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Lead Azide and PETN Reactions at Sub-mm Geometries

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Workshop on “Synthesis of Advanced Energetic Materials-The Path Forward”,
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Introduction
Explosives Scaling

- **Desire to reduce the amount of sensitive explosives in energetic components**
  - **Safety**
  - **Undesirable materials (e.g., lead)**

- **Stewart suggested:**
  - **Acknowledge transient behavior at output or:**
  - **Select material with suitable critical parameters (reaction zone, critical diameter)**

Stewart, D.S., "Towards the miniaturization of explosive technology," Shock Waves, vol. 11, pp. 467-473, May, 2002.

Introduction
DDT

- Deflagration to Detonation Transition (DDT)
  - Secondary explosives – long length scales
  - Primary explosives – short length scales
- DDT mechanisms – how does burning grow to a detonation?
  - Conductive → Convective → Shock wave
  - Or:
  - Compaction → Plug Formation → Shock wave
- Understanding the process of DDT in small explosives is important for scaling

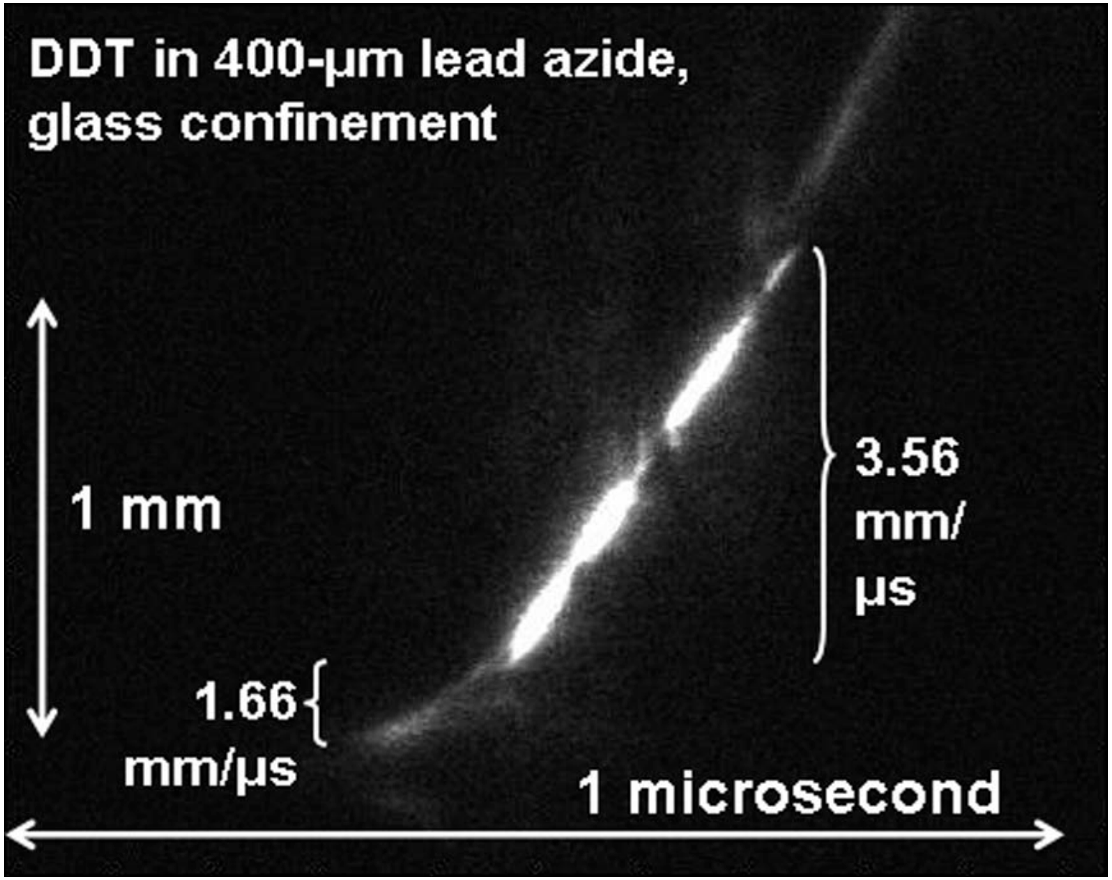
Dickson, P.M., Parry, M.A., and Field, J.E., "Initiation and Propagation in Primary Explosives," Ninth Symposium (International) on Detonation, Portland, OR, August 28-September 1, 1989, pp. 1100-1109.

Dickson, P.M. and Field, J.E., "Initiation and Propagation in Primary Explosives," Proceedings: Mathematical and Physical Sciences, vol. 441, pp. 359-375, 1993.

Ermolaev, B.S., Sulimov, A.A., Okunev, V.A., and Khrapovskii, V.E., "Mechanism for Transition of Porous Explosive System Combustion into Detonation," Combustion, Explosion, & Shock Waves, vol. 24, pp. 65-68, 1988.

Introduction
Lead Azide DDT in Glass Capillary

- Early work
- 400-µm inner diameter capillary
- Based on this result, further experiments were conducted to examine the DDT process in sub-mm lead azide charges



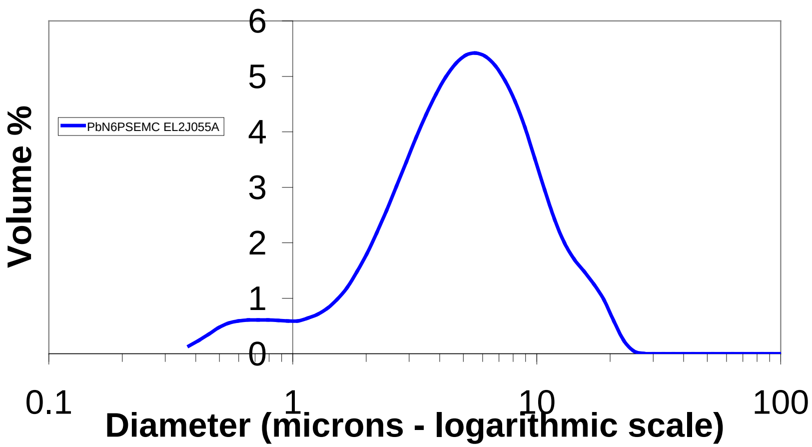
Madden, S.P., Tappan, A.S., Jung, P.C., Marley, S.K., Welle, E.J., and Pahl, R.J., "Rapid Data Analysis Methodologies for Streak Camera Images: Measurement of Detonation Velocity and DDT Distance of Lead Azide at Sub-Millimeter Diameters," 13th International Detonation Symposium Norfolk, VA, July 23 – 28, 2006.

Jung, P.C., "Initiation and Detonation in Lead Azide and Silver Azide at Sub-Millimeter Geometries," Mechanical Engineering, Master's Thesis Lubbock: Texas Tech University, December, 2006.

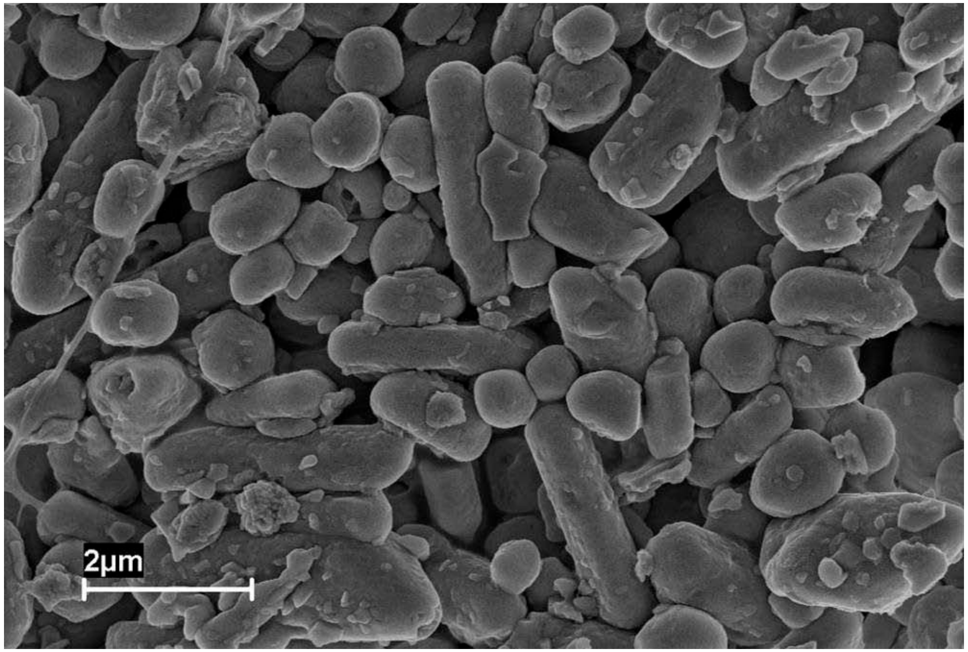
Lead Azide

- Colloidal lead azide synthesized by Pacific Scientific Energetic Materials Co., Chandler
- 6-µm average particle size

Lead Azide Coulter Light-Scattering Particle Size Analysis



Particle size analysis of lead azide.



SEM of lead azide.

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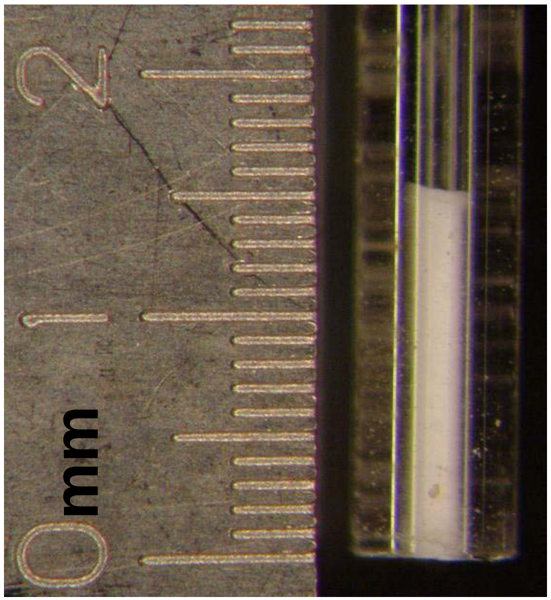
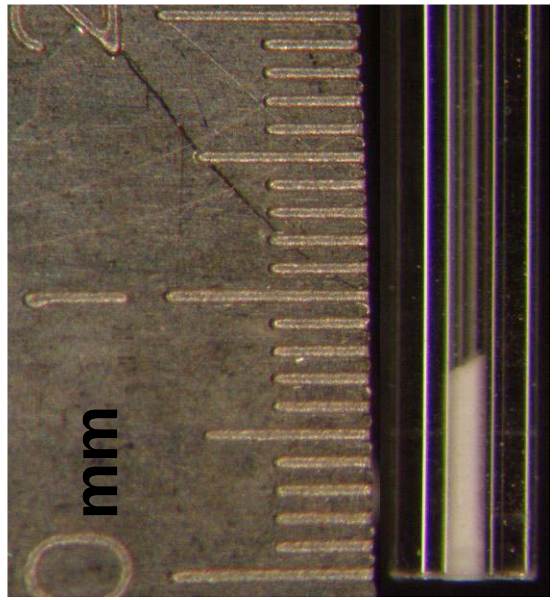
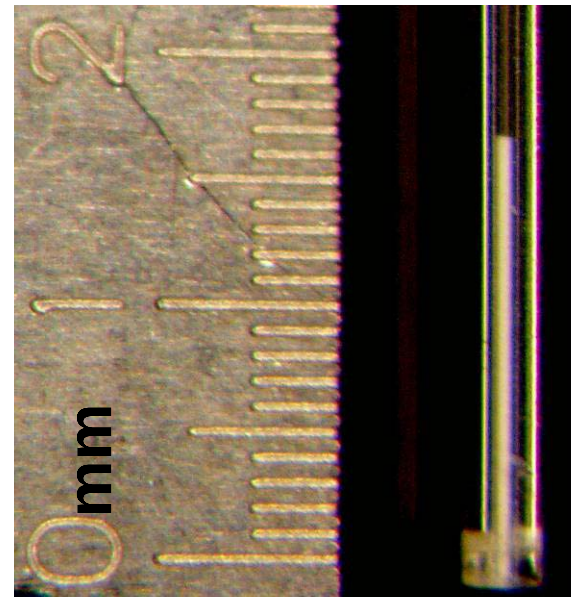
Lead Azide Charge Examples



