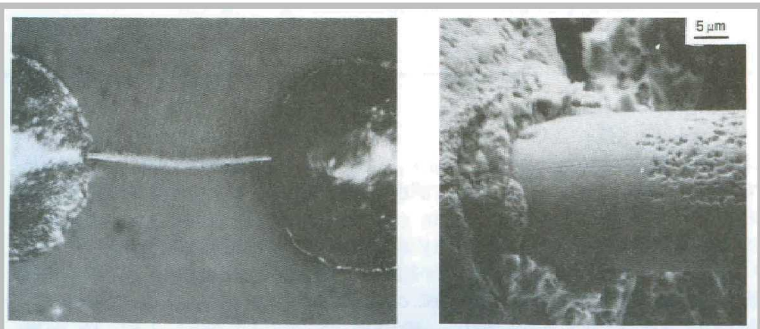
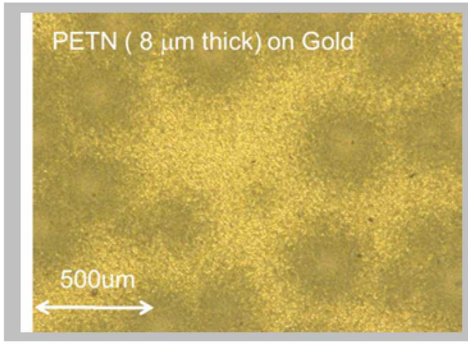
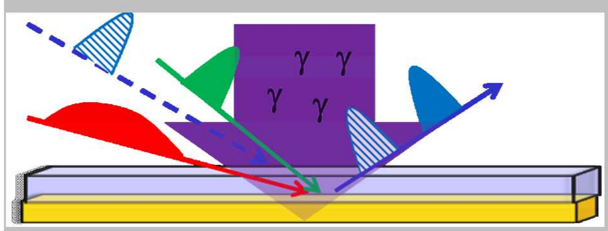
SAND2015-8194C







In situ study of surface-mediated explosive degradation using Heterodyne IR-Vis Sum Frequency Generation (HSFG).

Darcie Farrow, Robert Knepper, Barney Doyle, Elizabeth Auden, Ian Kohl, Kathleen Alam, Kevin Leung, David Enos, Laura Martin, Michael Marquez, Stephen Rupper

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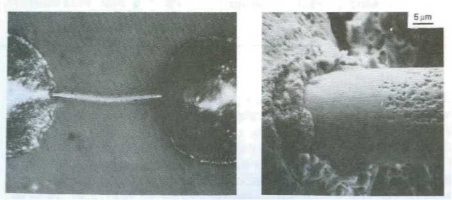
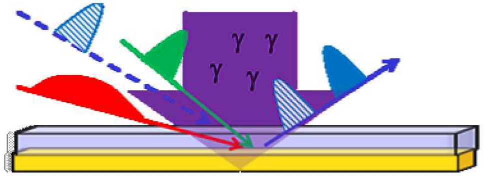
Outline

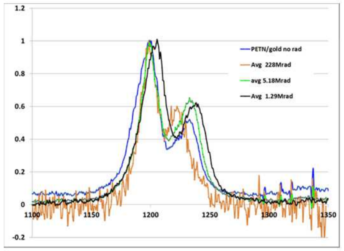
- Problem: Detecting unstable degradation products at buried organic/metal interface
  (explosive/metal bridge).



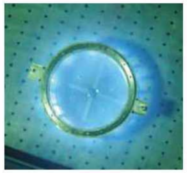
- Approach: Thin film model for bridge/Heterodyne SFG for intermediate detection.



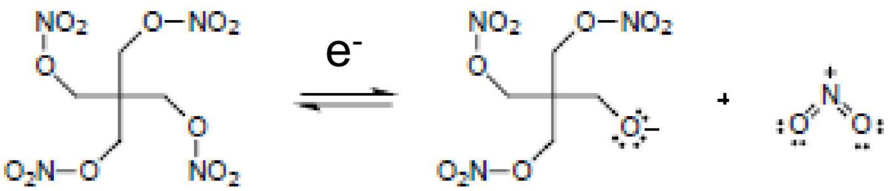
- IR-SFG of 3 PETN/metal interfaces.

