# Picosecond Time-Resolved Shock Compression of Energetic Materials

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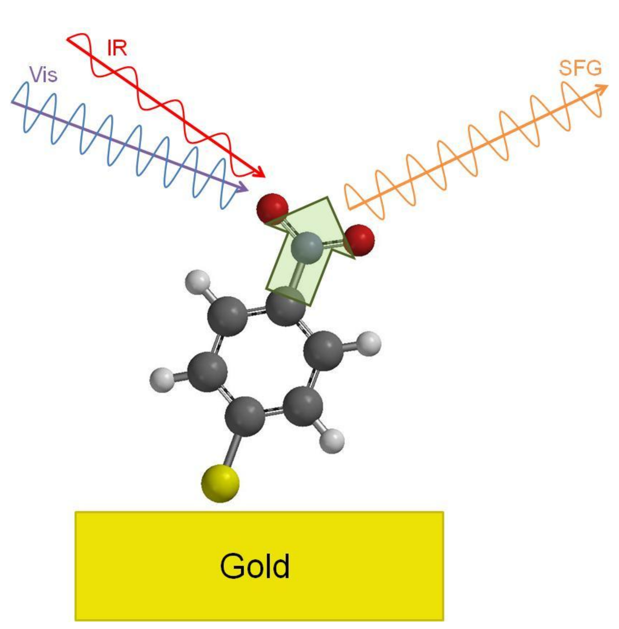
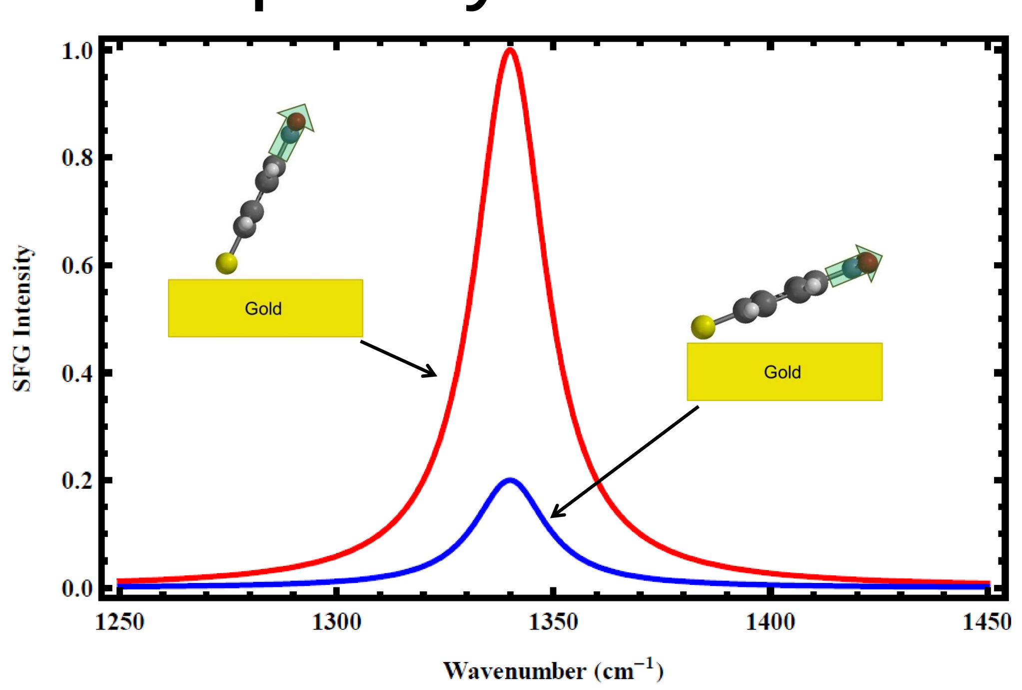
## Motivation

