White Paper for *Frontiers of Plasma Science Panel*

Date of Submission:	**June 19, 2015**

Indicate the primary area this white paper addresses by placing "P" in right column.
Indicate secondary area or areas by placing "S" in right column

	"P", "S"
• Plasma Atomic physics and the interface with chemistry and biology	P
• Turbulence and transport	
• Interactions of plasmas and waves	S
• Plasma self-organization	
• Statistical mechanics of plasmas	

Indicate type of presentation desired at Town Hall Meeting.

	"X"
Oral	X
Poster	
Either Oral or Poster	
Will not attend	

Title:	**Intense Coherent and Incoherent Table-top X-ray Generation from Plasmas**
Corresponding Author:	Prof. Jorge J. Rocca
• Institution:	Colorado State University
• email:	jorge.rocca@colostate.edu
Co-Authors:	Vyacheslav Shlyaptsev

1. The Research Frontier and Importance of the Scientific Challenge

Bright soft x-ray and x-ray radiation is a key enabling tool for new studies in plasma science, nanoscience and nanotechnology, materials science, chemistry, and biology. The great interest in its use has motivated the commissioning of free electron lasers. These accelerator-based machines produce extreme x-ray intensities that are opening new exciting opportunities in science. However they are large in size and their access is very limited. Dense plasma columns provide the opportunity to develop compact sources of bright x-ray laser radiation from atomic transitions that can be more readily accessible and can generate intense coherent beams at the site of the experiment or industry application. In turn, new advances in volumetric heating of hot near-solid density dense plasmas created by irradiation of aligned nanostructures of relativistic intensity can efficiently generate intense ultrashort flashes of incoherent hard x-rays for applications requiring intense flashes of multi-keV x-rays such as fusion plasma backlighters. The scientific challenge is a better understanding of laser-matter interaction that can facilitate the development of new plasma schemes that in combination with new laser driver technology can increase the efficiency ,power, and wavelength range of these x-ray plasma sources.

2. Approach to Advancing the Frontier and Need

2.1 Plasma-based soft X-ray lasers

Plasma-based soft x-ray lasers (SXRLs) are unique in that they can produce bright pulses of soft x-ray coherent radiation with high energy and narrow bandwidth on a table top. Compact lasers based on

collisional electron impact excitation of multiply ionized atoms are currently enabling applications in nano-scale imaging [1], dense plasma diagnostics [2], the development of analytic nano-probes [3], defect-free patterning of nano-structures [4], and other areas. Major challenges are to extend these [5-7] and other [8] plasma-based table-top SXRLs to shorter wavelengths, increase their average power, and shorten their pulse width from ps to fs. The wavelength scaling of the energy necessary to pump such lasers imposes a challenge to the realization of gain-saturated high repetition rate lasers at sub-10 nm wavelengths. Experiments at Colorado State University (CSU) recently succeeded in extending gain-saturated table-top lasers to 8.8 nm by amplification in an atomic transition of 29 times ionized La atoms in a laser-created plasma using traveling wave excitation with picosecond laser pulses, the shortest wavelength gain-saturated table-top laser reported to date [5]. The result, obtained using pump pulses with an unprecedentedly low energy, opens the prospect for bright high repetition rate plasma-based lasers at shorter wavelength and high repetition rates. The experiments made use of isoelectronic scaling to also obtain lasing in several shorter wavelength transition in lanthanide ions down to 7.9 nm in Ni-like Nd (Fig.1). Modeling suggests that these dense plasma amplifiers have the bandwidth necessary to sustain the amplification of SXRL pulses in the femtosecond range. Another challenge is to increase the average power of these lasers. Until recently the average power of plasma-based SXRLs was limited by the 10 Hz repetition rate of the solid state optical lasers that drive them. To overcome this limitation researchers at CSU developed a compact, directly diode-pumped chirped pulse amplification (CPA) laser based on cryogenically-cooled Yb:YAG that produces 1 Joule, 5 ps FWHM duration, λ=1.03 μm pulses at 100 Hz. This laser was used to drive a gain-saturated 18.9 nm SXRL in a Ni-like Mo plasma producing a record average power of 0.15 mW [6], increasing the repetition rate and average power of compact sub-20 nm lasers by an order of magnitude,