200-µm diameter charge



100-µm diameter charge

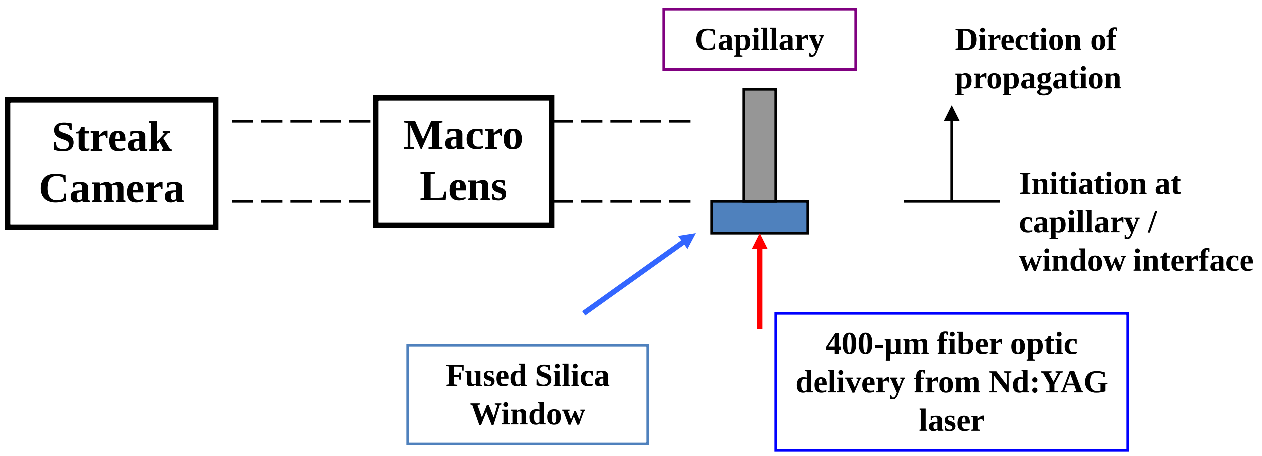


50-µm diameter charge

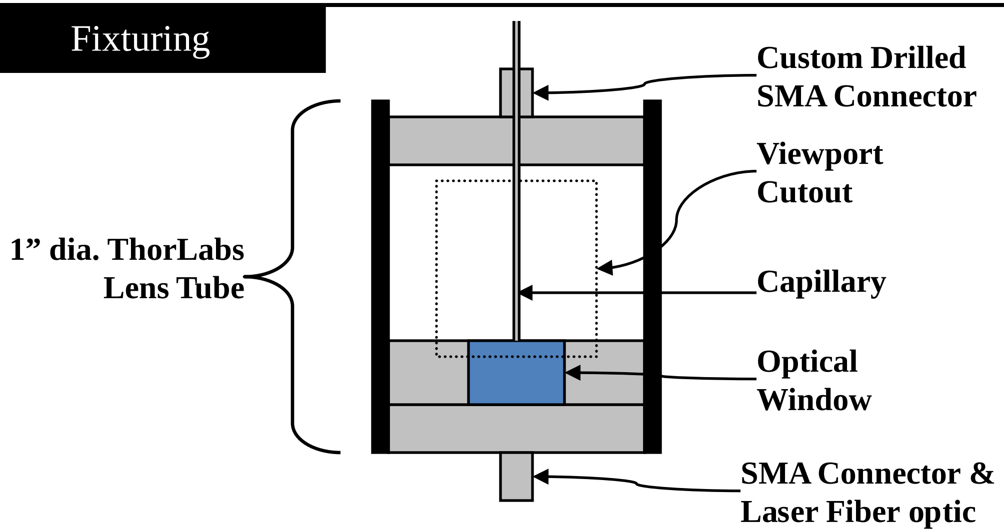
- **Dispersed with 1% dispersant**
- **Not pictured: 10-µm diameter charge**

