Photo-Thermal Interferometric (PTI) Particulate Absorption Monitor

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Introduction

Our project addresses the measurement of radiative forcing of aerosols, specifically energy-related aerosols, which is currently a focus of the Department of Energy (DOE) Atmospheric System Research (ASR) Program (AA Workshop, 2016). Aerosol particles affect the radiative balance of the earth directly, by scattering and absorbing solar and terrestrial radiation, and indirectly, by acting as cloud condensation nuclei.

Knowledge of the partitioning of solar and terrestrial atmospheric extinction between scattering and absorption is thus critical to the understanding of radiation transport through the atmosphere. At present, the uncertainties in the magnitude of aerosol-induced radiation forcing still pose a current limitation on the accurate quantification of both direct and indirect effects of aerosols on climate. Accurate measurement of the absorption by aerosols in the atmosphere is problematic because of their low magnitudes, as particle absorption far from urban centers may be less than 5 Mm^{-1}. The filter-based instruments which currently are the primary means of measuring absorption, while precise, have issues with accuracy in the presence of high non-BC backgrounds due to inherent issues with their operation and measurement methodology.

We chose to focus on PTI technology for specific reasons:

(1) PTI is one of two proven technologies (photoacoustic spectroscopy - PAS - being the other) that ***directly measure particulate absorption*** and can meet the stringent US DOE stated requirements (i.e., sensitivity goal of 1 Mm^{-1} (2σ at 10-s averaging) with a response time of 5 s);

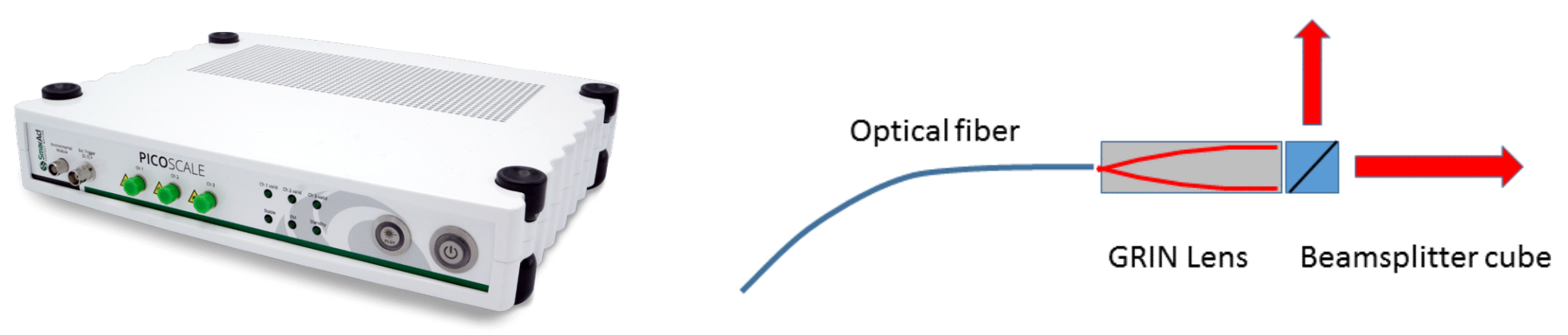
(2) PTI technology provides several potential advantages compared to PAS technology, including the ***freedom to operate over a wide range of frequencies*** that minimize overlap with environmental noise and that enable investigation of particle-dependent heating rates; and,

(3) PTI technology has not been developed to the extent of PAS technology (e.g., multiple commercial PAS instruments are currently on the market, whereas there is only one recently developed commercial PTI instrument).

Instrument Design

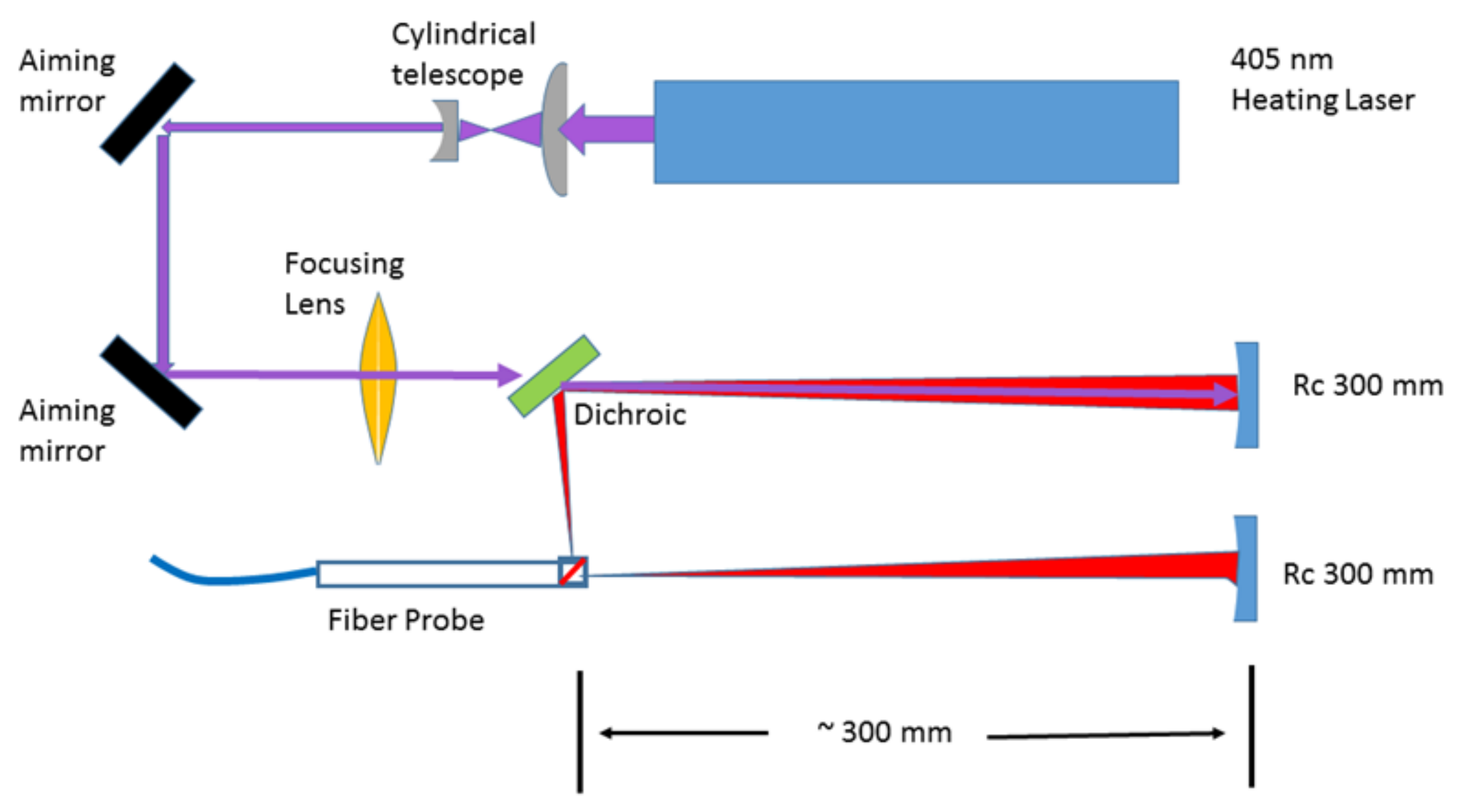
Interferometer: We chose a novel approach by testing the potential of using a commercial fiber-optic Michaelson Interferometer (MI), the Picoscale, sold by SmarAct. This system represents a new standard in commercially available, highly capable fiber-optic based interferometers for measuring distances, rotations, and vibrations for nanoscale machining. The appeal of using a commercially available interferometer is that it allowed us to focus exclusively on the configuration of the heating laser, optical overlap between the heating laser and interferometer arm, and the sampling configurations.

