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RPPR Final Report
as of 25-May-2018

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Major Goals: The far-IR presents an interesting challenge for the development of an optical and optoelectronic infrastructure, as the III-V semiconductors that enable such extraordinary access to other portions of the EM spectrum are not well-suited for the far-IR, due to significant optical loss. These losses arise from the coupling of EM radiation to both free-carriers and phonons. While free-carrier loss can be controlled to an extent by limiting free-carrier density (doping) in the design and growth of semiconductor devices/materials, loss due to phonons is intrinsic to semiconductors in this wavelength range, and cannot be controlled in a similar fashion.

Far-IR light propagating through a polar crystal, such as GaAs or GaN, couples strongly to optical phonons. The transverse optical (TO) and longitudinal optical (LO) phonon energies of the semiconductor, determined by atomic composition and crystalline structure, bookend a region of negative permittivity and the center of a region of high loss, the so-called “Reststrahlen band” of the material. While the Reststrahlen band of any III-V material may span only a narrow portion of the far-IR, the combined effect of each of these bands, and the losses which extend well past the individual bands, combine to make the far-IR, or Reststrahlen region, in many ways a forbidding wavelength range for optics and optoelectronic research.

It is well-known that (longitudinal) optical phonons can interact strongly with electronic transitions in semiconductor materials. More recently there has been increasing interest in engineering phononic modes, most often surface phonon polaritons, or SPhPs) which can couple strongly to light. However, linking electronically generated longitudinal optical (LO) phonons to free space light has never been demonstrated, for the simple reason that the transverse nature of electromagnetic radiation prevents coupling to longitudinal phonon modes.

Our Short Term Innovative Research program looked to demonstrate a potential device architecture for electrical generation of far-IR light by coupling electrically generated LO phonons to optical modes which subsequently couple to free space. The overarching motivation is the development of a novel category of device which we label “opto-phononic-electronic” or –OPE. The ability to strongly couple light to LO phonons constitutes the single greatest barrier to the development of an –OPE based optoelectronic infrastructure, and constituted a high-risk, high-reward endeavor ideal for the STIR program.

The goals of the 9-Month STIR program can be summarized in the following 5 program thrusts:

1) To design and engineer optical resonances in polar semiconductor materials, demonstrating the ability to engineer strong optical absorption at the LO phonon energy.
2) To grow and fabricate opto-phononic structures based on the designs developed in (1).
3) To demonstrate strong coupling and absorption of far-IR radiation at the LO phonon energy in a semiconductor material
4) To develop an experimental mechanism for measuring LO phonon populations in our fabricated structures.
5) To develop techniques for the generation of non-equilibrium LO phonon populations, and coupling of these LO phonons to free-space photons via intermediate surface modes.

The above major goals were ambitious in both their extent and potential impact, given the 9-month time-frame of the effort. As will be described in the “accomplished” portion of the report, however, we have successfully accomplished the majority of the major goals of the proposal, and have made significant strides towards the remainder of the program goals.

Accomplishments: The uploaded pdf document provides a more detailed, narrative description of the program’s accomplishments. Below we summarize the accomplishments of our STIR program.
Program Thrusts
1) The design and engineering of optical resonances in polar semiconductor materials, demonstrating the ability to engineer strong optical absorption at the LO phonon energy.
2) Growth and fabrication of opto-phononic structures based on the designs developed in (1).
3) Demonstration of strong coupling and absorption of far-IR radiation at the LO phonon energy in a semiconductor material
4) Development of an experimental mechanism for measuring LO phonon populations in our fabricated structures.
5) Development of techniques for the generation of non-equilibrium LO phonon populations, and coupling of these LO phonons to free-space photons via intermediate surface modes.
Accomplished
- Demonstrated coupling of mid-IR light to polar materials at the LO Phonon energy (Thrusts 1-3).
- Demonstration of strong field enhancement in ultra-thin polar materials (Thrusts 1-3).
- First demonstration of mid-IR emission from the Berreman Mode, validating the concept of outcoupling vibrational energy to free space photons via epsilon near zero modes (Thrusts 1-3).
- First demonstration of the interaction of multimode antennas with ENZ materials (Thrusts 1-3).
- Demonstration of the ability to engineer the coupling of mid-IR light to polar materials at the LO phonon energy using optical nanostructures (Thrusts 1-3).
- Ability to engineer field enhancement and field distribution using monochromatic multimode antennas (Thrusts 1-3).
- Developed custom Raman setup for characterizing phonon populations in III-nitrides (Thrusts 4-5).
- Conceived two electrically-injected devices for generating large population of LO phonons that are compatible with the results in Thrusts 1 – 3 (Thrusts 4,5).

