Construction of a setup for Ultrafast Electron Diffraction

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Chapter 1 Introduction

In their natural way of observing the world, humankind is naturally limited to objects that are roughly as thick as a hair, 100 μm. Time scales at which changes can be observed are limited to tens of milliseconds, being the reason that the refresh rate of television has been 50 Hz for decades: this is observed as real-time motion rather than as a series of individual images. Over the centuries, scientists have been searching for ways to look smaller and faster.

The nineteenth century photographer Eadweard Muybridge (1830 – 1904) invented a stroboscopic experiment to answer the highly debated scientific question whether all four legs of a horse at a gallop are off the ground at the same moment or not. His approach was to install twelve photocameras and to connect each one to a line that triggers the camera when it is touched by the passing horse. The images are shown in Figure 1.1. The conclusion from this experiment was that there is an instant that the horse is completely elevated from the ground. But apart from that, this was the first millisecond timescale experiment in the history: technology allowed him to observe something in detail which his own eye observes as one movement.

Thanks to the invention of lasers we are now able to study phenomena at a time scale that is twelve orders of magnitude faster than the Muybridge experiment: the femtosecond time domain. The PhD project of which this thesis is the written reflection was dedicated to the construction of an experimental setup for Ultrafast Electron Diffraction: a technique that allows studying processes that take place at this time scale, that is the natural time scale at which atoms and molecules interact and for instances atomic bonds are formed and destroyed.

1.1 Microscopy and Diffraction

Besides the motivation to push the limits of the shortest time scales that can be studied, a lot of interest in the history has gone to look at smaller and smaller objects. A very important person in this context is the Dutch scientist Antoni van Leeuwenhoek (1632 – 1723). In the 17th century there were already microscopes with magnifications up to 30x, but Van Leeuwenhoek made a microscope with a magnification of 480x. This allowed him to observe objects at the cellular level, among which red blood cells and human spermatozoids. This is also almost equal
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to a fundamental limit that applies when it comes to magnified observation with visible light: because of the diffraction limit objects that are smaller than the wave length cannot be observed.

Thanks to the development of quantum physics in the early twentieth century, and especially because of the realization that apart from a particle character, electrons also have a wave character, electron microscopy was developed in the 1920’s and 1930’s. As the De Broglie wavelength of electrons of e.g. 30 keV is 7 pm, the lower size limit of structures to be observed with an electron microscope is much smaller than that of a visible light microscope. Electron microscopy exists in two modes: transmission electron microscopy (TEM) and scanning electron microscopy (SEM). In the case of TEM, the approach is most similar to the approach in optical microscopy, whereas in the case of SEM the electron beam scans over the surface and the products of its interaction with the sample are probed.

Apart from microscopy as a way to directly observe objects, there is also diffraction which allows observing periodic structures in the reciprocal space. The main idea is that radiation impinging on a periodic structure will interfere constructively and destructively, resulting in a diffraction pattern; the most basic

Figure 1.1: Images of a horse at gallop from Eadweard Muybridge, who proved with this experiment that a horse has all his feet off the ground at a gallop. Image taken from the website of the Rijksmuseum in Amsterdam.
example being visible light interference from a grating. Since atoms in a solid are also placed in a periodic structure, a lattice will also serve as a grating for suitable wavelengths. The first experiments were done with X-rays in the 1910`s by Paul Peter Ewald and Max von Laue. Von Laue got the Nobel Prize in Physics in 1914 for his discovery of the diffraction of X-rays by crystals. The next year, the Nobel Prize was awarded to father and son Bragg for their further research into the field of X-ray diffraction. They formulated the direct relation between the angles under which diffraction is observed and the lattice distances:

\[ 2d \sin \theta = n \lambda \]

Where \( d \) is the distance between lattice planes, \( \theta \) is the angle, \( n \) is the diffraction order and \( \lambda \) is the wavelength. X-ray diffraction has proven itself in the course of the 20\(^{th}\) century to be very useful for the determination of the structure of solids. Besides in materials science it is also widely used in biology for studies of the structure of proteins. With the recent advent of X-ray Free Electron Lasers (X-FEL`s) it became possible to probe nanocrystals of proteins in a single shot of an ultrashort pulse of X-rays before they are destroyed by the deposited energy.

