



**Fundamentals of Femtosecond Laser Induced Damage of Solids: Advancement Through
Experimental, Computational and
Theoretical Development**

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14. ABSTRACT <p>Abstract</p> <p>Femtosecond laser induced damage (fsLID) is an exciting non-perturbative non-linear phenomenon with a wide range of applications. The present effort concentrates on understanding this phenomenon from experimental, theoretical and computational perspectives, with each effort benchmarking and providing feedback for the improvement of the other method. Among its chief achievements, understanding of the wavelength scaling of fsLID in several semiconductors have been well understood, with appropriate theoretical model development, notably the Keldysh-Vinogradov model of field induced free carrier generation and energy absorption. Large scale hybrid PIC computational method has been developed. In the current program, we have significantly extended our understanding of few cycle pulse (2 - 4 cycles, FCP) laser damage and ablation of solids and how it differs from that induced by longer (many cycle, > 10 cycles) pulses. FCPs typically exhibit bandwidth around 25% of the carrier frequency, while 100 fs pulses require ~1.5% bandwidth. It implies that first, the FCPs promote valence electrons to the conduction band at higher rate (due to lower order of multiphoton processes for the highfrequency part of the broad FCP spectrum); second, the FCPs produce free elec- trons with higher kinetic energy compared to those produced by 100-fs pulses. The higher kinetic energy supports deeper electron penetration into samples and reduced electron-particle collision time. The reduced collision time drives the laser-generated free-carrier plasma faster to quasiequilibrium and increases the rate of energy transfer to phonons. The increased rate of electron- phonon energy transfer supports reduced temperature of the laser-generated free-carrier plasma. Therefore, the FCPs are favorable for observing the transient processes at relatively lower fluence. However, also the transient effec</p>		
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Fundamentals of femtosecond laser induced damage of solids: advancement through experimental, computational and theoretical development: *Final Performance Report.*

Introduction:

Under the current AFOSR program, the PI's femtosecond solid dynamics laboratory (FSD lab) has established itself as a world leader in femtosecond laser damage physics. **Over the last six years, the program has graduated 7 PhDs, with many of them working in DoD and DoE labs or supported programs.** Over the years, FSD has developed strong collaborations AFRL RX (Materials Directorate) and RD (Directed Energy Directorate) on mid-IR laser technology and damage, Army Research Laboratory (RDE Com under Materials) and Adelphi Laboratory Center (ALC-ARL) [1–3] on strong field mid-IR solid interaction, including a synergistic collaborator role on laser solid non-linear interaction in an ongoing AFOSR MURI titled "Fundamental strong-field interactions with ultrafast, mid-infrared lasers" (to be concluded in 2020). The PI has collaborated with Laboratory for Laser Energetics (LLE) Rochester on developing metal dielectric hybrid ultrabroadband, low group velocity dispersion (GVD) optics with high damage threshold, which will be used in their MTW-OPAL 15 fs multi-joule high contrast OPA laser, to be used as a blue print for the EP-OPAL, the only 75 PW laser under consideration in the US at present. Lawrence Livermore National Laboratory's (LLNL) High Average Power Laser System (HAPLS) program also reached out to the PI to help them select broadband ultra-damage resistant dielectric mirrors for its next generation 10 Hz petawatt laser (L3 Beamline) amplifiers for the ELI-Beamline in the Czech Republic [4]. The lab was also selected as the laser induced damage threshold (LIDT) testing facility for the prestigious SPIE Laser Damage Competition two years in a row (2015-16) [5,6], where the top manufacturers of ultra-broadband damage resistant optics in the world participate. Currently, the PI is under contract with LLNL to test concept dielectric mirrors and gratings @ 2 micron wavelength for their futuristic high average power high wall-plug efficiency BAT (Big Aperture Thulium) laser framework [7,8], which would be capable of reaching petawatt levels of peak power at 1-100 kHz repetition rate. Relevant work by the team is described below briefly:

LIDA dynamics with few to single cycle pulses: In the current program, we have significantly extended our understanding of few cycle pulse (2 - 4 cycles, FCP) laser damage and ablation of solids and how it differs from that induced by longer (many cycle, > 10 cycles) pulses. FCPs typically exhibit bandwidth around $\geq 25\%$ of the carrier frequency, while ≥ 100 fs pulses require $\sim 1.5\%$ bandwidth. It implies that first, the FCPs promote valence electrons to the conduction band at higher rate (due to lower order of multiphoton processes for the high-frequency part of the broad FCP spectrum); second, the FCPs produce free electrons with higher kinetic energy compared to those produced by 100-fs pulses. The higher kinetic energy supports deeper electron penetration into samples and reduced electron-particle collision time. The reduced

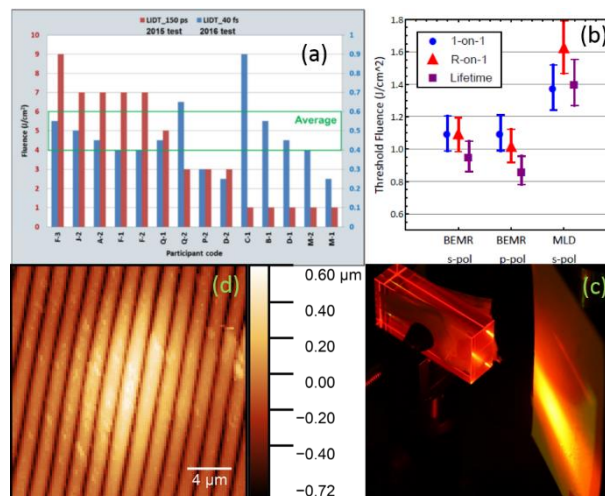


Figure 1 Notable examples of synergistic collaboration of Chowdhury Lab (a) Laser damage competition across the world 2015 & 2016, ultrabroad-band high reflector mirror testing (b) MTW-OPAL broadband enhanced metal reflector vs traditional multi-layer dielectric LIDT (c) Over 36% conversion into harmonics from MIR 3.8 micron 200 fs laser interaction with 4 cm poly-ZnSe (ARL-ALC) (d) Laser induced damage (blister formation) of MLD gratings with 2 micron, 100 fs pulses (LLNL).

