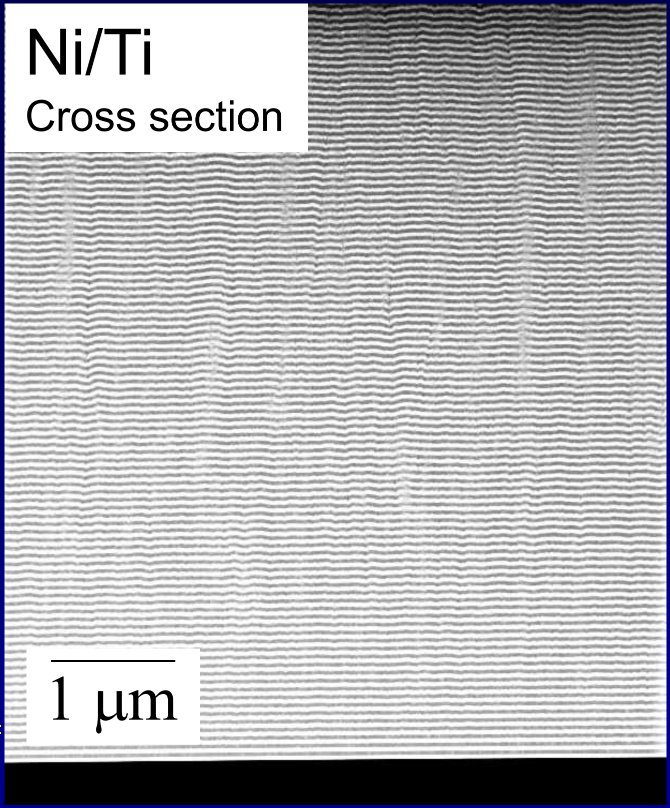
What are reactive multilayers?

- Vapor-deposited heterostructures consisting of two or more reactants that, when mixed, generate heat

  ✓ $M_1 + M_2 \Rightarrow M_1M_2$

  $M_1 + M_2O \Rightarrow M_1O + M_2$

- 10’s to 1000’s of individual layers
- Typical design employs single periodicity
- Total thickness 0.15 - 150 μm
- Ignited at a point
- Self-propagating high temperature reactions (deflagration)



Bilayer

ΔH_o (Ni-Ti) = - 34 kJ/mol atoms

Vapor deposited reactive multilayers were first reported by J. Floro J. Vac. Sci. Tech. A (1986).

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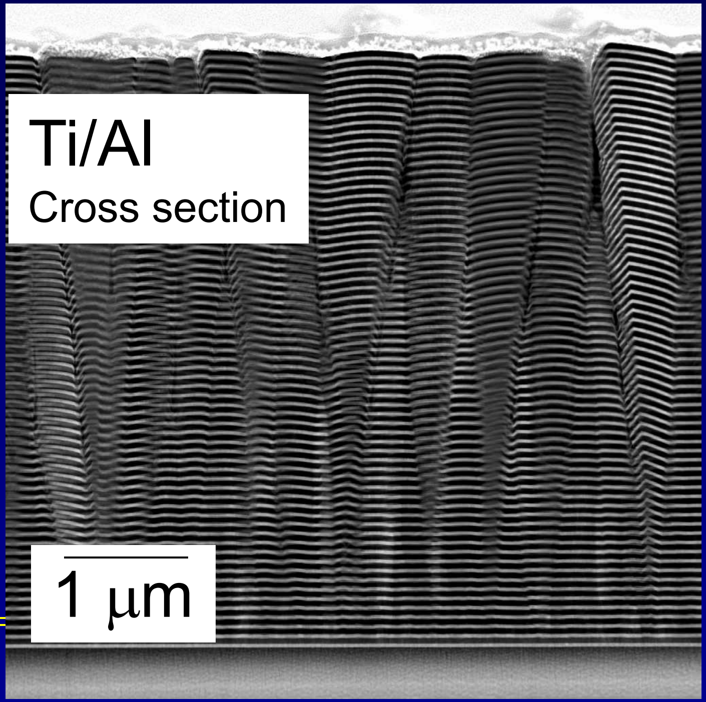

ΔH_o (Ti-Al) = - 38 kJ/mol atoms

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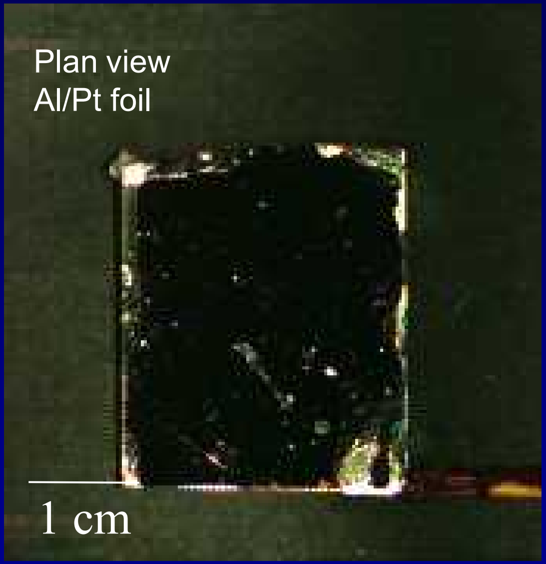

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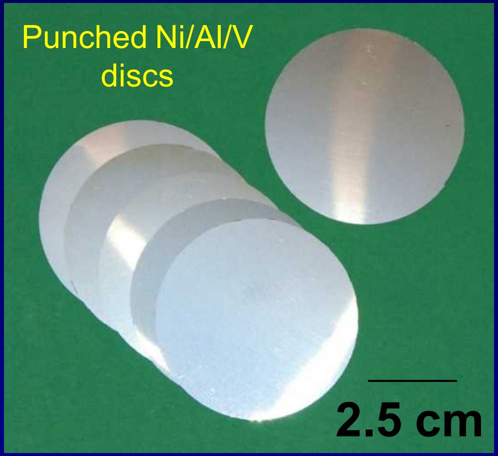
Reactive metal-metal multilayers have found use for advanced joining technology (soldering).

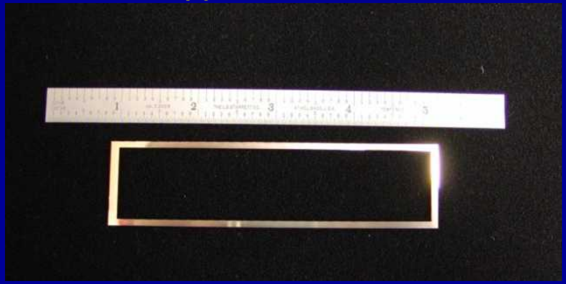
Punching

- Manual (1 disc/ 5 min.)
- > 90% success rate

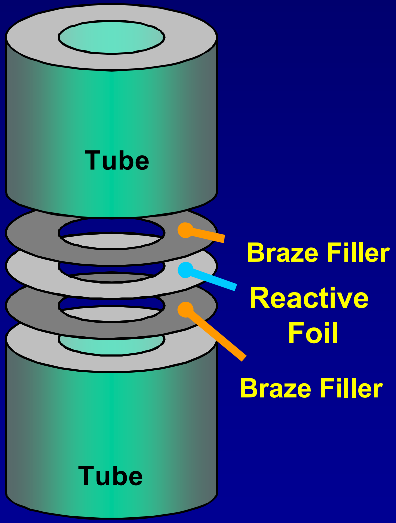


Serial laser cutting

-Ablation (1 disc/ 30 min.)
- > 99% success rate

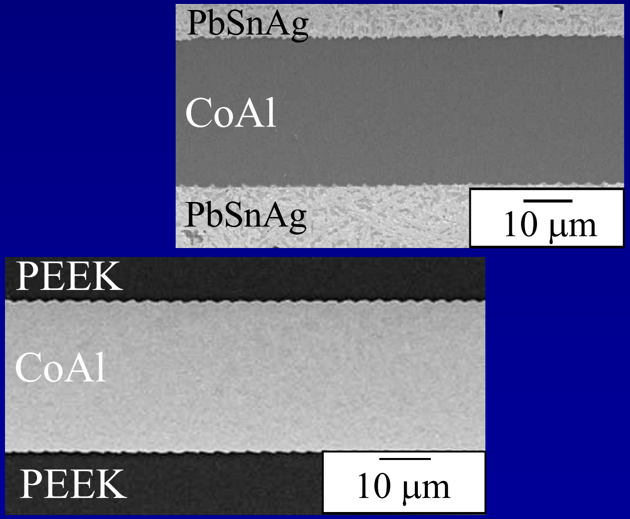


Fixturing and reaction



Assembly

Cross sections of soldered interfaces using exothermic foil as heat source.