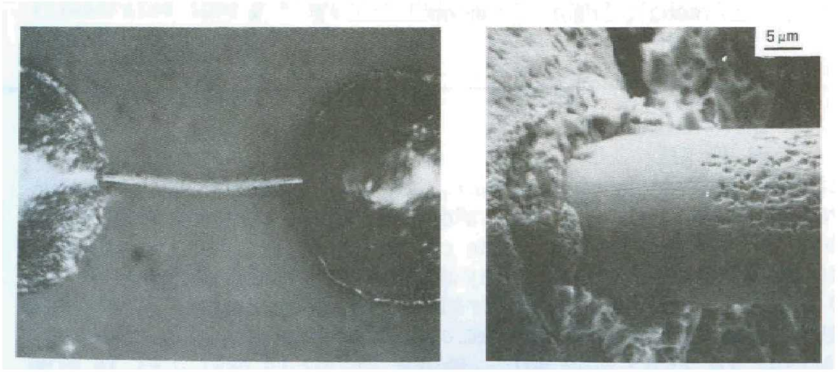


- Gamma radiation studies of PETN/gold samples.



Explosive/Bridgewire: Surface Enhanced Corrosion Chemistry

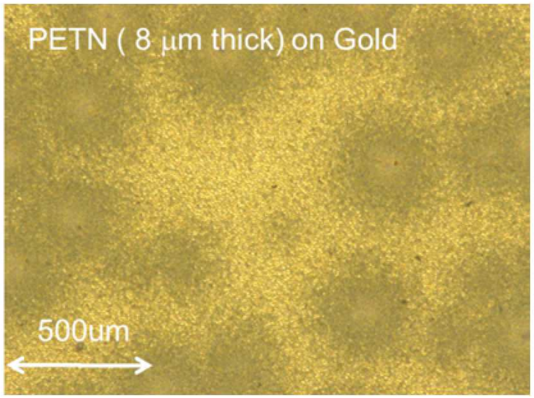


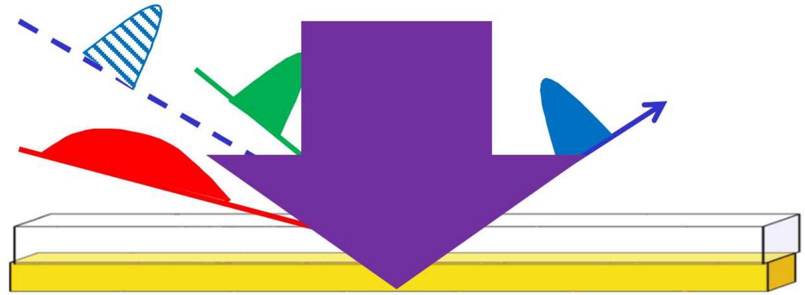


- Processes that degrade PETN may be enhanced at bridge wire interface *(auto-catalytic processes in low free volume, surface contamination, etc.)*

- Critical intermediates/products from the organic film may attack metal suface
  Currently no experimental method to identify corrosive species at wire/explosive surface in situ.