Combining the two discoveries – the wave nature of electrons and the diffraction of waves from lattices – led to the invention of electron diffraction in the 1930s. In 1937 Clinton Joseph Davisson and George Paget Thomson got the Nobel Prize for their experimental discovery of the diffraction of electrons by crystals. Nowadays electron diffraction is a widely used standard method to study solids; especially when the surface properties of the material are of particular interest. Generally there are three ways in which electron diffraction is carried out: the first one is using the diffraction in a TEM setup. This technique is suitable when the bulk properties of the material are of interest. The second one is reflection high energy electron diffraction (RHEED) which is very sensitive to structures at the surface and is performed in grazing incidence geometry. Typical electron energies are tens of keV`s. Also low (tens to hundreds of eV`s) energy electron diffraction (LEED) is used to probe surface properties but makes use of the property of low energy electrons that they scatter back when they are impinging the surface in a normal incidence.
1.2 Ultrafast lasers, Pump-probe Spectroscopy, Femtochemistry

Commonly phase transitions are studied by describing the properties of systems in equilibrium conditions before and after the phase transition has occurred. Knowledge of which out-of-equilibrium transient states are occupied following the primary excitation of the system can contribute tremendously to understanding the occurrence and nature of the transition. The fundamental time scale of breaking bonds between atoms is in the 100 fs regime and therefore this is also the time resolution which these experiments are required to have.

Access to this sort of knowledge was made possible by the introduction of ultrafast laser systems in the second half of the 20th century. After the first experimental realization of a laser in 1960, the developments went very fast with the first nanosecond pulses already in 1961 [1] and sub-picosecond pulses followed in 1974 [2]. In 1997 Douwe Wiersma and his group at our institute even made it to the Guinness Book of Records with their shortest event ever produced by mankind, with a 5 fs laser pulse [1,3]. Nowadays laser jack produce pulses as short as 100 as (attoseconds).

As soon as the femtosecond laser was introduced, a lot of scientists started using these lasers to do experiments. One of the pioneers in the field of femtochemistry was Ahmed Zewail, who received in 1999 the Nobel Prize in Chemistry for his studies of transition states of chemical reactions by femtosecond spectroscopy [1]. In these early-days femtochemistry experiments, a molecular beam was sent into a vacuum chamber and two femtosecond laser pulses were applied. The first one (the pump pulse) excites the system and defines the starting point of the dynamics.

1.3 Ultrafast Electron Diffraction

With the spectroscopic pump-probe techniques described before, a lot of information on the transient excited states during chemical reaction and phase transitions can be obtained. However, the picture cannot be complete without knowledge of the intermediate structure. This structure can be probed in both real space (microscopy) and reciprocal space (diffraction). This is the field of ultrafast X-ray science, ultrafast electron diffraction (UED) and ultrafast electron microscopy (UEM).
In all these techniques, an ultrafast probe is preceded by an ultrashort laser pulse to provide energy to the system and thereby start the dynamics and define a time origin (time zero for when the pump and the probe arrive at the same time). The probe arrives a defined delay time after the pump and this delay time defines the period that the system is allowed to evolve before it is probed. In principle, the experiment needs to be repeated for each time delay for which information is wanted (in contrary to the case of the Muybridge horse experiment where multiple cameras were triggered at different delay times in one single experiment). This also implies that the time resolution is limited by the duration of both the pump and the probe pulse, whereas the camera can fundamentally be slow (again different from the horse experiment where one of the crucial developments of Muybridge was the short shutter time of the cameras). The basic principle of an UED experiment is schematically shown in Figure 1.2.

The choice of the probe depends on what one wants to see. When structures are bigger and long-range order is destroyed, diffraction is the relevant probe to be used. Electrons are the better probe for thin samples since their elastic scattering
probability is much larger. Furthermore, electron sources can be much smaller and easier to handle than X-ray sources.

