



Ultrafast Imaging of Electronic Motion in Atoms and Molecules

**Martin Centurion
UNIVERSITY OF NEBRASKA**

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Final Report**

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Martin Centurion

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The AFOSR Program Manager currently assigned to the award

Enrique Parra

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Abstract

This project had both an experimental and theoretical component. In the experimental effort, a new setup was designed and constructed to deliver femtosecond electron pulses for scattering and diffraction experiments on a gaseous target of atoms or molecules. An optical setup was designed and constructed to compensate for the blurring of the temporal resolution due to the velocity mismatch between the laser and electrons, which we showed can be reduced to less than 100 fs. The charge and duration of the electron pulses were measured with a home-made faraday cup and laser-triggered streak camera, respectively. Both are retractable and can measure the beam in-situ. The gun was shown to generate pulses with more than a million electrons per pulse. The pulse duration was compressed from 20 ps to 700 fs, as measured with the streak camera. Active stabilization was implemented on the laser repetition rate and beam pointing to reduce the timing jitter. In the theory component of the project, we have calculated the distortion of a helium atom in an intense laser field in support of planned

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experimental measurements. We have developed a new theory for ultrafast electron diffraction from a time-varying coherent electronic state of a target atom or molecule that accounts for inelastic processes occurring within the bandwidth of the incident electron pulse. Finally, we have analyzed energy-resolved ultrafast electron diffraction from a time-varying coherent electronic target state. Simulations were carried out in all cases to illustrate the key points of our new theory and of the advantages of energy-resolved ultrafast electron diffraction.

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H.-C. Shao and A.F. Starace, "Use of Attosecond Electron Pulses to Image Electronic Motion in Atoms and Molecules," in Ultrafast Imaging and Spectroscopy, ed. Zhiwen Liu, Proceedings of SPIE Vol. 8845, 884504 (28 September 2013); doi:10.1117/12.2023522

H.-C. Shao and A.F. Starace, "Imaging Coherent Electronic Motion in Atoms by Ultrafast Electron Diffraction," Phys. Rev. A 88, 062711 (2013); DOI: 10.1103/PhysRevA.88.062711 This article has been selected by the editors of Physical Review A as an "Editor's Suggestion."

H.-C. Shao and A.F. Starace, "Imaging Electronic Motions in Atoms by Energy-Resolved Ultrafast Electron Diffraction," Phys. Rev. A 90, 032710 (2014). DOI: 10.1103/PhysRevA.90.032710

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6 Month extension granted

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Program Officer

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Abstract

This project had both an experimental and theoretical component. In the experimental effort, a new setup was designed and constructed to deliver femtosecond electron pulses for scattering and diffraction experiments on a gaseous target of atoms or molecules. An optical setup was designed and constructed to compensate for the blurring of the temporal resolution due to the velocity mismatch between the laser and electrons, which we showed can be reduced to less than 100 fs. The charge and duration of the electron pulses were measured with a home-made faraday cup and laser-triggered streak camera, respectively. Both are retractable and can measure the beam in-situ. The gun was shown to generate pulses with more than a million electrons per pulse. The pulse duration was compressed from 20 ps to 700 fs, as measured with the streak camera. Active stabilization was implemented on the laser repetition rate and beam pointing to reduce the timing jitter. In the theory component of the project, we have calculated the distortion of a helium atom in an intense laser field in support of planned experimental measurements. We have developed a new theory for ultrafast electron diffraction from a time-varying coherent electronic state of a target atom or molecule that accounts for inelastic processes occurring within the bandwidth of the incident electron pulse. Finally, we have analyzed energy-resolved ultrafast electron diffraction from a time-varying coherent electronic target state. Simulations were carried out in all cases to illustrate the key points of our new theory and of the advantages of energy-resolved ultrafast electron diffraction.

Accomplishments

Experimental component (Martin Centurion, P.I.)