Thin film model system

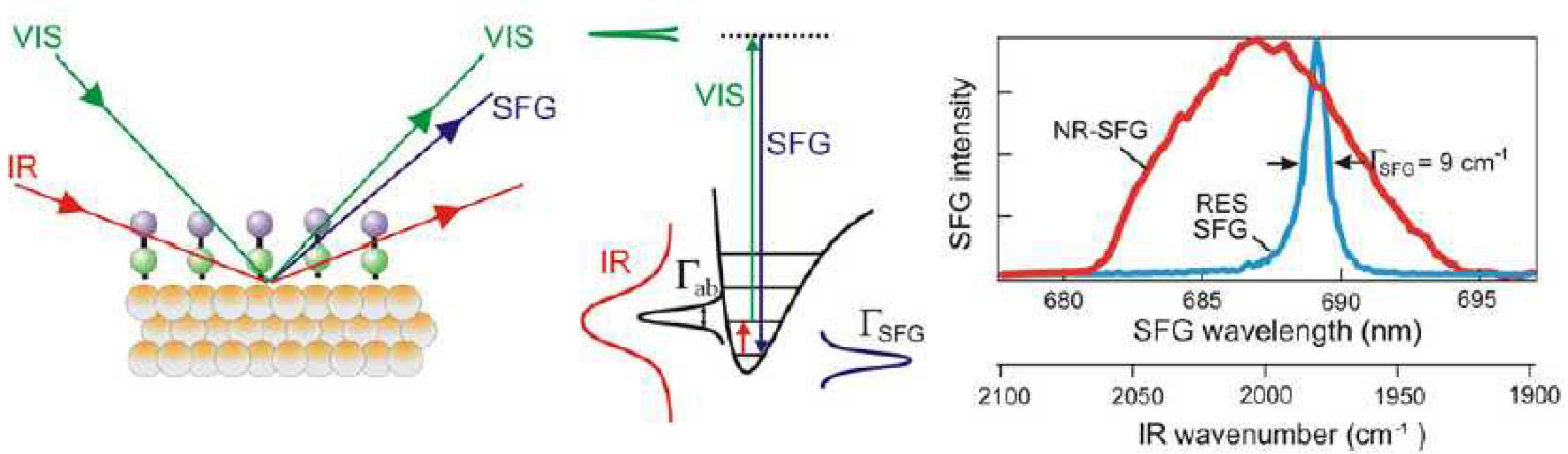




- Vapor deposited PETN on aluminum, copper and gold.
- Optically accessible samples (2-8 um PETN/1-2 um metal).
- Samples < 10 mg PETN. Samples can be accepted at radiation facilities on site not cleared for handling of explosive components.

Broad Band IR-Vis Sum Frequency Generation

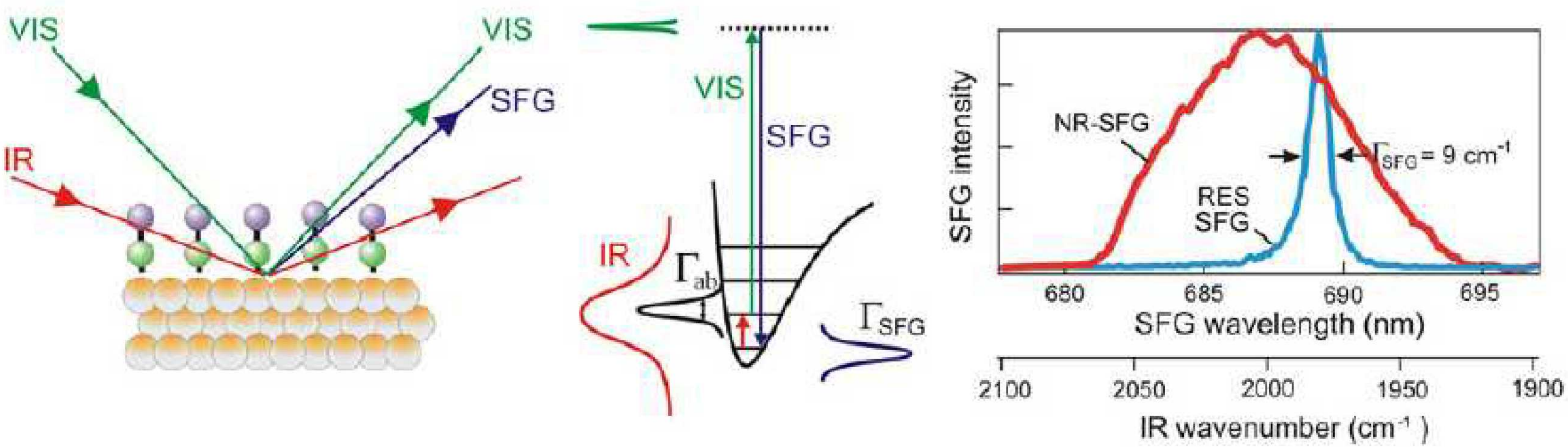
H. Arnolds, M. Bonn / Surface Science Reports 65 (2010) 45–66



