

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA, 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.
PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.

1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE Technical Report	3. DATES COVERED (From - To) -
-----------------------------	------------------------------------	-----------------------------------

4. TITLE AND SUBTITLE Ultrafast Spectroscopic Noninvasive Probe of Vertical Carrier Transport in Heterostructure Devices (Second year of three year progress report)	5a. CONTRACT NUMBER
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER

6. AUTHORS Grace Metcalfe, Blair Connelly, Stefan Svensson	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES Army Research Laboratory (ARL) 22,257.00 2800 Powder Mill Road Adelphi, MD 20783 -1138	8. PERFORMING ORGANIZATION REPORT NUMBER
--	--

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	10. SPONSOR/MONITOR'S ACRONYM(S) ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S) 62638-SE-DRI.1

12. DISTRIBUTION AVAILABILITY STATEMENT
Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT
Optical-based approaches have the advantage of being noninvasive and nondestructive, and can usually be performed without modification to the sample. We propose to merge two ultrafast spectroscopy methods, pump-probe spectroscopy and time-domain THz spectroscopy (TDTS), to realize a femtosecond-resolution double-pump-probe spectroscopy technique tunable from the visible through long-wave infrared (LWIR) wavelength range. Our approach allows us to examine not only the transient concentration due to an appropriate pump pulse, but also the time-resolved THz radiation induced by the probe pulse. By analyzing the resulting kinetic profiles, one can obtain

15. SUBJECT TERMS
Ultrafast spectroscopy, carrier transport, noninvasive probe

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE	UU		Grace Metcalfe
UU	UU	UU			19b. TELEPHONE NUMBER
					301-394-2864

Report Title

Ultrafast Spectroscopic Noninvasive Probe of Vertical Carrier Transport in Heterostructure Devices (Second year of three year progress report)

ABSTRACT

Optical-based approaches have the advantage of being noninvasive and nondestructive, and can usually be performed without modification to the sample. We propose to merge two ultrafast spectroscopy methods, pump-probe spectroscopy and time-domain THz spectroscopy (TDS), to realize a femtosecond-resolution double-pump-probe spectroscopy technique tunable from the visible through long-wave infrared (LWIR) wavelength range. Our approach allows us to examine not only the transient concentration due to an appropriate pump pulse, but also the time-resolved THz radiation induced by the probe pulse. By analyzing the resulting kinetic profiles, one can obtain critical information about the internal electric fields, radiative and non-radiative recombination rates, carrier drift and diffusion times, and electron and hole velocities.

TITLE: Ultrafast Spectroscopic Noninvasive Probe of Vertical Carrier Transport in Heterostructure Devices (Second year of three year proposal)

PRINCIPAL INVESTIGATORS: Drs. Blair Connelly (RDRL-SEE-I), Grace Metcalfe (RDRL-SE), Stefan Svensson (RDRL-SEE-I)

OBJECTIVE: To investigate the vertical carrier transport properties in Army-relevant semiconductor materials and optoelectronic (OE) devices by developing and applying ultrafast optical spectroscopy techniques.

APPROACH: Optical-based approaches have the advantage of being noninvasive and nondestructive, and can usually be performed without modification to the sample. We propose to merge two ultrafast spectroscopy methods, pump-probe spectroscopy and time-domain THz spectroscopy (TDTS), to realize a femtosecond-resolution double-pump-probe spectroscopy technique tunable from the visible through long-wave infrared (LWIR) wavelength range. Our approach allows us to examine not only the transient concentration due to an appropriate pump pulse, but also the time-resolved THz radiation induced by the probe pulse. By analyzing the resulting kinetic profiles, one can obtain critical information about the internal electric fields, radiative and non-radiative recombination rates, carrier drift and diffusion times, and electron and hole velocities.

RESULTS: In the second year, we successfully met our Q6 through Q8 goals of incorporating electrical testing capabilities into our system, investigating OE devices under operating conditions, extending the capabilities of our system into the IR range, and investigating new OE devices. We have made significant progress towards our Q5 goal of realizing a double-pump-probe system by designing and demonstrating a real-time THz acquisition scheme and optimizing our spectroscopy technique to enhance the detection sensitivity of THz generated by the probe beam. We expect completion of this milestone in Q9. We have also already made significant progress towards realizing third year goals of extending our capabilities into the IR range through experimental verification of our near infrared (NIR) pump-probe system and construction, initial testing and redesigning components of our optically-gated IR PL up-conversion setup, and of redesigning and growing IR transport measurement structures.