The foremost molecular responses to the extreme pressures and temperatures generated by shock compression are poorly understood. The purpose of this work was to design a simplistic platform to study the shock-induced initiation dynamics of molecular explosives with both high time and space resolution. As a preliminary experiment, ultrafast vibrational sum-frequency generation (SFG) spectroscopy was employed to probe the nitro symmetric stretch of a self-assembled monolayer (SAM) of a molecular explosive simulant. A laser-driven shock wave was propagated into the SAM, and dynamics of the molecules were monitored through variations in the SFG signal. Due to temporal resolution being limited by the shock transit time through the substrate, only nanometer scale thick samples could be utilized. The ability to visualize thin (~5-10 nm) layers of molecular explosives with high signal-to-noise is consequently a critical issue that must be confronted. Finally, techniques to probe molecules under conditions of static high temperature and hydrostatic pressure for comparative purposes were desired.

## Vibrational Sum-Frequency Generation




Symmetric nitro stretch of 4-nitrobenzenethiol (NBT)

$\omega_{SFG} = \omega_{VIS} + \omega_{IR}$

$I_{SFG}(\omega) \propto [\chi^{(2)}(\omega) E_{VIS} E_{IR}]^2$

where $\chi^{(2)}_{XYZ}(\omega) = N \langle\beta_{XYZ}(\omega)\rangle$

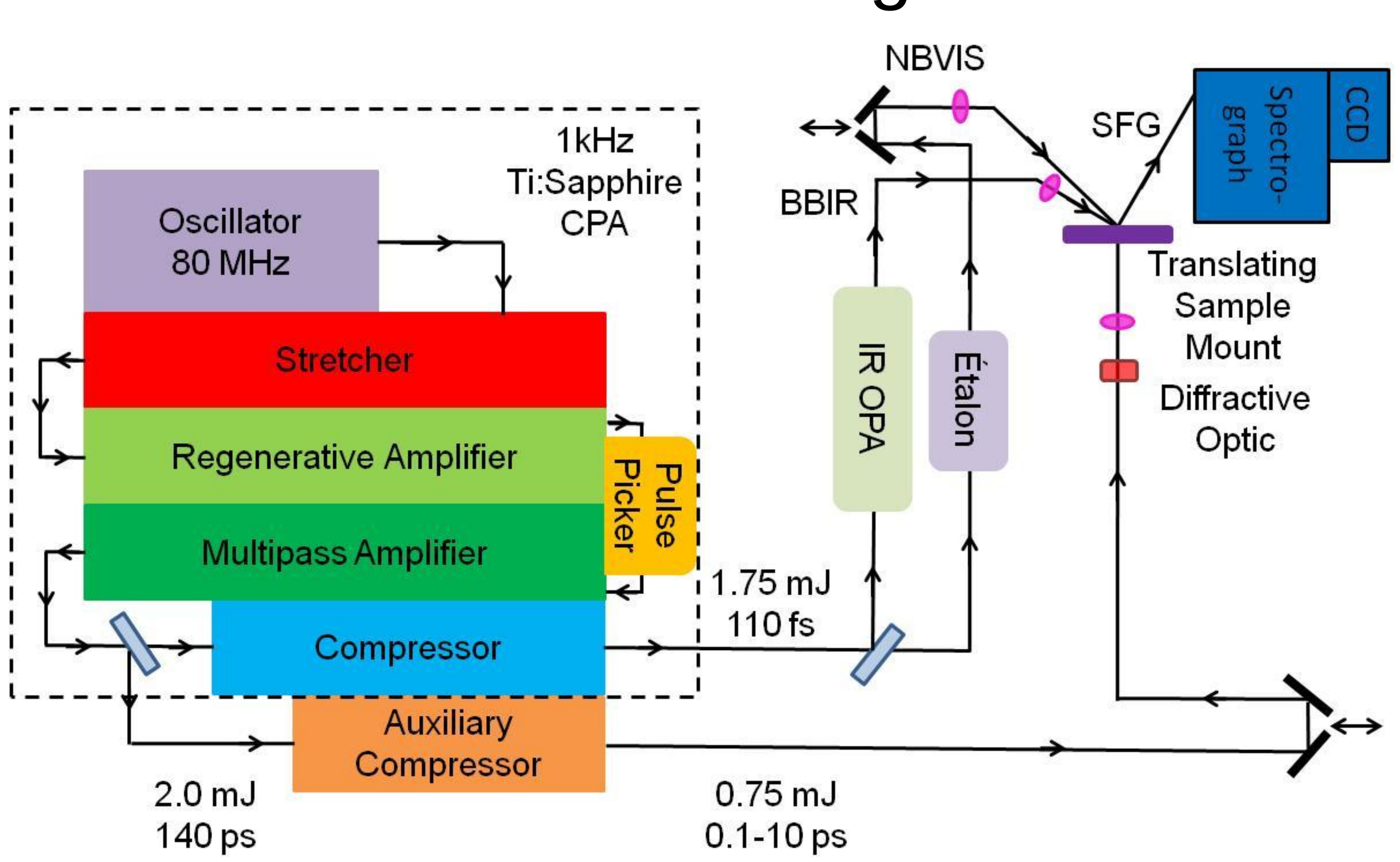
For centrosymmetric media (bulk):

