ABSTRACT

Coherent Control of Atoms and Molecules

by

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Whenever scientific tools are significantly advanced, new discoveries soon follow. My thesis work relies on advances in ultrafast laser technology that allow precise control over the shape of sub 20-fs laser pulses. This laser system enabled many new experiments that yielded significant advances in attosecond science, atomic physics, extreme nonlinear optics, quantum chemistry, coherent control, molecular physics, ultrafast optical phase modulation, EUV coherent imagines, and EUV radiation sources.

By using an automated learning control algorithm coupled to our deformable mirror pulse shaper, we have demonstrated the control of a process on an attosecond time-scale for the first time. We demonstrate the manipulation of the phase of an electron wave packet, allowing us to tailor the EUV spectrum generated by high-order harmonic generation. These optimal fields were discovered by a learning algorithm that both found an unknown optimal solution, but also discovered an unknown phase-matching mechanism that occurs in the interaction of a single atom and light pulse.

We control molecular vibrational and rotational coherences with the same learning algorithm, demonstrating selective control in room temperature molecules at atmospheric pressures. Moreover, we demonstrate that modifications to the algorithm can help automatically uncover the physics of the control mechanism found by the learning algorithm.

We also demonstrate a new pulse-compression technique using rotational phase modulation, where optical pulses were compressed by an order of magnitude after phase modulation by a molecular rotational coherence and subsequent propagation

through a transparent window. This technique will enable efficient pulse compression in the deep-UV.

Finally, we measure the coherence of EUV light generated by high-harmonic upconversion of a femtosecond laser. In phase-matched hollow-fiber geometry, the EUV light exhibits the highest inherent spatial coherence of any source in this region of the spectrum. We use this source to demonstrate the coherent image with first table-top EUV radiation. Since this source exhibits full spatial coherence at very short wavelength, this light source represents has smallest inherent effective source-size of any light source yet created. We also demonstrate that the spectrum of an optical field can be determined by measuring the interference of a double-slit pattern.