To analyze and improve our ultrafast spectroscopy technique to characterize increasingly complex heterostructures under operating conditions, we investigated the carrier dynamics in multiple quantum well (MQW) *p*-GaN/ *i*-InGaN/GaN MQW layer/ *n*-GaN solar cell structures (50 2.2-nm thick In_{0.2}Ga_{0.8}N wells and 4.9-nm thick GaN barriers). Transport was studied across numerous GaN/InGaN interfaces where the conduction and valence bands across the wells present a “sawtooth” potential resulting from large polarizations at the InGaN/GaN hetero-interfaces. Measurements in the MQW sample were made as a function of probe wavelength (above and below the well band edge), pump intensity and electrical bias (Fig. 1). Our 2nd year accomplishment of incorporating electrical testing capabilities required both successful integration into the complex geometry of our experimental setup and fabrication of more advanced InGaN/GaN devices.

When we probed the MQW InGaN sample above

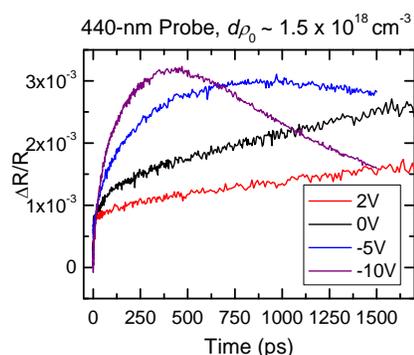


Figure 1: Differential reflection on MQW InGaN/GaN double heterostructure vs. bias.

the band gap, the curves demonstrated a rise to a peak followed by a slow decay. The data showed a clear dependence of the signal on electrical bias voltage (varied between +2 V and -10 V) with the peak location occurring at an earlier time when the reverse bias was increased, increasing the electric field across the *i*-layer and therefore carrier drift velocity. By modeling screening effects on the pump-probe signal detailed in studies of similar structures (InGaAs/GaAs and Ge/SiGe MQW *p-i-n* structures) [1, 2], we estimated a fast ~1-ps cumulative escape time from the wells, followed by a ~10-ps drift time for electrons and >500-ps drift time for holes across the *i*-InGaN/GaN MQW layer under -10 V bias. These slow time scales (relative to transport times in bulk InGaN [3]) demonstrate that the quantum wells and “sawtooth” potential significantly slow the carrier transport. Additional modeling efforts of the physical transport processes are in progress.

To study vertical transport properties in the IR, we are investigating two time-of-flight approaches by optically injecting carriers near the surface of an InAsSb *nBn* detector structure and detecting the arrival of carriers a known distance from the surface using (1) an optical probe of an ~2- μm bandgap GaInSb buffer layer below the detector structure and (2) photoluminescence (PL) from a “marker” well, with a narrower bandgap than the bulk material, embedded in the absorber layer. For approach (1), the visible pump-probe setup was expanded to operate in the NIR range, and we have demonstrated the operability of the system with a differential reflection result on the GaInSb buffer layer with ~200-fs temporal resolution. Optical probing from the backside of the sample necessitated considerable changes to the sample design. In the previously-used GaInSb graded layer, significant absorption of the ~40-nm bandwidth probe beam would occur in the graded layer, which obscured the differential absorption in the targeted buffer layer. We therefore modified the sample design to utilize an Al-containing AlInSb grading to widen the bandgap of the graded layer and provide optical access to the buffer layer from the backside of the sample (Fig. 2). This test structure has been successfully grown.

Observation of up-conversion of the 6- μm PL to ~1.03 μm for approach (2) has been complicated by a large background noise level at our InGaAs detector, but we are implementing several approaches to mitigate this challenge. The spatial overlap of the PL and gating pulse on the non-linear AgGaS₂ crystal has been improved by moving to a co-linear geometry. Further improvements to the spatial overlap consist of utilizing a longer focal length/smaller numerical aperture mirror(s) to focus the PL on the AgGaS₂ crystal at a smaller angle (telescoping the beam similar to a Cassegrain design), trying toroidal mirrors in the place of the off-axis parabolic to achieve a tighter focus of the PL, and acquiring a new cooled low-noise InGaAs PMT.

