Final Report
Quantum Phenomena in High Energy Density Plasmas

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Recent dramatic breakthroughs at JILA in the generation of coherent x-rays using the high harmonic generation (HHG) process have opened up many new areas of application, as well as demonstrating that, *after 25 years of investigation, we still do not know the limits of what might be physically possible using HHG*. One exciting discovery opens up a new and completely unanticipated possibility of implementing efficient (phase matched) HHG upconversion of deep-UV lasers in multiply-ionized plasmas, with potentially unprecedented conversion efficiency. HHG results from the extreme nonlinear response of matter to intense laser light: high harmonics are radiated as a result of a quantum coherent electron recollision process that occurs during laser field ionization of an atom. HHG represents the only route for generating coherent x-ray beams at wavelengths <6 nm on a tabletop, and the only way for generating coherent x-rays at high repetition rate for applications. To date, however, optimizing the conversion efficiency from laser to soft x-rays demanded a trade-off between two competing factors. The emission from each atom is largest for ultraviolet (UV) driving lasers, due to reduced quantum diffusion of the radiating electron wave function. However, macroscopic, phase-matched, buildup of bright HHG beams in the soft X-ray region favors mid-infrared (IR) lasers, to ensure that the emission from many atoms interferes constructively. In this case, HHG emerges as a broad supercontinuum (Fig. 1 (top)), ideal as a backlight for probing WDM.

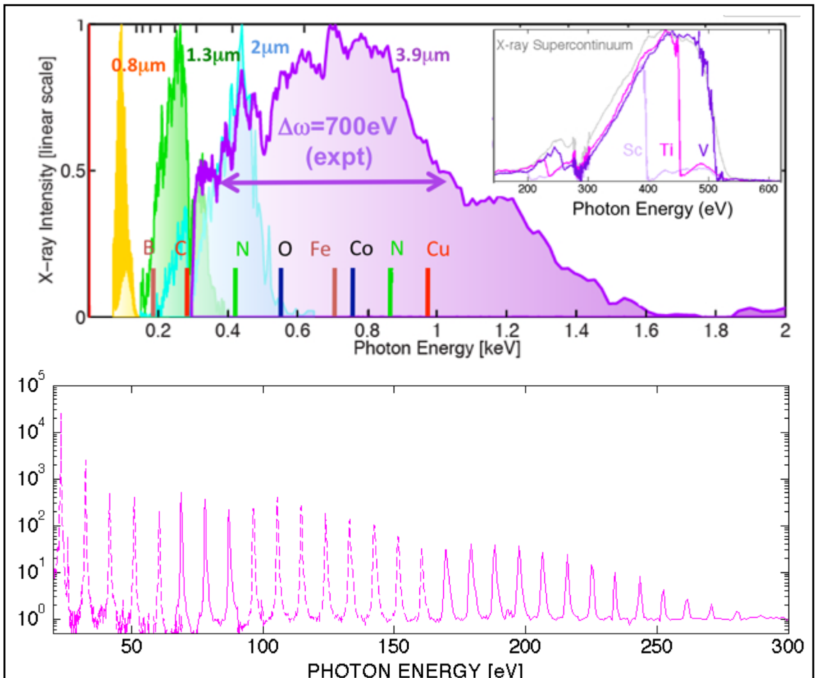

Fig. 1. Bright high harmonic generation in near neutral gases (top) and multiply ionized plasmas (bottom). (top) Bright coherent HHG X-ray super-continuum driven by mid-IR lasers, reaching photon energies > 1keV. The inset shows the first bright HHG in the water window at kHz rep rates, ideal as a backlight for probing WDM. (bottom) First bright HHG emission in plasmas (Ar^{5+}). Bright UV-driven HHG extends up to 280eV, with narrow line widths ideal for imaging WDM.

