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Understanding Laser-Cluster Interactions in the X-ray Regime

Edward Ackad
SOUTHERN ILLINOIS UNIVERSITY

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Air Force Research Laboratory
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14. ABSTRACT
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Abstract

This project proposed to determine if a purely microscopic model of laser-cluster interactions is valid and, if it is valid, use the model to understand the microscopic origin of observed macroscopic phenomena. The short term objective of determining a correct laser-cluster model also aids in the overarching objective of developing a microscopic laser-solid model. Using a hybrid quantum-classical model the laser-cluster interaction is simulated atomistically, tracking and moving each particle using molecular dynamics. Experimental signals were reproduced through the simulation and compared in order to validate or invalidate our model.

The work resulted in significant evidence for our model which can reproduce experimental results and explain the origin of different phenomena. We were able to calculate and propose an experiment to distinguish between the major rival models, giving a clear signal as to which is physically relevant and settle the decade-long controversy in the field. We've used our model to understand the underlying mechanisms involved in the resultant phenomena of the laser-cluster interactions.

Introduction

The new regime of ultra intense and short X-ray light-matter interactions is fundamentally different than any other regime since the wavelength of light is smaller than the atoms. It is my hypothesis that all new phenomena occurring in ultra intense and short X-ray light-matter interactions is the result of emergent properties of the system, not new fundamental processes. I therefore conjecture that a fundamental, microscopic theoretical/computational model of the interaction using well-known processes will be capable of replicating experimental results. The macroscopic properties then emerge through the evolution of the system due to its complexity, not some new fundamental property. This was and continues to be assessed by reproducing experimental results and proposing experiments to test emergent phenomenon found theoretically. The success of an atomistic model in reproducing homogeneous cluster signals and heterogeneous cluster signals strongly favors the idea that *all the phenomena observed in dense X-ray irradiated systems is emergent*.

Understanding X-ray Matter Interactions

Clusters are nanoscopic, condensed systems which bridge the gap between the gas and solid phases of matter. Their finite size, but almost solid density, can potentially make their properties very different from other forms of matter. Moreover, their interactions with intense lasers as well as their electronic properties are size dependent. Work done over the last decade on ultra-high intensity and ultra-short laser pulses interacting with rare-gas clusters has shown that the laser-cluster interaction is wavelength dependent. Cluster irradiation with high intensity lasers has been investigated since the 1990s [1] where a plethora of phenomena have been found, including: the emission of very energetic electrons (10s of keV), highly-charged ions (MeV) [2], X-rays [3], and neutrons [4]; each of which have promising scientific, industrial and biomedical applications. This proposal seeks to clear-up and expand our knowledge of laser-cluster interactions to new regimes now accessible experimentally as well as to increase our understanding of this nanoscopic phase of matter.

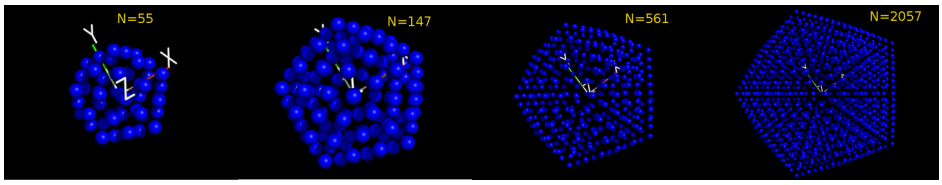

Illustration 1: Various sized rare-gas clusters.

All previously mentioned phenomena and investigations were the result of irradiation with wavelengths on the micron scale. Most effects were the result of the cluster absorbing large amounts of energy from the laser through inverse Bremsstrahlung heating (IBH) (the process by which radiation from the laser is absorbed by free electrons due to the oscillating electric field of the laser pulse). This process becomes much less efficient at small wavelengths scaling as λ^2, where λ is the wavelength.

The new generation of free-electron lasers, such as the DOE sponsored Linear Coherent Light Source (LCLS) at SLAC, opens the door to unexplored regimes of laser-cluster

interactions beyond even the extreme ultraviolet and well into hard X-rays. In this regime there are notable differences in the interaction of a cluster with a short wavelength laser pulse.

Single-photon ionization from the ground state of rare-gas atoms is accessible and a significant source of ionization. In X-ray pulses, inner-shell ionization (the ionization of an electron from a lower orbital than the valence shell) is also possible (and in many cases dominant over outer shell). In addition, unlike in the infrared (IR ~800 nm) and vacuum ultraviolet (VUV, ~100 nm), the IBH of freed electrons is negligible, decoupling cluster's plasma dynamics from the laser field [5,6]. However, electrons ionized from atoms often have a significant amount of kinetic energy and can collisionally ionize neighboring atoms in the cluster due to the clusters almost solid density.

