

**The intense laser-plasma interaction:
Modeling ion acceleration and laser damage.**

A Proposal Submitted to the
Ohio Supercomputing Center

Total Request: 30,000 Resource Units (Major Proposal)

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Abstract

We will use particle-in-cell (PIC) numerical simulations to explore the fundamental and applied light-matter interaction for intensities ranging from 10^{11} W/cm² up to 10^{22} W/cm². We are a combined experimental and computational group and the proposed computational program is closely tied to recent experimental runs performed using our lasers Scarlet and Gray, as well as the laser Draco (Dresden, Germany). In these experiments we demonstrated novel forms of laser driven ion acceleration and we performed precision laser damage experiments for benchmarking computational models. We have developed extensive PIC simulations to model these problems and achieved good agreement with experiment thus far. [REDACTED]

[REDACTED] In particular, we have observed what appears to be a new ion acceleration regime and we have achieved agreement between measured laser damage profiles and a novel simulation technique with no tuned parameters. The proposed work will explore and try to better understand the new ion acceleration regime which appears to be a kind of hybrid Target Normal Sheath Acceleration regime and to improve our laser damage model by exploring its predictions for shorter excitation times and by longer wavelengths.

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Proposal Outline

- I. Introduction**
- II. Previous Results**
- III. Proposed Research Program**
- IV. Conclusion**

I. Introduction

Overview

All ordinary, neutral matter ionizes when exposed to light at intensities on the order of 10^{12} W/cm² to 10^{13} W/cm². Current state-of-the-art intense field laser experiment explores light-matter interactions at intensities exceeding 10^{21} W/cm² and so these studies necessarily involve plasmas under extreme conditions. The study of phenomena under these conditions goes by several names, including High Energy Density Physics (HEDP).¹ HEDP is being aggressively explored by universities and national laboratories around the world for two reasons. The first is fundamental. All light-matter interactions begin with the force light exerts on the electronic system and in HEDP this interaction is highly relativistic for intensities significantly above 10^{18} W/cm² leading to unique phenomena such as relativistic transparency. This single fact turns ordinary phenomena at low intensities into a rich set of novel behaviors at high intensities. Even seemingly simple questions are still hotly debated after years of study. One example is the fundamental question of how the energy of the coherent interaction of the electrons with a laser quickly becomes an incoherent thermal or directed energy.^{2,3,4} The second reason for the broad interest in HEDP is practical. Intense laser-matter interactions can be used to generate intense beams of electrons, positrons, protons, ions, and x-rays that can be used as powerful experimental diagnostics and might be suitable for applications such as cancer therapy and the detection of hazardous materials.

The parameter range of HEDP laser-matter experiment is vast, ranging from kJ to MJ laser systems firing a few times a day at large national laboratories to mJ university systems operating

at kHz repetition rates. A new generation of university based laser systems has emerged in the last decade that occupies a middle ground with energies roughly from 1 to 100 J with repetition rates ranging from once per hour to 1 Hz. These laser systems have set the record for highest intensity and they all produce short laser pulses with pulse durations under 1 ps, permitting experiments with excellent time-resolution. The relatively high repetition rate of these lasers allows them to perform experiments that could not otherwise be attempted as well as to act as staging grounds for experiments on larger facilities where kJ and above pulse energies are available. The Ohio State University has one such system, Scarlet,^{5,6} and the PI also uses external facilities such as Astra (UK) and Draco (Germany).^{7,8} Scarlet currently generates up to 10 J pulses at 1 shot per minute with a pulse duration near 30 fs and a focused spot size of 3 μm (duration and size given as intensity full-width at half-maximum (FWHM)).

Most of the experimental work requires high-performance computing based numerical analysis for the interpretation of experimental results. Many of these experiments benefit greatly in the planning stage from modeling as well. [REDACTED]

[REDACTED] Attempted publication of our experimental results without intensive modeling would severely reduce the impact of our work, so the time requested in this proposal is crucial for the successful completion of this work. These experiments and the associated computational program are funded by multiple grants from the DOD and DOE.

Support is requested to study:

- Laser driven ion acceleration from very thin targets; data analysis on results from two runs.
- Laser damage of metal targets; data analysis on results from two runs.

