**White Paper for Frontiers of Plasma Science Panel**

**Date of Submission:** June 21st 2015

Indicate the primary area this white paper addresses by placing “P” in right column.
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| • Plasma Atomic physics and the interface with chemistry and biology | S |
| • Turbulence and transport | |
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| • Statistical mechanics of plasmas | P |

Indicate type of presentation desired at Town Hall Meeting.

| “X” |
| Oral (remotely) | X |
| Poster | |
| Either Oral or Poster | |

| Title: Resolving collective electron dynamics in nanoplasmamas with time resolution of 1 femtosecond |
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**Describe the research frontier and importance of the scientific challenge.**

We propose to create nano-sized plasmas at extreme conditions in high-repetition rate experiments to study the electron dynamics in such plasmas with ~ 1 femto-second time resolution approaching electron-electron collision times. In particular, this time scale is shorter than a single cycle of the infra-red (IR) laser driver (2.7 fs for a 800 nm laser pulse). Such measurements with unprecedented time-resolution allow studying fundamental aspects of light-matter interaction, for example the role of collective excitations, and fundamental equilibration processes over a wide range of plasma conditions. The results can be used to benchmark high-energy density physics models in the challenging regime of non-LTE plasmas.

Atomic clusters can serve as ideal model systems for exploring ultrafast laser-driven ionization dynamics of dense matter on the nanometer scale [1]. They absorb intense laser radiation with huge cross sections [2] and emit fast electrons [3], energetic photons [4], and charged ions [3,5] with MeV kinetic energy. Besides fundamental relevance for light-matter interactions and relaxation and transport processes in plasmas, also potential applications, e.g., extreme ultraviolet light sources, compact particle accelerators, or pulsed neutron sources, make a detailed understanding of laser-cluster processes desirable.

Much of the violent response of clusters to intense laser fields is related to the creation of a transient nano-plasma. A temporary resonant collective electron mode (plasmon) results in strongly enhanced absorption, as has been demonstrated in simulations [1,3] (cf. Fig. 1) as well as in experiments through increased yields of high-Z ions [6], electrons and x rays for optimal pulse conditions. When present, plasmon-enhanced coupling is more effective than higher order nonlinear absorption effects. The energy
capture results from resonant driving of quasi free electrons, either inherent to the system or created through inner ionization, over a transiently critical ionic charge density $\rho_{\text{crit}} = 3m_e\epsilon_0\omega_{\text{las}}^2/\epsilon$. Because of strong field amplification at resonance also the anisotropy of the electron emission is increased, as has been observed on metallic and large ($N \sim 10^5$) noble-gas clusters. The directional acceleration was traced back to the strong plasmonic field enhancement at the resonance, which supports emission and most efficient additional acceleration of electrons along the laser polarization [3], cf. Fig. 1. Several other effects stemming from the short-lived and dense nanoplasma in the Coulomb-exploding clusters have been investigated as possible mechanisms for high-charge ion production. These include tunnel ionization enhanced by fields from neighboring ions, space-charge fields, and resonant field amplification [7] and electron impact ionization [8,9] with atomic ionization thresholds lowered by plasma effects in the clusters [10].

So far, resonant collective heating of electrons and their plasmonic-field-driven acceleration could only be measured via the analysis of final reaction products [3,6]. The experiments proposed here aim at measuring in real-time the nanoplasma formation, the emergence and evolution of collective resonant heating, and the collisional relaxation of the cluster nanoplasma via spectrally resolved x-ray scattering from individual electrons. Such experiments became possible only with the advent of x-ray Free Electron Lasers like LCLS at SLAC in Stanford, CA, which provide a powerful, highly-collimated and ultra-short x-ray source that is required for low-cross section x-ray scattering measurements of small or low-density targets.

- Describe the approach to advancing the frontier and indicate if new research tools or capabilities are required.