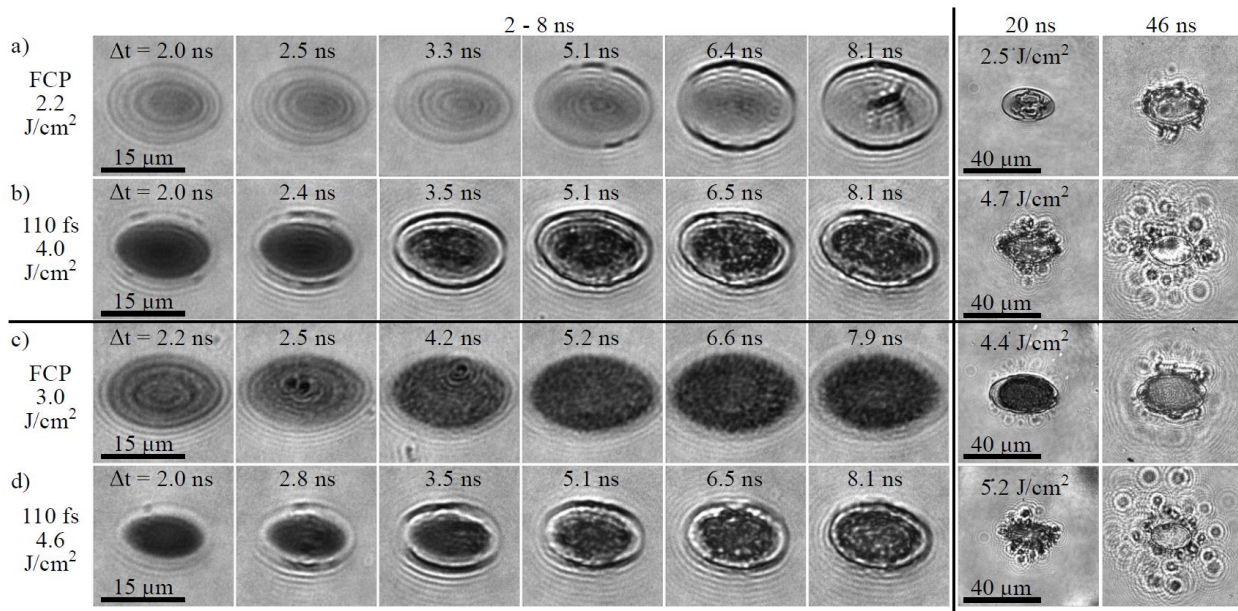


Figure 2 Time-resolved Surface Microscopy (TRSM) images of ablation dynamics from 2-8 ns (first six columns) and at 20 and 46 ns (last two columns). a) and b) correspond to FCP and 110 fs pulse ablation, respectively, on the quad-layer sample; c) and d) correspond to FCP and 110 fs pulse ablation on the double-layer sample. Note the change in scale for the 20 and 46 ns images. Taken from [15]

collision time drives the laser-generated free-carrier plasma faster to quasi-equilibrium and increases the rate of energy transfer to phonons. The increased rate of electron-phonon energy transfer supports reduced temperature of the laser-generated free-carrier plasma. Therefore, the FCPs are favorable for observing the transient processes at relatively lower fluence. However, also the transient effects are observed over larger range of fluences due to the increased rate of energy transfer to the lattice via phonons. This is how in the case of femtosecond laser damage, whose dynamical evolution spans around 10 orders of magnitude [9] in time may retain the 'memory' of the femtosecond impact long after the pulse has left.

Some key results involve measuring few cycle LIDT of few cycle optics for the first time [10], air and vacuum differences, anomalous polarization sensitive 'volcanic crater-like' blister formation on ultra-broad band chirped mirrors [11], stark differences in thin film ablation morphology induced by FCP compared with longer pulses, due to dynamic field modification inside material due to rapid ionization during FCP-solid interaction [12,13], and finally, capturing dynamics of damage/blister formation and ablation over a $0 - 4.5 \times 10^7$ fs, over seven orders of magnitude of time, using time resolved surface microscopy (TRSM) technique. With varying fluence, in single to multi-layer high bandgap dielectrics, we are able to capture electron-hole generation, recombination, rapid expansion of 'super-heated' material ('blistering'), oscillation of blister height, formation of fractures on the surface, and finally ablation. These results have already been disseminated via several CLEO and other conference presentations and multiple manuscripts are currently being prepared to publish these results. Part of this research was presented at SPIE Laser Damage 2019, and has won the best student research presentation award and it was

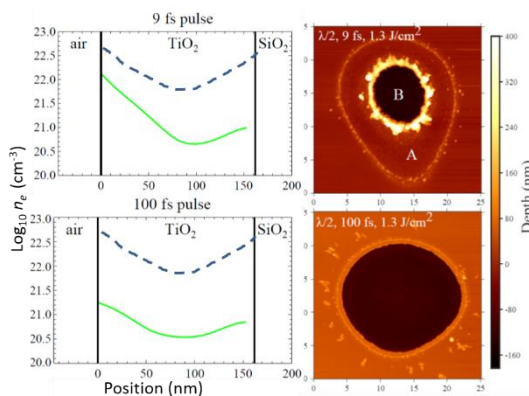


Figure 3 Rapid rise in intensity of FCP, compared with 100 fs pulses, causing dynamic, "modified" field distributions within optical thin films, affecting ablation crater morphology. Blue dashes show unscaled static field distribution in layer. (bottom row) 100 fs electron density follows static field distribution (top row) FCP modifies electron density and causes two step ablation crater. Ref [13]

published in SPIE Proceedings [14]. Other results (Fig. 2) later was submitted to Optics Letters and was recently accepted for publication. [15] Achieving these significant results establishing FCP damage and ablation as a different regime of femtosecond laser damage, were possible due to development of our expertise in generating, manipulating, controlling and fully characterizing FCP. *In fact, we are now able to ascertain pulse duration (FCP vs longer pulses) just by looking at thin film damage crater morphology. (see Fig. 3)*

