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**Understanding the femtosecond laser-solid interaction near and beyond the material damage threshold**

**Enam Chowdhury  
OHIO STATE UNIVERSITY THE**

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**05/23/2016  
Final Report**

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## **BRI: Understanding the femtosecond laser-solid interaction near and beyond the material damage threshold: *Final Report.***

### Abstract:

Under the recently completed BRI program, the PI has established a new femtosecond solid dynamics laboratory (FSD) dedicated to the study of the fundamentals of femtosecond laser damage as a function of various parameters, laser wavelength, pulsewidth, pulse number, experimental ambience (air/vacuum/other), polarization and angle of incidence, target solid material conductivity (metal/semiconductor/insulator), band gap, and surface morphology (nano/micro-structured surface), and the PI's team has worked synergistically in designing and performing experiments, analyzing results, and developing a unified femtosecond laser damage simulation framework, which allowed direct benchmarking of simulation results with experimental observation without any free parameters.

The brand new FSD Lab constructed under the BRI grant in the Physics Research Building at the Ohio State University has developed a wide ranging short pulse laser capabilities, from multi-milijoule pulses at kHz to single shot repetition rate, from pulsewidths 5 fs – 150 ps, from 200 nm to 4000 nm (~100 fs). FSD Lab has already established itself as a leading laboratory for studying short pulse laser damage under vacuum/air/other gas environments. The FSD began a collaboration with the High Average Power Laser System (HAPLS) program at LLNL in 2014 to help them select broadband ultra-damage resistant dielectric mirrors for its next generation 10 Hz petawatt laser amplifiers for the ELI-Beamline in the Czech Republic, whose first phase is already completed (please see support letter from LLNL group leaders). The lab was also selected as the laser induced damage threshold (LIDT) testing facility for the prestigious SPIE Laser Damage Competition 2015, where the top manufacturers of damage resistant optics in the world participate.

A comprehensive LIDT parameter exploration was performed, which resulted in presentations in five conferences, several conference proceedings, and a summary of results is being prepared to be submitted to Physical Review Letters titled "Near to mid IR femtosecond material damage threshold measurements". Single and multi-shot femtosecond laser damage threshold (fs-LDT) of materials have been studied across a range of band-gaps for *s*- and *p*-polarized light and it is found that conventional theoretical prediction on laser damage threshold based on Two Temperature Model (TTM) fails to capture the wavelength dependence of fs-LDT from near to mid IR.

A time-resolved diffraction microscopy technique has been developed in our lab to observe the formation of laser-induced periodic surface structures (LIPSS) from the interaction of a single femtosecond laser pulse (pump) with a nano-scale groove mechanically formed on a single-crystal Cu substrate (Yi Lab, OSU). The interaction dynamics (0–1200 ps) were captured by diffracting a time-delayed, frequency-doubled pulse (probe) from the nascent LIPSS induced by the pump using an infinity-conjugate microscopy setup. Understanding of LIPSS formation with mid IR light

pulses have been steadily progressing, and our group is first to demonstrate HSFL (LIPSS period  $< \lambda/2$ ) formation on germanium for 3 – 3.6 micron wavelength femtosecond pulses. Formation of low spatial frequency LIPSS (LSFL, with LIPSS period  $> \lambda/2$ ) with multiple femtosecond mid IR pulses of s and p polarization at oblique incidence angle showed that strong coupling between incident pulses and induced SPPs by multiphoton ionization is possible so much so that large amount of deposited energy is carried away from the most intense central region of the Gaussian focus [Austin et al Opt. Ex. 2015]. Formation of HSFL on Ge was also analyzed (submitted to APL), and it was found that instead of generation of surface plasmon polaritons, these nano-structures are formed by interference of incident pulses with surface scattered light generated by nano-roughness created from previous surface-pulse interactions. Surprisingly, such a phenomenon was not observed for silicon, and this is perhaps attributed to Ge having an order of magnitude larger  $\chi(3)$  than Si, which in Ge, results in the electron density to stay below critical density, allowing such an interaction to happen.

We have developed a new simulation technique for modeling the onset and evolution of laser damage due to the intense interaction of a laser and metal target. The simulation technique is based on the particle-in-cell (PIC) method for simulating plasmas and works by (1) implementing a Lennard-Jones pair-potential (LJPP) model within the PIC framework and (2) using a series of PIC simulations to follow target evolution over time-scales spanning six orders-of-magnitude, from the femtosecond scale of the laser interaction to the nanosecond scale of target evolution. To our knowledge, neither of these approaches has been demonstrated before. In the final phase of the BRI, we have bridged the gap between experimental and simulation results by performing benchmarking experiments, and were able to generate 2 micron diameter focal spot with few cycle pulses (5 – 35 fs). Such extremely spatio-temporally confined pulses interacted with single crystal Cu and generated craters for various fluence ranges, which provide straight forward templates for our PIC simulation to predict. We have also performed benchmarking simulations, which are producing crater depths, matching experimentally observed crater depths very well. To our knowledge, this is the first successful demonstration of benchmarking between experimental and theory/modeling efforts in the field of short pulse laser damage.

