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Full characterization of RF compressed femtosecond electron pulses using ponderomotive scattering

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Abstract: High bunch charge, femtosecond, electron pulses were generated using a 95 kV electron gun with an S-band RF rebunching cavity. Laser ponderomotive scattering in a counter-propagating beam geometry is shown to provide high sensitivity with the prerequisite spatial and temporal resolution to fully characterize, in situ, both the temporal profile of the electron pulses and RF time timing jitter. With the current beam parameters, we determined a temporal Instrument Response Function (IRF) of 430 fs FWHM. The overall performance of our system is illustrated through the high-quality diffraction data obtained for the measurement of the electron-phonon relaxation dynamics for Si (001).
Ultrafast electron diffraction with radio-frequency compressed electron pulses

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We report on the complete characterization of time resolution in an ultrafast electron diffraction (UED) instrument based on radio-frequency electron pulse compression. The temporal impulse response function of the instrument was determined directly in pump-probe geometry by performing electron-laser pulse cross-correlation measurements using the ponderomotive interaction. With optimal settings, a stable impulse response of 334 ± 10 fs was measured at a bunch charge of 0.1 pC (6.24 × 10^5 electrons/pulse); a dramatic improvement compared to performance without pulse compression. Phase stability currently limits the impulse response of the UED diffractometer to the range of 334 – 500 fs, for bunch charges ranging between 0.1 and 0.6 pC. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4747155]
Alignment of magnetic solenoid lenses for minimizing temporal distortions

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Abstract
An ubiquitous focusing element for charged particles is the magnetic solenoid lens. For the case of ultrashort electron pulses, we show here that misalignment of the lens, i.e. displacement or tilt, causes significant temporal aberrations on a femtosecond time scale. Pulse-lengthening is only minimized if the beam travels on the symmetry axis. We present an experimental procedure with periodic reversal of the magnetic field for aligning position and tilt with sufficient precision for reducing the aberrations to less than one femtosecond. This method will be instrumental for advancing ultrafast electron microscopy and diffraction to ultimate temporal resolutions.

Keywords: electron microscopy, ultrafast diffraction, attosecond science
Design and implementation of a flexible beamline for fs electron diffraction experiments

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Ultrafast Electron Diffraction (UED) has been widely used to investigate the structural dynamics of molecules and materials. Femtosecond (fs) electron bunches are used to obtain diffraction images of a specimen upon photo-excitation by a temporally delayed light pulse. The high cross-section of electrons makes it a very flexible tool for the study of light elements, monolayers and surfaces; at the same time, electrons can travel down to few nanometers (nm) and structural information from the bulk can also be retrieved. In this article, we discuss the design and implementation of a flexible beamline for fs electron diffraction experiments in transmission or reflection geometry. By the use of a radiofrequency (RF) compression cavity synchronized to our laser system, in combination with a set of electron optics, we demonstrate that we can control the beam properties in terms of charge per pulse, transverse spot-size on the sample and temporal duration of the bunches. The characterization of the beam is performed via a light-electrons cross-correlation experiment and we demonstrate an overall temporal resolution around 300 fs for bunches containing up to $10^5$ electrons at a repetition rate of 20 kHz.
Compression of single-electron pulses with a microwave cavity

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Abstract. Few-femtosecond to attosecond electron pulses are required for advancing ultrafast diffraction and microscopy to the regime of electrons in motion. Here, we report the combination of a single-electron source with a microwave cavity for pulse compression. In such an arrangement, the electron pulses can become significantly shorter than the laser pulses used for electron generation. This comes at the expense of an increase in energy spread. We report the use of an energy analyzer for characterizing microwave-compressed single-electron pulses. Phase effects, linearity, focal distances, incoming pulse durations and laser–microwave jitter are measured for three different synchronization approaches. The results demonstrate the applicability of a microwave cavity in the single-electron regime and identify jitter as the current limitation on the way to few-femtosecond, eventually attosecond pulses of single electrons.
Imaging of molecules in the gas phase with ultrafast electron diffraction

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ABSTRACT

A two-step algorithm is developed that can reconstruct the full 3-D molecular structure from diffraction patterns of partially aligned molecules in gas phase. This method is applicable to asymmetric-top molecules that do not need to have any specific symmetry. This method will be important for studying dynamical processes that involve transient structures where symmetries, if any, can possibly be broken. A new setup for the diffraction experiments that can provide enough time resolution as well as high currents suitable for gas phase experiments is reported. Time resolution is obtained by longitudinal compression of electron pulses by time-varying electric fields synchronized to the motion of electron pulses.

