LETTER

Induced transparency in the XUV: a pump-probe test of laser-cluster interactions

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Abstract

An experiment is proposed to distinguish between different laser-cluster atomistic models and their predictions. The induced transparency of rare-gas clusters, post-interaction with an extreme ultraviolet (XUV) pump-pulse, is predicted by using an atomistic hybrid quantum–classical molecular dynamics model. We find there is an intensity range for which an XUV probe-pulse has no lasting effect on the average charge state of a cluster after being saturated by an XUV pump-pulse: the cluster is transparent to the probe-pulse. Multiple complete experimental signals are calculated which include the effect of the pulse’s spatial distribution as well as the cluster size distribution. The calculated experimental signals and trends are also accomplished with the addition of an ionization potential lowering model that results in effectively removing the induced transparency effect. Thus, the proposed experiment is expected to either find the new phenomenon of induced transparency in clusters or give strong evidence for the existence of the enhanced ionization phenomenon, ionization potential lowering, in nanosplasmas.

1. Introduction

Ultra-intense laser pulses interacting with matter are more straightforward in the extreme ultraviolet (XUV) regime compared with the infrared regime as the interaction occurs primarily through photoionization. When a nanoscopic, dense arrangement of matter (cluster) is irradiated, secondary ionization events, such as collisional ionization, take place. Clusters have near solid density but their inter-cluster distance is so large that clusters do not interact with each other. Thus, they bridge the gap between the gas and solid phases of matter, keeping the many-body effects of dense matter but removing any outside energy dissipation channels. Additionally, clusters are often used to compare theoretical models with experimental results due to the relative ease of modeling the system and ability to change the behavior of the clusters by altering their size.

Experimental and theoretical laser-cluster interaction studies in the XUV regime are simpler to interpret than at other wavelengths, and it is an ideal regime to further test detailed atomistic models of laser-cluster interactions [1–4]. At longer wavelengths, even low intensity pulses have efficient processes to transfer energy from the pulse to the free electrons, heating the electron plasma (termed inverse bremsstrahlung heating, IBH) along the axis of the laser’s polarization [5–7]. At shorter wavelengths, photoionization occurs primarily from the inner shell electrons and leads to subsequent Auger ionization [8–12]. Thus, XUV pulses—which through photoionization access only valence shell electrons and where IBH is negligible for intensities <1016 W/cm2—present the ideal regime for experiments to probe the degree to which the ionization potential may be modified by the plasma environment [13–18].

As multiple models of laser-cluster interaction exist, new experiments are needed to allow the community to distinguish between the different models [16] as their founding assumptions are different. In this work, we report on the finding that the ionization in XUV-cluster interaction can become saturated when a gas-like...
ionization model is used for photoionization. Almost no additional ionization takes place when further irradiation of the cluster occurs. This occurs when a cluster is irradiated with an XUV pulse above a saturation intensity. A subsequent probe-pulse irradiating the cluster leaves no net effect on the ionization or total energy; thus the cluster is transparent to the probe-pulse. In contrast, repeating our calculations with a plasma-like model [14, 15], we found no such transparency for small clusters. Thus, an experimental search for induced transparency will be able to distinguish whether any enhanced photoionization mechanism plays a strong role (no transparency found) or not (transparency is found) in this parameter space. Experimental validation or invalidation of either model would be an important step forward in understanding light–matter interactions in nanoplasmas. A similar, shorter lived, phenomenon was observed by Nagler et al [19] using solid aluminum. The sample was irradiated by such a high intensity that the single electron in the accessible valence band became depleted. Further primary ionization for the valence band was energetically disallowed by less than 1 eV resulting in the sample becoming mostly transparent until the valence band was repopulated primarily by Auger decay shortly after the pulse. The main difference in the present work is that one model predicts no transparency and the other predicts complete long time (on the order of the plasma recombination time) transparency due to the depletion being of valence shell electrons (no subsequent Auger decay).