### **Final Report: Enam Chowdhury**

Under the current BRI program the PI has established a new femtosecond solid dynamics laboratory (FSD) dedicated to the study of the fundamentals of femtosecond laser damage, which has already established itself as a leading lab for studying short pulse laser damage. The FSD began a collaboration with the High Average Power Laser System (HAPLS) program at LLNL in 2014 to help them select broadband ultra-damage resistant dielectric mirrors for its next generation 10 Hz petawatt laser amplifiers for the ELI-Beamline in the Czech Republic, whose first phase is already completed (please see support letter from LLNL group leaders). The lab was also selected as the laser induced damage threshold (LIDT) testing facility for the prestigious SPIE Laser

Damage Competition 2015, where the top manufacturers of damage resistant optics in the world participate.

A comprehensive LIDT parameter exploration was performed, which resulted in presentations in four conference, several conference proceedings, and a summary of results is being prepared to be submitted to Physical Review Letters titled "Near to mid IR femtosecond material damage threshold measurements". Single and multi-shot femtosecond laser damage threshold (fs-LDT) of materials have been studied across a range of band-gaps for *s*- and *p*-polarized light and it is found that conventional theoretical prediction on laser damage threshold based on Two Temperature Model (TTM) fails to capture the wavelength dependence of fs-LDT from near to mid IR. Of particular interests are two anomalies, first—a large (up to 100%) increase in single shot *s*-LIDT vs *p*-LIDT (@ 45° AOI) in a range of wavelengths, where energy loss due to Fresnel reflection from zero order indices of refraction accounts for only 20-24% difference in LIDT for the wavelength ranges studied. This certainly points towards a dynamic effect in play by which

electrons accelerated by the *E*-field normal to surface for *p*-polarized pulses may cause the material to damage at lower fluences.

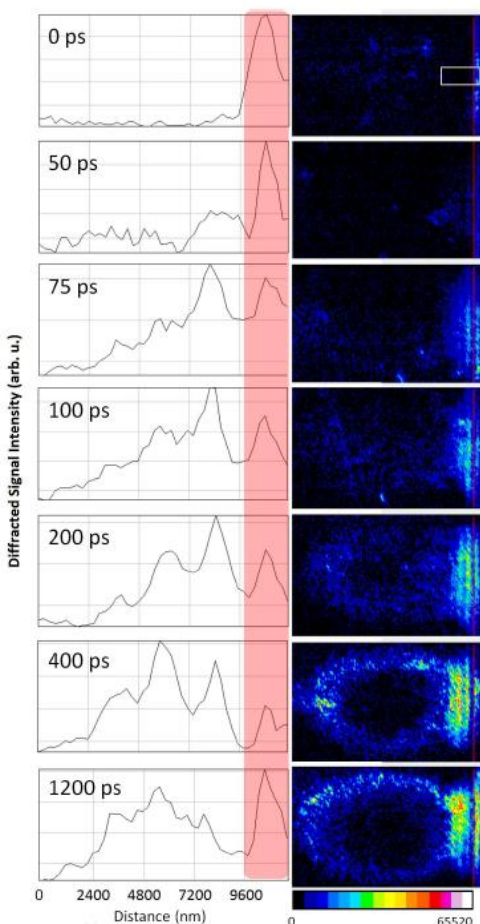


Figure. TRDM Images showing formation of LIPSS ridges

LIPSS were also generated on single crystal germanium after irradiation with multiple 3  $\mu\text{m}$  femtosecond laser pulses at a 45° angle of incidence (in collaboration with the Dimauro group, OSU). High and low spatial frequency LIPSS (HSFL and

A time-resolved diffraction microscopy technique has been developed in our lab to observe the formation of laser-induced periodic surface structures (LIPSS) from the interaction of a single femtosecond laser pulse (pump) with a nano-scale groove mechanically formed on a single-crystal Cu substrate (Yi Lab, OSU). The interaction dynamics (0–1200 ps) were captured by diffracting a time-delayed, frequency-doubled pulse (probe) from the nascent LIPSS induced by the pump using an infinity-conjugate microscopy setup. The LIPSS ripples were 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 surface plasmon polariton excitation at the groove edge predicts ripple period, melt spot diameter, and qualitatively explains the asynchronous time-evolution of LIPSS formation, which was observed by us for the first time.

