Progress on: “Coherent Terahertz Acoustic Phonon Scattering: Novel Diagnostic for Erosion in Plasma Thruster Discharge Chamber Walls”

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Progress on: “Coherent Terahertz Acoustic Phonon Scattering: Novel Diagnostic for Erosion in Plasma Thruster Discharge Chamber Walls”

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Outline of Talk

• Essential Ideas of Research Program
• Background and Objectives
• Progress and Future Work

Note: Phonons are quantized traveling elastic waves associated with the displacement field of atoms from their equilibrium positions.
Essential Ideas of Proposed Research

• We have recently\(^1\) obtained the first experimental evidence for the direct excitation of coherent nanosecond-pulsed high-frequency acoustic phonons in semiconducting doping superstructures by electromagnetic fields of the same frequency.

• A key life-limiting mechanism for most plasma thrusters is their internal erosion through nonequilibrium plasma-material interactions mainly in the form of energetic ion impact and induced lattice defects.

• It is well known that lattice defects can strongly scatter acoustic phonons.

• We shall measure the transmission of nanosecond-pulsed coherent terahertz acoustic phonons through thin layers of wall materials, principally the layered hexagonal boron nitride (h-BN), taken before and after the materials have been exposed to controlled amounts of energetic xenon ions in plasma thrusters.

• We shall model via MD the levels of ion-induced defects, and demonstrate a correlation between exposure time and phonon transmission measurements.

Background on Coherent Acoustic Phonon Generation

Motivation from early theoretical papers: Generation of narrowband coherent acoustic phonons by resonant absorption of far-infrared laser radiation in superlattices
Experimental Constraints for Phonon Generation/Detection

• Intense, nanosecond-pulsed, FIR laser source required for S/N and LA/TA mode discrimination in time-of-flight across 0.5-mm substrate
• Fast, sensitive (~nW) superconducting bolometer to detect nanosecond-pulses for TOF mode discrimination
• SC small-period grating-coupler required to (1) convert transverse laser E-field to a evanescent longitudinal component over depth of underlying DSL, and (2) protect bolometer from direct FIR laser radiation exposure.
• Boron-doped substrate (~15 Ω-cm) for Si:B uniaxial stress-tunable (0-4 kB) phonon spectroscopy (in progress)
• Jig for (a) bolometer shielding (pulsed-laser discharge noise) and (b) Si hyper-hemispherical lens coupling of laser radiation and (c) application of uniaxial stress
  Continued ....
**Phonon Experimental Arrangement**

- **Ultrashort-pulsed FIR Laser Radiation**: 1.22-mm, 5-ns, ~1-kW/mm²
- **Front Face**: Si Doping Superlattice (8.5 x 10¹² cm⁻²)
- **246-GHz LA Phonon**
- **Si:B (5 x 10¹³ cm⁻³) Substrate** (Piezophonon Spectroscopy)
- **Front Face**: Small-Period Niobium Grating Coupler (Line: 7 micron, Space: 3 micron)
- **Rear Face**: Granular Aluminum Bolometer: Straight Link (100-nm x 10 micron x 20 micron)
- **Uniaxial Stress Apparatus**: (~4-kBar)
- **34.5-nm period**
- **5 x 7 x 0.5 mm Sample Immersed in LHe @ ~1.75-K**
Theory of Delta-doped Doping
Superlattice as Resonant Absorber

Equation of motion of the lattice: \( \mathbf{M} \mathbf{N}_I [\dddot{\mathbf{\xi}}(r,t) + \Gamma \dddot{\mathbf{\xi}}(r,t)] = -C_{11} \nabla \times (\nabla \times \dddot{\mathbf{\xi}}) + \mathbf{F} \)

Force \( \mathbf{F} \) by harmonic FIR electric field on the delta-doped sheets consisting of \( N \) periods and period \( d \)

Displacement field has equal resonances at folded zone-center reciprocal lattice vectors, \( p=1,3,5 \ldots \)