Training Opportunities: The STIR funding supported one graduate research assistant at UT Austin, one graduate research assistant at Notre Dame, and one undergraduate researcher at Notre Dame. The graduate students were supported each for 6 months and materials were supplied for the undergraduate researcher.
The graduate student at UT Austin, Leland Nordin, was a second year PhD student at the time of the STIR program. Over the course of the project, Leland developed expertise in the design and modeling of IR optical structures using Rigorous Coupled Wave Analysis software, calculating the optical properties of structures fabricated from the polar material AlN. Leland also developed expertise in the set-up and operation of mid-IR spectroscopic techniques, including angle-dependent reflectivity and thermal emission measurements. Leland continued to learn molecular beam epitaxy (MBE) growth, and now has sufficient expertise to grow epitaxial samples on his own. Finally, Leland developed communication skills, presenting his work on this project at the Electronic Materials Conference in the summer of 2017 at Notre Dame University.
The graduate student at Notre Dame, Owen Dominguez, was a third year student at the time of the program. Owen developed expertise in designing and modeling structures for confining optical fields in the Reststrahlen band. Owen learned modeling skills using rigorous coupled wave analysis and finite element methods. He also gained significant experience in nanofabrication, including electron beam lithography. Owen also furthered his expertise in mid-infrared characterization of optical nanostructures. He presented his work at the Conference on Lasers and Electro-optics and the 2017 Electronic Material Conference, improving his skills in presenting and discussing technical data.
The undergraduate student at Notre Dame, Zhaoyuan (Andy) Fang, was a sophomore student at the time of the program. Andy worked closely with the graduate student on the project and gained general experience in a research lab. He also developed specific skills in rigorous coupled wave analysis, mid-infrared characterization of optical materials, and communication skills. Andy's work on engineering the dispersion of the Berreman mode is being prepared for publication, further developing his communication and writing skills.

RPPR Final Report
as of 25-May-2018

Results Dissemination: This program has resulted in one published peer-review publication, two publications in preparation, and multiple conference presentations.
- "Mid-infrared epsilon-near-zero modes in ultra-thin phononic films" was published in Applied Physics Letters, with Leland Nordin (UT Austin student) and Owen Dominguez (Notre Dame student) as the two lead authors.
- "Monochromatic Multimode Antennas on Epsilon-Near-Zero Substrates", with lead authors Owen Dominguez (Notre Dame) and Leland Nordin (UT Austin) as lead authors, is currently under preparation and is expected to be submitted in early May.
- "Engineering the dispersion of the Berreman mode in polar materials," with lead authors Zhaoyuan Fang (Notre Dame), Owen Dominguez (Notre Dame), and Leland Nordin (UT Austin) as lead authors, is under preparation and is expected to be submitted in late May.

All three publications reference the ARO STIR award number.