$\chi^{(2)}(\omega) = 0$

For non-centrosymmetric media (interface):

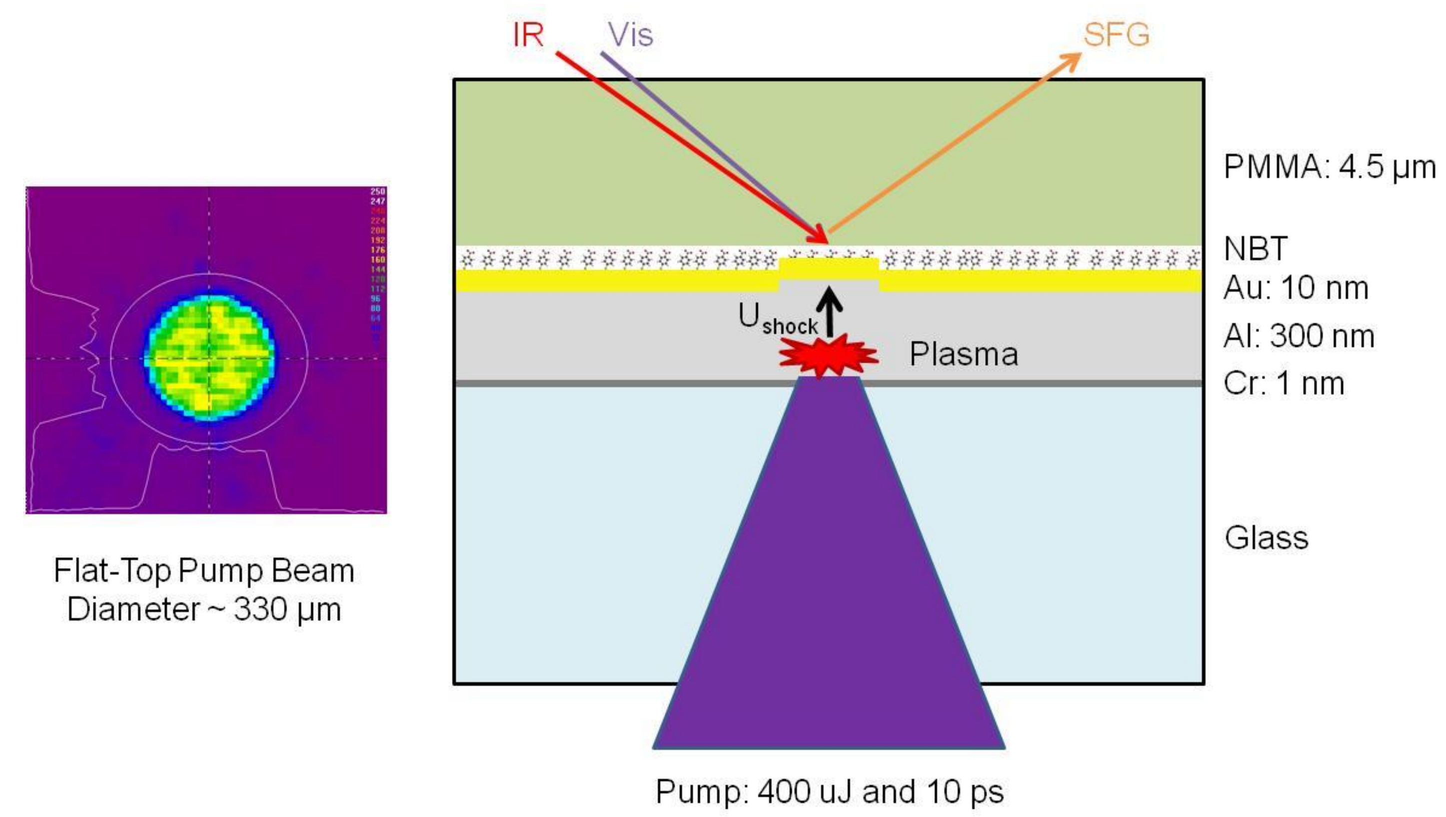
$\chi^{(2)}(\omega) \neq 0$

## Basic Laser Configuration

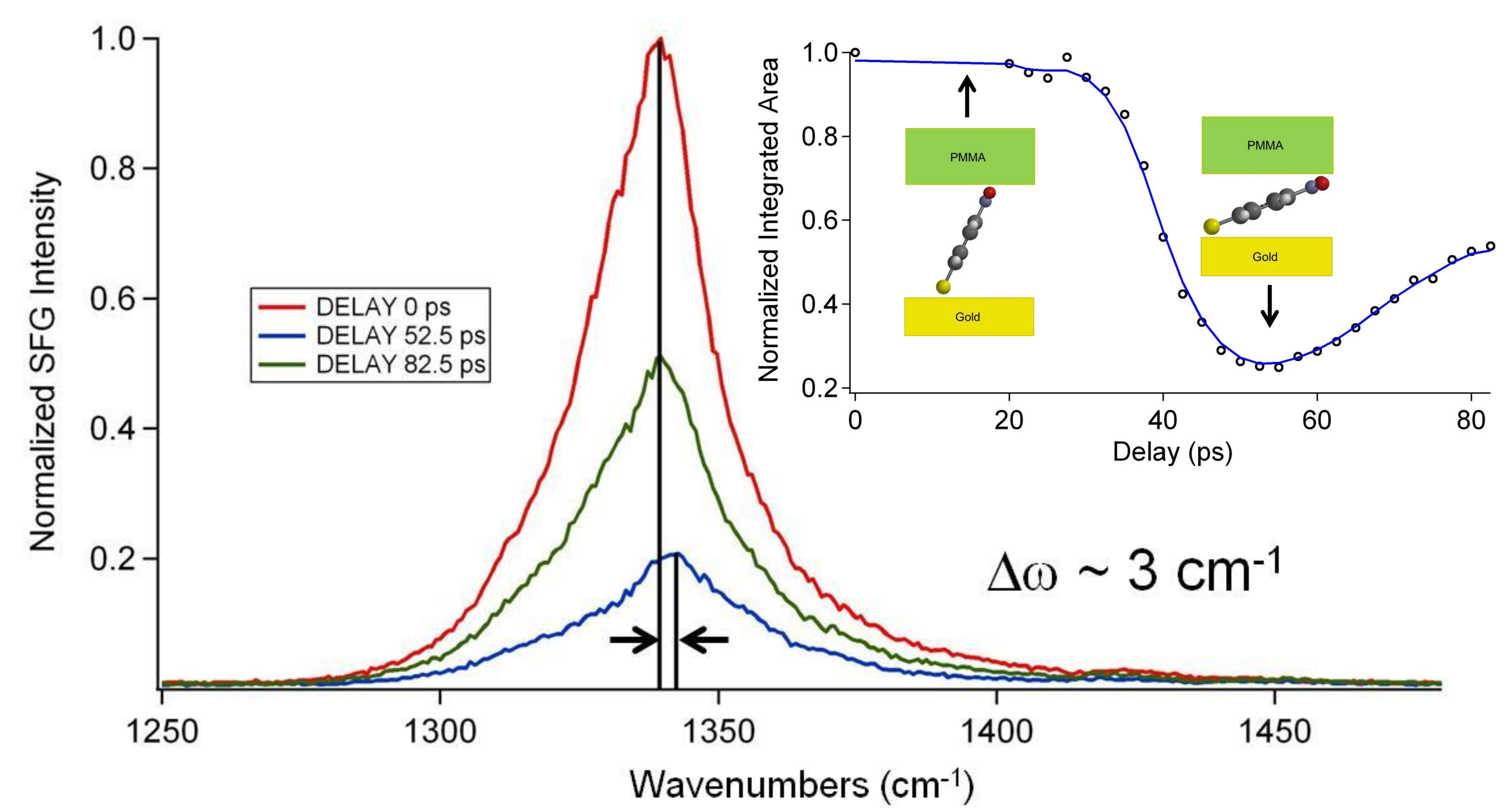



