Using Femtosecond Electron Diffraction to Study Dynamical Structures and Transient States

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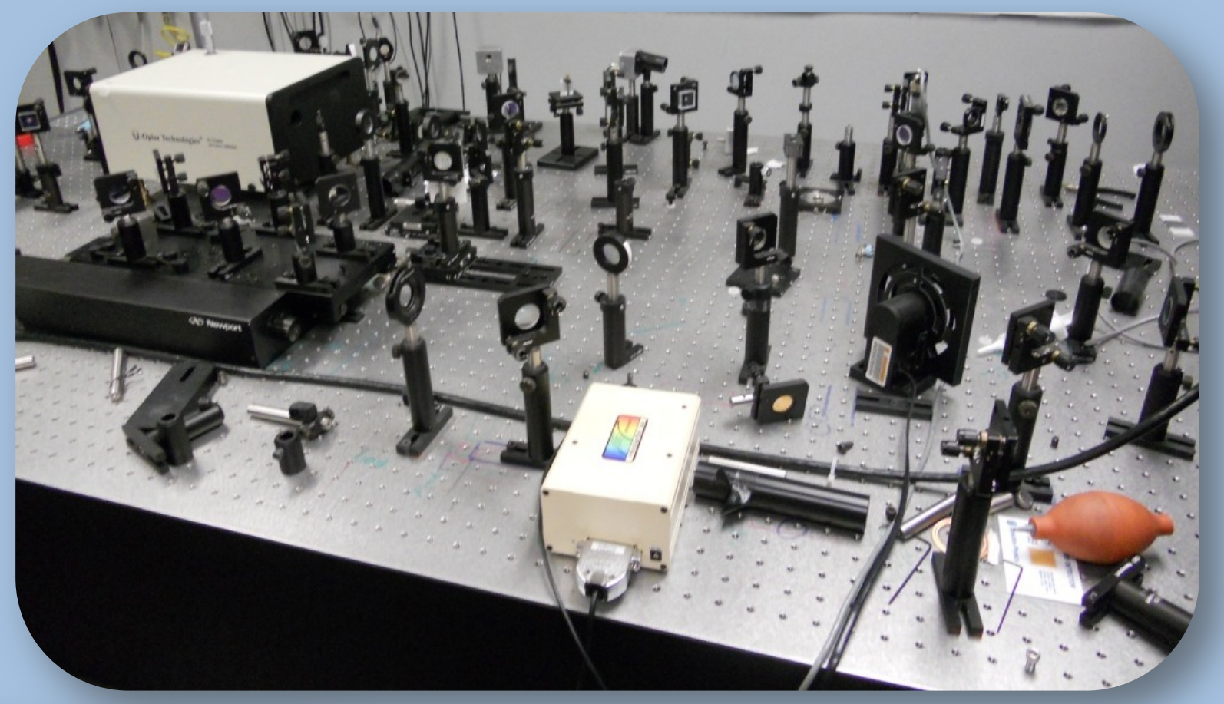
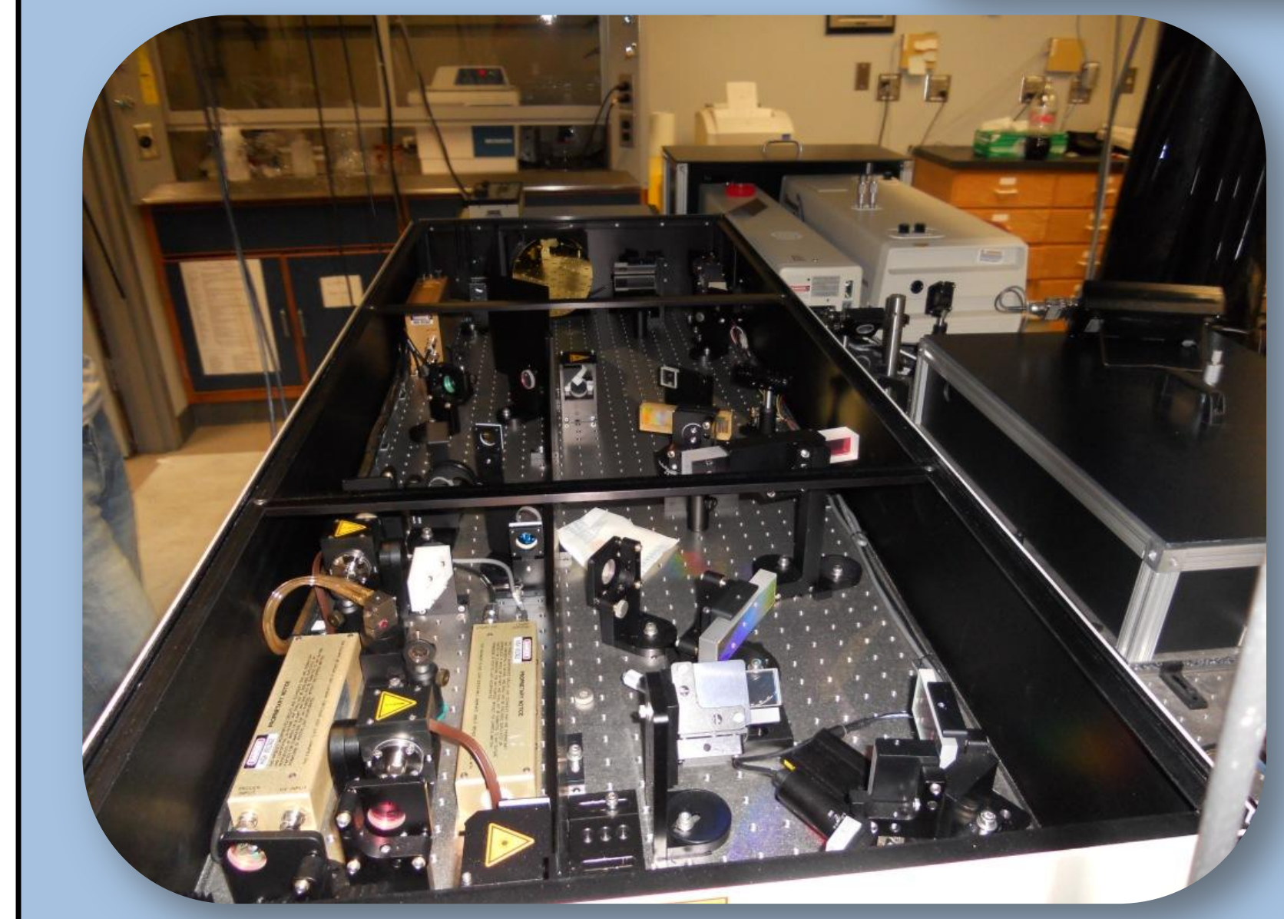
Abstract