- IR-Vis SFG signal sensitive to IR/ Raman active modes within a few monolayers of interface.
- Signal peaks when IR pulse resonant with vibration.
- Broad band SFG (fs IR pulse /ps Vis pulse)covering ~ 250 cm-1 of vibrational spectrum in each shot. *Time resolved measurements of reactive species possible w/ ps time resolution.*

Broad Band IR-Vis Sum Frequency Generation

H. Arnolds, M. Bonn / Surface Science Reports 65 (2010) 45–66

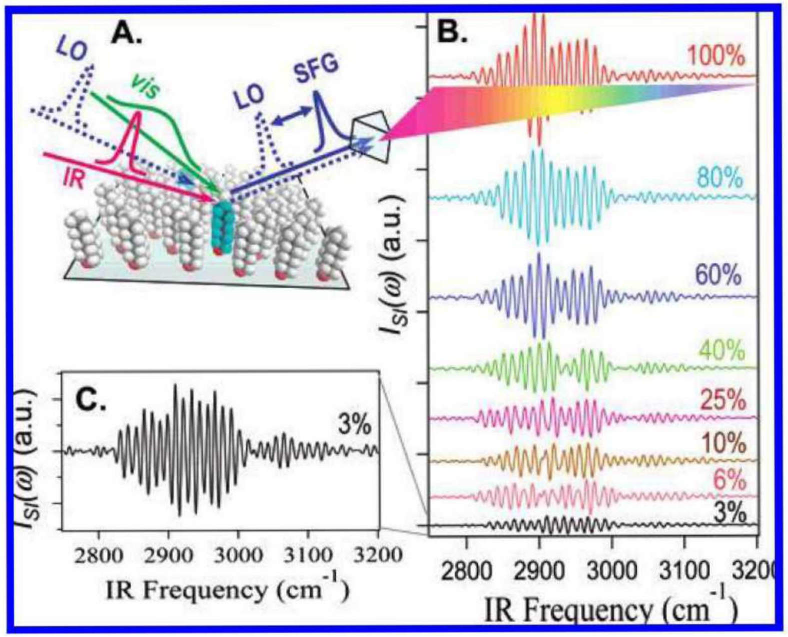


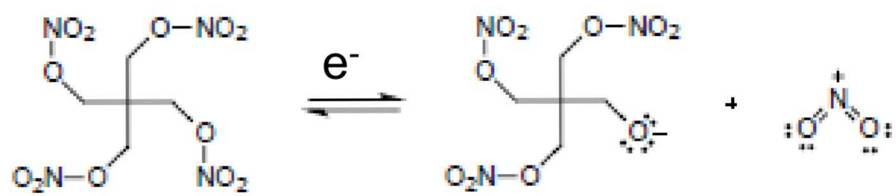
$I_{SFG}(N)$ is unfavorable for low surface coverage.

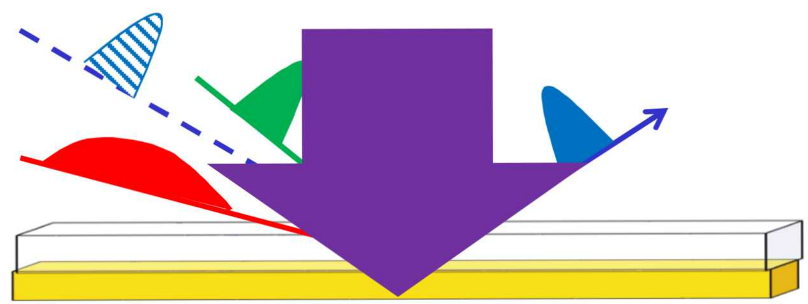
$$I_{\mathrm{SFG}} \propto |E_{\mathrm{SFG}}|^2 \propto |\chi^{(2)}|^2 = N^2 |\langle \beta^{(2)} \rangle|^2$$