Strong field Mid-IR regime damage and ablation: We have performed a methodical study of near to mid-IR single shot ablation threshold of three separate semiconductors (ZnSe, Si and Ge), and compared

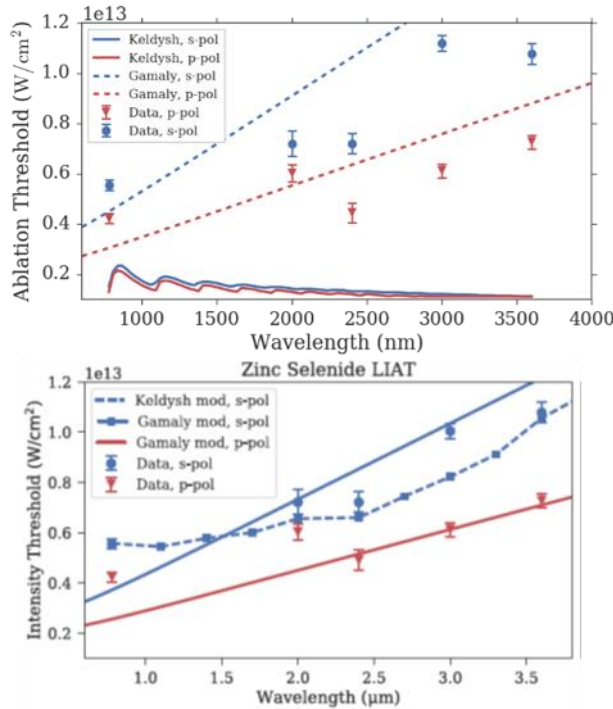


Figure 4 ZnSe ablation threshold data for ~100 fs pulses (top) vs conventional theoretical models (bottom) Using Keldysh-Vinogradov model with multi-band.

their threshold intensity/fluence behavior as a function of wavelength, to compare with existing theoretical approaches, and have been developing new theoretical approaches. Both pairs ZnSe and Si and Ge and Si provide interesting contrast and comparison from their physical properties, ZnSe and Si has large direct energy gaps (2.7 and 3.4 eV respectively), and Si and Ge are both shallow gap semiconductors (indirect band gaps of 1.1 and 0.67 eV, respectively), with Ge direct bandgap also being comparable, 0.8 eV. Both Si and Ge surfaces rapidly oxidizes to produce a 2-3 nm native oxide layer on top of single crystal samples, whereas ZnSe surface show no such layer with different chemical composition. At room temperature, single crystal ZnSe also differs from ultrapure high resistivity single crystal samples of Si and Ge in carrier concentration, where the former shows carrier densities below 10^5 cm^{-3} , the latter two exhibit concentrations on the order of 10^{14} cm^{-3} . We

observe tremendous differences between damage threshold vs wavelength behavior of ZnSe on one side and Si and Ge on the other. Si and Ge also differ starkly in their ablation behavior. *When threshold vs wavelength data of any of these three (ZnSe, Si, Ge) is compared to existing theoretical models (Keldysh and Gamaly two temperature models) as used widely in the field, they fail, in some instances by orders of magnitude (See Fig. 4)!* For the *s*-polarization case, in collaboration with our theory co-PI Gruzdev, we have developed a comprehensive model for laser ablation of ZnSe, integrating free carrier (FC) contribution from multiple valence/conduction band transitions, dynamically modifying dielectric function at the surface due to the change in FC density, and capturing strong optical field induced free carrier absorption (FCA), including the contribution from FC decoherence due to electron-phonon collisions (Keldysh-Vinogradov model [16]), details of which is described in more details in the theory section below. The

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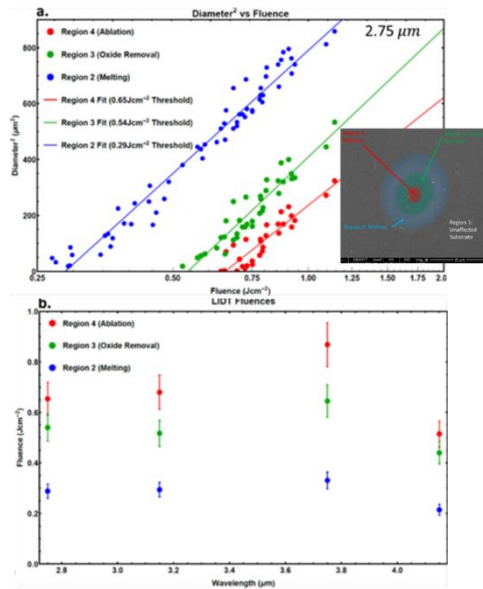


Figure 5 Multi-stage damage of silicon under strong mid-IR fields (200 fs pulses) with single *p*-polarized pulses. (top) LIDT determination of each region (bottom) LIDT vs λ for each region

other crucial modification needed was to discard the conventional notion of using FC critical density as the determining fluence/intensity threshold for damage/ablation, which decreases with wavelength as λ^{-2} [17], and underestimates damage threshold by significant amount (in some cases, by an order of magnitude or more). Combining all contributions, we were able to fit the ZnSe data extremely well (Austin PhD thesis, see Fig. 7). This model should be applicable to any wide band direct gap material laser induced ablation. We are preparing a manuscript to present this data and model in PRL and an extended version to PRB.