Background

In the relativistic regime the laser intensity is sufficiently high that an electron will accelerate to close to the speed of light within an optical cycle. By convention, this regime is often taken to begin when the normalized amplitude $a_0 = eE/m\omega c$ is unity or, roughly, when the laser intensity exceeds 10^{18} W/cm^2 . This field is of fundamental interest to the plasma science community because the ultrahigh intensity laser-plasma interaction involves high temperatures,^{9,10,11,12,13,14} enormous fields,^{15,16} and severe gradients and the behavior of matter in this regime is still not

well understood. It is not surprising that so much is not understood in this regime despite the intense attention it has received from talented research teams at universities and national labs around the world. The “phase space” of HEDP covers, currently, roughly four orders of magnitude in intensity from $10^{18} - 10^{22} \text{ W/cm}^2$, nine orders of magnitude in energy from mJ to MJ, and six orders of magnitude in time scale from about 10 fs to 10 ns. There is also great variety in the kinds of targets used, from gases to metals. Two experiments performed at, for example, 10^{20} W/cm^2 but using 100 fs and 10 ps pulses, are very different experiments. For example, the blow-off plasma in front of the target, which controls laser absorption, will vary widely over 10 ps in a way not possible over 100 fs. Much of the leading work in this field has taken place at large national facilities which can only provide a few dozen shots for any given experiment. Limited sampling of a vast phase space means there is much to be explored, hence the current vitality of the field.

Study of the laser-plasma interaction (LPI) has led to the observation of dramatic and useful effects including: the generation of large numbers of relativistic electrons;^{17,18,19,20,21,22,23} the acceleration of electron pulses using wake fields;^{24,25} the generation of short pulsed, MeV to GeV, ion beams via the target normal sheath acceleration and break-out after burner mechanisms;^{26,27,28,29,30,31,32} and the creation of dense clouds of positrons via the Bethe-Heitler effect.^{33,34} Understanding the propagation of a laser in an HEDP environment is challenging because the intensities used require a relativistic treatment. The laser-plasma interaction can cause the laser beam to change shape spatially and temporally due to interactions such as relativistic laser modification of the index of refraction.³⁵ The laser-plasma interaction also modifies the density and temperature profile of the plasma itself as gradients in the laser electromagnetic field push plasma particles.³⁶ The plasma-modified laser and the laser-modified plasma continue to interact until the laser is either absorbed, reflected or scattered. There are many absorption mechanisms, but all involve transferring energy to electrons that is then randomized, at least partially, by various mechanisms (collisions, vacuum heating, resonance absorption and $\mathbf{J} \times \mathbf{B}$ heating) that assume different relative roles that vary with intensity and density.³⁵

Particle-in-cell simulations

We propose to use numerical modeling to analyze recently concluded experiments using our lasers Scarlet and Gray, and the laser Draco. In particular, we will model realistic representations of these experiments in 3D and, where appropriate, in 2D3V. In 2D3V the simulation grid has 2 spatial dimensions, but vector quantities (particle momentum, associated current densities, the electric and magnetic fields) have all three components. This permits self-consistent electromagnetic field generation and propagation. Our primary tool is the commercial code LSP (Large Scale Plasma, ATK Mission Systems) for this effort.³⁷

Briefly, LSP is a particle-in-cell (PIC) based code that reduces the $\sim 10^{23}$ particles in a real experiment to, in our case, $\sim 10^8$ macro-particles residing in a gridded space. LSP employs a variety of algorithms to propagate the macro-particles and to calculate the self-consistent fields associated with them and any external fields present, such as a laser. It offers both explicit and implicit advancement for particles and fields and can be resistant to numerical heating, even when the Debye length is not resolved.³⁸ LSP has multiple models and algorithms to include the effect of collisions, ionization, radiative loss and other effects. In addition to performing purely kinetic simulations, LSP has two fluid models allowing it to solve problems hydrodynamically. Fluid and kinetic treatments can be incorporated into the same simulation in many cases and, for this reason, LSP is described as a hybrid code. This code can handle targets of arbitrary shape (subject to discretization errors) and content. LSP uses MPI to coordinate multiple processors and the simulation space can be divided into multiple grids, regions, and domains. Some of these aspects can be changed dynamically during run-time. Purchase of LSP includes full access to the source code. This is crucial since almost every project we have undertaken has required some modification of the code, either to fix bugs or to add new features. LSP technical support is provided by the developers and they are helpful in these efforts and responsive to requests for new features.

