Non-Linear Optical Material Characterization using Double Pump-Probe Spectroscopy

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Double Pump-Probe Collaboration

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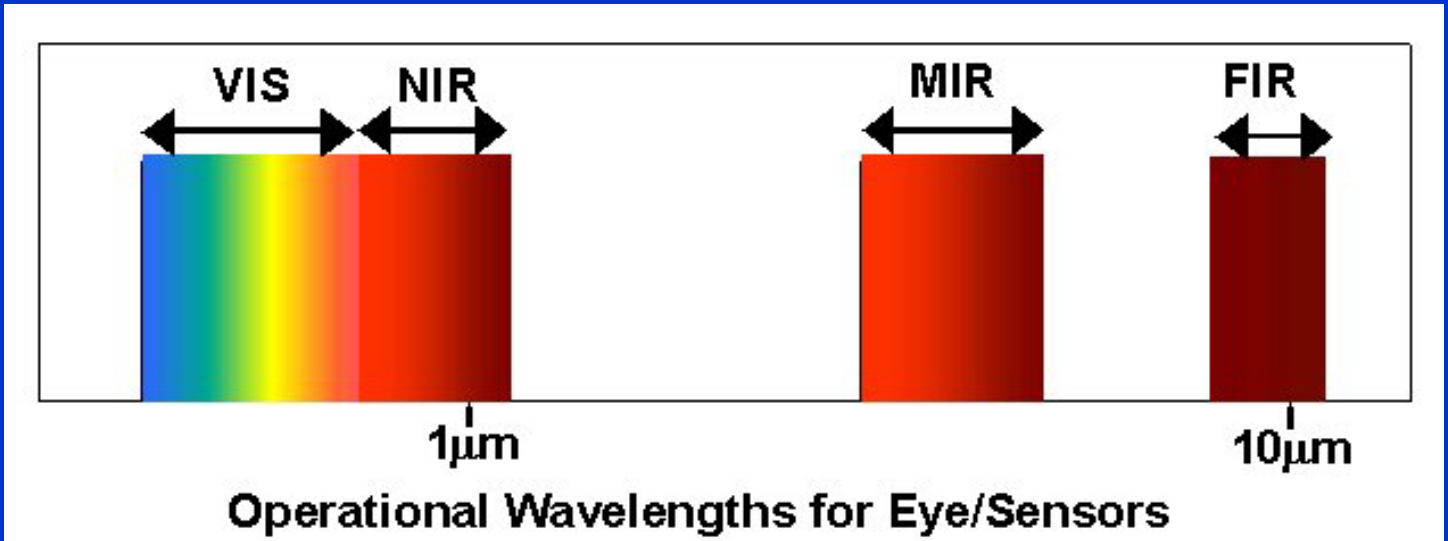
2LT's Richard Burgess, Dennis Mackin *and* **Joshua Hunter**

Outline

- Threat, Army Systems, Current Protection Levels
- General requirements
- Theory
- Experimentation at Adelphi
- Experimentation at USMA
- Questions

Army-Wide Sensor Protection Needs



- Direct View Optical (DVO) systems including soldiers' eyes
- Thermal Viewers
- I^2 Devices
- Camera Systems

Most of the optical spectrum is UNPROTECTED

DOD Laser Eye Protection Approaches

Fielded

Fixed Line Protection

Absorbing Dyes
(fielded)