Tasks of this research

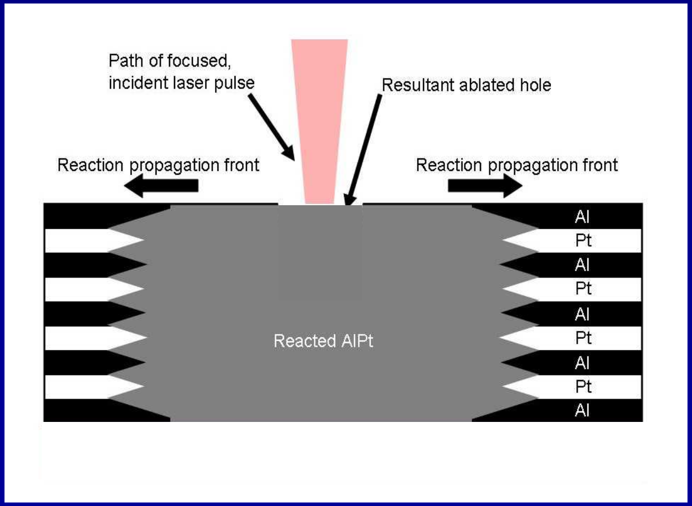
- Determine whether various reactive multilayers can be ignited at a point using pulsed lasers and determine the typical pulse energies required.

- Examine how ignition threshold is affected by

  multilayer design

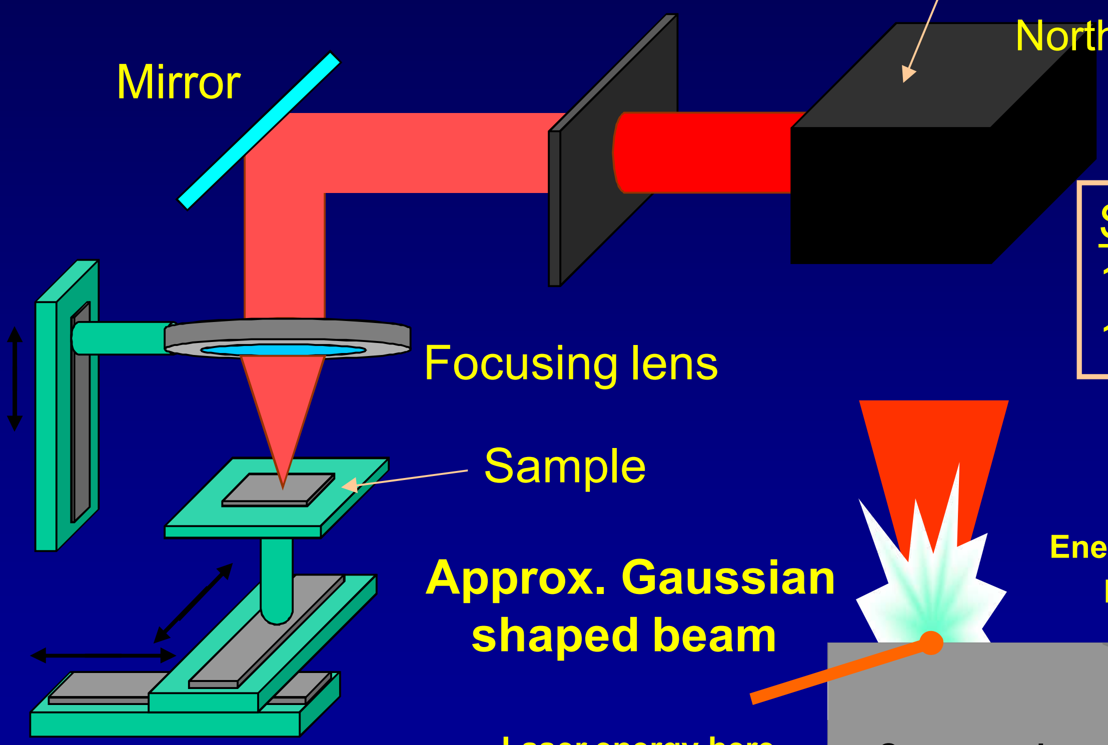
  pulse duration

Focus on a single material system: Al/Pt



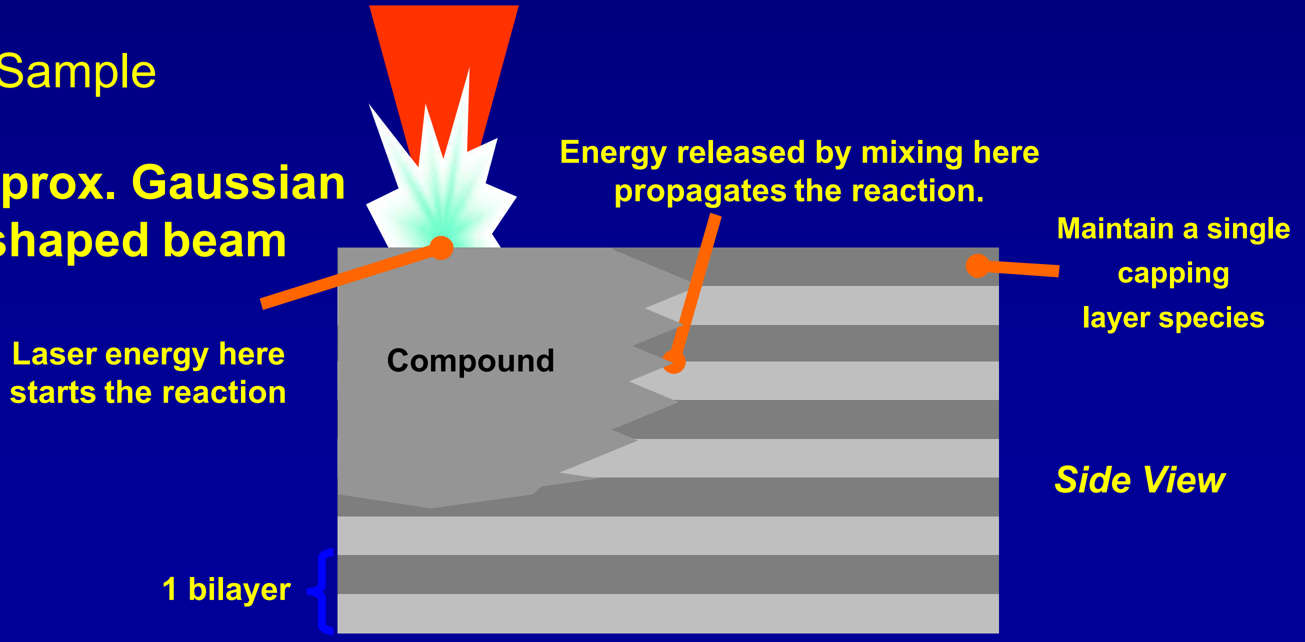
Experimental Schematic: Quantifying point ignition thresholds