We have designed, constructed and tested an electron source to deliver femtosecond electron pulses on an atomic or molecular target for time-resolved scattering and diffraction experiments. The setup comprises a laser triggered photocathode with DC acceleration, a beam transport line with two magnetic lenses, a resonant radio-frequency (RF) cavity for bunch compression, an experimental chamber that houses a laser-triggered streak camera and a detection chamber that houses a faraday cup and a phosphor screen that is imaged onto a CCD camera. We first describe each component, and then the results of the characterization.

I. Experimental setup

Electron pulse generation. The electron pulses are generated by shining a UV laser pulse on a solid photocathode. The UV pulse is generated using a home-made frequency tripler (from 800 nm to 267 nm). The electrons are accelerated in a static field to kinetic energies between 90 keV and 100 keV.

Beam transport. The beam is collimated and focused using two magnetic lenses, one immediately after the anode and a second further downstream (the position can be adjusted). Both lenses are solenoids outside the tube that transports the beam in vacuum.

Pulse compression. A temperature stabilized RF cavity is used to compress the electron pulses at the target position. The cavity has a longitudinal field that changes direction as the electron pulse traverses the cavity. The effect of the cavity is to reverse the velocity distribution of the pulse, accelerating the electrons in the trailing edge and slowing down the electrons in the front. After exiting the cavity, the pulse will self-compress. The distance to the temporal focus is determined by the magnitude of the electric field in the cavity. The cavity fields need to be synchronized with the laser pulses.

Synchronization. The synchronization is achieved using a synchronizer that takes a 75 MHz electronic signal from the femtosecond laser oscillator as the input. The input signal is multiplied by 40x to operate at a standard frequency of 3 GHz, where there are standard microwave components readily available. A signal power between 50 W and 200 W is delivered to the cavity. In order to fit the resonance of the cavity, the repetition rate of the laser was changed to 75 MHz. The repetition rate is actively stabilized with a feedback loop, using a frequency counter and a piezo mirror inside the laser cavity. The laser repetition rate is stable to better than 10 Hz RMS, which is sufficient to stay at the peak of the cavity resonance that has a width on the order of 100 kHz.

Laser system. The laser pulses from the oscillator are amplified to an energy of 2 mJ at a repetition rate of 5 kHz, duration of 50 fs and wavelength of 800 nm. A small fraction of the total pulse energy (about 0.2 mJ) is split to trigger the photocathode, after going through a third harmonic generator.

Velocity mismatch (VM) compensation. An optical setup was implemented to compensate for the velocity mismatch between the laser and electron pulses. At an energy of 100 keV, the electrons travel with roughly half the speed of light. The result is that as the electrons and laser traverse a sample, the time delay between them is not constant, which blurs the resolution of the experiments. Even for thin targets of less than 0.5 mm, VM will degrade the resolution to several picoseconds. We have shown that this can be compensated for a target as thick as 1 mm while keeping effect of VM to less than 100 fs, by using a laser pulse with a tilted intensity front. The laser pulse propagates at an angle to the electron beam, such that the longitudinal velocity of both pulses is match. Then, the intensity front of the laser is tilted such that it is parallel to the pulse front of the electrons. This can be achieved by reflecting the laser pulse from a diffractive element (a custom grating in this case) and then imaging the grating surface at the target position. The matching is only perfect in one plane, but the depth of focus can be designed such that there is a fairly good match throughout the target.

Streak camera. A miniaturized streak camera was built to measure the pulse duration. A parallel plate capacitor (with spacing of 0.5 mm) is charged to a potential difference of 2 kV. The capacitor is charged with a home-made pulsed high voltage power supply to

achieve a high electric field. The capacitor is discharged using a laser-triggered GaAs photoswitch. The small size of the streak camera results in a rapid oscillation of the field upon discharge (it behaves as an RLC circuit). The rapidly changing electric field allows us to measure the electron pulse duration with a resolution of 300 fs.

II. Measurements

Bunch charge. The charge in each pulse was measured using a home-made retractable Faraday cup that can be moved in and out of the beam path, directly in front of the phosphor screen detector. Figure 1 shows the results of the charge per pulse, as a function of the power of the UV laser beam incident on the photocathode. The data shows that more than a million electrons per pulse can be extracted with the available UV power.

