SLAC-PUB-16558

Measurement of high-dynamic range x-ray Thomson scattering spectra for the characterization of nano plasmas at LCLS[a)]

M. J. MacDonald,[1,2,b)] T. Gorkhover,[2,3] B. Bachmann,[4] M. Bucher,[2] S. Carron,[2] R. N. Coffee,[2] R. P. Drake,[1] K. R. Ferguson,[2,5] L. B. Fletcher,[2] E. J. Gamboa,[2] S. H. Glenzer,[2] S. Göde,[2] S. P. Hau-Riege,[4] D. Kraus,[6] J. Krzywinski,[2] A. L. Levitan,[7] K.-H. Meiwes-Broer,[8] C. P. O'Grady,[2] T. Osipov,[2] T. Pardini,[4] C. Peltz,[8] S. Skruszewicz,[8] M. Swiggers,[2] C. Bostedt,[9] T. Fennel,[8] and T. Döppner[4]

[1)] *University of Michigan, Ann Arbor, MI 48109, USA*
[2)] *SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA*
[3)] *Technische Universität, 10623 Berlin, Germany*
[4)] *Lawrence Livermore National Laboratory, Livermore, CA 94551, USA*
[5)] *Stanford University, Stanford, CA 94305, USA*
[6)] *University of California, Berkeley, CA 94720, USA*
[7)] *Franklin W. Olin College of Engineering, Needham, MA 02492, USA*
[8)] *Universität Rostock, D-18051 Rostock, Germany*
[9)] *Argonne National Lab, Lemont, IL 60439, USA*

(Dated: 9 June 2016)

Atomic clusters can serve as ideal model systems for exploring ultrafast (∼100 fs) laser-driven ionization dynamics of dense matter on the nanometer scale. Resonant absorption of optical laser pulses enables heating to temperatures on the order of 1 keV at near solid density conditions. To date, direct probing of transient states of such nano plasmas was limited to coherent x-ray imaging. Here we present the first measurement of spectrally-resolved incoherent x-ray scattering from clusters, enabling measurements of transient temperature, densities and ionization. Single shot x-ray Thomson scatterings signals were recorded at 120 Hz using a crystal spectrometer in combination with a single-photon counting and energy-dispersive pnCCD. A precise pump laser collimation scheme enabled recording near background-free scattering spectra from Ar clusters with an unprecedented dynamic range of more than 3 orders of magnitude. Such measurements are important for understanding collective effects in laser-matter interactions on femtosecond timescales, opening new routes for the development of schemes for their ultrafast control.

I. INTRODUCTION

Atomic clusters readily absorb intense laser radiation due to their extremely large cross section,[1,2] resulting in the emission of high energy electrons[3], ions[4] and x-rays. Previous measurements of ionization dynamics in such nano plasmas relied on the analysis of final reaction products and direct probing of transient nano plasmas was limited to coherent x-ray imaging.[5] Here we present a proof-of-principle experiment using incoherent x-ray Thomson scattering (XRTS)[6] to measure electron velocity distributions in clusters, which will ultimately enable time-resolved measurements of temperature, ionization, and collective electron dynamics on <100 fs time scales.

The small Thomson scattering cross section ($\sigma_T = 6.65\times10^{-25}\text{cm}^2$) in combination with the low average particle density in cluster jets poses a particular challenge for XRTS measurements. Being in the single photon counting regime, efficient mitigation of background signals is required. Furthermore, mid- or high-Z elements require high-dynamic range spectra to resolve inelastic Compton scattering near the strong elastic scattering component. Here we describe the setup for the infrared (IR) laser driven experiment and present the measurement of a high-dynamic range, near background-free XRTS spectrum from Ar clusters using an energy-dispersive pnCCD detector[7] in single-photon counting mode to integrate up to 200,000 shots.

II. EXPERIMENTAL SETUP

The experiment was conducted using the LAMP endstation at the AMO instrument[8,9] at the Linac Coherent Light Source (LCLS).[10] The clusters were created using an Even-Lavie source[11] with nozzle diameter of 150 μm and opening angle of 40°, operated at room temperature with a backing pressure of 80 bar. Based on common scaling, we expect argon clusters, Ar_N, of mean size $N = 60,000$ and atomic density of $n_{at} \sim 10^{17}$ cm^{-3} in the jet at 2 mm from the nozzle.