LSFL, 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 surface plasmon polaritons (SPP) and the incident laser pulses. A vector model of SPP coupling was introduced to explain the formation of *s*-polarized LSFL away from the center of the damage spot. Additionally, a new method was proposed to determine the SPP propagation length from the decay in ripple depth. This was 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 were used to corroborate these results while simultaneously offering insight into the nature of LSFL formation.

Laser interaction with nano-structures was studied by exploring the laser-induced femtosecond damage thresholds of Au and Ag coated pulse compression gratings with submicron grooves. These gratings differ from conventional metal-on-photoresist pulse compression gratings in that the gratings patterns are generated by etching the fused silica substrate directly. After etching, the metal overcoating was optimized based on diffraction efficiency and damage threshold considerations. The experiment on these gratings was performed under vacuum for single-shot damage using 800 nm laser pulses ranging in duration from 30 to 200 fs. The single-shot damage threshold, below which there is a 0% probability of damage, was determined to be within a 400–800 mJ/cm<sup>2</sup> range. The damage threshold exhibited no clear dependence on pulse width, but showed clear dependence on gold overcoat surface morphology. This was confirmed by electromagnetic field modeling using the finite element method, which showed that non-conformal coating morphology gives rise to significant local field enhancement near groove edges, lowering the diffraction efficiency and increasing Joule heating. Large-scale gratings with conformal coating have been installed successfully in the 500 TW Scarlet laser system.

### Final Report: Douglass Schumacher:

#### (1) *Development of a new approach for modeling ultrafast laser damage*

Many theoretical tools have been developed over the last several decades for the study of ultrafast (femtosecond scale)

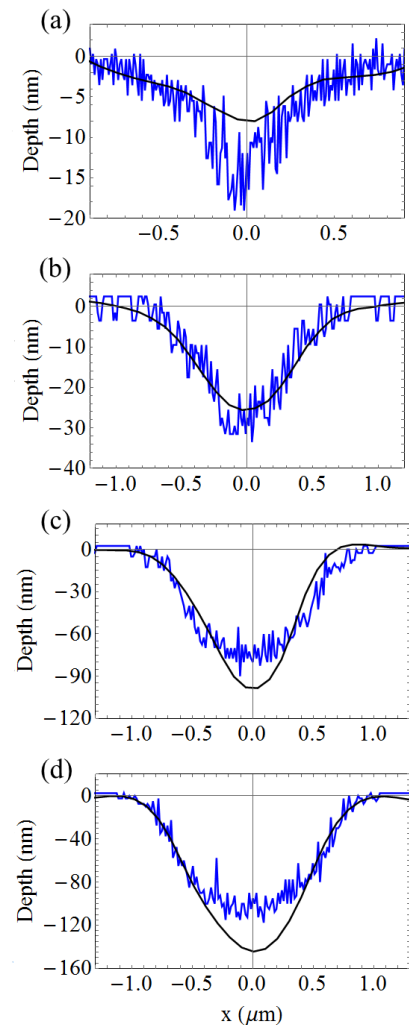


Figure. Shown are the crater depths produced from a single laser pulses measured experimentally (black) and predicted by simulation (blue) for fluences in J/cm<sup>2</sup> of (a) 1.8, (b) 2.8, (c) 5.5, and (d) 8.3.

laser damage. Many of these techniques fall into one of two categories: small spatial scale, ab-initio simulation techniques such as the molecular dynamics (MD) method; or large spatial scale, empirical, or rate equation models. None of these methods predict one of the primary and most important measures of laser damage – the damage crater morphology itself. We have developed the first simulation approach that can do this by combining the particle-in-cell (PIC) method with the Lennard-Jones pair potential (LJPP) model used by many MD codes to treat atomic motion after target heating. Because our method is based on PIC, it also naturally treats the initial laser-target interaction/heating as well. This LJPP PIC approach is completely new and took two years to develop. It was reported in 2015 in Optics Letters.

(2) *First model of laser-induced periodic surface structure (LIPSS) based on microscopic particle evolution from excitation to damage.*

LIPSS is an important and fundamental type of laser damage that produces sub-wavelength grating like structures that is already used for application, although much about it is not understood. We applied our simulation technique to an analysis of a recent experiment performed by Chowdhury and students and were able to reproduce the key features seen in the lab. We were able to identify surface plasma polariton excitation and follow the evolution of the LIPSS. The results have been submitted for publication to PRL.