In addition to the publications resulting from the STIR project, the work resulting from this project has been presented at multiple conferences. Conference presentations include:
- "Exciting Localized Modes in Polar Epsilon-Near-Zero Materials", O. Dominguez, L.J. Nordin, K. Feng, J. Lu, D. Wasserman, A.J. Hoffman, CLEO: Applications and Technology, Paper# JTh2A.113 (2017).
- "Strong Absorption from Berreman Modes in Thin AlN Films", L.J. Nordin, O. Dominguez, S. Dev, Z. Dong, A.J. Hoffman, and D. Wasserman, Electronic Materials Conference (2017).
- "Epsilon-near-Zero Mode Field Enhancement with Nanoantennas", O. Dominguez, L. Nordin, K. Feng, J. Lu, D. Wasserman and A. Hoffman, Electronics Materials Conference (2017).
- (Invited) "New Sources and Sensors for Mid- to Far-IR Optical Sensing", L. Yu, D. Jung, S. Dev, N. Yoon, L. Nordin, A.J. Hoffman, M.L. Lee, D. Wasserman, CLEO: Applications and Technology, Paper# AM2B.1 (2017).
- "Engineering the Coupling Between the Berreman Mode and Nanobar Antennas in Epsilon-near-zero Materials," O. Dominguez, L. Nordin, D. Wasserman, and A.J. Hoffman, CLEO (2018).

All of the above conference presentations reference support from ARO.

Finally, the work performed on the STIR program was discussed in numerous invited seminars over the past year. Professor Wasserman, as the recipient of an IEEE Distinguished Lectureship for the 2017-2018 term, had the opportunity to speak at a large number of institutions, and as part of his lecture, discussed the results from the STIR program. Specifically, this work was discussed during lectures at: University of Houston, University of Central Florida (CREOL), University of Virginia, Hampton College, Norfolk State University, City College London (UK), Chalmers University (Sweden), Hong Kong Polytechnic University, North Carolina State University, and University of Santa Barbara.

All presentations including results from the STIR program reference support from ARO.

Honors and Awards: Professor Wasserman was named an IEEE Photonics Society Distinguished Lecturer for the 2017-2018 term.

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: Daniel Wasserman
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI
Participant: Anthony Hoffman
Person Months Worked: 2.00 **Funding Support:**

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

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Conference Location: San Jose, CA
Paper Title: Exciting Localized Modes in Polar Epsilon-Near-Zero Materials
Authors: Owen Dominquez, Leland J. Nordin, Kaijun Feng, Junchi Lu, Daniel Wasserman, Anthony J. Hoffman
Acknowledged Federal Support: **Y**

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Paper Title: Engineering the coupling between the Berreman mode and nanobar antennas in epsilon-near-zero materials
Authors: Owen Dominguez, Leland J. Nordin, Daniel Wasserman, Anthony J. Hoffman
Acknowledged Federal Support: **Y**

Our STIR program focused on the III-nitride material AlN. This material system was ideal for our proposed exploration due to its strongly polar nature and its high energy LO phonons, which puts the AlN Reststrahlen band in the ~11-15μm range. This allows for a fundamental investigation of light/phonon interactions, with implications for the far-IR, but leveraging the more mature characterization techniques and equipment associated with the mid-IR (2-20μm) wavelength range. The primary focus of the STIR program was the demonstration of optical architectures designed to couple light into polar materials at those materials' LO phonon energy. Below we describe the primary results of the program, followed by a discussion of the investigation undertaken to integrate the results of the STIR program into active devices.

Coupling to Epsilon Near Zero Modes in Ultra-Thin Phononic Materials

In this thrust we investigated the coupling between free space light and the epsilon-near-zero mode supported in ultra-thin polar materials. Our ultimate goal was the demonstration of strong coupling of light into materials at the material LO phonon energy. While light cannot propagate in a polar material's Reststrahlen band, such materials have long been known to support surface modes at their interface with a dielectric material. These modes are typically referred to as Surface Phonon Polaritons (SPhPs). The interface between a phononic and dielectric material can sustain SPhP modes across the majority of the (typically narrow) Reststrahlen band. However, at lower energies, SPhP loss increases due to increased material absorption near the TO phonon energy. At the higher energy side of the narrow Reststrahlen band, approaching, but not at, the LO phonon, such modes can only exist when the real part of the material's permittivity is negative and larger in magnitude than the permittivity of the dielectric. At the LO phonon energy, the real part of the phononic material's permittivity is zero, and the imaginary part is small. However, traditional localized or propagating surface modes cannot be sustained at this epsilon-near-zero (ENZ) frequency, as the LO phonon is a longitudinal mode which will not couple to electromagnetic radiation, which is a transverse propagating mode.