II. Results from the existing ██████████ Allocation

The requested computing is to finish on-going analysis begun under the existing, and now depleted ██████████ Allocation. *Every publication and talk described below explicitly credits OSC.*

Peer-reviewed publications in 2016 based on OSC conducted research

- (I) P. L. Poole, A. Krygier, G. E. Cochran, P. S. Foster, G. G. Scott, L. A. Wilson, J. Bailey, N. Bourgeois, C. Hernandez-Gomez, D. Neely, P. P. Rajeev, R. R. Freeman, and D. W. Schumacher, “Experiment and simulation of novel liquid crystal plasma mirrors for high contrast, intense laser pulses,” *Scientific Reports* **6**, 32041 (2016).³⁹

We recently developed a novel technology for ultraintense laser targetry based on liquid crystals.⁴⁰ This technology allows us to form high quality thin films with tunable thickness as low as 10 nm on-demand and precisely aligned to the laser focus. This capability is currently unmatched by any other technology that we are aware of. We realized that this capability would also allow us to make reformable plasma mirrors (PMs) for ultrashort pulse laser contrast enhancement and demonstrated this recently at a run using the laser Astra in the UK. Plasma mirrors transmit the “junk” light referred to as pre-pulse that is in advance of the main pulse, Fig. 1. This is crucial since, with a peak intensity exceeding 10^{21} W/cm², even pre-pulse reduced in intensity by eight orders of magnitude will damage the target. The intense main pulse turns the PM “on” by exciting a plasma and is reflected. We demonstrated that quality PMs, until now based on high quality commercial optics, could be fashioned using liquid crystal films. We have also separately demonstrated film formation at 1 Hz and thus a 1 Hz PM device is now possible. This would have far reaching consequences in the development and operation of state-of-the-art laser facilities.

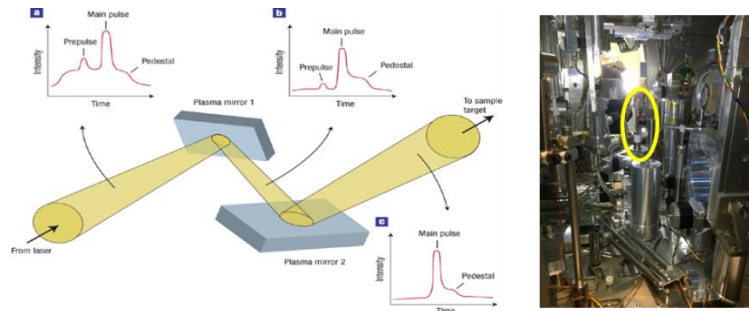


Figure 1: (left) The PM approach to pulse contrast enhancement shown with a double PM system.⁴¹ (right) Photo of experimental chamber configuration for the Astra experiment. A single liquid crystal film device was used, circled in yellow.

Despite the importance of PMs for pulse contrast enhancement, with many experiments performed today only possible with their use, fundamental modeling of PM operation has never been reported in the literature. This is because the natural numerical approach, PIC, is

not consistent with treating PM operation. PIC simulations generally model targets as collections of pre-ionized ions and electrons but, for PM simulations, this would result in a PM that begins in the “on” state. Most PIC simulations use a tunneling ionization model for photoionization, but plasma mirrors operate in the multiphoton ionization regime. Finally, for our approach a model for dielectric constant is required so that thin film interference is treated, but PIC codes generally do not incorporate such a model. We have performed a simulation that addresses all of these issues: the target begins with neutral atoms with the same index of refraction as the liquid crystal 8CB (1.53 for our conditions) and ionizes via a multiphoton ionization (MPI) model that we added to LSP. Finally, we included a collision model using capped Spitzer collision rates to treat plasma losses.

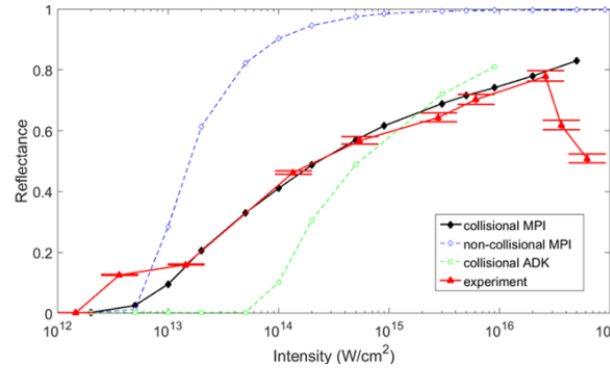


Figure 2: Comparison of experiment and PIC simulation for liquid crystal plasma mirror operation. (black) Astra results for liquid crystal plasma mirror reflectivity as a function of intensity. The simulation (red) is a good fit to experiment over three orders of magnitude in intensity. (The blue and green curves show the effect of different models, discussed below.)