While we will describe a proof-of-principle experiment, the general concept can be expanded in a straightforward way to other targets and excitation conditions. A 20 mJ/50 fs optical pump laser, operating at 800 nm and 120 Hz repetition rate, at the AMO endstation at the LCLS x-ray Free Electron laser at SLAC, Stanford, CA is used to heat an intense rare gas cluster beam to high charge states (e.g., $Z \sim 15$ in Xe$^N$) and temperatures ($T_e \sim 1$ keV). Coherent [11-13] and incoherent x-ray Thomson scattering (XRTS) [14-16] is used to probe the nanoplasma conditions, see Fig. 2. The XFEL, operating at $E_{\text{photon}} = 1.8$ keV, will be aligned nearly collinear to the optical laser and, with variable delay, will scatter off the heated nano-particles. The scattering signal consists of elastic and inelastic contributions. The elastic scattering derives from bound electrons and near-elastic scattering from electrons that closely follow the motion of the ions. Inelastic scattering is dominated by scattering from quasi-free electrons; only a small contribution comes from originally bound electrons that get promoted. The near-collinear pump-probe configuration as shown in Fig. 2 is chosen to avoid time-shear in the beam overlap region. Photon energy and scattering angle determine the scattering regime and what scale lengths are probed. An experiment conducted at 1.8 keV at a scattering angle of $\sim 25^\circ$ would be in the non-collective regime, i.e. the scattering will probe the motion of individual electrons. Inelastically scattered x-rays will be down-shifted by the Compton shift (0.4 eV) plus an additional offset from the Doppler-effect of $\pm \hbar \vec{k} \cdot \vec{v}$, cf. Fig. 2, with $\vec{v}$ the electron velocity vector. Hence, the scattering measurement maps out the electron velocity distribution along a chosen direction in the plasma. For example, by tuning the IR laser polarization by $90^\circ$, $\vec{k}$ will probe the electron velocity components parallel and perpendicular to the laser polarization axis, one at a time.

A high-efficiency x-ray spectrometer will collect the scattered photons towards a pnCCD detector [17], which allows for high-repetition rate (120 Hz) data acquisition in single photon counting mode. The experiments will be conducted in single shot acquisition, and a cross-correlation signal between LCLS beam and optical laser will be recorded for each shot to achieve $\sim 20$ fs accuracy in the optical delay measurement [18]. Even though the count rate of the described setup is low (ca. 1 photon per shot), the background-free nature of the measurement and the high repetition rate allow for achieving high-quality
scattering spectra within ~100 s integration time. The accuracy of the measurement is ultimately only limited by the integration time and the shot-to-shot stability of the measurement. The described proof-of-principle experiment can be improved by

- Operating the x-ray laser reliably at 1 fs pulse length with pulse energies on order of 1 mJ (this would allow to resolve the collective electron quiver motion shown in Fig. 1b)
- Increase repetition rate of FEL and IR pump laser to ca. 10 kHz (currently 120 Hz)
- Reduce IR pump pulse length to ~10 fs and provide pulse shaping capabilities to tailor plasma excitation
- Improve synchronization of FEL and IR lasers
- Develop 2D single photon counting detectors (like pnCCDs [17]) with up to 10 kHz readout rate
- Operate with high-flux, mono-disperse particle source

Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit.

By proper choice of the scattering geometry, the time-resolved scattering spectra will contain information on (i) the number of electrons participating in the resonantly-driven collective plasmon motion, (ii) the buildup and mean velocity of the collective motion, and (iii) the dissipation of energy into thermal nanoplasma energy. By varying the delay between the optical excitation and the x-ray probe, the electron-ion collision frequency (and thus the conductivity) will be measured via the direction-resolved analysis of the electron velocity distribution. Such measurements could help us gain a better understanding of conductivity at conditions at the ablator-fuel interface in ICF capsule implosions, which is important for improved modeling of hydrodynamic instabilities at this critical interface [19]. The number of residual bound electrons will be extracted from the elastically scattered radiation (Rayleigh signal), allowing the direct time-resolved measurement of the atomic ionization state. So far, the above fundamental observables are not well characterized in highly excited plasmas. Obtaining the described quantities in a time-resolved sense allows to inferring fundamental properties in nano-sized plasmas at extreme conditions with unprecedented time – resolution. Thanks to the high-repetition rate the experimental accuracy is ultimately only limited by the integration time. Besides fundamental relevance, an improved understanding of the light-matter interactions is also important for future particle and radiation sources [4], plasma waveguide generation [20], and laser induced dielectric modifications and micromachining [21].
References

Fig. 1: (a) Illustration of Surface Plasmon Assisted Rescattering in Clusters (SPARC, cf. Ref. [4]) leading to anisotropic electron velocity distributions in clusters that are driven close to surface plasmon resonance: Electrons are accelerated during their last transit through the cluster, (b) accelerated and some of them ejected along the polarization of the laser field, leading to (c) a distinct anisotropy of the high-energy electron emission. Adapted from Fennel et al., Phys. Rev. Lett. 98, 143401 (2007).

Fig. 2: Schematic of the experimental setup with near collinear IR pump and FEL probe beams. The scattering diagram shows the scattering vector $\mathbf{k}$, which determines the direction along which the electron velocity distribution $\mathbf{v}$ is measured. The latter determines the shape of the Doppler-broadened Compton profile (shaded) in the inelastic x-ray scattering spectrum.