Fig. 1 shows a schematic of the experimental setup. The gas clusters are manipulated using a pump-probe system with two 40 fs IR pulses at 800 nm from a Ti:Sapphire laser system with intensities up to 10^{15} W/cm^2. The free electron laser (FEL) was operated at 1811 eV to measure XRTS spectra from Ar. An incoupling mirror with a central hole for the FEL probe to pass through was used to make the IR and FEL beams colinear, interacting with the clusters 2 mm from the nozzle

[a)] Contributed paper published as part of the Proceedings of the 21st Topical Conference on High-Temperature Plasma Diagnostics, Madison, Wisconsin, June, 2016.
[b)] macdonm@umich.edu

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-76SF00515, FES, LDRD and NSF.

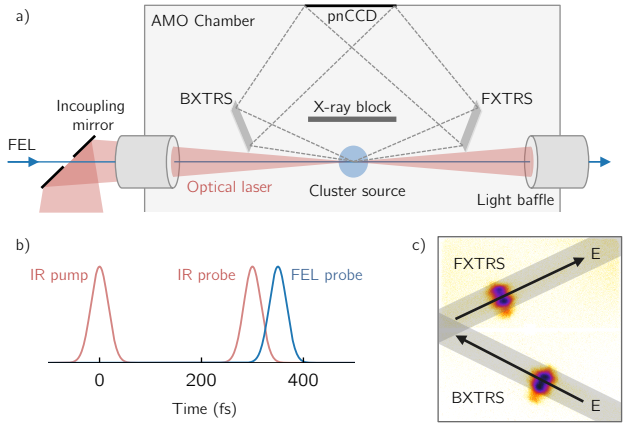

FIG. 1. a) Experimental geometry, b) pump-probe timing of IR pulses and FEL probe, and c) example raw pnCCD data showing the active regions of the detector for the forward (FXTRS) and backward (BXRTS) scattering spectra.

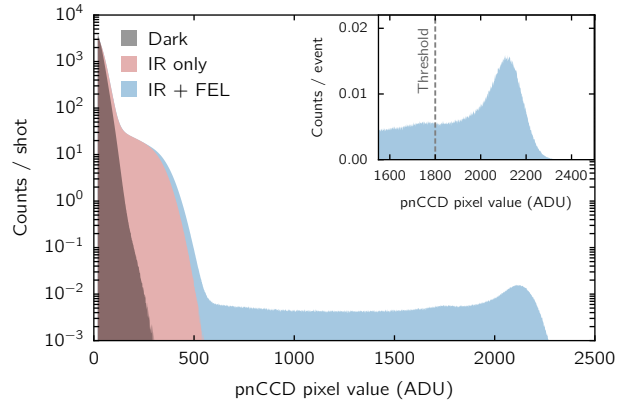

FIG. 2. Histogram of pnCCD ADU values for a dark run, an IR only run, and an IR + FEL run to show the contributions from each source. Counts from scattered x-rays are shown in the inset with the threshold used to remove background.

where the diameter of the cluster jet was ∼1 mm. The relative delay between the pump and probe IR pulses was set using a mechanical delay stage and the IR-FEL timing was measured for each shot using a spectrally-encoded time tool system.[12]

Two cylindrically curved PET (002) crystal spectrometers measured XRTS of the FEL probe in forward (FXRTS) and backward (BXRTS) scattering geometries at scattering angles of 28° and 152°, respectively. The details of the spectrometer design, setup, and characterization are described in detail in a separate publication.[13]

Single photon counting was enabled by the low noise pnCCD detector.[7] Two layers of aluminized polyimide film (700 nm polyimide + 100 nm Al each) were used to stop optical light and an x-ray block was in the direct line-of-sight between pnCCD and interaction region. Background was further reduced by the use of light baffles to limit diffuse IR reflections within the chamber.