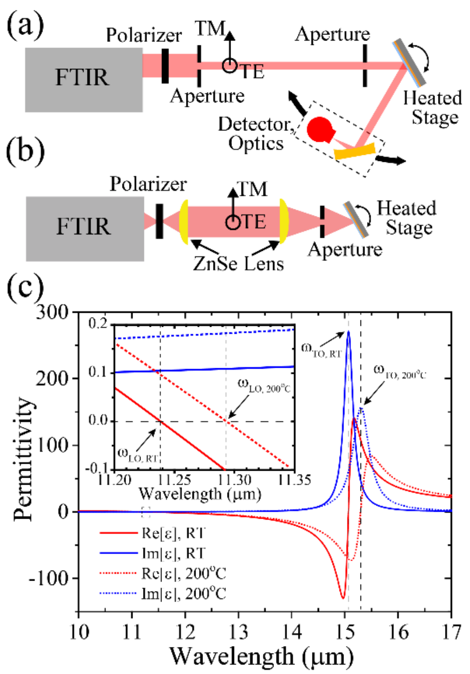

Figure 1: Experimental set-ups for angle-, polarization-, and temperature-dependent (a) reflection and (b) thermal emission spectroscopy. (c) Real (red) and imaginary (blue) permittivity of AlN at room temperature (solid) and 200 °C (dashed).

However, coupling to a polar material at, or near, the LO phonon energy is possible in thin layers of the material. In such a system, incident light couples to a leaky polariton mode generally referred to as the Berreman mode. The Berreman mode offers strong field enhancement in thin layers at near-ENZ frequencies, which for a phononic material, coincide with the material's LO phonon energy, offering a route towards strong coupling between light and longitudinal phonons via these hybridized modes. In this thrust we demonstrate coupling to such optical modes at mid-IR wavelengths in thin layers of AlN. Our samples are characterized experimentally by angle-, polarization-, and temperature-dependent infrared reflection and emission spectroscopy, and simulated using frequency domain mode matching software, which for planar structures reduces to a straightforward transfer matrix method (TMM). We demonstrated near perfect absorption and strong field localization in ultra-thin ($t < \lambda/100$) layers and mapped the Berreman mode dispersion as a function of layer thickness. In addition, we demonstrated that such structures can serve as

