
**NON-LINEAR OPTICAL STUDIES OF IR
MATERIALS WITH INFRARED FEMTOSECOND
LASER**

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1.0 SUMMARY

Intense femtosecond mid-IR (MIR) pulses from 2 - 4 micron wavelength at high repetition rate were used to explore non-linear effects into various mid infrared (MIR) materials like germanium and indium antimonide, including laser induced damage, defect generation, laser induced periodic structure (LIPSS) formation etc. Indium Antimonide, an important IR detector material, was studied with 2 and 1.55 micron femtosecond MIR lasers. LIPSS like structures were generated by 2 micron pulses. MIR studies with Ge revealed that it supports formation of two different types of LIPSS, the low spatial frequency and the high spatial frequency LIPSS (LSFL and HSFL, respectively), depending on surface fluence of the MIR femtosecond pulses, with different mechanisms. LSFL generation mechanism points towards creation of surface plasmons by the generation of carriers at and near the surface of the semiconductors with intense femtosecond light pulses, essentially turning them into metallic state instantaneously. Then, under successive pulse irradiation, the light pulses create enough surface roughness to form surface spatial frequencies needed to couple the surface plasmon polaritons (SPP) with the incoming light and generate periodic energy density profiles, which results in material removal and forms the LSFL morphology on the surface. HSFLs are created by short pulses at lower fluences, where the material never turns quite metallic, although its dielectric function is heavily modified by carrier generation by the short pulses. The short pulses are scattered from the roughened surface regions to create surface scattered waves (SSW), which, then interacts with the incoming waves, to accentuate energy absorption on the surface with specific spatial frequencies, whose spatial periods are significantly smaller than light wavelengths. These SSW then interact with other features on surface to create complex structures, in case of intense MIR irradiation on Ge. InSb also showed LSFL creation and deep surface damage under MIR irradiation. To study defect states created in InSb, short pulses at 1.55 micron wavelength were used to irradiate InSb surfaces when the defect states were simultaneously probed with Scanning Tunneling Microscopy (STM). It was found that the short pulse generated carriers can change the surface potential in the neighborhood of atomic defect states of InSb surfaces.

2.0 INTRODUCTION

Sources and detectors with suitable optical properties at infrared (IR) wavelengths are very important to various defense and civilian advanced technological applications. Some of the main applications are highlighted below:

- *Thermal imaging sensors, heat sensors, guidance sensors in atmosphere include various infrared semiconductor materials, and enhancing the efficiency of these detectors in cost effective ways have direct impact on DoD mission critical projects*
- *With earth's background blackbody radiation peaking around 9 μm wavelength, and air glow peaking around 3 microns, femtosecond lasers at 3 – 10 microns can provide tremendous advancement in undetected point to point communication, directed energy propagation, etc.*
- *Many molecular excitation bands lie between 3 – 10 microns wavelengths. A femtosecond laser based LIDAR can be a powerful tool to study atmospheric pollution, and possibly detect chemical/bio weapon signatures in real time.*
- *IR sources and detectors have a wide ranging applications in public sector from night vision cameras on cars helping avoid accidents to air pollution/pathogen monitoring in critical areas indoors (hospitals) and outdoors (cities).*

Studies of non-linear optical properties of relevant materials at IR wavelengths have been limited, because of lack of intense IR laser sources and detectors. Among relevant past studies, pico-second laser matter damage thresholds in a limited regime were studied by Simanovskii *et al* [1]. Kartashov *et al* [2], Fuji *et. al* [3] and Xu *et. al* [4] studied non-linear propagation and filamentation with MIR short pulse lasers. Non-linear frequency conversion of MIR light in ZnO crystal was studied by Ghimire *et al* [5]; Zhu *et al* used frequency up-conversion into visible wavelengths to analyze molecular resonances in MIR [6]. THz generation has also been tried with near IR (NIR) and short wavelength IR (SWIR) short pulse laser interaction with semiconductors like InSb [7].