When it comes to source development for UED, the brightness of the source is a very important quantity since it governs the relation between acquisition time and signal-to-noise ratio. However, the brighter the source, or the more electrons per pulse, the larger the Coulombic repulsion between the electrons. This leads to a bigger electron bunch and therefore worsens the time resolution of the experiment. There are multiple solutions possible for this space charge problem: compact gun designs in which there is only limited time for the electron bunches to grow in the propagation direction, electron guns with limited numbers of electrons per bunch (low brightness) with the ultimate goal of single-electron diffraction \cite{4,5}, or guns with active post-compression of the electron bunches, like in the case of our system \cite{6-9}.

New approaches in the field of source development include nanotip sources and cold atoms sources. Whereas in the traditional sources, photoelectrons are extracted from front-illuminated photocathodes, or back-illuminated thin metal films, in these new types of sources photoelectron generation is more complicated. A tip-based source \cite{10-12} consists of nanotips from which photoelectrons are extracted. The energy spread of the resulting electron bunch is smaller due to the very high field that can be achieved in this geometry. Furthermore, the small source size (which is smaller than the incoming laser beam size) leads to a high transverse coherence length. This is an interesting development; however, experimental times to collect a useful amount of data are very long. Therefore, the development of cold atom sources \cite{13} is even more promising. A magneto-optical trap (MOT) is built to reduce the atoms’ kinetic energy to an effective temperature of 10 K. Photoelectrons originating from these atoms have a very narrow thermal broadening and therefore a long coherence length with large numbers of electrons per pulse.

One of the early breakthrough experiments of this field was an observation of ultrafast melting of an aluminium thin film by Siwick \textit{et al}. \cite{14}. In this study polycrystalline, 20 nm thick Al films were irradiated with 120 fs near-IR laser pulses and subsequently probed with 600 fs electron pulses. From the evolution of the diffraction pattern after irradiation, an atomic description was deduced of
how the heating of the lattice takes place and how the energy is redistributed in
the material. From the time scale at which the process takes place, it was
concluded that the disordering is not electronically driven. During the first few ps,
the aluminium is in a superheated state that only exists transiently; afterwards
the energy is redistributed and the lattice is disordered after 3.5 ps to a state
where no short-range order is detectable anymore (see Figure 1.3).

In Siwick’s experiment a substantial number of electron bunches was needed for
each time point of the experiment. The use of MeV sources allows for single shot
UED. At relativistic energies, the space charge forces are highly suppressed and
therefore the number of electrons in a sub-picosecond electron bunch can be
orders of magnitude higher than in lower energy regimes. One of the leading

Figure 1.3: UED images of polycrystalline aluminium at different pump-probe delays. Taken from
[14].
groups in this respect is the group of Pietro Musumeci at the University of California, Los Angeles (UCLA). They have used this technique to follow laser-induced melting of a single crystal of gold with single shots per time point [15]. One step further is performing a real-single-shot experiment [16]. A 20 ps long electron bunch was streaked with an RF cavity after arriving at the sample to transfer the information in the time domain to a direction perpendicular to the propagation direction of the electrons. In this way, all information of the time evolution of the diffraction spots was contained in a single shot. The result of this experiment is shown in Figure 1.4.

Besides these melting phase transitions, also more exotic phase transitions are studied by means of UED. A nice example of these studies is the study of charge density wave materials (CDW) [17,18].

Charge density wave materials are materials in which there is periodic modulation in the density of conduction electrons, causing a periodic lattice displacement.
(PLD). The structure of such a material, $4Hb$-$TaSe_2$, is shown in Figure 1.5 (a) and the PLD is schematically shown in Figure 1.5 (c). This PLD can be observed experimentally by electron diffraction where it can be seen as a superstructure with satellite peaks ordered around the normal Bragg peaks, as shown in the diffraction pattern in Figure 1.5 (b). As the CDW state is a low temperature state, the transition from the commensurate to the incommensurate CDW state can be studied by means of ultrafast electron diffraction. The phase transition can then be seen as a decrease of intensity in the satellite peaks because the CDW state is suppressed upon (ultrafast) heating.