<u>Laser systems:</u>
Spectra-Physics Hurricane - Ti:sapphire
Lambda Physik LPX-300 – KrF Excimer
Northrump Grumman – Diode fiber



<u>Single pulse laser ignition</u>
150 fs, 1 ps, 30 ns; 800 nm
1 ds – 10 μs; 808 nm

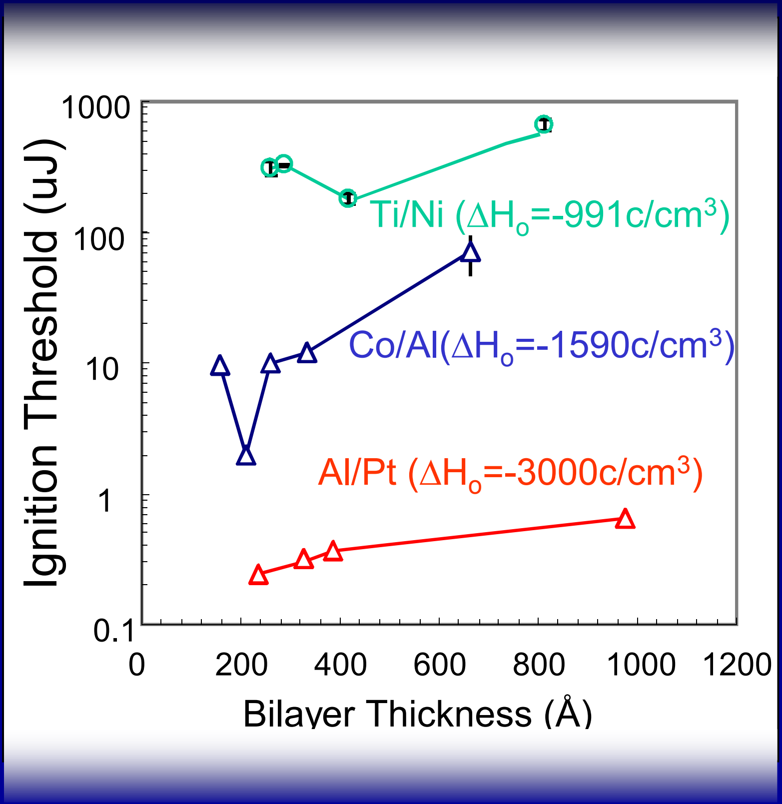
Approx. Gaussian shaped beam



Reactive foils are ignited by pulsed lasers.

Single pulse, single point ignition with 30 ns excimer laser, 248 nm spot size (dia.) = 8 μm

- Nanosecond pulse thresholds span a range of pulse energies ,< 1 μJ to 500 μJ.
- Differences between metal pairs is attributed to enthalpy, likely affected by mass transport and thermal transport.
- Data suggests an effect of multilayer design (i.e., bilayer thickness).



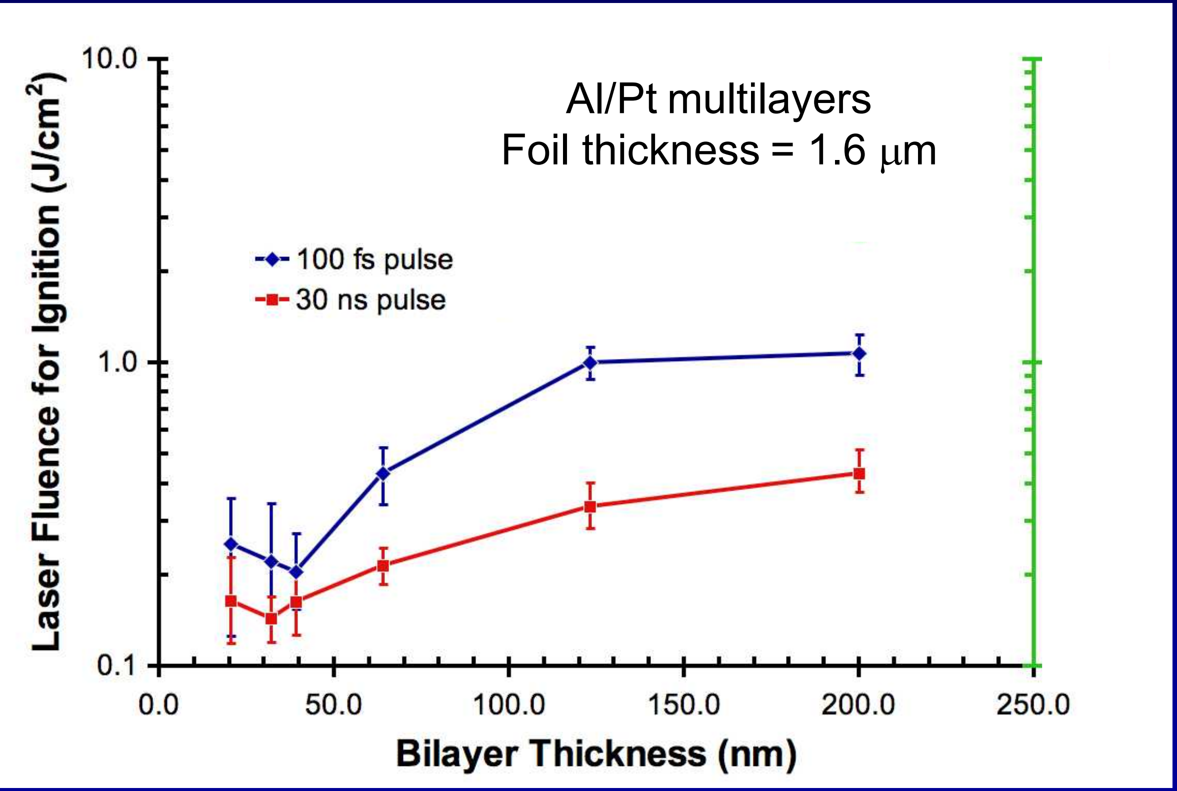
Y. Picard, J. P. McDonald, D.P. Adams, S.M. Yalisove Appl. Phys. Lett. 93 (2008).

Results compensate for reflectivity losses (assumed to be linear %R's)

Threshold fluence varies with multilayer design

Single pulse, single point ignition of Al/Pt with Ti:sapphire laser, 800 nm spot size (dia.) = 27.2 ± 0.4 μm.