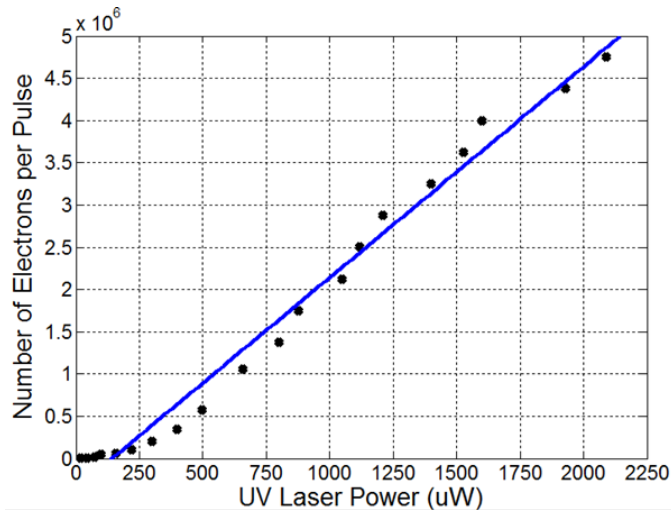


Figure 1. Number of electrons per pulse vs average power of the UV laser beam.

Laser stabilization. The stability of the laser beam is crucial for achieving a high temporal resolution. The repetition rate of the laser oscillator was fixed at 74.9976 MHz to match the value of the resonance of the RF cavity after the signal is multiplied by a factor of 40. The repetition rate is actively stabilized using a feedback loop with a frequency counter and a piezo mirror inside the laser cavity. Figure 2 shows the result of the stabilized laser repetition rate. The repetition rate can be fixed with an RMS error less than 10 Hz. The beam pointing of the amplified laser pulses was also actively stabilized to reduce the pointing jitter. A pointing correction system was built with two actuated mirrors and two CCD cameras. Figure 3 shows the results, where the pointing drift was removed and only a pointing jitter of around 10 microradians remains. This jitter is too fast to fix with actuated mirrors, and most likely reflects the shot to shot jitter of the laser.

Velocity mismatch. In order to test the performance of the laser pulse tilting optical setup, the tilt angle and pulse duration were measured as a function of position. Figure 4 shows the results of the measurements. The values were measured in two different ways, using an interferometric setup and using second harmonic generation. The measurement is described in more details in a publication [P. Zhang, J. Yang, M. Centurion, NJP 16 083008 (2014)]. The optimal tilt angle is 56.7 degrees, and a mismatch of a few degrees does not significantly affect the velocity matching. The measurement shows that the angle does not change significantly over a distance of several millimeters. The pulse duration in principle only reproduces the original pulse duration at the image plane, and increases away from this plane. The measurement shows that the pulse duration stays below 100 fs for a distance of about 1 mm, and below 200 fs for a distance of about 2 mm. This is more than sufficient for a gas target for scattering experiments, which typically has a diameter of less than 0.5 mm.

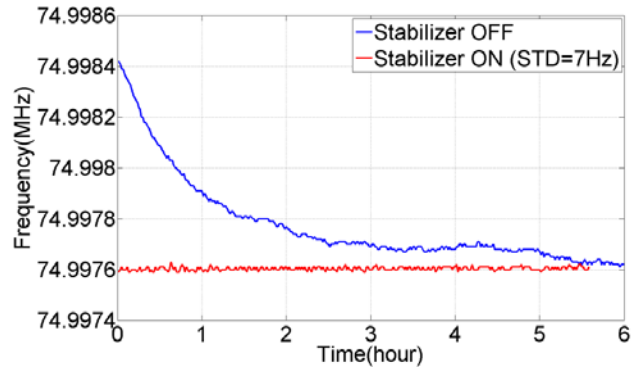


Figure 2. Active stabilization of the femtosecond oscillator repetition rate.