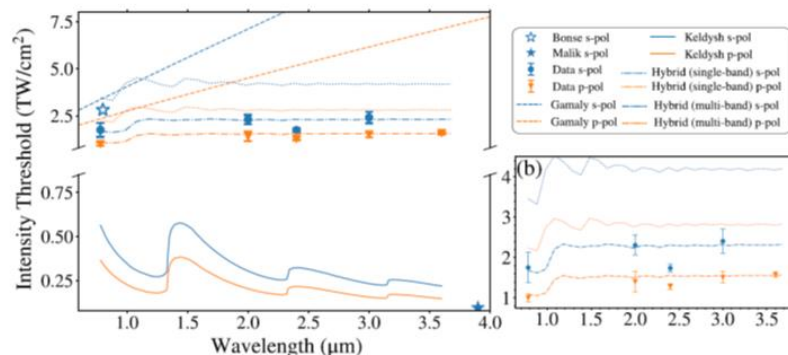


Figure 6 (a) Experimental single-shot damage/ablation intensity threshold versus wavelength in Ge for both p- and s-polarization light. For comparison, the single-shot melting threshold at 0.8 μm , with 130 fs pulses [17] and multi-shot damage threshold at 3.9 μm with 255 fs pulses [19] are shown as well. The former was performed on a heavily p-doped sample as opposed to the high purity, undoped sample used in this work. This is likely the cause of the discrepancy between the two measurements. Also plotted are the predicted thresholds from Gamaly's model and the standard application of the Keldysh model to laser damage, i.e., the onset of damage upon reaching plasma critical density. A modification to the latter model was made by including the contributions from all valence bands in Ge and by using the onset of ultrafast melting as the criterion for ablation. For comparison, single band results are also shown. (b) Zoom-in of the data points in (a).

versions. None of the models can explain the abrupt drop in threshold behavior for all three regions beyond 3.7 micron wavelength. Most likely this is due to enhancement of collisional ionization from ponderomotive heating of free carriers. However, such effects in silicon is extremely difficult to model because silicon is an indirect bandgap material and in it, there exists many pathways to phonon assisted ionization, complicating the approach. The silicon results were recently published in Scientific Reports [18]. We are also in collaboration with the premier TDDFT group in Japan, but, 200 fs pulse modeling is beyond their capabilities currently. Efforts will be undertaken to shorten the pulse-width for our source during the last two years of the project. Similar experiments with germanium single crystals were carried out as well, whose analysis also reveals breakdown of theoretical models [19] (Fig. 6).

Using our knowledge of strong field MIR laser interaction with Si, we were (in collaboration with Shvets group) able to the first team in the world to demonstrate photon acceleration in silicon metasurfaces-on-sapphire and observed time-resolved ultra-short pulse induced resonance shift (> 100 nm shift) due to strong field generated electron hole plasma dynamically modifying the dielectric function of the meta-surface. This technique will open door to future peta-Hertz optoelectronics switching and control. This work has been published in Nature Communications. [20]

Simulation

In the current project, we focused on developing this approach so that it could be used quantitatively without

It turns out that due to the presence of native oxides, Si and Ge femtosecond laser ablation and damage is more complicated. For example, in the case of Si ablation, there exists three stages of damage: first, the most intense central portion exhibiting material ablation, second, the annular area surrounding the ablated region shows 'native oxide removal' height change (~ 2 nm), and the outermost damage region, and the final outer damaged region represents ~ 50 nm thick melted/amorphous silicon (corroborated by Raman and high resolution transmission electron microscopy). The threshold behavior of all three regions were determined across the mid-IR wavelengths and compared to conventional Keldysh and Gamaly models and their modified

tuning of physical parameters over a range of conditions. This would both allow it to be benchmarked against experiment and to be useful as an analytic tool. We describe some of the primary innovations developed. (This work was primarily done by a single graduate student, Dr. Alex Russell [21], who graduated in 2018 and was hired by Voss Scientific, LLC, the company that developed LSP. Two new students have since been trained on PIC modeling in the intervening time, one of whom will perform the proposed work if funded. It is expected this work will be the basis of that student's dissertation.)

- *Collisions.* As an intense laser excites a target strong spatial gradients in temperature arise (10^5 degrees / μm or more) that vary rapidly on a femtosecond scale. The skin depth and reflection coefficient are likewise varying in space and time in a fashion that significantly impacts the absorbed energy distribution. In a metal where the initial absorption is small, small changes in the reflectivity have a dramatic effect. In a dielectric where ionization initiates the process, there is dramatic temporal variation in the direction normal to the surface and in the attainment of critical density. *We developed a new PIC collision algorithm that is fully kinetic and can treat non-equilibrium situations. Further, it can be readily modified to implement different or multiple collision models. None of our benchmarking would have been successful without this [22].*
- *We added multiphoton, Keldysh, and avalanche ionization models to the existing tunneling ionization model in LSP, since these are required for treating dielectrics.* This is somewhat harder than it looks since intensity based models are not natural to implement in an electric field based code with locally generated fields that are varying in space and time and a time and space varying index of refraction. However, we found that the intensity could be derived from the local magnetic field with reasonable accuracy.
- Our initial effort divided a simulation into three phases (femtosecond, picosecond, nanosecond), each running an optimized sub-simulation with the output of one becoming the input to the next. For technical reasons, target evolution under the pair potentials was not initiated until ~ 60 ps after the laser excitation began. This was valid for the first simulation covering the femtosecond laser excitation (< 100 fs) where little ion motion can occur, but this was problematic for the picosecond simulation because both thermalization and some ion motion occurs. Thermalization between electrons and ions requires up to 60 ps because the thermal gradients lead to the rapid spread of energy to unilluminated regions of the target that will eventually ablate. *We eliminated this artificial distinction with the pair potential algorithm now switched on immediately after the laser and we also implemented a TTM model within the PIC framework. PIC, MD and TTM can now be used self-consistently as needed within a single simulation. The TTM rate equations implemented are a restricted version that complements the PIC algorithm [21].*
- Our initial effort simply used Lennard-Jones potentials. *We implemented an embedded atom model along with the capability to use density functional theory (DFT) where available. Although less important for simpler cases of laser damage, this is vital for modeling the damage plume where very large variations in density are encountered.* Using embedded atom and DFT, we were able to model nanoparticle formation in the damage plume.
- *We showed that the damage crater could be modeled in 2D3V by comparing 2D3V and 3D simulations.* Essentially, in 2D3V, the laser mode is modeled as a line focus whose cross-section is the same as that of a true 3D laser. The cross-sectional damage profiles were nearly identical in copper. In part, this is because the primary thermal gradient is in the longitudinal direction (normal to the surface) and this dominates the problem. In dielectrics this is less true and 2D3V is wholly inadequate for modeling the plume [21].