- Thresholds vary with bilayer thickness.
- Threshold increases with bilayer thickness for a large range of this dimension (affected by mass transport)



Results compensate for reflectivity loss of Pt (assumed to be the linear R = 0.716)

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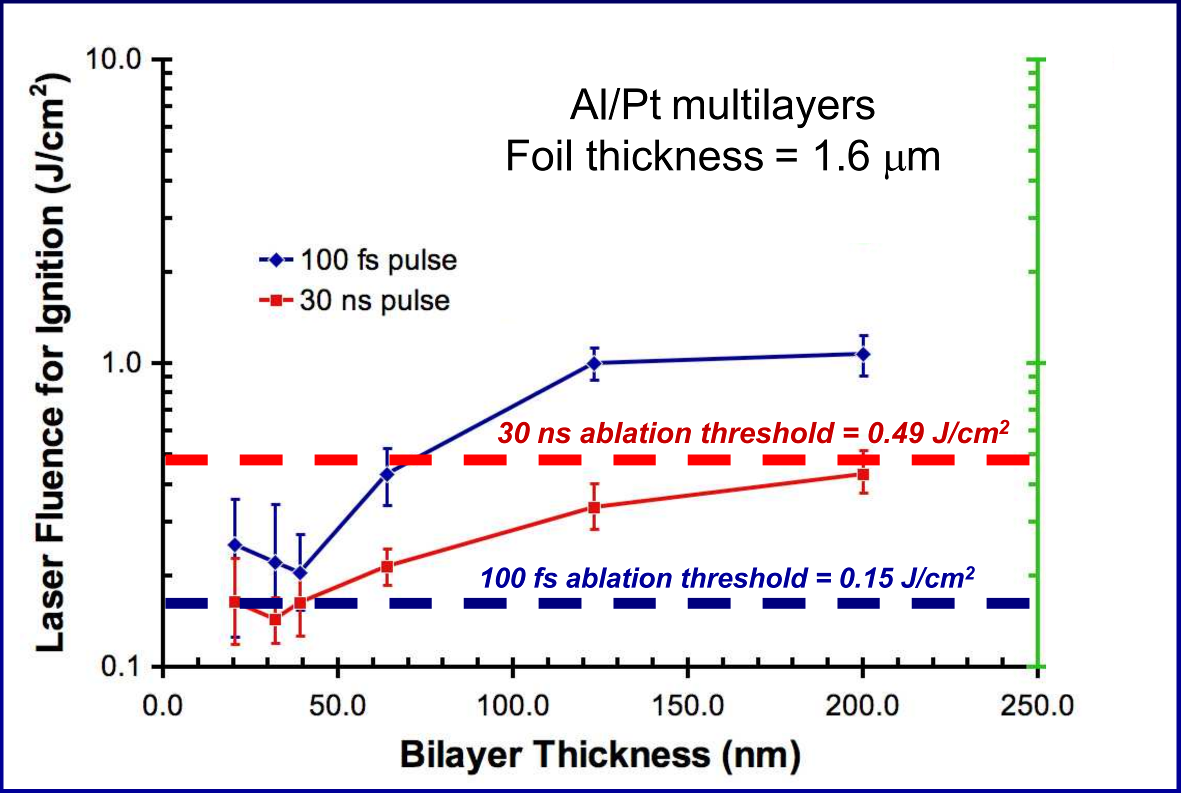
- Thresholds vary with bilayer thickness.
- Threshold increases with bilayer thickness for a large range of this dimension (affected by mass transport)
- Data shown with ablation thresholds demonstrates strategies for laser machining preforms for joining.
  (i.e., use fs laser light for this reactive pair)

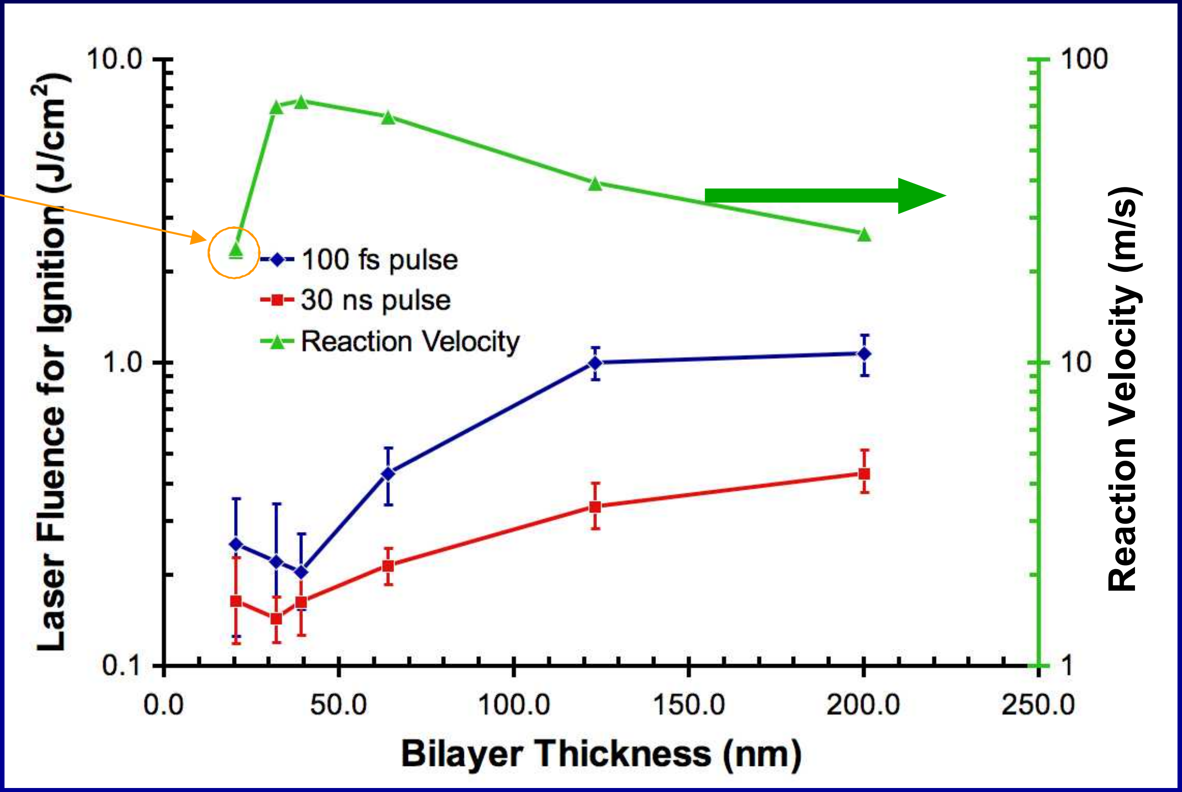


Results compensate for reflectivity loss of Pt (assumed to be the linear R = 0.716)

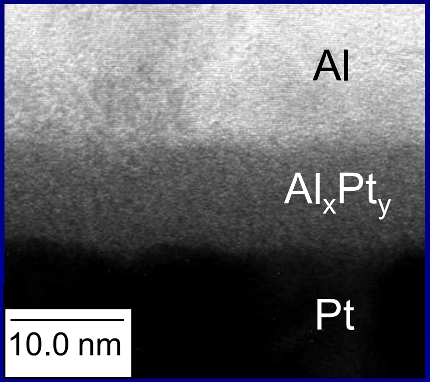
Laser induced ignition: Effects of multilayer design

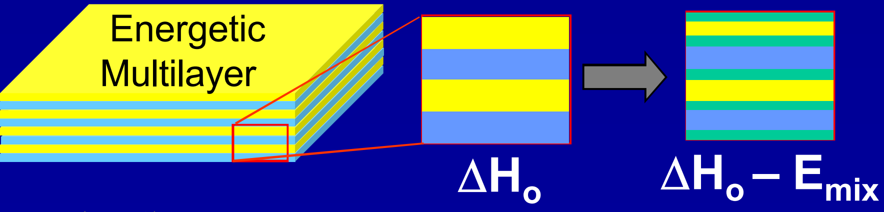
Single pulse, single point ignition of Al/Pt with Ti:sapphire laser

Reduced velocity and increased ignition fluence are due to the effects of rel. large amounts of premixed reactants (present before testing)