Experiment Setup

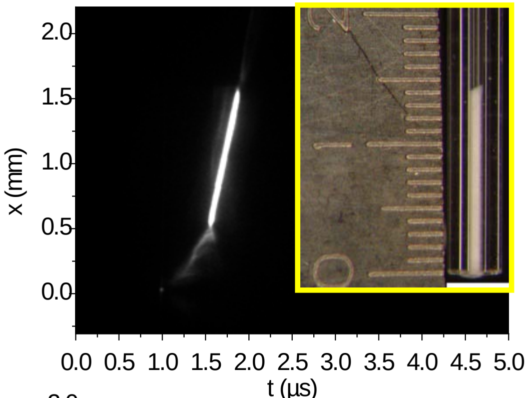
Optical Train

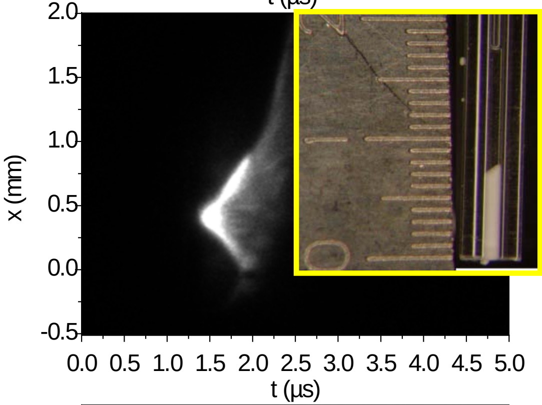


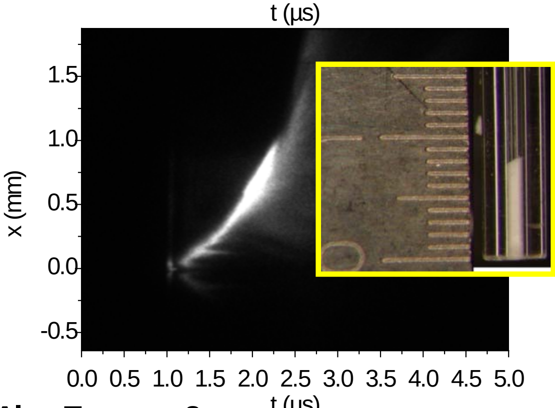
Fixturing



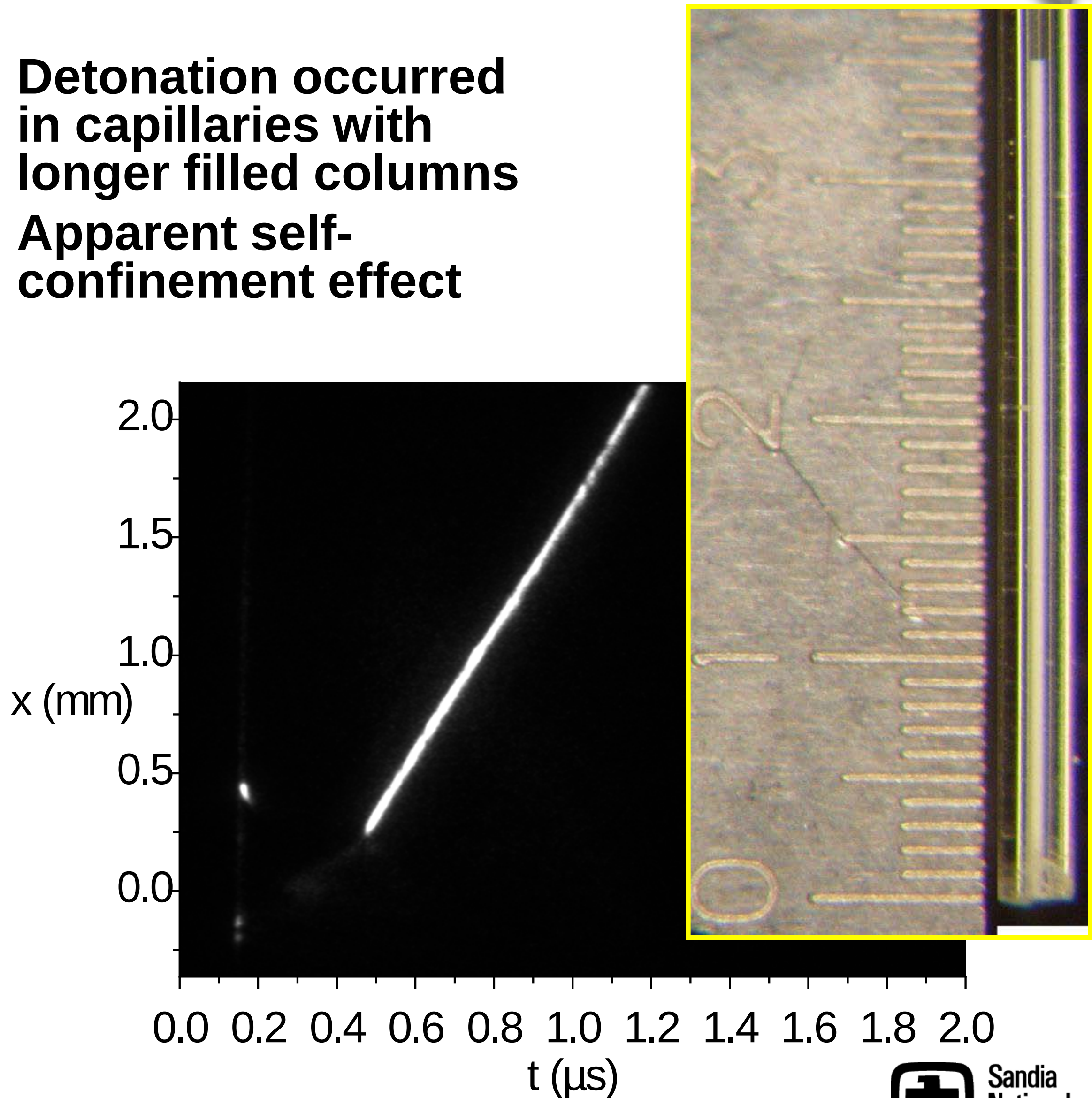
Lead Azide Reaction Comparisons





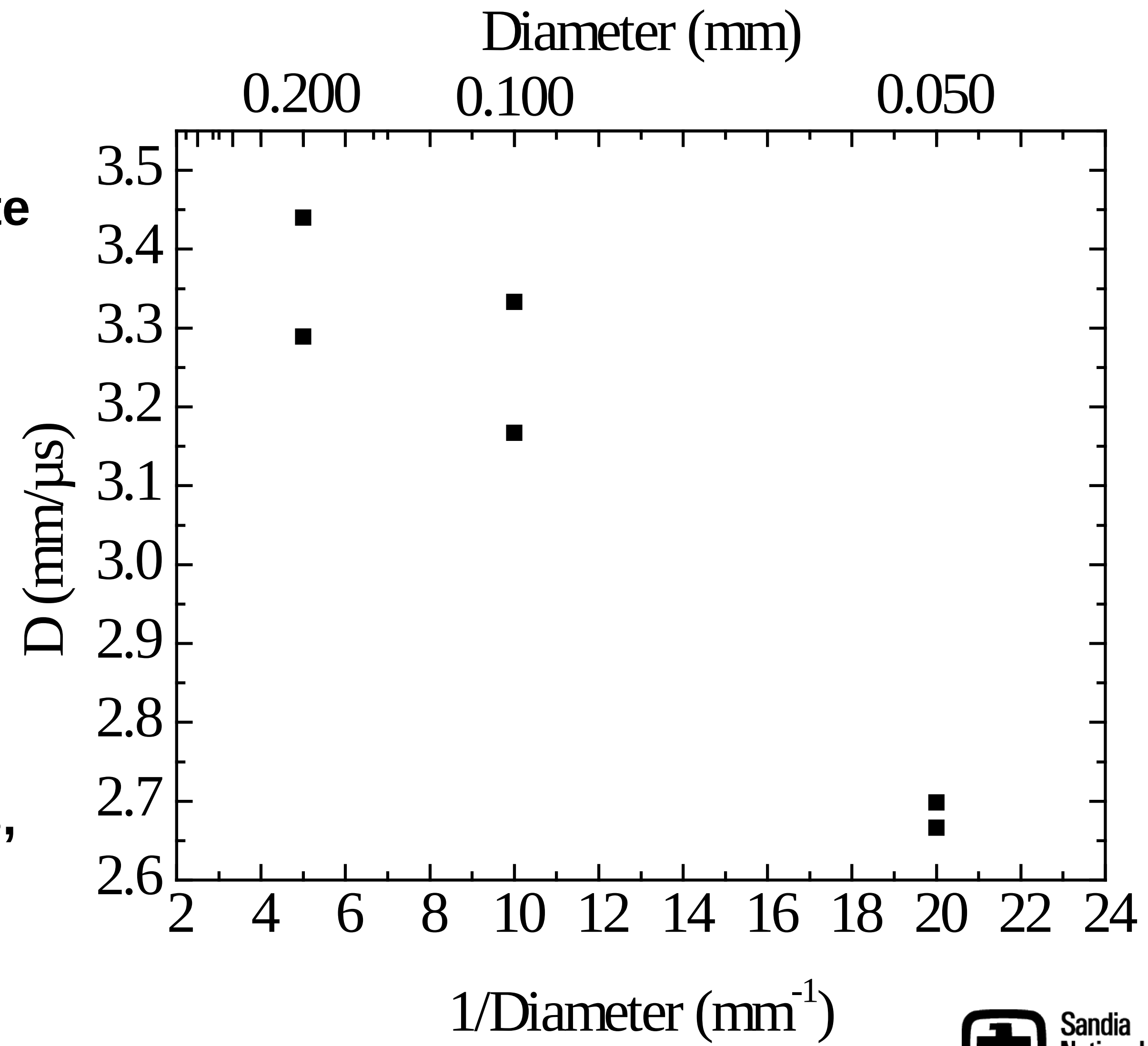


- **Detonation occurred in capillaries with longer filled columns**
- **Apparent self-confinement effect**



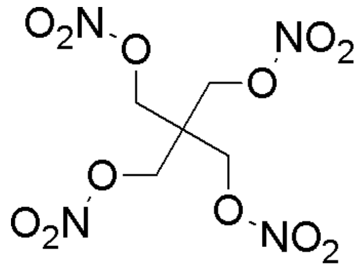
Diameter Effect in Lead Azide

- Six capillaries achieved steady detonation (50-, 100-, and 200-μm)
- Difficult to separate diameter (known) effect from possible density (unknown) effect
- For small sizes, diameter approached particle size and may have had an effect on the density
- For these samples, the critical diameter is less than 50-μm



Introduction

- **Data for small-scale explosive behavior of high-density PETN do not exist**
  - **Difficult to prepare small-scale samples**
  - **Low-density PETN data exist (ca. 50% TMD)**
  - **High-density PETN data exist only for PBX with 20% binder**
    - **Critical diameter of 0.222 mm (polycarbonate confinement)**



PETN

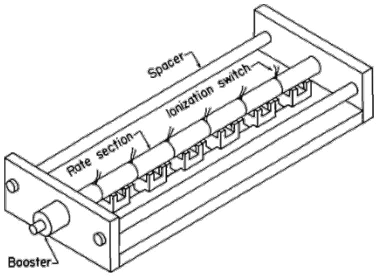
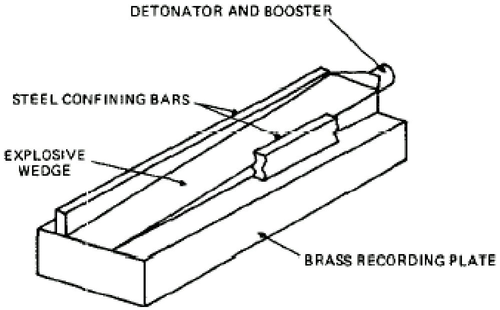


Fig. 1. Schematic of a typical rate-stick assembly.

Rate stick experiment.



Detonation failure experiment.

Campbell, A.W. and Engelke, R., "The Diameter Effect in High-Density Heterogeneous Explosives," *6th Symposium (International) on Detonation, Coronado, CA, August 24–27, 1976.*

Gibbs, T.R. and Popolato, A., "LASL Explosive Property Data," pp. 289–290, University of California Press, Berkeley, Los Angeles, London, 1980.

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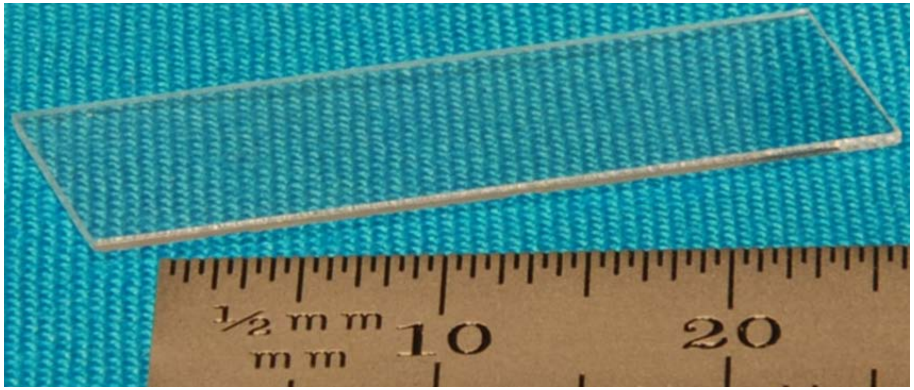
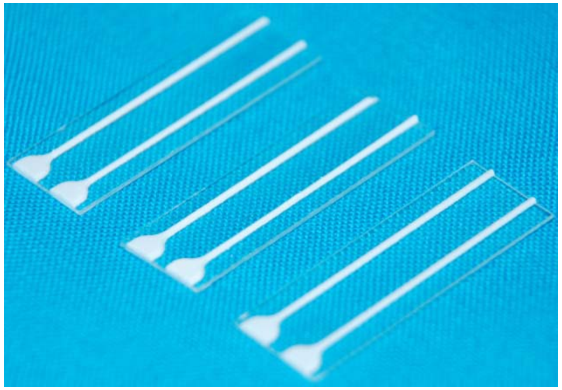
Physical Vapor Deposition Can Be Used to Make Small-Scale PETN Samples

- **Physical vapor deposition is used to sublime/evaporate PETN from a hot source onto a cool substrate**
- **Substrates are 0.5   10.0   30.0 mm fused silica**
- **Shadow masks are used to pattern lines of different widths**
  - **0.40, 0.60, 0.80, 1.00, 1.50, and 2.00 mm**
- **Deposition times control thicknesses (0.13–0.53 μm)**



Photograph of bare substrate prior to deposition.



Photograph of deposited PETN films.