Under the program just completed, we undertook to study strong non-linear interaction of MIR short pulses with IR materials Ge and InSb, where we studied laser induced damage and surface structure formation and surface defect states interacting with short MIR pulses.

3.0 METHODS, ASSUMPTIONS, AND PROCEDURES

Intense femtosecond MIR pulses from 2 - 4 micron wavelength at a 1 kHz repetition rate were used to explore non-linear effects into various MIR materials like germanium and indium

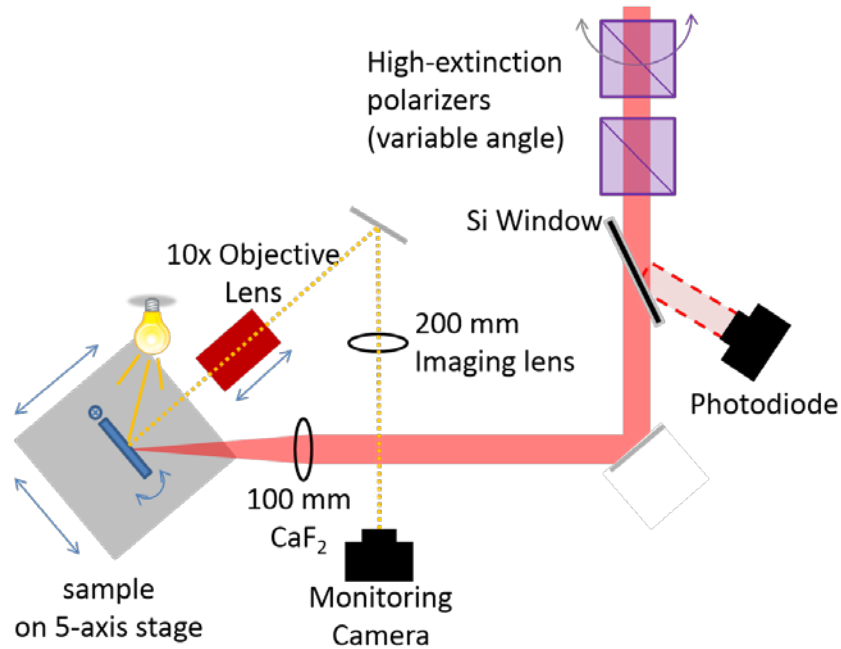


Figure 1: Setup of MIR Non-Linear Studies of Ge and InSb

antimonide, including laser induced damage, defect generation, laser induced periodic structure formation (LIPSS) etc. Indium Antimonide, an important IR detector material, was studied with 2 and 1.55 micron femtosecond MIR lasers.

Scanning Tunneling Microscopy (STM) study of InSb has also taken effect in collaboration with Professor Jay Gupta of Ohio State University (OSU) Physics, where we observe various defect states of surface of $\langle 110 \rangle$ InSb at 6k temperature. We have obtained preliminary results on how InSb surface defects forms under ultrafast pulse irradiation in ultra-high vacuum environment.

4.0 RESULTS AND DISCUSSION

LIPSS like structures were generated by 2 micron pulses, but with 45 degree angle of incidence, they were found to make a V type structure with the internal V angle of ~ 90 degrees, with peak of 'V' pointing to the laser k vector. Femtosecond pulse interaction with atomic scale resolution was also studied in InSb surfaces where characteristics of atomic defect states were strongly effected by 1.55 micron short pulses. It was found that Ge supports formation of two different types of LIPSS, the LSFL and the HSFL, depending on surface fluence of the MIR femtosecond pulses, with different generation mechanisms. LSFLs are formed with relatively higher laser fluences, when due to the influence of high density carrier generation by intense MIR pulses, Ge surface is metallized rapidly, which then supports surface plasmon modes; as surface is roughened by successive such pulses, the laser can be effectively coupled to the surface plasmon polaritons (SPP), which propagates and carries laser energy, resulting in surface structure enhancement which supports the SPP modes. As many as 7 pulses were used in MIR to generate LSFLs, with angle of incidence from 0 – 77 degrees, and with *s*- and *p*-polarized light pulses. HSFL formation in germanium was also studied with varying angle of incidence and

polarization. The period of these structures varied from $\lambda/3$ to $\lambda/8$. A modified surface-scattering model, where it is stipulated that surfaces roughened by previous laser pulses act as scattering site, creating surface scattered waves (SSW) in all directions, which vectorially interfere with incoming light wave to create these structures, mostly parallel to the laser polarization direction, including Drude excitation and the optical Kerr effect explains the spatial period scaling of HSFL across the MIR wavelengths.