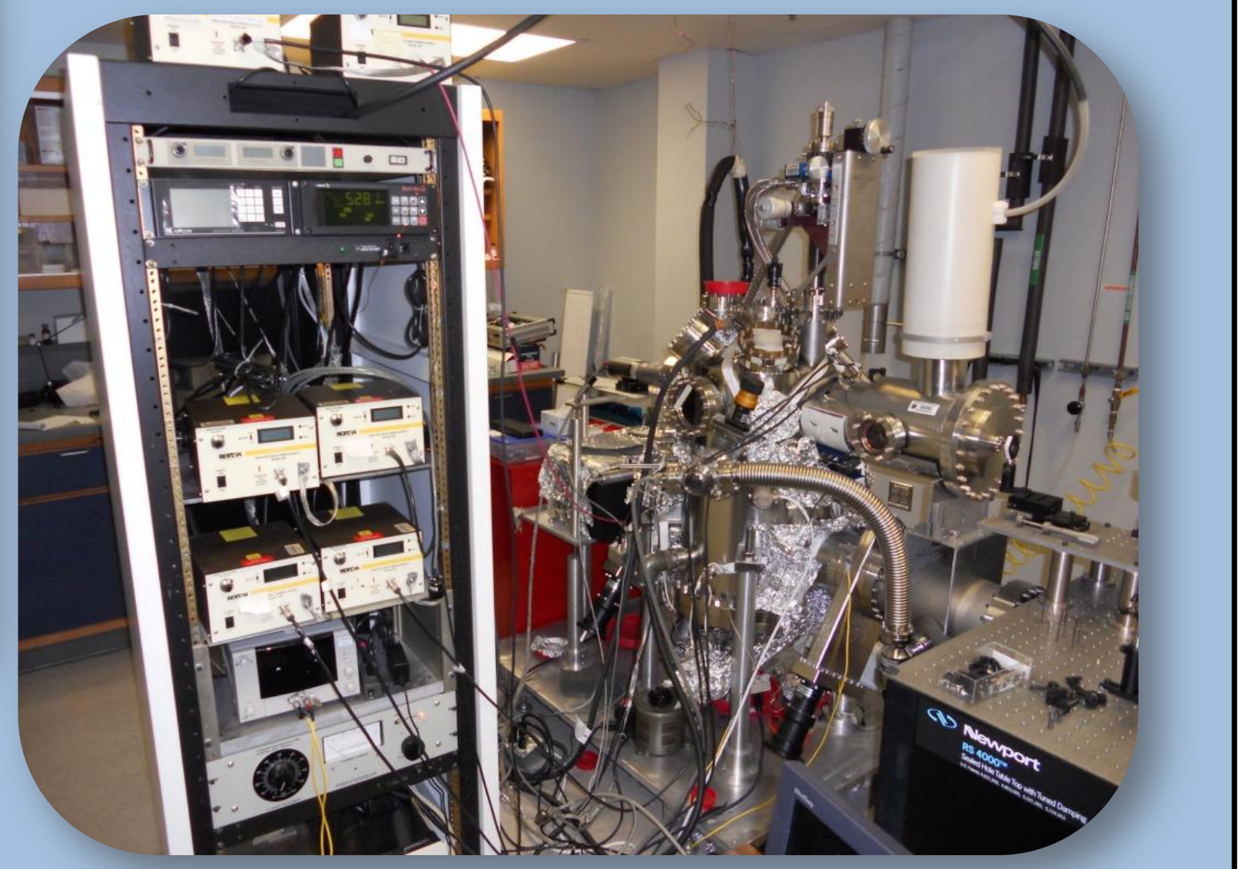
Femtosecond Electron Diffraction (FED) is an new imaging technique that could achieve both high spatial and temporal resolution in dynamical studies of material structures. With it, we can take snapshots of dynamical structures or transient states that only exist on sub-picosecond time scale. By taking a series of these snapshots at different time delays, we can construct the structural dynamics in real-time and study new structure functions associated with these dynamics.