At all wavelengths, provided there is sufficient intensity, a nanoplasma is formed in the cluster. The decoupling of the electronic motion from the electric field of the laser significantly changes the character of nanoplasma [7]. The dominant ionization mechanism which generates the nanoplasma remains fairly controversial at shorter wavelengths. Experiments on clusters in the VUV regime near 100 nm [1] observed unexpectedly high ionic charge states and much larger energy absorption than predicted by existing models [8]. This sparked a concerted theoretical effort over the last decade [2-6]. All of these studies justified their models by the appearance of a small population of Xe^{7+} or Xe^{8+} at the peak of the laser pulse, consistent with what was reported by the experiment. However, the intensity from this experiment [1] was recently revised [7] to be an order of magnitude lower than originally reported. Thus it is not clear if any of these mechanisms can account for the experimental result as none predicted higher charge states at the higher intensity. This necessitates a revisiting of the models to bridge the gap between theory and experiment.

These different models have continued to be used at even shorter wavelengths to explain current results in the extreme ultraviolet (XUV, ~30 nm) with moderate success. We propose a model that is simple, based firmly in well known atomic physics, and has been successful in reproducing the results of recent experiments. Our results for the 100 nm experiment are being prepared for submission.

All models of the laser-cluster interaction include both single-photon ionization and collisional ionization. In most models, ionization is treated as a single step process where the electron is promoted from the ground state to the continuum (often the continuum of the atom, but remaining bound to the cluster, so called inner-ionization). In our model, we augment the standard ionization channels to include the possibility of collisional excitation. An electron can first be promoted from the ground state to an electronic excited state (for Xe I, for instance, from the 5p to the 6s) by a collision of a freed electron. This excited electron can subsequently be ionized by being promoted from the excited state to the continuum. While the whole trip can be energetically the same, breaking the process up into two steps means each step requires less energy. This allows an electron with less kinetic energy (compared with single-step ionization) to execute the process. In a plasma, the energy distribution is, on average, Maxwellian and thus

there are many more electrons with enough energy to excite an atom than there are who can ionize an atom directly. This ionization pathway leads to: higher charged states in the cluster and collisionally reduced photo absorption [14].

We use a hybrid approach that has proven highly successful in reproducing the results of experiments [14,15]. The motion of the particles is solved using molecular dynamics (mainly on gpus), individually tracking every particle. The probability of an ionization to occur is calculated using experimental data and calculated cross-sections for: single-photon ionization, Auger decay, collisional excitation and ionization. Lastly, we also allow for ionized electrons to recombine with ions. A large number of simulations are performed with different random seeds to obtain the correct macroscopic behavior. The enormous amount of computation needed is only within our reach due to the cutting edge implementation of the molecular dynamics on the gpu.

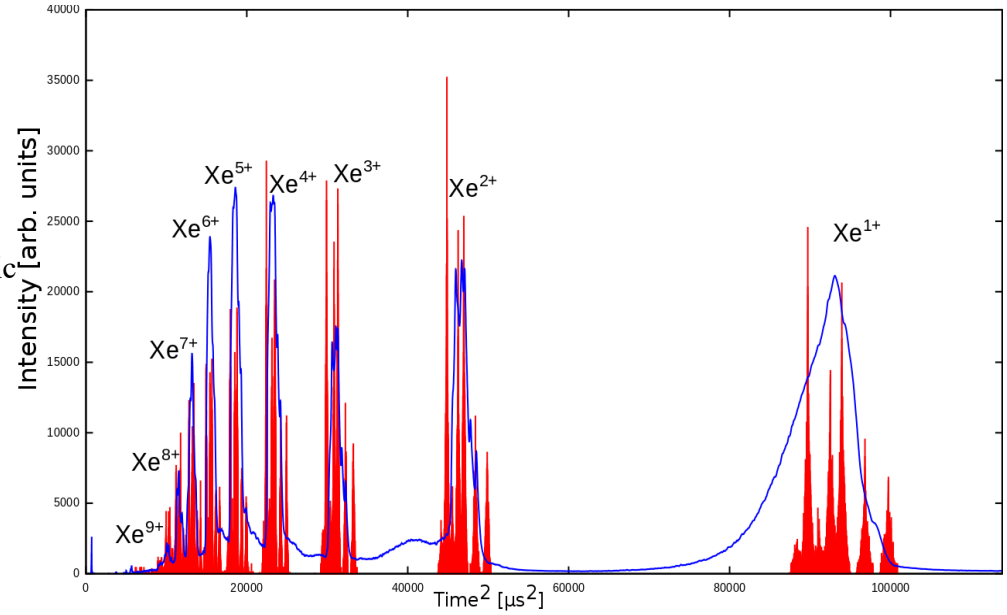

Figure 1: Comparison of the experimental time-of-flight ion signal obtained in [17] (blue line) with our calculated signal (red boxes) [18].