Reflective Filters
(limited fielding)
- Rugates
- Holograms
- Dielectric Stacks

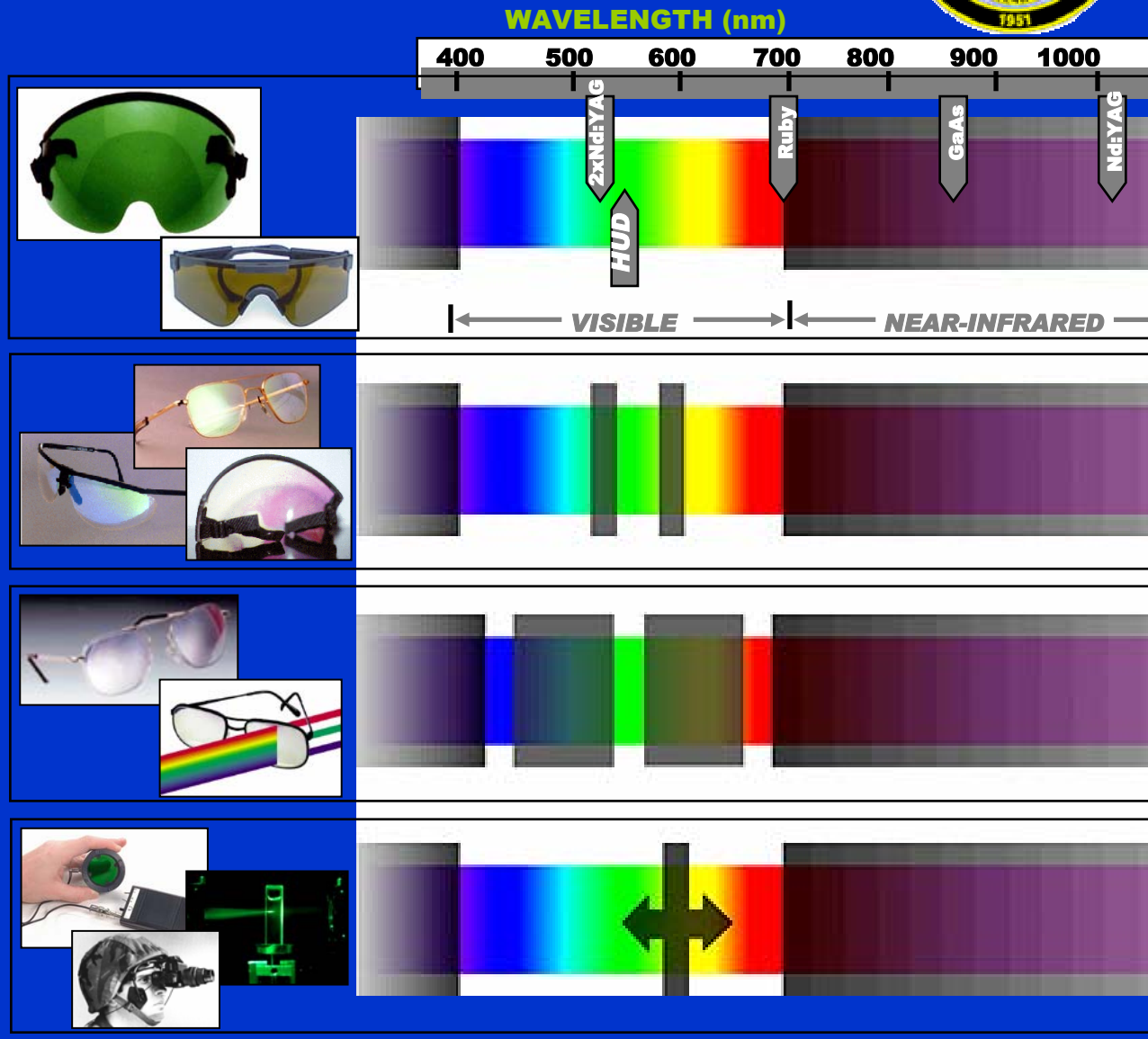
Development

Tristimulus Filters
- Combination of Dyes & Reflective

Agile Protection

Tunable Filters

Optical Limiters*



* (laser light initiates material response)