Keywords: Ultrafast electron diffraction, 3-D molecular structures, laser induced alignment, phase retrieval, electron pulse compression.
INITIAL RESULTS FROM STREAKED LOW-ENERGY ULTRA-FAST ELECTRON DIFFRACTION SYSTEM

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Abstract

Radiabean, in collaboration with UCLA, is developing an inexpensive, low-energy, ultra-fast, streaked electron diffraction (S-UED) system which allows one to reconstruct a single ultrafast event with a single pulse of electrons using an RF deflector. The high-frequency (GHz), high voltage, phase-locked RF field in the deflector enables temporal resolution of atomic events as fine as sub-100 fs. In this paper, we present an overview of the system being developed and the initial experimental results. We also discuss the challenges based on our design of a UED system that incorporates a novel, high-resolution dielectric-loaded RF deflector and a solid-state X-band amplifier.

INTRODUCTION

Time-resolved observation of atomic motion is one of the frontiers of modern science, and advances in this area will greatly improve our understanding of many basic sciences. One technique under active development in this area is ultrafast electron diffraction (UED). UED has already been used to study solid state phase transitions [1], gas phase reactions [2], strongly coupled systems [3] and surface dynamics [4]. To improve the resolution of their UED measurements, researchers need shorter electron bunches and methods increase temporal resolution on the detected electrons. By placing an RF deflecting cavity immediately after a sample, the time-dependent, attractor electron beam can be “streaked,” transforming the temporal evolution of the diffraction pattern from the sample into a transverse image [5]. In this project, we are developing a complete S-UED system based on a dielectric-loaded RF deflector and a novel solid-state power amplifier (SSPA), see Fig. 1. The system also includes the requisite electron gun, laser, magnetic optics, and imaging components.

HARDWARE

The UED system was assembled in the Pegasus Lab at the University of California, Los Angeles. The beam line was initially installed and commissioned during fabrication of the dielectric deflector. During this time we conditioned the gun to its operating voltage and worked through the alignment procedure for the magnetic optics. The diagnostics were also tested during this time and the results were compared to earlier simulations. The important components of the system are described in further detail below.

Electron Gun

The 100 kV gun shown in Figure 2 was purchased from a Dutch company, Acc-Tech. The HV conditioning and initial photo-beam measurements were carried out without insertion of the radio-frequency deflector in the beam line to avoid the initial alignment process less difficult. Although the purchased version of the DC gun is not operable under ultra-high vacuum due to use of elastomer O-rings, it is appropriate for our UED application which only requires vacuum of 10⁻⁶ Torr.

Dielectric Loaded Deflector

The deflector design was performed using the 3D code HFSS. We eventually decided to pursue a dielectric loaded structure since it saved us time and resulted in a device that was easier to manufacture than an all-copper version for such low charge. At the same time, Radiabean wanted to develop a medium-power (few hundred watts) RF power source using solid-state devices, and the two technologies were good compliments to each other. Figure 3 shows a 3D model of our 6 cm long
Direct measurement of synchronization between femtosecond laser pulses and a 3 GHz radio frequency electric field inside a resonant cavity

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We demonstrate a method to measure synchronization between femtosecond laser pulses and the electric field inside a resonant 3 GHz radio frequency (RF) cavity. The method utilizes the Pockels effect in a crystal inside the RF cavity by measuring the retardation of the components of polarization as a function of RF phase. Resolution of the setup used is shown to be 29 ± 2 fs (root-mean-square, rms), with timing jitter between the laser pulses and the RF field inside the cavity of 96 ± 7 fs (rms). The method provides a tool to reduce jitter and improve time-resolution in ultrafast electron diffraction experiments. V 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4823590]
Ultrafast Electron Diffraction of Thin Metal Films and Metal Nanoparticles

Master Thesis by Rosario A. Stornante

Supervisors
Prof. Petra Rudolf
Prof. Giuseppe G.N. Angilella

Academic Year < 2014/2015 >

Abstract
This thesis has been carried out at the group of Surfaces and Thin Films of the Zernike Institute where a facility for Ultrafast electron diffraction (UED) was developed. With this pump-probe technique, structural transitions (such as phase transitions or simply atomic disorder caused by temperature), induced by photo excitation, can be studied at the atomic scale with picosecond time resolution. A femtosecond laser is used to excite the sample; the laser generates also electrons from a solid copper cathode, which are accelerated to 30 keV and used as a probe.