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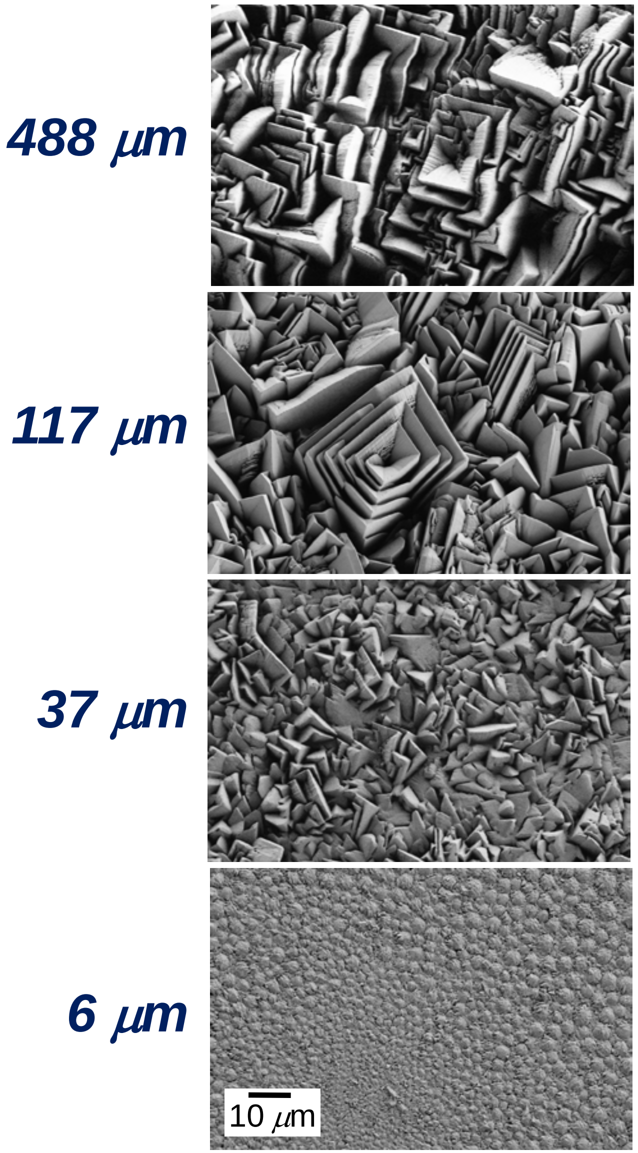
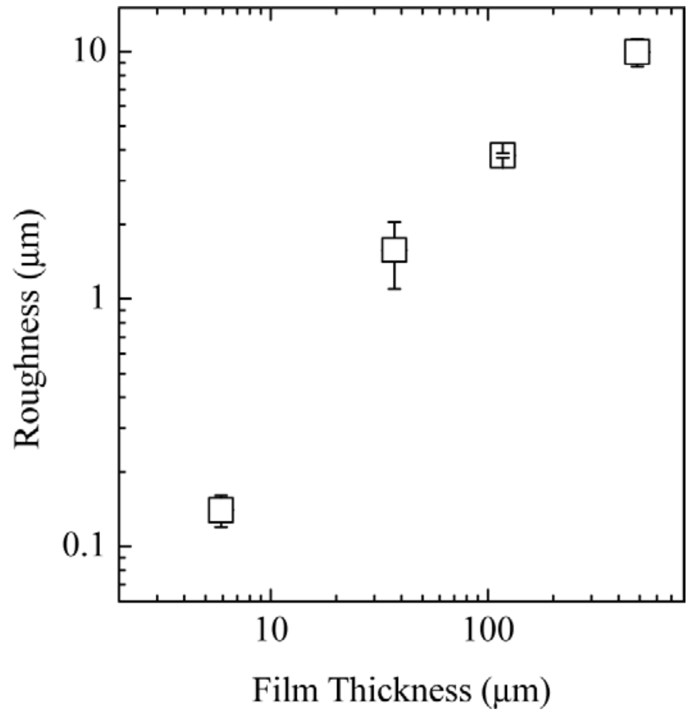
PETN Surface Roughness Evolution

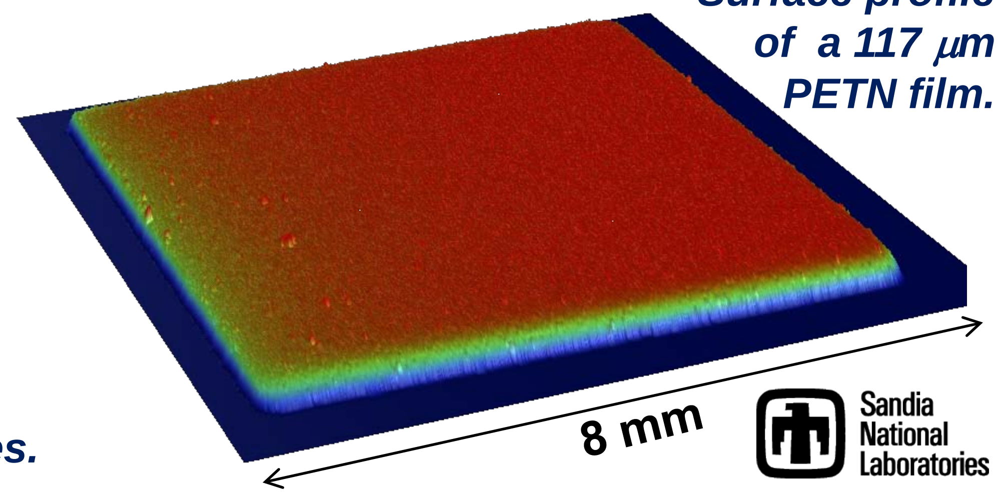


SEMs of different film thicknesses.



Surface roughness increases according to a power law with increasing film thickness.

Surface profile of a 117 μm PETN film.



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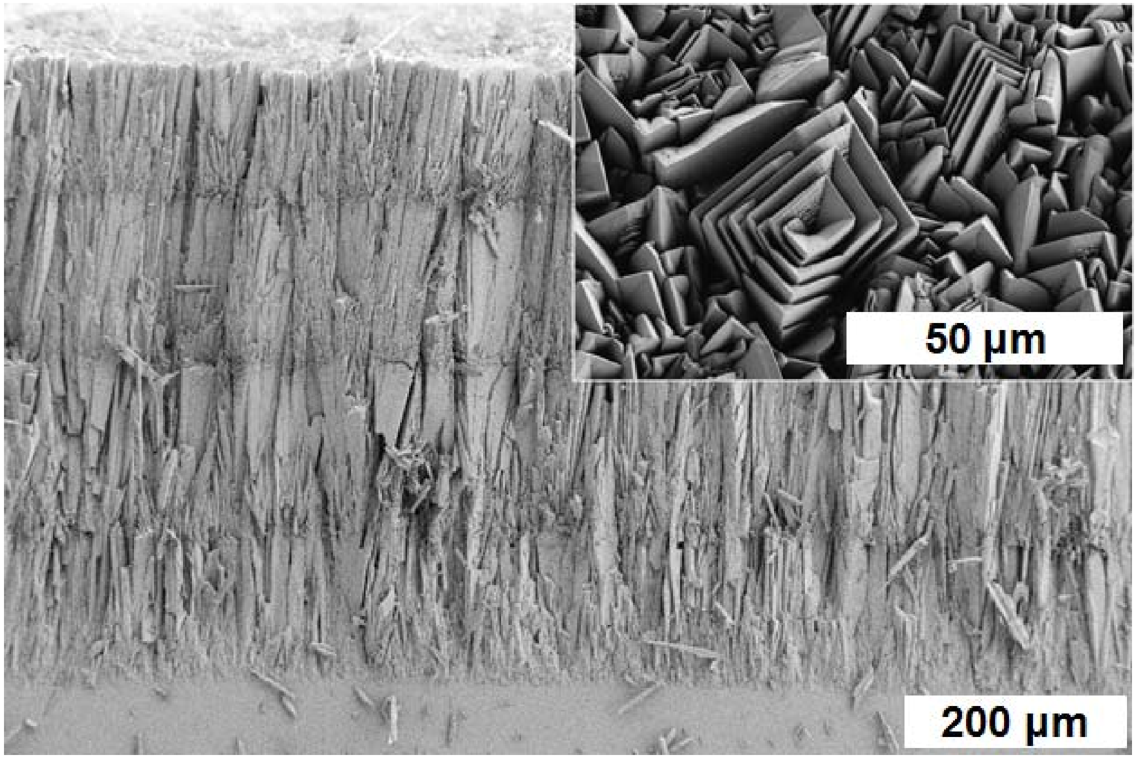
PETN Films Have High Density and Fine Grain Structure

- 1.41−1.50 g cm^{-3} (79−84% theoretical maximum density (TMD) of 1.778 g cm^{-3})
- Density gradient through thickness – densest at substrate
- Columnar grains of PETN elongated in the direction of film growth

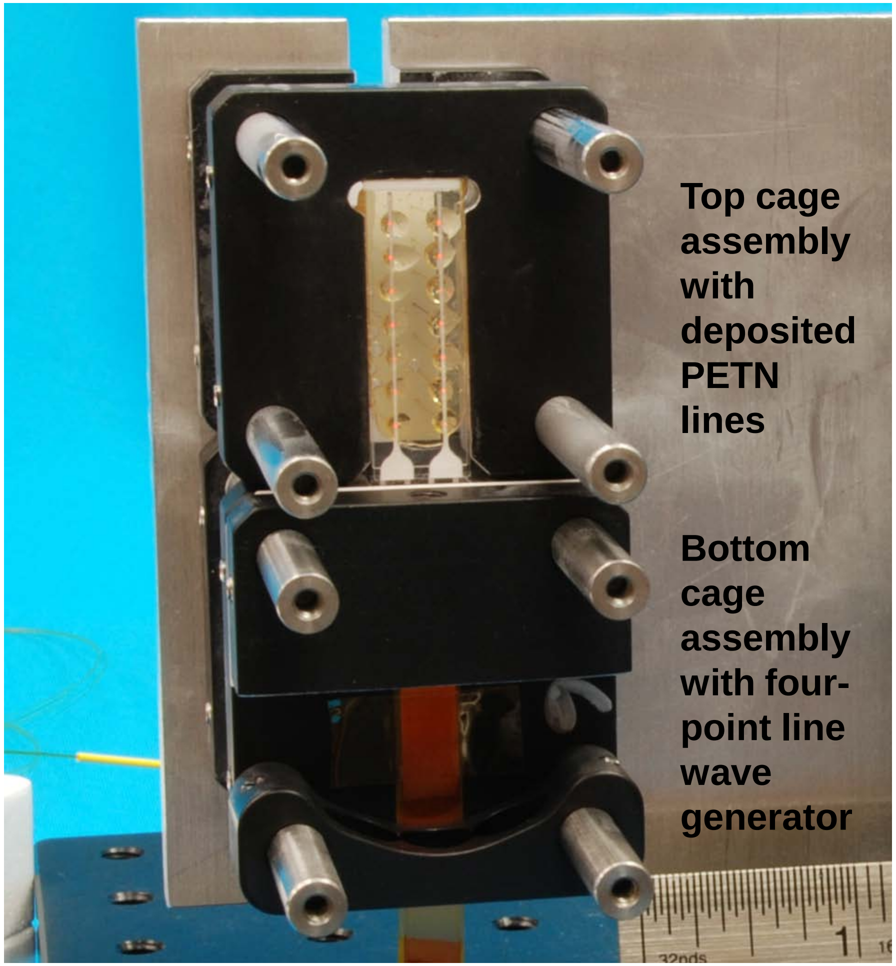


Scanning electron micrograph of fractured PETN film on fused silica. Inset shows top surface of deposited film.