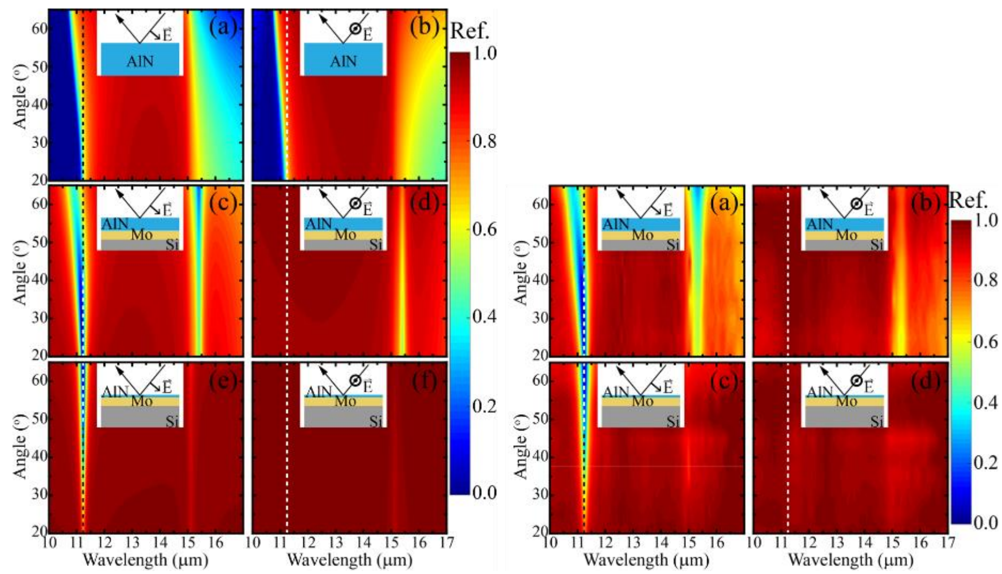

Figure 2: Left: TMM-simulated (a,c,e) TM- and (b,d,f) TE- polarized reflection from (a,b) semi-infinite AlN, (c,d) t=1.2μm thick AlN on 100nm Mo and (e,f) t=100nm thick AlN on 100nm Mo. Right: Experimental (a,c) TM- and (b,d) TE-polarized reflection from (a,b) t=1.2μm thick and (c,d) t=100nm thick AlN layers on Mo. Dashed vertical lines show the LO phonon energy. Insets show schematics of samples simulated and the polarization of the incident light.

polarization-dependent selective thermal emitters at or near the LO phonon energy, with minimal angular dispersion.

Figure 1 shows schematics of our experimental set-ups, as well as the permittivity of our AlN at both room temperature and at 200 °C (necessary for simulations of our thermal emission experiment). Figure 2 shows the simulated (left) and experimental (right) reflection data from the samples studied in this work. As shown in the insets, these samples consist of an optically thick Molybdenum layer upon which is deposited AlN of varying thicknesses (here we present AlN layer thicknesses of 1.2 μm and 100nm). In the simulated reflection, we include the reflection from semi-infinite AlN for comparison. Strong absorption features are observed at or near the LO phonon energy for the finite-thickness AlN, corresponding to coupling of incident light to the Berreman mode. For the thicker AlN, the mode is somewhat dispersive, quickly moving away from the LO phonon energy with increasing incidence angle. However, the ultra-thin AlN shows coupling to a remarkably non-dispersive mode, with strong and narrowband absorption largely tied to the LO phonon energy. Simulations of field intensities in the ultra-thin AlN show a factor 12 enhancement of the electric field strength in the AlN layer. Because the emissivity of a material is equivalent to its absorptivity, we are able to

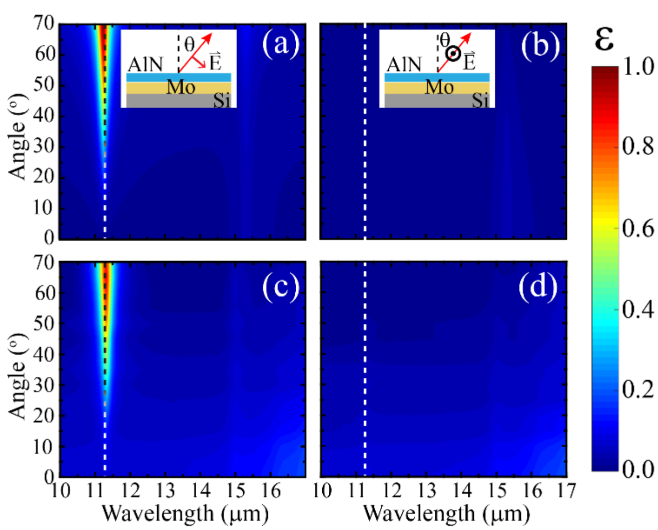

Figure 3: TMM-simulated and (c,d) Experimental (a,c) TM- and (b,d) TE-polarized emissivity (ε) from t=100nm thick AlN layer on Mo. Strong, narrowband and spectrally fixed emissivity peak at the AlN LO phonon energy (dashed vertical lines) is observed in the TM-polarized data, corresponding to thermally excited emission from the ENZ mode.

utilize the thin AlN layers as spectral selective thermal emitters. The experimental and simulated emissivity of the 100nm AlN sample is shown in Figure 3. Here we observe extremely narrowband thermal emission from the TM polarized Berreman mode, with virtually no angular dispersion.