We then take the process a step further to reproduce the full experimental signal. Recent experimental advances at the LCLS have allowed experimentalists there to image a single disintegrating cluster, instead a distribution of clusters with different sizes [16]. This provided the cleanest single-shot data ever obtained for clusters. However, most experiments are not performed under such sterile conditions and the experimental signal contains a mix of different cluster sizes (due to the way clusters are formed) and of different laser intensities irradiating the cluster (due to the clusters being in or away from the focal spot of the laser). Thus the experimental signal in most experiments is a composite of many differently sized clusters being irradiated at a range of intensities. We simulate a large number of clusters (to get statistically convergent results) at different intensities and build a composite picture of the experimental signal. This is done by post-processing the aforementioned results disallowing further ionization and using dynamic time-steps. We propagate the particles all the way to the detector in a procedure which mimics the experimental setup. Our signal can then be directly compared with

the published results as shown in figure 1 for the experimental ion time-of-flight data from reference [17] is compared with our calculated spectrum [18]. The multiple peaks for each charge state are due to the isotopic distribution of xenon which is needed to match the full experimental signal.

With the ability to directly compare our model with experiments for validity and explore, in detail, the laser-cluster interaction, we were able to resolve the lingering questions about the ionization mechanisms in clusters from the vacuum ultraviolet of the LCLS's predecessor, FLASH in Germany, to the hard X-rays of the LCLS.

The following three research tracks deal with different aspects of the laser-cluster interaction.

Track 1: Ionization mechanism investigation

Motivation

The mechanism for rare-gas cluster ionization when irradiated with intense short wavelength radiation remains a controversial and active area of research. The theoretical follow-ups to the 2002 Nature paper by Walbntiz *et al.* [1] illustrate the ongoing debate. In the decade following that publication many attempts have been made to explain the results [2-6]. It is unlikely that any of these provide the complete picture as Bostedt *et al.* [7] recently published a revised measure of the laser's intensity which was around an order of magnitude lower than reported in the original Nature publication. None of the theoretical works saw any higher charge states when compared with the experiment which is expected as they were solving for a higher intensity than the original experiment reported [1]. This necessitated revisiting the question of what mechanism caused such dramatic ionization and even more so because these same mechanisms were used to explain subsequent experiments at even shorter wavelengths.

The laser-cluster interaction is highly dependent on the wavelength of the laser. In the infra-red (IR), tunnel ionization and IBH dominate. The subsequent plasma dynamics are driven by the laser field itself [10,19]. This is not true at shorter wavelengths. Experiments on clusters in the vacuum ultraviolet (VUV) regime near 100 nm [1] observed unexpectedly high ionic charge states and a much larger energy absorption than predicted by existing models [8]. This sparked a concerted theoretical effort over the last decade. Santra *et al.* proposed an enhancement to inverse bremsstrahlung heating (IBH) due mainly to using self-consistent potentials for ions [2] and later plasma screening effects [3]. Jungreuthmayer *et al.* proposed a many-body dielectric recombination scheme for the strongly-coupled plasma electrons in the cluster whereby the electrons could then be driven by the laser field to reionize [4]. Siedschlag *et al.* proposed allowing photo-ionization to occur above the classical potential barrier of the neighboring ion [5]. Ziaja *et al.* incorporated many of these and found they all played various roles depending on the intensity [6]. All these works sought validation of their models by attempting to reproduce

the highest ionic charge states seen by Walbnitz et al. [1], Xe^{8+}. They did this by irradiating clusters of about the same size as in the experiment with the peak of the laser pulse. After the laser pulse, they examined what the highest charge state was.

The models made the assumption that the time-of-flight field (the field that accelerated the disintegrated charged fragments of the cluster to the detector) was strong enough to ionize any weakly bound electrons. Further, all the models made the assumption that the ionized electrons would not recombine and lower the charge state seen by the detector. For instance, as a Xe^{8+} leaves the semi-neutral core of the cluster, upon disintegration, few electrons become ensnared in its potential. In previous models, these electrons are assumed to be ionized by the time-of-flight field. Our model makes neither of these assumptions, but allows for any of those processes to happen.

These models did not see any charge state higher than Xe^{8+}. It is unlikely that these models increase the ionization sufficiently to still see Xe^{8+} with the revision of the peak intensity to 8×10^{12} W/cm^2 from 7×10^{13} W/cm^2 and with the full-width-at-half-max of the pulse's duration updated to 50 fs from 100 fs, all resulting in pulses that were 5 times less intense than the theoretical work. This necessitated revisiting the problem, especially since these models are still used to explain more recent experiments.

This work was completed and submitted for review to the New Journal of Physics and as of this writing is under review. It includes a comprehensive explanation of the experimental results as well as the underlying ionization mechanisms to settle the decade's long debate. Using the actual experimental parameters for the pulse we were able to mimic the experimental signal and see all the same charge state peaks as seen in figure 2.