Knepper, R., Tappan, A.S., and Wixom, R.R., "Controlling the Microstructure of Vapor-Deposited Pentaerythritol Tetranitrate (PETN) Films," *14th International Detonation Symposium, Coeur d'Alene, ID, April 11–16, 2010.*

Detonation Velocity Measurement Experiment

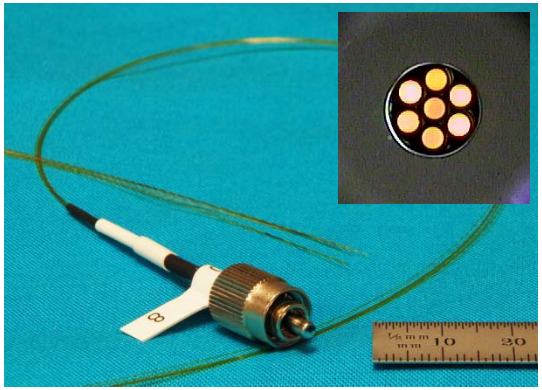
- **Detonation in deposited PETN lines is achieved by a four-point line wave generator**
- **Up to two experiments are conducted at once**
- **Cage assembly accommodates different PETN line thicknesses**
- **PETN confined within fused silica and epoxy**



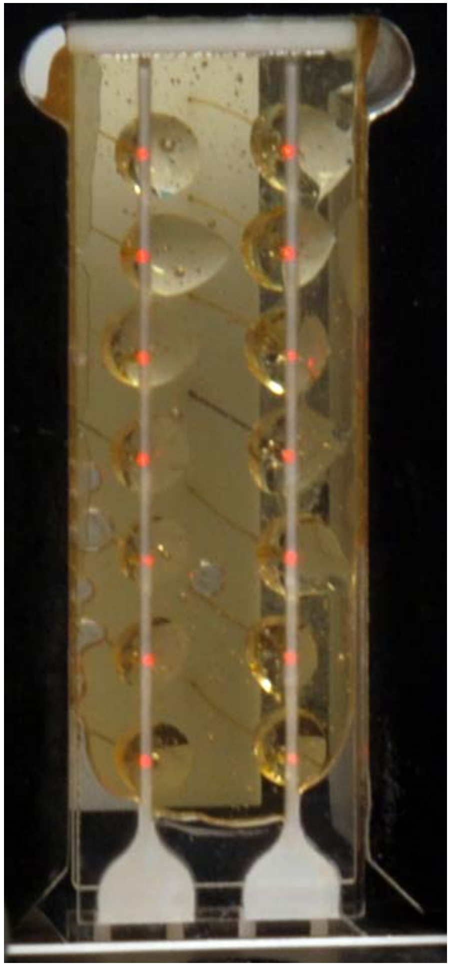
Photograph of experiment used to measure detonation velocity.

Optical Fiber Probe Is Used to Measure Detonation Velocity

- Optical fiber probe consists of seven 100 μm core silica fibers terminated in a six-around-one connector
- Optical fibers inserted through laser-machined holes in fused silica lid and bonded with epoxy
- Polished or pre-cleaved at lid
- Data acquisition with Si photodetector

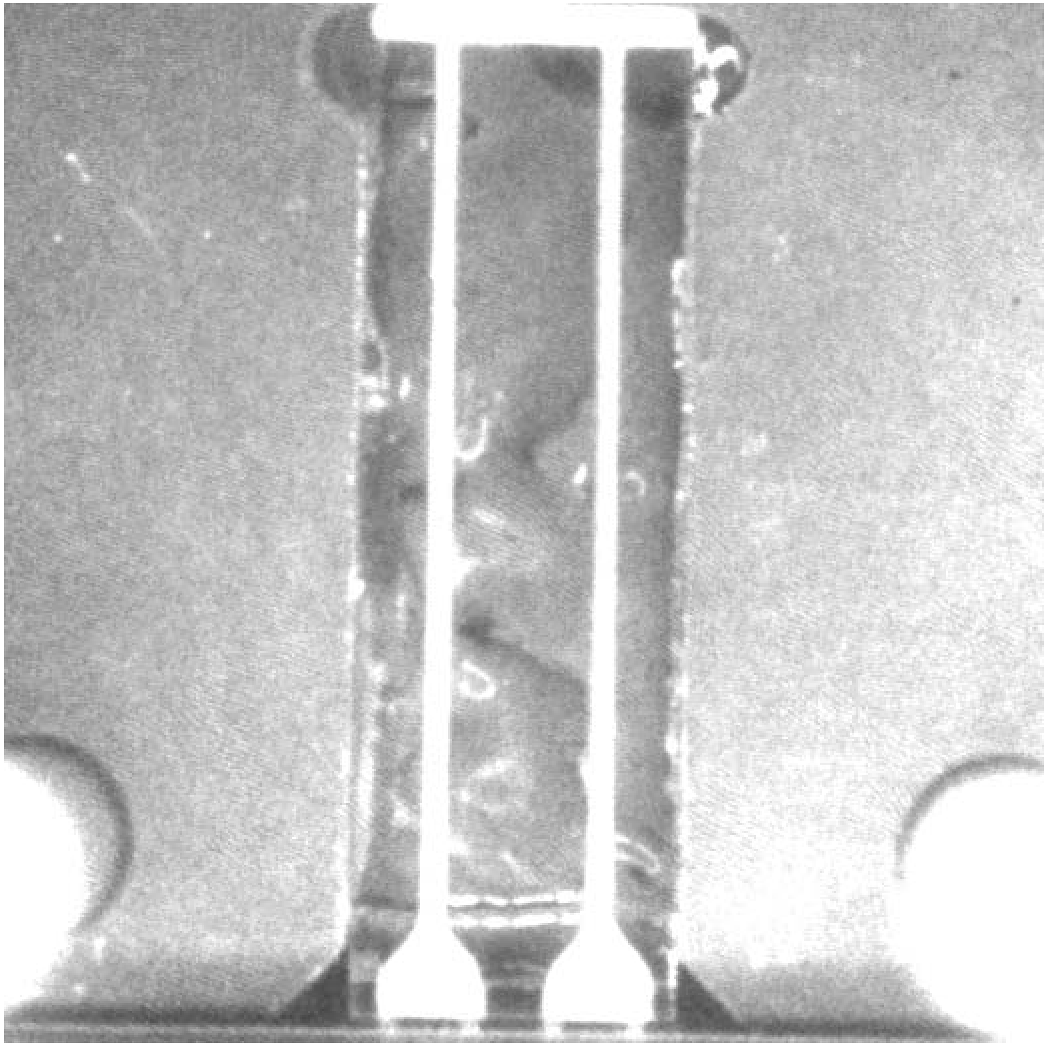


Photograph of optical fiber probe with inset showing six-around-one connector.



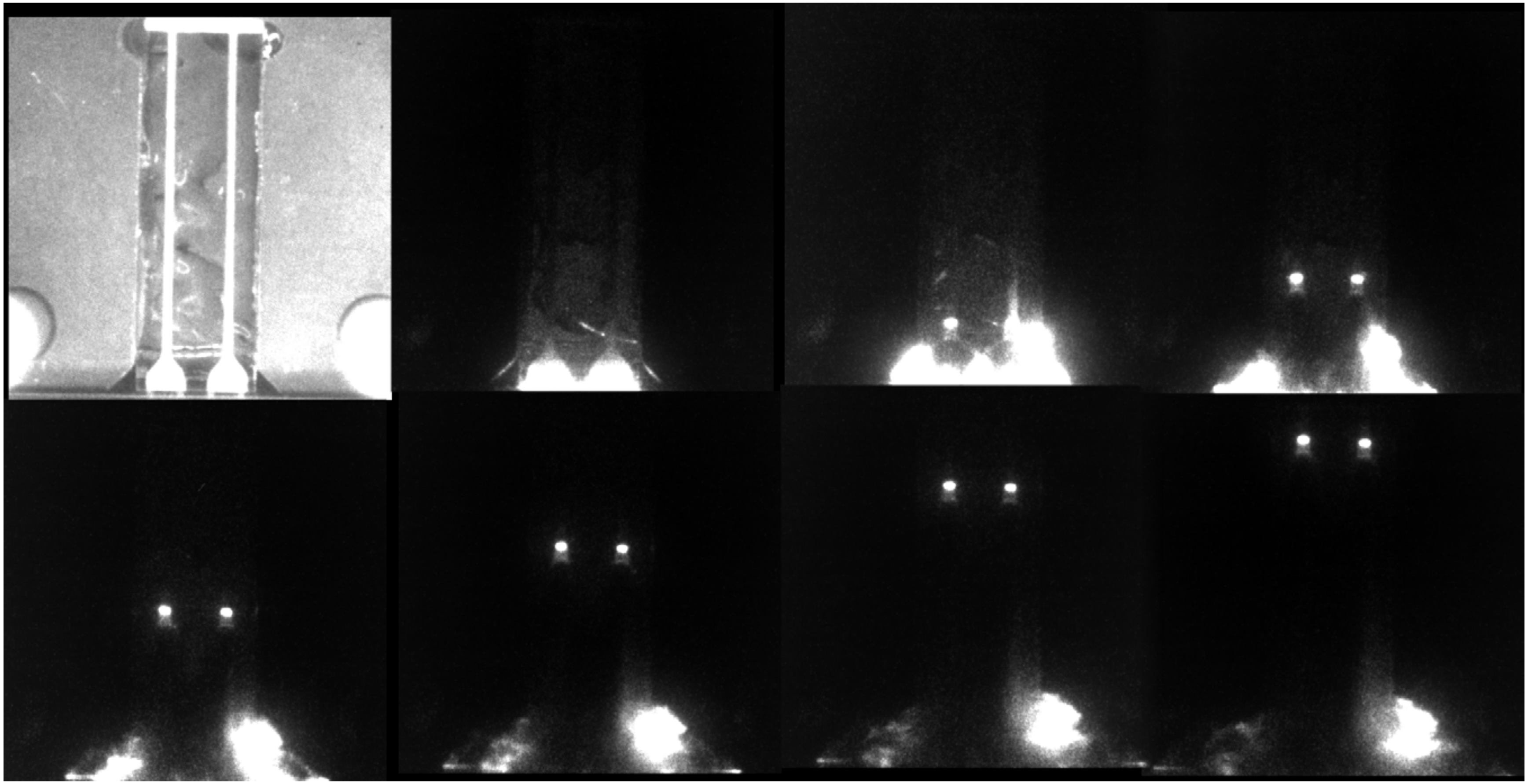
Photograph of optical fiber probe lid on deposited PETN. Optical fibers illuminated to show position.

Framing Camera Used for Qualitative Detonation Information



Framing camera images of detonation in deposited PETN lines. 1.67 million frames per second (1/600 ns), 20 ns exposure time.

Framing Camera Used for Qualitative Detonation Information



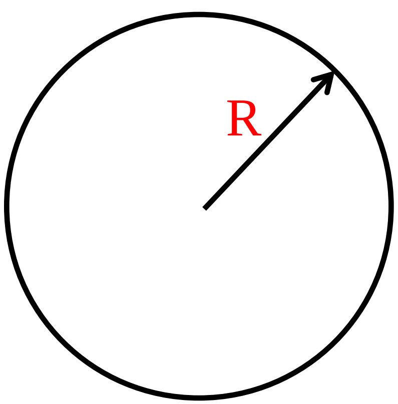
Framing camera images of detonation in deposited PETN lines.
1.67 million frames per second (1/600 ns), 20 ns exposure time.