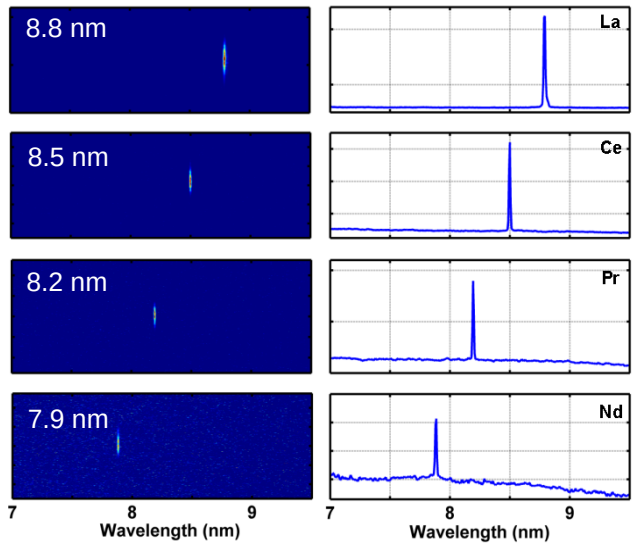

Fig. 1. Spectra of of the axial emission from plasmas created irradiatiing slabs of lanthanide materials of increasing atomic number with intense optical laser pulses. Lasing is observed at progressively shorter wavelengths in the 4d $^1S_0$-4p$^1P_1$ line of the Ni-like lanthanide ion sequence, down to 7.9 nm in nickel-like Nd.

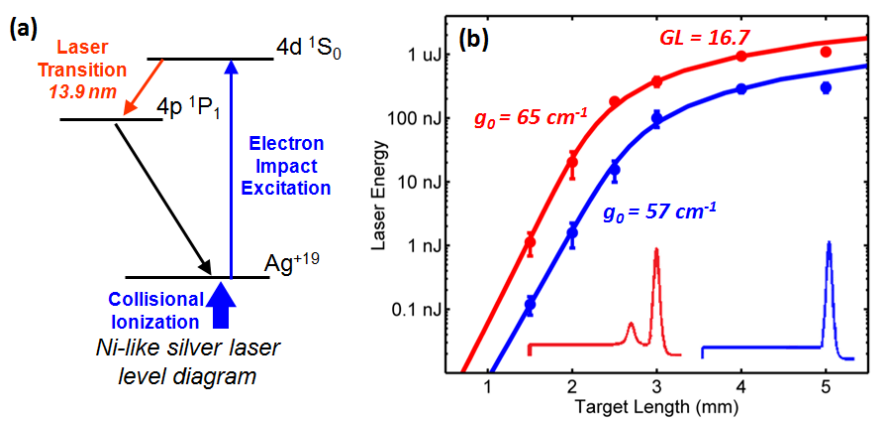

Fig. 2. Effect of adding a small (12% of main pulse energy) short pre-pulse in the output pulse energy output of a λ=13.9 Ni-like Ag laser pulse. (a) Excitation mechanism of the collisionally excited λ = 13.9nm Ni-like Ag SXRL. (b) Measured SXRL output pulse energy as a function of target length for pump pulse sequences with (red) and without (blue) added short pulse. The addition of a 12% picosecond-duration pulse before the main driver pulse (red traces) results in a higher gain and a ~3 fold SXRL energy enhancement.

the highest value reported to date for coherent table-top source at these wavelengths. Recent studies conducted in Europe [9] and CSU [7] show that the performance of collisional SXRLs depends strongly on the temporal profile of the laser pump energy used to create and heat the soft x-ray plasma]. For example, the addition of a small picosecond pre-pulse with only 12% of the energy of the main pulse a few tens of picoseconds ahead the main pulse increased by ~ 300 % the pulse energy produced by a λ = 13.9 nm Ni-like Ag SXRL (Fig.2). Model simulations show that the addition of the small pre-pulse causes the times of maximum Ni-like ion density and electron temperature to coincide, allowing us to obtain the 3X increase in SXRL energy and a record 0.1 mW average power at λ=13.9 nm. This result shows that tailoring of the plasma excitation has great potential to increase the efficiency and power of these SXRLs. Technologically what is needed is new compact high energy laser drivers capable to heat the necessary plasmas at high repetition rates (kHz).