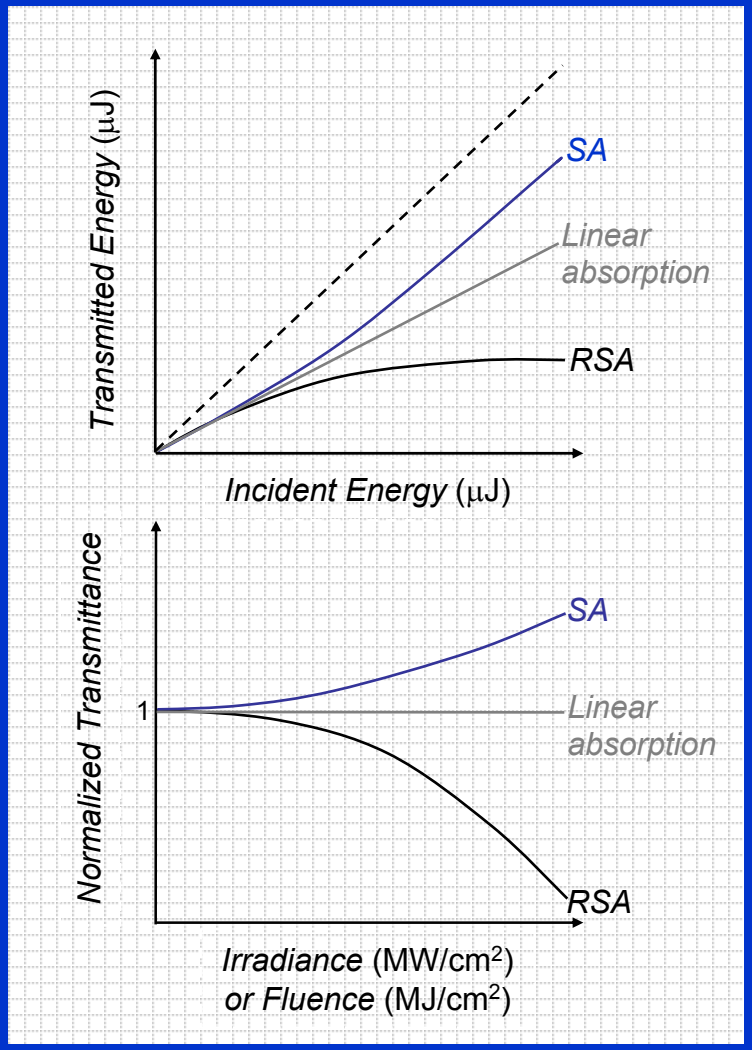
General Requirements for Damage Protection

- Bandwidth
- Optical Density
- Broad Temporal Response
- Dynamic Range
- Damage Threshold
- High Linear Transmission
- Color
- Protection Over Entire FOV of System
- Long Shelf-life, Rugged, Low Cost, Environmentally Stable, Light Weight

Pulse width Protection Requirements

- Currently concentrating in ns regime
- Future will bring wider array of temporal threats that we can not ignore
- Material response time must be significantly faster than the pulse to insure leading edge of pulse does not cause damage.

Theory of Optical Limiters



- Most materials exhibit **saturable absorption** (SA).

  Fraction of the incident energy absorbed by the material **decreases** at high intensity or fluence.

