SLAC-PUB-16807

High Resolution X-ray Thomson Scattering Measurements from Cryogenic Hydrogen Jets using the Linac Coherent Light Source[a)]

L. B. Fletcher,[1, b)] U. Zastrau,[2] E. Galtier,[1] E. J. Gamboa,[1] S. Goede,[1, 2] W. Schumaker,[1] A. Ravasio,[1, 3] M. Gauthier,[1] M. J. MacDonald,[1, 4] Z. Chen,[1, 5] E. Granados,[1] H. J. Lee,[1] A. Fry,[1] J. B. Kim,[1] C. Roedel,[1] R. Mishra,[1] A. Pelka,[6] D. Kraus,[7, 6] B. Barbrel,[7] T. Döppner,[8] and S. H. Glenzer[1]

[1)] *SLAC National Accelerator Laboratory, Menlo Park, CA, USA*
[2)] *European XFEL, Schenefeld, Germany*
[3)] *Laboratoire pour l'Utilisation des Lasers Intenses, Palaiseau Cedex, France*
[4)] *University of Michigan, Ann Arbor, MI, USA*
[5)] *University of Alberta, Edmonton, Alberta, Canada*
[6)] *Helmholtz Zentrum Dresden-Rossendorf, Dresden, Germany*
[7)] *University of California Berkeley, Department of Physics, Berkeley, CA, USA*
[8)] *Lawrence Livermore National Laboratory, Livermore, CA, USA*

(Dated: 20 June 2016)

We present the first spectrally resolved measurements of x-rays scattered from cryogenic hydrogen jets in the single photon counting limit. The 120 Hz capabilities of the LCLS, together with a novel hydrogen jet design[1], allow for the ability to record a near background free spectrum. Such high-dynamic-range x-ray scattering measurements enable a platform to study ultra-fast, laser-driven, heating dynamics of hydrogen plasmas. This measurement has been achieved using two Highly Annealed Pyrolytic Graphite (HAPG) crystal spectrometers to spectrally resolve 5.5 keV x-rays elastically and inelastically scattered from cryogenic hydrogen and focused on to Cornell-SLAC Pixel Array Detectors (CSPAD)[2].

I. INTRODUCTION

Accurate characterization of hydrogen from the strongly coupled cryogenic state to the high energy density (HED) plasma state is a crucial goal towards advancing our understanding of astrophysical objects[3] as well as achieving laser driven fusion[4]. The growing interest in understanding and predicting the properties of dense hydrogen plasmas necessitates the development of accurate in situ temperature and density probes. X-ray Thomson scattering (XRTS) is a powerful diagnostic that can be used to accurately characterize and measure the key properties of dense plasmas such as the temperature, density, and ionization[5]. The characteristics of the Linac Coherent Light Source (LCLS) have allowed for the development of dynamic, high-precision XRTS experiments to be performed with unprecedented resolution[6,7]. In this letter, we will demonstrate a platform that can be used to measure the properties of hydrogen through the use of spectrally resolved scattering of 5.5 keV x-rays detected by single-photon counting with the ability to reproduce the spectral distribution to a fitting precision of $\pm 1\%$ as compared to first-principle calculations.

A. X-ray Thomson scattering

X-ray Thomson scattering is the scattering of x-ray radiation by electrons in a material that are either free, weakly bound, or tightly bound to the ion. The total scattered x-ray spectrum demonstrates a unique intensity profile and spectral shape that depends on a combination of scattering contributions that are distinctly related to the temperature, density, and ionization state of the plasma[8,9]. In this paper, the hydrogen is probed in a cryogenic state, therefore all the scattering comes from bound and weakly bound electrons.

II. EXPERIMENT

The experimental setup can be observed in figure 1. Here a 5 μm diameter hydrogen jet is created by injecting liquid H_2 from a helium cooled cryostat into a vacuum chamber whereby the liquid will undergo evaporative cooling resulting in a liquid/solid cylindrical jet of pure hydrogen. Separately the jet is then probed with x-rays at photon energies of 5.5 keV produced from the Linac Coherent Light Source 5 mm away from the cryostat nozzle. The LCLS beam has a duration of $\sim$70 fs at a repetition rate of 120 Hz. Thus the collected scattered x-rays are gated by the time duration of the x-ray laser and accumulated for 80 seconds ($\approx$10,000 events).

Scattered x-rays from the jet are collected at fixed angles of θ=10° and θ=170°. Such a geometry allows for direct measurements of the static structure factor from elastically scattered x-rays in the forward direction[10] and can be properly normalized from inelastically scattered x-rays in the backward scattering geometry following the f-sum rules of particle conservation[11].