III. REMOVAL OF IR LASER BACKGROUND

Fig. 2 shows a histogram of detected pixel intensity values, measured in analog digital units (ADU), normalized by the number of shots for each run. Three runs are presented to show the contribution of each source: a dark run, an IR only run, and an IR + FEL run, containing 8,280, 12,067, and 190,784 shots, respectively. The histogram shows that the IR laser contributes the vast majority of the signal, ∼0.1 IR photons/pixel/shot, but this contribution can be easily removed by setting a minimum threshold above ∼500 ADU. The scattered x-ray peak is located above 1800 ADU, where a minimum threshold is set to remove additional background as shown in the inset, where the counts per shot are shown on a linear scale. A small peak appears near 1750 ADU, which is most likely Al K-α fluorescence at 1486 eV from the pnCCD filtering or other Al components in the chamber. In addition to the XRTS measurements made by the pnCCD, the slope of the histogram data can be used to infer a temperature for the hot plasma.

IV. FEL ENERGY JITTER CORRECTION

The FEL at LCLS has a RMS photon energy jitter of $\sim 0.1\%$ at 1.8 keV. These variations reduce the spectral resolution of scattering measurements when integrating over many shots by convoluting the output spectrum with this distribution. This can be avoided by measuring the centroid of input spectrum for each shot.

To do this, the energy of the electrons before the undulator was recorded for each shot, which is directly correlated to the photon energy of the FEL. A contour plot of the measured BXRTS spectral intensity as a function of electron energy is shown in the inset of Fig. 3. The peak of the scattered intensity, which is dominated by elastic scattering, as a function of electron energy was fit using a linear regression with the best fit line shown in the inset of Fig. 3, with $E_{\text{photon}}(eV) = 604.9E_{e^-}(\text{GeV}) - 2027$.

To remove the effect of electron energy jitter the photon energy of each detected photon was corrected according to this formula and centered at 1811 eV. This reduced the full width at half maximum of the BXRTS spectrum by 18% (1.7 eV) as shown in Fig. 3.

V. XRTS SPECTRA FROM COLD ARGON CLUSTERS

Fig. 4 shows the result for a XRTS spectrum from argon clusters at a scattering angle of 152°, integrated over 175,000 shots. On average, 1 photon is detected within the spectral window shown in Fig. 4 every 2 shots. For this measurement the FEL was defocused to a 200 μm

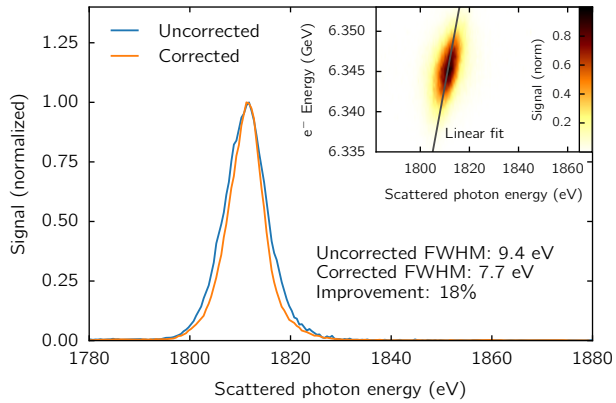

FIG. 3. Comparison of scattering spectra with and without input photon energy correction. The elastic peak of the scattering is a linear as a function of electron energy (inset), which can be used to correct for variations in probe photon energy.

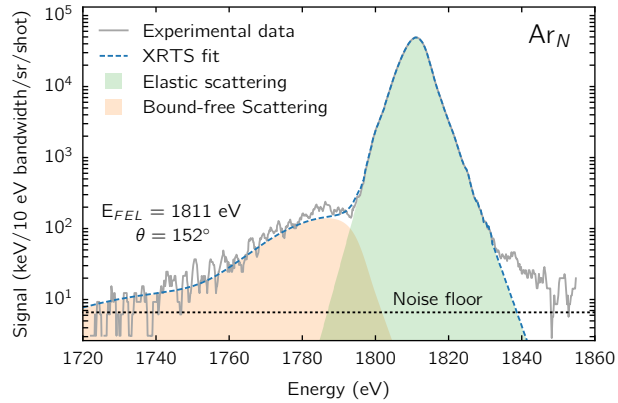

FIG. 4. Scattering spectrum at $\theta = 152°$ and fit from unheated Ar clusters demonstrating the ability to measure elastic and inelastic features with high-dynamic range using single photon counting and a high repetition rate system.