Selected simulation results.

These, and other features, are implemented as needed for a given problem. The femtosecond simulation usually requires half a day using several hundred cores on The Ohio Supercomputer where all of this work was done and where we have an annual allocation sufficient for the needs of the proposed project. However, DOD computer time will be applied for to support extended 3D modeling. A full 2D simulation including

crater formation can be done in several days. A 3D simulation requires 1-2 weeks.

To test our approach, we performed precision laser damage experiments and ab initio modeling with no adjustable parameters. Performing our own experimental measurements was crucial since target preparation can have a dramatic effect on the resulting morphology and we were able to carefully characterize the laser in space and time. The simulations began with the experimentally measured laser pulse and a single crystal copper target. Material parameters such as the pair potentials and temperature dependent collision rates (electron-ion, electron-electron; the pair-potentials establish ion-ion interactions) used established values from the literature so there was no tuning of physical parameters to match the experimental results. The measured damage crater cross-section and simulation results are shown in Fig. 7 as obtained with no scaling applied. As can be seen, the agreement is good. To our knowledge, although the field of laser damage is decades old, this is the first time crater morphology was correctly predicted from such a first-principles simulation [23].

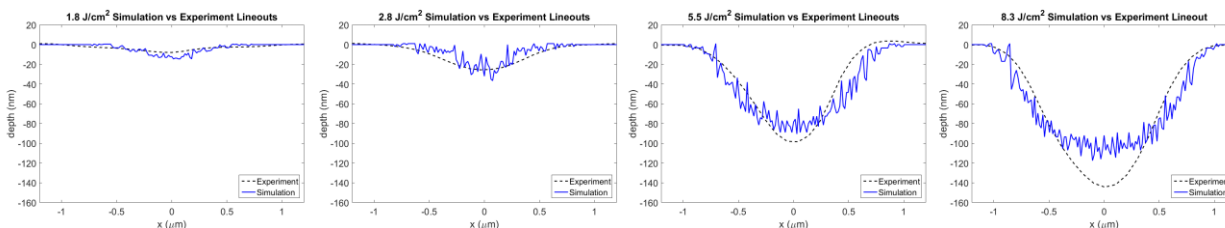


Figure 7 Results of benchmark tests: Experiment (black dotted line) and PIC prediction (blue solid line) at 4 fluences as indicated above each plot. The experimental profiles are smooth due to the limited spatial resolution of the profilometer used.

It was crucial that the collisionality of the copper be modeled including electron-electron, electron-ion and electron-lattice collisions during the laser interaction up to electron temperatures of ~ 10 eV. There was significant departure from thermal equilibrium in the electronic system during the laser pulse and, of course, the electrons and ions were far from equilibrium as well. The collision rates varied significantly between

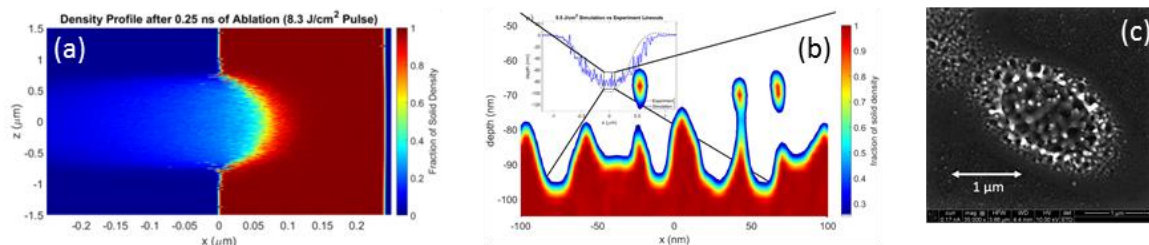


Figure 8 Evolution of the heated region. Simulation results: (a) Damage plume before damage is complete, (b) Nanodroplet formation leading to surface texture. For comparison, (c) Experimental micrograph of a damage spot.

0 and 10 eV; electron-ion dominated at first and then electron-electron dominated later. Figure 8(a) shows the damage plume. The rough surface predicted by the simulations, Fig. 8(b), was in fact observed. The experimental profiles shown are smooth due to the spatial resolution of the profilometer used, but later SEM measurements showed high spatial frequency variation at the scale predicted by the simulations on a different, but similar target to that used for the benchmark measurements, Fig. 8(c).