A. HAPG crystal spectrometer design

Figure 2 shows the details of the spectrometer design fielded inside the MEC target chamber. A 50 μm thick

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

[b)] Author to whom correspondence should be addressed: lbfletch@slac.stanford.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.

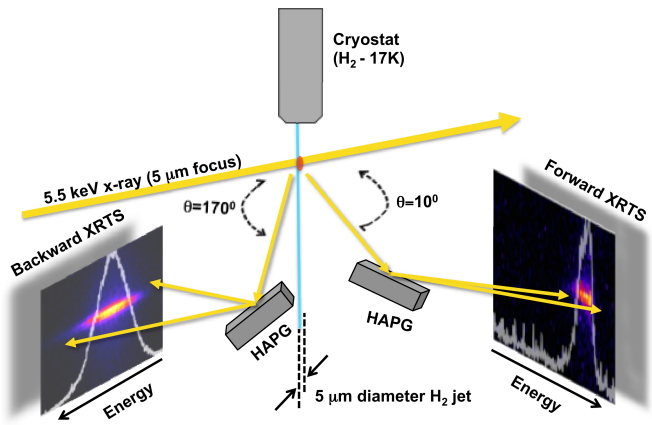

FIG. 1. Experimental configuration at the MEC (Matter in Extreme Conditions) end-station of the LCLS showing a 5.5 keV LCLS x-ray beam focused to 5 μm scattering from a 5 μm diameter H_2-jet. The scattered photons are captured on two x-ray crystal spectrometers; (left) Backward HAPG crystal spectrometer resolving inelastic Compton scattered x-rays at $\theta = 170°$; (right) Forward HAPG crystal spectrometer resolving elastically scattered x-rays at $\theta = 10°$.

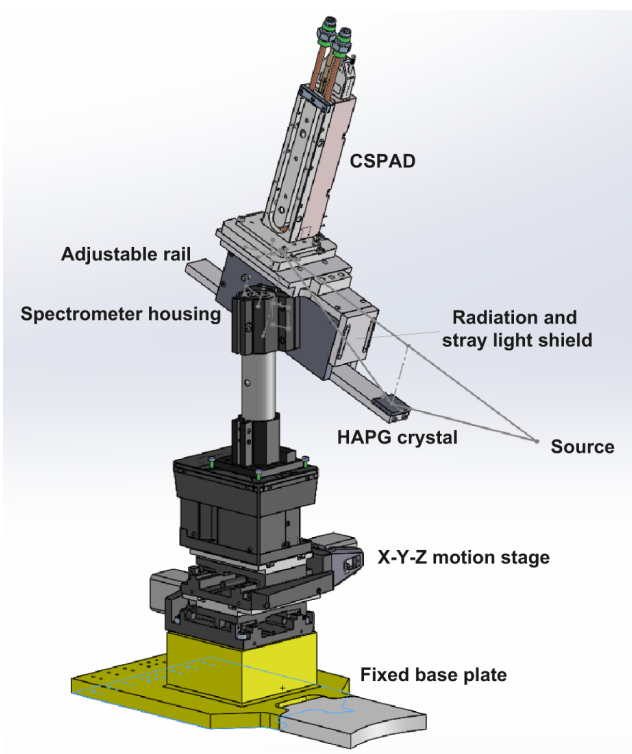

FIG. 2. Computer-aided design (CAD) model of the entire spectrometer setup and configuration.

HAPG crystal of dimensions 30x32 mm and a radius of curvature (ROC) of 77.5 mm is used to focus the scattered x-ray signal in the non-dispersive direction on to a CSPAD oriented in a Von Hamos geometry to increase the collection efficiency[12]. The spectrometer is a fixed mono-angle spectrometer with the crystal on an adjustable rail, that can be moved to cover x-ray energies from 7 keV (280 mm focal length) to 4 keV (150 mm focal length). For this experiment, measuring 5.5 keV x-rays, it is positioned at 217 mm focusing distance. The spectral dispersion follows directly from the geometry, and is calculated to be ∼1.22 eV/100 μm pixel at 5.5 keV.

III. RESULTS AND ANALYSIS

Due to the low scattering cross section of hydrogen atoms, small collection angle, and inherent inefficiencies involved with crystal spectrometers, only one photon can be collected per event necessitating the need for ultrabright x-rays sources that operate at high repetition rates.