The Picoscale interferometer operates three separate channels and provides a direct measure of the displacement distance between the two arms of the interferometer on each channel. The electronics and optics are housed in a single enclosure with connections for three fiber optic cables and operated by a proprietary software program. The Picoscale uses a temperature stabilized distributed feedback (DFB) laser diode coherent light source at 1550 nm with 150 µW of power in each interferometer arm. Phase modulating the laser at high frequency generates a set of harmonics of the modulation frequency, which provides quadrature information on the phase shift between paths.



Smar-Act PicoScale Interferometer and fiber optic probe. The probe head is ~4 mm in diameter.

PTI optical design tested during this study.



Advantages:

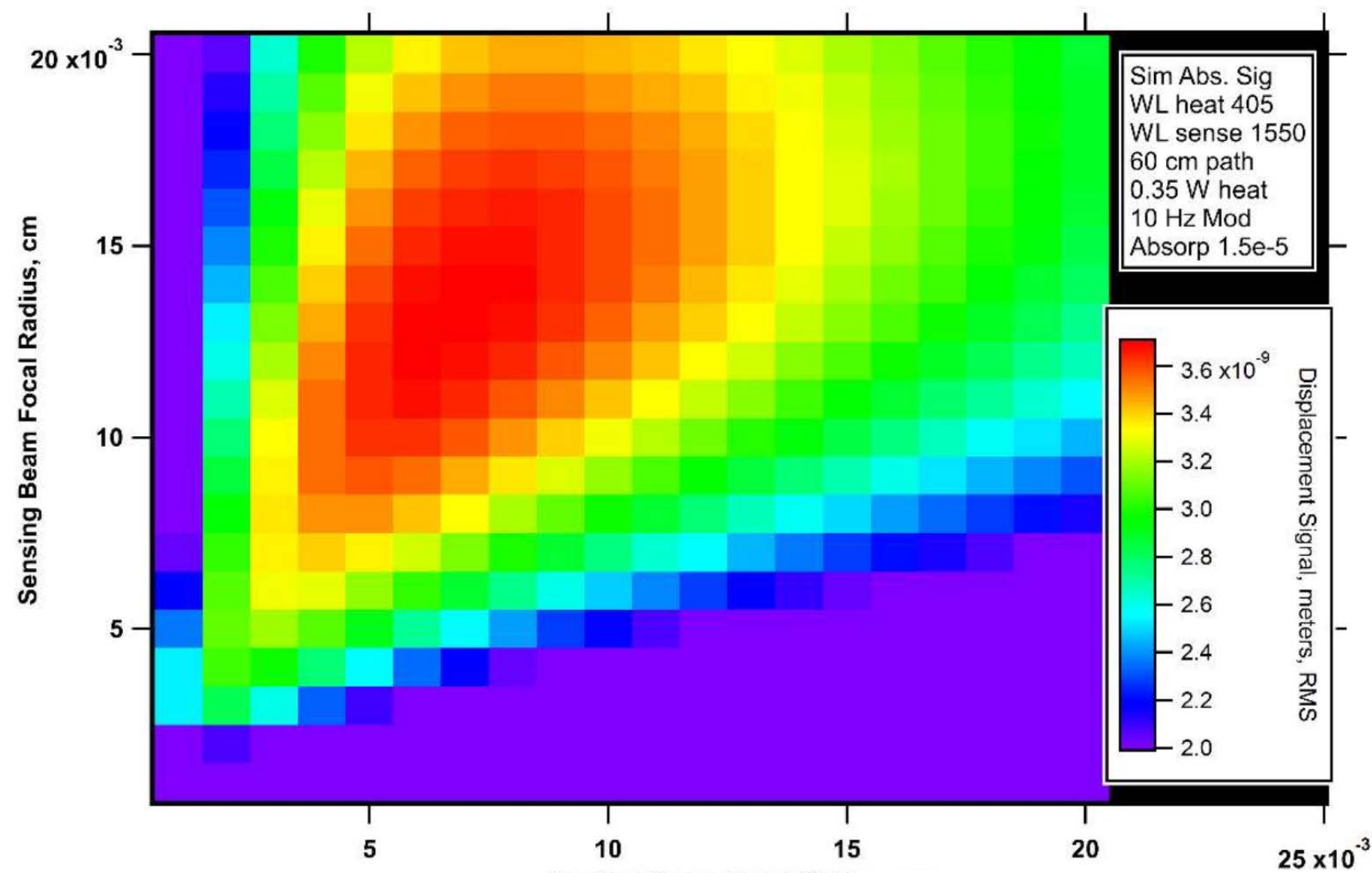
- The interferometer, along with control and calibration software is a commercial product and thus is readily available.
- The two-beam design allows long path lengths with a small length difference, promising high sensitivity and leveraging advantages of common-mode rejection.
- The fiber optic probe allows flexibility in optical system design. Its output beams can be matched into a variety of optical designs with long paths.