2.2 Efficient generation of ultrafast x-ray pulses from aligned nanostructure arrays irradiated with laser pulses of relativistic intensity

The efficient generation of intense ultra-short bursts of X-ray radiation is essential for backlighting nuclear fusion experiments, and is also of high interest for probing ultrafast structural changes in materials, high resolution x-ray flash imaging, and other applications. Dense plasmas produced by irradiation of materials with intense femtosecond laser pulses are bright emitters of X-ray pulses originating from an area roughly the size of the laser focus spot. A consequence of the high plasma density is a fast radiative lifetime that produces a short burst of X-rays, typically several picoseconds in duration. Efforts to increase the X-ray yields have included the use of structured target surfaces to improve the coupling of the laser energy into materials [10-17], that yielded conversion efficiencies of 10^{-4} -10^{-2} for X-rays >1keV. When nanowire targets were employed, hard X-ray yields of more than 40X compared to polished flat targets were achieved [12,15]. Recent experiments have shown that the trapping of femtosecond laser pulses of relativisitc intensity deep within ordered nanowire arrays (Fig 3, and Fig 4a) volumetrically heats plasmas with electron densities nearly 100 times greater than the typical critical density to multi-keV temperatures (Fig. 4c), generating a new dense ultra-hot large-volume plasma regime with extreme degrees of ionization (Ni^{+26}, Au^{+52}) that favors the highly efficient generation of X-rays [17]. This approach can provide record X-ray conversion efficiency by depositing laser energy deep into the array, heating a volume several µm in depth with average densities corresponding to 12 percent solid density to KeV temperature. In this scheme ultrafast heating of vertically aligned nanowire targets with femtosecond laser pulses of ultra-high

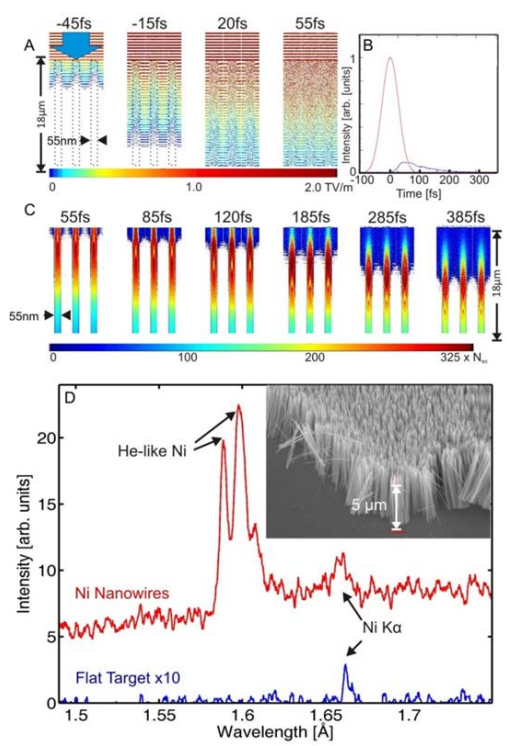

Fig.4. Particle-in-cell simulation and spectra from plasma generated by fs laser pulse irradiation of a vertically aligned array of 55 nm diameter Ni nanowires. (A) PIC simulations of the penetration of the laser beam electric field in an array of 18 µm long Ni wires with an average atomic density of 12% solid density irradiated at an intensity of 5 x 10^{18} W cm-^{2} by a λ= 400 nm, 60 fs FWHM duration laser pulse. Times are measured with respect to the peak of the laser pulse. (B) Computed impinging (red contour) and reflected (blue contour) laser intensity. (C) Computed electron density evolution. The electron density scale is units of critical density (N_{ec}=6.8x10^{21} cm^{-3}). (D) Single-shot X-ray spectra comparing the emission from an irradiated array of 55nm diam., Ni nanowires to that from a flat polished Ni target. The nanowire target plasma spectra is dominated by He-like Ni line emission. The only line observed in the flat target plasma spectra is the Ni-Kα line. The inset is an scanning electron microscope image of the array of vertically aligned nanowires. [17]

