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XUV Frequency Comb Development for Precision Spectroscopy and Ultrafast Science

R Jason Jones  
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07/28/2015  
Final Report

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This program will explore novel experimental approaches, aided by numerical simulations, to advance the science and technology needed to develop a next-generation XUV frequency comb. The overall objective will be the development of a coherent source capable of making a significant impact in precision spectroscopy and ultrafast science from the IR to the XUV. A new technique for highly sensitive measurements of optical nonlinearities and ionization dynamics is also proposed which utilizes the resonant response of the fsEC itself. This technique will enable the investigation of ways to control the ionization levels inside the fsEC. Furthermore, we propose to develop a coherent dual-comb XUV system. Dual-comb spectroscopy incorporates a second "local oscillator" frequency comb to enable heterodyne detection of individual comb components. We show that sufficient XUV power levels can now be achieved to enable the extension of this approach for the first time to the XUV. The dual-comb system will enable a systematic study of these noise properties in the XUV for the first time and provide insight to the underlying

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## Final Report

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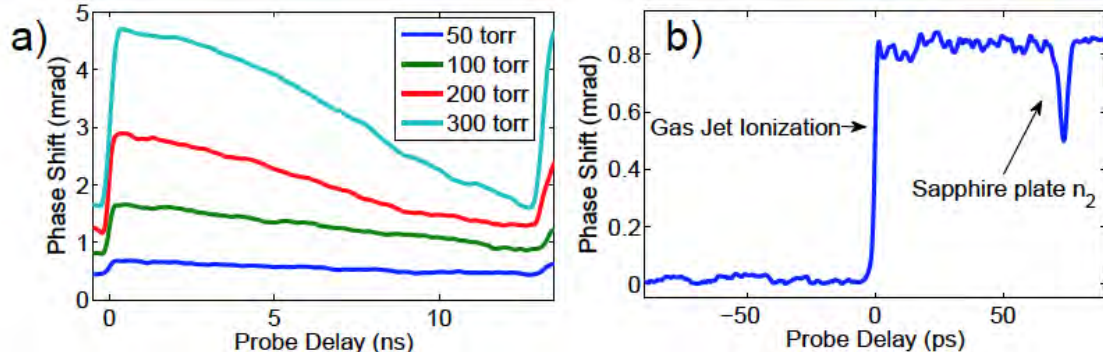
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The primary goals of this grant were the development of next generation fs enhancement cavities for intracavity HHG (iHHG) and the first demonstration of a coherent dual-frequency comb system in the VUV/XUV. During this grant period we have completed most of the key milestones. Much of our initial effort had been on the construction and testing of our next generation dual-frequency comb high power IR source and passively isolated xuv vacuum chamber, which is now completed. The source consists of 2 home-built and phase coherent ~50 W, ~100fs Yb fiber frequency combs. The mutual phase stabilization between both combs and the fs enhancement cavity (fsEC) is achieved using optical phase locks with two common cw lasers (at 1064nm and 1052nm). This system serves as the driving laser source for our work on intracavity high harmonic generation with fs frequency combs. Due to the phase coherence of the dual-comb source, we can now resolve individual frequency comb components in the IR. The next step was to observe and characterize *individual* fs comb components of the high harmonic light in the VUV and XUV, which to date had *not* been demonstrated. This would enable a simple and robust approach to direct frequency comb spectroscopy in the VUV and XUV. A very significant and recent achievement that enabled this is the coupling of 2 independent fs comb pulse trains into a *single* fsEC for the first time. This has enabled us to now demonstrate (i) measurements of ionization dynamics with a continuously and precisely controlled intracavity pump-probe delay and (ii) the generation of spatially overlapped dual-comb high harmonic pulse trains that have been used for the *first* demonstration of dual-comb detection of individual comb components generated from intracavity high harmonic generation. Finally, numerical simulations provided valuable insight to the limitations caused by intra-cavity ionization dynamics.