Introduction

The methods for executing successful FED require precise tunings of many different instruments, including laser system, motion stage control, electron gun, vacuum system and CCD imaging. Since the dynamics we study mostly occur on a picosecond level, we must go into the femtosecond region to be able to take clear pictures. This demands the use of femtosecond laser pulses to achieve. At the same time, atomic level lattice change could also be achieved in our system. These allow us to study dynamical structure changes with both high temporal and spatial resolutions.

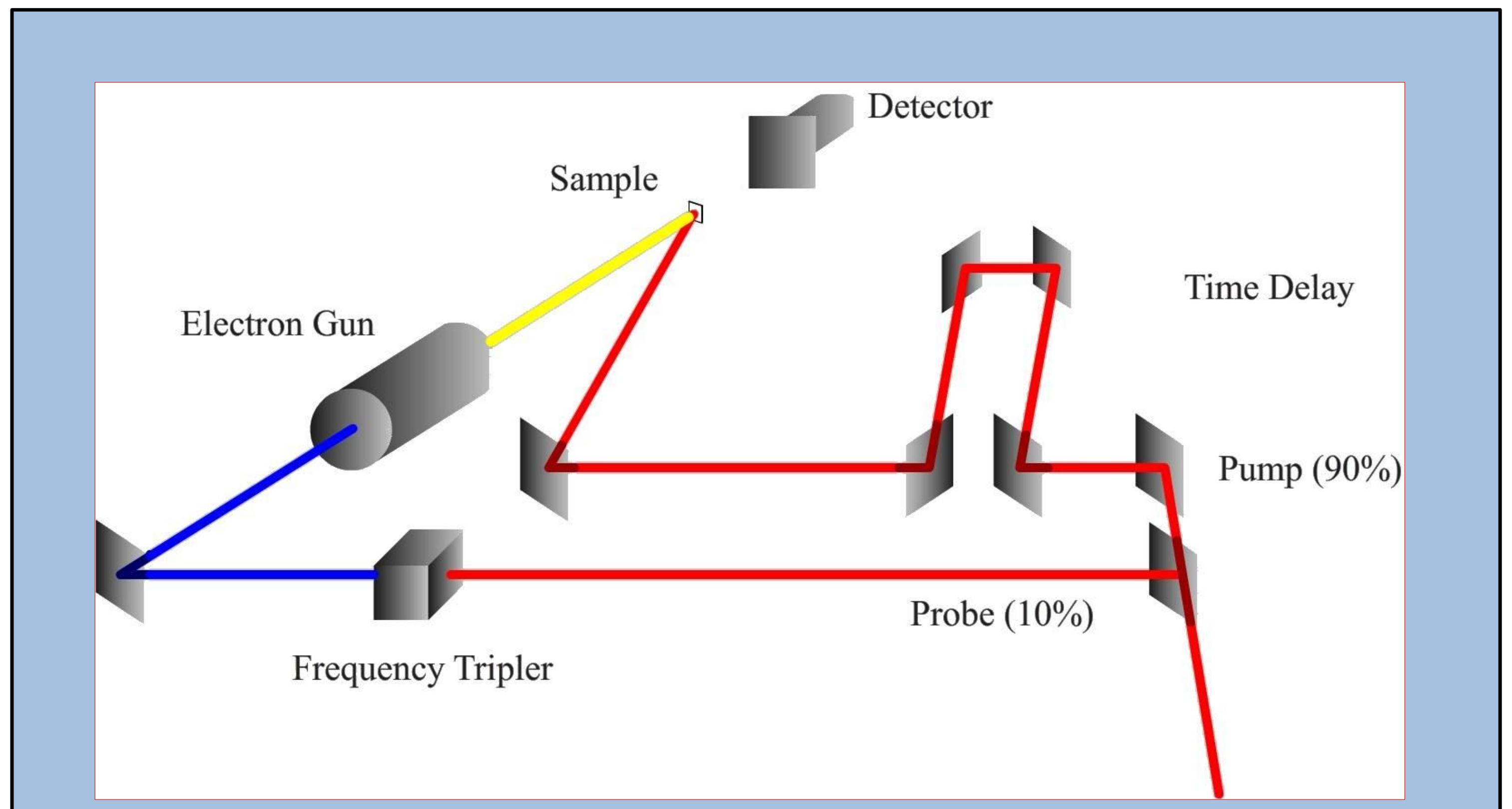






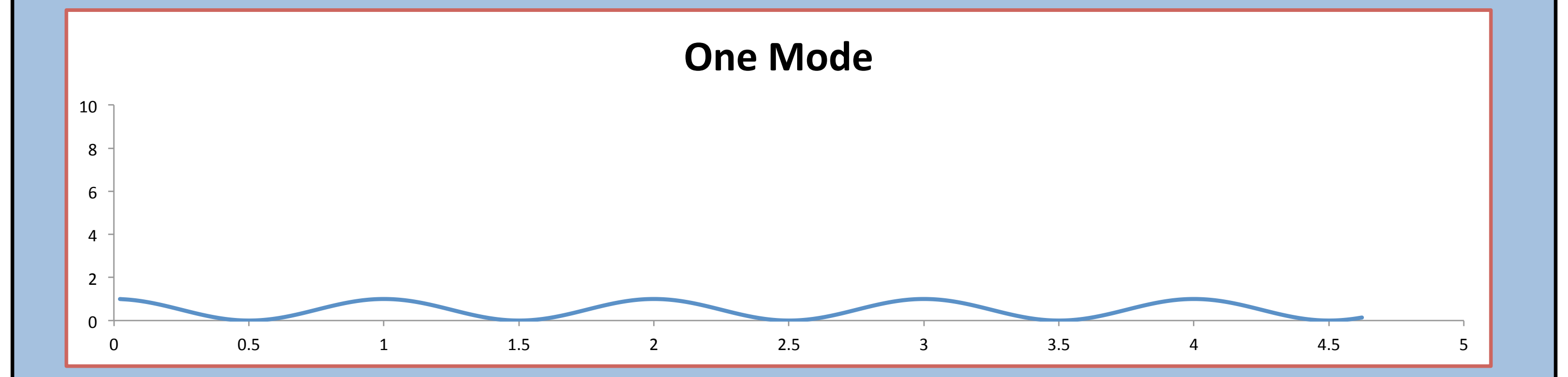
Femtosecond Electron Diffraction System

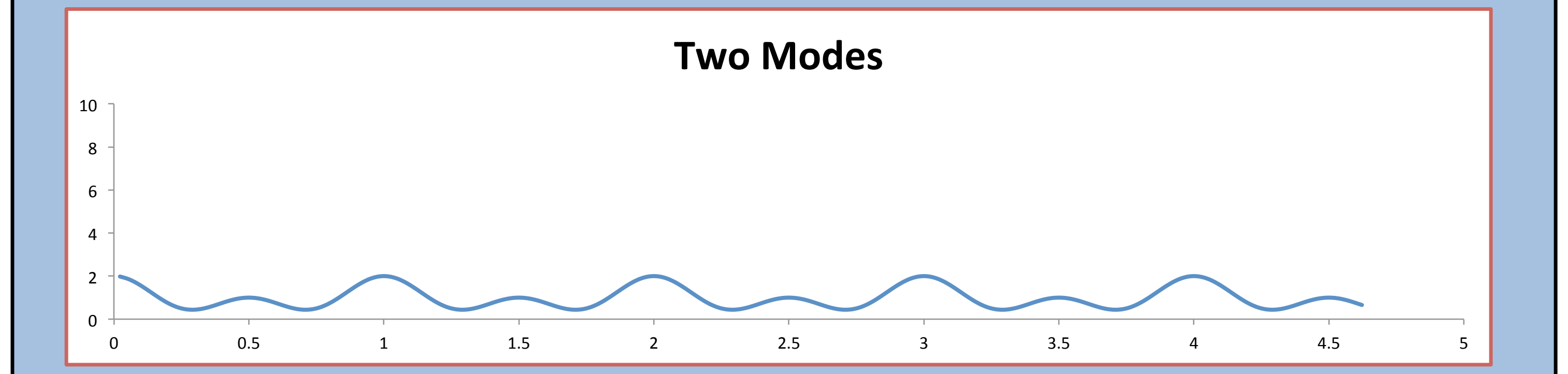
The setup we use is quite complex altogether. It starts with an amplified femtosecond laser unit which produces pulses close to 50fs with a wavelength in the infrared spectrum near 800nm. The pulses are then sent through an optical array where the beam is split into two separate beams of strength, 90% and 10%. These are our pump and probe beams, respectively. The pump branch also contains a motion stage where, by controlling the difference in optical path lengths, we can control the delay time of when we pump and when we probe. The pump pulses are firstly focused on the sample to initiate some dynamical change. The probe pulses are sent to an electron gun, where they are converted into electron pulses, and then probe the laser-induced structure change by electron diffraction. The resulting patterns are then recorded using a CCD camera.



Femtosecond Laser Pulses

To achieve femtosecond laser pulses, we begin with a continuous YAG laser at a wavelength of 532 nm. The continuous laser beam is sent into a Ti:Al_2O_3 oscillator, where 532 nm photons are absorbed, and photons at infrared range are emitted. Within the oscillator cavity, only the modes at resonance frequency could get magnified. By carefully choosing these modes and compensating the optical path difference within the cavity, these modes can get coherently magnified and keep a constant magnification ratio, which is known as “mode-locking”. After this is achieved, the interference of these modes give a spike like peak in the time domain. The interference of these model in the time domain are shown in following figures.