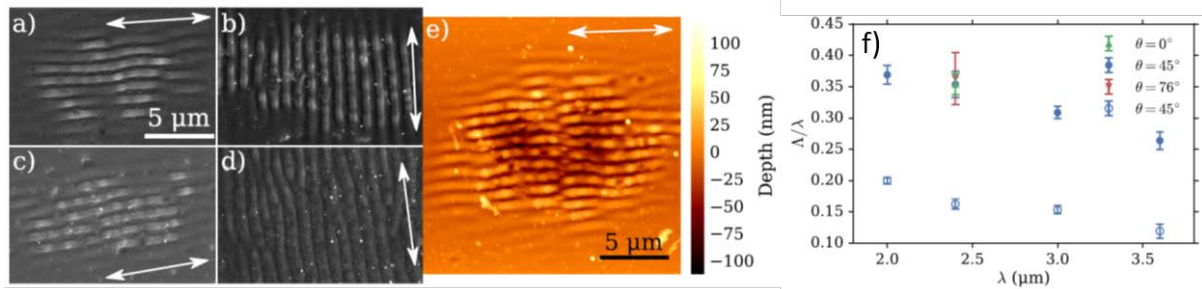


Figure 2: (a)–(d) Example SEM Images of Central HSFL Comparing p- and S-polarization HSFL at 3.6 Microns (a, b, respectively) and 3.0 Micron Wavelengths (c, d, respectively). All Damage Spots were formed using 100 Pulses at an Angle of Incidence 45° . The Orientation of the HSFL is found to Remain Parallel to the Polarization (arrows). No Significant Difference in Period is observed. (e) AFM Image of Central HSFL on Ge Showing the Surface Morphology (same laser conditions as in (a)). (f) HSFL Period Δ (normalized to the laser wavelength) as a Function of k . The HSFL were Produced using 100 pulses of P-Polarized Light. Sipe’s Model of HSFL Formation would Predict a Constant Δ/λ Across Wavelengths; the Observed Deviations are Consistent with the Model which Considers Modification of the Refractive Index of Ge after Laser-Excitation. Also Plotted are the Shortest Consistently Observed Periods of the Peripheral HSFL (unfilled circles)

Understanding of LIPSS formation with MIR light pulses have been steadily progressing, and our group is first to demonstrate HSFL (LIPSS period $\leq \lambda/4$) formation on germanium [8], [9] for 2 – 3.6 micron wavelength femtosecond pulses, which shows how femtosecond laser with non-normal incidence interact with semiconductor surfaces to excite SPPs that can propagate in a direction other than along laser k -vector component on the surface (s-pol), and can carry significant energy away from the peak of laser focus. As Ge is a very attractive MIR material (with transparency from 2 - 12 microns), with high third order non-linearity ($\chi^{(3)}$) it is considered a candidate material for next generation opto-electronic and opto-plasmonic devices. In a more recent paper [10] the formation of HSFL in Ge using sub-bandgap, MIR, ultra-short laser pulses was systematically studied while varying multiple laser parameters including wavelength, angle of incidence and polarization. The formation of the central HSFL is consistent with a Sipe-Drude-Kerr (SDK) surface scattered wave model of LIPSS formation. This is in contrast to the LSFL formation mechanism in the same wavelength regime, where higher fluences generate a metallic surface layer, allowing for the excitation of SPPs and their subsequent interference with the incident laser light. The inclusion of Drude excitation in the SDK model allows for an estimate of the electron density after laser-excitation. These estimates were significantly influenced by the Kerr effect because of the highest third-order susceptibility of Ge. With these effects taken into account, it was found that central HSFL on Ge seem to form optimally when