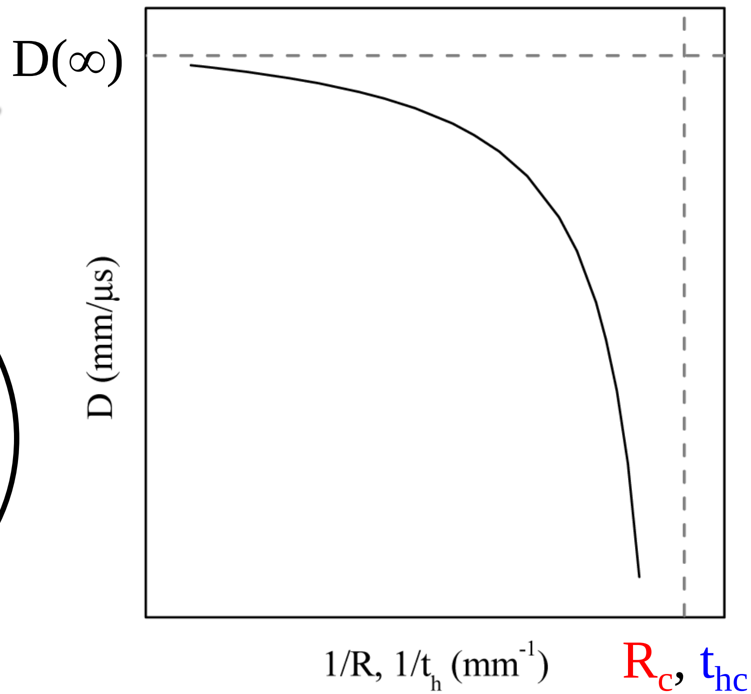
Data Analysis Is Conducted Using the Standard Critical Diameter Form

$$D(R) = D(\infty)\left[1 - \frac{1}{R}\left(\frac{A}{1 - R_c\frac{1}{R}}\right)\right]$$

$$D(R) = D(\infty)\left[1 - \frac{1}{t_h}\left(\frac{A}{1 - t_{hc}\frac{1}{t_h}}\right)\right]$$

Critical diameter configuration.





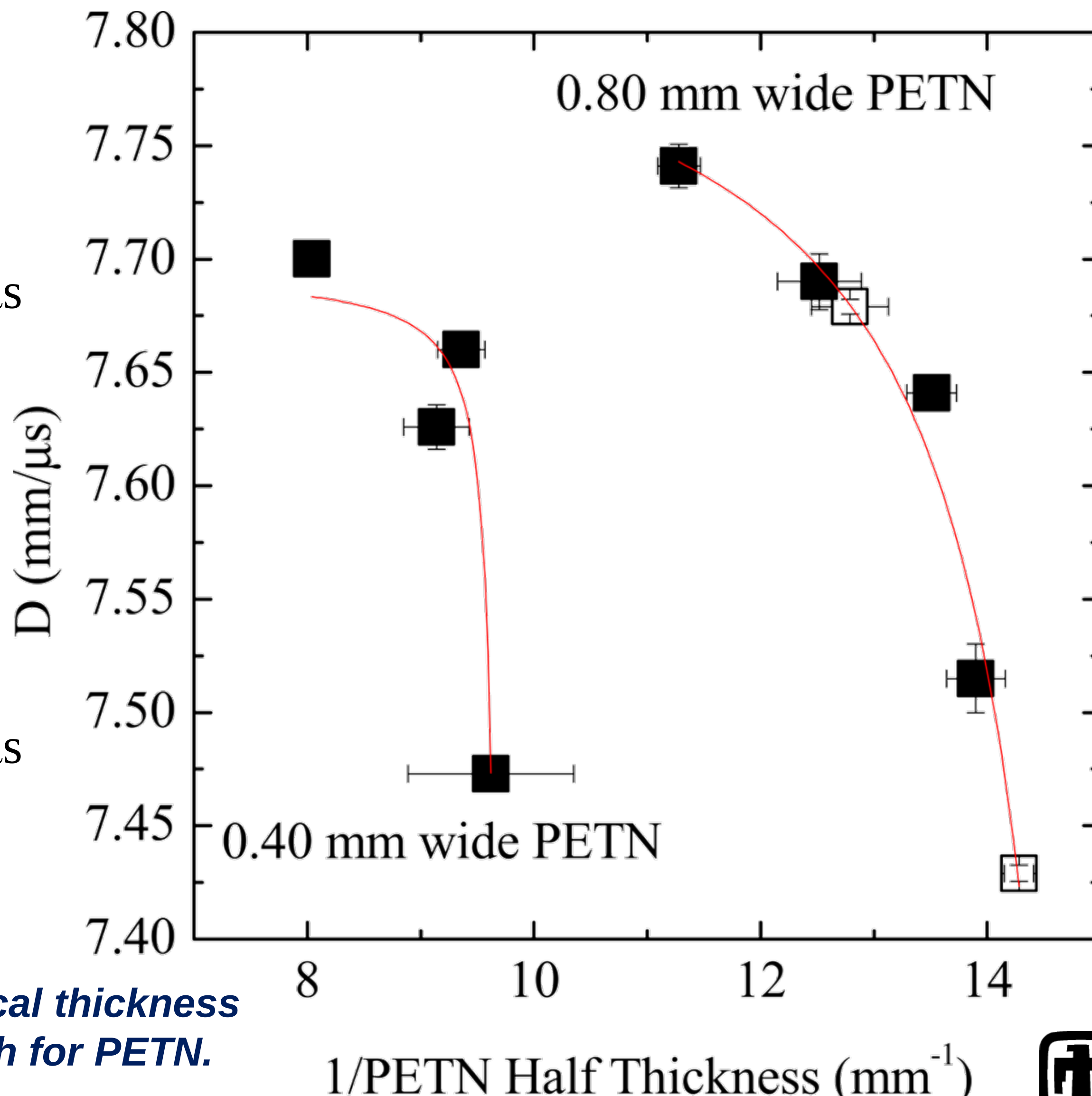
Critical thickness configuration.

Campbell, A.W. and Engelke, R., "The Diameter Effect in High-Density Heterogeneous Explosives," *6th Symposium (International) on Detonation, Coronado, CA, August 24–27, 1976.*

Critical Thickness Measured for PETN

0.80 mm wide PETN
D(∞) = 7.82   0.05 mm/μs
t_c = 0.131   0.003 mm

0.40 mm wide PETN
D(∞) = 7.70   0.05 mm/μs
t_c = 0.206   0.001 mm



Critical thickness graph for PETN.

Conclusions

- Physical vapor deposition used to produce high-density PETN samples with small geometries
- Density and surface roughness change with film thickness
- Critical thickness shows dependence on PETN film width
  - 0.206 mm (0.40 mm wide)
  - 0.131 mm (0.80 mm wide)
- Critical thickness less than 0.13 mm for films at "infinite width"
- Unsteady light intensity in thin, narrow films

- Funding: Joint Department of Defense/Department of Energy Munitions Technology Development Program
- Thanks to: Marc Basiliere, Rosa Montoya, Adrian Casias, David Saiz, Thomas Gutierrez , M. Barry Ritchey

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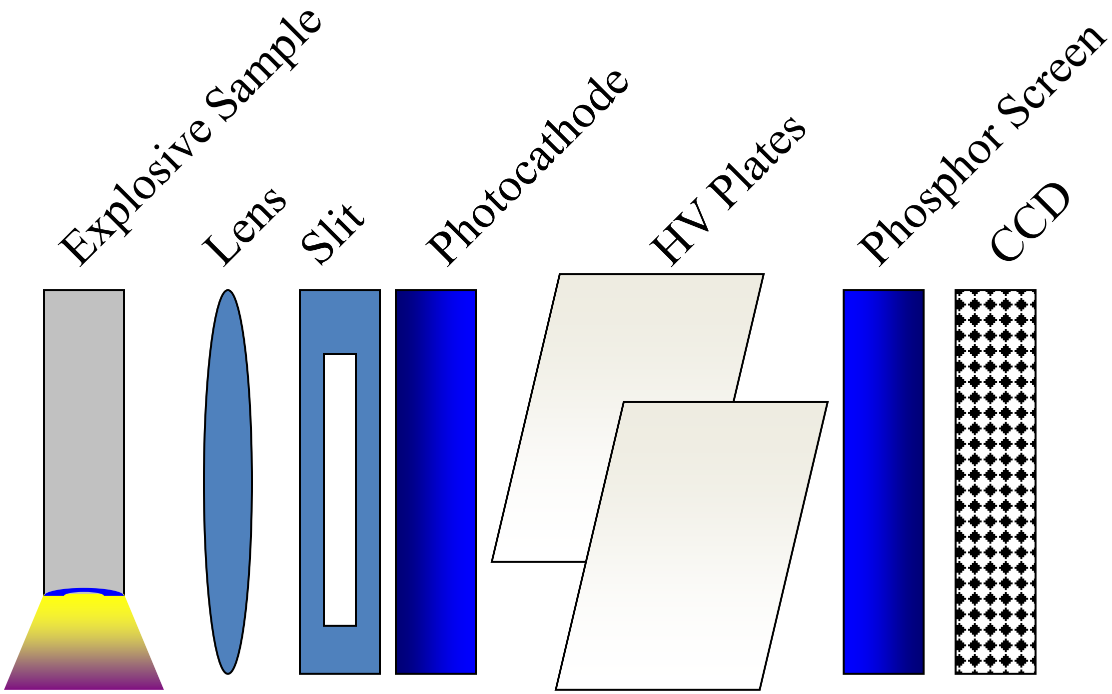
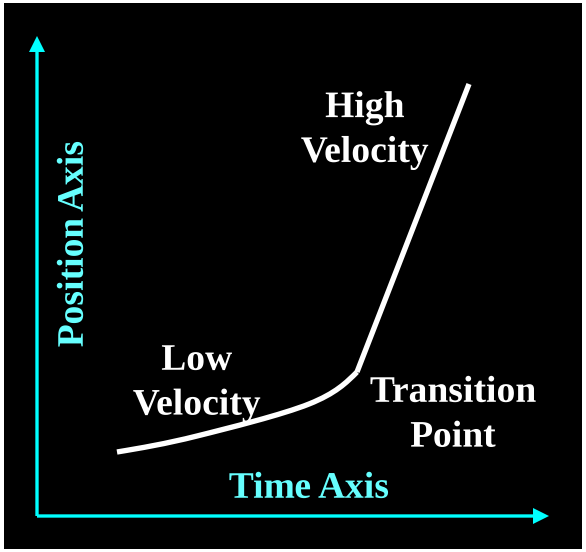
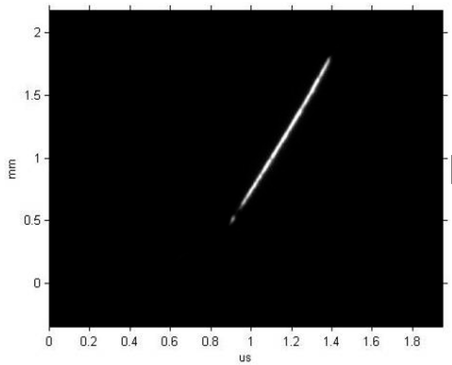
Streak Camera and Analysis

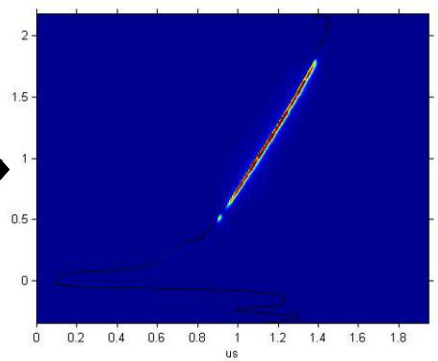


Streak camera block diagram.