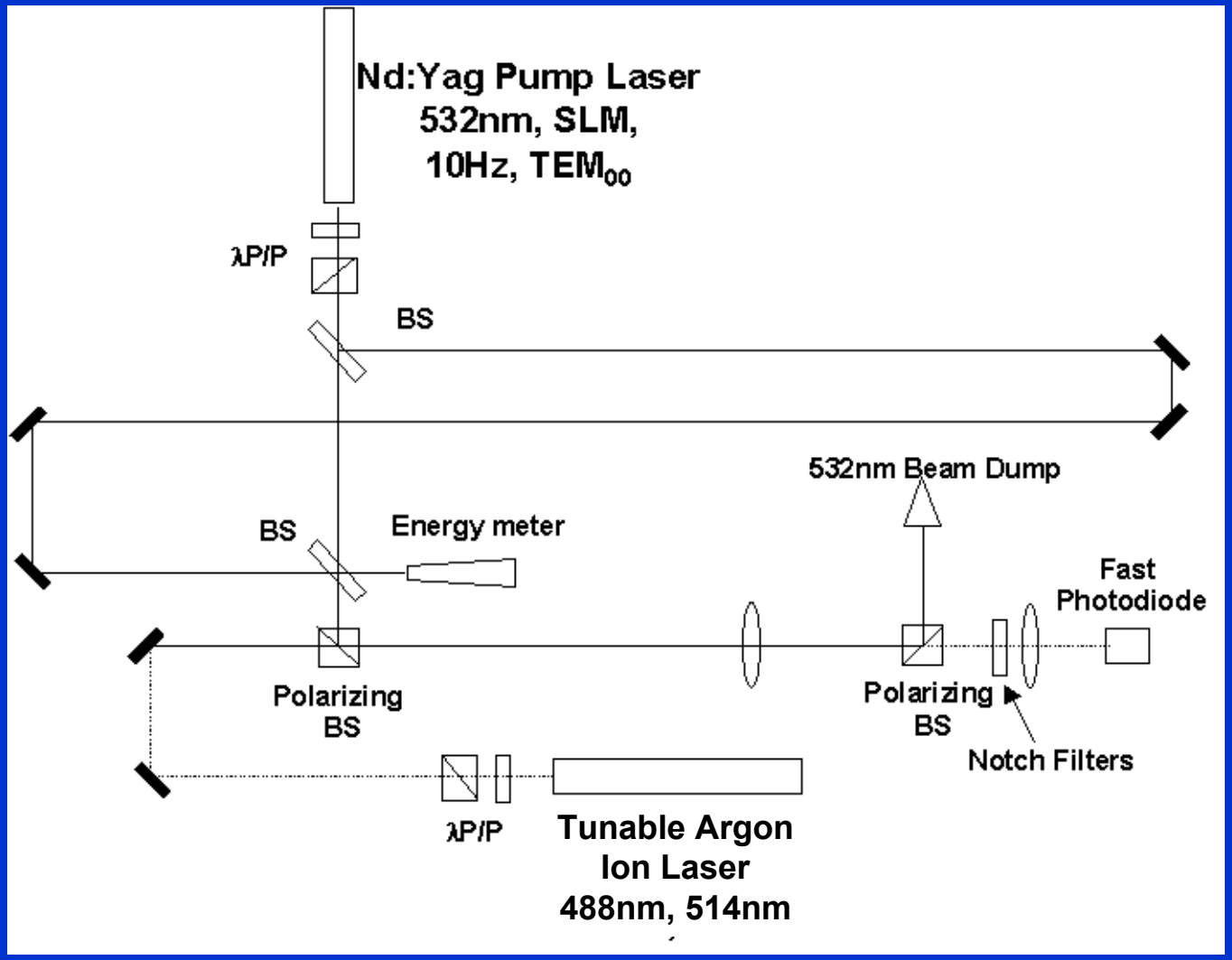
- Materials exhibiting **reverse saturable absorption** (RSA) are important for optical limiting.

  Fraction of the incident energy absorbed by the material **increases** at high intensity or fluence.