(3) *Quantitative benchmark of our new algorithm.*

We have worked with Chowdhury to benchmark our new approach against experiment. Chowdhury performed a series of measurements under tightly controlled and well characterized conditions. Matching experimental conditions exactly and with no tuned parameters, we then modeled the same laser shots and resulting damage. The results are shown in the figure. The match is very good. To our knowledge, such a comparison between experiment and fundamental simulations has never been attempted or achieved before. These results are being prepared for publication. With this result, we have achieved the primary goals described in our proposal and are now working to extend the method and explore new phenomena.

### **Final Report: Gennady Shvets**

Efficient, broadband, damage-resistant pulse compression gratings (PCG) are crucial components of current large scale high power short-pulse laser systems. The performance and resistance to damage by laser pulse radiation of gold coated fused silica PCG was studied and found to be strongly dependent on the gold layer profile, with conformal layer gratings being preferable to non-conformal gratings. To explain the observed experimental results numerical simulations modeling radiation impinging on the grating were conducted for various gold layer profiles. The less conformal profiles showed stronger field enhancement at the surface, thus higher damage susceptibility and worse grating efficiency. With the simulation model it would be possible to evolve gold layer profiles leading to improved grating characteristics provided manufacturing techniques that allow sufficiently fine control over the shape of the layer are developed.

LIPSS form on a wide variety of materials and with a wide variety of characteristics such as ripple period and orientation. This has led to the necessity for different physical mechanisms

being invoked in order to explain their formation and properties. In the experimental study of LIPSS formation in germanium at oblique incidence, LSFL and HSFL were observed for both S and P polarized light. The established mechanism for describing LSFL is the resonant excitation of surface plasmon polaritons on the ionized surface and their interference with the laser field, with phase matching setting the position of the resonance. However, the model used so far at normal incidence did not give correct dependences for groove depth and groove period with increasing number of pulse repetitions when applied to the oblique incidence case. We carried out full wave electromagnetic simulations of the laser field coupling to a SPP due to the periodic structure. In order to find agreement with experimental observations we had to drop the previously taken assumption of infinite metalized medium and instead considered a thin ionized layer of thickness varying with the number of pulses. This model allowed us to adequately describe measurement observations for both light polarizations. The thin ionized layer approach used also merits further exploration when applied to some HSFL formations.

### **Final Report – Sheikh Akbar**

We have been in close collaboration with Dr. Enam Chowdhury's group in the past two years and this effort has benefited our research in both materials science and laser physics. While the collaboration greatly expanded our capabilities for materials processing and characterization, it also led us to think differently from an interdisciplinary point of view. The formation of laser-induced periodic surface structure (LIPSS) is a universal phenomenon that has been observed in a variety of materials. On the other hand, Akbar's group has shown that periodic nanostructures can also be grown via a spontaneous self-assembly process involving surface diffusion and strain energy relaxation (Stress-Induced Periodic Surface Structures, SIPSS). These two processes take place in very different time scales (femtoseconds for LIPSS, hours for SIPSS), however, can result in very similar features. The collaboration has prompted to test a hypothesis that LIPSS is a type of laser-activated SIPSS and their combination can be exploited to create novel structures that have potential of generating properties such as field enhancement, super-wettability, and negative permittivity and permeability. This idea has further led to the generation of multiple research grants. Specifically, this effort has led to the following outcomes.

### **Final Report: Allen Yi**

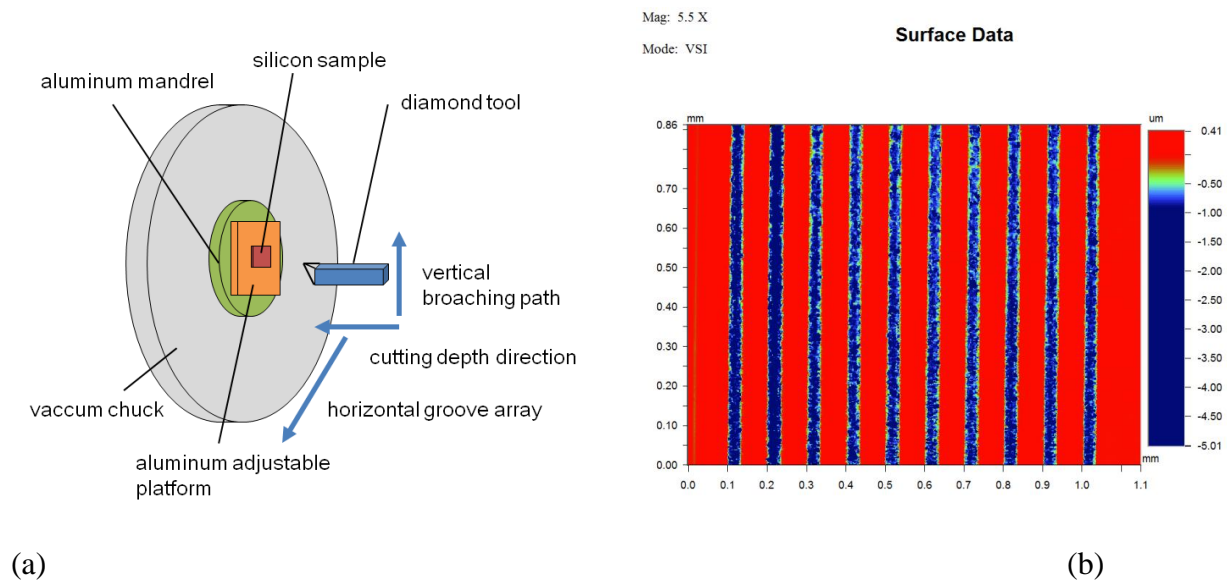
Our research had two components, (1) focused on fabrication of micro/sub-micro features using broaching process as illustrated in figure 1a on the 350 FG (Freeform Generator from Moore Nanotechnology, Inc., Keene, New Hampshire) or cleanroom process; (2) focused on working with Chowdhury group in characterizing surface damage profiles with nm depth precision using