### *Dual-comb fsEC measurements of extreme optical nonlinearities.*

Our experimental and theoretical work highlights the significant role photo-ionization plays in the performance of iHHG. In addition to affecting the phase matching conditions, the electron plasma generated by the high intensity circulating pump pulse places a fundamental limitation on the maximum intensity available for HHG. In the course of this grant period we demonstrated a novel pump-probe technique for monitoring intracavity plasma dynamics. The basic idea is that with a strong “pump” laser locked to the fsEC, a time delayed probe pulse (injected in either the same or in a counter-propagating direction) can be used to detect the plasma-induced phase shift resulting from the ionization of the gas target by the pump. With the current dual-comb system coupled to a single fsEC, we can now

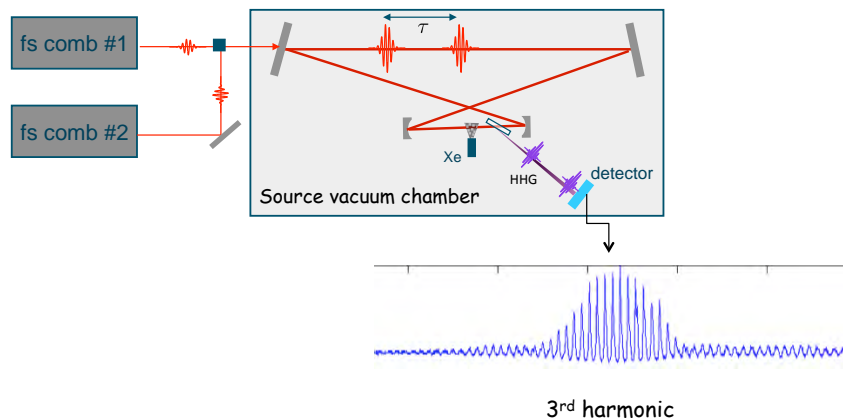
precisely adjust the delay between the pump (fs comb #1) and probe pulse (fs comb #2) from fs timescales up to the maximum round trip time of  $\sim 13$  ns, enabling a near “real-time” picture of the evolving plasma density seen by the probe beam. In Fig. 1 we show the results from one such measurement taken using xenon gas. In this experiment the pump and probe pulses were *counter-propagating* inside the fsEC. The shift of the fsEC due to the plasma produced from the pump pulse is monitored by the error signal from the probe beam. The error is proportional to the shift of the fsEC frequency, which in turn is proportional to the nonlinear phase acquired by the probe pulse each round trip. This phase is then proportional to the plasma density. One can clearly see in Fig. 1a the intracavity plasma dynamics as the pump pulse ionizes the gas target each round trip, followed by the plasma decay before the pump again arrives at the gas target. The decay mechanisms of the plasma (spatial evolution and possible recombination) and conditions that may affect its overall levels inside the fsEC are of great interest. In Fig 1b we show a zoomed in region in which the nonlinear response of both the xenon target and the sapphire plate (used for output coupling of the VUV/XUV) is observed. Through this new approach we can observe such dynamics directly for the first time and with a very high signal-to-noise, timing resolution, and phase sensitivity. This approach is dramatically more sensitive than standard interferometric techniques for measuring optical nonlinearities due to the resonant nature of the fsEC. This work has been published in multiple conference proceedings. A published version of the work is expected to be accepted for publication in summer 2015.



**Figure 1. Intra-cavity measurement of ionization dynamics using the dual-comb system (a), and a zoomed in region showing the nonlinear response due to both the gas jet ionization and the sapphire plate (b).**