DPP Spectroscopy Technique

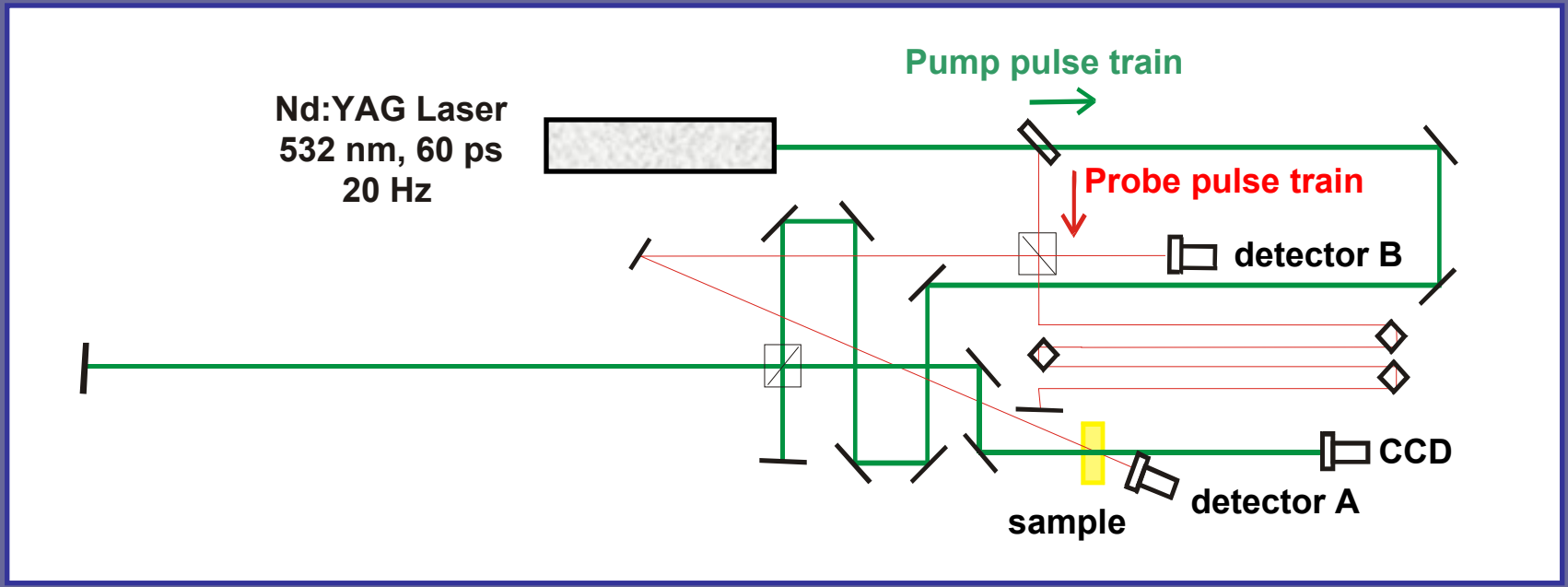
- Two strong “non-linear” pulses initiate nonlinear response in material of interest
- “Weak” probe pulse measures change in differential transmission

ns Double Pump/CW Probe



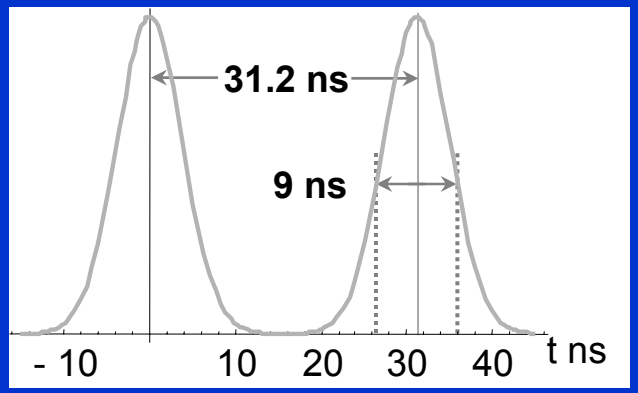
* K.J. McEwan, et. Al., Mat. Res. Soc. Symp. Proc., vol 597, pp 395-406, 2000

USMA Picosecond Double Pump-Probe Experimental Set-up



Complementary double pump-probe experiments have been established at ARL and USMA

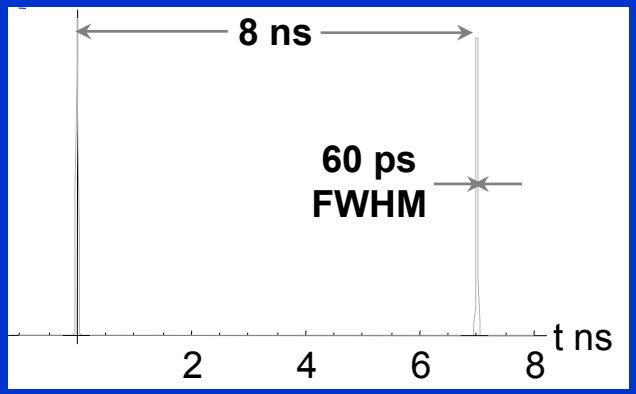
ARL



- ARL experiment employs a long (9-ns) pump pulses with a CW probe beam.

Experiment based on: K. J. McEwan, G. Bourhill, J. M. Robertson, and H. L. Anderson, J. Nonlinear Opt. Phys. Mat. 9(4), 451-568 (2000).

USMA



- USMA set-up uses short (60-ps) pulses for both pump and probe.