spot, delivering 0.25 mJ per shot onto the target. We estimate that on average an energy of 0.2 eV per atom is absorbed in the clusters, which can thus be considered cold. The data is shown on an absolute scale based on spectrometer efficiency and its spectral sensitivity.[13] The spectral bandwidth of 10 eV was chosen as it is close to the spectral resolution of the measurement (7.7 eV). The spectrum is fit using elastic scattering from bound electrons and an inelastic scattering contribution from ionization of M-shell electrons,[14] where the ratio of elastic to inelastic scattering of 215 is found in the data. There is some uncertainty on the exact FEL spectrum in the lower wings of the elastic scattering profile. Here we assumed exponential functions that are consistent with the slope at high elastic scattering signal levels. This assumption might explain the deviation of the data from the fit at energies above 1835 eV. The noise floor for this measurement is almost four orders of magnitude below the peak signal. Because of the near background-free measurement, the noise floor could be further lowered by increasing the number of shots used for signal integration.

VI. CONCLUSION

We have presented a proof-of-principle experiment to measure x ray Thomson scattering from a cluster jet target at LCLS. Background signals were successfully mitigated by selecting a narrow range of ADU values from the energy-dispersive pnCCD detector. Despite a very low count rate, we demonstrated the ability to measure high-dynamic range scattering spectra with a clear inelastic scattering component from Ar clusters. These results show great promise for future experiments that will use XRTS to study laser-driven ionization and relaxation dynamics in clusters with sub-100 fs time resolution.

ACKNOWLEDGMENTS

This material is based upon work supported by the NSF Graduate Research Fellowship Program under Grant No. 2013155705 and by the U.S. DOE Office of Science, Fusion Energy Science under FWP 100182. B.B., S.H.-R., T.P. and T.D worked under the auspices of the U.S. DOE by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and acknowledge support from Laboratory Directed Research and Development Grant No. PLS-15ERD-026. C.B. acknowledges support from the U.S. DOE, Office of Science, Office of Basic Energy Sciences, Division of Chemical, Geological, and Biological Sciences, under Contract No. DE-AC02-06CH11357.

[1] T. Ditmire, R. A. Smith, J. W. G. Tisch, and M. H. R. Hutchinson, Phys. Rev. Lett. **78**, 3121 (1997).
[2] T. Fennel, K.-H. Meiwes-Broer, J. Tiggesbäumker, P.-G. Reinhard, P. M. Dinh, and E. Suraud, Rev. Mod. Phys. **82**, 1793 (2010).
[3] T. Fennel, T. Döppner, J. Passig, C. Schaal, J. Tiggesbäumker, and K.-H. Meiwes-Broer, Phys. Rev. Lett. **98**, 143401 (2007).
[4] T. Döppner, T. Fennel, T. Diederich, J. Tiggesbäumker, and K. H. Meiwes-Broer, Phys. Rev. Lett. **94**, 013401 (2005).
[5] C. Bostedt et al., Phys. Rev. Lett. **108**, 093401 (2012).
[6] S. H. Glenzer and R. Redmer, Rev. Mod. Phys. **81**, 1625 (2009).
[7] L. Strüder et al., Nucl. Instrum. Methods Phys. Res., Sect. A **614**, 483 (2010).
[8] D. J. Bozek, Eur. Phys. J. **169**, 129 (2009).
[9] K. R. Ferguson et al., J. Synch. Rad. **22**, 492 (2015).
[10] P. Emma et al., Nat Photon **4**, 641 (2010).
[11] U. Even, Advances in Chemistry **2014**, 636042 (2014).
[12] M. Harmand, R. Coffee, M. R. Bionta, M. Chollet, D. French, D. Zhu, D. M. Fritz, H. T. Lemke, N. Medvedev, B. Ziaja, S. Toleikis, and M. Cammarata, Nat Photon **7**, 215 (2013).
[13] E. J. Gamboa et al., J. Inst. (In preparation).
[14] G. Gregori, S. H. Glenzer, F. J. Rogers, S. M. Pollaine, O. L. Landen, C. Blancard, G. Faussurier, P. Renaudin, S. Kuhlbrodt, and R. Redmer, Physics of Plasmas **11**, 2754 (2004).