Disadvantages:

- The interferometer sensitivity is far less (1-2 orders of magnitude) than the folded-Jamin design in the BNL research-grade PTI system.
- The method of operation and the setup and calibration software are proprietary.
- The optical fiber is sensitive to vibrations and movement.

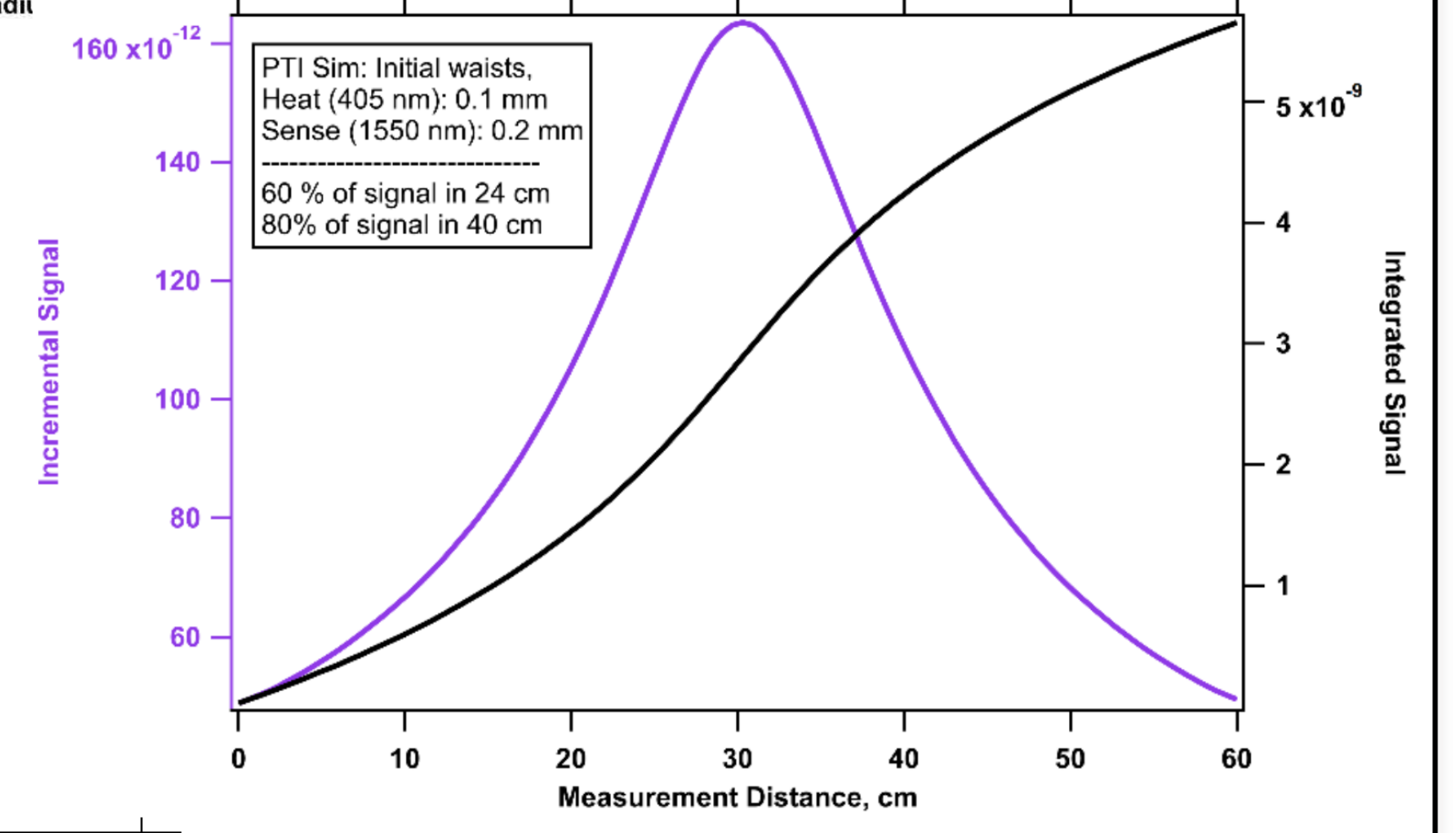
PTI model

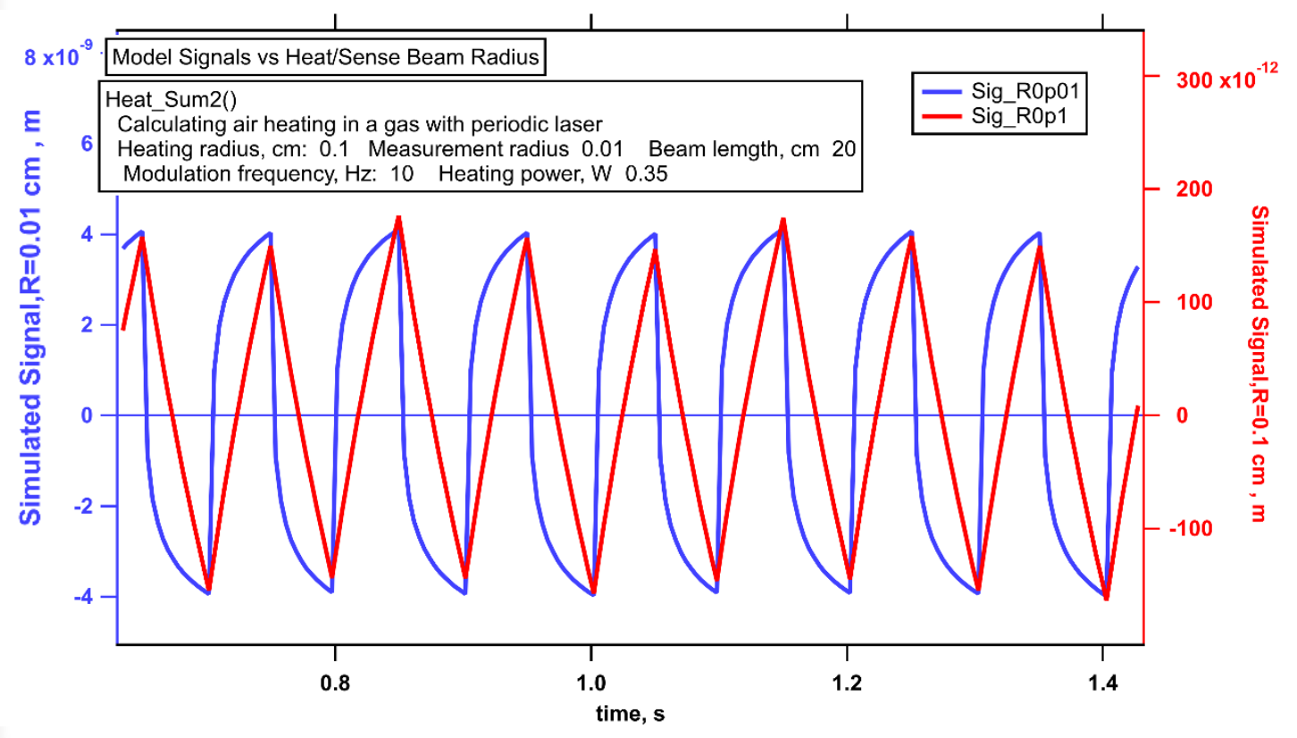
We modeled the heat flow from a single laser-heated particle and from an ensemble of particles that may be treated as a homogeneous absorber, like a gas. In PTI, small absorbing particles are heated by a modulated excitation laser, and the heat is dispersed diffusively into the surrounding air. The rise in temperature of the air produces a small region of reduced number density (i.e., change in refractive index), which is measured by one arm of an interferometer. The heating laser is modulated in time, and the interferometer-measured phase shift is detected at the modulation frequency.