Mean acoustic power flux at fundamental resonant frequency \( f=s/d \):

\[
\langle I_a \rangle = Z \langle \left( \frac{\partial \mathbf{\xi}}{\partial z} \right)^2 s^2 \rangle = \rho_m s^3 \left( \frac{en_d^{(2d)} E_0}{\rho_m d} \right)^2 \left( \frac{2\pi}{d} \right)^2 2(\omega \Gamma)^2
\]
The width of the resonance determined by finite number of periods \( N \):

\[
\Gamma \approx \Delta \omega = \frac{\omega}{N}
\]

At normal incidence, mean electromagnetic power flow, in terms of field strength within DSL (cgs):

\[
\langle I_{EM} \rangle = \frac{cE_0^2 (\sqrt{\kappa} + 1)^2}{32\pi}
\]

Conversion efficiency of electromagnetic to acoustic intensities:

\[
\frac{\langle I_a \rangle}{\langle I_{EM} \rangle} = 4 \frac{(en_d^{(2)}N)^2}{m^{sc}(\sqrt{\kappa} + 1)^2}
\]

\[
\eta = 2.4 \times 10^{-8} \text{ i.e., } -76 \text{ dB}
\]

Using silicon doping superlattice parameters:

\( N = 30 \)

\( n_d^{(2)} = 8.5 \times 10^{12} \text{ cm}^{-2} \)

\( d = 34.5 \text{ nm} \)

Delta-Doped Superlattices for 246-GHz Experiments*

*Courtesy of Michael Oehme and Jorg Schultze of U-Stuttgart IHT
Logfile of a nipi structure (~10 min/period)
Representative Doping Profiles by SIMS
Development of Cavity-Dumped Nanosecond-Pulsed FIR Laser*

- MW grating-tuned TEA CO$_2$ laser as optical pump for molecular gas laser.
- Molecular gas in custom resonator lases via vibrational-rotational transitions.
- Insert high-resistivity Si wafer in-situ at beamwaist in molecular gas resonator (low-loss for p-polarized radiation).
- Optically-switch the Si with 532-nm frequency-doubled YAG laser radiation in ns-timescale.
- Intense, cavity-dumped output with 5-ns pulse width result.

Details of ns-pulsed, cavity-dumped THz laser design

**FIR laser resonator schematic diagram.** 

- **M1**: 10-cm diameter Au-coated concave copper mirror, radius of curvature 845 mm;  
- **M2**: 10-cm diameter Au-coated concave copper mirror, radius of curvature 1275 mm;  
- **Silicon**: <100> float-zone, double-sided polished, 100-mm diameter x 0.5 mm, 7000 Ω-cm resistivity;  
- **L1**, **L2**, plano-convex TPX lens, 159-mm and 105-mm focal length, respectively;  
- **M3**: 75-mm diameter flat Au-coated Pyrex mirror;  
- **L3**: 50-mm diameter, -100-mm focal length plano-concave BK7 spherical lens, visible AR coating;  
- **L4**: -40 mm focal length cylindrical BK7 lens.  

*Not shown*: modified *Gentec DD-250 TEA CO2 pumping laser* (focused TEA laser radiation enters through 2-mm coupling hole in **M2**), *Spectra-Physics GCR-150* frequency-doubled YAG laser output entering through **L4**. The FIR laser output is coupled into a segmented 246-GHz corrugated waveguide.
Recent improvements to the FIR

- in-situ mode-matching Newtonian telescope, employing custom TPX lenses, to couple the output into a 3 m long 246 GHz corrugated waveguide for beam transport to the sample access window of the floor-mounted liquid helium cryostat.
- a recycling cryogenic absorption pump for the isotopic methyl fluoride gain medium.
- After the 246 GHz laser radiation propagates through corrugated waveguide, routinely obtain power levels at the cryostat access window of 5 kW in 6 ns pulses, at a pulse repetition rate of 10 pulses per second.
Pulse profiles of cavity-dumped nanosecond-pulsed FIR laser and optical-switch*