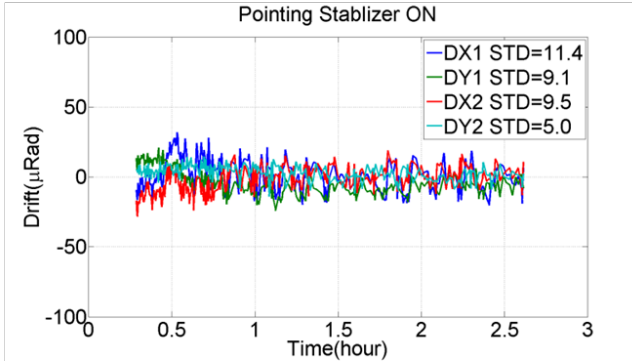
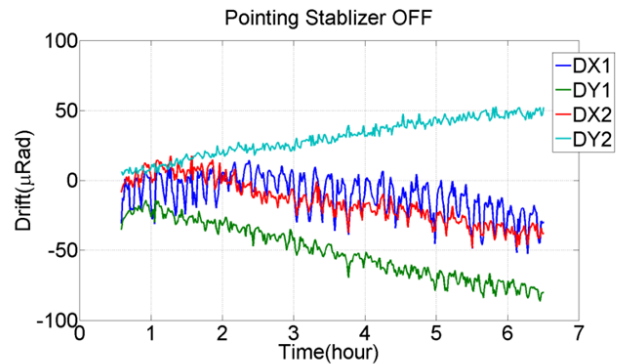


Figure 3. Beam pointing stabilization.

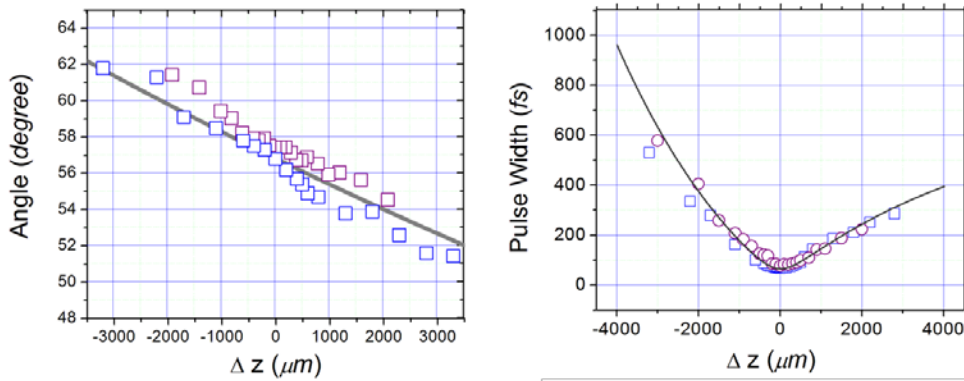


Figure 4. Tilt angle and pulse duration produced by the pulse tilting setup. The initial duration of the laser pulse in this experiment was 60 fs. The red points are the results of the interferometric measurement, and the blue squares are the result of a second harmonic generation measurement. The black line is the theoretical prediction.

Pulse duration. The pulse duration was measured using the streak camera. The discharge frequency of the streak camera was measured to be 2.6 GHz, sufficient for a resolution of 300 fs, which is better than most commercial streak cameras. Figure 5 shows the pulse duration measured for electron pulses containing 2×10^5 electrons/pulse. In order to find the minimum pulse duration, both the amplitude and phase of the electric field in the RF cavity need to be optimized. The minimum pulse duration achieved so far was 750 fs, while the duration of the uncompressed beam was about 20 ps.