We have also explored damage for the cases of near-IR/aluminum and mid-IR/ZnSe including modeling of multiple valence bands. Fig. 14 shows results for aluminum in a series of simulations applying an analysis common in the literature [24]. The depth and width of the final crater is plotted for varying incident pulse fluence using 780 nm, 80 fs pulses focused to a 1 μm spot. We compare to experimental results [25][26] that extrapolated such data (red and blue lines) to measure the damage threshold. The simulation derived thresholds are 0.10 J/cm^2 (width based) and 0.12 J/cm^2 (depth based) while the experimental results range from $0.12 - 0.14 \text{ J/cm}^2$, about a 20% difference. This is a good result given there was no tuning in our technique and we have no detailed information on pulse contrast or spatial mode as we did when comparing to our own experiments. We also performed a mid-IR study of copper damage. Mid-IR pulses tend to be longer, so 120 fs long pulses were used and the target thickness was $8 \mu\text{m}$ so that breakthrough was not possible. Pulse fluences of 2 J/cm^2 and 5 J/cm^2 and wavelengths of $1 \mu\text{m}$, $3 \mu\text{m}$, and $5 \mu\text{m}$ were modeled. We found that absorption fell with increasing wavelength (2 J/cm^2 : 0.07 to 0.02 going from $1 \mu\text{m}$ to $5 \mu\text{m}$; 5 J/cm^2 : 0.17 to 0.04 , same wavelength range). The absorption was not linear in fluence. Analysis of the simulation explains these results due to the rapidly varying laser-target/plasma interaction which results in a dynamically changing complex index of refraction (hence skin depth and reflectivity). A new result is increased kinetic effects. In particular, at longer wavelengths we see increased ballistic (non-thermal) motion both into and transverse to the target, even though the heated region is somewhat smaller. The transverse spreading appears irregular or chaotic, but can be captured because our model is intrinsically kinetic and does not require a thermal distribution. More analysis is required for these initial results, however.

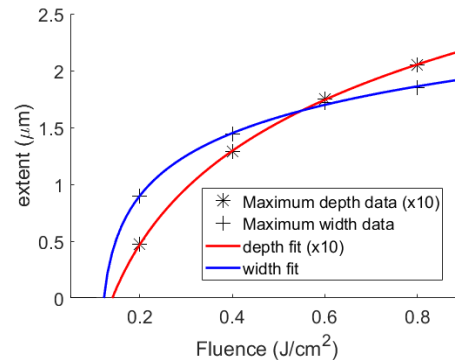


Figure 9 Simulation predicted crater depths and widths as a function of incident pulse fluence.

Theory: Special feature of the mid-infrared laser pulses is a relatively small magnitude of photon energy. It supports generation of free carriers with low kinetic energy and large collision time. Therefore, the free-carrier plasmas generated by intensive mid-infrared femtosecond laser pulses are characterized by low electron-particle collision rates. Optical response and absorption of such low-collision-rate electron-hole plasmas cannot receive a proper theoretical treatment by the usual models of free-carrier generation based on a rate equation for conduction-band electron density. Those models improperly combine the Keldysh photoionization model with the Drude model of free-carrier optical response and use high collision rates obtained from fitting some experimental data. As a result, the traditional rate-equation models failed to qualitatively reproduce wavelength scaling of damage and ablation thresholds by ultrashort mid-infrared laser pulses. The theoretical part of this project was focused on developing a simple, computationally effective and inexpensive, but physically correct model of ultrafast generation and transient optical response of free carriers under the approximation of low rate of electron-particle collisions. The Keldysh photoionization model and the Vinogradov equation for the rate of free-carrier absorption were employed as the basic elements of the newly developed model. Preliminary simulations with the newly developed model demonstrated significant departure from the traditional rate-equation models and reproduced the trend of wavelength scaling of

damage and ablation thresholds. The major theoretical work of this project was focused on two tasks:

A) checking, improvement of the mathematical formulation, and numerical implementation of the **multi-band Keldysh-Vinogradov** model of electron-hole plasma generation and ultrafast heating proposed during the first year of this project [27,28];

B) theoretical analysis of the pathways of laser-driven electron excitations in silicon.

Task A. The Keldysh-Vinogradov model includes inter-band electron transitions from several valence bands to a conduction band characterized by the transition rates evaluated from the Keldysh formula [29]. The intra-band dynamics of the conduction-band electrons is described by the Vinogradov equation [30]. In contrary to a majority of the traditional models of the plasma generation in crystals, the Vinogradov equation assumes low electron-particle collision rate and takes into account the laser-driven modifications of the conduction band. Those approximations are favorable for generation of highly non-equilibrium energy distribution of the conduction electrons. Proper simulation of the distribution was one of the major objectives of the theoretical efforts this year. The challenges met during the simulations were attributed to the fact that the model consists of several nonlinear differential equations nonlinearly coupled to each other. The simulations of the energy distribution of the conduction-band electrons were successfully done with improved simulation codes and reduced steps of energy variations in energy-momentum space. Obtained energy distributions are qualitatively similar to those obtained by more elaborated (and time consuming) methods, e. g., numerical solving the Boltzmann equation.

Another significant challenge was to integrate the Keldysh-Vinogradov model into a larger-scale simulation model that incorporated laser-induced variations of the optical response and reflectivity. For that model, the total absorption rate was obtained as a sum of the intra-band contribution evaluated by the Vinogradov equation and the inter-band contribution evaluated from Keldysh formula [16]. That absorption was treated as an imaginary part of a complex refractive index and was introduced into the Kramers-Kronig relations to obtain the real part of the index. At this step, the simulations produced questionable results that did not match some test cases. The reasons of the mismatch are under investigation now, and additional testing is under run to indentify particular point of the mismatch. Approaches to fix this issue are under consideration including reformulation of the Keldysh-Vinogradov model in terms of integral equations with smooth and continuous kernels.

Task B. Experimental data on laser-induced ablation of silicon with mid-IR ultrashort laser pulses demonstrated wavelength scaling that was not reproduced by any standard model of electron excitation, e. g., the Keldysh formula and the Gamaly model of strong instant ionization. However, the original formulation of the Keldysh-Vinogradov model cannot be applied to simulate those data because of the specific band structure of silicon that does not meet the assumptions of the Keldysh-Vinogradov model. In this connection, a separate analysis was done with a support from simple estimations of inter-band transition rates. The analysis suggests that the ultrafast effects

observed at mid-IR wavelengths correspond to the transition of energy absorption and deposition mechanisms from photon-dominated to field-dominated (also referred to as quasi-static). This conclusion is supported by the fact that the amount of the energy associated with laser-driven electron oscillations (i. e., the ponderomotive energy) exceeds photon energy at mid-IR wavelengths. Therefore, exchange by the ponderomotive energy during electron-particle collisions transfers more energy than absorption of a single photon. This transition has not been discussed in any previous publication and is a novel concept in the field of ultrafast laser-solid interactions. This discovery, along with experimental work, has been published recently in Scientific Reports [18].