- Model suggests that the maximum PTI signal will be realized when the pump beam is ~0.075 cm and the probe beam is ~0.015 cm.
- BNL PTI is close to these operating conditions

- Incremental signal along the length of the beam, for optimal beam focal radii that maximize PTI signal.
- Enables better design of optical cell, including mirror purges, etc.





- The signal wave shape transitioned from triangular to more rectangular, depending upon heat/sense beam radii.
- Shape also affected by gas absorption compared with mirror heating signals.
- Wave shape dependence on heating hints at potential application to size-resolved absorption measurements

Prototype Results

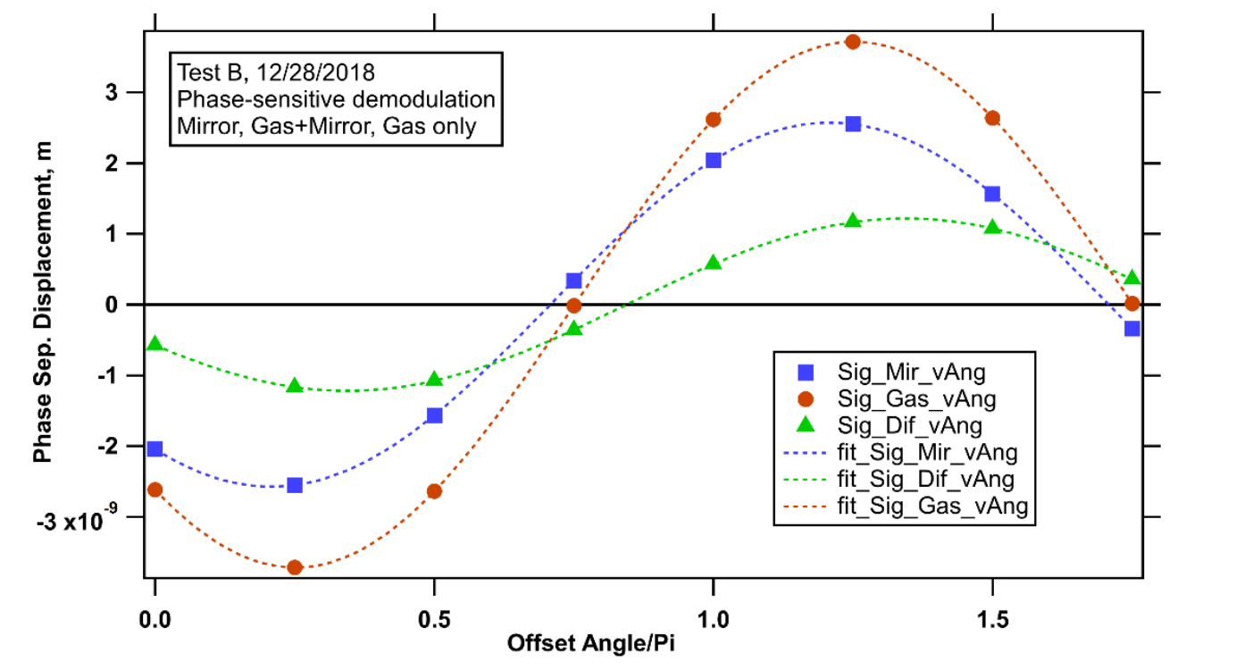
We achieved the following results during this project, using a 405 nm heating laser:

(1) developed a phase sensitive demodulation of the PTI signal to remove mirror heating signal from the gas/particle signals;