REFERENCES:

1. Claussen, S.A., et al., Optics Express, 2010. **18**(25): p. 25596-25607.
2. Wang, H.S., et al., IEEE Journal of Quantum Electronics, 1997. **33**(2): p. 192-197.
3. Connelly, B.C., et al., physica status solidi (c), 2014. **11**(3-4): p. 682-685.

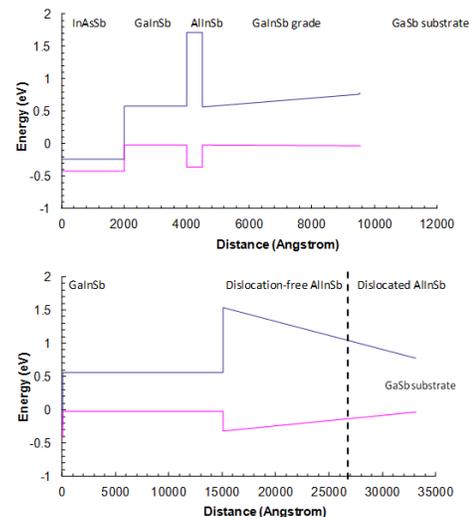


Figure 2: (top) Bandgap profiles of a Ga-graded on GaSb with a GaInSb VS for 2 μm absorption. A possible AlInSb barrier-layer is also shown as well as an InAsSb layer for LWIR absorption. (bottom) Bandgap profiles of the selected Al-graded on GaSb with a GaInSb VS for 2 μm absorption.

TRANSITIONS:

1. B.C. Connelly *et al.*, "Ultrafast Carrier Transport in an InGaN/GaN Multiple Quantum Well *p-i-n* Heterostructure using Differential Reflection," *submitted to the Conference on Lasers and Electro Optics (CLEO)*, May 2015.
2. B.C. Connelly *et al.*, "Time-Resolved Electroabsorption Measurement of Carrier Velocity in Inverted Polarity, In_{1-x}Ga_xN/GaN Heterostructures due to Internal Electric Fields," *International Conference on the Physics of Semiconductors (ICPS)*, August 10-15, 2014.
3. N.T. Woodward *et al.*, "The Origin of Lateral Electric Fields in *c*-plane III-V Nitrides," *International Conference on the Physics of Semiconductors (ICPS)*, August 10-15, 2014.
4. B.C. Connelly *et al.*, "Time-resolved electroabsorption measurement of carrier velocity in inverted polarity, In_{1-x}Ga_xN/GaN heterostructures due to internal electric fields," *physica status solidi c* 11(3-4), 682-685 (2014).
5. N.T. Woodward *et al.*, "Anomalous terahertz emission in *c*-plane InN due to in-plane transport," *physica status solid c* 11(3-4), 686-689 (2014).
6. B.C. Connelly *et al.*, "Time-Resolved Electroabsorption Measurement of Carrier Velocity in Inverted Polarity, In_{1-x}Ga_xN/GaN Heterostructures due to Internal Electric Fields," *International Conference on Nitride Semiconductors (ICNS)*, B12.01 2013.
7. N.T. Woodward *et al.*, "Anomalous Terahertz Emission in *c*-plane InN due to In-plane Transport," *International Conference on Nitride Semiconductors (ICNS)*, B12.05 2013.
8. B.C. Connelly *et al.*, "Temperature-Dependent Carrier Transport in a *p*-GaN/*i*-InGaN/*n*-GaN Solar Cell Heterostructure using Ultrafast Spectroscopy," *Conference on Lasers and Electro Optics (CLEO)*, CTh1M.7, 2013.
9. N.T. Woodward *et al.*, "Polarization and Interface Effects on THz Emission from *c*-plane InGaN/GaN Heterostructures," *March Meeting of APS*, Y23.00010, 2013.
10. B.C. Connelly *et al.*, "Time-Resolved Electroabsorption Measurement of Electron Velocity in InGaN Heterostructures due to Internal Electric Fields," *March Meeting of APS*, T23.00010, 2013.