The simulation results are compared against experiment in Fig. 2. There is excellent agreement across three orders of magnitude in intensity including the optimal operating point of 10^{16} W/cm². For the lowest and highest intensities the model diverges from experiment, we believe due to treating the liquid crystal 8CB used for this work as a collection of its constituent atoms and, also, not resolving the Debye length at the highest intensities. The disagreement at low intensities is important since it affects our ability to predict contrast enhancement and further work will be done on this in the future. However, such a comparison between simulation and experiment has never been achieved before for this problem to our knowledge and it is a key achievement of this project. The green curve in Fig. 2 shows the effect of using the ADK tunnel ionization model used by most PIC codes for photoionization – it completely fails to capture the low intensity behavior illustrating the

need for an MPI model. Similarly, the blue curve shows the results if the collision model is not used – without collisional dephasing, plasma losses are not modeled and the reflectivity is unrealistically high.

Manuscripts submitted for publication in 2016 based on OSC conducted research

- (1) *R. A. Mitchell, D. W. Schumacher and E. A. Chowdhury, “First model of laser-induced periodic surface structure based on microscopic particle evolution from excitation to damage,” submitted to Physical Review Letters.*

As described in previous reports, we recently showed for the first time that the PIC method could be adapted to the target heating and damage problem.⁴² This was done by adding an atomic pair potential model to the PIC integration of the equation of motions which usually only include the interaction between monopoles (and currents). In this work, we used our model to treat laser-induced periodic surface structure (LIPSS), a damage phenomenon where the resulting damage pattern is in the form of a self-induced grating structure. This is a highly studied phenomenon and we demonstrated that it could be modeled from first principles, permitting analysis of the formation mechanism which we showed involved the excitation of surface plasma polaritons.

- (2) *A. M. Russell and D. W. Schumacher, “Extending the Nanbu Collision Algorithm to Non-Spitzer Systems and Application to Laser Heating and Damage,” submitted to Physical Review E.*

PIC methods cannot directly treat particle collisions due to the finite spatial resolution imposed by the spatial grid. The Nanbu collision algorithm is a widely used approach to restore the effect of collisions using a Monte Carlo treatment in a way that PIC codes can efficiently implement.⁴³ The algorithm as introduced and generally used implements collisions with Spitzer rates which are appropriate for hot or relatively low density plasmas. Spitzer rates are invalid for studying the initial heating of a target, however, due to their divergence with decreasing temperature. As described above, we have introduced a new approach to PIC modeling for treating laser damage. In that work, we used a non-kinetic (not particle based) approach to treating collisions, but in general kinetic approaches are needed because of the non-thermal distributions of heated electrons that arise. In this work, we demonstrated a modification of the Nanbu algorithm that can work for a general collision model and applied it to the initial phase of the target heating problem.

Talks and presentations based on work done at OSC:

During 2016 we have thus far given two invited talks at the BELLA-i Workshop and the ELIMED Workshop; three seminars at Voss Scientific, Lawrence Livermore National Laboratory, and Lawrence Berkeley National Laboratory; two project reviews at the NNSA SSAP Meeting and the DAPRA PULSE Project Review; and several contributed presentations. There will be two contributed talks at the upcoming APS Division of Plasma Physics Meeting and one at the upcoming SPIE Laser Damage Meeting.

III. Proposed Research Program

Support is requested to continue studies of:

1. Laser driven ion acceleration from very thin targets; data analysis on results from two runs.
2. Laser damage of metal targets; data analysis on results from two runs.

(1) Laser driven ion acceleration.

Laser driven ion acceleration, in particular proton acceleration, is a major topic in HEDP. We have completed two ion acceleration runs facilitated by our liquid crystal technology. One used our laser Scarlet and the other used the laser Draco in Dresden, Germany. Both runs indicate a new acceleration mechanism from those identified thus far.⁴⁴ Specifically, the spectrum is like that expected from the so-called Target Normal Sheath Acceleration (TNSA) mechanism, but the resulting ion energy is surprisingly high, up to 25 MeV, using as little as 2-3 J/pulse in tightly focused, 40 fs pulses (this is a record). Also, the optimal targets were surprisingly thin (as low as 10 nm). In a long series of simulations we believe we have identified the numerical and physical criteria that must be satisfied for successful simulations in this ultraintense, thin target regime. In particular, we have shown that 2D simulations (the standard in this area) exaggerate the proton energy by at least a factor of 2 due to an artificially slow fall-off of the accelerating electric fields in space and time. Results from our 3D simulations are compared to experiment in Fig. 3.