the electron density is approximately half of the surface critical density. Additionally, a mechanism was proposed for the formation of HSFL near the boundary region of damage spots, which stipulates that SSWs reflect off of the sharp boundary. The subsequent interference between the incident light, the incoming SSW and the RSSW seems to form the peripheral HSFL with spatial frequencies double that of central HSFL. Finally, two possible mechanisms were introduced to qualitatively explain the formation of the amorphous layer in the HSFL region. To identify the mechanism for quantitative agreement with observations, further studies at different pulse numbers as well as a measurement of the TPA coefficient for Ge in the MIR and femtosecond regimes at TW/cm² intensities are needed. We are working on understanding various aspects of surface engineering of Ge with MIR femtosecond pulses, including HSFL formation in Ge, including smallest wavelength where HSFL ceases to form.

InSb is another extremely relevant MIR detector material for which ultra-fast laser surface interaction effects is not well known. We have performed extensive non-normal interaction studies with InSb <100> single crystal using 2 micron 90 fs laser pulses, which reveal ‘wake-like’ surface structures oriented along specific crystal axis. We believe that such an effect have previously not been seen in literature. We were able to generate LIPSS ridges parallel and perpendicular to the polarization of 45° angle of incidence pulses, with ~1.6 and ~2.5 micron periods respectively. We are actively investigating these phenomenon.

STM study of InSb has also taken effect in collaboration with Professor Jay Gupta of OSU Physics, where we observe various defect states of surface of <110> InSb at 6k temperature. We have obtained preliminary results on how InSb surface defects forms under ultrafast pulse irradiation in ultra-high vacuum environment. It turns out that when the STM tip, with a bias between it and the surface it scans, is brought near the surface of a semiconductor, a tip induced band bending creates a small depletion region on the surface right below the tip [11], which can be mitigated by photon induced electron-hole carrier generation. This mitigation effect is called surface photo-voltage effect. Under specific atomic defect sites on <111> InSb surfaces, our efforts have detected unusually large surface photo-voltage effects due to 1550 nm femtosecond pulses. A manuscript is being prepared on this finding.

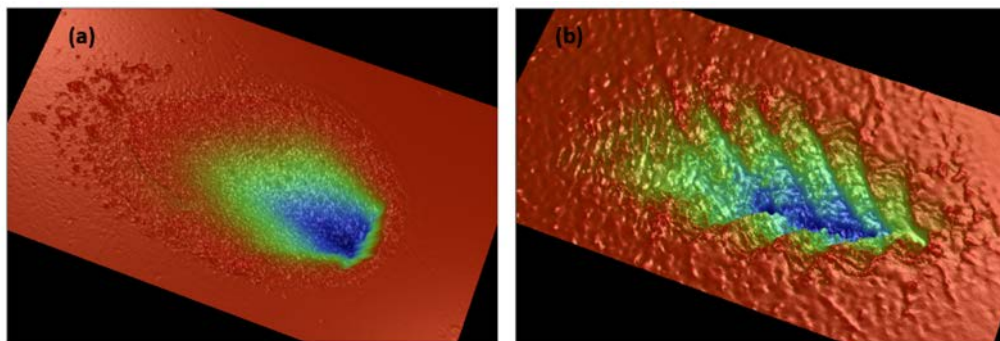


Figure 3: Insb Damage (a) and LIPSS formation (b)