Such a study was performed on $4Hb$-$TaSe_2$ by the group of Heinrich Schwoerer at Stellenbosch University [17]. The transient intensity of both the Bragg spots and

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*Figure 1.5: (a) Crystal structure of $4Hb$-$TaSe_2$. (b) Equilibrium state diffraction pattern. (c) Phase transition between the normal state (left) and the commensurately modulated state (right). Image taken from [17].*
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the CDW satellite spots was followed as a function of delay time between the excitation with a pump pulse (780 nm) and a probe electron bunch. The experiment was repeated at different fluences. The main results are shown in Figure 1.6.

Upon excitation, two processes take place: the first process (at a time scale shorter than 400 fs) is the change of electron distribution due to the electronic excitation. Due to coherent electron-phonon coupling, the atoms move to their equilibrium state that corresponds to the high-temperature phase. In the diffraction pattern this process is characterized by a depletion of the CDW spots and an increase of the Bragg spots. Followed by this process, incoherent phonons result in a Debye Waller effect (more disorder due to heating of the lattice), which can be seen as a decrease of the intensity of both the Bragg and the CDW spots.

Based on a BCS analysis of the transient PLD as a function of fluence, it was determined that the photo-induced ultrafast transition is a second order phase transition which is different from the normal thermal phase transition which is first order.

In a follow up study [18] the metamorphosis between the nearly commensurate (NC) and incommensurate (IC) CDW phase of 1T-TaS$_2$ was studied with the same experimental technique. The domain sizes were estimated by following the widths of the diffraction spots corresponding to the CDW as a function of time. At the short time scale (1.5 ps) the IC peak width goes up by a factor of 2, indicating a domain size of about 4 nm; however after 100 ps the widths go back to the lattice peak width, indicating a domain size larger than the coherence length of the electron pulse (8 nm).
Figure 1.6: Results of the UED experiment on 4Hb-TaSe2. In (a) the surrounding of a Bragg spot is shown with the satellite peaks clearly present before T0 and vanishing after T0. In (b) the intensity of the Bragg peaks is shown as a function of delay time; in (c) the corresponding CDW peak intensity is shown. Image taken from [17].
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1.4 The scope of this thesis

At the Zernike Institute for Advanced Materials (Surfaces & Thins Films group) at the University of Groningen we have built an ultrahigh vacuum (UHV) setup to do UED. UHV conditions allow for the study of clean surfaces, which is important when the surface and interface properties of the material in study are relevant. Our setup is among the first in the world where UED and surface science meet. It is also unique in the possibility to switch between reflection and transmission geometry, which makes it very flexible for a variety of materials and samples of interest.

The PhD project of which this thesis is the final product was mostly dedicated to the building up phase and the commissioning phase of this experimental setup. Chapter 2 describes the function and characteristics of all components of the setup, as well as some preliminary experimental results which aim to prove the temporal resolution of the experiments.

Chapter 3 and 4 describe the first two real experiments that were performed in this new laboratory. In the first experiment we produced a bilayer foil, consisting of chromium and gold and studied the interface dynamics by means of UED. Chapter 4 describes the experiment that resulted from collaboration with the group of Mehmet Açet at the University of Duisburg-Essen (Germany). The dynamics of the martensite-austenite phase transition in Heusler alloys were studied.

The last chapter (Outlook) describes a possible experiment that would make full use of the strengths of our experiment setup: a study on the phase transition in which the superconducting phase in thin films magnesium diboride ($\text{MgB}_2$) is destroyed. Angle Resolved Photoelectron Spectroscopy measurements were carried out to study this phase transition from the electronic point of view and UED measurements would complement this with structural information. Therefore it would be a very promising and interesting future experiment making use of all capabilities of the UHV UED setup.
References