The experimental setup allows measurements both in transmission and in reflection: in the second case the difference between the group velocity of the electrons and that of light is compensated by the wave front tilt of the laser. The diffraction image is recorded using a ultrasensitive charge-coupled device. The system is equipped with a preparation chamber connected with the analysis chamber, for the growth of the samples, equipped with various evaporators and a XPS system. The samples are mounted on a cryostat which allows cooling with liquid nitrogen or liquid helium.

The purpose of this work was to complete the calibration of the UED system and run the first experiments to verify its proper operation. Measurements in transmission were performed on a film of chromium and gold of 45 nm (5 nm Cr 40 nm Au) and on nanoparticles of cobalt, smaller than 10 nm. A measurement in reflection on a crystal of silver was also performed. The data obtained were interpreted by means of a theoretical model, and compared with literature references. Results show a sub-picosecond resolution, as well as very good signal to noise ratio. Future experiments will involve ultrathin films that show phase transitions of greater interest. Some of these films, as very sensitive to oxidation, will be prepared in situ in the preparation chamber, making full use of this facility.
Abstract

Femtosecond Electron Diffraction and Spectroscopic Studies of a Solid State Organic Chemical Reaction

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2014

Photochromic diarylethene molecules are excellent model systems for studying electrocyclic reactions, in addition to having important technological applications in optoelectronics. The photoinduced ring-closing reaction in a crystalline photochromic diarylethene derivative was fully resolved using the complementary techniques of transient absorption spectroscopy and femtosecond electron crystallography. These studies are detailed in this thesis, together with the associated technical developments which enabled them. Importantly, the time-resolved crystallographic investigation reported here represents a highly significant proof-of-principle experiment. It constitutes the first study directly probing the molecular structural changes associated with an organic chemical reaction with sub-picosecond temporal and atomic spatial resolution – to follow the primary motions directing chemistry.

In terms of technological development, the most important advance reported is the implementation of a radio frequency rebunching system capable of producing femtosecond electron pulses of exceptional brightness. The temporal resolution of this newly developed electron source was fully characterized using laser ponderomotive scattering, confirming a $435 \pm 75$ fs instrument response time with $0.20$ pC bunches. The ultrafast spectroscopic and crystallographic measurements were both achieved by exploiting the photoreversibility of diarylethene. The transient absorption study was first performed, after developing a novel robust acquisition scheme for thermally irreversible reactions in the solid state. It revealed the formation of an open-ring excited state intermediate, following photoexcitation of the open-ring isomer with an ultraviolet laser pulse, with a time constant of approximately $200$ fs. The actual ring closing was found to occur from this intermediate with a time constant of $5.3 \pm 0.3$ ps. The femtosecond diffraction measurements were then performed using multiple crystal orientations and a large number of different samples. To analyse the results, an innovative method was developed in which the apparently complex ring-closing reaction is distilled down to a small number of basic rotations. Immediately following photoexcitation, sub-picosecond structural changes associated with the formation of the intermediate are observed. The rotation of the thiophene rings is identified as the key motion. Subsequently, on the few picosecond time scale, the time-resolved diffraction patterns are observed to converge towards those associated with the closed-ring photoproduct. The formation of the closed-ring molecule is thus unambiguously witnessed.
Towards attosecond 4D imaging of atomic-scale dynamics by single-electron diffraction

Dissertation
an der Fakultät für Physik
der Ludwig–Maximilians–Universität München

Vorgelegt von
Alexander Gliserin
aus Kriwoj Rog, Ukraine

München, den 17. Februar 2014

Abstract

Many physical and chemical processes which define our daily life take place on atomic scales in space and time. Time-resolved electron diffraction is an excellent tool for investigation of atomic-scale structural dynamics (4D imaging) due to the short de Broglie wavelength of fast electrons. This requires electron pulses with durations on the order of femtoseconds or below. Challenges arise from Coulomb repulsion and dispersion of non-relativistic electron wave packets in vacuum, which currently limits the temporal resolution of diffraction experiments to some hundreds of femtoseconds. In order to eventually advance the temporal resolution of electron diffraction into the few-femtosecond range or below, four new concepts are investigated and combined in this work: First, Coulomb repulsion is avoided by using only a single electron per pulse, which does not repel itself but interferes with itself when being diffracted from atoms. Secondly, dispersion control for electron pulses is implemented with time-dependent electric fields at microwave frequencies, compressing the duration of single-electron pulses at the expense of simultaneous energy broadening. Thirdly, a microwave signal used for electron pulse compression is derived from an ultrashort laser pulse train. Optical enhancement allows a temporal synchronization between the microwave field and the laser pulses with a precision below one femtosecond. Fourthly, a cross-correlation between laser and electron pulses is measured in this work with the purpose of determining the possible temporal resolution of diffraction experiments employing compressed single-electron pulses. This novel characterization method uses the principles of a streak camera with optical fields and potentially offers attosecond temporal resolution. These four concepts show a clear path towards improving the temporal resolution of electron diffraction into the few-femtosecond domain or below, which opens the possibility of observing electron densities in motion. In this work, a compressed electron pulse's duration of 28±5 fs full width at half maximum (12±2 fs standard deviation) at a de Broglie wavelength of 0.08 Å is achieved. Currently, this constitutes the shortest electron pulses suitable for diffraction, about sixfold shorter than in previous work. Ultrafast electron diffraction now meets the requirements for investigating the fastest primary processes in molecules and solids with atomic resolution in space and time.
Abstract