Atomistic cluster models in and beyond the vacuum ultraviolet (\(\lambda \leq 100\) nm) to date include single and multi-photon ionization as well as collisional ionization processes based on the photoionization rates and ionization potentials of single atoms. Additional processes have been included in order to explain higher charge states found in experimental results. The most fundamental is collisional excitation which has been incorporated as augmented collisional ionization [20]. Collisional excitation and then ionizing from an excited state is a well-known atomic gas-phase phenomenon and thus is expected to play a major role in shaping the laser-cluster dynamics. Photoexcitation is excluded from most models since the energy and angular momentum conservation laws make photoexcitation (single photon driven atomic bound state to bound state transition) negligible, unless the laser is tuned to a resonance [21]. Further mechanisms come from collective effects known to occur in large, dense plasmas. These involve the screening of an electron bound to an ion by neighboring ions and quasifree electrons. Electron screening is often implemented using the Debye screening model [22–24]. The ion screening model has been implemented using the ion-sphere or muffin-tin model [25, 26], while the ionization potential lowering (IPL) model has been recently implemented for rare-gas clusters [14, 15, 27]. These plasma mechanisms attempt to distort the atomic potentials and allow ionizations to occur which would otherwise be energetically impossible for photoionization. Work which only included augmented collisional ionization using the local ionization threshold model [20, 28], which treats the cluster potential as a constant potential perturbation, has successfully reproduced the results of multiple experiments [29], including experiments where Auger ionization is dominant [30]. Thus, the question arises: are the plasma effects on ionization, such as ionization potential lowering, negligible in small clusters? If so, there would be a fundamental behavioral difference between nanoplasmas and other plasmas.

In the following, we shall refer to a model which removes the effects of the plasma prior to ionization as a gas-like ionization model (GIM). Foundationally, GIMs such as the local ionization threshold model, are predicated on the principle of superposition, reducing the complex dynamics of the laser-cluster interaction to a series of individual atomic processes [31]. The plasma environment then modifies the energetics of the ionized electron, post-ionization. A plasma-ionization model (PIM) is foundationally predicated on a modification of the atomic processes due to the plasma prior to ionization. They modify the ionization process due to the surrounding plasma by changing the cross-section and/or ionization potential from its gas phase values. PIMs, such as ionization potential lowering (IPL), must assume that at least one excited final state exists at the correct energy with the correct angular quantum numbers. For instance, photoionizing the valence 3s electron from an Ar\(^{+1}\) by a 33 nm photon in the PIM model requires there to be a p-state (\(\Delta l = 1\)) with an energy of around 86 eV (\(E_{IP} - E_{l} = 124 \text{ eV} - 38 \text{ eV}\). All PIMs in the single-photon ionization regime (from the VUV to x-ray), to date, have also assumed these barrier modifications only occur in photoionization. It is unclear why this should be the case since collisional processes can transfer arbitrary amounts of energy and angular momentum.

2. Methods

Our implementation of a gas-like ionization model (GIM) is a hybrid approach wherein the particles are treated as classical charge distributions whose motion is solved by molecular dynamics. The ionization rates are determined from a mix of experimental (when available) and theoretical cross-sections in the gas phase [20]. During ionization, the perturbation on the ions due to the cluster environment (the nanoplasma) has been shown to be well represented by our local ionization threshold model [28], which maintains the use of atomic ionization potentials. Using the local ionization threshold model, we include single- and multi-photon ionization, collisional ionization, augmented collisional ionization [20], and many-body recombination [30].
The photoionization cross-sections used for argon are \( \sigma^{(1)} = 5.0 \times 10^{-18} \text{cm}^2 \) for single-photon ionization [32] and \( \sigma^{(2)} = 10^{-30} \text{cm}^4/\text{s} \) (taken as an upper limit from reference [33]) for two-photon ionization. The larger the value of \( \sigma^{(2)} \), the smaller the range of intensities in which induced saturation will occur. Thus taking an upper limit gives a conservative estimate of the saturation effect.

Our plasma-ionization model (PIM) adds the ionization potential lowering (IPL) mechanism to the gas-like ionization model (GIM) following the over-barrier inner-ionization of reference [15]. The new ionization potential is given by \( I_p^* = I_p - \Delta_{\text{env}} \) where \( I_p \) is the ionization potential in gas and \( \Delta_{\text{env}} \) is the environmental shift due to the plasma [17]. The environmental shift is given by \( \Delta_{\text{env}} = U_{\text{ion}} - U_{\text{barrier}} \) where

\[
U_i = \frac{\sum_{i=1}^{N} k_i q_2}{|r_i - r_j|} \tag{1}
\]

\( N \) is the number of particles excluding the ionizing ion, \( k \) is Coulomb’s constant, \( r_i \) is the location of target ion with index \( j \) or the location of the barrier in the case of \( U_{\text{barrier}} \), and \( r_j \) is the location of the particles. The location of the barrier is determined by the stationary point in the potential between the ionizing ion and its nearest neighboring ion. \( I_p^* \) is artificially given a lower bound of 0.1 eV for situations where the \( I_p^* < 0 \).