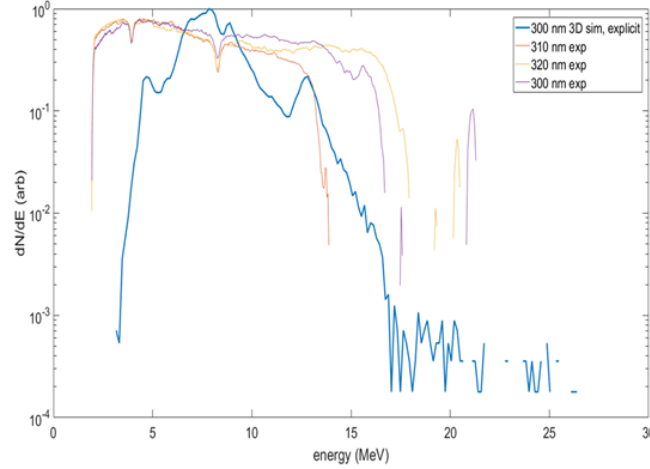


Figure 3: Comparison of experiment and PIC simulation for accelerated proton spectra from the Draco run for a 300 nm target. The simulation is shown in blue along with three shots taken with similar parameters. Note the log scale.

Although there are significant differences in the overall spectral shape, this level of agreement is state-of-the-art for this problem. In particular, the maximum proton energy of 14-18 MeV is well represented. In the TNSA mechanism, the ions are accelerated in the sheath field at the boundary of the expanding plasma coming from the target and travel roughly normal to the target. In the Radiation Pressure Acceleration (RPA) mechanism, light pressure directly acts on the electronic system to create a quasi-static electric field that accelerates the ions in the laser axis direction. The simulation shows the protons traveling in the target normal direction which is what is seen in the experiment, but radiation pressure appears to be critical nonetheless. The combination of high energy, thin target optimized, target normal proton acceleration suggests a different mechanism or unusual combination of mechanisms than that observed previously. We believe clarification of the acceleration mechanism would be high impact given the importance of ion acceleration in the literature and the high energies obtained here with such low energies. For protons, 50 to 85 MeV has been demonstrated in multiple experiments, but using up to 100 J pulses.⁴⁵

Proposed program. We need to further benchmark our approach by running 3D simulations for other target thicknesses measured in the experiments which cover a range from 10 nm to 2 μm . At a minimum, we need a set of simulations for 10 nm, 100 nm, and 300 nm (used for Fig. 3). If we can model all three thicknesses using the same simulation technique with a level of agreement similar to that shown in Fig. 3, we will have a very successful benchmark. This will allow us to understand the underlying acceleration mechanism as well as predict how it can be optimized for upcoming experiments scheduled for 2017 on Scarlet and other laser facilities in the U.S. and Germany. Also, although the experiment only measured results for p-polarized light, simulations comparing s- and p- polarizations would be highly instructive

since the angle of incidence was $\sim 45^\circ$, meaning there is a large difference in the *optical* electric field normal to the target during excitation.

(2) *Laser damage.*

This program examines laser damage⁴⁶ of materials at laser intensities in the range of 10^{12} W/cm² to 10^{16} W/cm² – as much as eight orders of magnitude lower than the intensities associated with the HEDP experiments described above. Laser damage is a fundamental problem in its own right, but is also useful for understanding phenomena at the highest intensities since the target evolution as the pulse turns on, before the peak arrives, can be crucial. Finally, laser damage is the basis of the important practical applications of laser surgery and laser machining. One of the primary tools currently used to model laser damage is molecular dynamics (MD) simulation.⁴⁷ PIC, on the other hand, is not generally used in this field. MD is effective because of its realistic treatment of interatomic potentials, allowing realistic models of void formation and other processes that begin the damage process. However, the optical interaction is not generally treated realistically and MD simulations are currently limited to small ~ 100 nm (often 10 nm) sized regions, whereas the damage morphology that must be understood typically extends over several microns. PIC approaches, however, can readily treat aspects of the problem over the micron scale lengths required, including the interaction with the laser, but do not incorporate interatomic interactions.