Optical parametric amplifier (OPA) → Broadband IR (BBIR) pulse (250 cm$^{-1}$ FWHM)
Étalon → Narrowband 800nm 'visible' (NBVIS) pulse (10 cm$^{-1}$ FWHM)

## Original Substrate Design

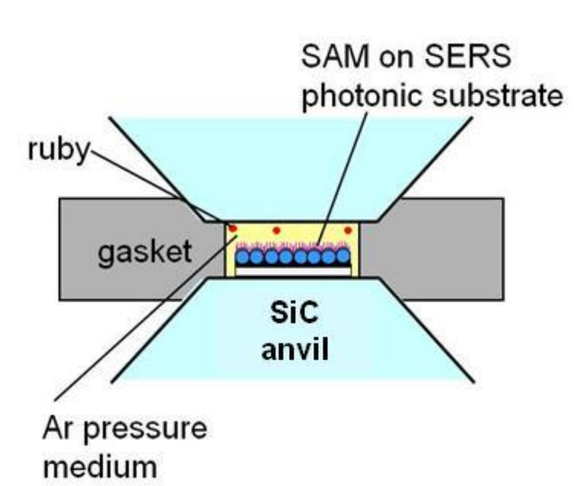


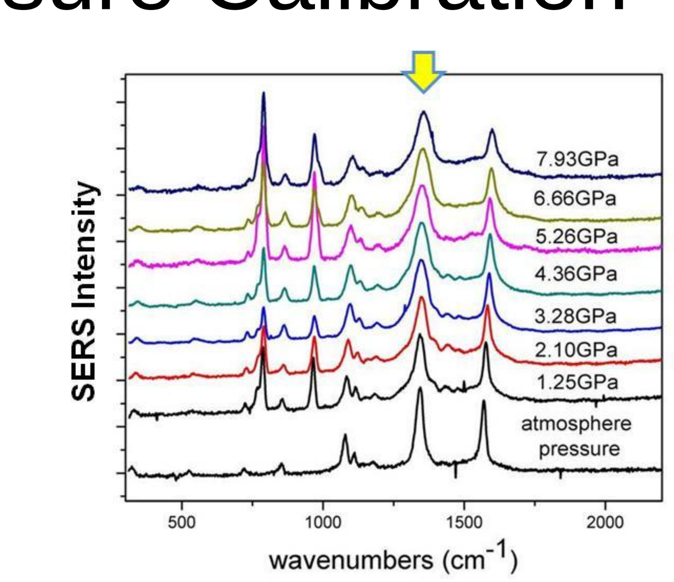

## Shock-Loading of NBT SAM




Time zero denotes incidence of pump beam onto sample.