Under current support from this grant in work published in Science in 2015, we discovered a new regime of bright HHG in highly-ionized plasmas driven by intense UV lasers, that generates bright harmonics to photon energies >280eV (Fig. 1 bottom). This was an unexpected result – all prior theory predicted that phase matched HHG using UV laser wavelengths would be limited to <30 eV photon energies. This new regime is still not well understood; however, the experimental data shown in Fig. 1 (bottom) precipitated the realization that the approximations used in HHG phase matching models to date are well-satisfied ONLY when using mid-IR lasers to drive HHG from weakly ionized gases. In contrast, our experiemnts suggest that using UV driving lasers, the high linear and nonlinear UV refractive indices of large neutral atoms and ions may compensate for free electron plasma dispersion, *even in multiply ionized plasmas*. The resulting HHG from ions has estimated conversion efficiencies of 10^{-3} - 10^{-6} in the extreme ultraviolet (EUV) and soft X-ray regions – possibly higher than any other approach to date. Moreover, the well-separated narrowband HHG peaks produced by UV lasers are ideal for applications in imaging.

Highlights of this JILA OFES/NNSA research to date

1. Publications and Patents: Since 2009, we published *10 peer-reviewed papers and patents in top journals* including Nature, Physical Review Letters, Nature Photonics, PNAS etc.
2. Awards: Since 2009, students, postdocs and faculty received *16 national and international awards*, including NSF and DOE Graduate Fellowships, a Plasma Thesis Award, the ACS Zewail, APS Schawlow Prize, and election to the National Academy of Sciences.
3. 2012 and 2014 Student Internships at DOE NNSA Labs: During 2012, grad student and NSF Fellow *Dennis Gardner* worked with an LLNL team led by Mathias Frank, and participated in experiments at LCLS to investigate x-ray damage of thin membranes. Graduate student and DOE NNSA Stewardship Science Fellow *Ben Galloway* did a 12-week practicum at LLNL during 2014, working with Bradley Pollock and Felicie Albert on betatron radiation.
4. Human resources impact for DOE NNSA: *Dr. Franklin Dollar*, the 2013 Dawson Thesis prize winner, is Native American. *Dennis Gardner* is Hispanic and an NSF Fellow. Past students include *Dr. Richard Sandberg* (now staff at LANL) and *Jim Holtsnider* (now at the State Dept.). NSF Fellow *Chris Mancuso* is a former marine who served in Iraq as a linguist.
5. Leveraging: Three students will work on the project (2 part-time). Support is requested for only one student since two students won highly competitive NSF/DOE Fellowships.
6. Impact on Technology Transfer: High power laser and cryogenic technologies developed at JILA are commericalized and widely used, inluding at DOE labs (Sandia, LANL, LLNL).
7. Research breakthroughs: We made 3 discoveries under OFES support since 2012 –
   - We made the first experimental observations of plasma breakdown and shock formation in individual nanoplasmas (JILA/LLNL/NRL collaborations).
   - We discovered that bright phase-match high harmonic generation in multiply-ionized plasmas can be generated by using intense UV driving lasers (as discussed above).
   - We demonstrated a new plasma explosion imaging technique, where the ion momenta identify the specific location of dense plasma formation within individual nanoparticles.

Research result #1: Observation and Control of Shock Shells in Nanoplasmas:

Nanoparticles absorb many times more laser energy than gaseous species, enabling extreme physics with tabletop laser sources. Indeed, previous experimental studies have observed the production of high-energy ions and even nuclear fusion in femtosecond laser-irradiated nanoplasmas. However, although theoretical studies had predicted for over a decade that shock waves could be generated in nanoplasmas, they had not been observed experimentally because the shock propagation is highly dependent on particle size - requiring either a very uniform target, or the ability to observe *individual* nanoplasmas. Nanoplasma shock waves might allow for the practical generation of quasi-monoenergetic high-energy ions, neutrons from fusion processes, or ultrafast X-ray bursts.