One Mode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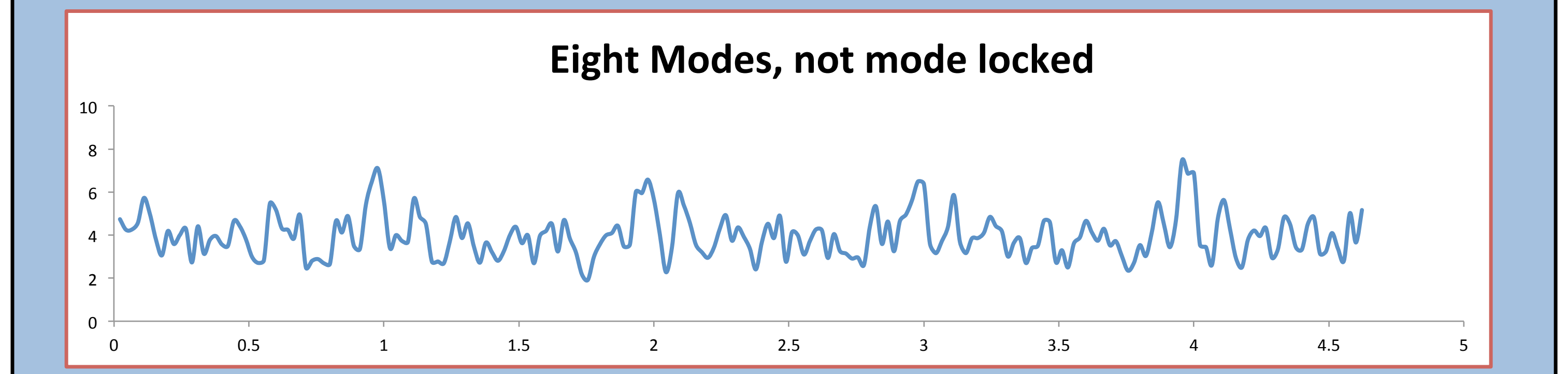
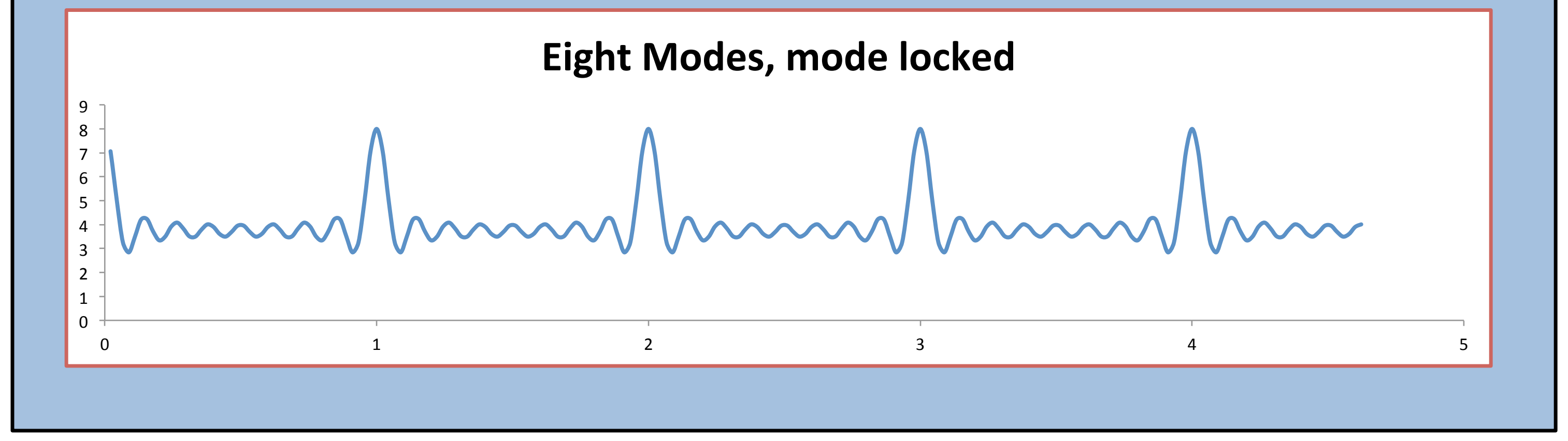
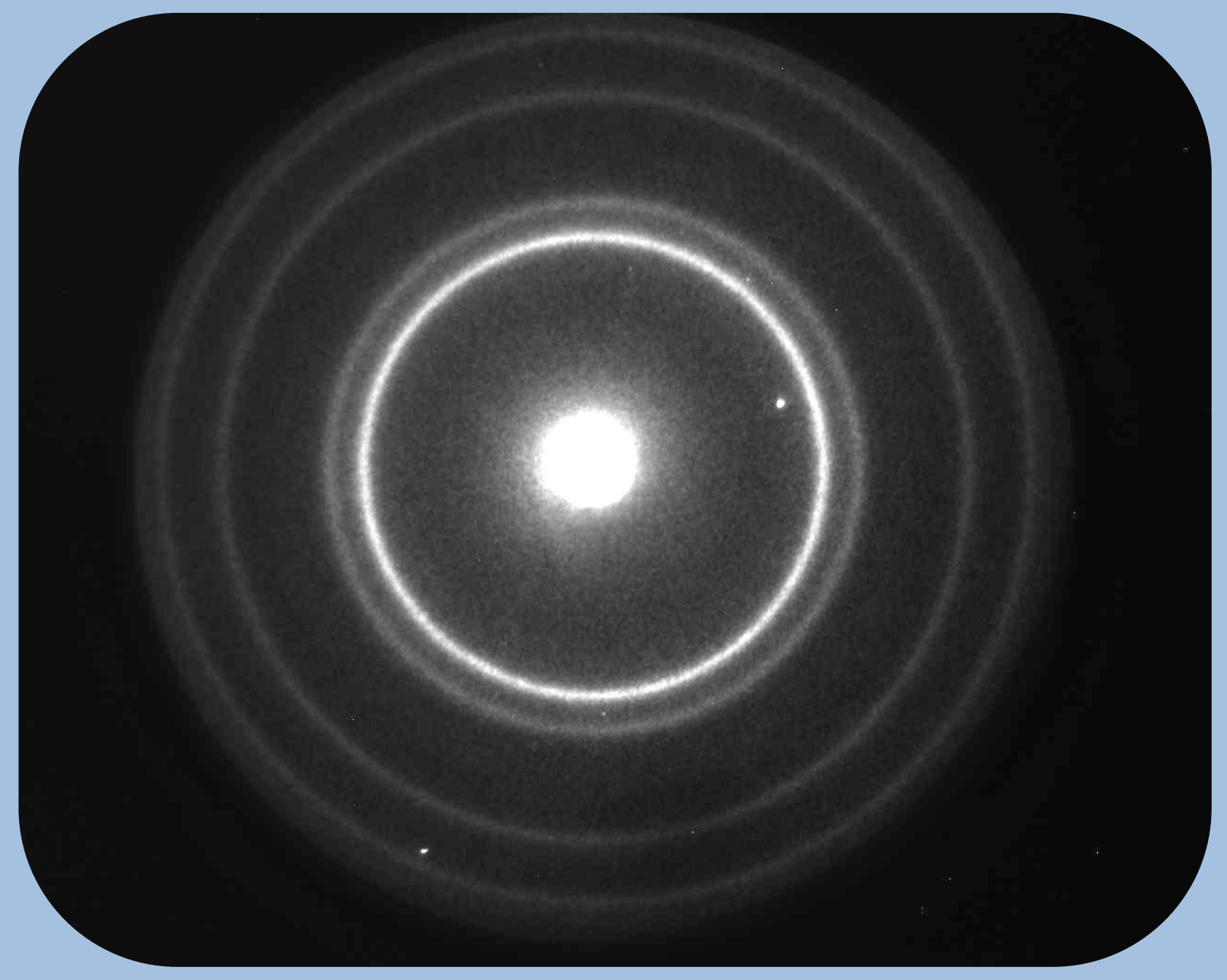

Eight Modes, not mode locked


Eight Modes, mode locked

Diffraction Patterns

When an electron beam is projected at our samples, which range in thickness from 10nm to 30nm, the beams are scattered due to interactions with the crystal structure of the sample. On the directions satisfying Bragg equation, the scattering are constructively intensified, on the other directions, the scattering are deconstructive. Following figure is an example of these diffraction patterns, which contains information about crystal structure. The IR pump excites the sample and initiate thermal or coherent change of the crystal structure, and the probe pulse monitor these structure change at different time delays. By analyzing these diffraction patterns and combining with Bragg equation, we could derive the thermal motion of the structure from intensity change of the Bragg rings. We could also derive the coherent motion of lattice from the radius change of the Braggs rings. Since the time width of pump pulses in our FED setup is roughly 50fs and our probe is around 350fs, we are able to study a structure change with duration time as short as 400fs.



Ni Diffraction Pattern

Conclusion

Femtosecond electron diffraction enables researchers to explore questions dealing with ultrafast thermal motion and coherent motion of lattice in crystals. The use of FED allows for time resolved studies of the atomic realm giving us greater insight into non-equilibrium, or transient states that only exist in very short duration time, and are not reachable on normal equilibrium conditions.

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