As described in Section II, we have a new approach that can be seen as combining the approaches of PIC and MD. Despite our recent initial successes, see reference 42 and the two manuscripts submitted for publication described in Section II, we have not until recently performed careful benchmarks against experiment. Although this may seem odd, there are generally a large range of results for nominally similar conditions in the literature, and benchmarking is not possible. There are many reasons for this: not all laser systems are equally well characterized nor experiments well diagnosed but, also, laser damage can depend sensitively on the laser and targets. For example, copper targets behave differently depending on the amount of oxidation present. These issues stand in the way of careful benchmarking. It is also worth noting that, before our approach, an *ab initio* method for predicting damage morphology has not existed against which to compare experiment. Recently, Prof. Enam Chowdhury of the OSU physics department has performed a highly characterized study of laser damage in copper using

the laser Gray, with the experimental conditions selected to facilitate comparison to our model. In particular, the laser was tightly focused to reduce the size of the simulation grid required. The results of his experiment and our simulation results, performed this year, are compared in Fig. 4.

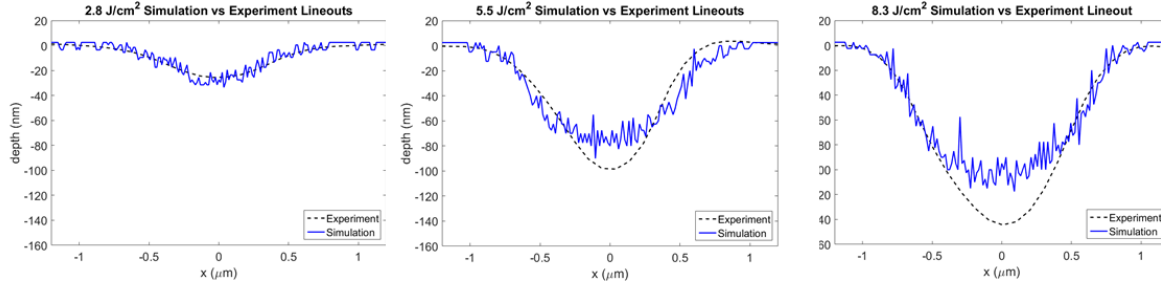


Figure 4: Comparison of experiment (black) and PIC simulation (blue) for laser damage morphology using clean, smooth, single crystal copper targets. Each plot is a cross-section from the damage crater which was nearly circular in both cases. From left to right are single shots at the indicated laser fluences. The laser pulse was 40 fs in duration and focused to a 1 μm waist with a center wavelength of 800 nm.

To our knowledge, such a comparison has never been done before and we note that there were no tuned parameters in the simulations. Each began with a bare copper target and an incident laser, following the target evolution over 6 orders of magnitude in time scale: from femtoseconds for the laser excitation to nanoseconds for the final target evolution. The agreement is very good, although it lessens with increasing fluence. It should be noted that the smoothness of the experimental line is due to the spatial resolution of the microscopy used. Scanning electron microscopy shows a bumpy surface similar to that predicted by the PIC simulations.

Proposed work. We would like to complete two tasks in order to publish. First, Prof. Chowdhury has collected a second set of shots using 5 fs pulses and performed in vacuum. (Vacuum measurements are rare in this field but may be crucial since ionization of the air in front of the target may have a significant effect.) We need to run another set of simulations so that this data can serve as an additional benchmark. Second, analysis of the simulations shown in Fig. 4 indicates that the bumpiness of the damage crater is due to void-like formation inside the target as it ablates. The formation of these structures has been studied in MD simulations and might be a good way to compare our approach against that of MD for a case where we have quality experimental results. This analysis may require additional computer time. Finally, in what would constitute a new study, we would like to run simulations for longer wavelengths than the 800 nm

used in the results of Fig. 4. Prof. Chowdhury has performed experiments at wavelengths exceeding $2\text{ }\mu\text{m}$ and has obtained interesting results. Our PIC analysis may prove useful here.

IV. Conclusion

Thus far in 2016 we have published one paper in Scientific Reports and submitted two papers to Physical Review Letters and Physical Review E based in part or in whole on simulations performed at OSC. We have also given multiple invited talks, seminars, and contributed presentations. In addition to this, we have new results on ion acceleration and on laser damage that are intriguing and agree reasonably well with recent experiments. This request is for additional computer time so that these studies can be completed and submitted to high impact journals.

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