The cross-sections for the PIM photoionization is determined by adding the environmental ionization potential shift, \( \Delta_{\text{env}} \), to the actual photon energy, \( E_\gamma \), to obtain the new photon energy, \( E_\gamma^* = \gamma + \Delta_{\text{env}} \). This gives an underestimate of the cross-section.

Results for this PIM (using our gas-ionization model and adding the over-barrier inner-ionization IPL mechanism) are a lower bound compared with other photoionization-only IPL models such as reference [14]. Additionally, extending IPL to collisional processes is expected to have an even more dramatic increase in the overall ionization amount [25], and, thus, our results are the most conservative estimates for differences between the GIM and PIM.

Any plasma-ionization model (PIM) which is dependent on the density of the plasma will not find a region of induced transparency. This would imply that an ionization potential lowering (IPL) mechanism, or some other PIM, was needed even for nanoplasmas. On the contrary, experimental verification of induced transparency would validate the GIM in the nanoplasma regime and place limits on the strength of any density-dependent PIM. Further, this would show a significant difference between continuum plasmas and nanoplasmas.

In this paper, the average ion charge state (AICS) is used as a measure of the overall ionization of a cluster since atoms are not detected and the AICS can be obtained by fitting the time-of-flight (TOF) signal.

We propose using 33 nm XUV pulses. The saturation will occur because a 33 nm photon is insufficient to further ionize an \( \text{Ar}^{2+} \) ion. However, PIMs, such as IPL, decrease the ionization potential and thus the pulse can continue to ionize the ions to much higher charge states resulting in no induced transparency region. Using a pump–probe setup, the existence of the induced transparency can be tested and give evidence for either model.

The methodology to distinguish the two models is as follows. A pump-pulse above the saturation intensity irradiates the cluster. The probe-pulse follows 25 fs later. The GIM predicts that the pump-pulse saturated the cluster and so the probe-pulse has no effect on the AICS. The PIM predicts that the AICS increases with the intensity of the probe-pulse due to PIMs allowing for further photoionization beyond the \( \text{Ar}^{2+} \). A delay of 25 fs ensures that the probing of the nanoplasma occurs while it is still dense, minimizes the pulse overlap, and occurs far from the time recombination becomes dominant.

This methodology is complementary to previous proposals in larger clusters [3]. Additionally, it is the first to include the role of collisional excitation which is known to play a dominant role in the XUV [34].

### 3. Results

To show the induced transparency effect in our gas-like ionization model (GIM), we solved the interaction of argon clusters (\( N = 147 \)) irradiated by two XUV pulses at \( \lambda = 33 \text{ nm} \) (37.6 eV) with a 25 fs delay. Both pulses have a full-width-at-half-maximum of 10 fs to minimize the pulse overlap while irradiating the cluster when the density is still high (enhancing the likelihood of ionization potential lowering or IPL effects which depend on the plasma density).

An intensity scan is first performed by calculating the average ion charge state (AICS) resulting from a pump-pulse at each peak intensity. The AICS of the cluster after disintegration (when the cluster field is much less than the time-of-flight field) is used as a measure of the overall ionization of the cluster, and the cluster is first considered to be at the focus of the laser pulse. The average is taken over all ions; ions containing classically bound electrons have their charges decreased accordingly.

The dotted cyan curve in figure 1 shows the AICS versus pump intensity when an \( \text{Ar}_{147} \) cluster is irradiated by a pump-pulse. As pump intensity is increased from \( 10^{13} \text{ W/cm}^2 \) to \( 10^{14} \text{ W/cm}^2 \), the AICS starts to increase very gradually. At around \( 10^{14} \text{ W/cm}^2 \) the AICS increases dramatically until a saturation at an intensity of about
10^{15} \text{ W/cm}^2 where the AICS is about 3.1. The start of the plateau in AICS is what we term the ‘saturation intensity’. Further increases of the pump intensity beyond 10^{15} \text{ W/cm}^2 only marginally increases the AICS until after the AICS plateau, around 10^{16} \text{ W/cm}^2. This plateau is the region of intensity where induced transparency takes place as photoionization in the GIM has saturated. The small increase in the AICS in the plateau region is due to multiphoton ionization and IBH. Even at $I = 10^{17} \text{ W/cm}^2$, more than 50% of the ionization is due to collisional ionization, almost exclusively through augmented collisional ionization. However, at the saturation intensity ($10^{17} \text{ W/cm}^2$), augmented collisional ionization accounts for well above 90% of all ionizations, while multiphoton ionization only accounts for a few percent. Thus, the primary ionization mechanism remains collisional even at the highest intensities. The saturation intensity as a function of cluster size is shown in the supplemental materials (online at stacks.iop.org/JPCO/2/051002/mmedia). It shows a decrease in the saturation intensity as the cluster size increases.