TEM

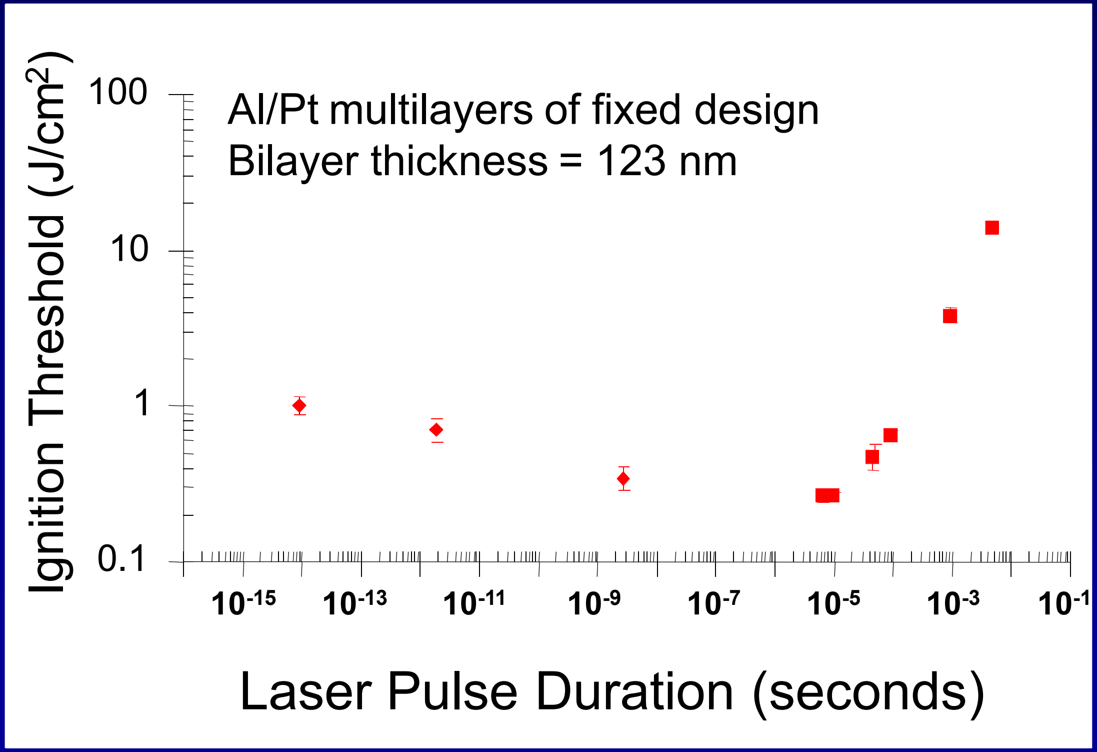




Laser induced ignition: Effects of pulse duration

- Decreasing thresholds for ignition when decreasing pulse duration from ds to ms to 10 μs (peak power is efficient at stimulating reaction).
- Increased thresholds when using ps, fs light (attribute to ablation, etc.)

Single pulse, single point ignition of Al/Pt with 800 nm (◆),808 nm (■) light spot size (dia.) = 91 μm



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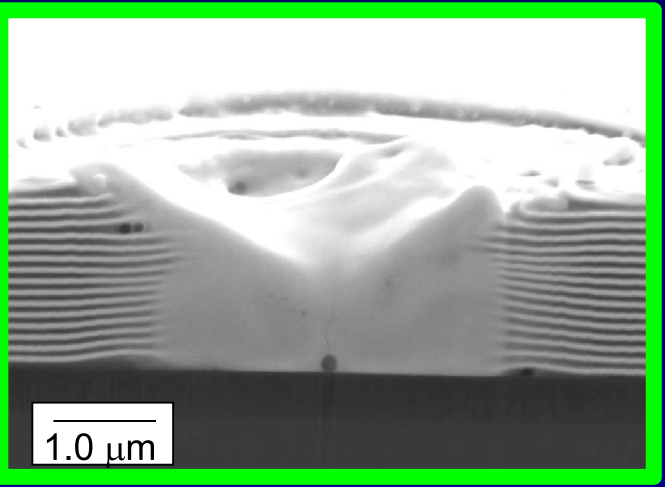
Ignition involves a competition of energy input and various loss mechanisms

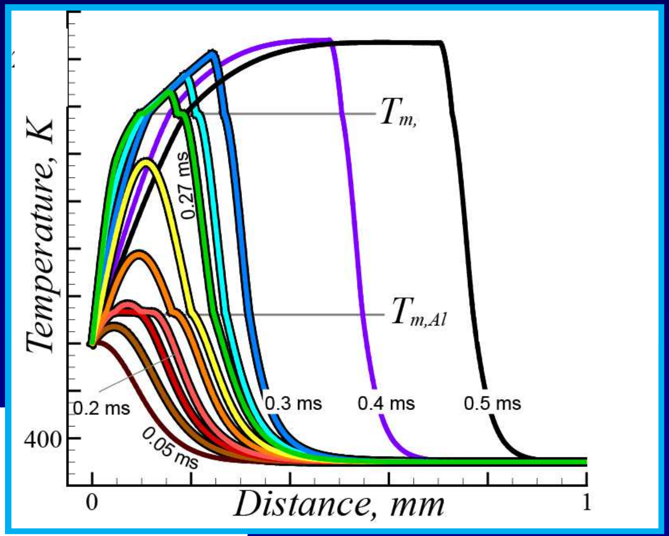
Ablation for ultrashort pulse
- loss of energetic materials

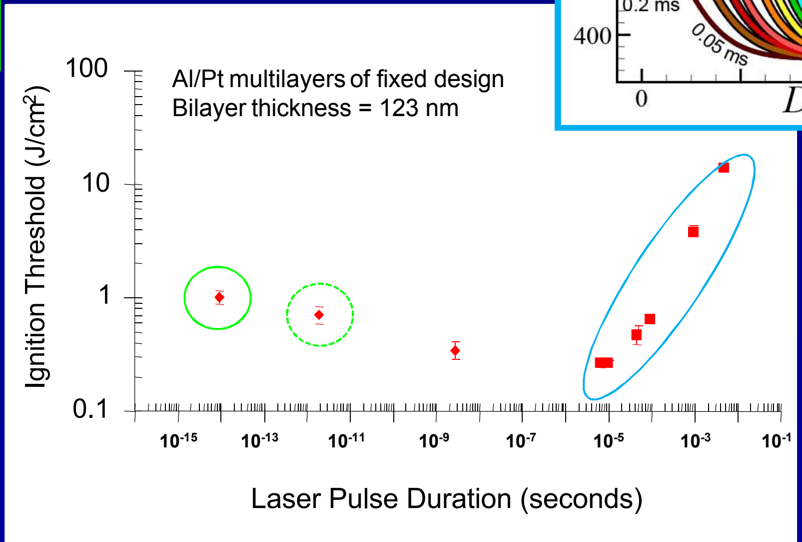


Al/Pt multilayer
irradiated with 80% threshold
(100 femtosecond pulse duration)

then sectioned by FIB

Finite difference simulations
of reactive multilayer ignition
- Losses via thermal conduction
and latent heat of melting





Summary

- **Pulsed laser irradiation is a useful method for detailed studies of reactive multilayer ignition.**