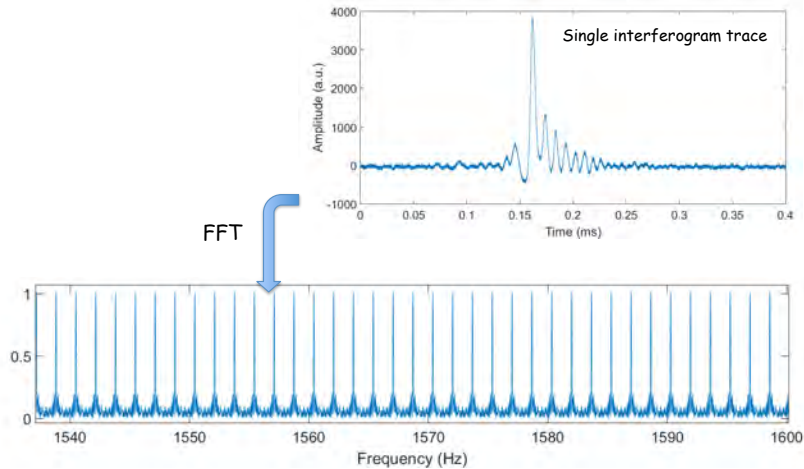
### *Dual-comb intracavity HHG*

With the current dual-comb system coupled to a single fsEC, we can now controllably and precisely tune the delay between two intracavity pulses from zero up to the maximum round trip time in the fsEC without a mechanical delay arm. Due to the optical phase locking between both laser systems and the fsEC, the relative delay between intracavity pulses can be fixed, and/or scanned through the entire delay range at a rate of up to  $\sim 10$  Hz (the maximum difference in repetition rates that the fsEC can support). Fig. 2 shows a schematic of the optical layout and an intracavity cross-correlation for the 3<sup>rd</sup> harmonic UV beams. This system enables dual-comb spectroscopy using any higher order



**Figure 2. Intra-cavity cross correlation of the 3d harmonic from each fs comb.**

harmonic. In a more general sense, it enables collinear VUV/XUV pulse pairs to be delivered with attosecond timing control, useful for not only spectroscopy but time resolved studies as well. *By recording this interferometric cross-correlation in a phase-coherent manner for longer time scales, we are able to resolve individual comb components generated from iHHG for the first time (see Fig. 3).* This should be easily extended to higher harmonics in the near future, enabling robust and precision spectroscopy from the UV to the XUV using direct comb spectroscopy. These results have also been presented and published in the proceedings of multiple conferences. We expect acceptance of peer-reviewed results towards the end of summer 2015.



**Figure 3. Top plot shows a single cross-correlation interferogram between the 3<sup>rd</sup> harmonic beams. The lower figure shows a zoomed in picture of the resulting Fourier transform, demonstrating detection of individual comb components generated from iHHG>**

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### Program Manager

The AFOSR Program Manager currently assigned to the award

Enrique Parra

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## Abstract

The primary goals of this grant were the development of next generation fs enhancement cavities for intracavity HHG (iHHG) and the first demonstration of a coherent dual-frequency comb system in the VUV/XUV. During this grant period we have completed most of the key milestones. Much of our initial effort had been on the construction and testing of our next generation dual-frequency comb high power IR source and passively isolated xuv vacuum chamber, which is now completed. The source consists of 2 home-built and phase coherent ~50 W, ~100fs Yb fiber frequency combs. The mutual phase stabilization between both combs and the fs enhancement cavity (fsEC) is achieved using optical phase locks with two common cw lasers (at 1064nm and 1052nm). This system serves as the driving laser source for our work on intracavity high harmonic generation with fs frequency combs. Due to the phase coherence of the dual-comb source, we can now resolve individual frequency comb components in the IR. The next step was to observe and characterize individual fs comb components of the high harmonic light in the VUV and XUV, which to date had not been demonstrated. This would enable a simple and robust approach to direct frequency comb spectroscopy in the VUV and XUV. A very significant and recent achievement that enabled this is the coupling of 2 independent fs comb pulse trains into a single fsEC for the first time. This has enabled us to now demonstrate (i) measurements of ionization dynamics with a continuously and precisely controlled intracavity pump-probe delay and (ii) the generation of spatially overlapped dual-comb high harmonic pulse trains that have been used for the first demonstration of dual-comb detection of individual comb components generated from intracavity high harmonic generation. Finally, numerical simulations provided valuable insight to the limitations caused by intra-cavity ionization dynamics.

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### Archival Publications (published) during reporting period:

David Carlson and R. Jason Jones, "Pump-Probe Intracavity Phase Spectroscopy," *Frontiers in Optics (FiO)*, OSA, Rochester NY. Paper FW1B.3 (2012).

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