Experiment based on: S. N. R. Swatton, K. R. Welford, R. C. Hollins, and J. R. Sambles, Appl. Phys. Lett. 71(1), 10-12 (1997).

Continuous wave and pulsed probes confer different advantages

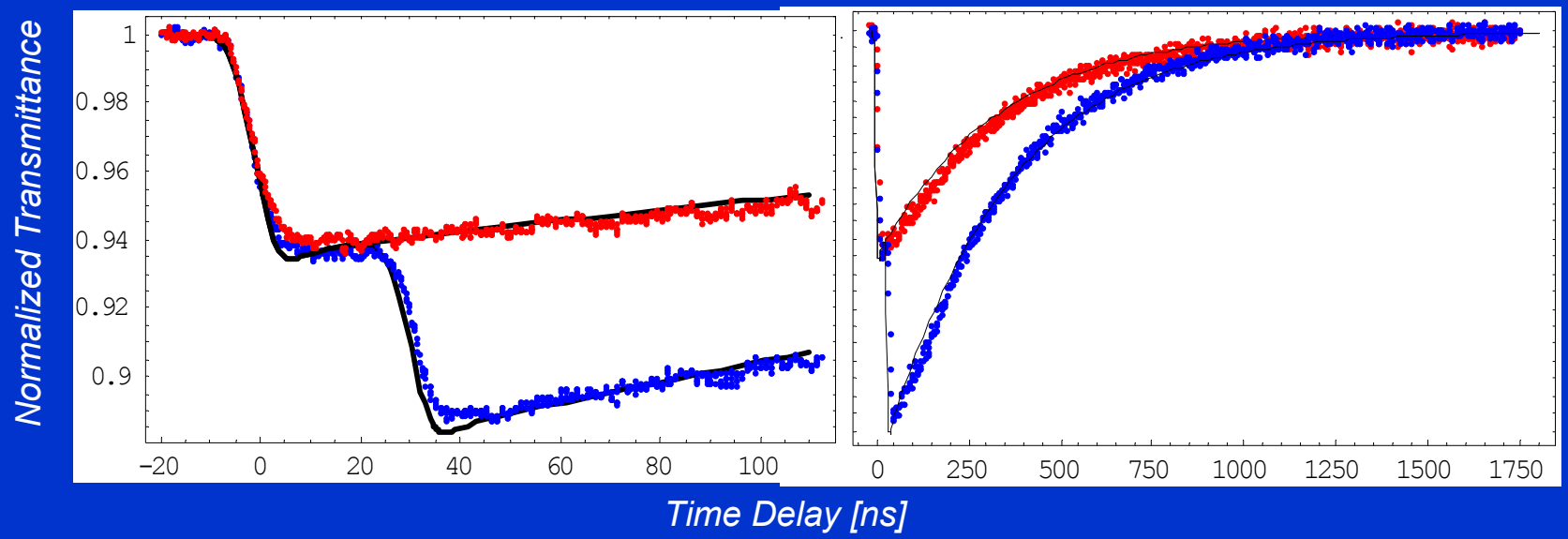
ARL

- CW probe at 514 nm
  - Speeds data acquisition, allows sample to be inter-rogated for 1000s of ns.
  - Pump and probe beams follow collinear paths through sample; beam overlap is not an issue.
  - Excited state absorption cross-sections at pump and probe wavelengths will, in general, differ.

USMA

- Pulsed probe at 532 nm
  - Probe beam traverses sample at a small angle to pump; beam overlap may vary slightly with time delay.
  - Accessible probe time delays limited by translation stage length to ~15 ns.
  - Pump and probe have same wavelength, so the variation of absorption cross-section with wavelength is not an issue.
  - Pico-second resolution offers more detail of the material response.