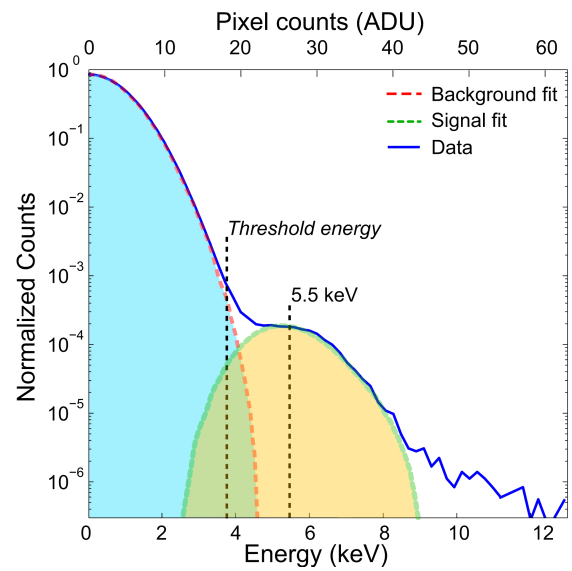

FIG. 3. A normalized histogram of the total accumulated x-ray signal. The histogram demonstrates two Lorentzian distributions that contribute to the measured signal. (light blue) Distribution of the background. (light orange) Distribution of the scattered 5.5 keV x-ray signal.

A detailed analysis of figure 3 shows that one scattering photon at 5.5 keV generates approximately 27 pixel counts on the detector. A threshold value of 18 pixel counts can be inferred from the histogram by choosing a value that minimizes the signal contribution from background (dark current) while also maximizing the contribution from the scattered signal. A comparison between Figure 4a and Figure 4b demonstrates that with the proper thresholding value one can separate the photons collected from the background with those that are scattered from the hydrogen, over an 80 second accumulation period, resulting in the ability to measure a near background free spectrum with a signal to noise (S/N) improvement of 20× at $\theta = 10°$ and 250× at $\theta = 170°$.

Figure 5 demonstrates the integrated XRTS spectrum from the forward (k=0.5 Å^{-1}) and backward (k=5.5 Å^{-1}) scattering geometries. The results are compared to first-principle calculations of the bound-free inelastic contribution using Impulse Approximation[13] and an average source function fit that is modeled using a non-linear optimization method to decompose the overlapping x-ray laser signals into its component parts. Impulse Approximation assumes that the electrons, which contribute to

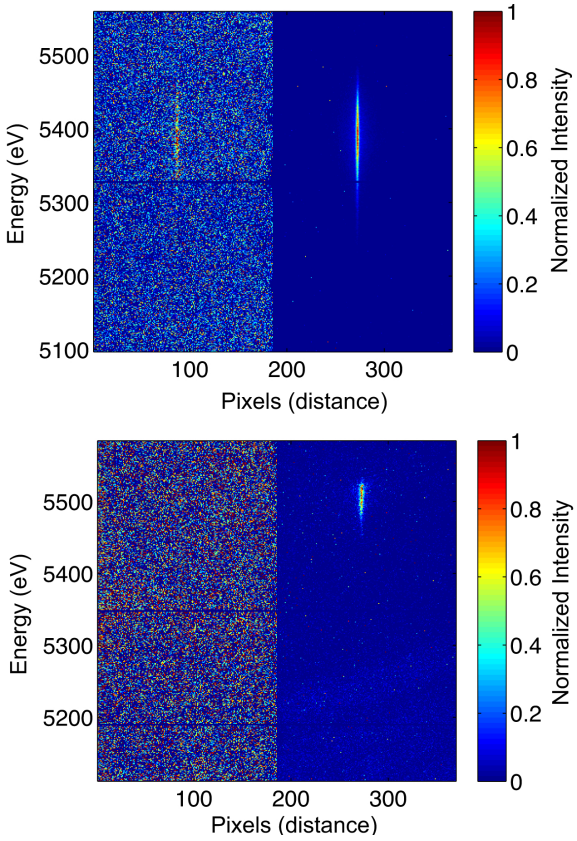

FIG. 4. Raw integrated spectrum of x-rays focused on the CSPAD. (top) Backward scattering geometry with and without thresholding.(bottom) Forward scattering geometry with and without thresholding. Including only the counts which exceed the threshold increases the S/N ratio of 20× for $\theta = 10^\circ$ and 250× for $\theta = 170^\circ$.