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Program Officer: Dr. Andrew Stickrath

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Awards, Publication and Presentation efforts:

Award:

1. [K.R.P. Kafka, N. Talisa, G. Tempea, D.R. Austin, C. Neacsu, and E.A. Chowdhury, *Few-cycle pulse laser-induced damage of thin films in air and vacuum ambience* oral presentation by Kyle Kafka was awarded the Best Oral Presentation Award for the SPIE Laser Damage 2016 conference at Boulder, CO. This is the most prestigious and world renowned conference in the field of laser damage.](#)
2. N. Talisa, M. Tripepi, B. Harris, J. Krebs, N. Talisa, M. Tripepi, B. Harris, A. Alshafey, J. Krebs, A. Davenport, E. Randel, C. S. Menoni, E. A. Chowdhury, E. Randel, C. S. Menoni, and E. A. Chowdhury, "Single-shot femtosecond laser- induced damage and ablation of HfO₂/SiO₂- based optical thin films: a comparison between few-cycle pulses and 110 fs pulses," Noah Talisa was awarded the Best Student Research presentation award in SPIE Laser Damage, 2019 in Boulder, CO.

Journal articles:

1. N. Talisa, A. Alshafey, M. Tripepi, J. Krebs, A. Davenport, E. Randel, C. S. Menoni, and E. A. Chowdhury, "Comparison of damage and ablation dynamics of thin film interference coatings initiated by few cycle pulses vs longer femtosecond pulses," *Opt. Lett.* **accepted**, 1–5 (2020).
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4. [Kevin Werner, Michael G. Hastings, Aaron Schweinsberg, Brian L. Wilmer, Drake Austin, Christopher M. Wolfe, Miroslav Kolesik, Trenton R. Ensley, Laura Vanderhoef, Anthony Valenzuela, and Enam Chowdhury, "Ultrafast mid-infrared high harmonic and supercontinuum generation with n₂ characterization in zinc selenide", *Optics Express* **27**, 2867 \(2019\). <https://doi.org/10.1364/OE.27.002867>](#)
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12. [Kyle R. P. Kafka, Noah Talisa, Gabriel Tempea, Drake R. Austin, Catalin Neacsu and Enam A. Chowdhury, *Few-cycle pulse laser induced damage threshold determination of broadband optics*, *Optics Express* 24, 28858 \(2016\).](#)

Conference Proceedings/Presentations:

1. Invited Talk, TU Darmstadt, Germany, December 2019
2. Invited Talk, LOA, Ecole Polytechnique, Paris, France December 2019,
3. E. A. Chowdhury, [Invited talk, "Surface structuring with femtosecond mid-infrared laser pulses", RAPID \(Research and applications of Photonics in Defense\), Miramar Beach, FL, 22-24 August, 2018](#)
4. E. A. Chowdhury, [Invited talk, "High damage threshold optics in ultra-intense laser systems: State of the art and Next Gen Optics" ICUIL \(International Conference on Ultra-Intense Lasers\), Lindau, Germany, 9-14 September, 2018](#)
5. [Plenary talk HiLase Workshop, October 14-17, 2018 Dolny Brezany, Czech Republic](#)
6. [Seminar at Center for Energy Research, University of California San Diego, May 18, 2018](#)
7. Invited talk HPLSE, April 9-13, China 2018
8. *Invited talk, SPIE Defense and Security, Ultra-fast Bandgap Photonics, 2018, Orlando FL.*
9. *Invited talk, SPIE Defense and Security, Ultra-fast Bandgap Photonics, 2017, CA.*
10. *Invited Talk, SPIE Pacific Rim Laser Damage 2017, Shanghai, China.*
11. Invited Seminar, Sandia National Laboratory, Sept 2017.
12. E. A. Chowdhury, "Femtosecond laser damage: Past, Present and Future", Invited Tutorial at the SPIE Laser Damage, Boulder CO. 2017.
13. [E. Chowdhury, N. Talisa, K. Werner, D. Austin, V. E. Gruzdev, K. R. P. Kafka, "Ultrafast bandgap photonics with few cycle pulses" \(invited\); *Ultrafast Bandgap Photonics II, part of SPIE Defense + Commercial Sensing, Anaheim CA, 9-13 April 2017.*](#)

