CHAPTER VI

Summary

Controlling the dynamics of atoms and molecules gives us new abilities to manipulate matter. This work was enabled by new ultrafast laser technology and the lessoned learned by 30 years of attempts at using coherent light to control atoms and molecules. It has only been in recent years that very successful coherent control experiments have emerged. My work has demonstrated that very high-order nonlinear systems can not only be controlled, but that we can understand the control mechanisms involved. By selectively optimizing a single harmonic order in the HHG spectrum, the learning algorithm found an unexpected solution. This new solution taught us about a new phase-matching mechanism that occurs in extreme nonlinear optics. This demonstrates that a learning algorithm can non-only control a very high-order nonlinear system, but it can also teach us new science.

The work presented in this thesis is an initial contribution to what will, in my view, become a widely applied approach to the manipulation of matter. As new applications for coherently controlled atoms and molecules are found, interest in this field will grow. This new ability to manipulate matter will likely create new field of technology in the future.

Control over HHG, and in particular, sculpting a quasi-monocromatic HHG spectrum is directly applicable to time-resolved coherent EUV imaging applications and monitoring chemical reactions on a surface [152]. Furthermore, the increase in conversion efficiency makes this source more practical for applications that require a large photon flux.

Controlled molecular rotational wave packets have, in this thesis, been shown enable a new pulse compression technology that promises to provide sub 5 fs laser pulses in the deep-UV, which are particularly interesting for studying excited-state molecular dynamics.

Furthermore, the molecular control experiments I performed were done nominally at STP conditions, and is a promising avenue for laser-catalyst chemistry with macroscopic yields.

6.1 Towards EUV radiation for table-top coherent imaging

This thesis described work that separately showed both the control over the HHG spectrum by tailoring the driving pulse shape, and the use of HHG radiation for coherent imaging applications. One solution to the shaping of the HHG spectrum demonstrated the selection of a single harmonic order, creating a nearly monochromatic EUV spectrum. A monochromatic spectrum is desirable for coherent imaging applications since a broad spectrum can average out observed fringes, and reducing resolution. Applying the shaped HHG spectrum to coherent imaging will provide greater resolution for HHG microscopy experiments.

Gabor holography has limited applicability for imaging with a relatively low-resolution CCD camera. To improve the resolution, other imaging schemes should be explored. The most straightforward improvement of resolution will be to magnify the hologram with an optical component placed between the object and the CCD camera. We are currently constructing an off-axis reflective Swarzchid object for this purpose. However, each optical element offers a reflectivity of ~ 30 %, so the magnification comes at a high cost in terms of photons. Furthermore, the aberrations of this objective are very sensitive to alignment.

Other approaches depend on the type of object to be imaged. For the case of mostly opaque objects, imaging can be accomplished with phase retrieval algorithms, and the resolution can be higher than the detector with the correct geometry. Hybrid holography, which is a variation of Lensless Fourier Transform microscopy, has led to microscopy with 1 μm resolution using a CCD camera with 24 μm and 545 nm light [159]. This variation of the Leith-Upatnieks off-axis hologram uses a reference wave with a low carrier spatial frequency that is introduced with an opaque mask with a small hole that serves as a reference beam and a larger window in which the object is placed. The object image is retrieved from the inverse Fourier transform of the portion of the spatial frequency distribution at the carrier frequency. These two approaches are just two of the many techniques used for coherent imaging in the visible domain that will be explored for application to the EUV.

6.2 The future of laser-controlled chemical reactions

This thesis has demonstrated that control over molecular degrees of freedom at STP is possible using pulses that are not resonant with any particular electronic transition. In the past, work has focused on control in crossed-beam experiments or in systems with excitation levels well below a percent. We have observed behavior that indicates we have modified the reactivity of two products with a shaped light pulse. However, since a light pulse can also initiate a simple photochemistry reaction, great care must be taken to show that the reaction is not simply due to the breaking of the weakest bond, thus providing new products that may react. The goal is to demonstrate a concerted reaction in which a specific pulse shape enhances the reaction between two molecular reactants.

The apparatus used in the experiments described in this thesis was a hollow-fiber that was filled with the reactants. The benefit of macroscopic conversion makes this setup difficult to use for learning control experiments to manipulate chemical reactions since each laser shot substantially changes the concentrations of each chemical in the cell. Therefore, tracking the changes in the product concentrations becomes difficult. A better approach is to make a cell flow so that the initial conditions are reset for each trial pulse shape that only measures the effect of the reactivity of the trial pulse shape. Once an optimal "catalyst" laser pulse is discovered, the system can be operated in a "production mode" where laser pulses can be focused into large volumes of reactants in order to drive the desired chemical reaction and synthesize a target product.