10% surface coverage ➡ drop in intensity by factor of 100

Heterodyne detection to increase sensitivity





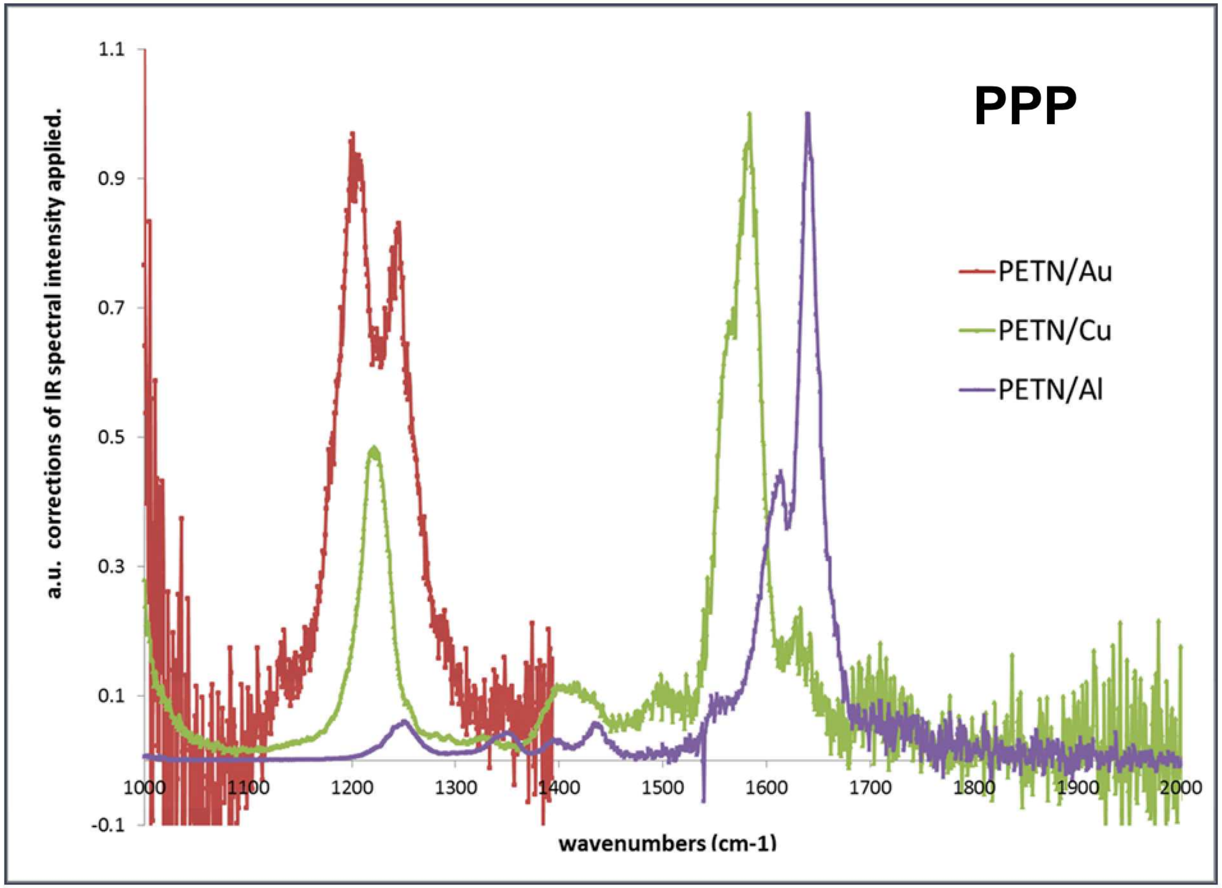


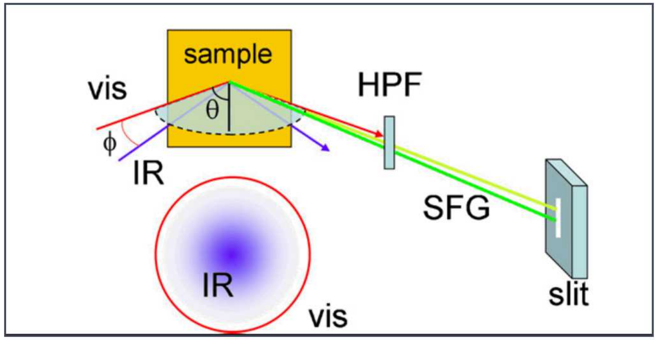
$$I_{SI} \propto 2\mathrm{Re}[E_{SFG}E^{*}_{LO}] \propto N\langle\beta^{(2)}\rangle$$