FIR laser output transported to floor-mounted cryostat via 3-m corrugated waveguide

Superconducting grating-coupling required for LA phonon generation and to protect bolometer

The decay length $\delta$ of the first-order diffracted evanescent $(p/(\lambda/n)<1)$ field is given* by:

$$\delta \approx \left| \frac{p}{2\pi \sqrt{1-(np/\lambda)^2}} \right|$$

where $p$ is the grating period and $\lambda/n$ is the optical wavelength of the FIR in the DSL. For a 10-µm grating period at 246-GHz in silicon, the decay depth is 1.6-µm, i.e, the same order as the DSL active region. A grating fill factor of 0.6 maximizes the longitudinal component. Skin depth of superconducting Nb is 39-nm.

Thin film patterning of bolometer and grating-coupler: image-reversal photolithography, dc-magnetron sputtering, and lift-off

Niobium Grating-Coupler (3 micron space, 7 micron line, thickness 500-nm = ~13 FIR skin depths) Thicker ones on the way!

Granular Aluminum/Palladium (0.35 mm²) Bolometer

Granular Aluminum/Palladium (200 mm²) Bolometer
Bolometer characterization for deconvolution\textsuperscript{1}

Bolometer’s characteristic current $I_m$ and thermal conductance $G$

In absence of phonon flux: $R_b^{(stat)} = \frac{R_b(T_S)}{1 - \left(\frac{I_b}{I_m}\right)^2}$


$G_{1.79K} = I_m^2 \alpha = 2.5 \times 10^{-7}$ W/K
Granular aluminum/Pd bilayer microbolometer operational parameters*

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bath temperature: $T_S$</td>
<td>1.791 K</td>
</tr>
<tr>
<td>Bias current: $I_b$</td>
<td>20 $\mu$A</td>
</tr>
<tr>
<td>Bolometer resistance: $R_b$</td>
<td>4.3 $\Omega$</td>
</tr>
<tr>
<td>Characteristic current: $I_m$</td>
<td>43.0 +/- 0.9 $\mu$A</td>
</tr>
<tr>
<td>Thermal conductance: $G</td>
<td>q_0$</td>
</tr>
<tr>
<td>Sensitivity: $\alpha</td>
<td>_i$</td>
</tr>
<tr>
<td>Characteristic Time: $\Lambda(\tau)$</td>
<td>3.2 (2.9) ns</td>
</tr>
<tr>
<td>Heat Capacity: $C$</td>
<td>7.2x10^{-16} J/K</td>
</tr>
<tr>
<td>Responsivity: $\mathcal{R}$ (f=50 MHz)</td>
<td>9.9-kV/W</td>
</tr>
</tbody>
</table>

Predicted bolometer signal $(\eta=9.4 \times 10^{-9})$

For spectral analysis we will use the technique of burning a hole into the spectrum of the emitted phonons by resonance scattering\(^1\). Stress tuned boron acceptors in silicon are used as resonant scatterers for several reasons: the ground state forms a two level system under uniaxial stress and the splitting is proportional to the applied stress. Resonance scattering is very effective so that an acceptor concentration as low as \(10^{13}/\text{cm}^3\) is sufficient. The homogeneous width of the resonance is typically some percent of the splitting.

For example, the ground state splitting energy \(E\) (GHz) as a function of stress \(X\) (kBar) along a [100] direction is given by:

\[
E = (392 \text{ GHz/kBar}) X
\]

Sample held between two stainless pistons (not shown is new ‘rapid prototype’ sample jig to hold a silicon hyperhemispherical lens coupler in contact with SL)
Sample jig for shielding, stress apparatus and silicon lens coupling

Ni-plated polymer sample holder for uniaxial stress capability, silicon lens coupling, micro-coaxial (semi-rigid) connection, and shielding (In-foil surrounds all edge faces).
First evidence of coherent 246-GHz LA phonon generation with no evidence of extraneous broadband phonons*  