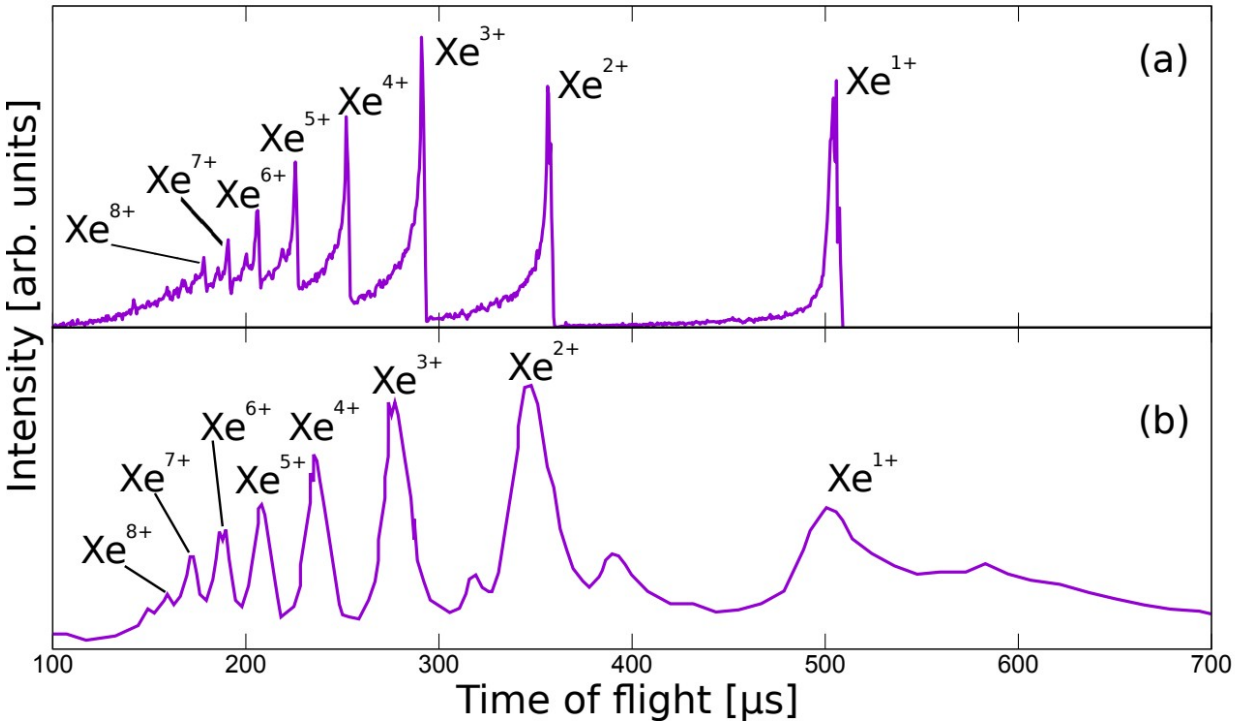

Figure 2: (a) the predicted time-of-flight signal for a 100 nm pulse with a full-width-at-half-max of 50 fs and a peak intensity of 2.8x10^{12} W/cm^2 and is compared with (b) which is the experimental signal.

Projects

Controlling collisional ionization with a second long-wavelength pulse

Question: Does collisional ionization dominate the ionization in the XUV regime?

This work will model a hypothetical experiment which would use an initial extreme ultraviolet (XUV) pulse to create a nanoplasma. Following soon afterwards would be a long-wavelength pulse of insufficient intensity to ionize on its own (via tunnel ionization or multi-photon ionization). The second pulse would only interact with the cluster via inverse bremsstrahlung heating (IBH), resulting in increased collisional ionization. As the intensity of the long-wavelength pulse is increased, collisional ionization will increase dramatically due to IBH being proportional to λ^2, where λ is the wavelength of the laser.

The extreme ultraviolet (XUV) regime (defined for this proposal as from around 60 nm to 30 nm) is uniquely suited for an investigation of the importance collisional ionization processes play in laser-cluster interactions. In this regime, single-photon ionization of neutral atoms as well as from some low charge states is possible. However, inner-core ionization is not energetically

possible, even for the neutral atoms. This simplifies the photo-ionization picture significantly. Further, the wavelength of the laser is short enough that IBH contributes a negligible amount of energy to the electrons. Thus the only communication between the laser and the cluster is through single-photon ionization.

This work was completed and incorporated and published along with other work. We have found that collisional ionization is by far the dominant ionization mechanism within the XUV and X-ray regime, contrary to much of the prevailing models which favored photoionization. This question is a core part of each and every piece of work we have done.

Induced Single-Photon ionization Transparency - Testing barrier lowering

Question: Does barrier lowering occur [5], and can it be detected in an experimental signal?