J. AM. CHEM. SOC. 2008, *130*, 2271−2275

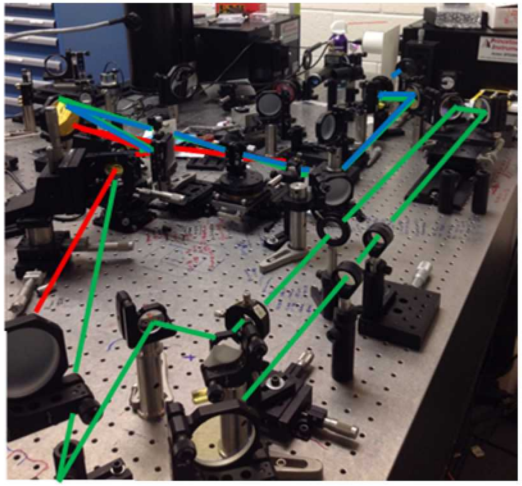
Detection limit lowered from 40% to <3%.

Traditional SFG Spectra of PETN/metal interface.

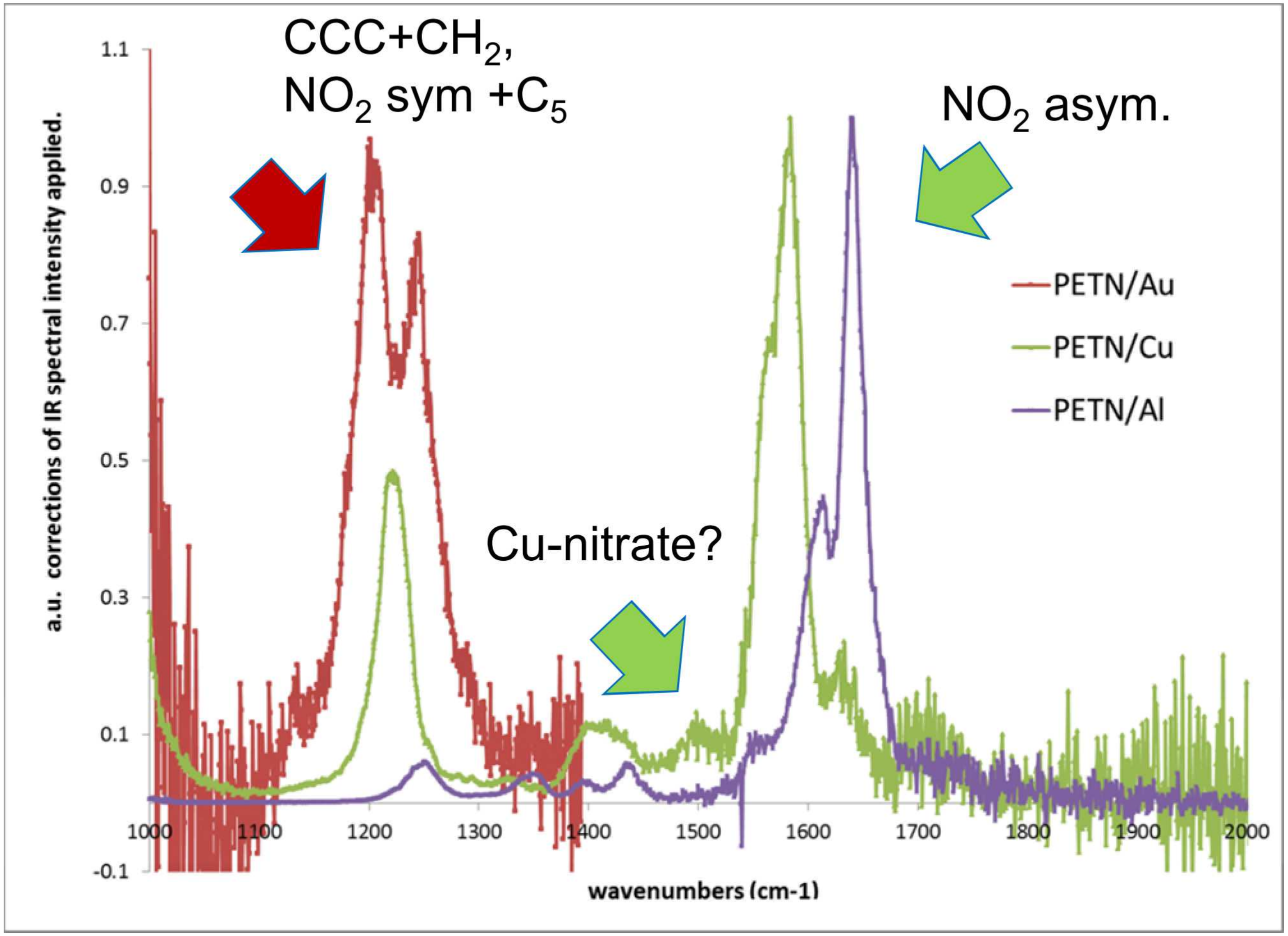




A. Lagutchev et al. / Spectrochimica Acta Part A *75 (2010) 1289–1296*



Kinetics Platform: Reactive Surface



- Copper nitrate present from oxidation of PETN on copper (degradation products). 1-2 day process at RT.
- Look for proposed gamma interaction with gold leading to PETN breakdown.