Arrival of ballistic LA phonon signal at appropriate TOF and of same pulse width as generating cavity-dumped FIR laser

... Higher Time Resolution

Leakage of cavity-dumped FIR laser radiation through grating-coupler

Arrival of ~ 2-nW ballistic coherent LA phonon signal at appropriate TOF and of same pulse width as generating cavity-dumped FIR laser

Absence of any delayed larger, incoherent TA phonon signal – (expected if LA phonon signal was simply incoherent heat pulse)
Extraneous TA phonons are seen in other heat pulse experiment with same samples

Conclusions from 246 GHz LA phonon generation

• Detected LA phonon absorbed power of \(~2\) nW -- in good agreement with coherent phonon generation theory of Ruden and Dohler.

• Time-of-flight (60-ns) in agreement for ballistic LA phonons across 0.5-mm thick substrate.

• No evidence of any larger TA phonon pulse as detected previously for similar samples in a heat pulse experiment – implies no incoherent broadband phonon generation (heating), which in turn implies LA pulse is likely coherent.

• Next steps for 246 GHz phonon experiments: Increase grating-coupler thickness to reduce leakage; use large bolometer to increase signal strength; deploy Si:B spectrometer, increase N
Background on acoustic phonon scattering

• ‘Obstacles’ in the path of propagating phonons can result in scattering, and such obstacles may include other phonons, crystalline interfaces, grain boundaries, impurity atoms, structural defects such as vacancies and dislocations, change in atomic mass (isotopes), and, in general, any features that change the bond stiffness, bond orientation, or mass of adjacent atoms from those of the host lattice. (James P. Wolfe, *Imaging Phonons: Acoustic Wave Propagation in Solids*, (Cambridge University Press: New York, 1998) Chapters 8-9.)

• Point imperfections were shown to scatter as the fourth power of frequency (analogous to the familiar Rayleigh scattering of photons), dislocations as the first power, and grain boundaries independently of frequency. (P.G. Klemens, “The Scattering of Low-Frequency Lattice Waves by Static Imperfections”, *Proc. Phys. Soc. A* 68, 1113 (1955).

• Early experiment on broadband acoustic phonon-pulses scattered from 40-keV aluminum-implanted damage at sapphire surfaces, a region implanted by the lowest dose ($10^{14}$ cm$^{-2}$) was easily identified by the change in the magnitude of specularly reflected phonon modes (LA mode affected most) due to defect scattering even though this region was found to be indistinguishable from virgin sapphire when measured with Rutherford back channeling (RBC). (Hamid bin Rani, S C Edwards, J K Wigmore and R A Rollins, “Observation of heat pulses scattered from ion bombardment damage at sapphire surfaces”, *J. Phys. C: Solid State Phys.* 21, L701 (1988)).
Estimate the expected attenuation of transmitted 1.04 THz coherent LA phonons through a thickness h-BN with lattice defects resulting from xenon ion bombardment.

- The isotopic scattering rate of acoustic phonons is given by

$$\tau^{-1}(\nu) = \frac{2\pi^3}{3} V_0 g \nu^2 D(\nu)$$

where $V_0$ is the volume per atom, $g$ is a factor measuring the magnitude of the mass fluctuation ($g=2.02 \times 10^{-4}$ for silicon) $\nu$ is the phonon frequency and $D(\nu)$ is the one-phonon density of states per unit volume. In the case of h-BN, both $V_0$ and $D(\nu)$ are reported by Kern, G. Kresse, and J. Hafner, “Ab initio calculation of the lattice dynamics and phase dynamics of boron nitride”, Phys. Rev. B 59 (13), 8551 (1999): $V_0=8.69 \times 10^{-30} \text{ m}^3$, and by fitting the density of states to a quadratic, $D(\nu) = 5.47 \times 10^{-10} \nu^2 \text{ s}^3/\text{m}^3$.