## Hydrostatic Pressure Calibration

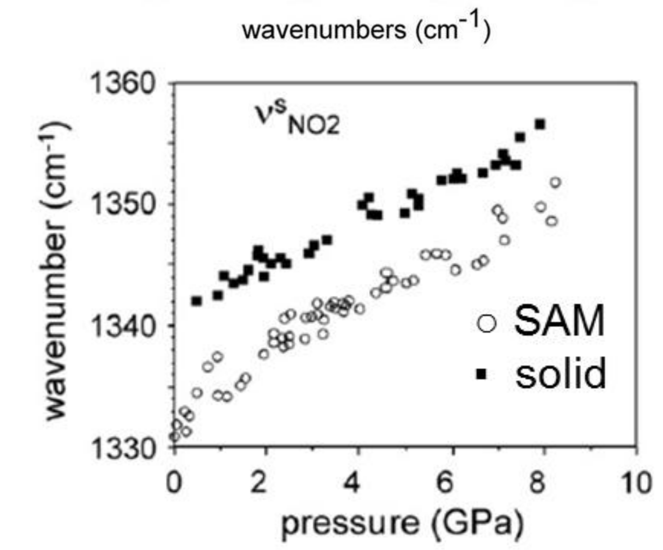


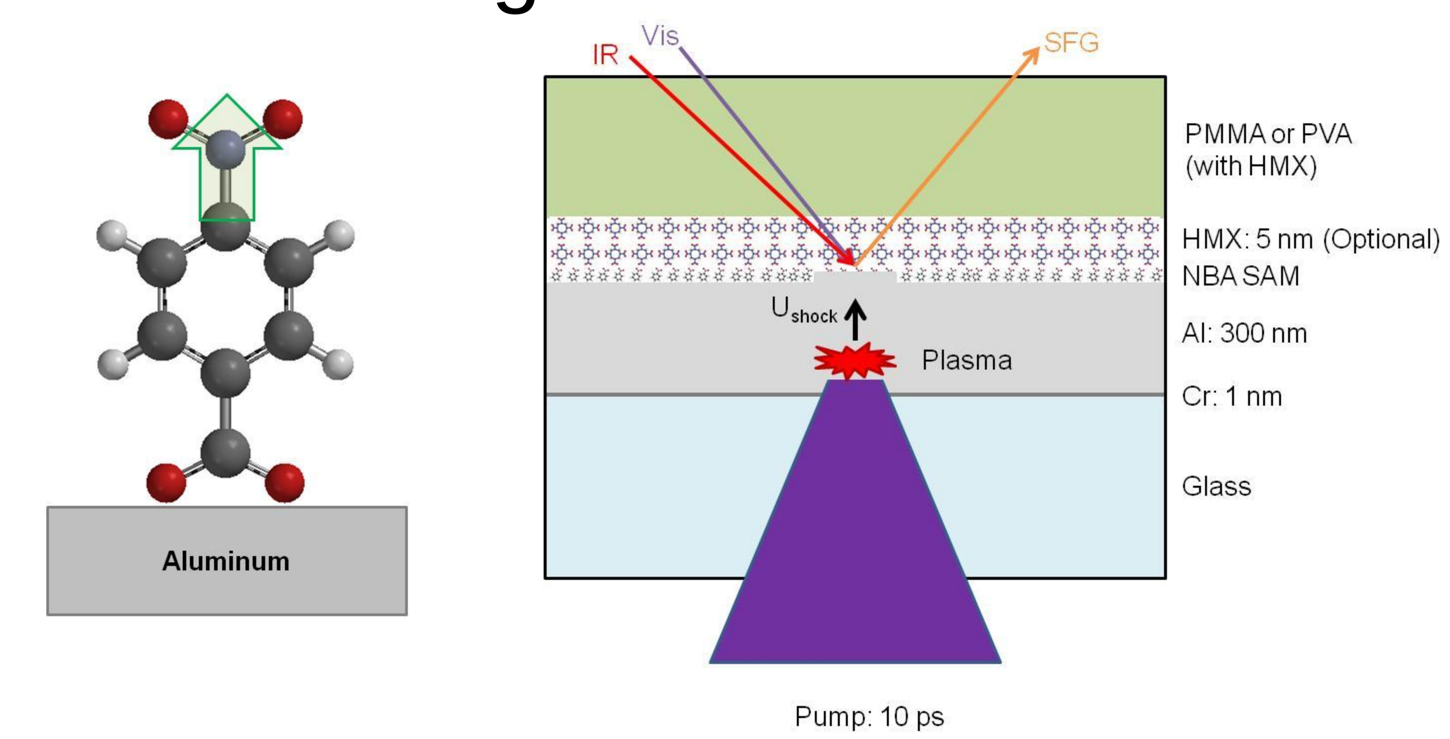

NBT symmetric nitro stretch (yellow arrow)