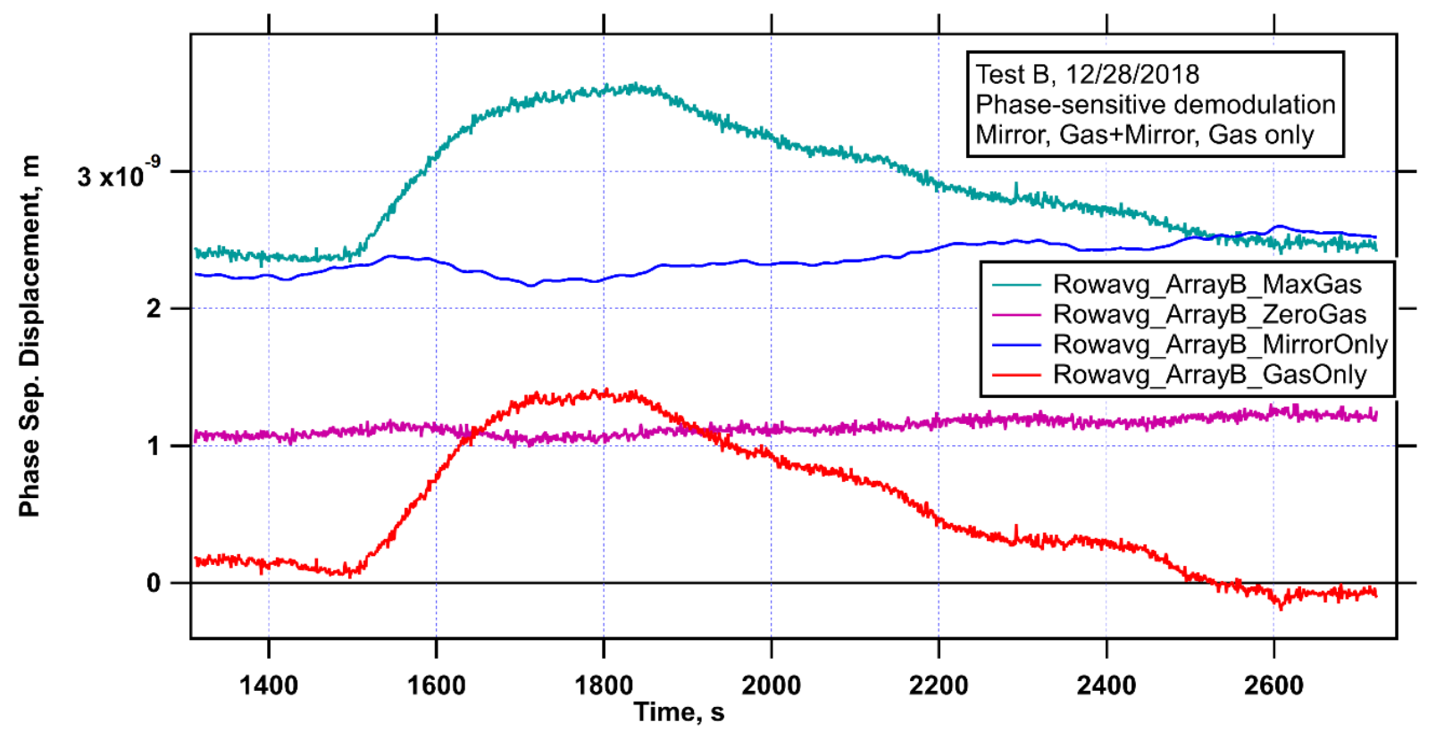
(2) conducted multiple gas-phase NO_2 calibrations to test prototype sensitivity; and,

(3) quantified the noise levels and compared with manufacturer's stated levels.

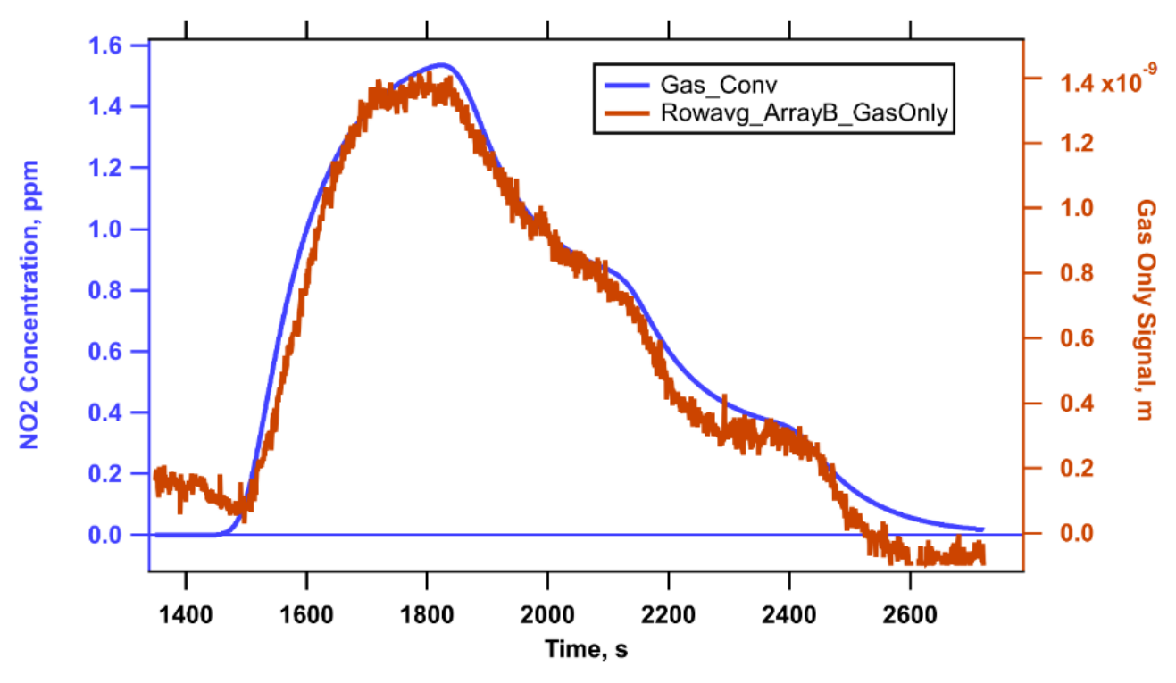
- **Phase sensitive demodulation** of gas-phase NO_2 signal and mirror heating, due to their different rates of heating the surrounding gas.