- **Ignition threshold of reactive multilayers**

  - varies with material system

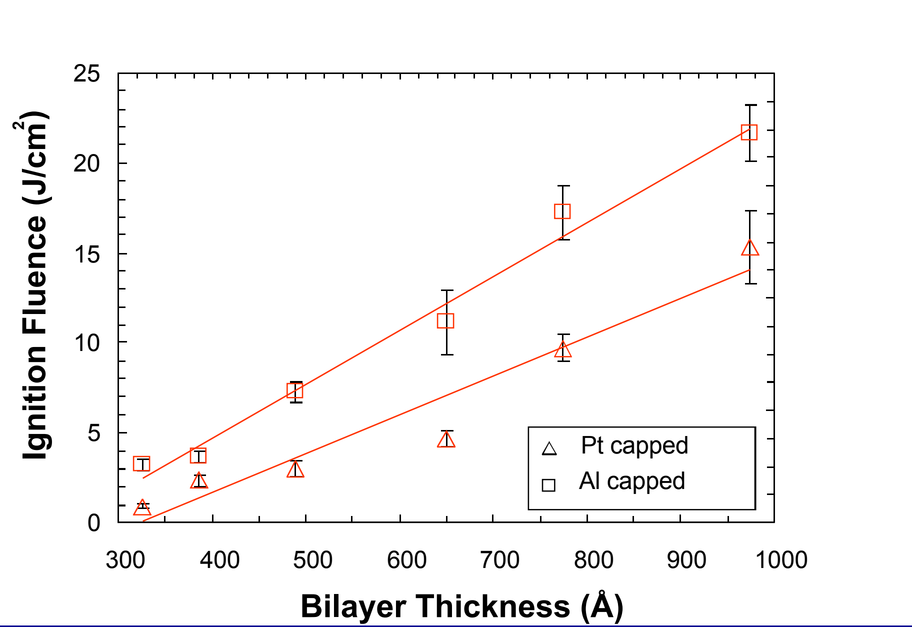
  - is affected by multilayer design (for a given reactive pair) and

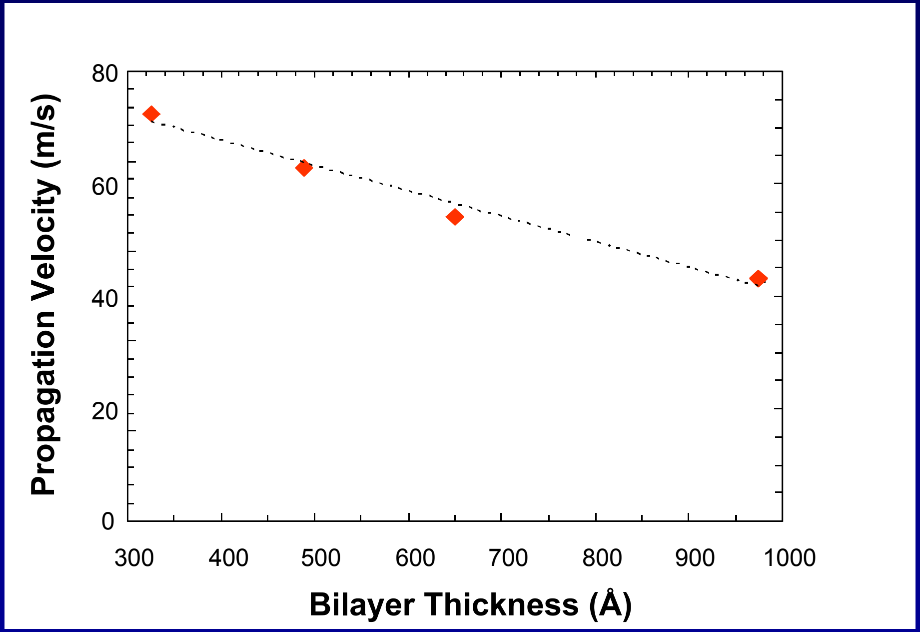
  - varies with pulse duration (for a given reactive pair of fixed multilayer design).

BACKUP SLIDES

Laser ignition of Al/Pt: Influence of capping layer reflectivity

Example: Single pulse, single point ignition
with 120 ns Ti: sapphire laser, 800nm
spot size = 8 μm

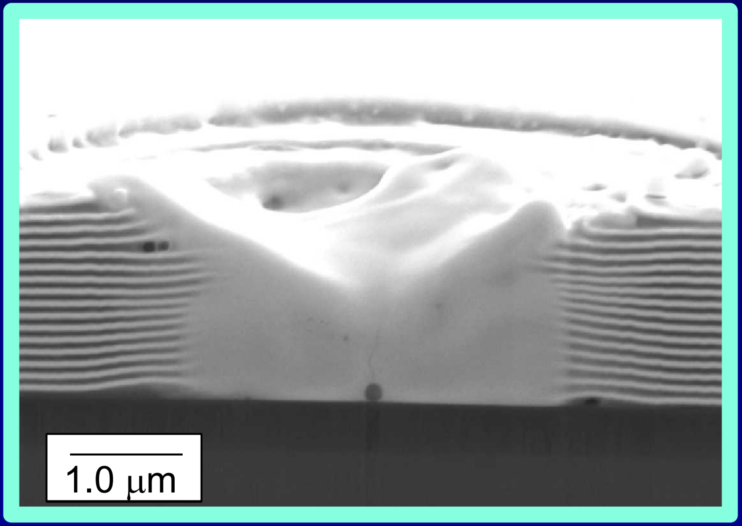




Difficulty in evaluating heat-affected-zones

Al/Pt multilayer
irradiated with 80% threshold
(100 femtosecond pulse duration)

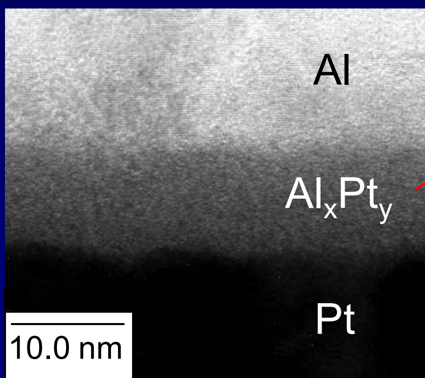
then sectioned

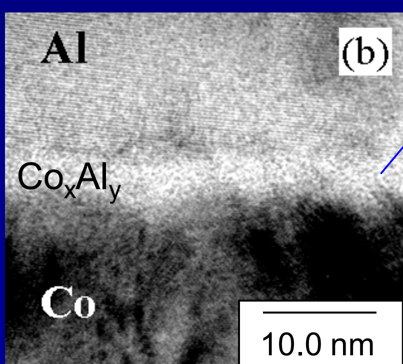


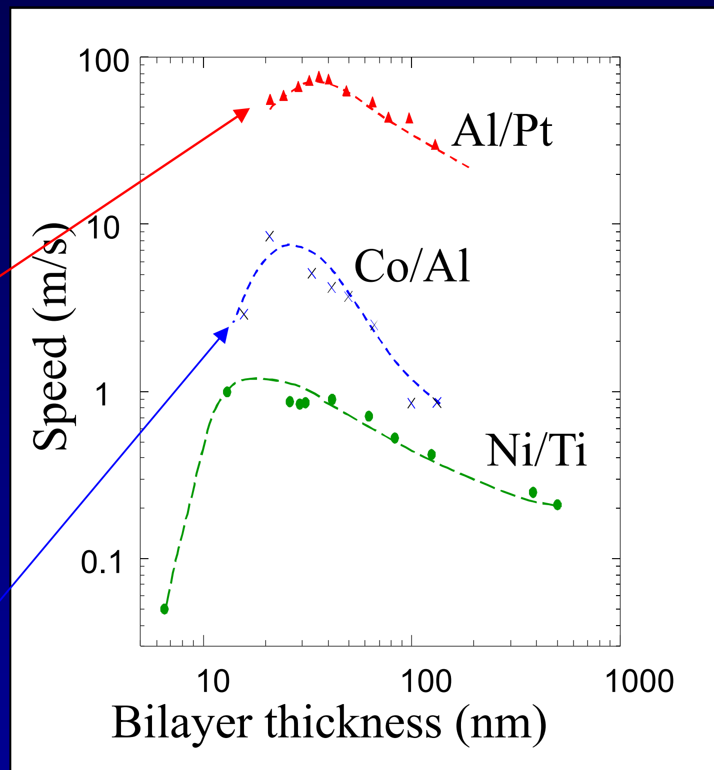
Reaction zone could be solely the result of laser Irradiation