In work published in Physical Review Letters and highlighted in APS Focus and as an Editor's suggestion, we recently made the *first experimental observation of shock shells in nanoplasmas*. Furthermore, by exploring a unique laser parameter space, we also demonstrated that these shock rings can be controlled and enhanced using two time-delayed laser pulses. Previous efforts used high laser intensities of 1x10^{16} W/cm^2 to ionize large collections of noble gas clusters less than 10 nm in diameter. In our experiments, we use larger nanoparticles, ranging from 40 to 200 nm diameter, which allows us to create a nanoplasma using far lower laser intensities (~1x10^{14} W/cm^2) and to examine the ion and electron momentum distributions resulting from the explosion of a single nanoplasma. *Moreover, by performing the experiment on individual nanoparticles, instead of a group of nanoparticles with broad size distributions, we can clearly identify the shock physics that occurs during the rapid expansion of a laser-created nanoplasma. Most importantly, we note that this new capability is unique compared to the large national lab facilities - these experiments cannot be performed there due to low shot rates.*

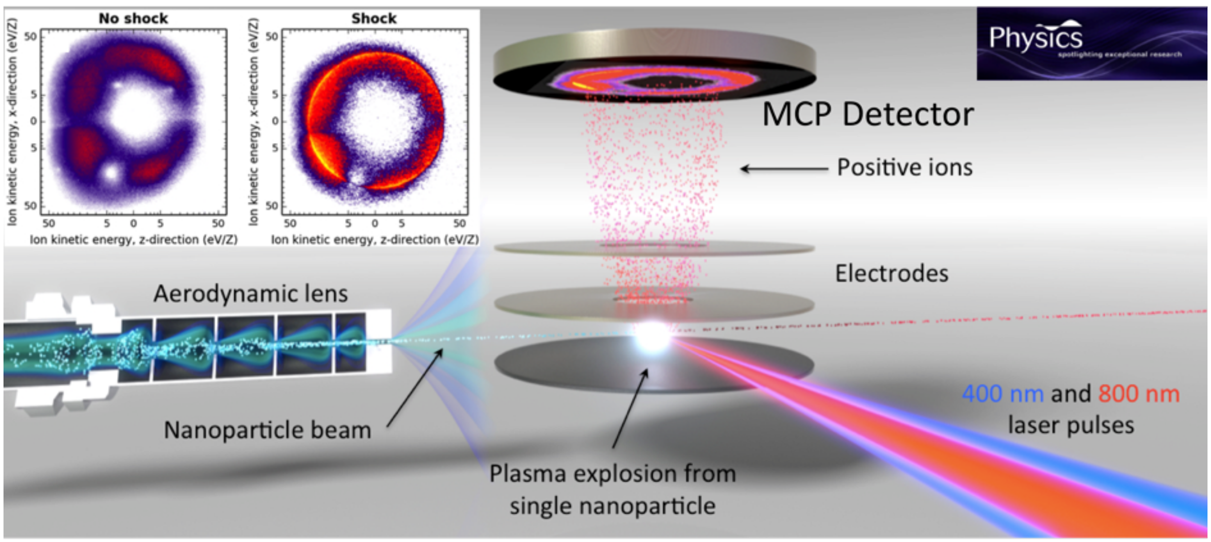

Fig. 2: Imaging shocks in nanoplasmas. An aerodynamic lens produces nanoparticles which are irradiated by one or two time-delayed laser pulses to generate an expanding nanoplasma. (left inset) The photoion angular distribution from a NH_4NO_3 nanoparticle typically displays a broad ion distribution. However, if the particle size, laser intensity, and laser pulse time-delay are tuned appropriately, a sharp shock wave (yellow) appears.

Our measurements of photoelectrons and photoions from single ~100 nm nanoparticles are made possible by the unique combination of a velocity map imaging (VMI) spectrometer, a nanoparticle aerosol source, an aerodynamic lens which collimates the nanoparticle aerosol, and a femtosecond laser with a focal volume small enough to only probe a single nanoparticle (Fig. 1). We find that the nature of the nanoplasma explosions is very sensitive to the laser intensity, the particle size, and the composition of the particle, and that optimized generation of shock rings occurs only with the right combination of these parameters.

