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Interaction of clusters with intense, few-cycle, long wavelength fields

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OHIO STATE UNIVERSITY THE

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11/05/2015  
Final Report

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## Final Performance Report

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Subject: Final Performance Statement to Dr. Enrique Parra

Contract/Grant Title: Interaction of clusters with intense, few-cycle, long wavelength fields.

Contract/Grant #: FA9550-12-1-0047

Reporting Period: 15 March 2014 to 14 September 2015

Principal Investigators: Louis F. DiMauro and Pierre Agostini

Institution: The Ohio State University, Department of Physics, Columbus, OH 43210

Grant accomplishments: Over the grant period, the DiMauro/Agostini group has completed most of plans for the study of intense laser-cluster interactions. First, we have finished the construction of a time-of-flight (TOF) cluster beam machine (shown in Fig. 1), which is integrated into the existing Hettrick XUV spectrometer. This dual mode machine allows us to observe photons and electrons/ions, both of which convey essential and complementary information about laser-matter interaction. In addition, this machine is designed to mount both atom and cluster targets under identical experimental conditions so that any cluster effect can be explicitly distinguished from that of atom.

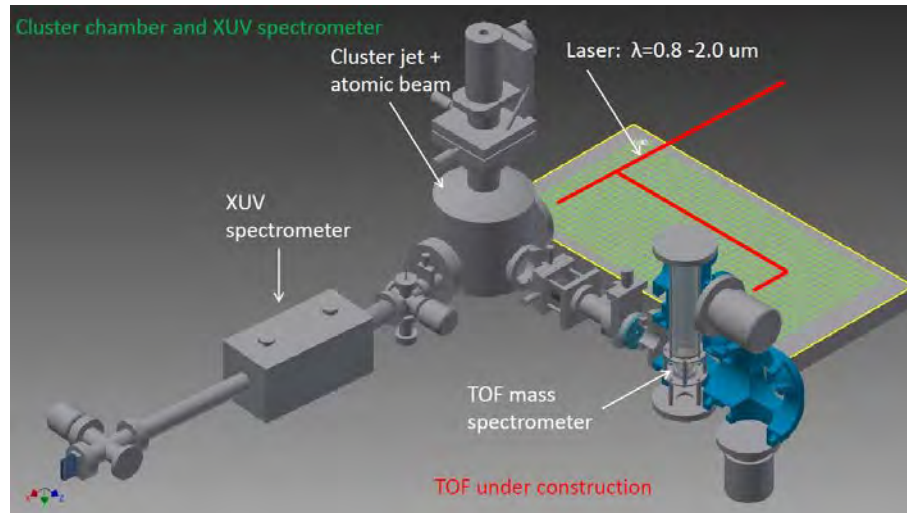


Figure 1: Schematic view of the OSU time-of-flight ion/electron cluster beam spectrometer.

From the photon spectrometer, we have carried out high-order harmonic generation (HHG) experiments. This experiment was intended to find how the cluster behaves in HHG process and clarify its utility as a potentially brighter HHG medium. We have found that van der Waals clusters (or noble-gas clusters) act more like ensembles of many atoms due to weakly delocalized electrons, and thus the main HHG process follows a 3 step quasi-classical model, which is widely accepted in atomic targets. The experimental evidences from our experiments include group delay measurements, ellipticity dependence of yield,

and cutoff energy- the results of those measurements appear to be quite similar to the atomic targets from the direct comparison studies.

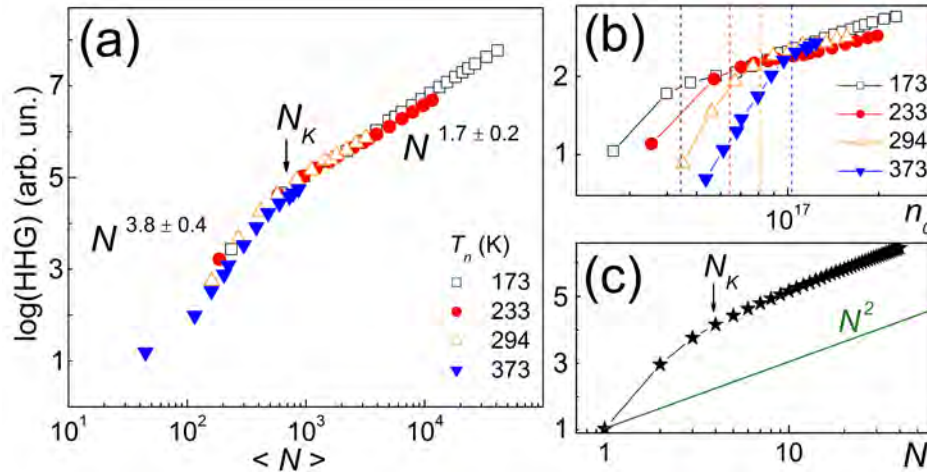


Figure 2: HHG yields in various temperature and pressures (a) against the cluster size,  $\langle N \rangle$ , and (b) atomic density,  $n_0$ . (c) Calculation of HHG yield as a function of the cluster size.