This work modeled an experiment with two short XUV pulses impinging on the cluster in rapid succession. The first pulse was of sufficient intensity that it would completely saturate the single-photon ionization channel. This would mean that every ion that could be ionized in gas form has been ionized in the cluster. For instance, a gas of argon irradiated by 50 nm wavelength (24.8 eV) laser would only ionize up to Ar^{2+} since the ionization potential of Ar^{2+} is to high (27.6 eV).

A second XUV pulse at the same wavelength then probes the cluster to test if the channel is actually saturated. If enhanced photo-ionization mechanisms contribute significantly to the ionization of the cluster, then the second pulse will create a significant change in the ionization of the cluster [5,9]. Otherwise, the second XUV pulse will not interact with the cluster (due to the saturation of the single-photon ionization channel) and will leave it largely unchanged. Theis could begin to determine why barrier lowering has not yet been observed at the LCLS [17,20].

This work was recently published titled, “Induced transparency in the XUV: a pump-probe test of laser-cluster interactions” in the Journal of Physics Communications. In it, we show how our model differs significantly from the dominant laser-cluster model and show how experimental signals would be different and thus the proposed experiment could differentiate the models which would tell us about the underlying ionization mechanisms.

This work found a new state of matter in the XUV regime, the induced transparency state, where the system can no longer communicate with the laser. Additionally, due to nature of our model, we can predict that this is a common phenomena in intense XUV-matter interactions and will happen for any dense material at a range of wavelengths.

Track 2: Investigating intense laser-cluster interactions in the X-ray regime

Motivation

Many interesting and useful phenomena have been discovered in the infrared regime of laser-cluster interactions [10-13]. The new generation of free-electron lasers opens the door to unexplored regimes of laser-cluster interactions beyond even the extreme ultraviolet and well into hard X-rays. Further, new experimental techniques seek to probe the cluster's ultrafast dynamics directly [20-22]. At the LCLS this has been pushed to the most pristine situation where experiments can now be conducted on single clusters with an image being the probe [17]! Thus, as with previous experiments, the disintegration fragments are measured and carry information about the breakup, but at the LCLS an image is also taken giving direct information from within the cluster during the ultrafast process. The level of detail now available from experiments will allow us to build detailed models of the laser-cluster interaction in the X-ray regime. Moreover, we can use the LCLS as a probe to image the ultrafast processes generated by laser-cluster interactions from other regimes by changing the pump pulse. We can thus create the conditions for clusters to generate neutrons as in reference [13], for instance, then probe the cluster during this process to determine the specifics of the mechanism(s) as never before.

The imaging pulse, while a probe, is still disturbing to the system causing an immense amount of ionization. Most of the ionization is not from the initial single-photon ionization but from the subsequent Auger decay ionization. These secondary electrons will flood the cluster during imaging. Understanding their effects will allow us to separate out the effects of the pump from the probe pulse. Further, the LCLS imaging pulses on their own will explore a new regime of laser-cluster interaction in the X-ray regime where new phenomena and collective effects maybe just waiting to be discovered.

Mysteries have already begun to surface at the LCLS. Recent experiments seem to hit an ionization barrier at Xe^{26+}. Models which proposed a lower photo-ionization barrier due to the plasma [5,9,23] should enable higher charge states, yet none have been reported [17,20].

Projects

Laser-cluster modeling in the X-ray regime

Question: Can an atomistic model of laser-cluster interactions explain the results of recent Linear Coherent Light Source (LCLS) experiments?

This project extended our model of laser-cluster interactions to the hard-X-ray regime. Currently our model does include Auger decay for Xe at the so-called “4d giant resonance” with the correct timing delays (lifetimes) and ratios (single vs double ionization) all taken from experimental data. Our model has be augmented to allow for the on-the-fly calculation of photoionization, auger decay cross-sections and branching ratios using XATOM. This was incorporated using the Year 4 extension and now allows us to model not only more regimes in terms of wavelengths, but also any element in atomic form. Results using this new upgraded model are forthcoming.

Heterogeneous clusters and charge transfer

Question: How is charge transferred from one element to another in core-shell systems?

Experiments have recently been performed with mixed clusters consisting of a core of xenon and an outer shell of argon [24]. The clusters were irradiated with an intense X-ray pulse at the so-called “4d giant resonance” of xenon around 90 eV. At the resonance frequency, xenon is around 10 times more likely to photo-ionize from the inner-shell 4d state than from the 5p valence shell. Subsequent to the inner-shell photo-ionization is the Auger decay which ionizes another 1-2 electrons. When xenon gas was irradiated by the same laser pulse, charge states up to Xe^{9+} are detected with virtually no Xe^{1+}. When a cluster of xenon is irradiated by a similar pulse charge states up to Xe^{9+} are still detected but with a large Xe^{1+} signal. In the heterogeneous cluster of xenon and argon highly-charged argon was detected but a small Xe^{1+} signal was the only charge state of xenon detected. The charge must have transferred from the xenon inner-core to the argon outer core through a combination of collisional ionization and recombination.