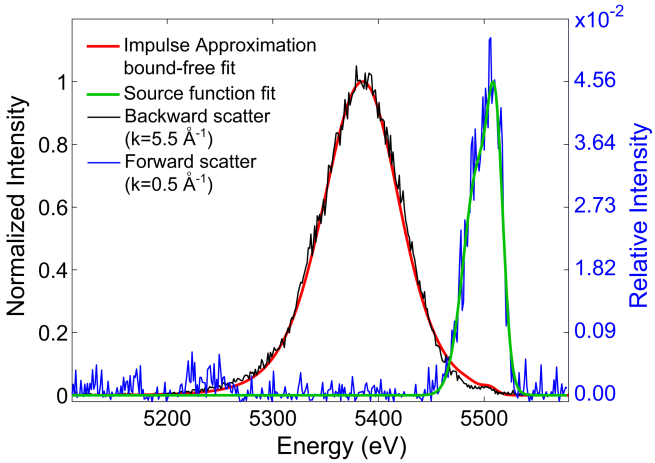

FIG. 5. The total measured scattered x-ray signal, spectrally resolved after single photon thresholding is applied to the analysis. (blue) Forward elastically scattered x-rays at k=0.5 Å^{-1}. (green) Source function fit. (black) Inelastically scattered x-rays in the backward k=5.5 Å^{-1} geometry. (red) Impulse Approximation bound-free calculations/fit.

the scattering, may be treated as free rather than bound. The model uses plane wave hydrogenic wavefunctions to calculate the momentum distribution where the energy difference from the initial and final states approaches zero using a constant instantaneous potential.

There is considerable agreement with the measured bound-free spectrum and the Impulse Approximation model. A fitting error of ±1% is determined by the calculated RMS (root mean squared) of the pairwise differences between the noise in the data compared to the fit of the theoretical dynamic structure factor. Monitoring this distribution as the temperature and density of the hydrogen plasma increases, one can accurately measure the ionization state of the plasma, as the electrons will transition from a bound state to a free state resulting in a distribution that is directly related to the density and temperature of the system[14], changing both the width and the shape.

IV. CONCLUSIONS

In this report we show that in the single photon counting limit, accurate measurements of x-ray scattering from hydrogen can be made with high signal to noise. The results from this study have been compared to first-principle calculations and demonstrate a reproducible spectrum with a fitting error of ±1% that can be achieved from 120 Hz high-rep rate data accumulation. This high-dynamic-range x-ray scattering measurement can be used as a platform to study ultra-fast, laser-driven, high temperature heating dynamics of hydrogen plasmas in the collective and non-collective scattering regime using an ultrabright x-ray source.

ACKNOWLEDGMENTS

This work was performed at the Matter at Extreme Conditions (MEC) instrument of LCLS, supported by the DOE Office of Science, Fusion Energy Science under contract No. SF00515 and supported under FWP 100182 and DOE Office of Basic Energy Sciences, Materials Sciences and Engineering Division, contract DE-AC02-76SF00515.

[1] J. B. Kim, S. Goede, and S. H. Glenzer, Rev Sci. Inst. (2016), to be published.
[2] S. Herrmann *et al.*, Nucl. Instrum. Methods A **718**, 550 (2013).
[3] T. Guillot, Science **286**, 72 (1999).
[4] J. D. Lindl *et al.*, Phys. of Plasmas **11**, 339 (2004).
[5] S. H. Glenzer and R. Redmer, Rev. of Mod. Phys. **81**, 1625 (2009).
[6] L. Fletcher *et al.*, Nat. Phot. **9**, 274 (2015).
[7] S. H. Glenzer *et al.*, J. of Phys. B **49**, 092001 (2016).
[8] J. Chihara, J. of Phys. F **17**, 295 (1987).
[9] J. Chihara, J. of Phys.: Condensed Matter **12**, 231 (2000).
[10] D. O. Gericke, J. Vorberger, K. Wünsch, and G. Gregori, Phys. Rev. E **81**, 065401 (2010).
[11] P. Nozières and D. Pines, *The theory of quantum liquids* (Perseus, 1999).
[12] U. Zastrau, A. Woldegeorgis, E. Förster, R. Loetzsch, H. Marschner, and I. Uschmann, JINST **8**, P10006 (2013).
[13] P. Eisenberger and P. Platzman, Phys. Rev. A **2**, 415 (1970).
[14] O. Landen, S. Glenzer, M. Edwards, R. Lee, G. Collins, R. Cauble, W. Hsing, and B. Hammel, J. of Quant. Specros. and Radia. Transfer **71**, 465 (2001).