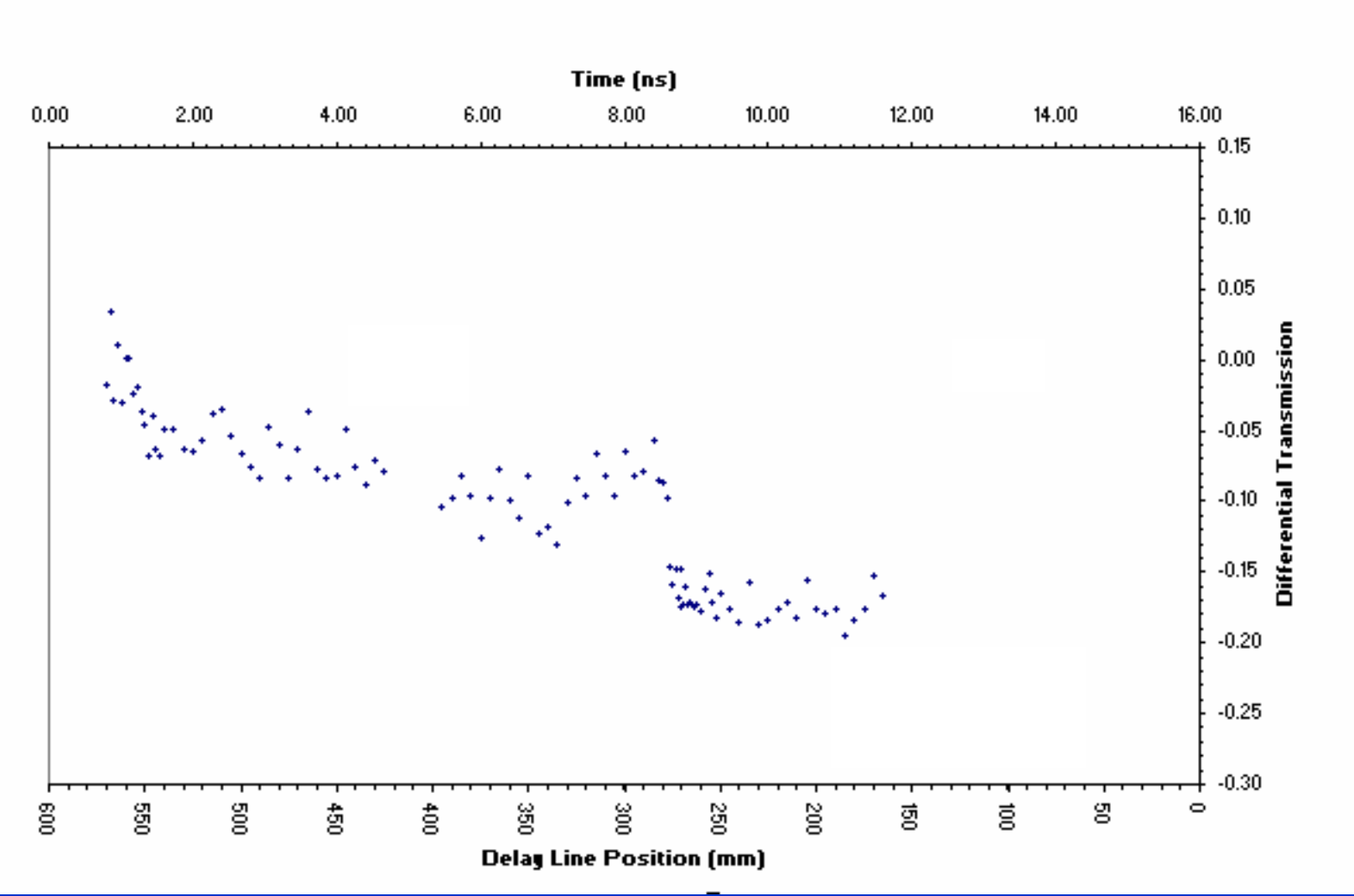
ns Theory vs. Experiment: Single and Double Pump-Probe Results for C_{60}



Normalized transmittance of a 7-mW CW probe by a 1-mm sample of 4.25-mM C_{60} in the presence of 9-ns FWHM, 2.85 μJ pump pulses, focused to a spot of radius 150 μm.

- In **black** are the predictions of the theoretical **model**.
- In **red** are data from an experiment employing a **single pump pulse**.
- In **blue** data from an experiment employing **two pump pulses** with a peak-to-peak separation of 31.2 ns.

ps Theory vs. Experiment: Double Pump-Probe Results for SinC



Questions?