The results from this first thrust are an important first step towards the demonstration of the coupling of LO phonons to free space photons. The results from this work were published in Applied Physics Letters in 2017.

Accomplishments (Thrust 1-3)

- *Demonstrated coupling of mid-IR light to polar materials at the LO Phonon energy.*
- *Demonstration of strong field enhancement in ultra-thin polar materials.*
- *First demonstration of mid-IR emission from the Berreman Mode, validating the concept of outcoupling vibrational energy to free space photons via epsilon near zero modes.*

Monochromatic Multimode Antennas on Epsilon Near Zero Substrates

Our work on planar AlN films demonstrated the viability of our larger goal of coupling vibrational modes to free space photons utilizing epsilon near zero modes. The advantage of utilizing the Berreman Mode lies in the relative ease of fabrication, as the mode is confined in an entirely planar structure. However, emission from these structures is strongest at large angles, and electrical access to the device becomes challenging, as the addition of any metal to the structure alters the optical modes. Thus, in parallel to investigating the Berreman Mode, we also explored the potential for antenna-coupling of free space photons to AlN at the LO phonon energy. In this work we demonstrate coupling between antenna modes and the epsilon near zero Berreman mode for a system consisting of metallic rod antennas fabricated upon a subwavelength AlN layer above a Mo groundplane. We show that antenna modes pin at an effective zero-index spectral position. For increasing antenna lengths, we are able to observe pinning of the fundamental antenna mode, as well as the two subsequent harmonics, over a narrow wavelength range, resulting in what is effectively a monochromatic, multimode response for the coupled system. The response of the coupled system is observed experimentally, and simulated using finite element methods, Fig. 4. Additionally, finite element models indicate a significant (45x) enhancement of the local fields on resonance. A coupled resonator model is developed to intuitively model the optical response of the system, shown in Fig. 4 (right). These results offer a mechanism for engineering the optical

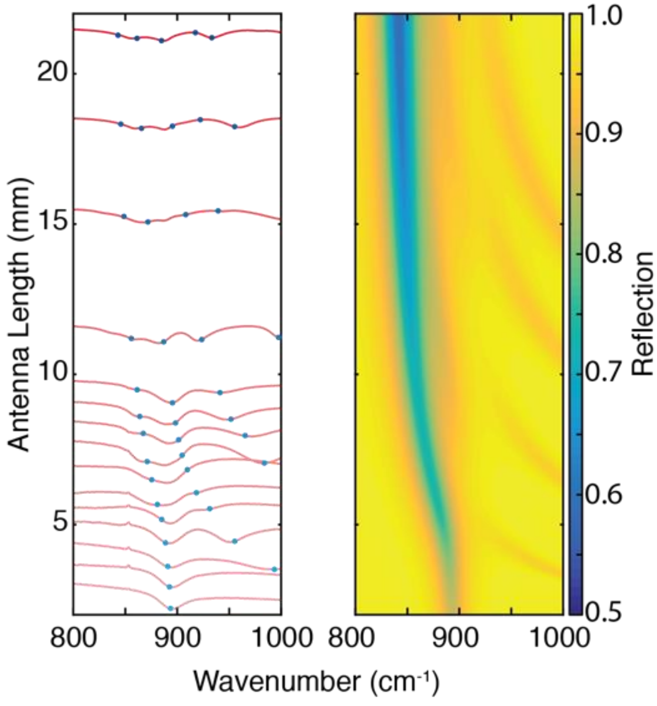

Figure 4: (left) Measured reflection spectra versus nanoantenna length offset such that the average reflectance is at the measured antenna length. The dots indicate the spectral location of modes using a coupled harmonic oscillator model. (right) Reflection versus wavenumber and antenna length calculated using a coupled harmonic oscillator model assuming coupling between each antenna mode and the Berreman mode. The uncoupled Berreman mode is at 895 cm^{-1} as seen for the shortest antenna lengths.

response of antenna systems by engineering the local dielectric environment, with potential applications in sensing, infrared optoelectronics and thermal emission control.