We have modeled this experiment and *obtained excellent agreement with the experimental signals*. Additionally, *we have found a general empirical law for how the average charge state of the inner cluster changes with respect to the amount of outer layers*. This is almost ready for publication, we are still working on a theoretical explanation of the empirical law before publishing.

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Results from Subaward at the University of Ottawa

by Lora Ramunno

Mechanism of Laser Induced Filamentation in dielectrics

Laser filamentation in transparent material has a wide range of applications, from three dimensional manufacturing to biological technologies. Various experimental results showed that femtosecond laser pulse filamentation in fused silica strongly depends on laser focusing conditions. However, the physical mechanism governing each regime has not been fully understood. For the first time, single and multiple re-focusing of the laser pulse in interaction of femtosecond laser pulse with fused silica, and consequent single and multiple damage zones (filaments) have been observed in our extensive three-dimensional, high resolution FDTD (finite-difference time-domain) simulations. We show that Kerr nonlinearity plays a crucial role in loose laser focusing regime, while it is not an important factor in tight laser focusing regime, where geometrical focusing becomes important. Our simulation results agree well with existing experimental findings. In addition, the improved analytical model prediction gives a reasonable estimate of the shift from Kerr nonlinearity regime to linear geometrical focusing regime.

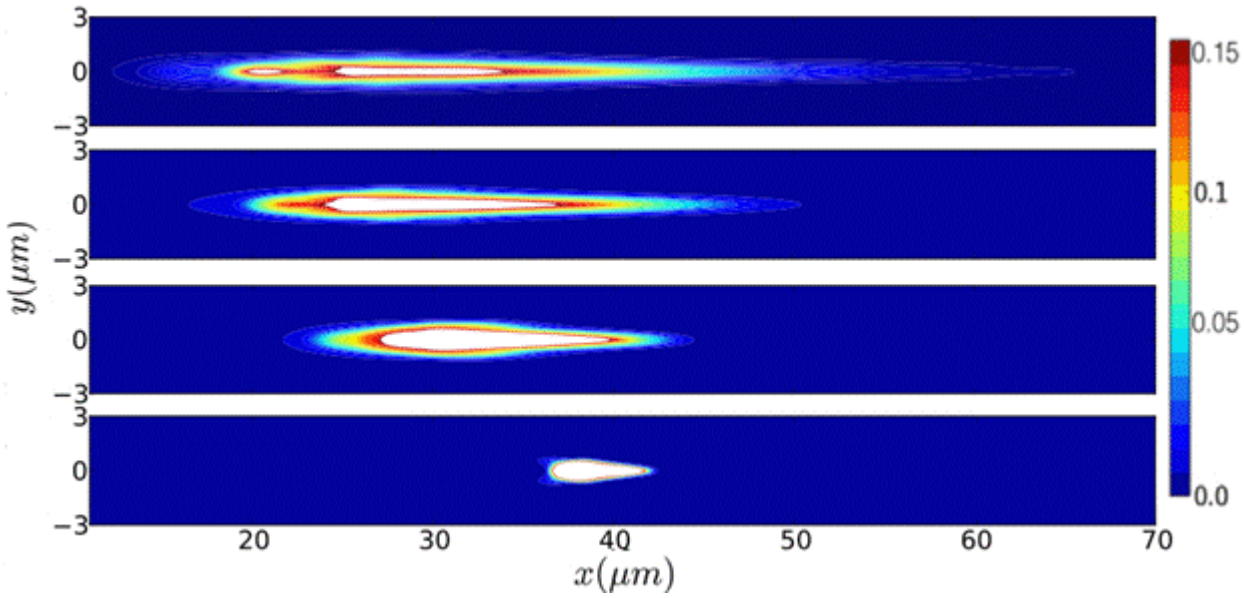


Fig. 1. Contour plots of electron density from top to bottom for w_0 = 2., 1.5, 1., 0.69 μ*m* for fixed energy, showing filament formation in different focusing regimes.