Reaction zone could also be affected by a partial (incipient) reaction wave that terminated close to the exposed area

Propagation rate is affected by multilayer design







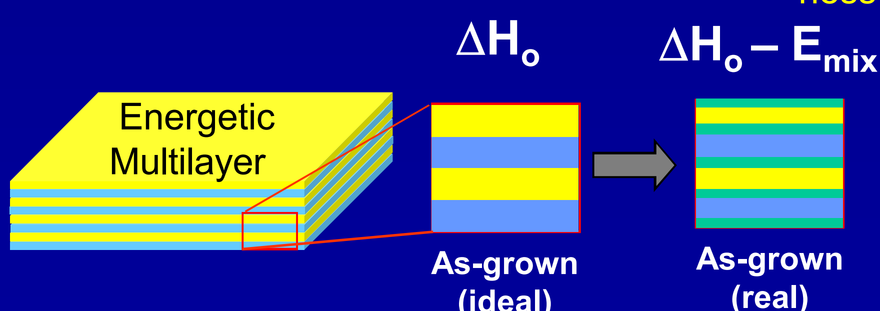
- Propagation speed increases with decreasing bilayer thickness for a large range of designs.

consistent with R. Armstron (1990)

$$V^2 = \frac{3A\exp(-E/RT_c)RT_c^2\lambda^2}{(1/4t_{BL})^2E(T_c-T_o)}$$

- Differences in speeds (for diff. materials) owe to a larger ΔH_o and differences in mass/thermal transport.

- Propagation speeds affected by pre-mixed reactants when bilayer thickness is small.

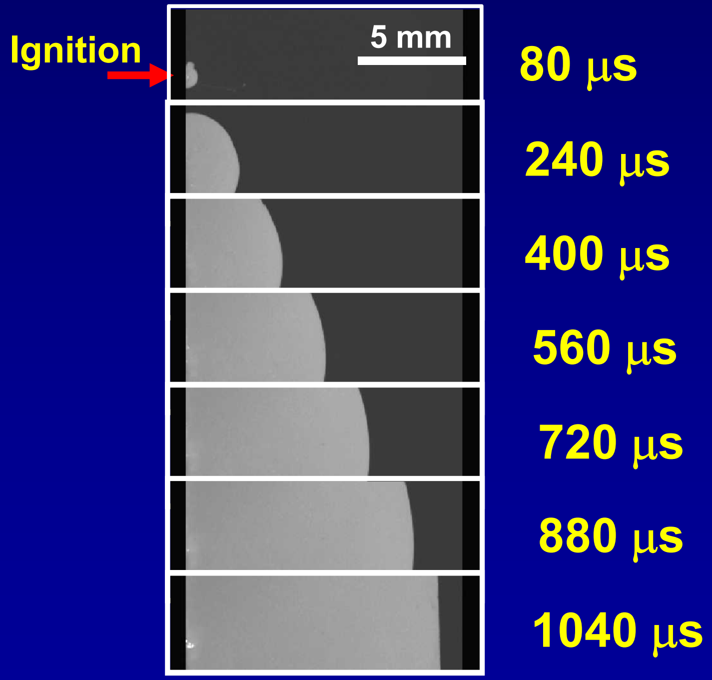


Y. Picard, J. P. McDonald, D.P. Adams, S. Yalisove Appl. Phys. Lett. 93 (2008).

Al/Pt multilayers exhibit stable reaction modes characterized by a circular wavefront

Ex. Al/Pt multilayer
in plan view

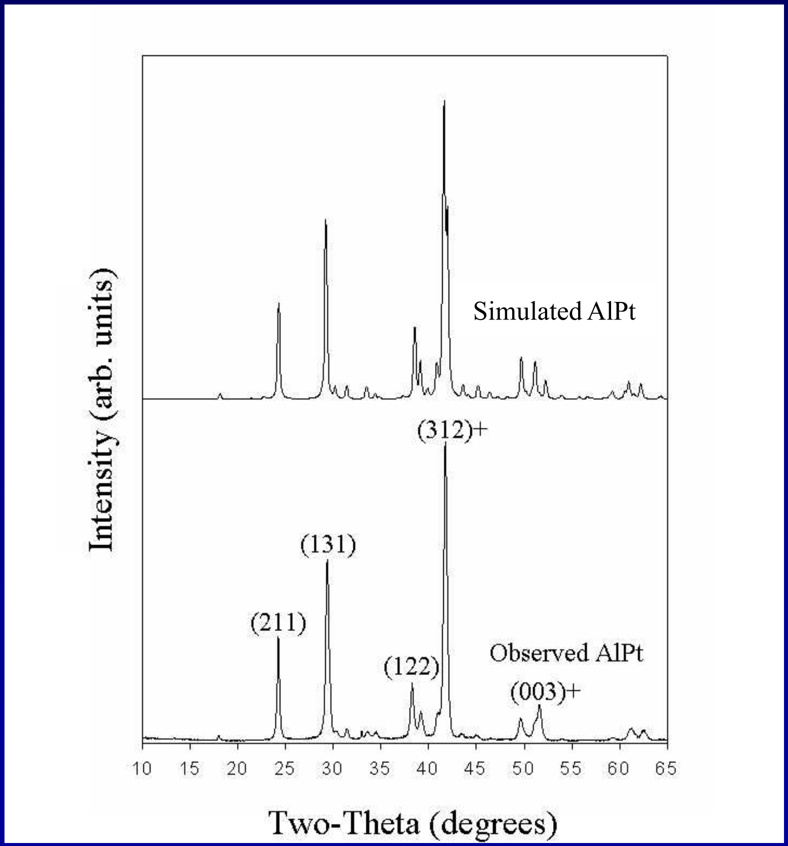
- Reaction front is smooth when observed at the micrometer scale
- No evidence of oscillations or spin waves



21 nm bilayer thickness

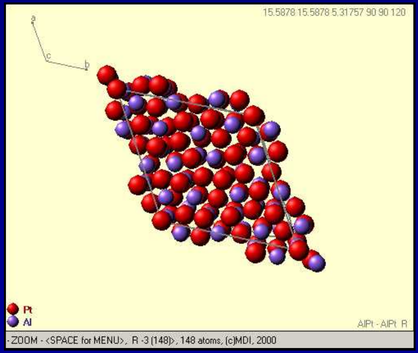
Phase formed is not always according to equilibrium diagram

New phase of Al-Pt discovered.



$Al_{0.5}Pt_{0.5}$ is cubic (FeSi structure) under equilibrium conditions but rhombohedral as reacted film.

New rhombohedral phase has 39 formula units in the unit cell.