Nevertheless, the inherent delocalization of the electron wave function, albeit weak, is found to be the reason why the cluster produces higher HHG yields than the monomer counterpart. The partially delocalized wave function induces a larger dipole oscillation, in other words, higher yields, when the returning electron wave function interferes with that of the parent ion at recombination. The effect of the small delocalization of the wave function has been manifested in our 1-D calculation, which is based on Lewenstein model including the partially delocalized initial wave function. As we have observed in the experiments (Fig. 2[a]), the model produces a rapidly growing HHG yield as a function of the cluster size, faster than a quadratic growth in the monomer (Fig. 2[c]). However, this enhancement effect vanishes beyond a certain cluster size due to the weak delocalization. Practically, this size limit still to favor atomic gases as a table top XUV source, although the problem can be potentially overcome by producing a high density gas of small clusters.

From the TOF cluster beam machine, we have observed electron and ion emissions from the laser-cluster interaction in IR and mid-IR range. Our investigations have focused on the high energy ion emissions and the electron energy distribution in the various wavelength. First, we have observed that the laser heated cluster can store a significant amount of photon and kinetic energies before the cluster explodes and emits energetic particles during its relaxation process. Furthermore, the amount of heating energy varies with the wavelength of the driving fields. This wavelength dependent cluster heating is the first experimental observation to our best knowledge and will provide us with new and deep physical insights into the laser-matter interaction. The result on the cluster heating is shown in Fig. 3(a) where the ion energy against the wavelength is plotted. The ion energy is found to decrease with increasing wavelength from 0.75 to 2.0  $\mu\text{m}$ . This result can be interpreted by a resonant like heating mechanism, Inverse Bremsstrahlung (IB) collisions, in which the initially ionized electrons absorb photon energies through a collision with an ion, and then the hot electrons impact ionize the existing ions and neutrals, resulting in highly charged ions. This heating mechanism is, in principle, most efficient near the plasma

frequency, which is about 0.3  $\mu\text{m}$  in the wavelength, and it is rapidly suppressed as the driving wavelength is detuned.

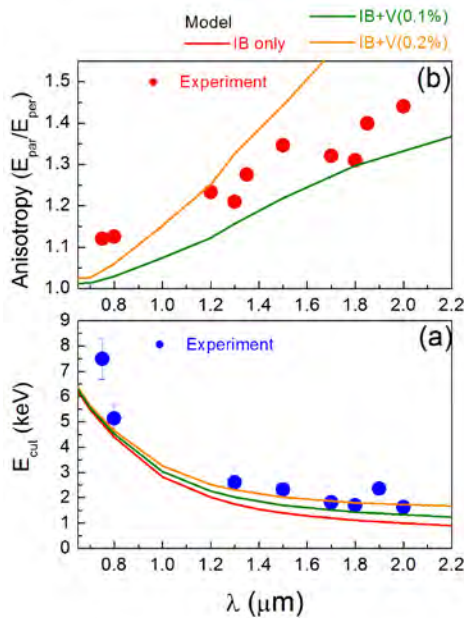


Figure 3: Wavelength dependent (a) ion energies and (b) anisotropies. The solid curves represent our model which consists of IB and vacuum heating mechanisms.

heating in IB. The other evidence is the measurement of the sensitivity of the ion energy to CP light, which is expected to be higher in a longer wavelength. The idea is that a larger quiver motion in a longer wavelength suppresses the vacuum heating more due to larger deflection of the electron trajectory in a CP light. We have observed that reduction of the ion energy by changing from LP to CP light appears to be more significant as the wavelength is increased.

We have concluded that the two heating mechanisms, IB and vacuum, do exist in cluster heating especially at mid-infrared wavelengths. IB heating is dominant, since it occurs over the whole volume, whereas the vacuum heating contributes only to the surface portion, which is relatively small. Our simple model, which combines those two heating processes, qualitatively reproduces the ion energy and anisotropy as shown in Fig. 3 (a) and (b), respectively.