The plasma-ionization model (PIM) result follows the GIM result until the saturation intensity, shown as the long-dashed blue curve in figure 1. As the intensity of the pump-pulse increases beyond the saturation intensity, the AICS continues to increase at an almost constant rate. Thus, as expected, the PIM allows the pump-pulse to continue to ionize the cluster well beyond the GIM. Unlike the GIM, photoionization accounts for over 60% of the ionization for all charge states below Ar^{3+} for the PIM at the peak-only pump intensity of $I = 10^{17} \text{ W/cm}^2$.

To demonstrate induced transparency, a pump-probe setup is modeled. The pump-pulse is fixed at $2.5 \times 10^{15} \text{ W/cm}^2$, just above the saturation intensity. The intensity of the subsequent probe-pulse (25 fs later) is scanned from $10^{13} \text{ W/cm}^2$ to $10^{17} \text{ W/cm}^2$. The dashed-dotted red curve in figure 1 shows average ion charge state (AICS) versus probe intensity for the GIM. The AICS begins and remains saturated until the intensity of the probe-pulse exceeds about $10^{16} \text{ W/cm}^2$. This is when the probe-pulse reaches high enough intensity for IBH to become significant [7]. Below this intensity, the additional probe-pulse does not meaningfully increase the AICS from what it was after the pump-pulse; this is the basis for terming the phenomenon induced transparency, since it is as if the cluster were effectively transparent to the probe-pulse. With the same pump-probe setup, the PIM gives significantly different results, shown as the solid green curve in figure 1. Even at the lowest intensities, the probe-pulse causes further ionization of the cluster due to the photoionization not being limited to the Ar^{3+} ion. The additional IPL-ionized electrons add their excess energy to the electron plasma in what is termed ionization heating [35].

Why is there a plateau in the AICS for the gas-like ionization model (GIM)? An analysis of the charge state distribution versus time shows that the irradiation of the cluster by the pump-pulse at the saturation intensity ionizes all possible targets via photoionization and collisional ionization. Thus, during the pulse there are no more targets to further photoionize. This is the high intensity limit to the previously observed collisionally reduced photoabsorption where clusters absorb less photons due to fast collisional ionization depleting ionizable targets [29]. The result is the same saturated AICS, both with and without the probe-pulse. The plasma-ionization model (PIM) which includes the IPL mechanism does not give rise to a plateau in the AICS, but instead the AICS continues to rise. This is expected since the mechanism by which induced transparency occurs, the saturation of the photoionization channel, is removed [14, 17, 35].
We now consider calculations that correspond more directly to what an experiment would detect in the absence of pulse clipping (removing the lower intensity spatial wings of the pulse). In any cluster beam, there is a log-normal distribution of cluster sizes and the laser pulse has a spatial extent. Most clusters are not irradiated by the peak fluence. A complete experimental signal includes the integration over the size distribution of the clusters as well as over the spatial extent (different intensities) of the laser pulse(s). The time-of-flight (TOF) signal is generated, using the methodology from reference [30], by integrating over four cluster sizes ($N = 55, 147, 309$ and 561) in a log-normal distribution centered at $N = 150$ and eighteen intensities assuming a Gaussian spatial pulse shape (see supplemental figures 1, 2 and 3 is available online at stacks.iop.org/JPCO/2/051002/mmedia for some of the full TOF signals and integrated charge state distributions). Each TOF signal is then integrated to obtain the integrated average ion charge state (AICS) signal. The integrated AICS is then plotted as a function of the peak pump (probe) intensity for the pump-only (pump-probe) setup in figure 2. In the pump-probe setup, the pump-pulse is fixed at $I = 2.5 \times 10^{15}$ W/cm$^2$ (see supplemental figure 4 for the effect of cluster size on the saturation intensity). The pump-only AICS for the GIM (dotted cyan) and PIM (long-dashed blue) follow similar trends as a function of the pump-pulse’s intensity with the PIM signal increasing almost linearly and the GIM signal giving a slowly rising plateau. The AICS values are lower due to the inclusion of lower intensity regions of the pulse(s). The GIM signal (dot-dashed red), as a function of the probe’s intensity with the pump-pulse fixed at $2.5 \times 10^{15}$ W/cm$^2$, shows the same trend as at the peak of the pulse remaining unchanged until about $2.5 \times 10^{15}$ W/cm$^2$ where it rises slowly along with the pump-only GIM signal. The most significant change due to the integrations (intensity profile and log-normal size distribution) is in the PIM pump-probe (solid green) signal which also has the pump-pulse’s intensity fixed at $2.5 \times 10^{15}$ W/cm$^2$. In the pump-probe setup, clusters were assumed to interact with the same intensity region of both pulses, i.e., the pulses were assumed to be spatially identical and focused at the same location.