Mapping Organic Molecular Motions with RF Compressed Femtosecond Electron Diffraction

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2014

This thesis reports on the development of a novel ultrabright femtosecond electron diffraction setup and its application towards monitoring primitive atomic motions during a photoinduced phase transition in an organic molecular crystal. By using the time-dependent electric field of a 3 GHz radio-frequency cylindrical cavity, high flux electron bunches are compressed to a few hundred femtoseconds in duration. A grating-enhanced ponderomotive scattering technique is used to fully characterize the temporal characteristics of the high brightness electron pulses. A minimum temporal instrument response of 420 femtoseconds (full-width-half-maximum) for 200 femtocoulomb electron bunches is reported. The instrument response is limited by the synchronization jitter between the radio-frequency waves and the laser oscillator. To mitigate this problem, a photo-triggered streak camera is used to first characterize the arrival time jitter of the electron pulses and then time-stamp the precise arrival time of each individual electron pulse with respect to the laser. This reduces the synchronization contribution to the instrument response time down to 30 femtoseconds RMS (root-mean-square). This new electron source is used to study the molecular dynamics of a photoinduced insulator-to-metal phase transition in the organic crystal (EDO-TTF)$_2$PF$_6$. A 60 femtosecond optical pulse at 800 nm induces the phase transition in the crystal and ultrabright electron pulses are applied to monitor the evolution of the Bragg intensities. A structure refinement algorithm is applied to calculate the transient structures during the first few picoseconds of evolution. A transiently locked intermediate state is discovered and reveals the primary mechanism behind the transformation towards the metallic state.
Ultrafast single-electron diffraction at 100 keV and investigation of carbon-nanotube dynamics

Dissertation
an der Fakultät für Physik
der Ludwig–Maximilians–Universität München

vorgelegt
von Daniel Kreier
aus Schweinfurt


Abstract

Time-resolved electron diffraction is a powerful tool to observe ultrafast structural dynamics in materials and molecules with atomic spatial as well as temporal resolution. Due to Coulomb repulsion, however, the use of only single-electrons or few-electrons per pulse is inevitable to reach the shortest pulse durations. Electrons have rather high scattering cross sections and thus experiments in transmission require ultrathin samples in the nanometer-range, making sample preparation very challenging. Up to now, ultrafast single-electron diffraction was only demonstrated at an electron energy of 30 keV; these measurements were performed in our group at the “UED1-beamline”.

This work introduces our second-generation beamline, “UED2”, where the electron acceleration voltage is upgraded from 30 to 100 keV, which allows the investigation of significantly thicker samples. This is decisively widening the range of complex materials that can be studied. In the experiment, electron pulses are generated by a two-photon photoemission process and the long-term stability of the source is shown. The samples can be placed in transmission as well as grazing incidence geometry. To achieve phase-matching between the optical and electron pulses, tilted optical pulses can be applied. We figured out that to avoid temporal distortions in tilted pulses, a geometry must be chosen in which the propagation direction of the tilted pulses is perpendicular to the grating’s surface. Furthermore, temporal distortions for ultrashort electron pulses caused by misaligned magnetic lenses are examined. It is found that a displacement or tilt of the lens causes significant temporal aberrations on a femtosecond time scale and pulse-lengthening is only minimized if the beam travels precisely on the symmetry axis. An experimental procedure detailed here for aligning lens-position and -tilt reduces the aberrations to less than one femtosecond.

For the “UED2-beamline”, a new laboratory was established and a first time resolved electron diffraction experiment at this beamline performed. Anisotropic ultrafast atomic motion in carbon-nanotubes was observed, revealing the nature of the system’s chemical bonds, which vary from relatively weak van der Waals to strong covalent interactions.

In summary, it is thus shown that ultrafast electron diffraction at 100 keV with single/few electrons per pulse is an excellent method to study ultrafast atomic-scale dynamics even in complex solid samples with the highest possible resolution in space and time.