our interferometric optical depth profiler Wyko NT9100. This played a crucial role in determining laser induced damage over many materials and a large wavelength range.

In the ultraprecision machining process, to achieve the uniform depth for the micro grooves, an aluminum adjustable fixture (platform) had to be designed. Using this adjustable fixture, different samples could be machined efficiently as the sample could be leveled easily. This arrangement increases achievable surface roughness because total machine time is reduced and wear of the tool is kept at minimal.

Before the broaching process, four corners of the sample were measured by an ultraprecision gage of nanometer precision. The measurements were used to orient the workpiece such as single crystal copper such that cutting depth would gradually and automatically decrease. The cutting depth of 1  $\mu\text{m}$  spacing grooves was controlled in a linear fashion with a step size which was interpolated using the measurements of the four corners. The exact cutting would also be designed to cope with the random thermal drifting on the spindle that was generated when the spindle was locked.

Samples of various materials such as copper, aluminum and silicon were machined using this process the surfaces were investigated under a white light noncontact interferometer, the Wyko NT9100 as shown in Figure 1b. In addition, these samples were undergone laser ablations and then were characterized using the Wyko interferometer as well. This method has proven to be essential to this program (see publications).



**Figure 1:** (a) Ultra-precision diamond machining of laser target (b) Surface texture of the diamond machined surface

## Publication and Presentation efforts:

### Journal articles:

1. Jian Cheng, Mingjun Chen, Kyle Kafka, Drake Austin, Jinghe Wang, Yong Xiao and Enam Chowdhury, *Determination of ultra-short laser induced damage threshold of KH<sub>2</sub>PO<sub>4</sub> crystal: Numerical calculation and experimental verification*, AIP Advances 6, 035221 (2016).
2. Robert A. Mitchell, Douglass W. Schumacher, and Enam A. Chowdhury, *Modeling crater formation in femtosecond-pulse laser damage from basic principles*, Opt. Lett. 40, 2189 (2015)
3. 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).
4. 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)
5. Robert A. Mitchell, Douglass Schumacher, and Enam Chowdhury, *Modeling femtosecond pulse laser damage using particle-in-cell simulations*, Optical Engineering 53(12), 122507 (December 2014)
6. H. Li, P. He, J. Yu, L. J. Lee & A. Y. Yi, "Localized rapid heating process for precision chalcogenide glass molding," *Optics and Lasers in Engineering* **73**, 62–68 (2015).
7. Patrick Poole, Simeon Trendafilov, Gennady Shvets, Douglas Smith, and Enam Chowdhury, *Femtosecond laser damage threshold of pulse compression gratings for petawatt scale laser systems*, Optics Express, Vol. 21, Issue 22, pp. 26341-26351 (2013)

### Submitted for publication:

1. Drake Austin , Kyle Kafka , Yu Hang Lai , Zhou Wang , Kaikai Zhang , Hui Li , Cosmin Blaga , Allen Y Yi , Louis F. Dimauro , Enam A Chowdhury, "High spatial frequency laser induced periodic surface structure formation in germanium under strong mid-IR fields", under review at App. Phys. Lett.
2. "First model of laser-induced periodic surface structure based on microscopic particle evolution from excitation to damage," R. A. Mitchell, D. W. Schumacher, E. A. Chowdhury, submitted to Phys. Rev. Lett.
3. Z. Niu, H.A. Ansari, E.A. Chowdhury, S.A. Dregia, S.A. Akbar. "Self-assembly of nanoislands and dopant induced step faceting on miscut YSZ-(001) surfaces". Submitted to Small.

