1. Introduction
   1. Ultrafast dynamics in condensed matter
      1. Timescales and processes in solids
   2. Attosecond Transient Absorption Spectroscopy (ATAS)
      1. Overview of the technique
         1. Induced dipole picture (gas phase)
         2. XUV probe of population transfer/state changing picture (condensed phase)
         3. Why are you doing it with HHG? Contrast HHG vs synchrotron. Include figure of pulse duration vs photon energy, showing different light sources (synchrotrons, HHG sources, XFEL, etc)
         4. Comparison of absorptive to reflective measurements
            1. Complex refractive index
            2. Sample requirements and preparation
            3. Pointing stability (in reflection, sample is an XUV optic)
      2. Previous work, what is the state of the art?
         1. Previous ATAS/ATRS measurements in Ge, Si, etc.
         2. Motivation for long-wavelength studies in condensed matter
      3. What is physically observable via ATAS, and what isn’t?
         1. Limited k-space information (requires single crystal)
         2. Transmission geometry measures Im[n], not Re[n]
      4. Interpretation of data
         1. Most common method: TD-DFT
   3. High harmonic generation (discussed in context of XUV light source)
      1. 3-step model, temporal & spectral profile of light
      2. Phase matching considerations (with an eye towards increasing flux)
         1. Pressure-length product
         2. HHG yield vs wavelength scaling
         3. Optimal phase matching pressure vs wavelength
2. Experimental Apparatus
   1. Overview of TABLe, including main parts of beamline. Refer reader to Steve’s thesis for details on target chamber & photon spectrometer.
   2. Laser system
      1. SpitFire/OPA description & specifications
      2. Beam routing from laser bay to TABLe
      3. Active pointing correction
      4. SpitFire/OPA tips & tricks
   3. Vacuum system
      1. The need for vacuum (XUV propagation, air-sensitive equipment)
      2. Design goals
         1. Light source & interferometer w/ modular end stations
         2. Mag-lev pumps for low vibration
         3. Compact vacuum chambers --> split-level optical table
      3. Details of the vacuum system
         1. Generation chamber
            1. Gas throughput specifications
            2. CaF2 window
            3. Focusing geometry

Supported focal lengths

telescope

2-color generation

* + - * 1. XYZ translation stage and modular gas nozzle holder
        2. Vacuum aperture
        3. Rough vacuum feedthrough for HPC
      1. Differential pump chamber
      2. IR Diagnostic port & metallic filter
      3. Mirror chamber
         1. CaF2 window
         2. Ellipsoidal mirror motorization
         3. HM location: before or after EM
      4. Target chamber & 2nd differential pump chamber
         1. Designed to accommodate an electron spectrometer (not implemented)
         2. Sample holder (gas/condensed phase)
         3. Vacuum aperture & differential pumping
      5. 2nd filter chamber & photon spectrometer
         1. VLS gratings and motorization
         2. MCP/P
         3. Movable detector plane (cage & crank)
         4. IR diagnostic mirrors / port
      6. OMRON vacuum safety & rough vacuum systems
  1. Interferometer design
     1. Design goals
        1. In-air pump arm design for optical convenience (future nonlinear upgrades)
        2. XUV propagation in vacuum
        3. 3-to-1 XUV demagnification for high-intensity IR studies (cite Dietrich K.)
     2. XUV optics
        1. Requirements: reflective, ultra-smooth, gold coated
        2. Comparison of XUV mirror options (toroid, ellipsoid, multilayer mirrors)
        3. Our ellipsoidal mirror specifications (Zeiss)
        4. FRED simulations of ellipsoidal mirror focus
        5. Knife edge measurements at XUV focus
     3. Pump arm focusing
        1. Hole mirror
           1. The need for collinear IR/XUV geometry
           2. HM placement: before or after EM
        2. Description of optics used
        3. *LightPipes* simulations of IR focus
     4. Pulse energy control, calibration & monitoring
     5. Delay control & delay calibration
     6. XUV-IR spatial overlap
        1. Sensitivity of ATAS signal to misalignment of XUV/IR focal spots (simulations)
        2. Sensitivity of XUV-IR overlap integral to interferometer’s input pointing (simulations)
  2. XUV photon spectrometer (committee is OK with some overlap of Steve’s dissertation)
     1. Basic description (VLS gratings, flat field)
     2. MCP/P
        1. Basic specifications
     3. 2D CMOS sensor
        1. Specifications of lens and camera
        2. Description of data axes (spatial / spectral)
     4. Spectral calibration
        1. Argon Fano resonances
        2. Harmonic counting scheme
        3. Pi-plate & phase grating scheme
        4. Numerical fitting & the Jacobian