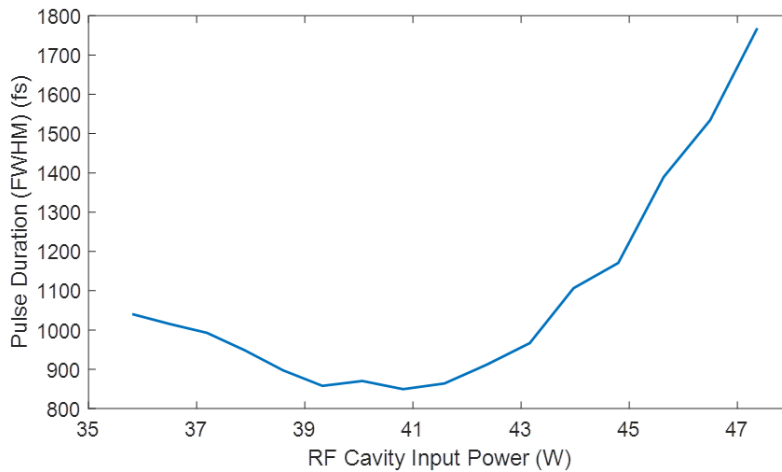


Figure 5. A sample scan of pulse duration vs RF power into the cavity. The phase of the cavity needs to be optimized as well to reach the minimum pulse duration.

Theoretical component (Anthony F. Starace, co-P.I.):

The distortion of the He ground state charge distribution in a Ti:sapphire laser field of intensity 10^{13} W/cm², as exhibited in the elastic scattering cross section for a 25 keV electron beam, was calculated to be about 6% smaller for small momentum transfers.

We have developed a new theory appropriate for scattering of a coherent electron beam from a time-varying electronic state that includes the role of inelastic scattering processes. We consider pump-probe processes in which a laser pulse creates a coherent superposition of target states that are probed by incident electron pulses. Varying the pump-probe delay time, the delay-dependent scattering intensities record the ensuing electronic motions. The key idea for describing the time-dependent scattering is to use a coherent wave function comprising the wave packets of both the projectile electron and the target. The latter are localized in space and time, so scattering events can be defined and analyzed properly. The scattering intensities, having the target information, are obtained by following the development of the wave packets. Our results for a 10 keV 100 attoseconds (FWHM) electron pulse diffracted by a coherent superposition of 3p+4p states of the H atom show that inelastic processes are significant for small scattering angles. However, the time variation of the total diffraction signal (including both elastically and inelastically scattered electrons) is found to remain significant, indicating the ability of ultrafast electron diffraction to detect electron motion in atomic and molecular targets. This work has been published [H.-C. Shao and A.F. Starace, "Imaging Coherent Electronic Motion in Atoms by Ultrafast Electron Diffraction," *Phys. Rev. A* **88**, 062711 (2013); DOI: 10.1103/PhysRevA.88.062711] and was selected by the editors of *Physical Review A* as an "Editor's Selection."

We have also developed a time-dependent scattering theory for energy-resolved ultrafast (attosecond) electron diffraction. This work shows that by energy-resolving particular inelastic transitions one can obtain valuable information on time-varying target electronic states. This work has been published [H.-C. Shao and A.F. Starace, "Imaging Electronic Motions in Atoms by Energy-Resolved Ultrafast Electron Diffraction," *Phys. Rev. A* **90**, 032710 (2014). DOI: 10.1103/PhysRevA.90.032710]. It was also the subject of an invited talk presented on 12 December 2015 at the 14th International Symposium on Ultrafast Intense Laser Science, 9-13 December 2015, Kauai, HI.

Archival publications during reporting period:

P. Zhang, J. Yang and M. Centurion, "Tilted femtosecond pulses for velocity matching in gas-phase ultrafast electron diffraction," *New J. Phys.* **16** 083008 (2014). DOI:10.1088/1367-2630/16/8/083008

H.-C. Shao and A.F. Starace, "Use of Attosecond Electron Pulses to Image Electronic Motion in Atoms and Molecules," in *Ultrafast Imaging and Spectroscopy*, ed. Zhiwen Liu, Proceedings of SPIE Vol. **8845**, 884504 (28 September 2013);

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H.-C. Shao and A.F. Starace, “Imaging Coherent Electronic Motion in Atoms by Ultrafast Electron Diffraction,” *Phys. Rev. A* **88**, 062711 (2013); DOI: 10.1103/PhysRevA.88.062711 *This article has been selected by the editors of Physical Review A as an “Editor’s Suggestion.”*

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Change in AFOSR program manager: None

Extensions granted or milestones slipped: We have received a 6-month extension.

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