<u>Research result #2: Nanoscale absorption of intense lasers using plasma explosion imaging</u>

In work recently published in ACS Nano, we demonstrated a new technique that we call *plasma explosion imaging*, where we first create a localized dense plasma in a nanoparticle and then use the momentum of the ejected ions to infer the location of this plasma within individual nanoparticles (collected using a velocity-map imaging spectrometer). The location of the plasma indicates where breakdown initiates due to enhanced fields in the particle, and provides a map (image) of light absorption on the nanoscale - in an intensity regime that has not been explored to date. Using this unique capability, we found that the photoion momentum distributions resulting from the nanoplasma spark vary considerably from particle to particle because they depend on the nanoscale composition, as well as the shape and orientation of the nanoparticles. We find that nanocrystals of salts explode asymmetrically, whereas TiO_2 aggregates exhibit homogeneous absorption and explosion, while the explosion of metal nanoparticles depends on their size. This work directly reveals how nanoparticles and nanoplasmas respond to intense laser fields above the damage threshold, as part of a room temperature nanoparticle is transformed into a warm dense plasma – a regime that is extremely challenging to model.

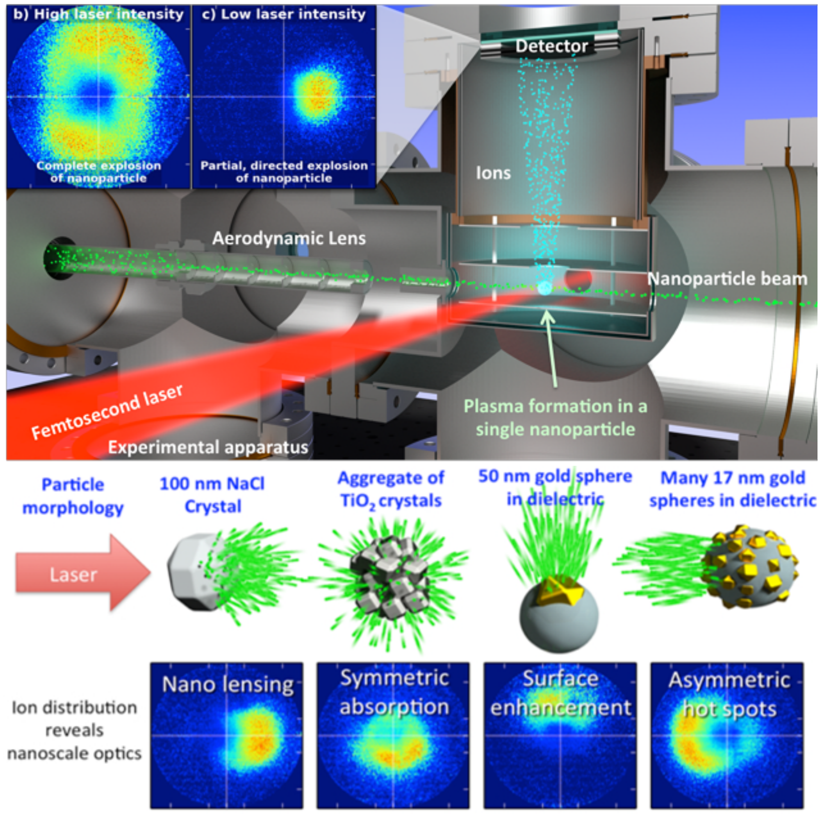

Fig. 3: Plasma explosion imaging of single nanoparticles. A single nanoparticle interacts with a tightly focused 40fs laser pulse. At high laser intensities (>10^{14} W/cm^2), it transforms into a plasma, ejecting ions in all directions (left top inset). (bottom) When a particle encounters low laser intensity, plasma forms in a small region of the nanoparticle due to field enhancement, and ions are ejected in a specific direction that depends on the structure, composition and orientation of the particle.