Mechanism of Void Array Formation in PDMS

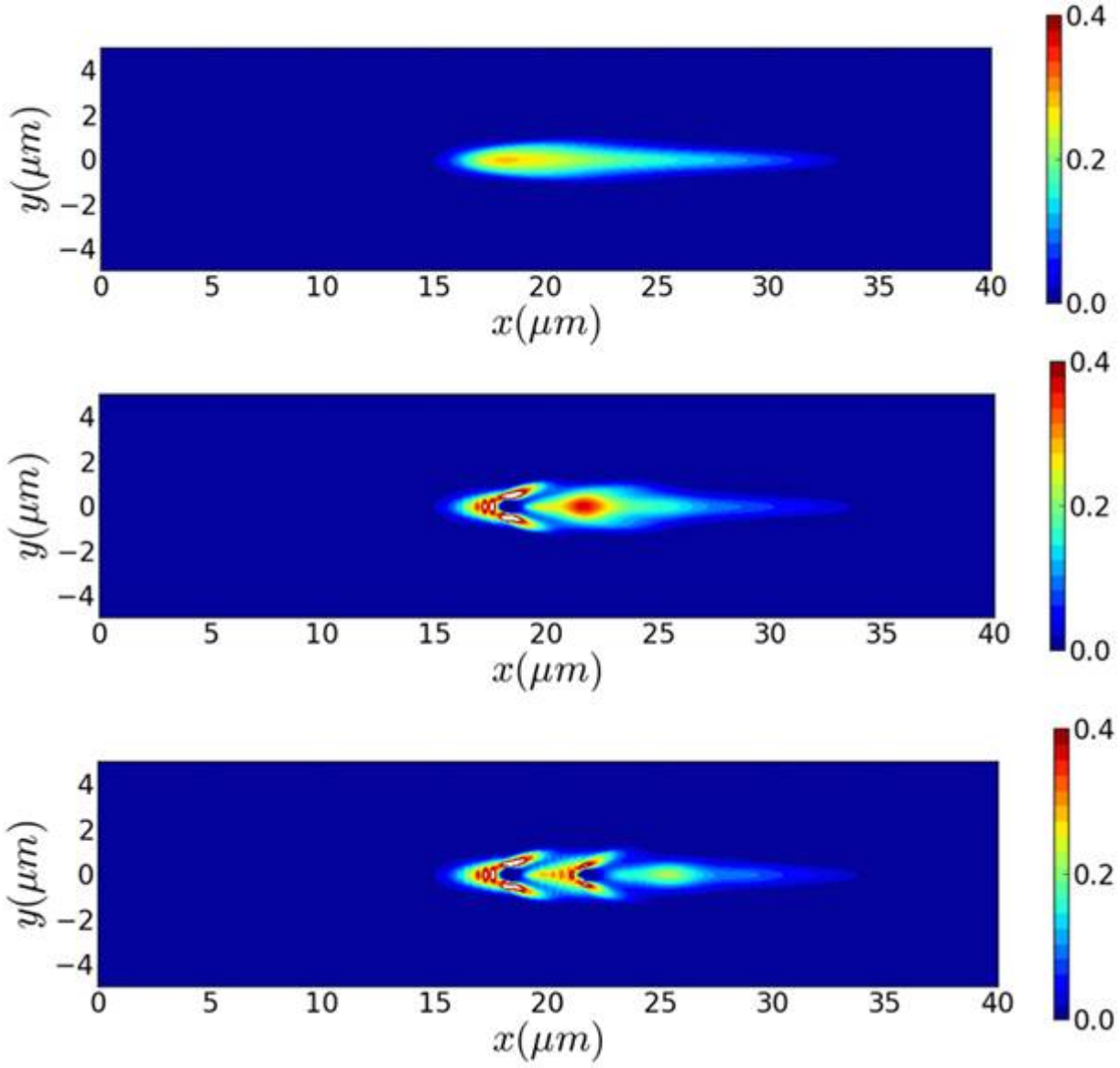
The void formation in dielectrics is a result of micro-explosions. The material in fact is pushed sideways from the laser focus resulting in void formation. However void array formation is the result of multi-laser irradiation. Experimental results show that increasing the laser energy in single pulse interaction results in elongated single void and

Fig. 2 Contour plot of final electron density normalized to ncr in a) bulk of PDMS b) with one pre-recorded void located at x = 18.4 μmc) with two pre-recorded voids located at x = 18.4, 22.3 μm.

not a void array. The mechanism of void array formation is still unclear. We propose a new method that clearly demonstrates the void array formation and confirms the experimental results by using three dimensional FDTD simulations. First we study the interaction of a single laser pulse with PDMS. Then we model the first damage structure (void) as pre-recorded into the medium and show the generation of the second void as a result of field enhancement in front of the first void. To finish we model the two generated voids as pre-recorded in the medium and show the third void generation in interaction of the third laser pulse with medium. Our method shows that this can continue and successive voids will be generated as a result of multi-laser interaction with material.