contrast overcomes the limitations imposed by the critical density surface to volumetric heating of plasmas for the production of highly super-critical (~100 times critical density), multi-keV temperature plasmas [17]. Such extreme plasma parameters had only been reached previously using multy-kJ laser energies to induce spherical compression. The large heated plasma volume relative to plasmas from flat solid targets significantly lengthens the plasma hydrodynamic time, which combined with a greatly decreased radiative lifetime that results from the increased collisional excitation rate associated with a nearly two orders of magnitude increase in plasma density respect to solid targets, results in a greatly decreased cooling-time to hydrodynamic-time ratio, greatly increasing the X-ray yield.

Irradiation of arrays of vertically aligned 55 nm diameter Ni nanowires at an intensity of $5x10^{18}$ Wcm^{-2} with ultra-high contrast (λ= 400 nm) pulses of ~ 60 fs FWHM duration showed strong emission from He-like Ni (Ni^{+26}) lines (Fig. 4D). This spectrum differs greatly from that for a polished flat target irradiated at the same conditions which only shows line emission from the Ni Kα line (with 10 X magnified scale in Fig.4 D). The Kα emission is produced mainly by high energy electrons, while generation of the He-like ion transitions requires a hot thermal plasma to generate and excite the highly stripped ions. It is remarkable that the He-like line emission from the nanowire target exceeds the intensity of the Kα line at this irradiation intensity, since in previous work with Cu foils the emission from the Kα lines was only surpassed at irradiation intensities $>2x10^{20}$ Wcm^{-2}. The aligned Ni nanowire target produced an increase of more than 50 times in X-ray flux in this 7-8 keV spectral region. Similar enhancements were also observed in the 1 keV photon energy region in agreement with previous experiments [11]. Similarly, irradiation of Au nanowire arrays created near solid-density Au plasma with an extraordinarily high degree of ionization. Au nanowire spectrum in the λ=4.5-5.25 Å region displays strong Au M-shell emission with unresolved 4-3 lines from ions ranging from Co-like (Au^{+52}) to Ga-like Au (Au^{+48}) [17]. The spectra show increases of up to 100X in the emission in this photon energy region as compared to a polished flat target irradiated with the same pulse. Filtered photodiode measurements showed a 50X emission increase for photon energies > 8 keV. The greatly increased hydrodynamic to radiative lifetime ratio of these plasmas and their increased absorption promises to result in unsurpassed X-ray yields.

3. Impact and societal benefits

The study of this frontier of plasma science, the physics leading to the efficient conversion of optical laser light into coherent and incoherent X-ray radiations, will both increase fundamental knowledge of intense light-plasma interactions and can result in compact ultra-bright sources of X-ray radiation that will enable advances in several fields of science and technology. New compact plasma-based X-ray lasers can make possible for example the visualization of nano-scale dynamic phenomena by sequential single-shot imaging, can diagnose very dense plasmas with densities and gradients that are beyond the reach of optical and ultraviolet lasers, make possible the development of analytic nano-probes that can map in 3-D the chemical composition of biological micro-organisms, and enable the defect-free patterning of nano-structures.

References.