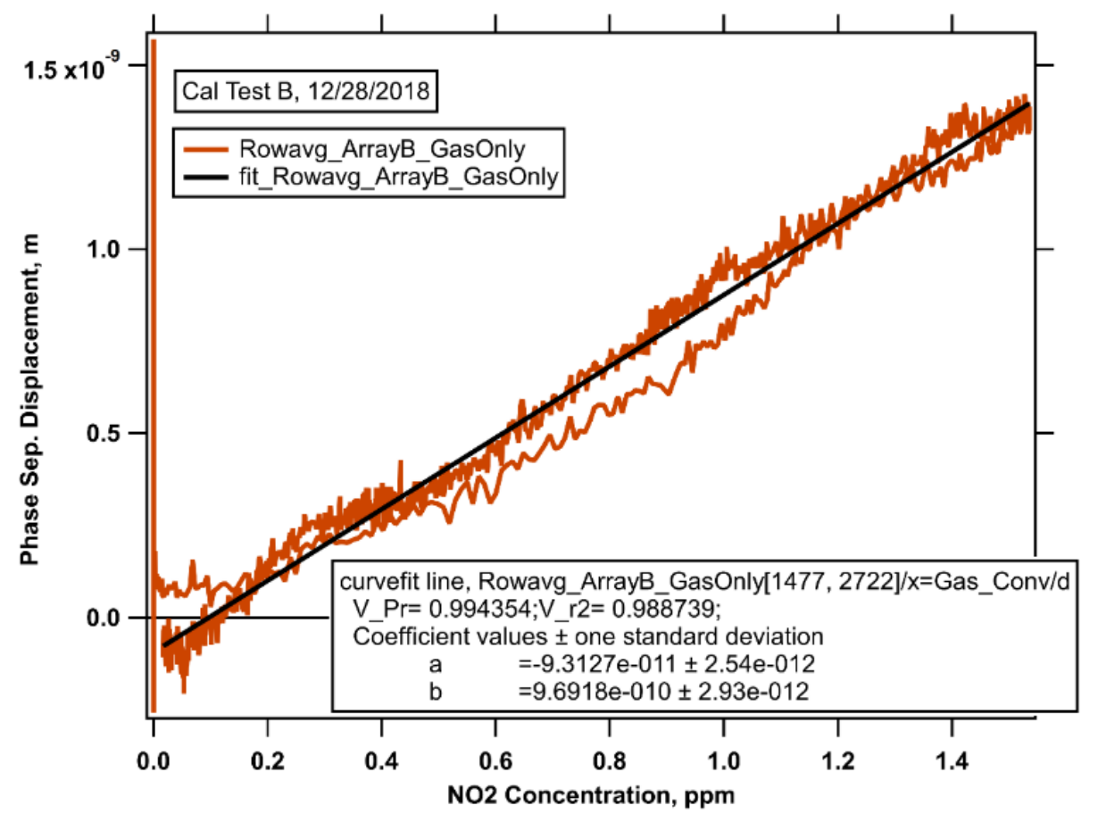


Fit results:	Condition	Amplitude	Phase	Phase/pi
blue	NO2 = 0	3.716e-9	2.360	0.7114
brown	NO2 +Mirror	2.574e-9	2.235	0.7512
green	NO2 only sig.	1.218e-9	2.652	0.8441

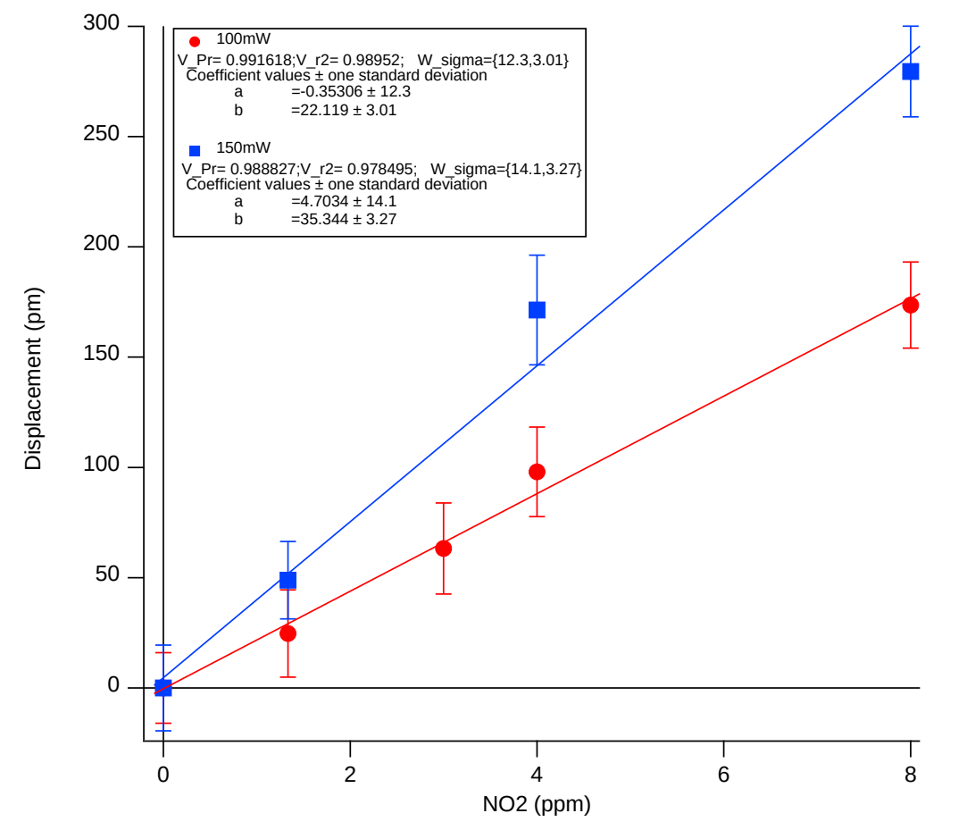


NO_2 calibration





curvefit line, Rowavg_ArrayB_GasOnly[1477, 2722]/x=Gas_Conv/d
V_Pr= 0.994354;V_r2= 0.988739;
Coefficient values ± one standard deviation
a =-9.3127e-011 ± 2.54e-012
b =9.6918e-010 ± 2.93e-012



100mW
V_Pr= 0.991618;V_r2= 0.98952; W_sigma={12.3,3.01}
Coefficient values ± one standard deviation
a =-0.35306 ± 12.3
b =22.119 ± 3.01

150mW
V_Pr= 0.988827;V_r2= 0.978495; W_sigma={14.1,3.27}
Coefficient values ± one standard deviation
a =4.7034 ± 14.1
b =35.344 ± 3.27

- NO_2 gas phase calibration exhibiting a sensitivity of ~1000 pm / ppm NO_2 (1σ, 1s), equivalent to ~1 ppb NO_2/pm.
- NO_2 calibrations done with 405nm heating laser powers of 100mW (red line) and 150 mW (blue line) demonstrate linear dependence on heating laser power.
- Our lowest measurement of spectrally resolved noise (1 σ, 10 Hz, 1 second) is 15 pm.
- This is approximately 50% higher for our current setup than provided by the manufacturer.