Surface Enhance Chemistry: PETN/Gold

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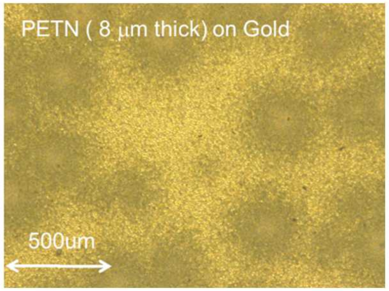
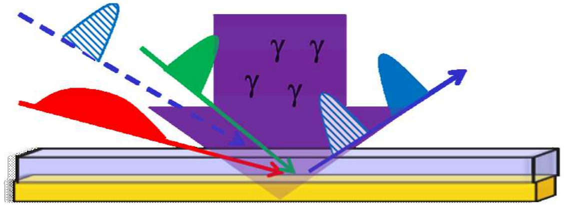
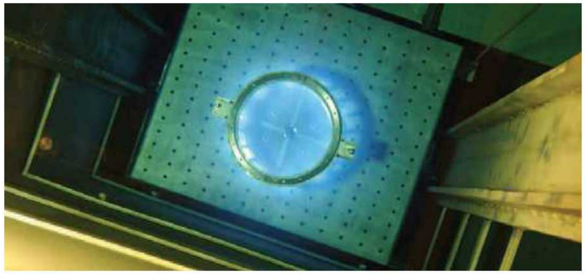
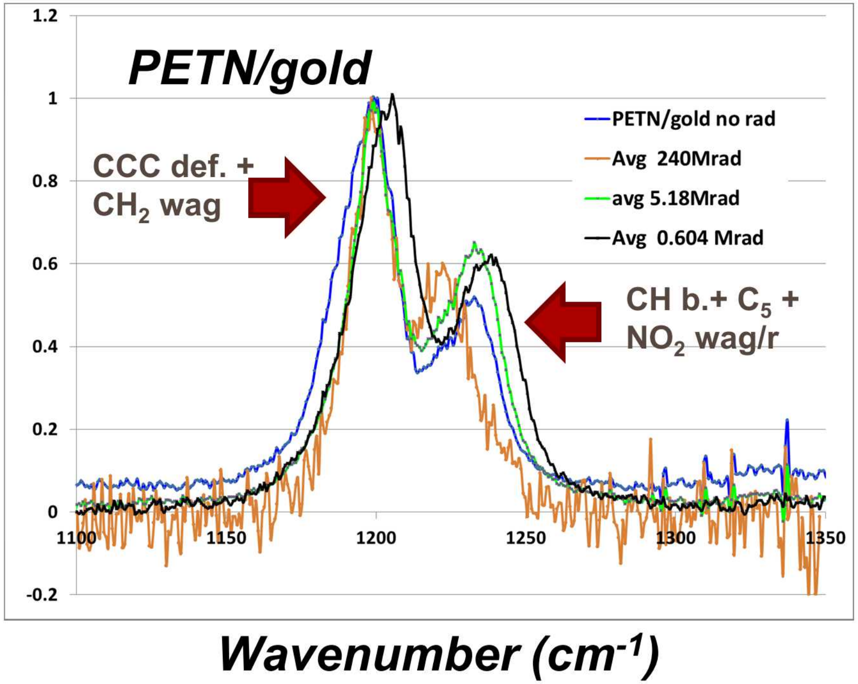
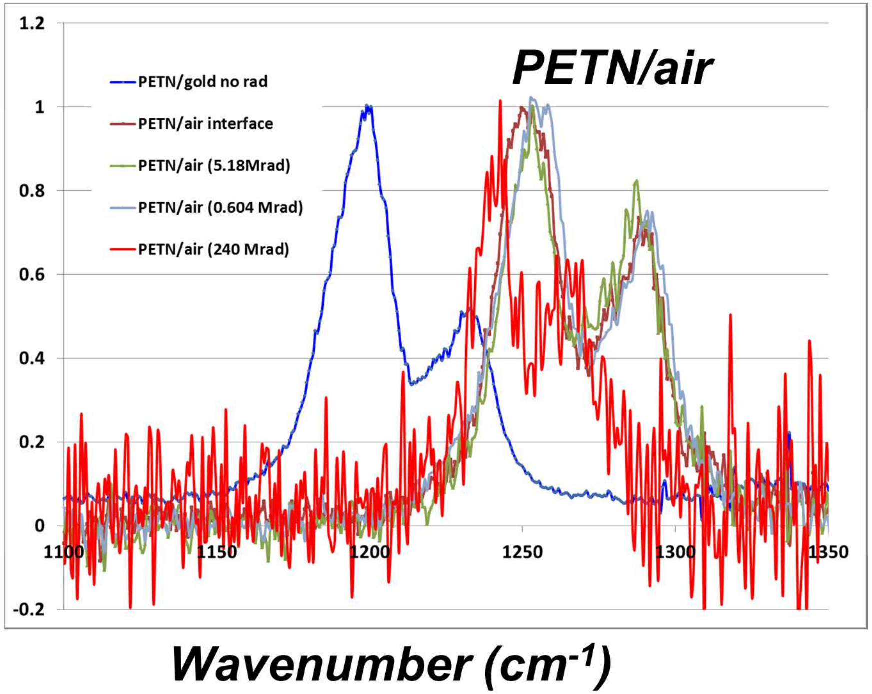




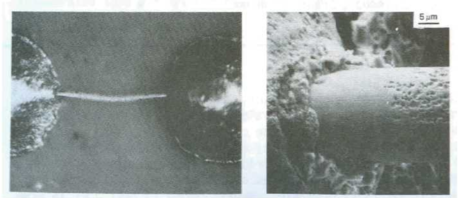






In Conclusion:

- Applying HSFG to the detection of unstable corrosion agents trapped at gold/PETN interface.



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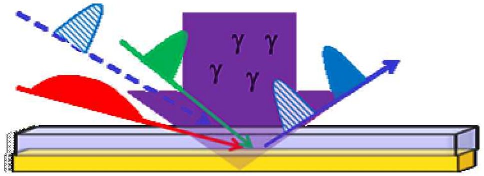
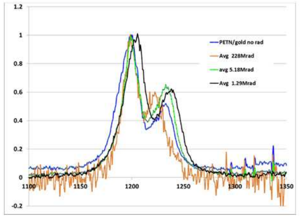


- Demonstrated platform for corrosion chemistry with traditional SFG for PETN/Cu and PETN/ gold interfaces.



- Next step to compare HSFG/SFG and IR/Raman detection limits for degradation products at surface.