Near-infrared femtosecond laser machining initiated by ultraviolet multi-photon ionization

Laser micro-machining has been vastly studies for various applications. The nonlinear interaction between femtosecond laser pulse and transparent media such as fused silica leads to localized damage structures. Nanoscale structures can be generated by pulse



energy control and other methods. Alternatively, using ultraviolet (UV) pulses can control the size of the damage structure at nanometer scale. It is known that the laser pulses with shorter wavelength have larger cross section which can excite free electrons from valance band to the conduction band by photoionization. Therefore, UV lasers can be used to

generate seed electrons and a long wavelength pulse to create electron avalanche. The energy of UV pulse is kept below the damage threshold, so that the damage can only form within the seeding volume which is identified by UV pulse. This method propose a possible UV laser nano-machining. In this paper we focus on multi-photon and possible avalanche processes by a UV and near infrared pulses to study the mechanism of damage threshold and control of the size of the damage structure defined by UV pulses. UV pulses are more efficient in multi-photon ionization than NIR pulses. The band gap of fused silica is 9eV and only two photon is needed for 266 nm pulses to generate electrons from valance band to conduction band. While for 800 nm pulses, this number is 6. After this ionization process the electrons will be self-trapped to form exotics (STE) in 150 f s which has a lifetime of 30 − 300 ps. In dielectrics the nonlinear ionization rate strongly depends on the band gap. Excited states of atoms and molecules ionize more easily than their ground states because the ionization potential is decreased. Similarly we expect STE states in dielectrics to ionize easier than the unexcited bulks. These STE generated by UV pulse can be again excited to become free electrons. Therefore the process is as follows: The seed laser pulse (266 nm) generates the excitons (STE) which is localized, then the second pulse (800 nm) deposits energy in the localized region without modifying the surrounding medium. This enables making spatial resolution. The energy of UV pulse is kept below the damage threshold, so that the damage can only form within the seeding volume which is identified by UV pulse. This method propose a possible UV laser nano-machining. In this project, we focused on multi-photon and possible avalanche processes by a UV and near infrared pulses to study the mechanism of damage threshold and control of the size of the damage structure defined by UV pulses. we use a two-step method to study the UV/IR interaction with fused silica. In the first step, the near UV laser pulse with wave length of 266 nm interacts with fused silica. The first laser pulse generates self-trapped excitons which have lower band gap energy as of bulk of fused silica. After this ionization process the electrons will be self-trapped to form exotics (STE) which has a lifetime of 30−300 ps. The second step is to model the exciton population in fused silica and shoot 800 nm laser pulse. It should be noted that in the second step, the build up electrons from excited states (excitons) found in the first step act as seed electrons. Therefore we adapted the shape of the carrier density distribution from UV interaction with silica and implemented it in the medium at the same location observed previously in the simulation domain. The adapted exciton distribution will have a lower ionization threshold. The pre-generated excitons can become free electron via four photon absorption as discusses in literature. The rest of the medium is six photon absorption for 800 nm. We modeled the excitons as an oval layer with 4 photons. A

sample result (second step) of my high resolution FDTD simulation is shown in Fig. 3. The damage area is localized to 4- photon absorption volume. The electron density in the rest of the medium is much lower than the damage threshold ($0.15n_{cr}$, where n_{cr} is the critical electron density). We performed simulations with bulk of fused silica (without excitons) and no damage area appeared, showing that the localization of the damage area is due to excitons. We also found out that the Kerr nonlinearity does not play an important role in the interaction (due to low laser intensity $3.7e13W/cm^2$). Also avalanche ionization does not play a role for 60 fs laser pulse that we used.

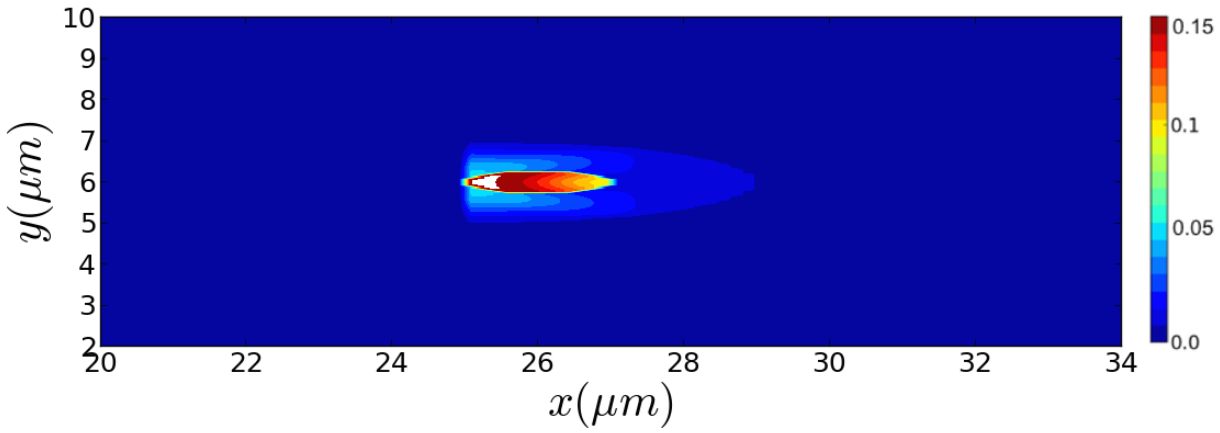


Fig. 3 electron density distribution. The surface of the silica is located at x=25 microns.