### Peer reviewed Conference Proceedings Published:

1. Enam Chowdhury, Kyle R. P. Kafka, Drake R. Austin, Kevin Werner, Noah Talisa, Boqin Ma, Cosmin I. Blaga, Louis F. Dimauro, Hui Li, Allen Yi, Ultra-fast bandgap photonics in mid-IR wavelengths, Proc. SPIE **9835** SPIE Defense+Security, 9835-43 (2016).
2. Enam Chowdhury, Kyle R. P. Kafka, Robert A. Mitchell, Alex M. Russell, Kevin Werner, Noah Talisa, Hui Li, Allen Yi, Douglass W. Schumacher, *Single-shot femtosecond laser ablation of copper: experiment versus simulation*, Proc. SPIE **9632**, Laser-Induced Damage in Optical Materials: 96320R (2015).

3. 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*, Proc. SPIE **9632**, Laser-Induced Damage in Optical Materials: 96321C (2015).
4. 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* Proc. SPIE **9632**, Laser-Induced Damage in Optical Materials: 96320C (2015).
5. Robert A. Mitchell, Douglass Schumacher, Enam Chowdhury, *First principles simulation of laser-induced periodic surface structure using the particle-in-cell method*, Proc. SPIE **9632**, Laser-Induced Damage in Optical Materials: 96320Y (2015).
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7. Kyle Kafka; Drake Austin; Jian Cheng; Simeon Trendafilov; Gennady Shvets; Hui Li; Allen Yi; Cosmin I. Blaga; L. F. DiMauro; Enam Chowdhury, *Laser induced periodic surface structure formation in germanium above laser damage fluence by mid IR femtosecond laser irradiation*, Proc. SPIE **9237**, Laser-Induced Damage in Optical Materials: 92371S (2014).
8. Robert A. Mitchell, Douglass Schumacher, Enam Chowdhury, *Using Particle-In-Cell simulations to model femtosecond pulse laser damage*, Proceedings SPIE **9237**, Laser-Induced Damage in Optical Materials 92370X (2014).
9. Robert A. Mitchell, Douglass Schumacher, Enam Chowdhury, *Modeling femtosecond pulse laser damage on conductors using Particle-In-Cell simulations*, Proceedings SPIE **8885**, 88851U (2013).

Conference Presentations (excluding Conference Proceedings publications above):

1. *Few-cycle Pulse Laser Induced Damage Threshold (LIDT) determination of Critical optical Components for next generation ultra-intense laser systems*, Enam Chowdhury, Noah Talisa, Kyle Kafka, Kevin Werner, Hui Li, Allen Yi, Gabriel Tempea, Catalin Neacsu, International Conference on Extreme Light, Bucharest, Romania 2015.
2. *Self-assembled Nano-structures on Miscut YSZ(001) Surfaces*, Z. Niu, H.A. Ansari, E.A. Chowdhury, S.A. Dregia, S.A. Akbar. Materials Science and Technology (MST), Columbus, OH 2015.
3. *Laser-Induced Periodic Surface Structures from Single femtosecond Pulses*, K. Kafka, D. Austin, J. Chen, H. Li, S. Lambert, L. Hanzlik, S. Akbar, S. Dregia, A. Yi, E. Chowdhury, Gordon Research Conference, Multi-photon Processes, 2014
4. *Adapting Particle-In-Cell simulations to the study of short pulse laser damage*, Robert Mitchell, Douglass Schumacher and Enam Chowdhury, DPP 2014, Bulletin of the American Physical Society **59** (2014).
5. *Laser induced damage threshold (LIDT) Measurement of wide bandgap dielectrics with few-cycle femtosecond laser pulses*, Noah Talisa, Kyle Kafka, Kevin Werner, Drake Austin, Hui Li, Allen Yi, Matthew Foster, Matthew Le, Shivam Tickoo, Enam Chowdhury, 3<sup>rd</sup> High Power Laser Workshop, SLAC National Accelerator Laboratory, CA, 2015

Final Report for AFOSR Award FA9550-12-1-0454 titled, "Understanding the femtosecond laser-solid interaction near and beyond the material damage threshold". Program Officer: Dr. Enrique Parra

6. *Femtosecond laser solid interaction near material damage threshold at mid IR wavelengths*, Enam Chowdhury, Drake Austin, Kyle Kafka, Cosmin Blaga, and Louis F. DiMauro, Quantum Information and Measurement, Berlin Germany, 2014.

PhDs supported by this grant:

1. Dr. Robert Mitchell (2015), "Understanding femtosecond-pulse laser damage through fundamental physics simulations", now working at Voss Scientific.
2. Z. Niu Material Science and Engineering (2016)
3. K. R. P. Kafka, Drake Austin (Chowdhury) and H. Li (Yi) had been funded by the BRI towards their PhDs and they are in their final year.