- Scattering rate for 1.04 THz acoustic phonons in h-BN, assuming a mass fluctuation $g$ (due to vacancy defects) equal to 20%: $\tau(1.04 \text{ THz}) = 2.30 \times 10^{10} \text{ s}^{-1}$.

- The attenuation factor $\alpha$ for ballistic phonons of frequency $\nu$ through a thickness $d$ of material is given by $\alpha(\nu)=1-\exp(-d/\nu \tau(\nu))$. The phonon velocity for LA phonons along the c-axis (normal to the layers) of h-BN is $v_{LA[001]}=3.44 \times 10^3 \text{ m/s}$.

- For $d=10 \text{ nm}$ thick layer with $g=0.2$, we find $\alpha(1.04 \text{ THz})=7\%$. 
Background of the transfer of hexagonal boron nitride layers to Si(100)

- Large-scale (cm$^2$) hexagonal boron nitride layers will be fabricated and transferred to our silicon doping superlattice Si(100) substrates by Dr. Pulickel Ajayan’s group at Rice University, [L. Song et al., “Large Scale Growth of Atomic Layers of h-BN”, Nanoletters 10, 3209-3215 (2010).]
Background of the transfer of hexagonal boron nitride layers to Si(100)

- Process flow for transfer
Progress to date on 1.04 THz coherent phonon generation

- New doping superlattices appropriate for 1.04 THz have already been supplied. The parameters are as follows: 30 periods of 8.1 nm period with a 2-D doping concentration of 3E13 cm$^{-2}$.
- In order to avoid the generation of broadband (heat pulse) phonon generation in the substrate through THz radiation absorption in the grating coupler, we must use a grating material with a superconducting energy gap higher than 1.04 THz. (The gap frequency for our current Nb gratings is 670 GHz.) NbTiN is an ideal candidate and Arthur Lichtenberger at the University of Virginia has already fabricated NbTiN grating couplers on our new Si superlattices. NbTiN films deposited at 400 °C have a $T_c=16$ K and $\rho_{n, 20K} \sim 110 \, \mu\Omega\cdot$cm, with a superconducting gap of up to 1.15 THz [Cecil et al., “Investigation of NbTiN Thin Films and A1N Tunnel Barriers With Ellipsometry for Superconducting Device Applications”, IEEE Trans. Appl. Supercond. 17(2), 3525 (2007).]
- Experiments at 1.04 THz LA phonon generation without h-BN are in progress.
- We have demonstrated ns-pulsed 50 kW peak power 1.04 THz laser radiation (to be published) (next slides)
- h-BN layer transfer is nearly finished for a small set of samples.
High-Power 1.04 THz Radiation Available (from CH$_3$F pumped by 9R20)

![Graph showing THz Laser Radiation: 1.04 THz, 55 kW, 10 pps. Gaussian Fit, Width 4.55 ns.](image)
Custom Metal-Mesh Scanning Fabry-Perot Interferometer with Finesse of 2.0 at 1 THz
Metal-Mesh Fabry-Perot interferogram verifies 1.04 THz laser radiation

---

**Experimental F-P Scan of 1.04 THz laser radiation**

**Theory (k=21.08 mm\(^{-1}\), f=2.12)**

---

**Fabry-Perot Response (arb units)**

---

**distance (mm)**

---

**Model**

Fitted (User)

**Equation**

\[ C(1+H^2\sin(F+B))/X^2 \]

**Reduced Chi-Sqr**

0.00115

**Adj. R-Square**

0.91019

---

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<th>Value</th>
<th>Standard Error</th>
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<tr>
<td>B</td>
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<td>B</td>
<td>21.80869</td>
<td>0.00385</td>
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Acknowledgements