BNL PTI

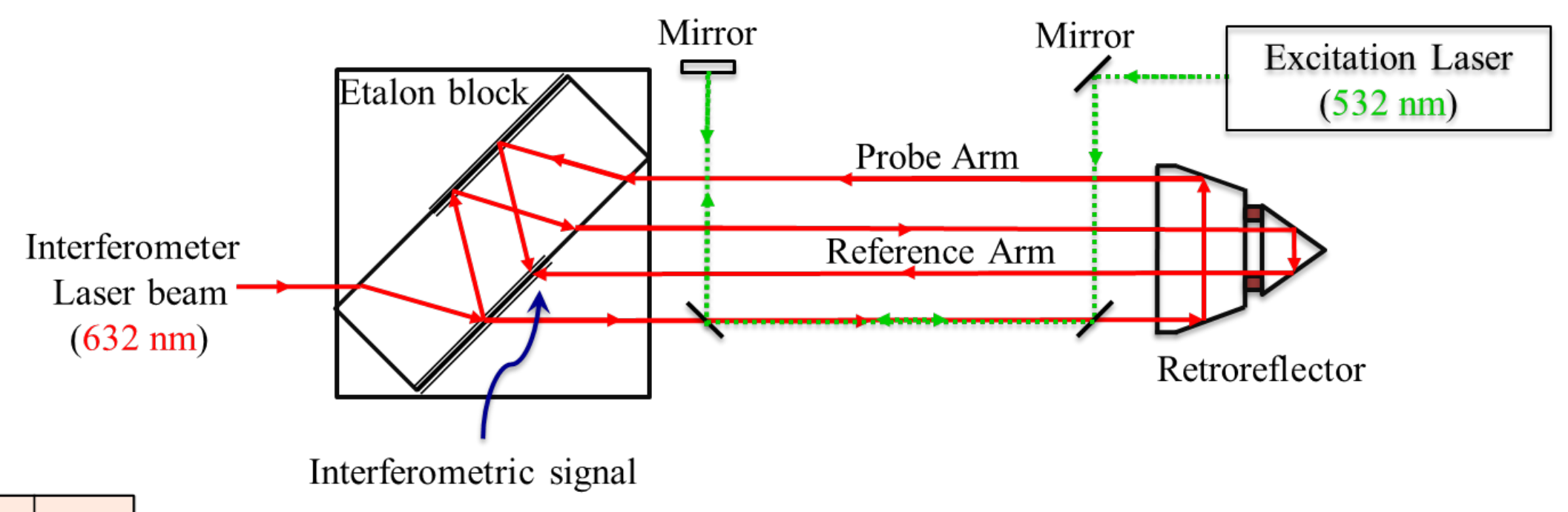
The PTI developed at BNL utilizes a Folded Jamin Interferometer design based on proprietary quadrature technology. During this study, BNL investigated whether the BNL PTI could be

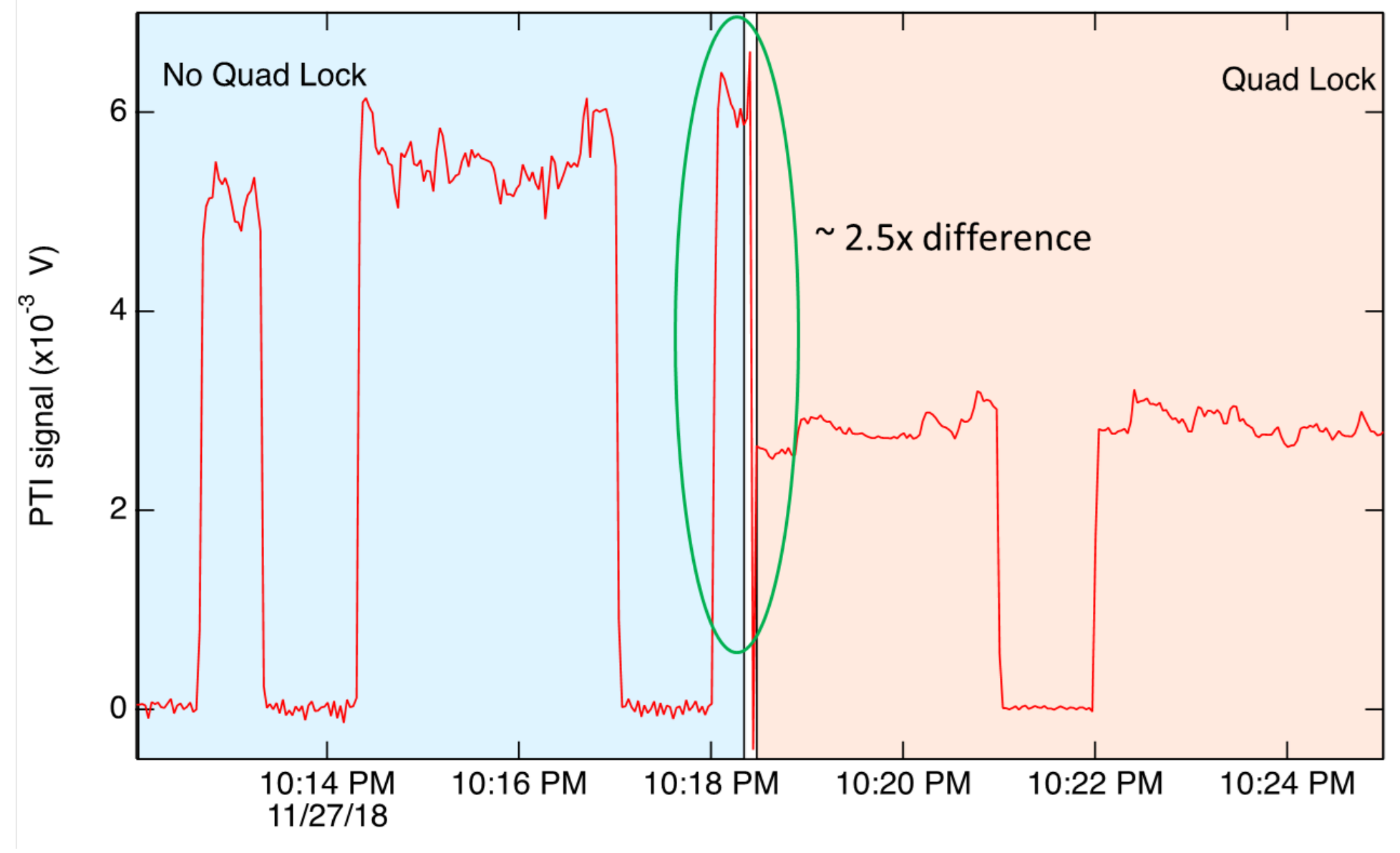
(1) operated with increased overlap between the heating beam and the interferometer arm.