14. "Femtosecond Laser Damage in mid IR wavelengths", Enam Chowdhury, Invited talk SPIE Pacific Rim May 2017, Shanghai, China.
15. E. A. Chowdhury, K. R. P. Kafka, N. Talisa, D. Austin, G. Tempea, and C. Neacsu, "Few cycle pulse Damage morphology of an ultra-broad band chirped mirror," in *Conference of Laser Electro Optics*, 2017.
https://doi.org/10.1364/CLEO_AT.2017.JTu30.4
16. *SPIE Laser Damage Symposium aka Annual Symposium on Optical Materials For High Power Lasers, Broomfield, CO, USA, 22-25 September 2019* V. Gruzdev, O. Sergaeva, D. Austin, E. Chowdhury, "Free-carrier dynamics in wide-band-gap crystals: Drude model versus Vinogradov equation" (poster);
17. *The International High-Power Laser Ablation Conference VIII (HPLA-20), Santa Fe, NM, USA, 26 – 29 March 2018* O. Sergaeva, V. Gruzdev, D. R. Austin, E. Chowdhury, "From Keldysh-Drude model to Keldysh-Vinogradov model: Fixing of some fundamental issues" (poster).
18. *SPIE Laser Damage Symposium aka XLIX Annual Symposium on Optical Materials For High Power Lasers, Boulder, CO, USA, 24-27 September 2017* Drake R. Austin, Kyle R. P. Kafka, Yu H. Lai, Zhou Wang, Kaikai Zhang, Hui Li, Cosmin I. Blaga, Allen Y. Yi, Louis F. DiMauro, Vitaly E. Gruzdev, Enam A. Chowdhury, "Wavelength dependence of the mid-IR ablation threshold of ZnSe" (oral);
19. *SPIE Laser Damage Symposium aka XLIX Annual Symposium on Optical Materials For High Power Lasers, Boulder, CO, USA, 24-27 September 2017* Vitaly E. Gruzdev, Olga Sergaeva, Drake Austin, Enam A. Chowdhury, "Beyond the Drude model: the Keldysh-Vinogradov model of ultrafast generation and heating of electron-hole plasma" (poster);
20. *32nd International Union of Radio Science General Assembly and Scientific Symposium (URSI GASS-2017), Montreal, Canada, 19-26 August 2017* V. Gruzdev, D. Austin, O. Sergaeva, E. Chowdhury, "Simulations of ultrafast laser-induced excitation and heating of electron sub-system of semiconductors with the Vinogradov equation and multi-band Keldysh formula" (oral)
21. *CLEO-2017, San Jose, CA, May 14-19, 2017* V. Gruzdev, D. R. Austin, O. N. Sergaeva, E. Chowdhury, "Beyond the Drude Approach: a Keldysh-Vinogradov Model of Dynamics of Ultrafast Laser-Induced Electron Excitation" (oral), STh4J.6, *CLEO: Science and Innovations*;
22. *SPIE Laser Damage Symposium: Annual Symposium on Optical Materials For High Power Lasers Boulder, CO, USA, 27-30 September 2016* V. Gruzdev, D. R. Austin, E. A. Chowdhury, "Influence of multiple energy bands on the photoionization of non-metal crystals" (oral)
23. "First principles simulation of the dynamics of transient warm dense matter during the formation of ultrashort laser pulse induced damage using the particle-in-cell method," Alex M. Russell and Douglass Schumacher, *Laser-Induced Damage in Optical Materials 2017*, SPIE Proceedings **10447**, 104470H (2017). DOI: 10.1117/12.2280543.
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25. Mid-IR applications of resonant semiconductor metasurfaces: from "perfect" diffraction gratings to tunable harmonics generation (Conference Presentation) Gennady B. Shvets; Maxim R. Shcherbakov; Zhiyuan Fan; Melissa Bosch; Kevin Werner; Noah Talis Talisa; Enam Chowdhury *Proceedings Volume 10928, High Contrast Metastructures VIII*; 109280W (2019) <https://doi.org/10.1117/12.2512064>

Event: [SPIE OPTO](#), 2019, San Francisco, California, United States

26. Manifestations of photon acceleration in semiconductor metasurfaces (Conference Presentation) [Maxim R. Shcherbakov](#); [Kevin Werner](#); [Zhiyuan Fan](#); [Noah Talisa](#); [Enam Chowdhury](#); [Gennady Shvets](#) [Proceedings Volume 10722, Plasmonics: Design, Materials, Fabrication, Characterization, and Applications XVI](#); 107220T (2018) <https://doi.org/10.1117/12.2321398>

Event: [SPIE Nanoscience + Engineering](#), 2018, San Diego, California, United States

27. Nonlinear Manifestations of Photon Acceleration in Rapidly Evolving Semiconductor Metasurfaces, Maxim R. Shcherbakov, Kevin Werner, Zhiyuan Fan, Noah Talisa, Enam Chowdhury, and Gennady Shvets CLEO (Conference on Lasers and Electro-Optics) OSA Technical Digest (online) (Optical Society of America, 2018), paper FTh1D.2 https://doi.org/10.1364/CLEO_QELS.2018.FTh1D.2
28. Strong Field Laser induced surface structuring in Mid-IR wavelengths, Enam Chowdhury, Noah Talisa, Kevin Werner, Shler Irani, and Drake Austin, High-Brightness Sources and Light-driven Interactions OSA Technical Digest (online) (Optical Society of America, 2018), paper HT3A.7 (<https://doi.org/10.1364/HILAS.2018.HT3A.7>)
29. Scanning Tunneling Microscopy Studies of Er Adatoms on GaAs (110), Smith, Rebekah; Benjamin, Anne; Werner, Kevin; Chowdhury, Enam; Gupta, Jay, APS March Meeting 2019.

Students Graduated:

PhD

1. K. R. P. Kafka, PhD. Dissertation Title, "Laser Induced Damage with Femtosecond Pulses". Dr. Kafka is a Scientist at the Laboratory for Laser Energetics at the University of Rochester, Rochester, NY.
2. Drake Austin, PhD. Dissertation Title, "Semiconductor Surface Modification using Mid- Infrared, Femtosecond Laser Pulses". Dr. Austin is currently a postdoctoral researcher at Air Force Research Laboratory in RX.
3. Kevin Werner, PhD. Dissertation title, "Ultrafast mid-infrared laser-solid interactions". Dr. Werner is Principal Laser Scientist at BAE Systems.
4. Alex Russell, PhD. Dissertation title, "Modeling Ultrashort Pulse Laser Damage with the Particle in Cell Method". Dr. Russel is a scientist at Voss Scientific.
5. Zhiyuan Niu, PhD. Dissertation title, "Nanoscale Self-patterning and Engineering of YSZ Surfaces". Dr. Niu is employed in industry.

Undergraduates:

1. Evan Lang, 2013-2017, *Ultrafast fiber oscillator development*, Manufacturing Engineering Manager at EMCORE Corporation
2. Manh Le, 2014-2018 *UV ultrafast beamline development for nano-LED*, graduate student at Milchberg group at University of Maryland