1. S. Carbajo, I.D. Howlett, F. Brizuela, K.S. Buchanan, M.C. Marconi, W. Chao, E.H. Anderson, I. Artioukov, A. Vinogradov, J.J. Rocca, and C.S. Menoni, “Sequential single-shot imaging of nanoscale dynamic interactions with a table-top soft X-ray laser,” Optics Letters **37**, 2994 (2012).
2. M.A. Purvis, J. Grava, J. Filevich, D.P. Ryan, S.J. Moon, J. Dunn, V.N. Shlyaptsev, and J.J. Rocca, “Collimation of dense plasma jets created by low energy laser pulses and studied with soft X-ray interferometry,” Phys. Rev. E **81**, 036408 (2010).
3. I. Kuznetsov , J. Filevich, F.Dong, M. Woolston, W. Chao, E. Anderson, E. Bernstein, J.J Rocca, and C.S. Menoni, “Three dimensional nanoscale molecular imaging by extreme ultraviolet laser ablation mass spectrometry, Nature Communications **6**, Art. No. 6944 (2015).
4. L. Urbanski, W. Li, J.J. Rocca, C.S. Menoni, M.C. Marconi, A. Isoyan, and A. Stein, “Defect tolerant extreme ultraviolet lithography technique,” J. Vac. Sci. Technol. B **30**, 06F502 (2012).

5. D. Alessi, Y. Wang, D. Martz, B. Luther, L. Yin, D. H. Martz, M.R. Woolston, Y. Liu, M. Berrill, and J.J. Rocca, “Efficient excitation of gain-saturated sub-9 nm wavelength table-top soft X-ray lasers and lasing down to 7.36 nm,” Physical Review X **1**, 021023 (2011).
6. B.A. Reagan, K.A. Wernsing, A.H. Curtis, F.J. Furch, B.M. Luther, D. Patel, C.S. Menoni, J.J. Rocca, “Demonstration of a 100-Hz repetition rate gain-saturated diode-pumped table-top soft X-ray laser,” Optics Letters **37**, 3624 (2012).
7. B.A. Reagan, M. Berrill, K.A. Wernsing, C. Baumgarten, M. Woolston, J.J. Rocca, “High-average-power, 100-Hz-repetition-rate, tabletop soft-x-ray lasers at sub-15-nm wavelengths,” Physical Review A **89**, 053820 (2014).
8. Y. Avitzour and S. Suckewer , “ The feasibility of achieving gain in transition to ground state of CVI at 3.4 nm”. Journal of Optical Scoeity of America B, **24**, 817, (2007)
9. R. A. Banici, G. V. Cojocaru, R. G. Ungureanu, R. Dabu, D. Ursescu, and H. Stiel, "Pump energy reduction for a high gain Ag X-ray laser using one long and two short pump pulses," Optics letters **37**, 5130-5132 (2012).
10. Murnane M.M. et al. Efficient coupling of high-intensity sub-picosecond laser pulses into solids. Appl. Phys. Lett., **62**, 1068 (1993).
11. Gordon S.P., Donnelly T., Sullivan A., Mamster H., Falcone R.W. X-rays from microstructured targets heated by femtosecond lasers. *Opt. Lett.*, **19,** 484-487 (1994).
12. Kulcsár G. et al. Intense Picosecond X-Ray Pulses from Laser Plasmas by Use of Nanostructured “Velvet” Targets. *Phys. Rev. Lett.*, **84**, 5149-5152 (2000).
13. Rajeev P.P, Taneja P., Ayyub P., Sandhu A.S., Kumar G. Metal Nanoplasmas as Bright Sources of Hard X-Ray Pulses. *Phys. Rev. Lett*, **90**, 115002 (2003).
14. Sumeruk, H.A. et al. Control of Strong-Laser-Field Coupling to Electrons in Solid Targets with Wavelength-Scale Spheres. *Phys. Rev. Lett.*, **98**, 045001 (2007).
15. Mondal S. et al. Highly enhanced hard x-ray emission from oriented metal nanorod arrays excited by intense femtosecond laser pulses. *Phys. Rev. B*, **83**, 035408 (2011).
16. Ovchinnikov A.V. et al. Characteristic X-rays generation under the action of femtosecond laser pulses on nano-structured targets. *Laser and Particle Beams*, **29**, 249–254 (2011).
17. M.A. Purvis, V.N. Shlyaptsev, R. Hollinger, C. Bargsten, A. Pukhov, A. Prieto, Y. Wang, B. Luther, L. Yin, S. Wang, J.J. Rocca, “Relativistic plasma nano-photonics for ultra-high energy density physics,” Nature Photonics **7**, 796 (2013).