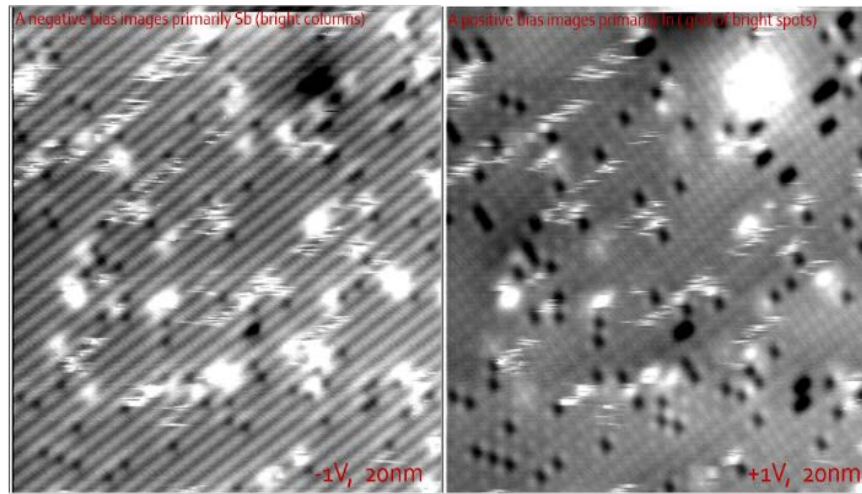


Figure 4: (Left) A Negative Bias Images Primarily Sb Atoms (bright diagonal columns) Bright Blobs and Triangles and Dark Circles are Surface Layer Defects or Adatoms. Bright streaks are mobile Adatoms, Imaged as they Move across the Sample. (Right) A Positive Bias Images Primarily in Atoms (bright grid-like spots). Bright Correspond to Bright Defects in Negative Bias Image. Dark Rectangles Correspond to Dark Circles at Negative Bias. The Streaks are the same. Each Image is 20 x 20 nm in Area.

5.0 CONCLUSIONS

In Conclusion, we have studied strong non-linear interaction in MIR regime between femtosecond light pulses and solid states of Ge and InSb, two very important IR materials, both for defense and commercial applications. We have established wavelength scaling of damage features formed on surfaces due to intense MIR pulse irradiation and we also have performed preliminary study of atomic scale surface defect interaction with short pulses of light. Future efforts are planned to explore wavelength scaling of InSb laser damage and surface features and how the successive pulses change InSb surface states.

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APPENDIX A — Publications and Presentations

Journal Articles:

1. D.R. Austin, K.R.P. Kafka, Y.H. Lai, Z. Wang, K. Zhang, H. Li, C.I. Blaga, A.Y. Yi, L.F. DiMauro, and E.A. Chowdhury, *High spatial frequency laser induced periodic surface structure formation in germanium under strong mid-IR fields*, J. Appl. Phys. **120**, 143103 (2016).
2. K. R. P. Kafka, D. R. Austin, H. Li, A. Y. Yi, J. Cheng, and E. A. Chowdhury, *Time-resolved measurement of single pulse femtosecond laser-induced periodic surface structure formation induced by a pre-fabricated surface groove*, Optics Express **23**, 19432 (2015).
3. Drake R. Austin, Kyle R. P. Kafka, Simeon Trendafilov, Gennady Shvets, Hui Li, Allen Y. Yi, Urszula B. Szafruga, Zhou Wang, Yu Hang Lai, Cosmin I. Blaga, Louis F. DiMauro, and Enam A. Chowdhury, *Laser induced periodic surface structure formation in germanium by strong field mid IR laser solid interaction at oblique incidence*, Optics Express **23**, 19522 (2015).

Conference Proceedings/Presentations:

- 7.0 E. Chowdhury, K.R.P. Kafka, D.R. Austin, K. Werner, N. Talisa, B. Ma, C.I. Blaga, L.F. DiMauro, H. Li, and A. Yi, *Ultra-fast bandgap photonics in mid-IR wavelengths*, Proc. SPIE 9835, p. 983519 Ultrafast Bandgap Photonics (2016).
- 8.0 Enam Chowdhury, Kyle R. P. Kafka, Robert A. Mitchell, Kevin Werner, Noah Talisa, Hui Li, Allen Yi, Douglass W. Schumacher, *Single-shot femtosecond laser ablation of copper: experiment versus simulation*, Paper 9632-26, SPIE Laser Damage 2015, Boulder CO.
- 9.0 Kyle R. P. Kafka, Enam Chowdhury, Raluca A. Negres, Christopher J. Stolz, Jeffrey D. Bude, Andy J. Bayramian, Christopher D. Marshall, Thomas M. Spinka, Constantin L. Haefner, *Test station development for laser-induced optical damage performance of broadband multilayer dielectric coatings*, Paper 9632-51, SPIE Laser Damage 2015, Boulder CO.
- 10.0 Christopher J. Stolz, Lawrence Livermore National Lab. (United States); Kyle R. P. Kafka, Enam Chowdhury, The Ohio State Univ. (United States); Matthew S. Kirchner, *Broadband low-dispersion mirror thin film damage competition* Paper 9632-11, SPIE Laser Damage 2015, Boulder CO.