Accomplishments (Thrust 1 - 3)

- *First demonstration of the interaction of multimode antennas with ENZ materials.*
- *Demonstration of the ability to engineer the coupling of mid-IR light to polar materials at the LO phonon energy using optical nanostructures.*
- *Ability to engineer field enhancement and field distribution using monochromatic multimode antennas.*

Characterizing and generating non-equilibrium phonon populations

Our ultimate goal is the generation of photons from non-equilibrium populations of optical phonons. Developing methods for (1) characterizing and (2) generating non-equilibrium populations of LO phonons is needed for this end-goal. Our approach to characterizing phonon populations is to measure the Stokes and anti-Stokes components of scattered radiation while a device is under operation; the phonon population is related to the ratio of the Stoke to anti-Stokes signal. We therefore developed a custom Raman spectroscopy system compatible with electrically-active devices comprising a 785 nm Raman laser, appropriate edge filters, a broadband beamsplitter, a 20 X objective, and a high-resolution fiber-coupled grating spectrometer with a liquid nitrogen cooled CCD. The setup is shown in Fig. 5. We have used this custom setup to measure the Raman spectrum (Stokes and anti-Stokes) of a thin film of AlN. We are upgrading our Raman setup with a second Raman laser at 532 nm and a higher power objective that will enable us to characterize phonon generation in small active regions.

Over the course of this program, we have developed two approaches to optical phonon generation in electrically-injected devices in III-nitride devices. The first is to implement a Schottky diode using single-crystal, n-type GaN. Here, majority charge carriers are injected into the high-field reverse biased GaN depletion region at energies above the GaN conduction band edge. LO phonons are then generated as these high energy carriers scatter back to the GaN conduction band edge. The generation of optical phonons in reverse-biased Schottky diodes is a known outcome, and an effect that has been studied for well over a decade. Our second approach, is to design (Al)GaN hetrostructure devices with intersubband transitions at the GaN LO phonon energy. An exemplary design comprises AlN/GaN/AlN resonant tunneling devices (RTDs) with intersubband states separated by the GaN LO phonon energy. Injection into the RTD's excited state will allow for a phonon-mediated intersubband transition (IST) and the generation of a LO phonon.

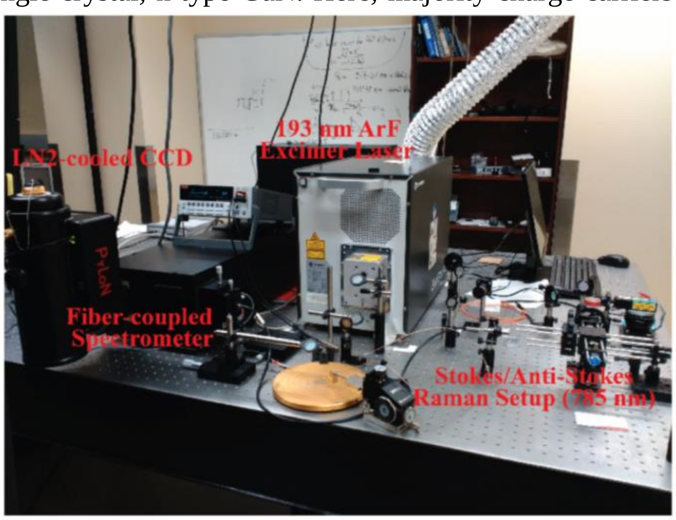

Figure 5. Photograph of the custom Raman spectroscopy setup capable of measuring the Stokes and anti-Stokes components of scattered light. Also shown is a 193 ArF Excimer laser that can be used to optically excite wide bandgap semiconductors and generate non-equilibrium phonon populations.

The high current densities allowed in III-nitride devices will ideally offer the potential for significant generation of non-equilibrium phonons.

Accomplishments (Thrust 4 and 5)

- *Developed custom Raman setup for characterizing phonon populations in III-nitrides*
- *Conceived two electrically-injected devices for generating large population of LO phonons that are compatible with the results in Thrusts 1 – 3.*