We have also performed TOF experiments by observing electrons in which the laser intensity is moderate (below saturation intensity  $\sim 1 \times 10^{14} \text{ W/cm}^2$ ) so that we understand better what we have observed in the HHG experiment. It is found that the electron energy distribution resembles the classical curve in atomic targets, yet the detailed structure is quite different from the atomic case. Most notable features are broader angular distribution and higher cutoff energy compared to those of the atom. It seems that the electron trajectory and thus rescattering process are much more complicated in the cluster than in the atomic

Another heating mechanism that exists but is not clearly observed in the ion energy is Brunel-like vacuum heating. The vacuum heating arises due to the fact that electrons exposed to the vacuum are driven by the external field back and forth and convey the electron kinetic energy into the cluster when they return. The heating energy is expected to increase as a function of the wavelength, since the electron energy is basically ponderomotive energy. Although the ion energy was not sensitive enough to reveal the vacuum heating effect, we have observed evidence of it by other experiments- anisotropic ion emission and sensitivity to circularly polarized (CP) light as a function of the wavelength. From the ion measurement both in the parallel and perpendicular direction to the linear polarized (LP) laser, we have found that the anisotropies of the ion yield and energy progressively grow with increasing the wavelength. The result manifests the inherent preferential heating along the polarization direction in a field driven process, in contrast to the random

target. As the laser intensity is increased further beyond the saturation intensities, the electron spectrum completely loses the features of the atomic case. We will focus on the study of the electron energy distribution for the rest of the project.

In summary, we have accomplished the aim of the project on the laser-cluster interaction within the period of the grant. We have performed the first studies of laser-cluster interaction in the mid-infrared spectral region. We have finished the construction of TOF spectrometer cluster beam machine by which several intriguing phenomena have been observed. The results are in analysis and manuscript preparation stage. In addition, we have published the result from HHG in Physical Review Letters.

Archival publications during reporting period: “Size-Dependent High-order Harmonic Generation in Rare-Gas Clusters”, Phys. Rev. Lett. **113**, 263401 (2014).

Changes in research objectives: none

Change in AFOSR program manager: none

Extensions granted or milestones slipped, if any: Yes, the grant was given a 6-month no-cost extension with a new end date of September 14, 2015.

New discoveries, inventions or patent disclosures during this reporting period:  
The cluster (or plasma) heating mechanism is found to be wavelength dependent, which enables us to better understand the laser-matter interaction. (Article in preparation).

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<b>6. AUTHOR(S)</b> Louis F. DiMauro Pierre Agostini	<b>5d. PROJECT NUMBER</b>
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<b>9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)</b> USAF, AFRL DUNS 143574726 CODE: FA9550 AF Office of Scientific Research 875 North Randolph Street, RM 3112 Arlington VA 22203 Sharon B. Taylor 703-696-7340	<b>10. SPONSOR/MONITOR'S ACRONYM(S)</b> USAF, AFRL, AFOSR
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The goal of this proposal was to develop a more complete understanding of the fundamental physics associated with intense laser-cluster interactions. The physics is explored by tuning the wavelength of the intense laser in the mid-infrared spectral region (1-4.5 micron). Applications are in the perspective included the production of highly charged, energetic ions, x-ray photons or high harmonics light, and hence attosecond pulses and attophysics. The proposed investigations have provided new insights into the physics of nano-plasmas over this unexplored region of parameter space.

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**Grant/Contract Number**

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FA9550-12-1-0047

**Principal Investigator Name**

The full name of the principal investigator on the grant or contract.

Louis DiMauro

**Program Manager**

The AFOSR Program Manager currently assigned to the award

Dr. Enrique Parra

**Reporting Period Start Date**

03/15/2012

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10/14/2015

**Abstract**

The goal of this proposal was to develop a more complete understanding of the fundamental physics associated with intense laser-cluster interactions. The physics is explored by tuning the wavelength of the intense laser in the mid-infrared spectral region (1-4.5 micron). Applications are in the perspective included the production of highly charged, energetic ions, x-ray photons or high harmonics light, and hence attosecond pulses and attophysics. The proposed investigations have provided new insights into the physics of nano-plasmas over this unexplored region of parameter space.

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“Size-Dependent High-order Harmonic Generation in Rare-Gas Clusters”, Phys. Rev. Lett. 113, 263401 (2014)

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

None

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

None

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

Yes, the grant was given a 6-month no-cost extension with a new end date of September 14, 2015.

**AFOSR LRIR Number**

**LRIR Title**

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