<u>Research result #3: First Evidence of Phase Matched High Harmonic Generation in Plasmas:</u>

High harmonic generation from tabletop-scale femtosecond lasers results from an extreme quantum nonlinear response of atoms to intense laser light, extending well into the soft X-ray region to photon energies >1 keV. To date however, efficient HHG in the soft X-ray region demanded a trade-off between two competing factors. High harmonics are radiated as a result of a coherent recollision process that occurs during laser field ionization of an atom. This physics dictates that the HHG emission per atom is brightest for short wavelength UV driving lasers (<0.8µm), to reduce quantum diffusion of the electron wavepacket as it propagates away from, and then back to its parent ion. However, generating bright, phase matched, HHG beams in the soft x-ray region requires long wavelength mid-IR driving lasers (>0.8µm), to ensure that the laser and the X-ray light both propagate at the speed of light *c* through an extended medium (Fig. 4).

In exciting recent work, we demonstrated that by driving HHG using intense 0.27µm lasers, we can *simultaneously maximize both the microscopic and macroscopic soft X-ray yields*. Our simple models suggest that for UV driivng lasers, lower quantum diffusion dramatically increases the HHG emission per atom, while the higher linear and nonlinear indices of the atoms and ions compensate the smaller negative index of the generated free-electrons. *This allows coherent HHG build up even in a multiply-ionized plasma, for the first time with sufficient flux for applications (Fig. 4)*. Moreover, we observe a bright flat plateau of HHG from ions for the first time: flux measurements indicate efficiencies comparable or greater than can be achieved using any other approach to date over the entire EUV to soft X-ray spectral range.

<u>3.4 Impact on Technology and Personnel at DOE and DoD Labs</u>

Two graduate students spent summers at LLNL. (NSF Fellow Dennis Gardner, SSGF Fellow Ben Galloway) Past iterations of this grant have funded students now working at the NNSA labs and in govt. (*Dr. Richard Sandberg*, LANL, *Jim Holtsnider*, State Department) Cryocooled high average power laser technology developed at JILA has been commercialized by KMLabs, and are now in use at DOE and DoD labs (Sandia, LANL, LLNL, LBL, Wright Patterson).