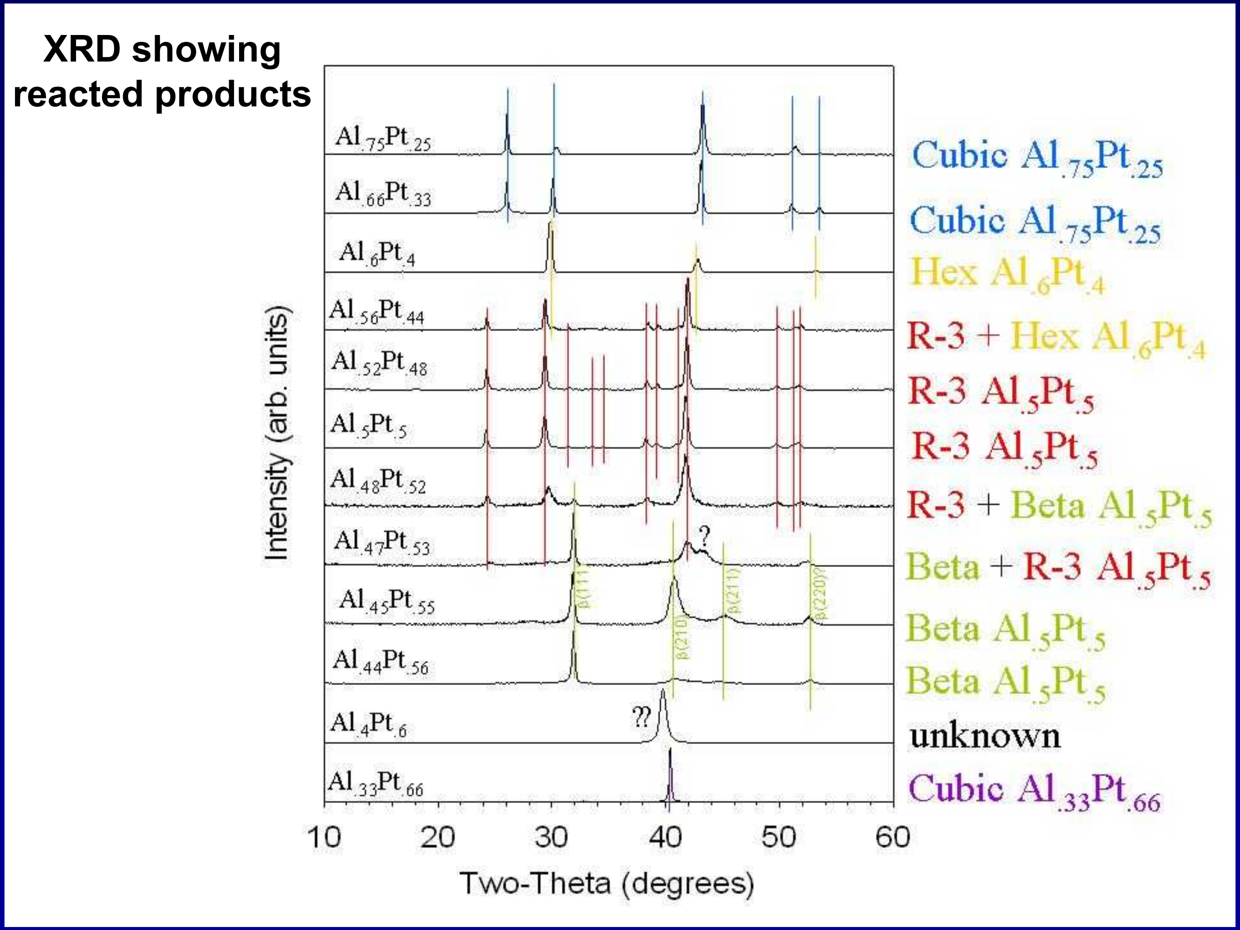
Suggests that rapid quench rates (est. -3.9×10^{8} C/sec) can influence phase formation.

D.P. Adams, M. Rodriguez
J. Mater. Res. 21 (2006).

A wide range of compositions often exhibit self-propagating reactions

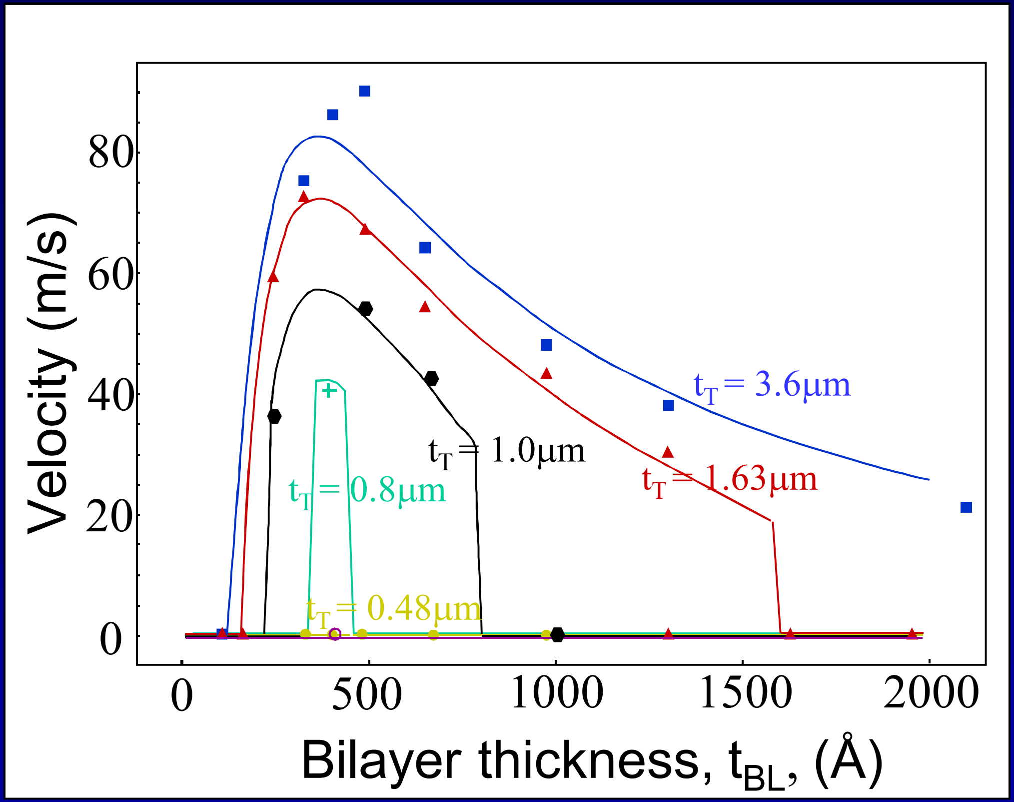
ex. Al/Pt

from $Al_{.75}Pt_{.25}$ to $Al_{.33}Pt_{.66}$



Propagation rates are influenced by total multilayer thickness

Example: Al/Pt on substrate



A variety of metal-metal multilayers have been evaluated at Sandia

Exothermic Materials (2-50 micron thick foils)	Composition of foil	**Heat of reaction** (kJ/mol atoms)	Propagation speeds (m/s)	Ignition Temperatures (°C)
Sc/Au	ScAu	-111 #	10 - 40	tbd
Y/Au	YAu	-110 #	8 - 15	tbd
Ti/B	TiB_2	-102 #	10 - 30	tbd
Al/Pt	AlPt	-100	15 - 95	tbd
Ni/Al	NiAl	-60 #	6 -10	200 - 300
Co/Al	CoAl	-58	0.3 – 10	280 - 450
Y/Ag	YAg	-44 #	0.5 – 0.8	tbd
Sc/Ag	ScAg	-43 #	0.2 – 0.5	tbd
Ni/Ti/B	$Ni_{.43}Ti_{.48}B_{.09}$	-38	0.5 - 4.5	340 - 500
Ni/Ti/C	$Ni_{.43}Ti_{.48}C_{.09}$	-37	1 – 5.0	320 - 480
Sc/Cu	ScCu	-36	0.2 - 0.9	tbd
Ni/Ti	NiTi	-34	0.1 – 1.0	280 - 400
Y/Cu	YCu	-32 #	0.2 – 0.4	tbd

Compare with

$\Delta H_o (Al_2O_3)$ =
-1670 kJ/mol

Reference deBoer