APPENDIX B – Abstracts

1. D.R. Austin, K.R.P. Kafka, Y.H. Lai, Z. Wang, K. Zhang, H. Li, C.I. Blaga, A.Y. Yi, L.F. DiMauro, and E.A. Chowdhury, *High spatial frequency laser induced periodic surface structure formation in germanium under strong mid-IR fields*, J. Appl. Phys. **120**, 143103 (2016).

Abstract: Formation of high spatial frequency laser induced periodic surface structures HSFL in germanium by 90 fs mid-IR pulses at a 1 kHz repetition rate with wavelengths between $\lambda = 2$ and 3.6μ was studied with varying angle of incidence and polarization. The period of these structures varied from $\lambda/3$ to $\lambda/8$. A modified surface-scattering model including Drude excitation and the optical Kerr effect explains the spatial period scaling of HSFL across the MIR wavelengths. Transmission electron microscopy shows the presence of a 30 nm amorphous layer above the structure of crystalline germanium. Various mechanisms including two photon absorption and defect-induced amorphization are discussed as probable causes for the formation of this layer.

2. K. R. P. Kafka, D. R. Austin, H. Li, A. Y. Yi, J. Cheng, and E. A. Chowdhury, *Time-resolved measurement of single pulse femtosecond laser-induced periodic surface structure formation induced by a pre-fabricated surface groove*, Optics Express **23**, 19432 (2015).

Abstract: Time-resolved diffraction microscopy technique has been used to observe the formation of LIPSS from the interaction of a single femtosecond laser pulse (pump) with a nano-scale groove mechanically formed on a single-crystal Cu substrate. The interaction dynamics (0-1200 ps) was captured by diffracting a time-delayed, frequency-doubled pulse (probe) from nascent LIPSS formation induced by the pump with an infinity-conjugate microscopy setup. The LIPSS ripples are observed to form asynchronously, with the first one forming after 50 ps and others forming sequentially outward from the groove edge at larger time delays. A 1-D analytical model of electron heating including both the laser pulse and SPP excitation at the groove edge predicts ripple period, melt spot diameter and qualitatively explains the asynchronous time-evolution of LIPSS formation.

3. Drake R. Austin, Kyle R. P. Kafka, Simeon Trendafilov, Gennady Shvets, Hui Li, Allen Y. Yi, Urszula B. Szafruga, Zhou Wang, Yu Hang Lai, Cosmin I. Blaga, Louis F. DiMauro, and Enam A. Chowdhury, *Laser induced periodic surface structure formation in germanium by strong field mid IR laser solid interaction at oblique incidence*, Optics Express **23**, 19522 (2015).

Abstract: LIPSS, or ripples, were generated on single crystal germanium after irradiation with multiple $3 \mu\text{m}$ femtosecond laser pulses at a 45° angle of incidence. HSFL and LSFL LIPSS, respectively, were observed for both s- and p-polarized light. The measured LSFL period for p-polarized light was consistent with the currently established LIPSS origination model of coupling between SPP and the incident laser

pulses. A vector model of SPP coupling is introduced to explain the formation of s-polarized LSFL away from the center of the damage spot. Additionally, a new method is proposed to determine the SPP propagation length from the decay in ripple depth. This is used along with the measured LSFL period to estimate the average electron density and Drude collision time of the laser-excited surface. Finally, full-wave electromagnetic simulations are used to corroborate these results while simultaneously offering insight into the nature of LSFL formation.

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