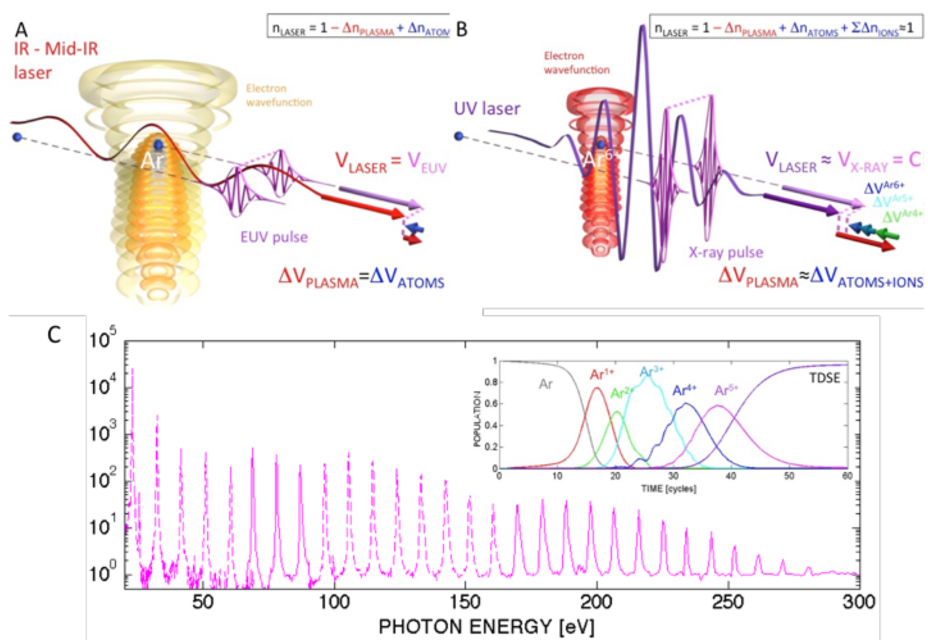

Fig. 4. Phase-matched or near-phase matched HHG in hightly-ionized plasmas. (A) "Traditional" phase matched HHG using IR lasers: the refractive index of a near-neutral gas (~5% ionization for Ar), balances plasma dispersion. (B) For UV lasers, the high linear and nonlinear indices of atoms and ions (>500% ionization in Ar) counterbalances plasma dispersion, enabling coherent HHG buildup in plasmas. (C) Bright HHG in Ar driven by 0.27µm lasers extends from the UV to 280 eV. (inset) Calculations show that above 30eV, HHG emerges from a multiple-ionized plasma.

<u>Publications and Patents for this grant since 2012</u>

1. D. Hickstein, F. Dollar, J. Ellis, K. Schnitzenbaumer, B. Palm, K. Keister, C. Ding, G. Petrov, J. Gaffney, M. Foord, S. Libby, G. Dukovic, J. Jimenez, H. Kapteyn, M. Murnane, W. Xiong, "Mapping Nanoscale Absorption of Femtosecond Laser Pulses using Plasma Explosion Imaging," *ACS Nano* ***8***, 8810 (2014).

2. D. Hickstein, W. Xiong, F. Dollar, J. Gaffney, M. Foord, G. Petrov, B.B. Palm, K.E. Keister, J. Ellis, C. Ding, S. Libby, J. Jimenez, H. Kapteyn, M. Murnane, "Observation and control of shock waves in individual nanoplasmas", *Phys. Rev. Lett.* **112**, 115004 (2014). *PRL Editors' Suggestion (1 in 8 papers); Highlighted in Physics 7, 28, "The Smallest Shock Wave".*

3. D. Popmintchev, C. Hernández-García, F. Dollar, C. Mancuso, J. Pérez-Hernández, M.C. Chen, A. Hankla, X. Gao, B. Shim, A. Gaeta, M. Tarazkar, D. Romanov, R. Levis, J. Gaffney, M. Foord, S. Libby, A. Jaron-Becker, A. Becker, L. Plaja, M. Murnane, H. Kapteyn, T. Popmintchev, "Efficient soft X-ray high harmonic generation in multiply-ionized plasmas: the ultraviolet surprise," *Science* **350**,1225 (2015).

4. T. Popmintchev, D. Popmintchev, M. M. Murnane, and H. Kapteyn, "Method for phase-matched generation of coherent VUV, EUV, and x-ray light using VUV-UV-VIS lasers," US Patent Application submitted 2013 US20150063385, Notice of Allowance 2015. Patent # 9,627,844.

<u>Student and Faculty Awards since 2012</u>

1. 2016 Honorary Degree of Doctor of Science, Uppsala University, Sweden
2. 2015 Honorary Degree of Doctor of Science, National University of Ireland
3. 2015 Elected to Member, American Philosophical Society
4. 2015 Honorary Degree of Doctor of Science, University College Dublin

5. 2015 Honorary Degree of Doctor of Science, Trinity College Dublin
6. 2014 Moore Foundation Experimental Investigator Award
7. H. Kapteyn and M. Murnane, University of Colorado Boulder Inventors of the Year (2014)
8. Franklin Dollar, International John Dawson Thesis Award (2013)
9. Henry Kapteyn, Elected to the US National Academy of Sciences (2013)
10. Ben Galloway, DOE NNSA Stewardship Science Graduate Fellowship (2013)
11. Margaret Murnane, Honorary Member, Royal Irish Academy (2013)
12. M. Murnane, Chair, President’s Committee for the National Medal of Science (2012-2014)
13. H. Kapteyn, Outstanding Alumnus Award, Harvey Mudd College (2013)
14. Chris Mancuso, NSF Graduate Fellowship (2012)
15. H. Kapteyn and M. Murnane, Lamb Award for Laser Science and Quantum Optics (2012)