Since finishing high school, I have participated in three research projects that have motivated me to pursue a career in interdisciplinary physics research. These studies have reinforced my fascination with the interaction between optical fields and molecular dynamics. Interfacing with subjects in biology, chemistry, physics, and computing, this phenomenon has developed rapidly in recent years to address many novel experimental and theoretical projects.

Following high school, I participated in Argonne National Laboratory (ANL)’s Pre-College Research Participation Program. I worked with Dr. Robin Santra to design a simulation to model High Harmonic Generation (HHG) under semi-classical principles. HHG results when an ionized electron generates electromagnetic radiation demonstrative not only of the driving frequency of the laser but also of additional high-order harmonics due to the presence of the charged parent atom. Understanding this phenomenon is essential toward devising an ultrafast imaging probe of orbital dynamics during chemical processes. Despite its promise, theoretical studies are troubled by the computational expense of modeling the quantum mechanical process. My study addressed approximating the system with a classical model of the electron during its propagation in the laser field to speed computation. I designed a simulation that described the electron’s motion through classical principles and computed the radiation spectrum. The spectra realized through this method were compared to those found in the literature, and demonstrated the model sufficient to render qualitatively consistent results, indicating it as a useful tool for testing qualitative hypotheses.

The following summer, I returned to ANL to work again with Dr. Santra as a DOE Science Undergraduate Laboratory Intern. During this summer, I completed two tasks. The first consisted of modeling the x-ray diffraction patterns of gold nanoparticles. X-ray diffraction is commonly used toward imaging fixed macromolecular crystals to determine molecular structure. My summer project investigated removing the crystalline requirement and extending this technique to resolve the dynamics of a single gold nanoparticle aligned within an optical trap toward investigation of novel behaviors of trapped nanoparticles. I considered particles, on average, of 100 nm in length, and modeled my laser source by the specifications of third-generation synchrotron light sources such as the Advanced Photon Source at ANL. To generate the diffraction pattern of the particle, I designed a program that would calculate scattering properties of the crystal and map the scattering photointensity onto an angular grid. The resulting diffraction pattern was considered in the small and wide angle regimes to demonstrate the applicability of the technique on two crystal length scales. Wide angle scattering resolved Angstrom length scales and indicated lattice alignment sensitivity through the intermittent appearance of Bragg diffraction peaks, while particle shape-sensitive small angle scattering responded to nanometer-scale alterations through the evolution of the intense scattering boundary surrounding the forward scattering peak. Discussions with experimentalists indicated that, while the scattering of individual particles in the wide angle would be difficult to distinguish from experimental noise, the photon yield of small angle scattering was sufficiently high to be used as a detection tool.

During this summer, I worked additionally in collaboration with Dr. Phay Ho to investigate the laser-induced rotational alignment of molecules modeled as rigid rotors. Working with a pre-constructed package, I examined the influence of molecular size toward the appearance of field-free alignment features. As this is a quantum mechanical process, it was proposed that massive molecules might cease exhibiting this effect, permitting classical modeling techniques in place of the more rigorous quantum mechanical ones. It was seen that, even in the case of heavier molecules, some alignment was still achieved at periodic intervals during the molecule’s field-free evolution, but that smaller alignment structures existing for smaller molecules outside of these brief intervals were eliminated. My contributions to this project were published in *The Journal of Chemical Physics*1.

Motivated by these studies of molecular alignment, I joined Dr. Tamar Seideman’s group at the beginning of my sophomore year at Northwestern. My work within this group has focused upon developing and implementing a model for the impulsive laser alignment of a biphenyl molecule. In general, the alignment of molecules is a critical consideration to many physical and chemical processes where traditional methods of achieving molecular alignment through strong static electric fields disrupt the novel behavior to be observed. The use of a short, intense laser pulse offers a feasible alternative by rotationally exciting a molecular species and creating periodic alignment revivals in the absence of the laser field. Biphenyl molecules, possessing electronically-critical torsional activity, are a relevant subject for laser alignment: it has been demonstrated that the dihedral angle adopted by biphenyl significantly alters its capacity for charge transport, so that control of the conformation of the molecule through laser alignment implies suitability toward applications demanding charge transport control, such as molecular junctions. A previous study of static torsion control revealed strong responsivity toward alignment with circularly polarized light. My project, motivated by this discovery, examined the dynamical torsional-rotational behavior exhibited by this molecule.

To simulate the process of alignment, I solved the time-dependent Schrödinger equation for the rotational-torsional wavefunction. The total Hamiltonian is separable into two terms: a molecular Hamiltonian which extended traditional diatomic molecule models to encompass torsional behavior; and a term to encompass the interaction between a circularly polarized electric field and the molecular induced dipole at non-resonant frequencies. The rotational-torsional wavefunction was expanded into a coupled basis, using the normalized Wigner d-matrices according to the rotation of the molecule; the torsional motion, troubled by the presence of the four-fold torsional potential barrier, was ultimately expressed using the special function solutions to the Whittaker-Hill equation. After solving a coupled system of differential equations for the wavefunction, it was possible to calculate observables indicative of the rotational and torsional alignment achieved. Adding torsional motions to the behavior of the biphenyl molecule was seen to detract from the laboratory-fixed rotational alignment achieved by biphenyl assumed torsionally-frozen. However, appropriate choice of laser intensity and duration revealed strong torsional and rotational alignment. The results of this study are very positive for further applications in molecular junction systems, where the field-free realignments of the torsional angle could be used to form an ultra-fast junction switch. Part way through the research I was selected to present this project at the Chicago Area Undergraduate Research Symposium2, and am currently writing an article for publication regarding this work.

My research experiences in interdisciplinary physics have reinforced my desire to pursue a career toward further exploring the interface between optical physics and molecular process. I enjoy the interdisciplinary these challenges, and hope to continue to work toward bettering our fundamental understanding of physical and chemical processes.

**References:**

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