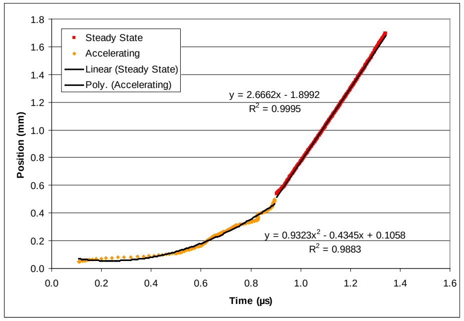




Raw image.



Edge detection.



x-t plot.

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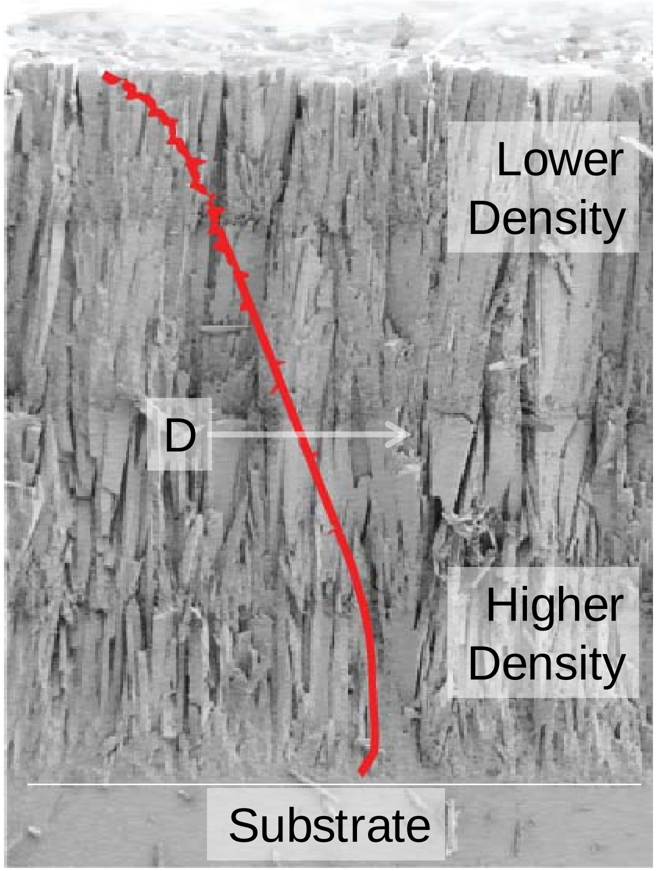
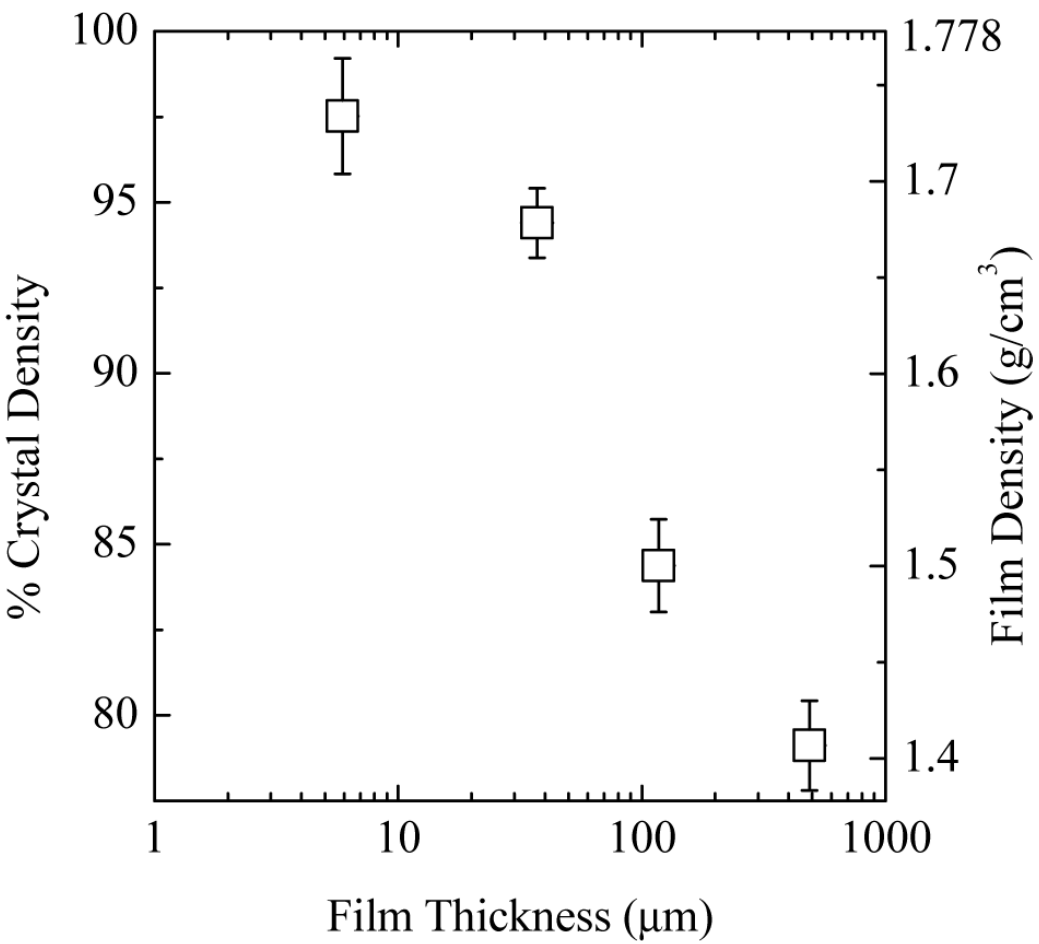
Density Decreases with Increasing Film Thickness

- **Density of deposited PETN at substrate interface is very high**
- **Self-shadowing as film grows results in voids**





Density of deposited PETN film versus film thickness.

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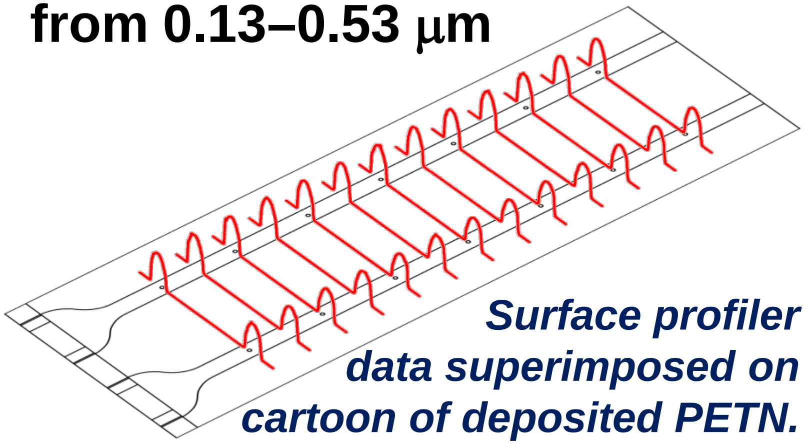
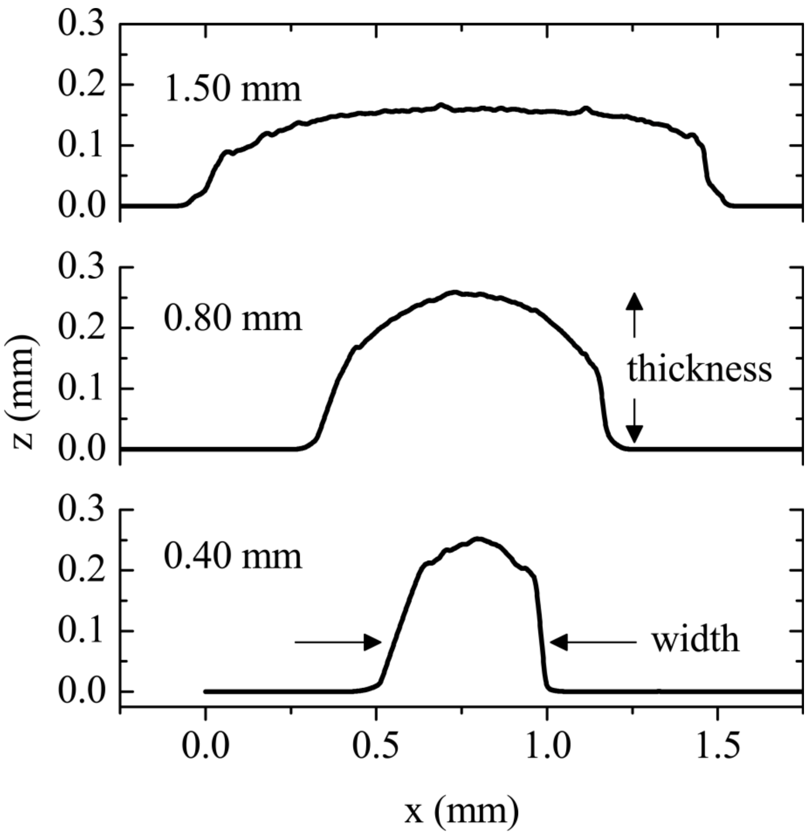
Surface Profiler Measurements

- Stylus surface profiler used for measurement
- Center 100 μm of scan defined as film thickness
- Each line thickness reported as average of 13 scans across film
- Film thicknesses varied from 0.13–0.53 μm



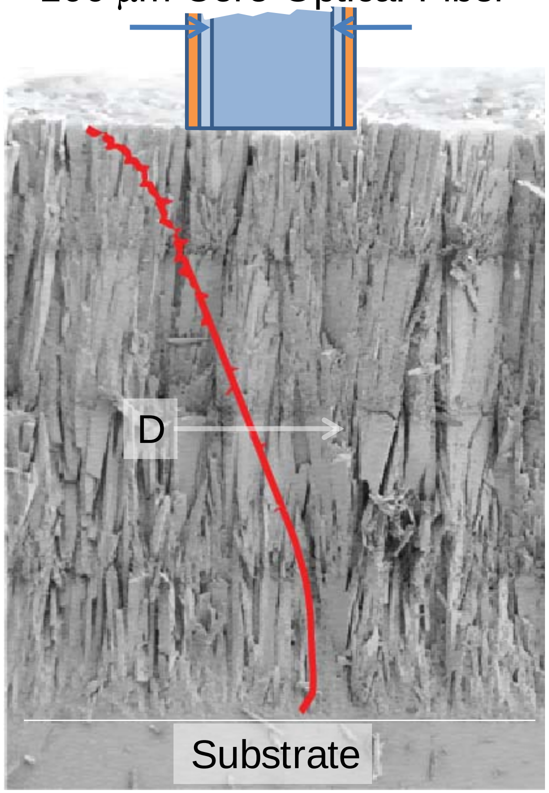
Surface profiler data superimposed on cartoon of deposited PETN.