(2) operated in a mode that does not require continuous quadrature lock.

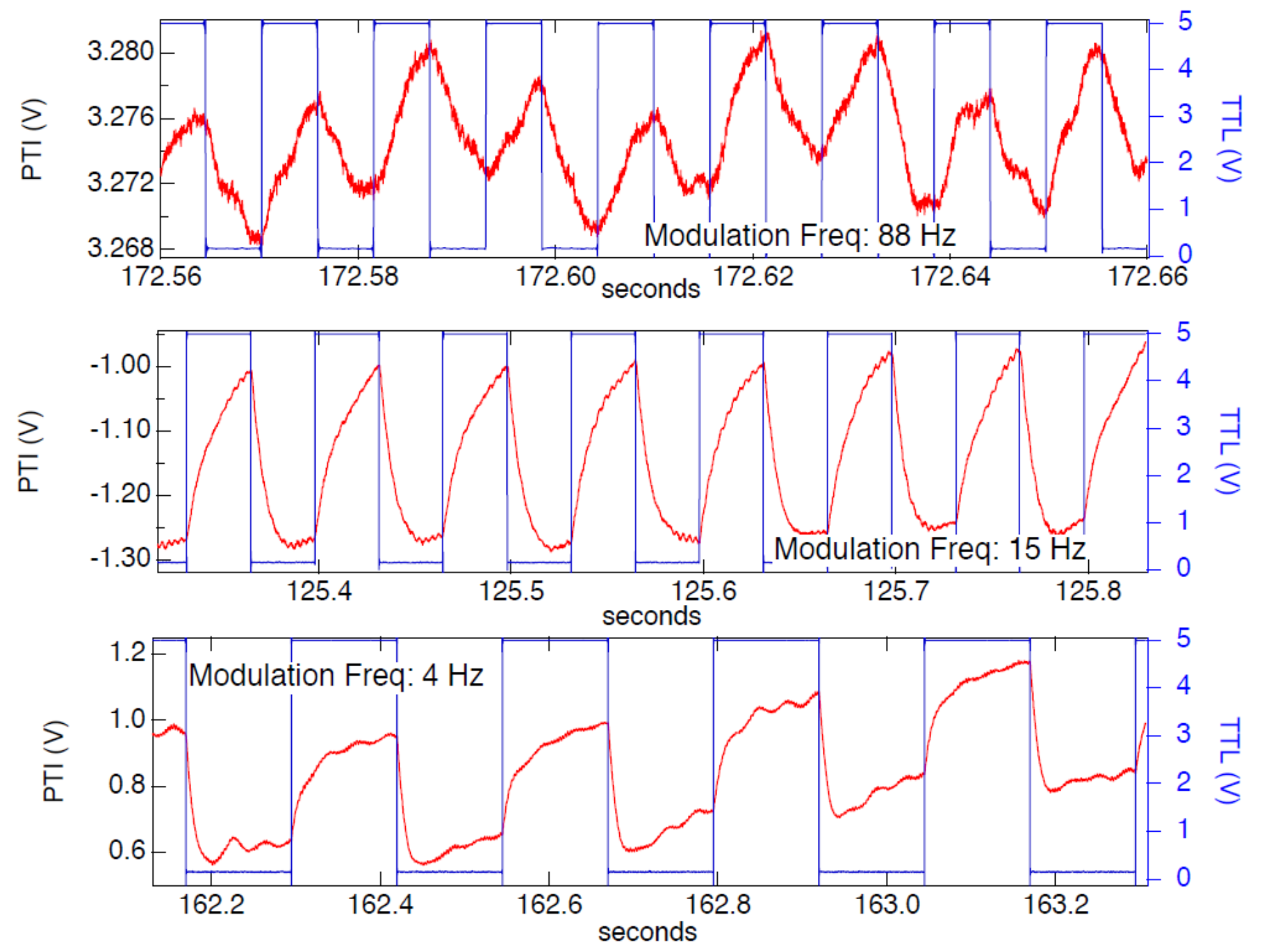
(3) operated at slower frequencies (i.e., higher signal levels) – currently restricted due to a slow RC-filter roll off on the feedback circuit.

- BNL PTI is based on a Folded-Jamin interferometer design
- Maximum common rejection of vibrations
- Preliminary studies using direct overlap of excitation laser and reference arm of the interferometer increased signal levels by a factor of 2-3X.





- Operating without active quadrature lock at typical 88 Hz, enabled an increase of ~2.5X in signal amplitude.
- Proof-of-principal that the PTI could be operated with intermittent quadrature lock to increase signal and reduce dependence upon proprietary circuits.



- Without active quadrature lock, the BNL PTI was able to operate at lower modulation frequencies
- Notice the change in signal shape with frequency – as predicted by the model

Conclusions and Acknowledgement

- Successfully constructed and tested multiple optical configurations of a prototype PTI system built around a commercial interferometer using a 405 nm 1W diode heating laser modulated at 10-100 Hz.
- Calibrated our prototype PTI system with absorbing NO_2 gas, showing a sensitivity of ~1000 pm / ppm NO_2 (1σ, 1s), equivalent to ~1 ppb NO_2/pm.
- Estimate of the noise level for our prototype PTI system is 24 Mm^{-1} in 1 second, or ~8 Mm^{-1} in 10 seconds.
- We demonstrated response to absorbing particle "puffs".
- Developed signal processing methods to investigate PTI signals, including phase demodulation methods that enabled us to separate sources of absorption signals (gas-phase or particle-phase from mirror signals).
- Identified and addressed environment-based acoustic and vibrational noise.
- Initiated the development of a numerical model of the photothermal interferometry (PTI) process for the purposes of interpreting results and illuminating technical development pathways.
- Our focus on the optical configuration encouraged the BNL team to reconsider their research-grade PTI system optical configuration, which led to several significant advances in their design.
- Our combined efforts provide a road map toward a proposed phase II PTI system that will meet the stringent DOE requirements, will be commercially viable, and will provide a new and exciting technique for measuring atmospheric particle-based absorption.

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References: AA Workshop (2016), Absorbing Aerosols Workshop Report, DOE/SC 0185, C. Cappa, R. Kotamarthi, A. Sedlacek, C. Flynn, E. Lewis, A. McComiskey, and N. Riemer, eds.