1.

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**Abstract**

Under the recently completed BRI program, the PI has established a new femtosecond solid dynamics laboratory (FSD) dedicated to the study of the fundamentals of femtosecond laser damage as a function of various parameters, laser wavelength, pulsewidth, pulse number, experimental ambience (air/vacuum/other), polarization and angle of incidence, target solid material conductivity (metal/semiconductor/insulator), band gap, and surface morphology (nano/micro-structured surface), and the PI's team has worked synergistically in designing and performing experiments, analyzing results, and developing a unified femtosecond laser damage simulation framework, which allowed direct benchmarking of simulation results with experimental observation without any free parameters.

The brand new FSD Lab constructed under the BRI grant in the Physics Research Building at the Ohio State University has developed a wide ranging short pulse laser capabilities, from multi-milijoule pulses at kHz to single shot repetition rate, from pulsewidths 5 fs – 150 ps, from 200 nm to 4000 nm (~100 fs). FSD Lab has already established itself as a leading laboratory for studying short pulse laser damage under vacuum/air/other gas environments. The FSD began a collaboration with the High Average Power Laser System (HAPLS) program at LLNL in 2014 to help them select broadband ultra-damage resistant dielectric mirrors for its next generation 10 Hz petawatt laser amplifiers for the ELI-Beamline in the Czech Republic, whose first phase is already completed (please see support letter from LLNL group leaders). The lab was

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also selected as the laser induced damage threshold (LIDT) testing facility for the prestigious SPIE Laser Damage Competition 2015, where the top manufacturers of damage resistant optics in the world participate.

A comprehensive LIDT parameter exploration was performed, which resulted in presentations in five conferences, several conference proceedings, and a summary of results is being prepared to be submitted to Physical Review Letters titled "Near to mid IR femtosecond material damage threshold measurements". Single and multi-shot femtosecond laser damage threshold (fs-LDT) of materials have been studied across a range of band-gaps for s- and p-polarized light and it is found that conventional theoretical prediction on laser damage threshold based on Two Temperature Model (TTM) fails to capture the wavelength dependence of fs-LDT from near to mid IR.

A time-resolved diffraction microscopy technique has been developed in our lab to observe the formation of laser-induced periodic surface structures (LIPSS) from the interaction of a single femtosecond laser pulse (pump) with a nano-scale groove mechanically formed on a single-crystal Cu substrate (Yi Lab, OSU). The interaction dynamics (0–1200 ps) were captured by diffracting a time-delayed, frequency-doubled pulse (probe) from the nascent LIPSS induced by the pump using an infinity-conjugate microscopy setup. Understanding of LIPSS formation with mid IR light pulses have been steadily progressing, and our group is first to demonstrate HSFL (LIPSS period  $< \lambda/2$ ) formation on germanium for 3 – 3.6 micron wavelength femtosecond pulses. Formation of low spatial frequency LIPSS (LSFL, with LIPSS period  $> \lambda/2$ ) with multiple femtosecond mid IR pulses of s and p polarization at oblique incidence angle showed that strong coupling between incident pulses and induced SPPs by multiphoton ionization is possible so much so that large amount of deposited energy is carried away from the most intense central region of the Gaussian focus [Austin et al Opt. Ex. 2015]. Formation of HSFL on Ge was also analyzed (submitted to APL), and it was found that instead of generation of surface plasmon polaritons, these nano-structures are formed by interference of incident pulses with surface scattered light generated by nano-roughness created from previous surface-pulse interactions. Surprisingly, such a phenomenon was not observed for silicon, and this is perhaps attributed to Ge having an order of magnitude larger  $\chi^{(3)}$  than Si, which in Ge, results in the electron density to stay below critical density, allowing such an interaction to happen.

We have developed a new simulation technique for modeling the onset and evolution of laser damage due to the intense interaction of a laser and metal target. The simulation technique is based on the particle-in-cell (PIC) method for simulating plasmas and works by (1) implementing a Lennard-Jones pair-potential (LJPP) model within the PIC framework and (2) using a series of PIC simulations to follow target evolution over time-scales spanning six orders-of-magnitude, from the femtosecond scale of the laser interaction to the nanosecond scale of target evolution. To our knowledge, neither of these approaches has been demonstrated before. In the final phase of the BRI, we have bridged the gap between experimental and simulation results by performing benchmarking experiments, and were able to generate 2 micron diameter focal spot with few cycle pulses (5 – 35 fs). Such extremely spatio-temporally confined pulses interacted with single crystal Cu and generated craters for various fluence ranges, which provide straight forward templates for our PIC simulation to predict. We have also performed benchmarking simulations, which are producing crater depths, matching experimentally observed crater depths very well. To our knowledge, this is the first successful demonstration of benchmarking between experimental and theory/modeling efforts in the field of short pulse laser damage.