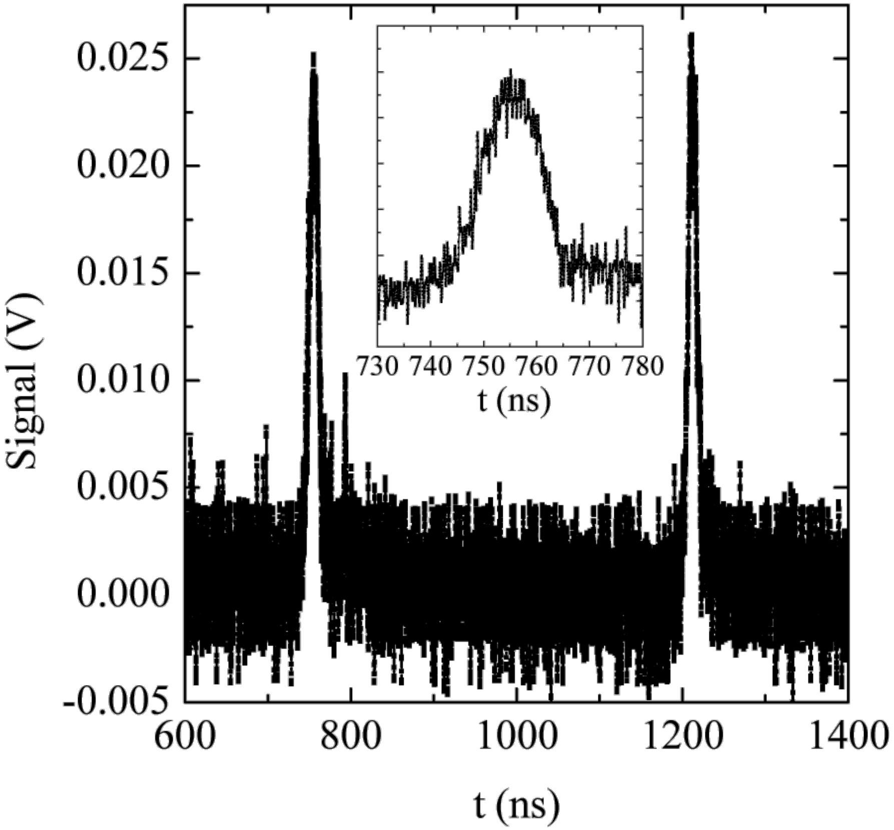


Surface profiler single line scans of 0.40, 0.80, and 1.50 mm wide vapor-deposited PETN films.

Optical Fiber Probe Signal Has Fast Rise and Fall Times

- **Full-width-half-maximum = 13 and 11 ns for graph at right**
- **0.1 mm core / 7.5 mm/μs = 13 ns**

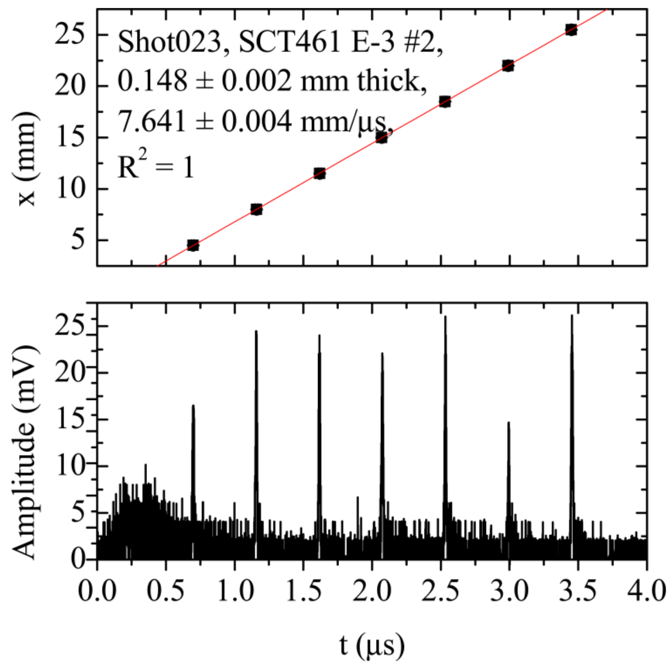


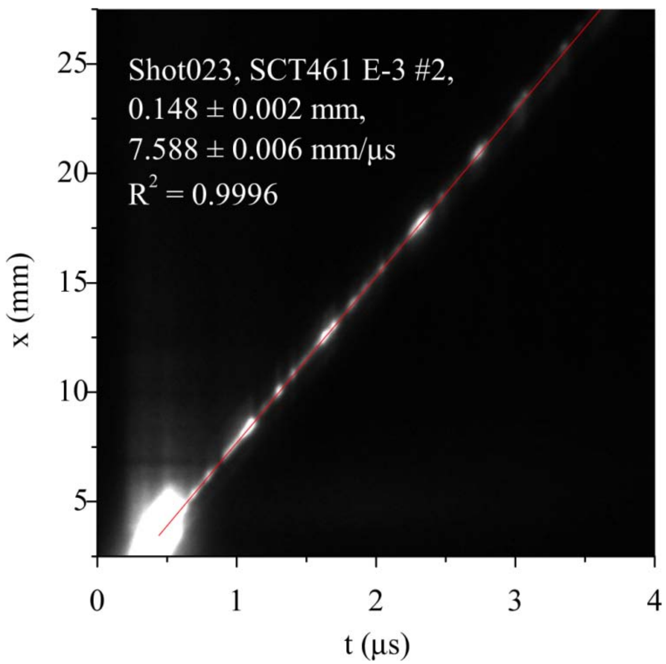


Optical signal from two of seven fibers used to measure detonation light. An expanded view of the left pulse is shown in the inset.

Optical Fiber Probe Velocity Correlates Well with Streak Camera

- Optical fiber probe and streak camera velocities agree to within 1% on multiple experiments
- Analysis of optical fiber probe data is less subjective
- When both are available, optical fiber data are used





Optical fiber data and streak camera data from the same experiment.

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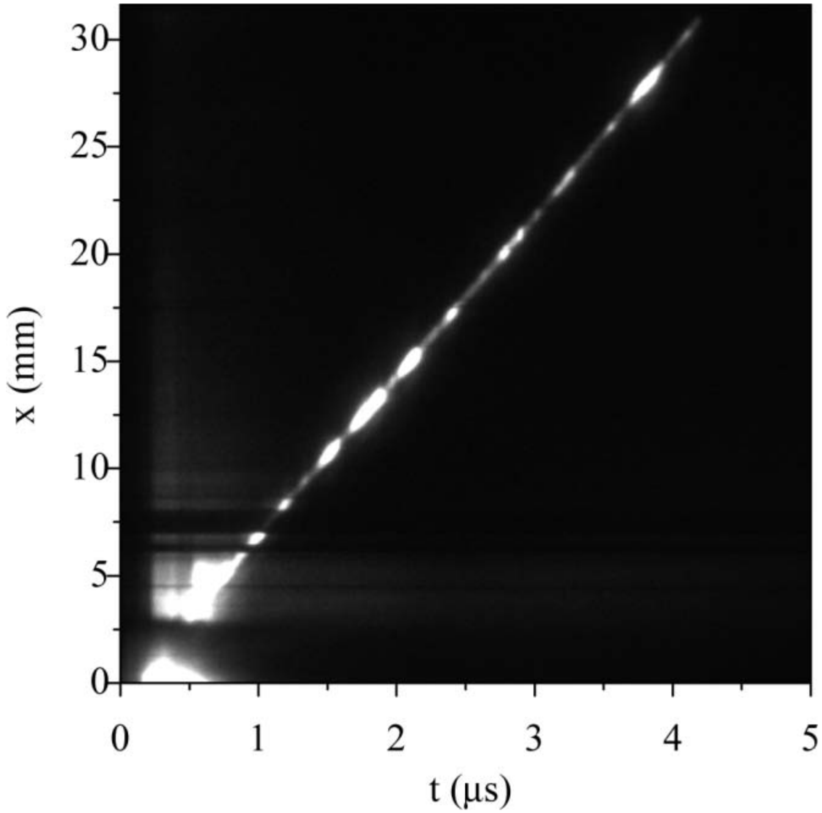
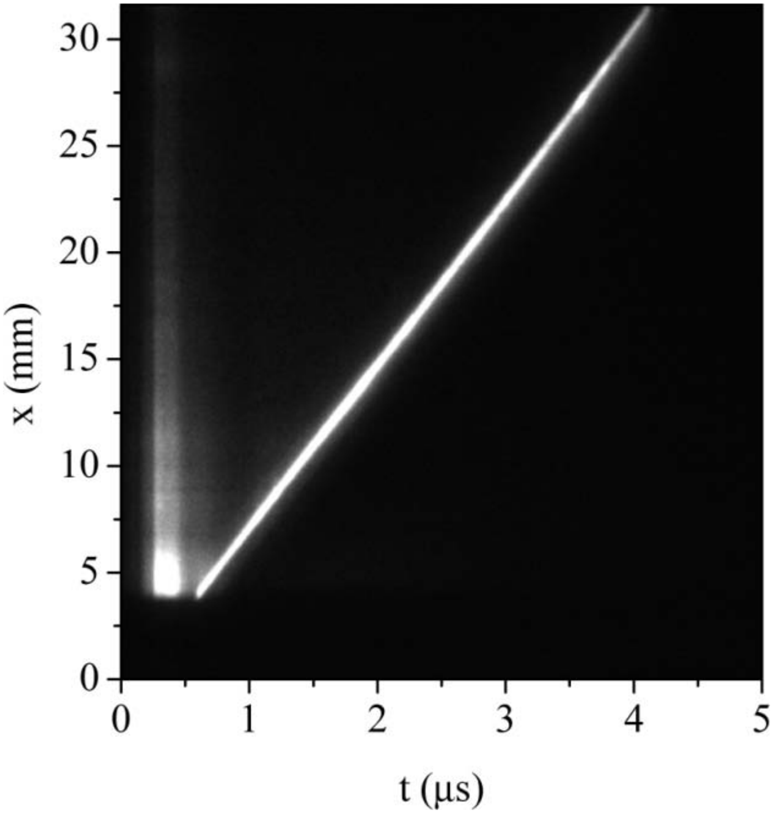
Evidence of Unsteady Detonation Near the Critical Thickness

- Thinner PETN films produced fluctuations in streak camera light intensity
- Not observed in thick films or thin films that were also wide
- No effect on velocity stability



Unsteady light intensity,
0.14   0.001 mm thick, 0.80 mm wide,
7.429   0.004 mm/μs, Shot018.



Steady light intensity,
0.132   0.004 mm thick, 1.00 mm wide,
7.691   0.002 mm/μs, Shot036.