The larger-sized cluster’s disintegration is slower and thus the cluster retains a higher density during the probe-pulse. This allows for additional ionization in the PIM and explains why the pump-only AICS is higher than the pump-probe signal. Thus, despite the fact that induced transparency only occurs for the highest intensities, its effect on the AICS is detectable in the full experimental signal.

Plasma-ionization model (PIM) results are more sensitive to the time delay. The sensitivity of the ionization potential lowering (IPL) mechanism arises from the rapid change of the barrier with the inter-ionic spacing, $\Delta_{env}$, is inversely proportional to the inter-ionic spacing and thus a change in the probe’s delay can significantly alter the average ion charge state (AICS). Changing the time delay results in a significant change in the peak AICS (not shown) and the fully integrated AICS (figure 3). A time delay of $10$ fs results in a small increase in the AICS (long dot-dashed black), while a $50$ fs time delay decreases the AICS significantly (very long dashed magenta). Above $I = 5 \times 10^{16}$ W/cm$^2$ there is almost no additional enhancement due to the pump-pulse (seen by the pump-probe-pulse result being roughly equal to the pump-only result). The full charge state distribution for the different time-delays is shown in the supplemental material (supplementary figures 5 and 6, available online at stacks.iop.org/JPCO/2/051002/mmedia), clearly showing the decrease in the population of the highest charge states as the time-delay decreases in the PIM.

A time delay up to $50$ fs only alters the gas-like ionization model’s AICS by less than $5\%$ from the results presented in figure 2 (not shown), and that change is due to the contribution of the unsaturated clusters. These clusters are not irradiated by the peak of the pulse and are thus not saturated. They are additionally ionized by the
probe-pulse. The increased time delay lowers the AICS since the ionized electrons are more likely to escape the cluster than have an inelastic collision due to the decreased cluster density.

An AICS measurement of negligible change, as a function of delay time, would provide strong evidence for the gas ionization model (GIM) and the existence of the phenomenon of induced transparency in the XUV in small clusters (as was found in solids in the x-ray regime by Nagler et al [19]). On the contrary, a significant change in the AICS as a function of the time delay would be clear evidence for a plasma ionization model (PIM), even for small clusters. This would be unexpected since the effect was not detected in Nagler et al for a solid and the ionization potential lowering (IPL) mechanism’s contribution decreases as the density of the cluster decreases. This decrease is due to the cluster’s disintegration and the effect must tend to zero as the density becomes that of a gas since no such mechanism has been observed in gas [14, 16, 36–38]. Lastly it is worth noting that our PIM used the most conservative assumptions and thus our results represent a lower bound on the experimental PIM results.

4. Conclusion

In conclusion, we have shown that an atomic-ionization laser-cluster interaction model predicts that it is possible to induce and probe transparency in the XUV regime using a pump-pulse setup. This effect is insensitive to the time delay between the pulses, and thus insensitive to nanoplasma density. This is in contrast to what ionization potential lowering models would predict. Experimental verification would either uncover a new transient state of matter and make a distinction between continuum and nanoplasmas, or provide clear evidence for plasma ionization models such as ionization potential lowering in small rare-gas clusters. It would also provide the field with valuable data to refine its models of photoionization in laser-cluster interactions not only in the XUV, but for any wavelength where photoionization plays a major role.

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