1. XUV light source design and apparatus performance
   1. The need for high XUV flux in ATAS experiments
   2. Harmonic gas sources
      1. Brief overview of relevant gas flow calculations
      2. Free expansion gas jet nozzle
         1. HHG performance vs pressure
         2. Shortcomings at longer wavelengths
         3. Low flux
      3. Low pressure cell
         1. Overview of design
         2. HHG performance
      4. High pressure cell
         1. Design goals:
            1. High flux
            2. High pressure for longer wavelengths
            3. Easily serviceable w/o relying on outside groups
         2. Overview of design
            1. Differential pumping
            2. Rough vacuum feedthrough into generation chamber
            3. Pressure monitoring
            4. Bellows specifications (movement, pressure limitations)
         3. Measured vs expected vacuum performance
         4. Performance relative to other designs
      5. Amsterdam pulsed piezo valve
         1. Basic description (large aperture for 2-source studies)
         2. HHG performance vs pressure
   3. Characterization of XUV source
      1. Knife edge measurements at XUV focus
      2. Harmonic yield stability
      3. XUV spectra optimized for various HHG conditions
      4. Measured transmission of metallic filters (Al, Zr)
      5. Ground state measurements of condensed matter samples
         1. Silicon
         2. Diamond
         3. Silicon nitride
         4. WS2
         5. Germanium
         6. WPOMs
         7. Cr2O3
   4. Characterization of interferometric stability
      1. Dietrich’s RABBIT data
      2. Two-omega oscillations in Argon
   5. Scaling of MCP response (yield & noise) with MCP voltage
2. ATAS experiments in Germanium
   1. Introduction
   2. Experimental considerations
      1. Sample requirements
         1. Thickness, large area
      2. Rastering of sample through focus to avoid heating, charge build-up
      3. XUV maps of samples
      4. IR propagation in thin films (TMM calculations, starting with *LightPipes* output)
      5. Orbital-resolved excitation probability vs wavelength (band structure calculations)
      6. Laser damage
      7. Estimation of excited carrier density
   3. Optimizing experimental ATAS parameters for Germanium
      1. Rep. rate (avoiding ms-scale excitation)
      2. IR pulse energy
      3. Harmonic spectrum (2-color, wavelength)
      4. Optimized ATAS Ge experimental results
      5. Post-experiment analysis: verification we didn’t permanently damage sample
   4. Data analysis
      1. Description of data pipeline
         1. Going from a 2D image to a 1D spectra
            1. Background subtraction
            2. Selecting a divergence window
            3. Normalization by exposure time, divergence window
            4. Integration over divergence window
         2. Energy calibration
         3. calculation
      2. Systematic noise sources in our experiment
         1. Harmonic yield drift
         2. Dark counts, background subtraction & ambient light
         3. Laser-induced sample damage
         4. Nonlinear MCP/P response?
      3. Methods to numerically correct for harmonic drift / noise
         1. Descriptive statistics: correlation between harmonic drifts
         2. Broad energy range harmonic normalization (doesn’t work that well)
         3. nearest-neighbor harmonic normalization (works best)
         4. SVD of harmonic spectra?
         5. Other methods?
      4. Frequency filtering to remove oscillations
   5. Physical interpretation of spectra (following Leone)
      1. Decomposition of spectral response
      2. Description of observed dynamics
3. Conclusion
   1. Experimental outlook, potential improvements to system
      1. Reflective focusing for generation & pump arms
      2. In-situ imaging of XUV/IR overlap
      3. Closed-loop control over HM2 pointing
      4. Optical improvements to minimize XUV/IR focal spot misalignment (guided by simulations)
      5. Isolated attosecond pulses for XUV continuum
      6. In-vacuo condensed matter sample cooling with gas jet to reduce sample heating
      7. Motorized retroreflector to study ps-scale dynamics
      8. Simultaneous XUV reference measurement (w/o sample)
      9. Simultaneous photoelectron & photoabsorption measurements
      10. Moving laser system closer to TABLe, or propagating under vacuum
      11. Replacing target chamber with doubly-differentially pumped chamber (for gas & liquid phase experiments)
      12. Upgrade HPC RV pump to a dry pump
4. Appendix
   1. How to use the vacuum system
      1. OMRON safety system
      2. pump down, venting, gate valve operation, arming & emergency venting
   2. How to align into the interferometer
   3. How to use the High Pressure Cell
   4. Required maintenance
      1. Turbo pumps
      2. Rough pumps
      3. Spitfire (gratings, fluids, filters)