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**Archival Publications (published) during reporting period:**

Journal Articles

1. Jian Cheng, Mingjun Chen, Kyle Kafka, Drake Austin, Jinghe Wang, Yong Xiao and Enam Chowdhury, Determination of ultra-short laser induced damage threshold of KH<sub>2</sub>PO<sub>4</sub> crystal: Numerical calculation and experimental verification, *AIP Advances* 6, 035221 (2016).
2. Robert A. Mitchell, Douglass W. Schumacher, and Enam A. Chowdhury, Modeling crater formation in femtosecond-pulse laser damage from basic principles, *Opt. Lett.* 40, 2189 (2015)
3. 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).
4. 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)
5. Robert A. Mitchell, Douglass Schumacher, and Enam Chowdhury, Modeling femtosecond pulse laser damage using particle-in-cell simulations, *Optical Engineering* 53(12), 122507 (December 2014)
6. H. Li, P. He, J. Yu, L. J. Lee & A. Y. Yi, "Localized rapid heating process for precision chalcogenide glass molding," *Optics and Lasers in Engineering* 73, 62–68 (2015).
7. Patrick Poole, Simeon Trendafilov, Gennady Shvets, Douglas Smith, and Enam Chowdhury, Femtosecond laser damage threshold of pulse compression gratings for petawatt scale laser systems, *Optics Express*, Vol. 21, Issue 22, pp. 26341-26351 (2013)

Peer Reviewed Conference Proceedings:

1. Enam Chowdhury, Kyle R. P. Kafka, Drake R. Austin, Kevin Werner, Noah Talisa, Boqin Ma, Cosmin I. Blaga, Louis F. Dimauro, Hui Li, Allen Yi, Ultra-fast bandgap photonics in mid-IR wavelengths, *Proc. SPIE 9835 SPIE Defense+Security*, 9835-43 (2016).
2. Enam Chowdhury, Kyle R. P. Kafka, Robert A. Mitchell, Alex M. Russell, Kevin Werner, Noah Talisa, Hui Li, Allen Yi, Douglass W. Schumacher, Single-shot femtosecond laser ablation of copper: experiment versus simulation, *Proc. SPIE 9632, Laser-Induced Damage in Optical Materials: 96320R* (2015).
3. 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, *Proc. SPIE 9632, Laser-Induced Damage in Optical Materials: 96321C* (2015).
4. 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 *Proc. SPIE 9632, Laser-Induced Damage in Optical Materials: 96320C* (2015).
5. Robert A. Mitchell, Douglass Schumacher, Enam Chowdhury, First principles simulation of laser-induced periodic surface structure using the particle-in-cell method, *Proc. SPIE 9632, Laser-Induced Damage in Optical Materials: 96320Y* (2015).
6. Drake Austin; Kyle Kafka; Cosmin I. Blaga; Louis F. Dimauro; Enam Chowdhury, Measurement of femtosecond laser damage thresholds at mid IR wavelengths, *Proc. SPIE 9237, Laser-Induced Damage in Optical Materials: 92370V* (2014).
7. Kyle Kafka; Drake Austin; Jian Cheng; Simeon Trendafilov; Gennady Shvets; Hui Li; Allen Yi; Cosmin I. Blaga; L. F. DiMauro; Enam Chowdhury, Laser induced periodic surface structure formation in germanium above laser damage fluence by mid IR femtosecond laser irradiation, *Proc. SPIE 9237, Laser-Induced Damage in Optical Materials: 92371S* (2014)

8. Robert A. Mitchell, Douglass Schumacher, Enam Chowdhury, Using Particle-In-Cell simulations to model femtosecond pulse laser damage, Proceedings SPIE 9237, Laser-Induced Damage in Optical Materials 92370X (2014).

9. Robert A. Mitchell, Douglass Schumacher, Enam Chowdhury, Modeling femtosecond pulse laser damage on conductors using Particle-In-Cell simulations, Proceedings SPIE 8885, 88851U (2013).

**Changes in research objectives (if any):**

NA

**Change in AFOSR Program Manager, if any:**

NA

**Extensions granted or milestones slipped, if any:**

NCE 01/31/2016.

**AFOSR LRIR Number**

**LRIR Title**

**Reporting Period**

**Laboratory Task Manager**

**Program Officer**

**Research Objectives**

**Technical Summary**

**Funding Summary by Cost Category (by FY, \$K)**

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

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**Appendix Documents**

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