• Art Lichtenberger, Professor of Electrical and Computer Engineering at U-Virginia, will supply the NbTiN grating-couplers
• Pulickel Ajayan, Professor of Mechanical Engineering & Materials Science at Rice University, will be providing the h-BN transferred layers
• Pawel Keblinski, Professor of Material Science and Engineering at Rensselaer Polytechnic University, will help model the acoustic phonon transmission through h-BN layers with prescribed numbers of defects
• Kolin Brown and staff at the West Virginia University Nanoengineering Shared Cleanroom Facility
• In addition to AFOSR Space Power and Propulsion Program, partial funding also from NASA EPSCoR.
Hall Thruster Plume Analysis
For Planning of Material Exposure Tests

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Introduction

• **Background:** AFRL/RQRS (Dr. Dan Brown) agree to place BN sample in plume of Hall thruster for sample exposure test. Location ~ 1 m and angle of 30 – 60 deg.

• **Objective:** Model Hall thruster plume to estimate ion flux and energy distribution to help plan exposure tests.

• **Methods:**
  – Hall thruster plasma discharge: hybrid PIC-fluid (HPHall)
  – Hall thruster plume: hybrid PIC/DSMC-fluid (MPIC)
  – Boron nitride sputtering: molecular dynamics (HOOMD)
Background: 6 kW Hall Thruster

- AFRL exposure test will likely use 6 kW Hall thruster:
  - Nominal operating conditions as shown
  - Modeling of:
    - Thruster (HPHall)
    - Plume (MPIC)
    - Sputtering (HOOMD)

<table>
<thead>
<tr>
<th>Operating Condition</th>
<th>Mass Flow Rate</th>
<th>$I_d$</th>
<th>$V_d$</th>
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<tr>
<td>Nominal</td>
<td>20 mg/s</td>
<td>20 A</td>
<td>300 V</td>
</tr>
</tbody>
</table>
Technical Approach: Codes Employed

- **HPHall (Fife, 1998)** utilizes hybrid fluid-particle method:
  - Xenon species modeled as particles (PIC-MCC)
  - Electrons modeled as quasi-1D fluid, solved at smaller time scale (“sub-cycle”)
  - Provides detailed information for the near-field plasma (VDF, plasma properties)

- **MPIC (Cai, 2005)** utilizes hybrid fluid-particle method:
  - Xenon species modeled as particles (PIC-DSMC)
  - Electrons modeled as 2D fluid
  - Provides detailed information for the plasma plume (current density, IEDF, etc.)
Technical Approach: Numerical Details

- HPHall:
  - 1,530 cells (quadrilateral), axisymmetric
  - ~300,000 particles
  - run for 100,000 iterations with $\Delta t_{\text{PIC}} = 5.0 \times 10^{-8}$ s

- MPIC:
  - 3,191 cells (unstructured), axisymmetric
  - ~2,500,000 particles
  - run for 450,000 iterations with $\Delta t = 1.0 \times 10^{-6}$ s
Simulation Results

Electron Density (m^-3)

HPHall:
Plasma density

MPIC:
Ion Current Density
Simulation Evaluation

HPHall:
Plasma potential along channel centerline

MPIC:
Ion current density along 1 m arc
Plume Ion Energy Distribution
Near Location of Material Sample

Location: 1 m from thruster, 30 deg. from axis
Summary

• Modeling of AFRL plume environment completed:
  – HPHall used for unsteady simulation of plasma discharge
  – Time averaged results from HPHall used as input to MPIC plume simulation
  – Assessment of both HPHall and MPIC results using existing experimental data sets

• In the location of the exposed sample:
  – Ion current density of 0.1 – 1.0 mA / cm²
  – Ion energy distribution centered around 200 eV
Future Plans

• Development and application of molecular dynamics code HOOMD for boron nitride sputtering:
  – Efficient implementation on GPU architecture gives speedup of $>50$ compared to traditional CPU
  – Based on plume study, HOOMD analysis will focus on ion impacts around 200 eV

• Molecular dynamics analysis will be extended to study defect production:
  – Follow same approach as Lehtinen (2010)
  – Ultimately will allow direct comparison with measurements gathered in sample exposure