Frequency shift for NBT SAM 2.1 cm$^{-1}$ / GPa

**Δ ω = 3 cm$^{-1}$ → Δ P = 1.4 GPa LOW!**

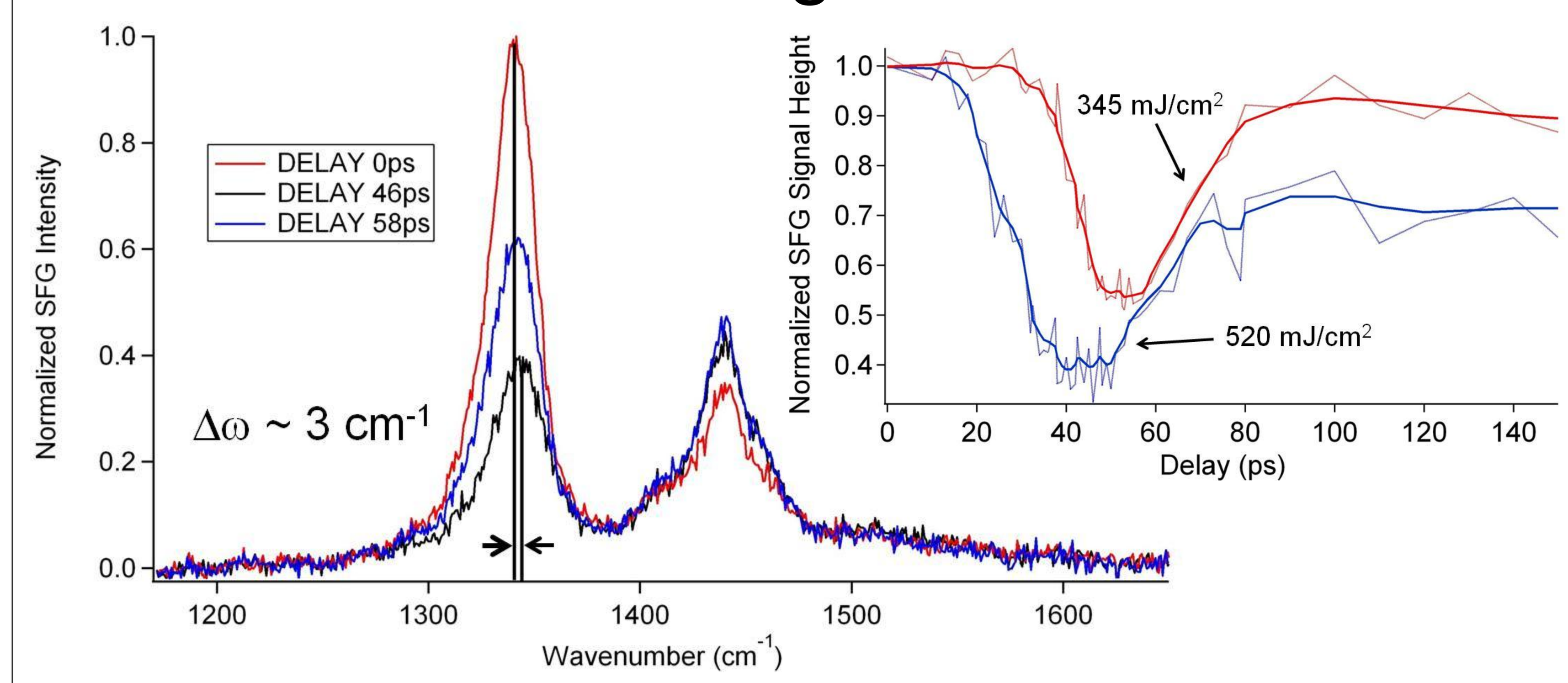



## Redesigned Substrate



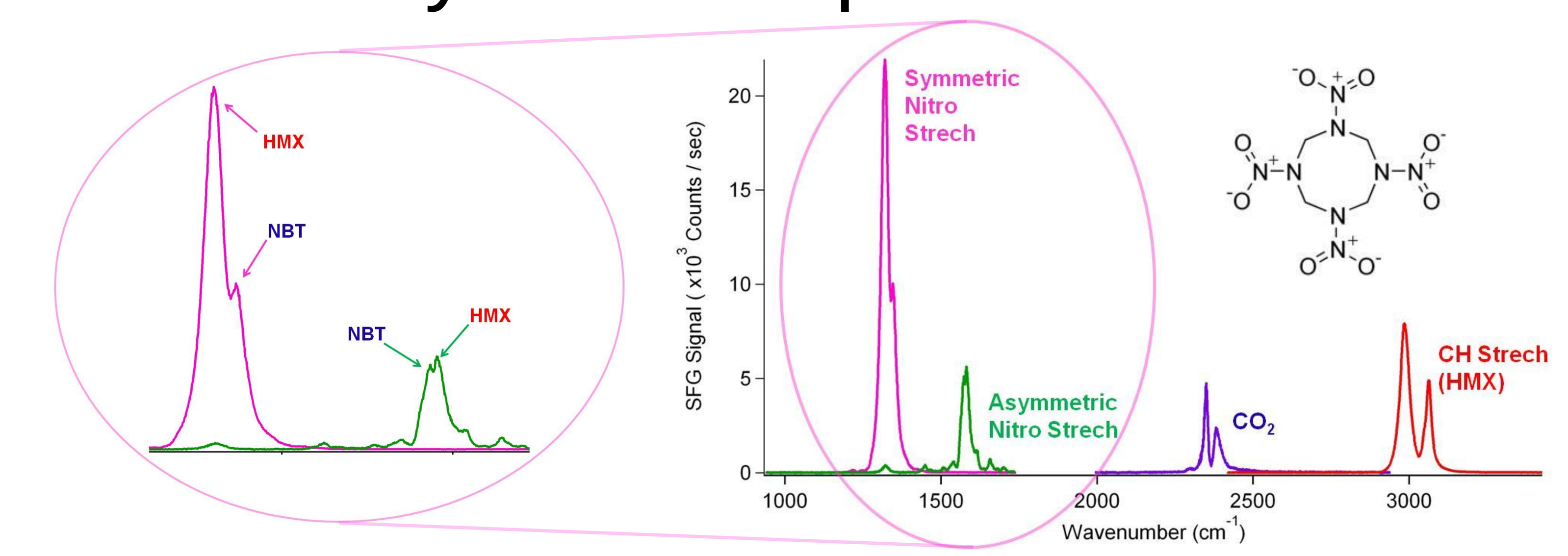

Forming monolayer of 4-nitrobenzoic acid (NBA).

## Shock-Loading of NBA SAM




Time dynamics shown for symmetric nitro stretch of NBA (lower frequency peak). Spectra taken with a pump pulse fluence of 520 mJ/cm$^2$.

## Thin Layer SFG Spectrum of HMX




Asymmetric nitro stretch and $CO_2$ offset by a factor of 5 and 50, respectively.

## Conclusions

Utilizing ultrafast vibrational SFG spectroscopy, the laser-driven shock compression of explosive simulant monolayers was probed with both high time and space resolution. Based on hydrostatic measurements, only pressure jumps of ~1.4 GPa where generated in the SAMs. However, irreversible changes (chemical effects or substrate deformations) were induced at higher pump fluences. The ability to recover high signal-to-noise signals from a thin, ~5 nm thick, layer of HMX was also demonstrated.

## Acknowledgements

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